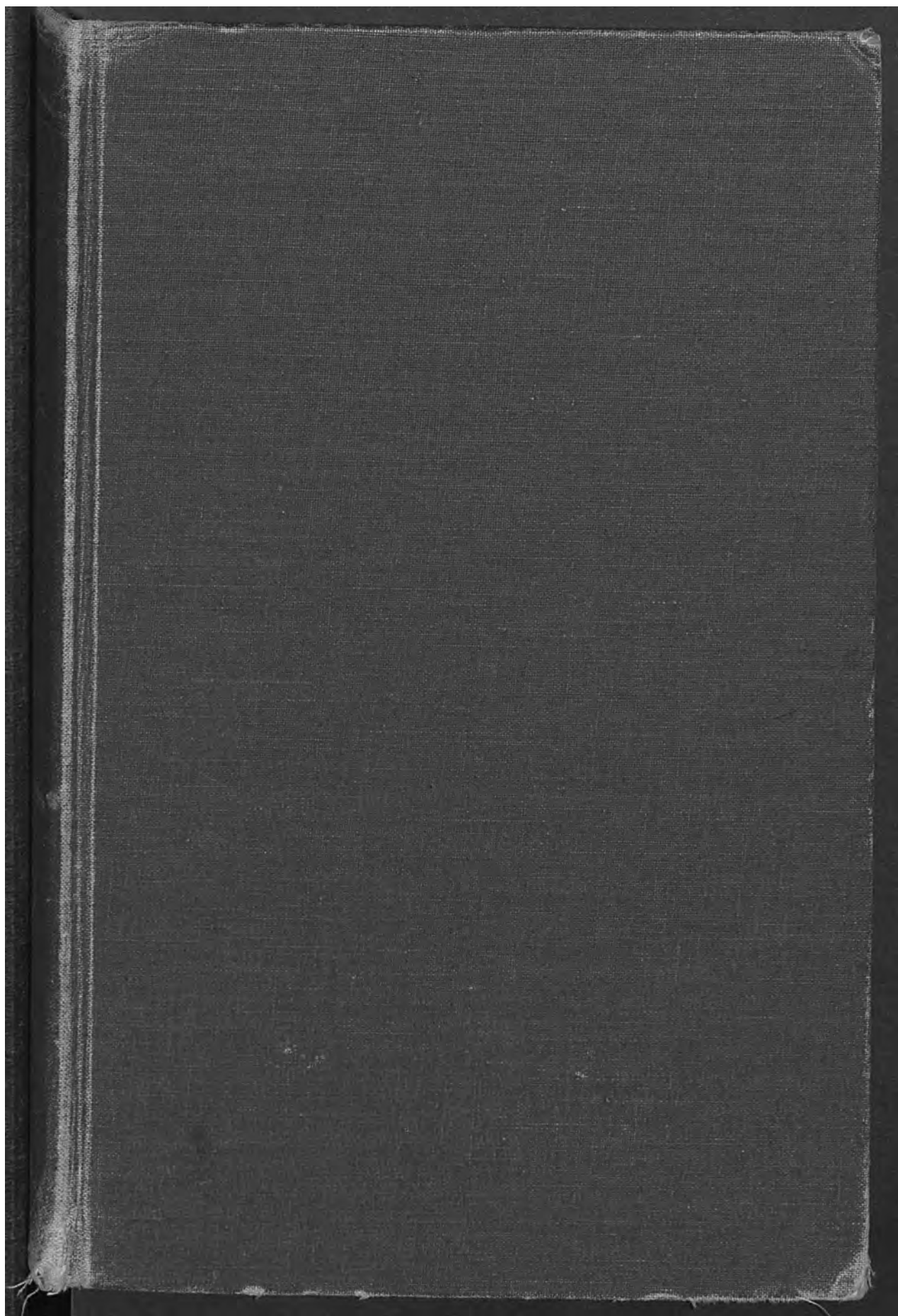
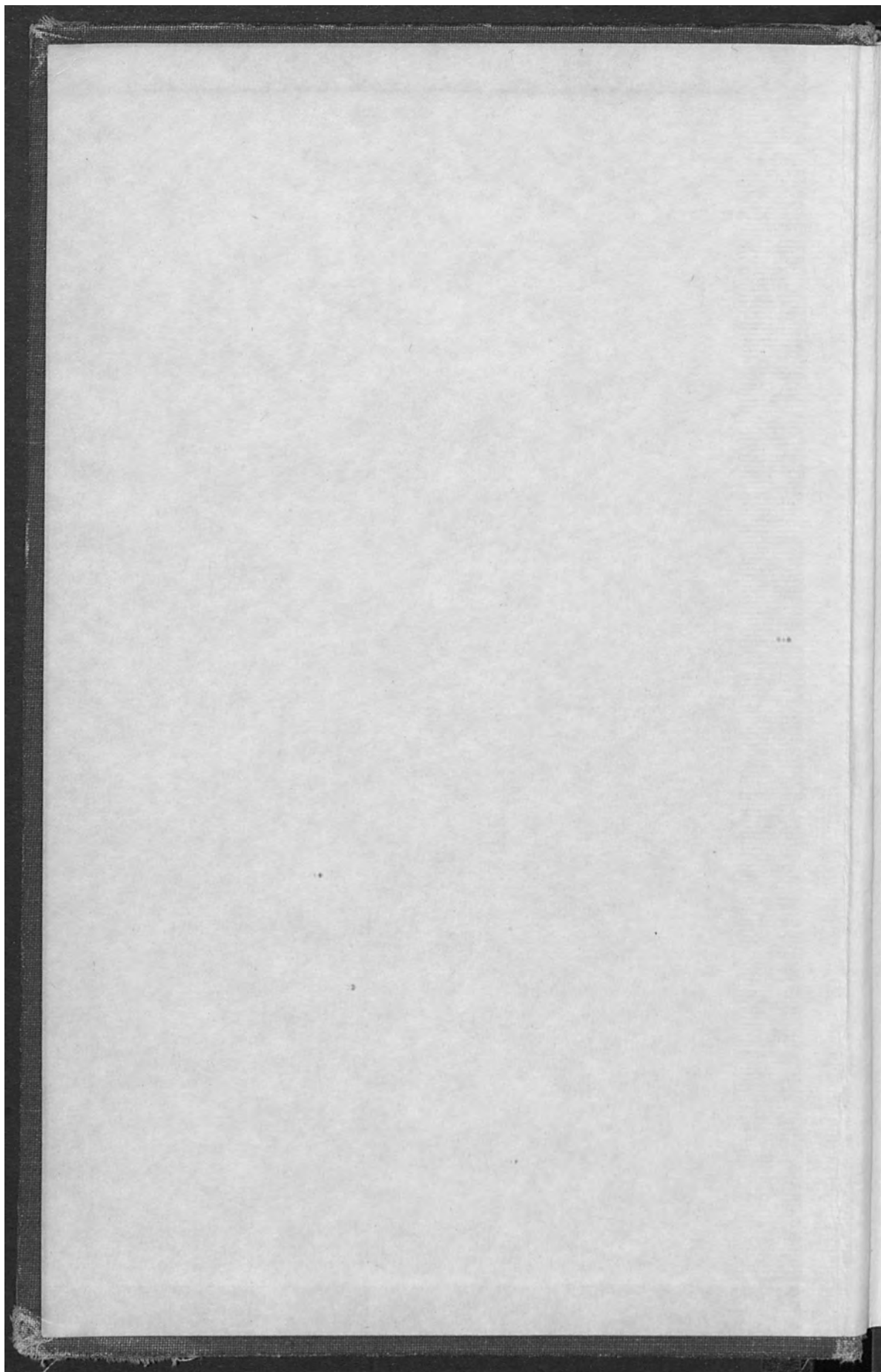
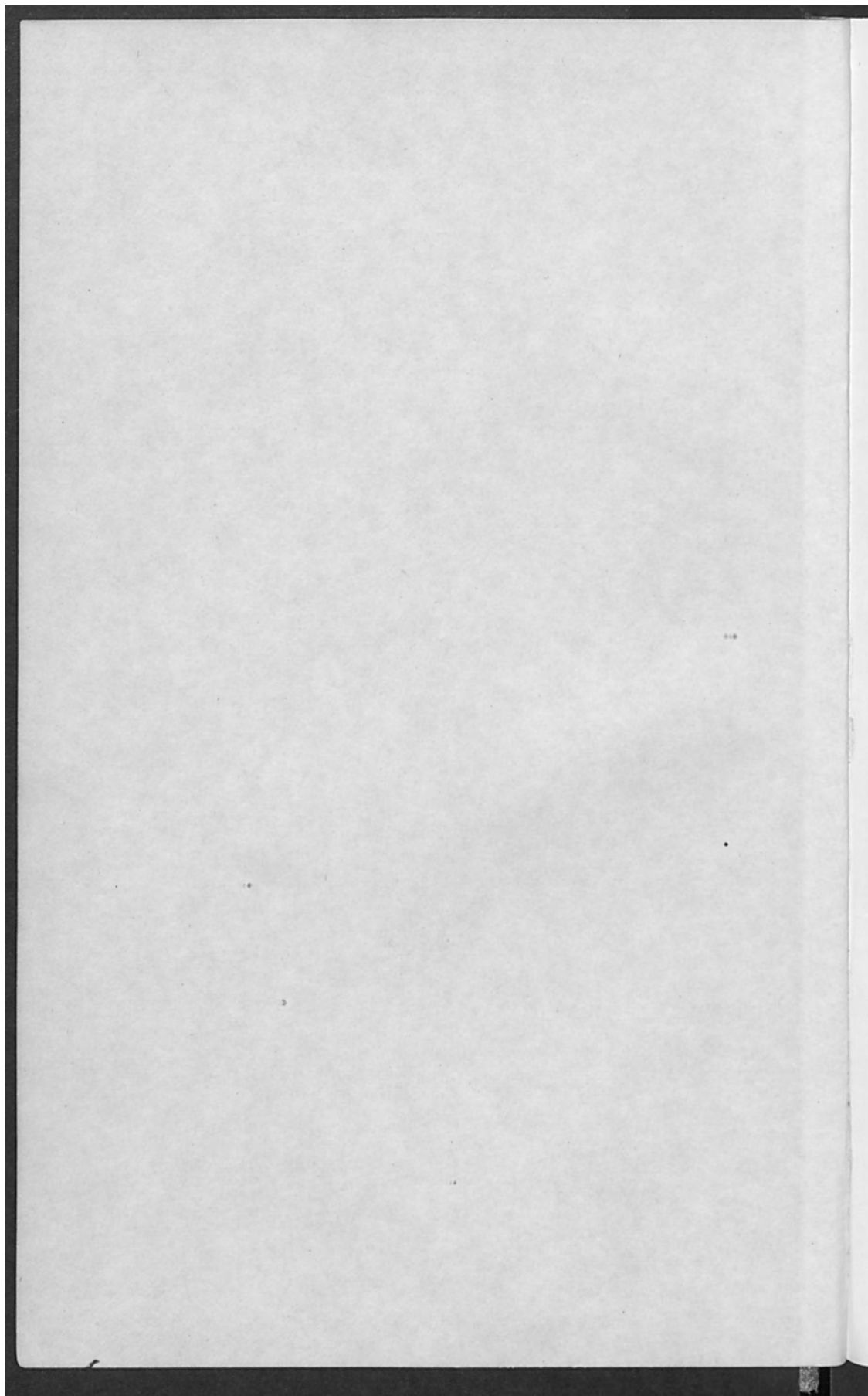






R. Babick



First International Conference on
ELECTRON AND ION BEAM SCIENCE AND TECHNOLOGY

THE ELECTROCHEMICAL SOCIETY SERIES

THE CORROSION HANDBOOK

Edited by Herbert H. Uhlig

ABSTRACTS OF THE LITERATURE ON
SEMICONDUCTING AND LUMINESCENT
MATERIALS AND THEIR APPLICATIONS
Compiled by Battelle Memorial Institute

ELECTROCHEMISTRY IN BIOLOGY AND MEDICINE

Edited by Theodore Shedlovsky

VAPOR-PLATING

By C. F. Powell, I. E. Campbell, B. W. Gonser

HIGH-TEMPERATURE TECHNOLOGY

Edited by I. E. Campbell

STRESS CORROSION CRACKING AND EMBRITTLEMENT

Edited by William D. Robertson

ARCS IN INERT ATMOSPHERES AND VACUUM

Edited by W. E. Kuhn

TECHNOLOGY OF COLUMBIUM (NIOBIUM)

Edited by B. W. Gonser and E. M. Sherwood

MECHANICAL PROPERTIES OF INTERMETALLIC COMPOUNDS

Edited by J. H. Westbrook

THE STRUCTURE OF ELECTROLYTIC SOLUTIONS

Edited by Walter J. Hamer

TRANSACTIONS OF THE SYMPOSIUM ON ELECTRODE PROCESSES

Edited by Ernest Yeager

IODIDE METALS AND METAL IODIDES

By Robert F. Rolsten

MODERN ELECTROPLATING, SECOND EDITION

Edited by Frederick A. Lowenheim

FIRST INTERNATIONAL CONFERENCE ON ELECTRON AND ION BEAM SCIENCE
AND TECHNOLOGY

Edited by Robert Bakish

First International Conference on
ELECTRON AND ION BEAM
SCIENCE AND TECHNOLOGY

Edited by
ROBERT BAKISH
Consulting Engineer
Englewood, New Jersey

Sponsored by
THE ELECTROTHERMICS AND METALLURGY DIVISIONS
OF THE ELECTROCHEMICAL SOCIETY, INC.



in cooperation with
THE METALLURGICAL SOCIETY OF THE AIME



JOHN WILEY & SONS, INC.
NEW YORK · LONDON · SYDNEY

Copyright © 1965 by John Wiley & Sons, Inc.
All rights reserved. This book or any part thereof
must not be reproduced in any form without the
written permission of the publisher.

Library of Congress Catalog Card Number: 65-14260
Printed in the United States of America

FIRST INTERNATIONAL CONFERENCE
ON ELECTRON AND ION BEAM SCIENCE
AND TECHNOLOGY

Conference Committee and Session Chairmen

R. BAKISH, *Chairman*

DR. F. BENESOWSKI

Metalwerk Plansee
Reute Tirol, Austria

PROF. A. E. JENKINS

University of South Wales
N. S. W., Australia

DR. D. TYTGAT

Centre d'Etude Nucleaire
Mol-Donk, Belgium

DR. B. W. SCHUMACHER

Ontario Research Foundation
Toronto 5, Canada

PROF. J. CABELKA

International Institute of Welding
Bratislava, Czechoslovakia

M. BOSTON, Esq.

Torvac Ltd.
Histon, Cambridge, England

DR. A. H. BELK

College of Advanced Technology
Birmingham 4, England

PROF. M. VON ARDENNE

am Weisen Hirsch
Dresden, East Germany

PROF. E. REXER

Institute for Applied Physics
Dresden, East Germany

DR. J. A. STOHR

Centre d'Etudes Nucleaires
Saclay, France

DR. R. CASTAING

University of Paris
Orsay (S.-et-O), France

DR. J. G. SIEKMAN

Philips Research Laboratories
Eindhoven, Holland

DR. S. NAMBA

The Institute of Physical & Chemical
Research
Tokyo, Japan

DR. C. HAYASHI

Japan Vacuum Engineering Co. Ltd.
Yokohama, Japan

DR. O. WINKLER

Balzers AG
Balzers, Lichtenstein

DR. E. BAS

Swiss Federal Polytech Inst.
Zurich, Switzerland

PROF. G. MOLENSTEDT

University Tübingen
Tübingen, West Germany

DR. H. DIETRICH

W. C. Heraeus
Hanau-Main, West Germany

DR. W. WRIGHT

Micro Systems Inc.
Pasadena, California

DR. A. LAWLEY

Franklin Institute
Philadelphia, Penna.

DR. J. WESTBROOK

G. E. Research Laboratory
Schenectady, N. Y.

H. P. MUNGER

Republic Steel Company
Cleveland, Ohio

DOZENT DR. N. A. OLSHANSKI

Moscow Institute for Energy
Moscow, U.S.S.R.

DR. B. PATON, Director

Institute for Electro-Welding
Kiev, U.S.S.R.

DR. A. KIASPAUL
Hughes Research Laboratories
Malibu, California

DR. A. T. FORRESTER
Electro Optical Systems, Inc.
Pasadena, California

PROF. R. E. OGLIVIE
M.I.T.
Cambridge, Massachusetts

MR. R. T. LAMB
Ampex Corporation
Redwood City, California

Preface

This book contains the papers presented at the first International Conference on Electron and Ion Beam Science and Technology held May 3-7, 1964, in Toronto, Canada. These papers were selected by a committee of some twenty-five individuals, representing all countries where consequential work in the field is conducted; I wish to thank the committee for their effort.

The desire to establish the level of achievement in this technology the world over, and to present material in this area of science and technology which has not seen prior publication, was used as a guideline in the selection of the material. Moreover, attempts were made to assure both the quality of the presentations as well as to render as broad a coverage of the subject matter as possible. The content of this book defines the scope of applications of this powerful technology.

It is hoped that this record of the conference brings to fruition the effort of all the individuals who donated so generously of their time to make this event possible and successful. The record in itself is a milestone for what is to become a biannual event to be conducted under the sponsorship of the first conference, that is, the Electrothermics and Metallurgy Divisions of The Electrochemical Society and the Metallurgical Society of the AIME. The individuals who organized the first meeting are preparing the second conference which will be held April 17-20, 1966, at the Park Sheraton Hotel in New York. It is appropriate here to thank these societies for their interest in the conference and for the generous support given to date.

As the reader will be able to establish, this record reflects the state of affairs in Electron and Ion Beam Science and Technology circa 1964. It achieves this by presenting a good representative cross section of work being carried out in laboratories throughout the world today.

The book presents the material in a somewhat different order from that

presented at the conference. I have assumed my prerogative as editor to rearrange the presentations in the hope of producing a better organized book and a valuable reference for many years to come. As a matter of fact, I am certain that no individual working in this field could afford the luxury of not having this book at his desk.

The volume consists of seven sections as follows: (1) Physics of Electron and Ion Beams, (2) Electron and Ion Beams in Microelectronics, (3) Electron Beams Recording and Information Storage, (4) Electron Beams in Material Processing, (5) Electron Beams in Materials Joining, (6) Electron Beams in Microanalysis, and (7) Ion Propulsion. Most sections are preceded by a brief introduction presenting consequential historical events, achievement to date, literature of importance, and an indication of section content highlights. Such an arrangement will permit even the newcomer in the field to obtain a good appreciation of the material at hand and will briefly remind the worker actually involved in this technology of the recent and more distant events of consequence in the field.

In closing permit me again to thank those who made this book a reality and to say that it was both a pleasure and an honor to work with this dynamic group of forward-thinking individuals. Last, but not least, thanks are due to my wife, Ellen, for her patience in the course of the preparation of this manuscript.

ROBERT BAKISH

Englewood, New Jersey
January 1965

Contents

SECTION I	PHYSICS OF ELECTRON AND ION BEAMS	1
1.	B. W. SCHUMACHER, A Review of the (Macroscopic) Laws for Electron Penetration through Matter	5
2.	W. B. NOTTINGHAM, Fundamentals of Thermionic Energy Conversion	71
3.	K. AMBOSS, Verification and Use of Herrmann's Optical Theory of Thermal Velocity Effects in Electron Beams in the Low-Perveance Regime	86
4.	E. B. BAS AND G. CREMOSNIK, Experimental Investigation of the Structure of High-Power-Density Electron Beams	108
5.	EMIL C. MULY AND JAMES LITTON, JR., Energetic Electron Beams and Cold Cathode Boundary Conditions	133
6.	HELMUT SCHWARZ, Mechanism of High-Power-Density Electron-Beam Penetration in Metal	158
SECTION II	ELECTRON AND ION BEAMS IN MICRO-ELECTRONICS	187
7.	A. N. BROERS, Micromaching by Sputtering through a Mask of Contamination Laid Down by an Electron Beam	191
8.	P. A. EINSTEIN, Some Observations on the Formation of Tracks in Thin, Supported Metal Films Produced by an Electron Beam	205
9.	R. F. W. PEASE AND W. C. NIXON, Microformation of Filaments	220
10.	JOSEPH M. HLAVIN AND RICHARD A. FOTLAND, The Fabrication of Electronic Components Using Low-Energy Electron Beams	231
11.	KAZUO YAMAGISHI, Vacuum Deposition by Means of Electron Beam	245

12. SUSUMU NAMBA, KIYOSHI WATANABE, EIICHIRO GOTOH, AND KATSUO NISHI, Automatic Control of Electron-Beam Micromaching 264
13. HIROKAZU KIMURA, HIFUMI TAMURA, KAZUTOSHI IKEGAMI, AND KIYOSHI SAKASHITA, Fabrication of Microelement Resistors by Electron Beam and Their Characteristics 283
14. D. J. GARIBOTTI, E. H. MILLER, AND P. ANDERSON, Automation of Electron-Beam Processes for Microelectronics 293

SECTION III ELECTRON BEAMS IN RECORDING AND
INFORMATION STORAGE 337

15. T. E. EVERHARDT, Nonthermal Electron-Beam Recording 341
16. P. WHITE AND D. W. NORTH, An Electron-Beam Memory Utilizing Some Properties of Polymer Films 352

SECTION IV ELECTRON BEAMS IN MATERIALS PROCESSING 365

17. MANFRED VON ARDENNE, Some New Contributions in the Field of Electron-Beam, Ion-Beam, and Plasma-Beam Technology 370
18. B. E. PATON AND B. A. MOVCHAN, Electron-Beam Radial Heaters for Melting of Metals 406
19. C. HAYASHI, S. KASHU, M. KOSAKI, AND H. HIRUTA, Casting Titanium by Using an Electron-Beam Skull Furnace 415
20. HANS FIEDLER, On the Problem of Remelting Steel in an Electron-Beam Multichambered Furnace 428
21. A. ACCARY, A. TREILLOU, AND J. TROUVÉ, Electron-Beam Melting of Ceramic Materials 439
22. P. W. BARTON, E. J. HUGHES, AND A. A. JOHNSON, The Preparation and Properties of the Intermediate Phase Based on the Compound TiCo 460
23. H. E. DOERING AND P. SHAHINIAN, Effect of Electron Bombardment Heating on Surfaces of Tungsten 481
24. J. C. WILSON AND M. L. PICKLESIMER, Variable Gradient, Electron-Beam Heating Methods for Growing Single Crystals of Zirconium 502
25. C. S. TEDMON, JR., AND R. M. ROSE, Substructure and Impurities in Niobium Single Crystals Grown by the Electron-Beam, Floating-Zone Technique 521
26. E. REXER, On the Deformation, Recrystallization, and Welding of Molybdenum Single Crystals 541
27. D. R. HAY AND E. SCALA, Field-Aided Electron-Beam Zone Refining of Tungsten 550

CONTENTS

xi

28. L. M. GILLIN, A Multipurpose, High-Temperature Tensile-Testing Machine Using Electron-Beam Heating . . . 567

SECTION V ELECTRON BEAMS IN MATERIALS JOINING . . . 583

29. N. A. OLSHANSKIY, AND O. K. NAZARENKO, The Current Status of Electron-Beam Welding in the USSR . . . 587
30. J. L. SOLOMON, New Developments in Electron-Beam Welding . . . 606
31. J. W. MEIER, Recent Developments in Nonvacuum Electron-Beam Welding . . . 634
32. R. F. KLEIN, Electron-Beam Welding of Nuclear Fuel Cladding Components . . . 655
33. A. J. WILLIAMS AND C. F. MARTIN, Properties of Electron-Beam Welds in Ultra-High-Strength Steels . . . 674
34. MEL SCHWARTZ, Electron-Beam Welding of Brazed Refractory Joints . . . 713
35. E. B. BAS, G. CREMOSNIK, AND H. LERCH, A 30-kV Electron-Beam Welding Machine with Indirectly Heated Tungsten Cathode . . . 734
36. P. R. PAYNE, JR., A System to View Electron-Beam Welding by Closed-Circuit TV . . . 759

SECTION VI ELECTRON BEAMS IN MICROANALYSIS . . . 775

37. R. CASTAING AND G. SLODZIAN, Electron and Ion Beams in Microanalysis . . . 780
38. D. QUATAERT AND R. THIESEN, Quantitative Analysis Using an Electron Microanalyzer . . . 799
39. R. THEISEN, Detection Limits of Electron Microprobe Analysis—Application of "Increased Sensitivity Method" in Metallurgy . . . 813
40. SHELDON H. MOLL, Practical Methods for Experimentally Calibrated Concentration Determination in the Electron-Beam Microanalyzer . . . 825
41. V. LEROY, Comparison of the Diffusion of Several Substitutional Elements in Nickel and in Cobalt . . . 830

SECTION VII ION PROPULSION . . . 851

42. R. M. WORLOCK, The Surface Ionization Ion Engine . . . 853
43. H. HUBER, R. LE BIHAN, AND D. MAUGIS, A Cesium Plasma Capillary Emitter as a Source of Cesium Ions or a Source of Electrons of High-Current Density . . . 877
44. A. SEPTIER, F. PRANGERE, AND H. ISMAIL, A High-Intensity Mercury Ion Source of Duoplasmatron Type . . . 897

45. CHARLES D. HENDRICKS, Classical Physics in Modern Space Propulsion: Heavy Particle Beam Formation by Electrical Spraying of Liquids	915
46. PAUL D. READER, The Operation of an Electron Bombardment Ion Source with Various Gases	925
47. G. ROHRINGER, Design of Ion Optics on Digital Computers (Abstract)	936
INDEX	937

PHYSICS OF ELECTRON AND ION BEAMS

Section I

PHYSICS OF ELECTRON AND ION BEAMS

1. ELECTRON AND ION BEAMS	1
2. ELECTRON BEAMS	1
3. ION BEAMS	1
4. ELECTRON BEAMS	1
5. ION BEAMS	1
6. ELECTRON BEAMS	1
7. ION BEAMS	1
8. ELECTRON BEAMS	1
9. ION BEAMS	1
10. ELECTRON BEAMS	1
11. ION BEAMS	1
12. ELECTRON BEAMS	1
13. ION BEAMS	1
14. ELECTRON BEAMS	1
15. ION BEAMS	1
16. ELECTRON BEAMS	1
17. ION BEAMS	1
18. ELECTRON BEAMS	1
19. ION BEAMS	1
20. ELECTRON BEAMS	1
21. ION BEAMS	1
22. ELECTRON BEAMS	1
23. ION BEAMS	1
24. ELECTRON BEAMS	1
25. ION BEAMS	1
26. ELECTRON BEAMS	1
27. ION BEAMS	1
28. ELECTRON BEAMS	1
29. ION BEAMS	1
30. ELECTRON BEAMS	1
31. ION BEAMS	1
32. ELECTRON BEAMS	1
33. ION BEAMS	1
34. ELECTRON BEAMS	1
35. ION BEAMS	1
36. ELECTRON BEAMS	1
37. ION BEAMS	1
38. ELECTRON BEAMS	1
39. ION BEAMS	1
40. ELECTRON BEAMS	1
41. ION BEAMS	1
42. ELECTRON BEAMS	1
43. ION BEAMS	1
44. ELECTRON BEAMS	1
45. ION BEAMS	1
46. ELECTRON BEAMS	1
47. ION BEAMS	1
48. ELECTRON BEAMS	1
49. ION BEAMS	1
50. ELECTRON BEAMS	1
51. ION BEAMS	1
52. ELECTRON BEAMS	1
53. ION BEAMS	1
54. ELECTRON BEAMS	1
55. ION BEAMS	1
56. ELECTRON BEAMS	1
57. ION BEAMS	1
58. ELECTRON BEAMS	1
59. ION BEAMS	1
60. ELECTRON BEAMS	1
61. ION BEAMS	1
62. ELECTRON BEAMS	1
63. ION BEAMS	1
64. ELECTRON BEAMS	1
65. ION BEAMS	1
66. ELECTRON BEAMS	1
67. ION BEAMS	1
68. ELECTRON BEAMS	1
69. ION BEAMS	1
70. ELECTRON BEAMS	1
71. ION BEAMS	1
72. ELECTRON BEAMS	1
73. ION BEAMS	1
74. ELECTRON BEAMS	1
75. ION BEAMS	1
76. ELECTRON BEAMS	1
77. ION BEAMS	1
78. ELECTRON BEAMS	1
79. ION BEAMS	1
80. ELECTRON BEAMS	1
81. ION BEAMS	1
82. ELECTRON BEAMS	1
83. ION BEAMS	1
84. ELECTRON BEAMS	1
85. ION BEAMS	1
86. ELECTRON BEAMS	1
87. ION BEAMS	1
88. ELECTRON BEAMS	1
89. ION BEAMS	1
90. ELECTRON BEAMS	1
91. ION BEAMS	1
92. ELECTRON BEAMS	1
93. ION BEAMS	1
94. ELECTRON BEAMS	1
95. ION BEAMS	1
96. ELECTRON BEAMS	1
97. ION BEAMS	1
98. ELECTRON BEAMS	1
99. ION BEAMS	1
100. ELECTRON BEAMS	1

Section 1

PHYSICS OF
ELECTRON AND ION BEAMS

PHYSICS OF ELECTRON AND ION BEAMS

Progress in any area of scientific endeavor has been intimately connected with advances in the physics related to this area. In scientific problems related to electron and ion beams this is certainly true, and the digestion of the science and its conversion to technology have been very dependent on advances in the physics involved.

By virtue of the complexity and the breadth of the subject matter, which we can refer to as "Electron and Ion Beam Science and Technology," a corresponding breadth of problems are present. In the conference we were able to consider only a minute number of them. There are problems which the group working on conference planning thought to be of greatest importance and most directly related to possible advances in particle beam application systems. It is these problems which are discussed in this first section of the volume.

Although any significance in the order of the presentations in this section is fortuitous, I have attempted to arrange the papers so as to present a gradually unfolding picture of the problems considered.

The papers discuss a number of important aspects basic to the generation of electron and ion beams. They consider approaches and scientific methods, they report on material problems associated with the various systems, and, last but not least, they consider the problems of interaction of particles and matter and the consequential changes thereof.

It is hoped that these contributions will help to further the understanding of fundamental principles involved and indirectly effect the progress in electron and ion beam technology.

As one who has been indirectly involved with the physics of electron and ion beam generation and related subject matter, I feel that we, who use electron and ion beams for tackling of material problems, owe an immense debt to those who have painstakingly worked to expand the bounds of knowledge in beam generating systems.

For those who wish to obtain a bird's eye view into the physics of electron beams, I suggest referring to Harris (1962). But should you wish to become better acquainted with the many problems which one concerned with beam generation faces, the references listed below should be of considerable help.

Editor

REFERENCES

- von Ardenne, M., 1963, Physik Tabellen, Band I, VEB, Berlin.
Harris, L.A., 1962, in "Introduction to Electron Beam Technology," R. Bakish, Ed., John Wiley and Sons, New York, p. 21.
Klemperer, O., "Electron Optics," Cambridge United Press.
Pierce, J., 1954, "Theory and Design of Electron Beams," second edition, D. van Nostrand Co., Princeton, N.J.,
Slater, J.C., 1953, "Microwave Electronics," D. van Nostrand Co., Princeton, N.J.
Weber, E., 1950, "Electromagnetic Fields: Theory and Application," John Wiley and Sons, New York.
Zvorykin, V. K., G. A. Morton, E. G. Roamberg, J. Hillier, and A. W. Vance, 1945, "Electron Optics and the Electron Microscope," McGraw-Hill Book Co., New York.

A REVIEW OF THE (MACROSCOPIC) LAWS FOR THE ELECTRON PENETRATION THROUGH MATTER†

"The theory of electrons, on which I shall have the honour to lecture before you, already forms so vast a subject, that it will be impossible for me to treat it quite completely."

H. A. Lorentz 1906

INTRODUCTION

General Remarks about the Subject

Electron penetration through matter has been studied for over seventy years [1] and a review of the subject may seem somewhat out of date. But with the renewed interest in the use of electron beams as tools for various purposes many people will have to concern themselves with it and, I am sure, will thereby find that (a) an enormous amount of literature on this subject can be found,¹ (b) that this literature is rather evenly spread over the past with no particular peak period, (c) that neither the terminology nor the numerical values arrived at by various authors are in full agreement, (d) that abstracts and quotations even in most recent years

*Ontario Research Foundation, Department of Physics, Toronto, Canada.

†This paper was one of the four plenary presentations on the opening session of the conference.

¹My own file contains about 500 references.

can be completely wrong and misleading, (e) that few of the mathematical treatments of the subject are shedding much light on it, and even fewer give numerical results which agree with experiments, and (f) that an impropotionally large effort has gone in certain directions investigating mainly aspects for which a mathematical theory seemed possible.

The reason for this state of affairs is primarily to be sought in the complexity of all the processes which take place when a fast electron passes through matter. In addition, what we observe in any one experiment is often complicated by an interplay between several such processes, modified further by responses of the apparatus or observer.

I want to present here a, as I believe, logical description of the events and the observations which one can make, the laws which can be stated, and certain functions and numerical coefficients with which one can work. But I am not concerning myself with a review of the numerical values which are found in the literature. The discussion will also be restricted to what one can observe on a macroscopic scale, with only a few references to quantum mechanical theories of scattering, etc. It will be seen that this is indeed a study in its own rights.

In any discussion of electron penetration, etc., it is absolutely necessary to state the range of energy of the electrons which interact with the atoms because in the very low range of a few eV, and in the very high range, of several 10 MeV, different processes dominate the picture (to mention only tunneling or Ramsauer effect and radiation losses of Cerenkov effect) which are of no or little importance in the range of, say, 1 keV to 10 MeV. In this paper we will restrict ourselves to the last-mentioned energy range. We will also restrict ourselves to electron beams of such low current that the energy dumped into the absorber per unit time does not influence the penetration. In other words, conditions as we find them where the thermal action of electron beams is utilized, are not included in the discussion.

The laws of electron penetration through matter as reviewed here are of prime interest in connection with all applications of electron probes for analytical purposes, for instance in mass determination of objects in the electron microscope [2]-[7]; in connection with resolution and contrast in the electron-beam scanning microscope operating in the refelction, transmission or absorption mode; in connection with the X-ray production in the microprobe analyzer of the Castaing type [8]; in the electron beam thickness gauge for thin integuments [9], [10]; in the absorption and fluo-

rescence probes for supersonic rarefied gas flow [11]-[20] and plasma analysis [21]; in density, temperature [22], level and thickness gauges with beta rays; in attenuation and single-scatter gauges for high-altitude measurements [17], [23]; in all devices with secondary emission [24], [25]; in the interaction of beams with X-ray targets, fluorescence screens, photographic plates, and other detectors; in biological applications of electron beams; in radiation chemistry; and in connection with shielding designs. This long list shows that every effort toward a final clear understanding of all phenomena would well be justified.

Appearance of Electron Beams Passing through Matter

It is never wrong to look at a subject with our eyes and get as much information as possible from visual inspection. There are two methods for inspecting electron penetration into a homogeneous absorber. In the cloud chamber we see the individual electron path. And if we fire a beam into gas we see the resultant collective scatter distribution from the fluorescence of the gas. Figure 1a shows a cloud chamber picture. Figure 1b shows a beam fired

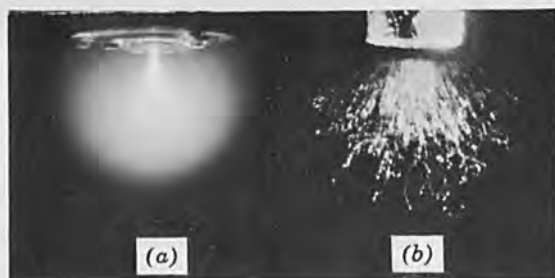


Fig. 1. (a) Top: cloud chamber picture of electron tracks coming from a beta emitter on the left (photograph courtesy Prof. Raether). (b) Lower half: Parallel beam of electrons of 60 keV, 60 A, 0.2-mm diameter fired into the atmosphere [13]. Note the irregular zigzag path of the individual electrons in the top picture and the short but intense central beam core followed by a brush or fountain-type scatter and backscatter in the collective process shown in the lower picture. Note also the rather sharp range (approximately 6 cm) with a nearly spherical boundary.

into the atmosphere. Individual electrons travel a very irregular zigzag path, collectively however they show a beautiful, regular, fountainlike scatter distribution. It extends over a nearly spherical region although the beam was originally parallel. This is typical for electrons where scattering dominates over energy loss. We may compare this with a beam of protons fired into nitrogen and hydrogen as shown in Figs. 2a, b. These particles lose their energy and come to rest before the beam has spread to any great extent.

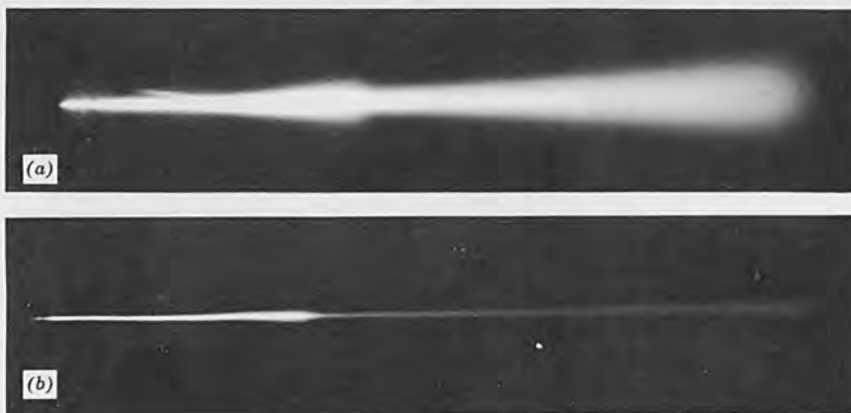


Fig. 2. (a) Top: Parallel beam of 0.6 MeV H^+ and H_2^+ ions fired into nitrogen of 140 torr. Range approximately 1.2 mg/cm^2 . (b) Lower half: The same beam fired into hydrogen of 400 torr. Note the insignificant scattering when compared with Fig. 1 [15].

Qualitatively we can say electron-electron scattering takes place between particles of equal mass, and therefore we get the strong deviation of the beam electron. The heavier ions do not deviate from their path in a collision with an electron although they may transfer energy to it (e.g., eject it from an atom, causing ionization). Collisions with the nucleus of an atom are comparatively rare in both cases. The scatter distribution of electrons of higher energy, where we have the relativistic increase in mass, approaches the one for ions.

Quantities of Interest

In order to describe the phenomena quantitatively we must first determine which quantities can be measured and are at the same

time physically meaningful. Figure 3 shows a typical electron path, assuming an electron of energy E_0 eV is fired into a homogeneous absorber of density ρ , and is traveling originally in the $+z$ direction. If it comes to rest at a certain depth R_z , we may call this the penetration depth or range R_z . It may be measured

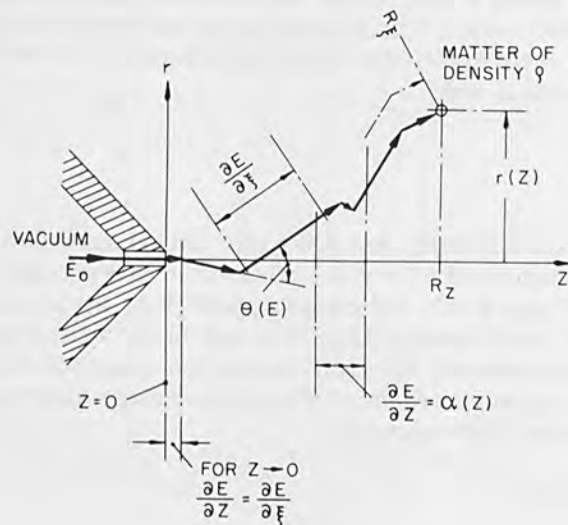


Fig. 3. Typical electron path and some quantities of interest. E_0 energy of the electron before entering the absorber. R_ξ length or range of the true (zigzag) path; R_z penetration depth or range; $\theta(E)$ scattering angle; $\partial E / \partial \xi$ energy loss per unit of true path; $\partial E / \partial z$ energy expended in a layer of unit thickness perpendicular to z .

in several ways as discussed later. The length of the true path may be denoted by R_ξ , and is of course larger than R_z ; R_ξ can only be measured on tracks in a cloud chamber or photographic emulsion. Scattering angles $\theta(E)$ are of interest and the probability distribution $\sigma(\theta) d\theta$ for scattering into a given angular range $\theta d\theta$. The energy loss $\partial E / \partial \xi$ along a section of the true path ξ is of interest; but this must be clearly distinguished from the energy expended in any one plane perpendicular to z and denoted $\partial E / \partial z = \alpha(z)$. Note that for small z values $\partial E / \partial z$ becomes identical with $\partial E / \partial \xi$ (because the electrons travel in the z direction), and this

allows us to measure $\partial E/\partial \xi$ directly [26]. We shall review what is known about the quantities mentioned above. Since scattering and energy loss are statistical processes, the values of R_z , etc., are of course subject to statistical fluctuations. To describe all processes fully would require knowledge of the directional flux density spectrum $\vec{\Sigma}_p(E, r, z)$ as a function of energy E at each point (r, z) , ($\vec{p}$ being a unit vector representing any direction of travel of the electrons). Σ is a sixth-order differential quantity [27]. Our present knowledge about this spectrum is extremely meager, as we shall see.

SCATTERING

In all experiments there are two basic situations which must be clearly distinguished. They are illustrated in Figs. 4a, b [28]. We may either have a thin but dense target (Fig. 4a) or a homogeneous one of lower density (Fig. 4b), and in each case have the same total mass between electron source and observation plane. In case (a) we measure essentially angular scatter distribution, in case (b) lateral beam spread.

Single Scattering

If the target in Fig. 4a is thin enough, most electrons will only be scattered once. Thereby they may or may not suffer from energy loss. Single scattering is very well investigated experimentally [28]-[35]. Figure 5 shows a typical angular distribution curve for the elastic and inelastic scattered fraction. By plotting intensity contours as a function of angle and energy loss in one plane [33], we can get the distribution charts shown in Figs. 6 and 7. Energy loss spectra show discrete lines [36]-[42].

Theoretical treatment of single scattering starts with the classical Rutherford formula. But for the scattering of an electron by an atom we have to consider the atom as a complete system. Figure 8 shows some results of different theories [43a]. In spite of much effort [43b], [44], [45] and various improvements on the theory, it is presently not more than a guide and the data we possess come primarily from experiments. A simple formula for the scattering cross sections cannot be given, but the following consideration is basic for the understanding of all electron scattering phenomena: It is essentially electron-electron scattering, and the number of electrons per gram of matter is nearly constant for all elements.

Therefore there is little dependence on atomic number z if cross sections are expressed in cm^2/g . The one notable exception is hydrogen.² Some further comments will be made in the section "Current Retained in a Beam."

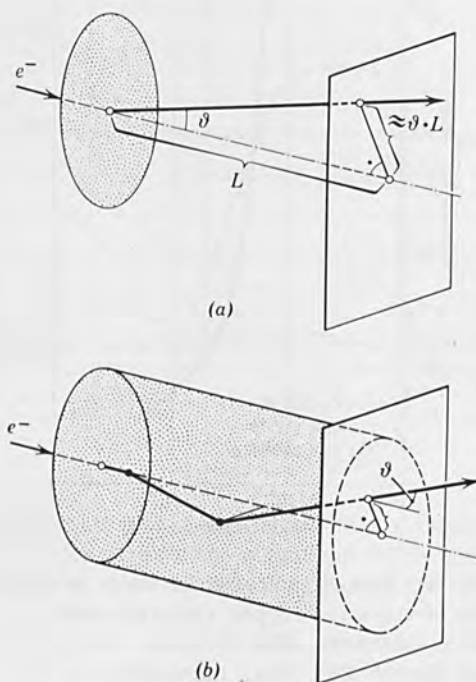


Fig. 4. The two basic situations in beam penetration experiments. (a) gives essentially angular scatter distribution, (b) lateral beam spread. After Jost and Kessler [28].

Plural Scattering

If knowledge about single-scattering is assumed, the scatter distribution after, say, ten scattering acts can in principle be obtained by purely mathematical treatment. The enormous amount of work in this direction has recently been reviewed by Scott [45]. As an example, for experimental results the measured angular distribu-

²This allows the determination of hydrogen content in hydrocarbons by means of beta-ray absorption [47].

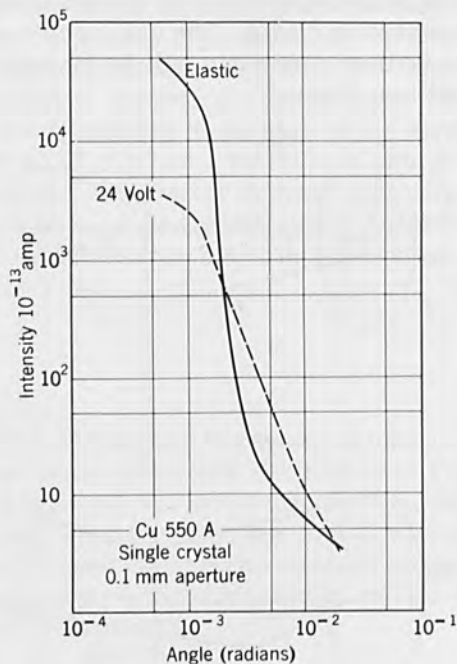


Fig. 5. Plot of scattered intensity as function of angle for copper single crystal of 550 Å thickness; after Simpson, McCraw, and Marton [33]. More information is contained in the newer type of plots of which Fig. 9 is an example.

tion for electrons with up to three units of energy loss is shown in Fig. 9. Another example is shown in Fig. 10 which gives the lateral beam spread (in an experiment according to Fig. 4b) for zero to forty scattering acts.

Scattering with Deep Penetration

Logically the next point in the discussion is scattering with the number of scattering acts going towards the infinite. But only special aspects of this process can presently be discussed. Neither theory nor experimental measurements are in any sense complete. The main difficulty arises from the fact that energy losses must be considered in addition to the angular scattering if we want to

come to any realistic results. The two factors just can no longer be treated separately. For this reason the dissipation of the beam energy shall be considered next.

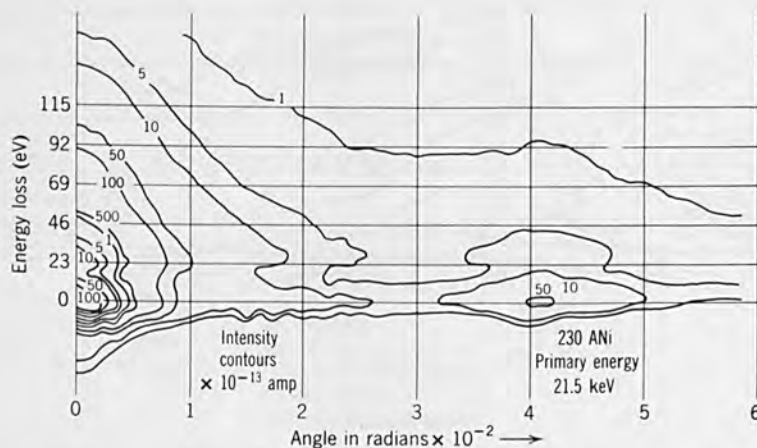


Fig. 6. Cartographic plot of the distribution of electrons scattered by 230 Å foil of nickel. Note the prominent diffraction maximum in both the elastic and characteristic loss peaks; after Simpson, McCraw, and Marton [33].

ENERGY DISSIPATION

Energy Loss $\partial E/\partial \xi$ along the True Path and True Range R_ξ

(Note re: Excitation Functions)

For $\partial E/\partial \xi$ we have the theory of Bethe [48] which was found to be in agreement with all measurements. In this case primary information comes from theory rather than from measurement. Extensive tables have been published [49], [50] giving $\partial E/\partial \xi$ as function of energy for many substances. The quantity $\partial E/\partial \xi$ is also called stopping power. Values for air obtained in an experiment according to Fig. 3 are shown in Fig. 22.

With the value $\partial E/\partial \xi$ known, the true range or total length of the track R_ξ can be obtained by integration. The above-mentioned tables also give R_ξ , and R_ξ for air and tungsten is included in Fig. 46.

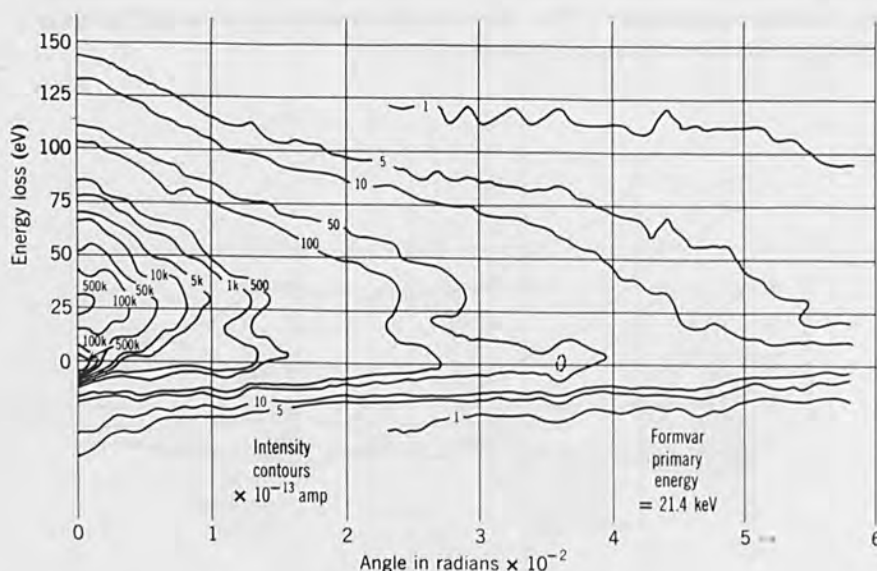


Fig. 7. Cartographic plot of the distribution of electrons scattered by Formvar. Note the similarity to the cartographic plots for metals except for the lack of prominent diffraction maxima; after Simpson, McCraw, and Marton [33].

For the better understanding of all energy transfer processes it seems indicated to say a few words about excitation and ionization processes which take up most of the energy of the beam particles. The important point to note is that these cross sections become nearly constant when the particle energy is large, compared with the threshold energy for the particular process. Near the threshold, however, the cross section can increase by factors of 3 to 10. This, it seems to me, is of importance in bringing about the sharply defined range and the sudden termination of the exponential attenuation process, as discussed later.

In Fig. 11, taken from the standard text on this subject [51], a few typical excitation functions are shown. In Fig. 12 evidence is presented for the role that inner shell or X-ray levels play [25], although their influence on the absorption curves, etc., may only be noticeable in special cases. (See also [36].)

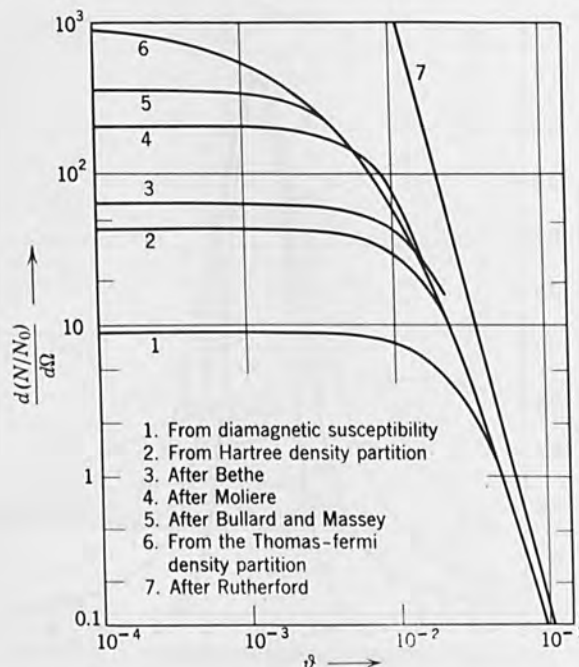


Fig. 8. Angular distribution of single scattered electrons from a carbon layer of 1×10^{-6} g/cm² according to various theories; after Lenz [43].

Energy Dissipation $\partial E/\partial z$ (Collectively); Depth-Dose Functions and "Partial" Depth-Dose Function; Residual Dose; Practical Range R_E

The energy absorbed in any stratum of an absorber can indirectly be determined from the difference between the beam energy and the energy of the fraction of electrons passing through and being backscattered. This has been done in a very complete series of experiments by Gentner [25] for electrons of 25.6 keV passing through a sectioned aluminum absorber. A detailed description will follow in the Section on particle dissipation. Kanter [59] and Makhov [65] have also made some calculations of this kind based on their measurements. But there exist more direct experiments.

Usually the ionization in a gas is taken as a measure for the energy dissipation, but we can just as well use the luminosity (Fig. 1b) which, with advantage, can be observed from some dis-

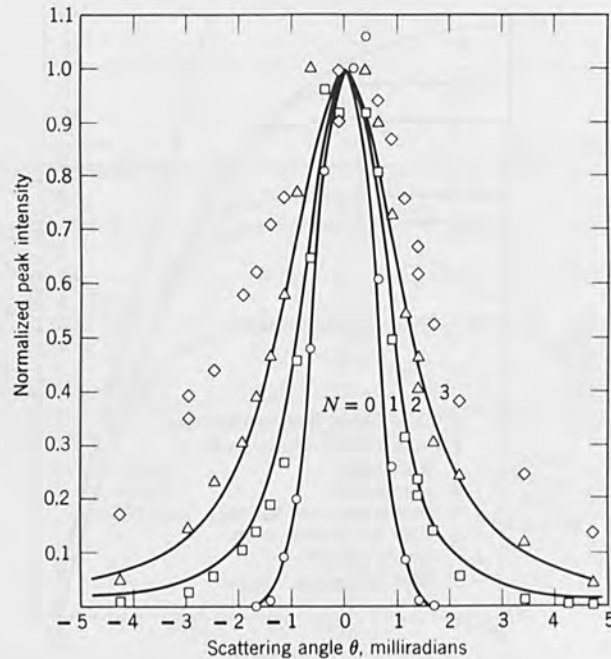


Fig. 9. Normalized angular distributions of the zero-loss, first-loss, second-loss, and third-loss peaks. Points are experimental peak heights. Smooth zero-loss curve is fitted to zero-loss points. First- and second-loss curves are computed from the zero-loss and first-loss points, respectively, by folding with the Bethe-Ferrell cross section, with $\theta_E = 0.370 \times 10^{-3}$ rad, corresponding to $\Delta E = 14.8$ eV; after Marton et al. [46].

tance. The true measure for energy dissipation, namely temperature (enthalpy) rise, would be hard to determine.

Figure 13 shows Gruen's apparatus [26] for measuring $\partial E / \partial z = \alpha(z)$ in a homogeneous absorber by measuring the light output through a narrow slit Δz as a function of z . A similar principle, using a thin fluorescent foil in a nonfluorescent glass block, was used by Olde and Brannen [52] to measure the energy dissipation of 1.5 MeV electrons in a solid absorber.

Figure 14 shows some curves measured by Gruen for various values of beam energy E_0 , and air as the absorber. Since the ab-

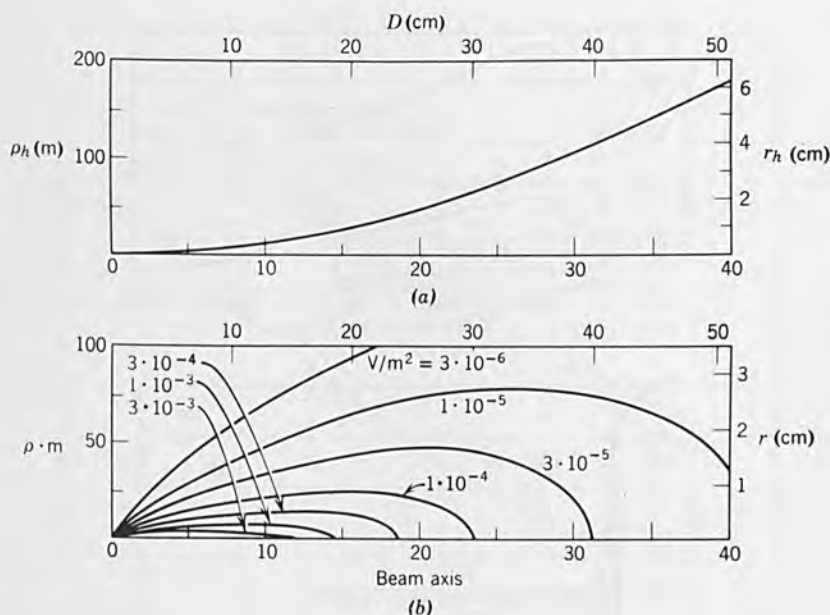


Fig. 10. Spread of a narrow electron beam entering from the left a space filled with Kr. Reduced scales on the lower and left margin expresses the density of the gas layer in terms of the mean collision number m ; the beam diameters are expressed in units of $\rho \cdot m$ (calculated with $R_L = 0.214A$). Special scales on top and right are for 100 keV electrons and Kr gas of 0.98 torr (0°C). In (a) the spread is shown by the "half-value width," meaning the radius which contains half of the primary intensity. In (b) the spread is shown by lines of equal current density, $V(\rho)/m^2$ being a (relative) measure for current density. From Jost and Kessler [28].

sorbed energy is customarily called radiation "dose," the functions $\alpha(z, E_0)$ are called depth-dose functions. The value of α for $z = 0$ is not zero but equal to $\partial E / \partial \xi$ as already mentioned (see also Fig. 3). The maximum of the energy dissipation lies below the surface and is followed by a rather straight descend of the $\alpha(z)$ curve. The extrapolation of this straight section to $\alpha = 0$ defines a value $z = R_E$, which we may call the practical range $R_E(E_0)$. The subscript E shall indicate that it was obtained from energy measurements, because there are also other definitions for the range to be discussed. The backscatter tail for $z < 0$ as shown in Fig. 14 could also be measured with the apparatus of Fig. 13 because the

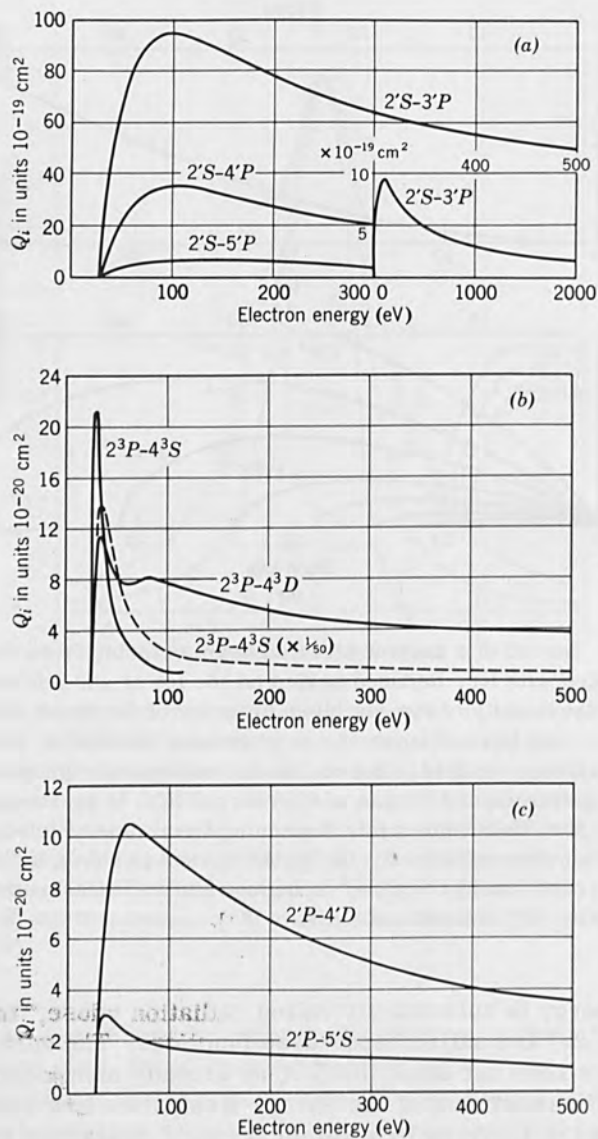


Fig. 11. Observed excitation functions obtained by Lees for He lines: (a) Three lines of the same series, λ 5015 (2^1S-3^1P), λ 3964 (2^1S-4^1P), λ 3614 (2^1S-5^1P). (b) λ 3888 (2^3S-3^3P), λ 4713 (2^3P-4^3S), λ 4472 (2^3P-4^3D). (c) λ 4922 (2^1P-4^1D), λ 4438 (2^1P-5^1S). From Massey and Burhop [51].

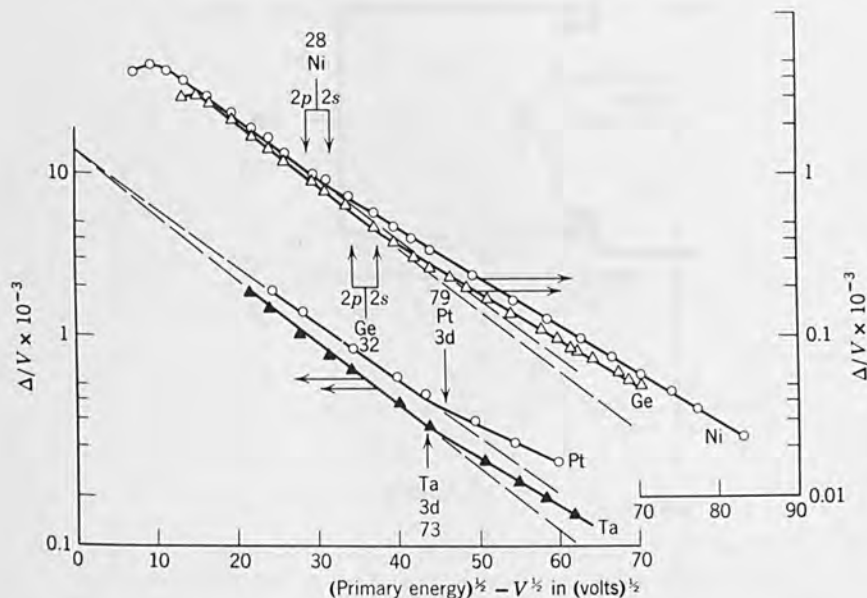


Fig. 12. Secondary electron yield as function of energy of the primary beam particle; from E. J. Sternglass [25].

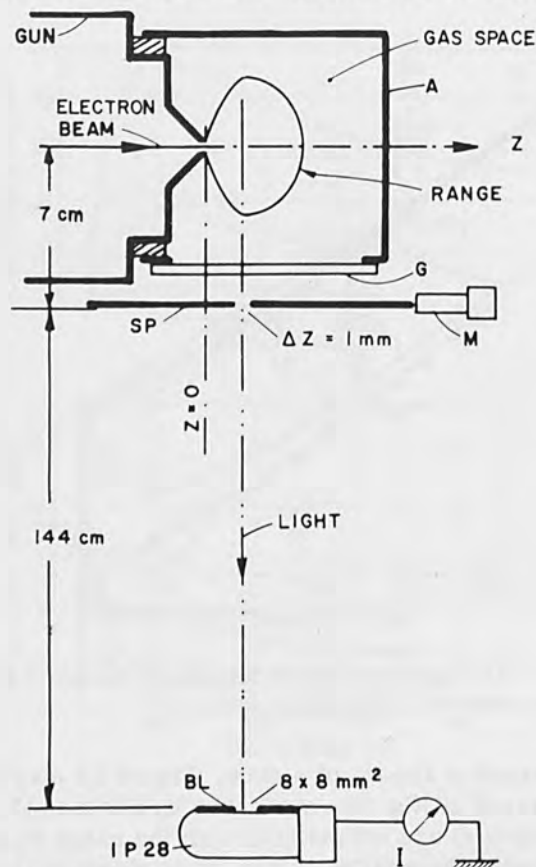
electrons emerged at the tip of a cone. Figure 14 may be compared with curve *E* in Fig. 29a measured by Gentner [25].

If the curves $\alpha(z)$ are normalized with the range R_E as unit of penetration length, the picture of Fig. 15 is obtained [26].

Evidently the shape of the normalized $\lambda(z/R)$ is little dependent upon the electron energy E_0 . There is some reason to suspect that this shape depends upon the atomic number Z of the absorber, as discussed later.

Integrating over the differential distribution function α or λ one can obtain a Residual-Dose-Function D_r , shown in Fig. 16. It gives the fraction of energy transmitted beyond a certain depth. It can be compared with the function η , discussed later, which gives the fractional current transmission, and which has obviously a different physical meaning. The extrapolation of the straight part gives a "range" $z/R < z/R_E$. This may explain the difference of curves b and c in Fig. 46.

For the energy dissipation with depth, a well-developed mathematical treatment exists, the theory of Spencer and Fano [53]-[55]. Figure 17 shows the good agreement with experiment; both curves are absolute, having been normalized to unit electron source



strength. Some of Spencer's curves for other energies are shown in Fig. 18.

Neither the theory of Spencer nor the experiments of Gruen of Olde and Brannen can tell us anything about the electron energy spectrum at a given depth z , nor do the depth-dose curves as such contain any information about the lateral distribution of the energy within a plane perpendicular to the z axis.

To get some information about the electron energy as a function of depth, we can place at various depths z a detector foil which has a well-defined threshold energy for detection. The only experiment of this kind seems to be the one of Castaing and Des-camps [8], where the X-ray intensity from thin tracer layers of copper, etc., imbedded in an aluminum absorber, was measured.

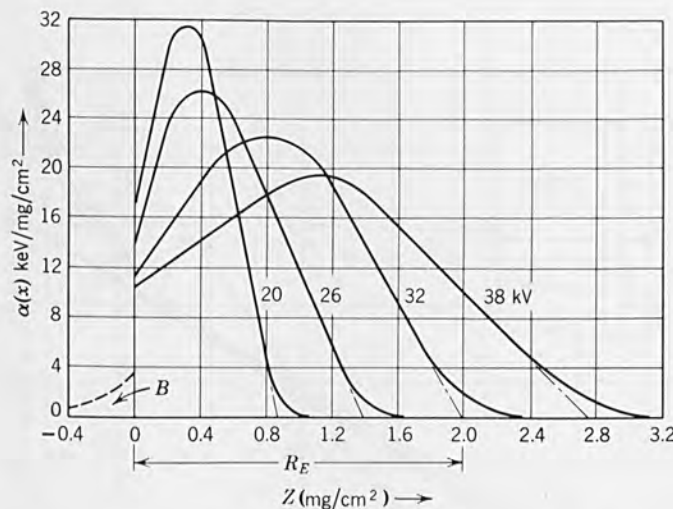


Fig. 14. Depth-dose functions as measured by Gruen in air; note the definition of the range R_E and the backscatter part B .

The results are shown in Fig. 19. We may call such curves (measured with a definite energy cutoff of the detector) "partial" depth dose functions.

Some information about the lateral distribution of energy can be obtained from Fig. 1b if the scattering field is photographed and if intensity contours for the luminescence are determined as shown in Fig. 20. From this plot we can, in principle, get the dose distribution in r and z by solving the related Abel-type integral equation, but no one seems to have done this.

The values for range R_E and energy loss $\partial E/\partial \xi$ resulting from the measurements of Figs. 13 and 14 are shown in Figs. 21 and 22. The range values can be represented by the formula

$$R_E = 4.57 \times 10^{-3} E_0^{1.75} \text{ mg/cm}^2 \quad (1)$$

(E_0 in keV)

for air; $5 < E_0 < 100$ keV

or
$$R_E = 814 E_0^{1.75} \text{ mg/cm}^2 \quad (1a)$$

(E_0 in MeV)

for air; $0.005 < E_0 < 0.1$ MeV

Extrapolated range values such as R in Fig. 14 are often called "practical range," in contrast to the values we get from the final

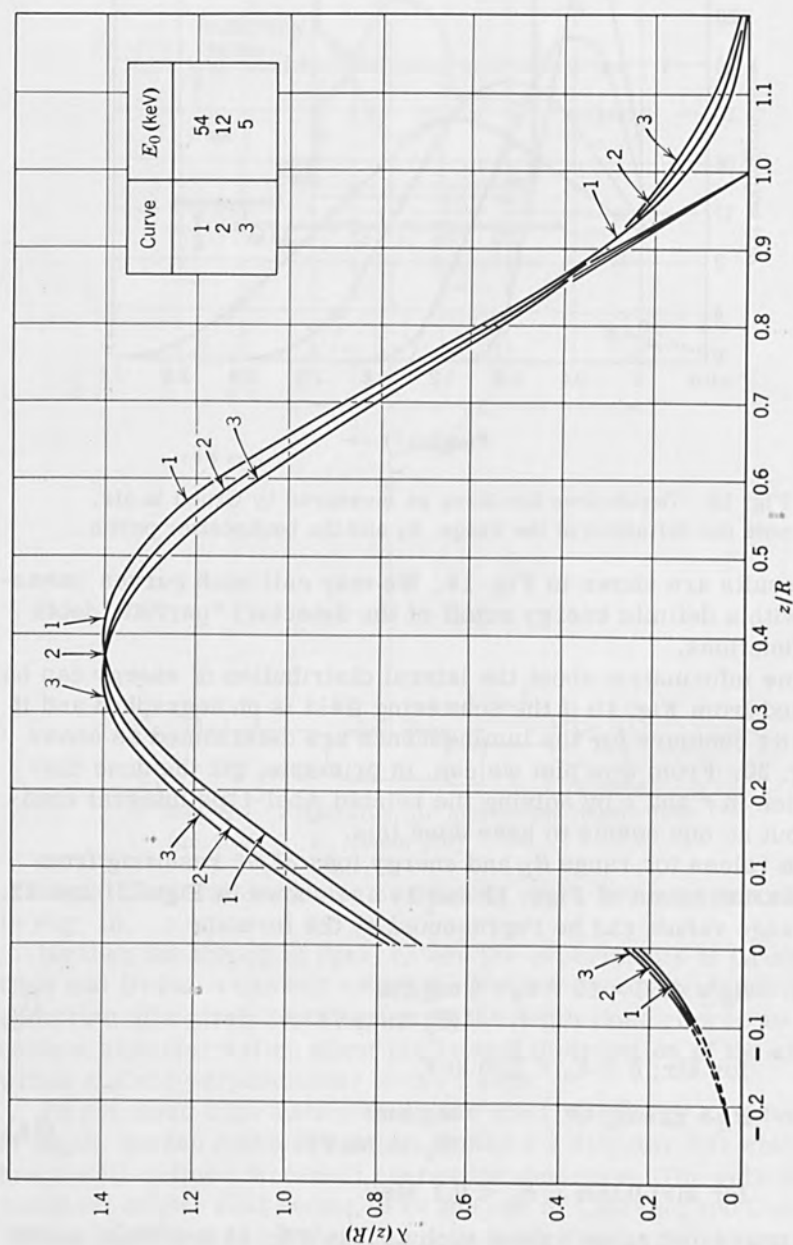


Fig. 15. Normalized depth-dose functions $\lambda(z/R) = (R/E_0) a(z)$ for air; A. E. Gruen [26].

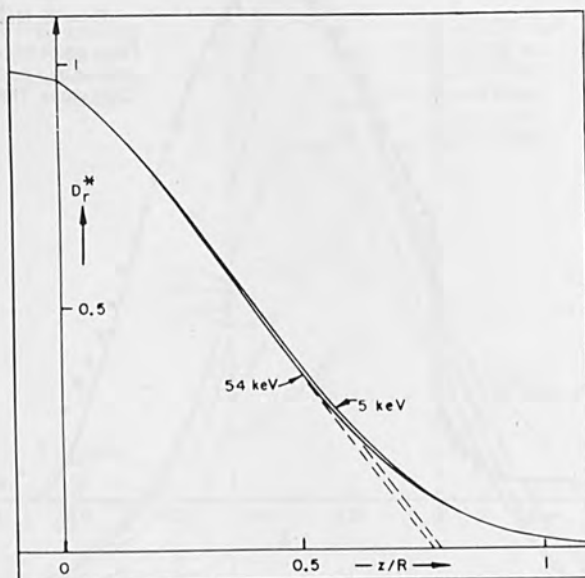


Fig. 16. Normalized residual dose function

$$D_r^*(z/R) = 1 - \int_{-\infty}^{z/R} (z/R) d(z/R)$$

convergence of the intensity curves with the z axis, values which are sometimes called "maximum ranges." This convergence, and therefore the maximum ranges, are poorly defined.

Range-energy relations for all energies have been reviewed by Katz and Penfold [58]. And although values for R_E , R_I , or other definitions, are not treated separately, we will list the relations already here. We find

$$R = 412 E_0 (1.265 - 0.0954 \ln E_0) \quad \text{mg/cm}^2 \quad (2)$$

for E_0 in MeV; $E_0 < 3$ MeV

$$R = 530 E_0 - 106 \quad \text{mg/cm}^2 \quad (3)$$

for E_0 in MeV; $2.5 < E_0 < 20$ MeV

These values are plotted in Fig. 46.

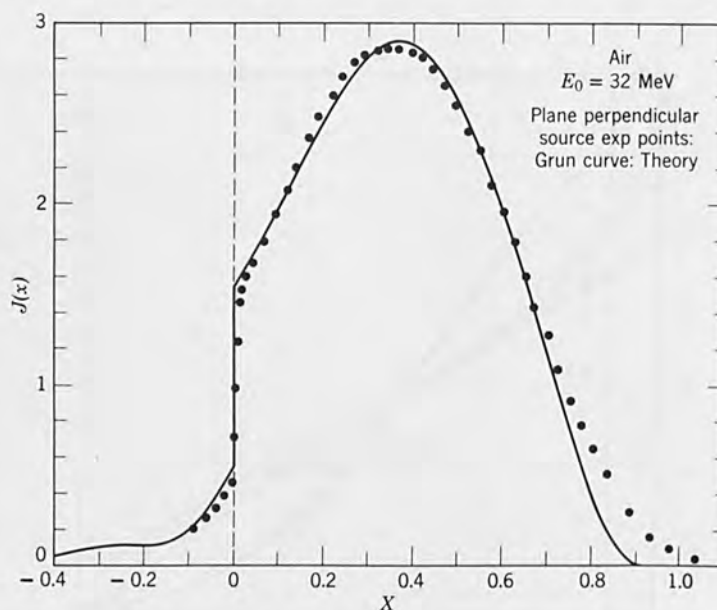


Fig. 17. Depth-dose distribution as derived theoretically by L. V. Spencer [54], solid line, compared with the experimental results of Gruen [26].

PARTICLE DISSIPATION I

Transmitted Current or Fraction of Transmitted Electrons η ;
Practical Range R_I , threshold Energy for Transmission E_c

In the earliest experiments about the passage of electron beams through matter, the current going through a thin foil was measured. Although such measurements can be made relatively easily and accurately, the interpretation of the results is a different matter.

Figure 23 shows the apparatus of Kanter [59] for measuring the fraction η of electrons of energy E_0 going through a thin film of thickness z , and also the angular distribution of these electrons. It is convenient to measure $I/I_0 = \eta(E_0)$ for constant z . Figure 24 shows typical transmission curves as function of energy E_0 . The straight part may be extrapolated to $\eta = 0$; the intersection determines a "critical energy" E_c for transmission. With a sufficient number of curves for various z , we may replot the results in a graph $\eta(z)$ for $E_0 = \text{const}$. This was done in Fig. 25. Here the inter-

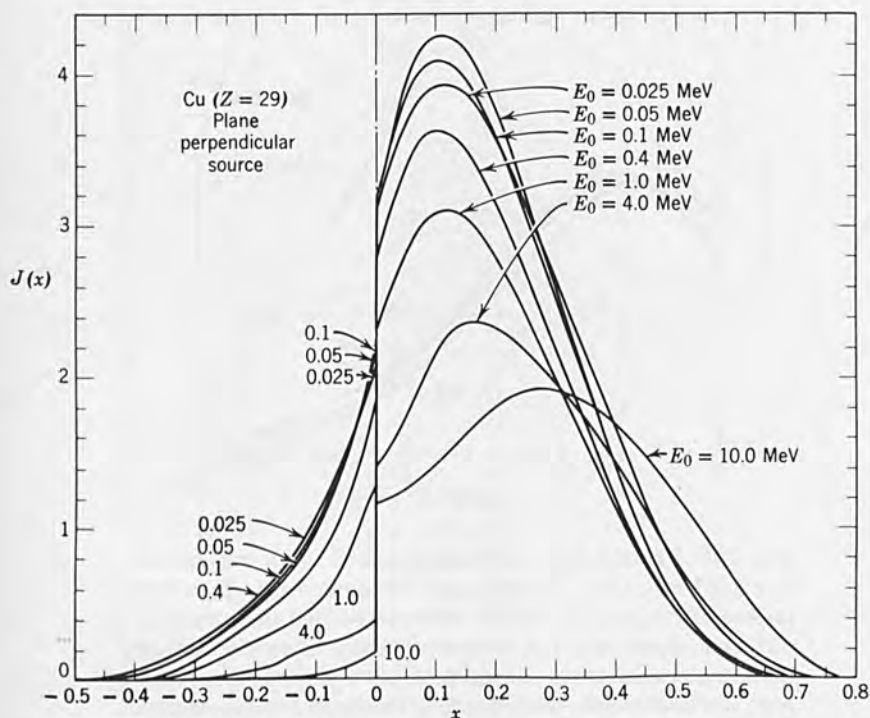


Fig. 18. Depth-dose curves as function of energy; Spencer's theory [54].

section of the extrapolated straight section with $\eta = 0$ gives another value of the practical range of electrons with energy E_0 . We call this range value R_I , the subscript I standing for "by current measurement." Since E_c and R_I are extrapolated values, the relationship depends upon the shape of the transmission curve and has not yet been formulated analytically, but $R_I(E_0)$ and $E_c(R_I)$ are not very different.

A summary of range values R_I is shown in Fig. 26. Results of many workers have confirmed that the range depends little, if at all, upon the atomic number of the absorber.

The shape of the fractional transmission function $\eta(z)$ or $\eta(E_0)$ seems to depend upon atomic number. A set of curves obtained by Kanter for low values of E_0 is shown in Fig. 27. The effect of atomic number on transmission curves in the energy range above 20 keV was noted in the earliest studies. However, Schonland [62] has shown that this effect appears much less pronounced if, for the

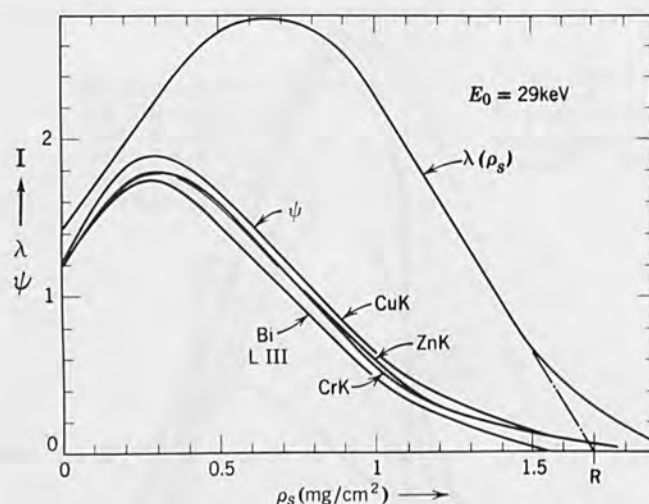


Fig. 19. Partial depth-dose functions ψ for X-ray excitation (redrawn from Castaing and Descamps [8]). Thin tracer layers of Cr, Cu, Zn, and Bi were imbedded in aluminum at different depth, $\psi = 1$ is said to correspond to the intensity from the tracer if suspended free in vacuum. For comparison, the total depth-dose curve λ is shown for the same probe voltage of $E_0 = 29$ keV; intensity values are not comparable.

initial current, not the beam current but beam current minus backscatter current is taken as I_0 . I believe that if the practical rather than the physical significance of the transmission curves is considered, then too much emphasis tends to be attached to it, as I hope to show here.

Besides the transmitted fraction, there are some electrons scattered backwards and some get trapped in the thin absorber film itself. All three fractions as a function of atomic number are shown in Fig. 28 for the special case that the electron energy E_0 is twice the critical energy E_c needed for onset of transmission. How these relationships can be used in an analytical electron microprobe system is illustrated in Fig. 29.

The fraction of electrons absorbed in a foil is, of course, related to the depth dose over a certain range z . But the relationship is not as straightforward as it may seem. For instance, the backscatter fraction from $z > z_d$, if z_d is the foil thickness, is missing. Therefore the exact relationship between the fractional transmission η

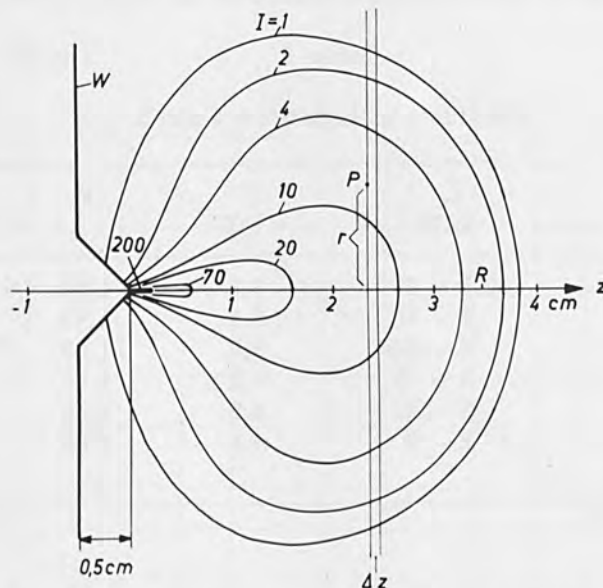


Fig. 20. Energy absorption for 32 keV electrons in air of $p = 363$ torr, 20°C . The curves are lines of constant luminous intensity obtained by densitometric measurements on a photograph similar to Fig. 1b. From A. E. Gruen [26].

and the residual dose D_r is not a simple one.

Gentner's measurements also reveal this relationship, as shown in Fig. 29a. Unfortunately those measurements were only made with one energy value and an aluminum absorber.

Normalized Functions $\eta(\chi, E_0)$

The first to consider normalization of transmission functions was apparently Seliger [64] followed by Makhov [65] and Kanter [59]. Whereas Gruen [26] showed that depth-dose functions can be normalized. In Fig. 30 the universal transmission function of Makhov is shown. For normalization he has chosen a foil thickness $\chi = 1$ at which η has fallen to $1/e$ of I_0 . (The special advantage he claims to gain by this choice is not quite clear.) He then finds for χ (which is obviously related to R_I).

$$\chi = CE_0^n$$

with values for C and n different absorbers as listed in Table I.

Table I
Constants of Equations 4 and 5

Substance	E_0 keV	C $\times 10^3$	n	p
Al	2.5 - 27	3.8	1.68	2
Si	2 - 20	3.4	1.65	2
Cu	2 - 20	5.8	1.53	2
Ge	2 - 18	6.5	1.47	2
Bi	3 - 20	4.2	1.44	1
Al ₂ O ₃	1 - 9	4.6	1.65	2
Pb				1

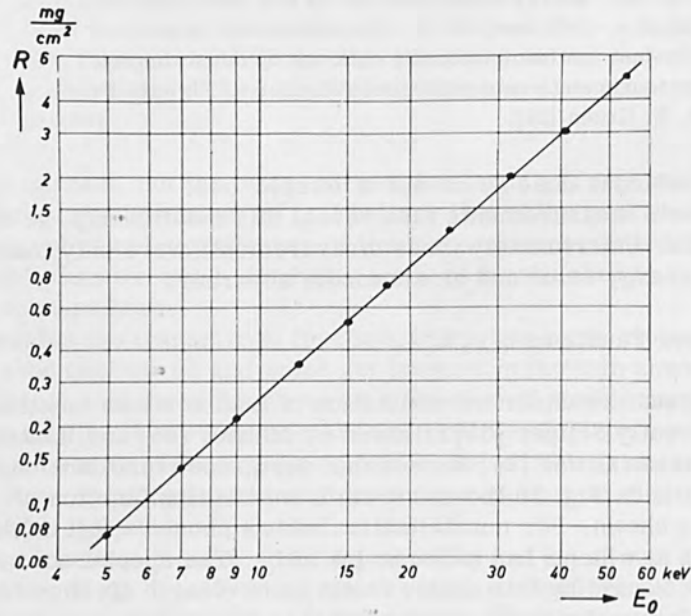


Fig. 21. Range R_E of electrons of energy E_0 measured and defined as shown in Figs. 13 and 14; A. E. Gruen [26].

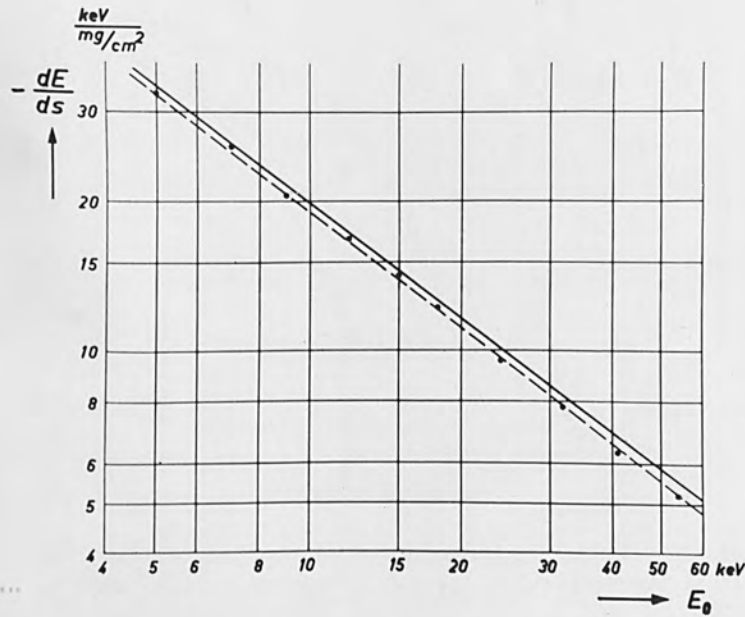


Fig. 22. Stopping power $\partial E / \partial \xi$ as function of E_0 measured and defined as shown in Figs. 3 and 13, A. E. Gruen [26].

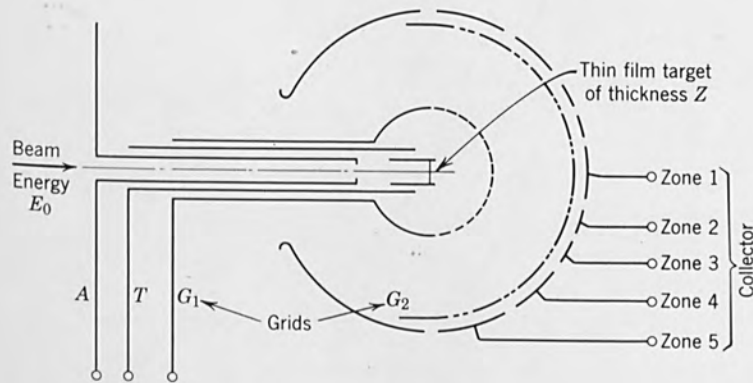


Fig. 23. Kanter's apparatus [59] for measuring fractional transmission curves η , and angular distribution of transmitted electrons, schematic.

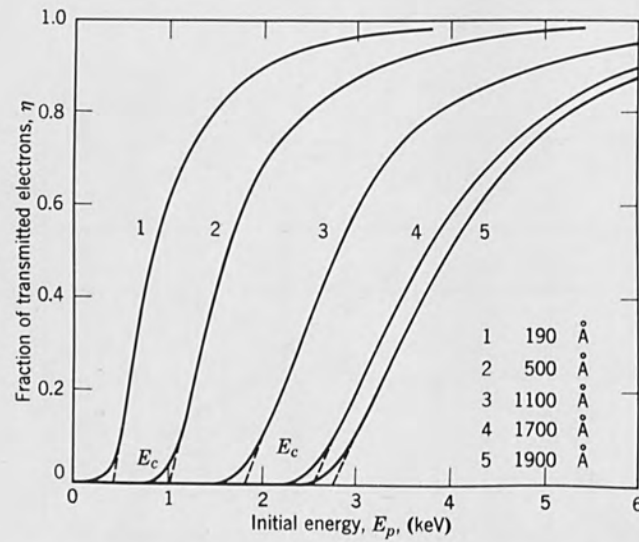


Fig. 24. Fractional transmission η as function of energy E_0 for carbon films of various thicknesses, from Kanter [59].

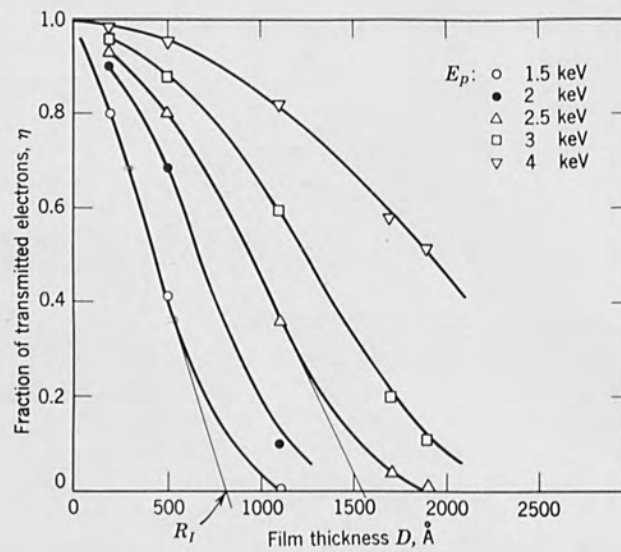


Fig. 25. Fractional transmission η as function of film thickness $z = D$ for various energies E_0 , from Kanter [59].

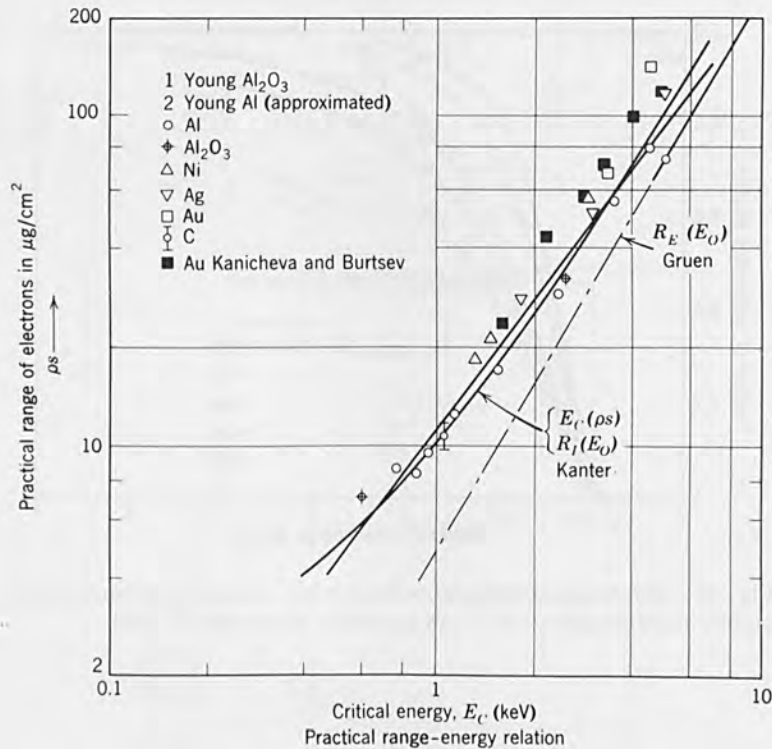


Fig. 26. Range-energy data from Kanter and others [60], [61] for $E_0 < 10$ keV; adapted from [59].

Finally he expresses the experimental results for the transmission by

$$\eta(z, E_0) = \exp \left[- \left(\frac{z}{C E_0^n} \right)^p \right] \quad (5)$$

[after Makhov]

with $p = 2$ for all measured substances except Bi and Pb where it is $p = 1$. This is hard to understand and can perhaps be explained by reference to Fig. 37, discussed later. Differentiating the curve in Fig. 30, or equation 5, gives the differential transmission curves of Fig. 31. As already mentioned, such differential curves are not representing particle distribution in a homogeneous absorber and are not directly related to the depth-dose curves. Figure 29a, derived from a more sophisticated experiment, is an exception.

Range values R_I based on transmitted current measurements seem to be shorter than values R_E based on energy measurements, as noted by Agu, Burdett, and Matsukawa [66]. There are certainly

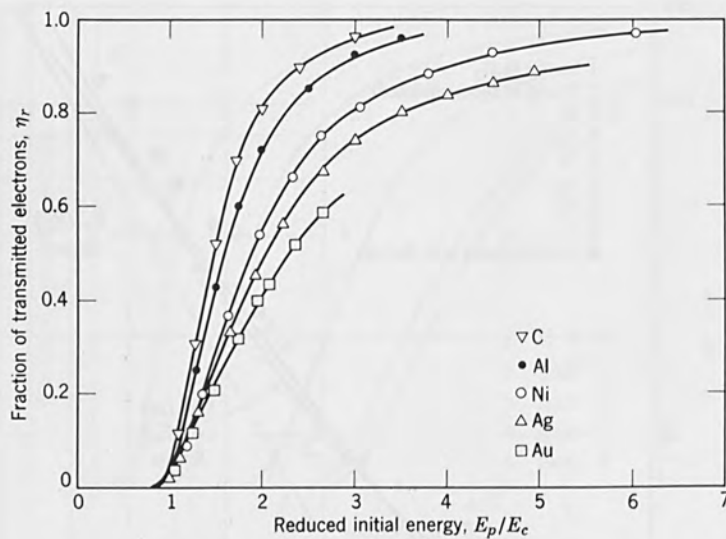


Fig. 27. Normalized transmission curves $\eta (E_0/E_c)$ as function of atomic number and $z = D > 15 \mu\text{g}/\text{cm}^2$; from Kanter [59].

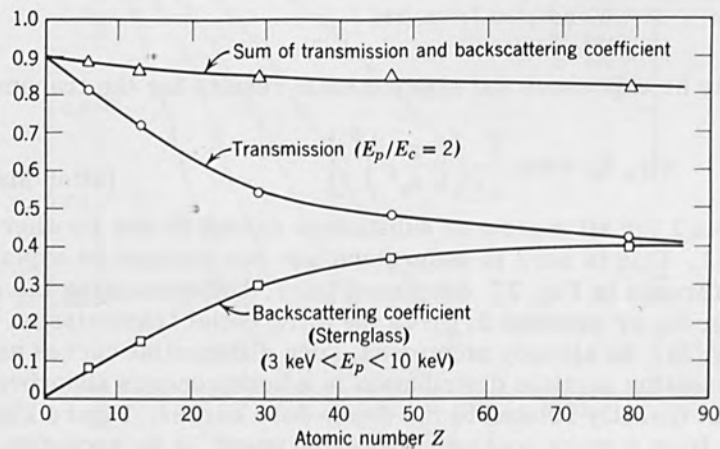


Fig. 28. Relation between transmission and backscattering coefficients as function of atomic number at $E_0/E_c = 2$; [59], [61].

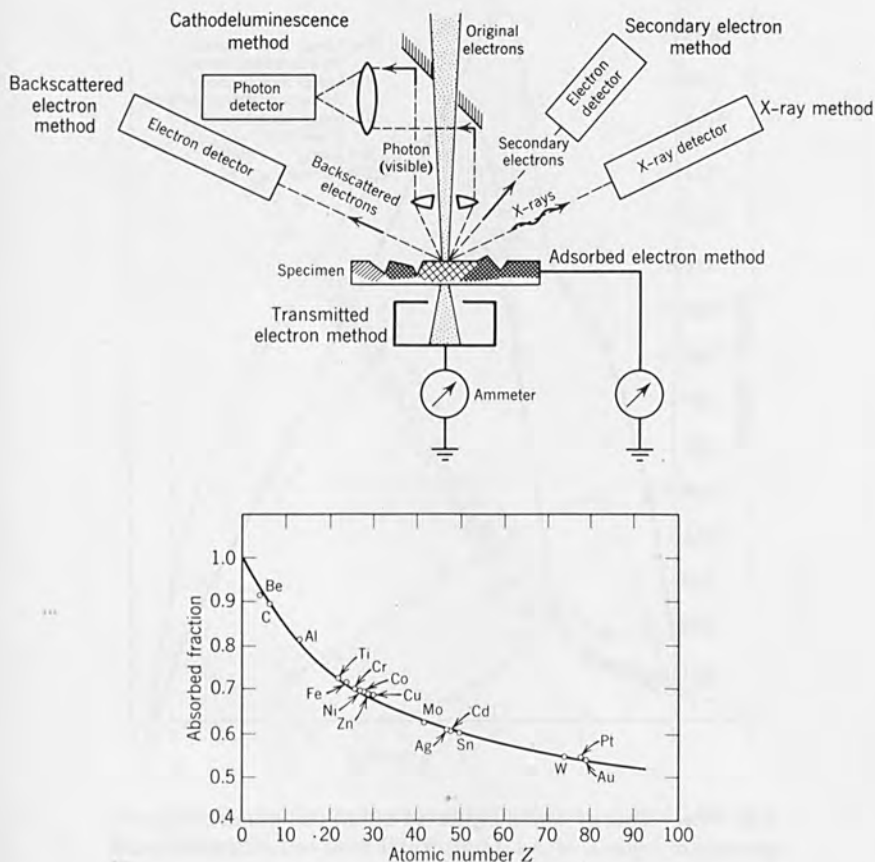


Fig. 29. Example of utilization of electron absorption, transmission, and backscatter in an analytical electron microprobe system; JEOL [63].

discrepancies in the presently available data, as a look at Fig. 46 will show. Experimental information is too incomplete to suggest how they may be resolved.

Most Probable Scattering Angle of Transmitted Electrons

Recent measurements of the angular distribution of the electrons [59] coming through foils of various atomic number are shown in Fig. 32. Similar results have been obtained earlier although not

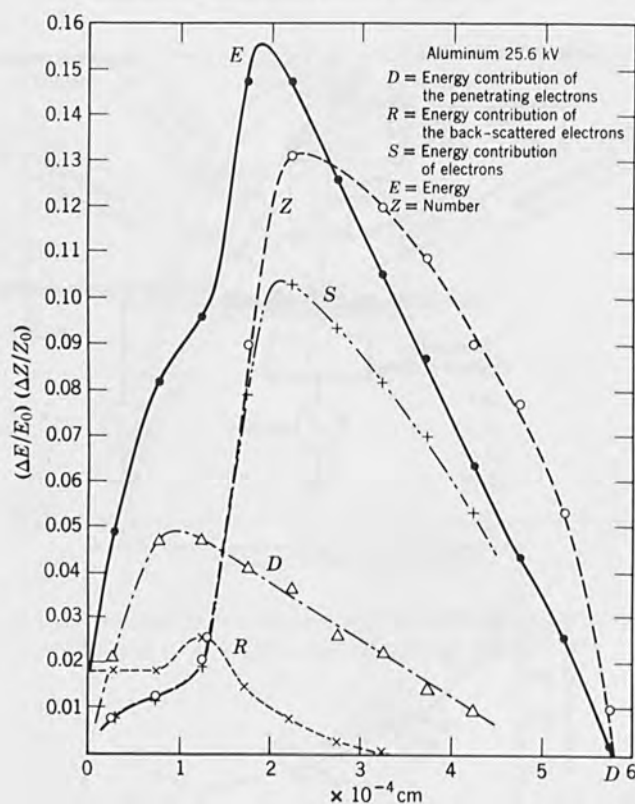


Fig. 29a. Energy and number of electrons absorbed in aluminum layers of $\Delta z = 0.5 \mu$ thickness but situated inside a homogeneous, thick aluminum absorber at various depth z (see original paper for details). Curve E = energy; curve Z = number; D = contribution of energy from electrons passing through; R = contribution from electrons backscattered from deeper layers; S = contribution from electrons stuck within Δz ; Gentner [25].

as complete. The most probable scattering angle approaches 45 degrees for z larger than half the range. The superficial interpretation which has been given to this fact has probably caused most of the confusion which we find when transmission and absorption "laws" are discussed. It has been said that for large-enough z , a state of "complete diffusion" is reached where the angular distribution no longer changes. There is no experimental support for this assertion. As for the correct interpretation of the foil experi-

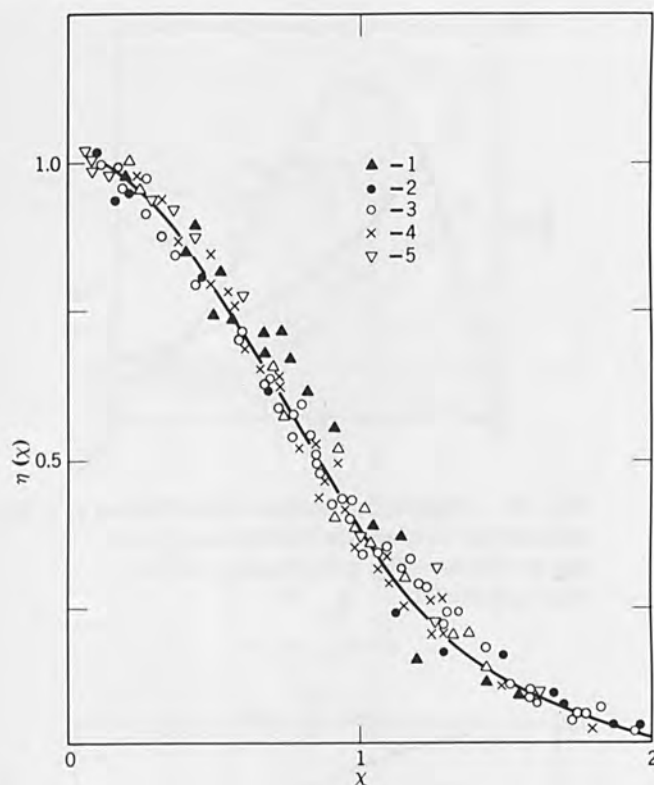


Fig. 30. Universal transmission function $\eta(\chi)$ of Makhov [65]; 1 Al, 2 Si, 3 Cu, 4 Ge, 5 Al_2O_3 ;

ments, we may compare the two situations shown in Fig. 33. A beam of diameter D small against the range R passes through an infinite plate of thickness $1/2 R$ or through a semisphere of radius $1/2 R$. Surely, the angular distribution of the electrons coming through in each case is different. It would not make sense to argue that one experiment is correct and the other is not. Any "significant" experiment of this kind would have to obey certain scaling laws, as used in similitude mechanics. How such scaling laws could be formulated for electron transmission is an open question.

Detailed measurements of the angular distribution and the energy spectrum of the scattered electrons under conditions approaching those inside a homogeneous absorber were made in the aforementioned study of Gentner. An aluminum absorber composed of two parts with a thin gap at a variable depth z was used. Electrons

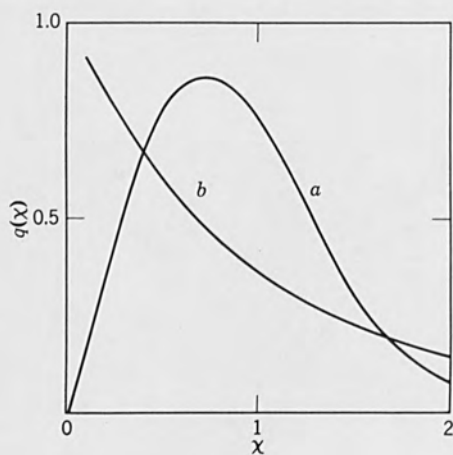


Fig. 31. Differential standard distribution function for transmitted electrons according to Makhov [65]; a for Al, Si, Cu, Ge, Al_2O_3 ; b for Bi;

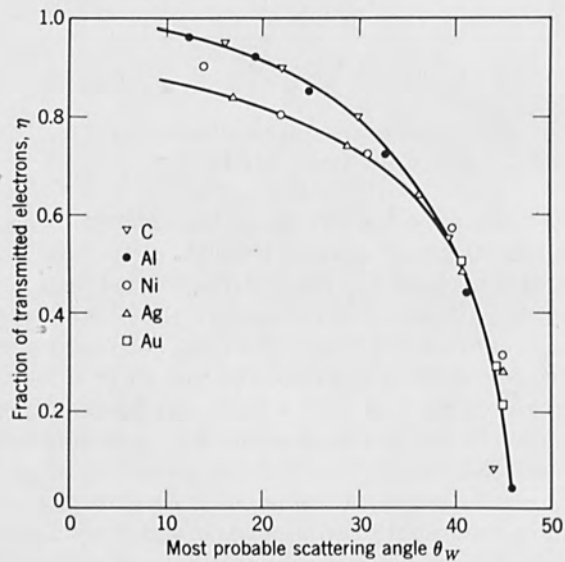


Fig. 32. Angular distribution of transmitted fraction of electrons; Kanter [59].

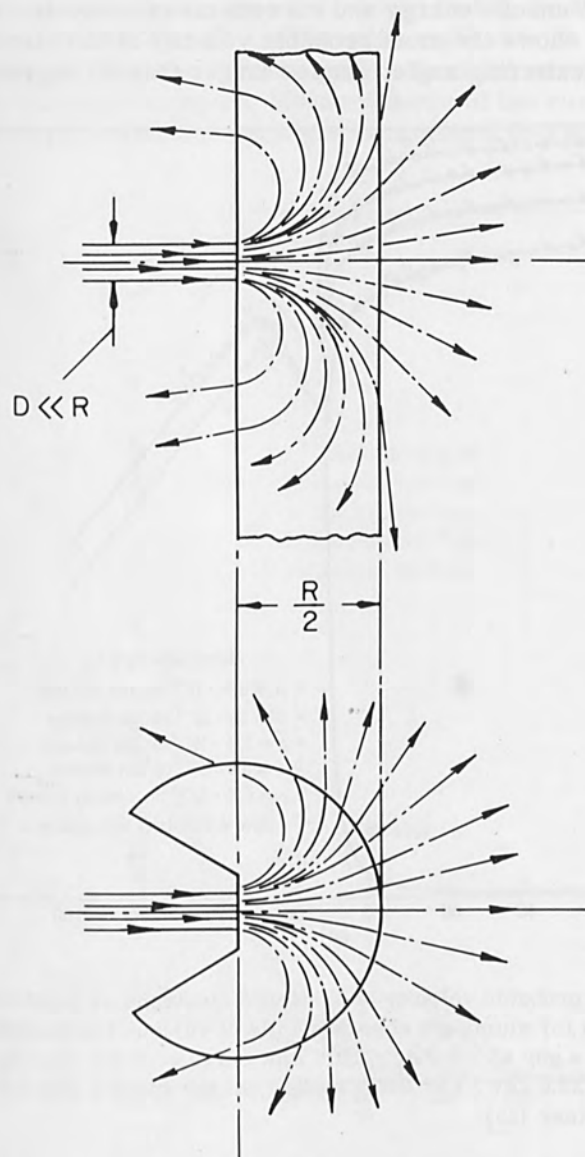


Fig. 33. Most probable scattering angle and considerations of similitude mechanics.

coming through this gap and representing scattering angles from about 80 to 100 degrees could be detected in addition to transmitted and back-scattered electrons. A magnetic spectrometer was used for simultaneous energy and current measurements.

Figure 33a shows the most probable velocity of the electrons as a function of scattering angle. Angles larger than 90 degrees de-

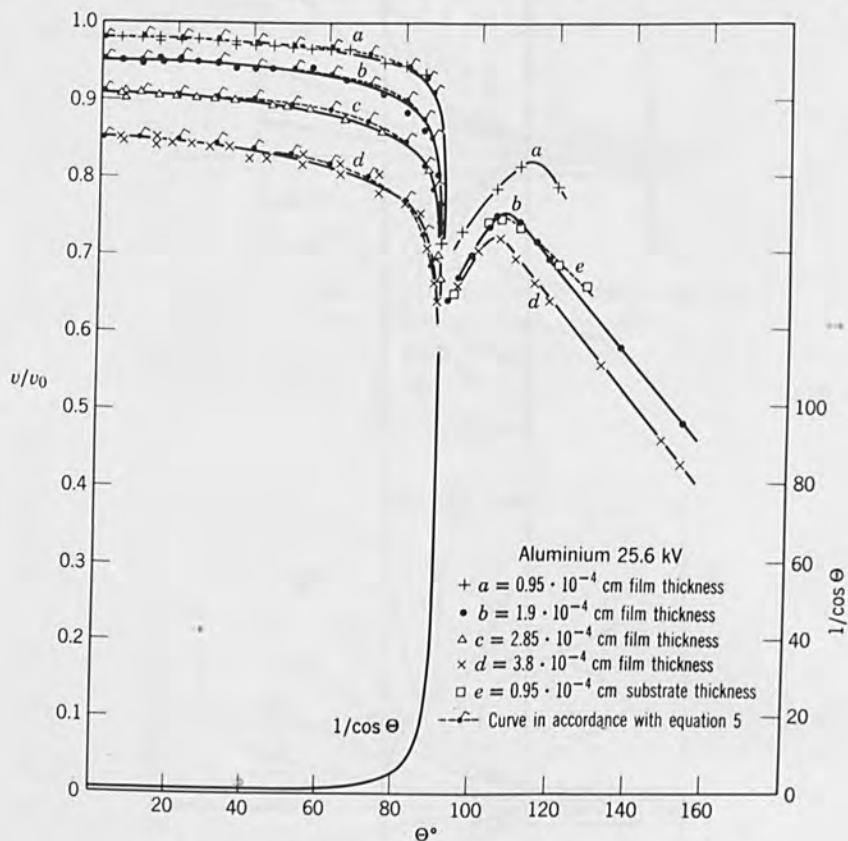


Fig. 33a. Most probable velocity of scattered electrons as function of scattering angle θ (i) for aluminum absorber foils of various thicknesses, curves a to d ; (ii) from a gap at $z = 0.95 \cdot 10^{-4}$ cm, curve e . V_0 = velocity corresponding to $E_0 = 25.6$ keV. For the complete energy spectra see the original paper of K. Gentner [25].

note backscattering. The high-energy loss for the electrons scattered close to 90 degrees is, as explained by Gentner, due to the

rather long path these electrons must have traveled nearly parallel to the absorber surface. Figure 33 would exemplify the situation. In a homogeneous absorber the 90 degree direction is in no way a preferred one. I suspect we would find there a smooth transition from the left side ($\Theta < 60^\circ$) to the right side ($\Theta > 130^\circ$) of the curves of Fig. 33a. Similar curves for total energy can be found in the original paper. Measurements of the current density as function of scattering angle are reproduced in Fig. 33b. The

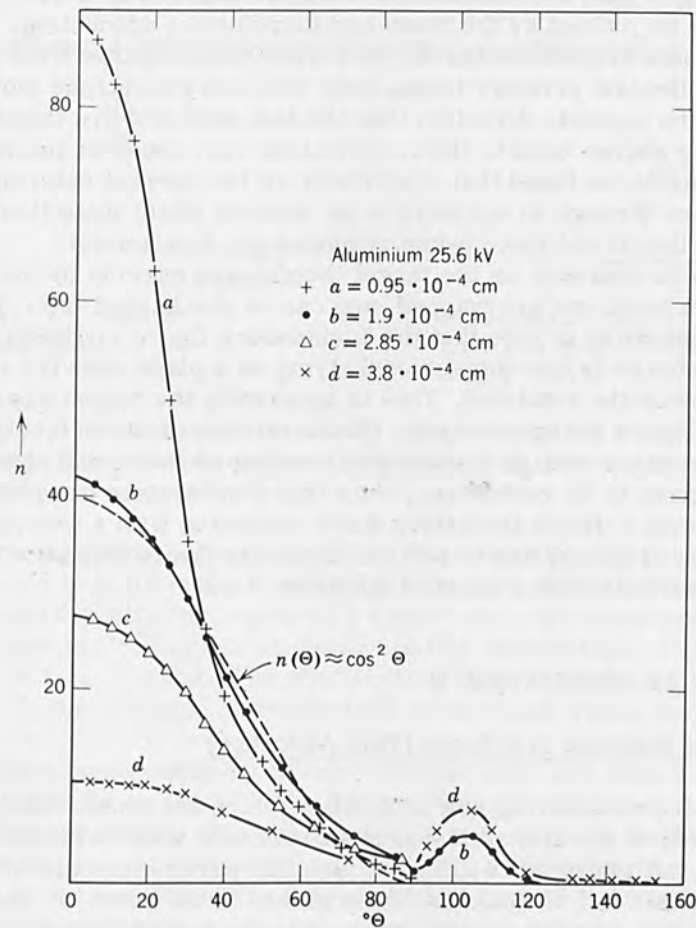


Fig. 33b. Current density n of scattered electrons as function of scattering angle Θ for various absorber thicknesses; Gentner [25].

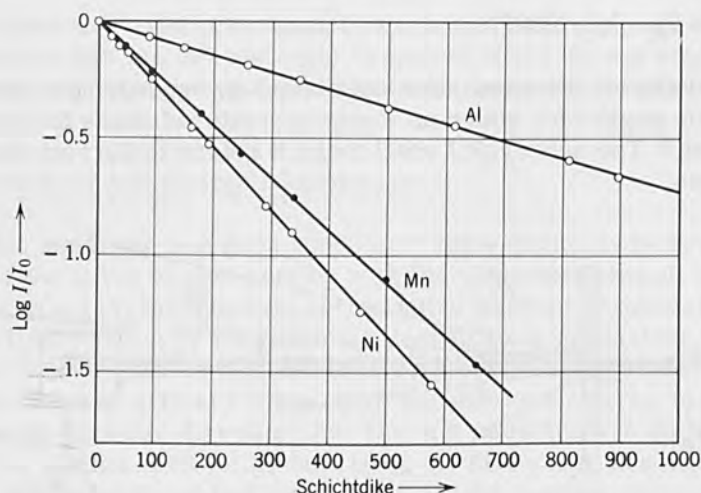


Fig. 35. Measurements by Reimer [69a] on the attenuation of an 80-keV electron beam by thin homogeneous films of Ni, Mn, and Al in a geometry as shown in Fig. 34.

coefficient per unit atom (not per unit weight) is considered, the curves of Fig. 35b are obtained, which are straight lines. Consequently we get for α_I

$$\alpha_I = 2.303 \frac{b}{A} Z^a \quad \text{cm}^2/\mu\text{g}$$

with the following coefficients a and b

Aperture		40 kV		60 kV		80 kV		100 kV	
μ	Radians	a	b	a	b	a	b	a	b
20	3.7×10^{-3}	1.08	.083	1.11	.054	1.15	.038	1.18	.030
50	9.3×10^{-3}	1.19	.048	1.20	.031	1.25	.023	1.28	.018
100	18.5×10^{-3}	1.23	.034	1.30	.019	1.34	.013	1.38	.009

(b in $\text{cm}^2/\mu\text{g}$).

Only hydrogen (and possibly helium) is not covered by this relationship.

Reimer has also analyzed how the exponential law must be modified for rough or inhomogeneous foils such as the ones shown in Fig. 36. Anomalous transmission due to diffraction effects has been discussed by Heidenreich [70]. What we measure when a thin

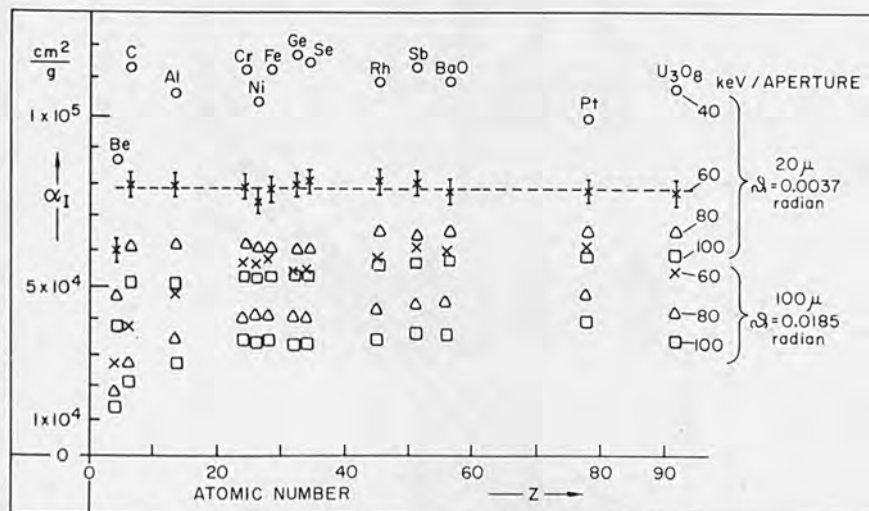


Fig. 35a. Measured values of the attenuation coefficient α_I as a function of electron energy and half-aperture β for which scattered electrons get "lost"; adapted from Reimer [69b]. The instrument used was an Elmiskop I with a focal length of the objective of $f = 2.7$ mm, and objective apertures of $d = 20 / 50 / 100 \mu$; angular apertures $\beta = d/2f$.

film, with increase in thickness, suddenly starts to crystallize is shown in Fig. 37, for Bi. This may shed some light on Markhov's results with Bi. Figure 43 summarizes the available experimental data on α_I , measured by Reimer, Hall, and Inone, and Zeiler and Bahr. Note the strong dependence of α_I on the half aperture and on E_0 . α_I is inversely proportional to E_0 . Remarkable is also the very high absolute value in the order of 10^5 cm²/g for 40-keV electrons, in comparison with the usually much lower values of the absorption coefficients for light and X rays. This is one factor which makes electron microscopy and certain density gauges such an exceptional method.

Current Density along the Original Beam Axis $\partial J / \partial z$ (Infinite, Homogenous Absorber)

If we have a gas as the absorber and we place a small current detector opposite the electron source at a variable distance s , then we measure another variable which was not yet considered, namely the current density J along the axis z . We find again

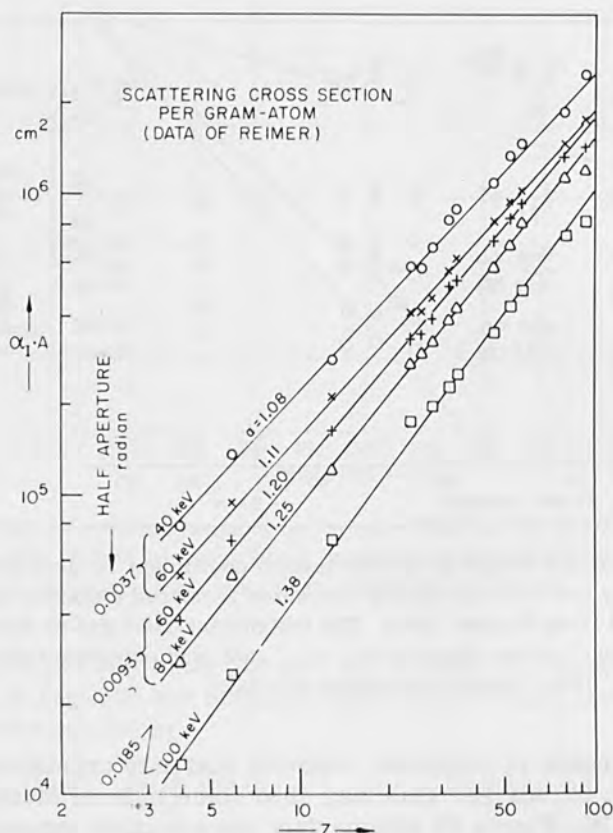


Fig. 35b. Scattering cross sections $\alpha \cdot A$ per gram-atom as function of electron energy and half-aperture α ; adapted from Reimer [69b].

an exponential attenuation law.

$$J = J_0 e^{-\alpha J \rho s} \quad (7)$$

This is the relation which Lenard found [71]. Since he did not express himself very clearly (although from the description of the apparatus and the experiments it is quite obvious that he meant current density), the relation was subsequently believed to hold for fractional transmission of the total current and for ionization, which it does not. Furthermore, the α_J values are orders of magnitude different from the α_I values defined before. There must be however, as we will see, a gradual transition which has not been explored.

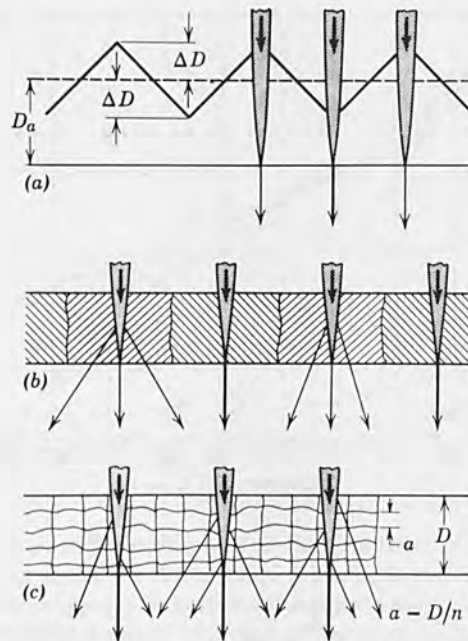


Fig. 36. Example for inhomogeneous films, (a) rough surface, (b) smooth but coarse crystalline, (c) smooth but fine crystalline; coherent electron scattering is indicated schematically; Reimer [69a].

Figure 38 shows a current detector in a scattering field of density ρ . Obviously the current density outside the center region is not constant and therefore the detector must be small if it is supposed to measure current density. For small values of s and a detector larger than the beam diameter, the detector will always catch the total current and no variation with s will be seen. But this does not reflect at all on electron scattering or absorption laws. For somewhat larger values of s , we come in a region where conditions resemble those in Fig. 34 and a sudden steep drop in current with s is found. For yet larger distances s , we begin to measure true current density where a small shift of the detector perpendicular to z will not alter the current reading. The steep middle section of the curve may be missing if the detector is very small. If the detector is very large it will not measure current density ever, but an undefined integral over various parameters.

Figure 39 shows a typical curve giving the current density J as

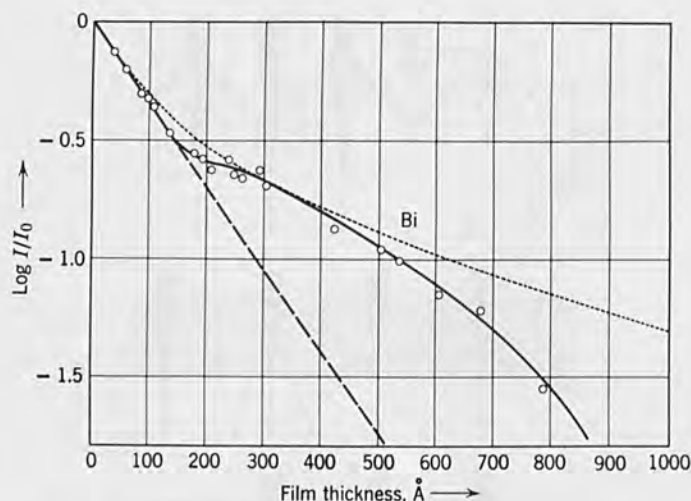


Fig. 37. Attenuation of 80-keV electron beam in thin films of Bi of various thickness; up to 150 Å the films are amorphous and smooth then the formation of irregularities can be seen in the electron micrograph; Reimer [69a].

function of s [11]. The detector was a shielded Faraday cage with 5-mm opening and about 120 degrees acceptance angle. For $s = 6$ cm (approximately $1/2 R$) the intensity perpendicular to s ($\pm X$ direction) shows a flat top and the slope of $J(s)$ approaches a constant value with $\alpha_J \approx 7000 \text{ cm}^2/\text{g}$.

The flat region for the current density curve perpendicular to the z axis may seem puzzling, but we must remember we have originally a parallel electron beam and not a point source with a $1/r^2$ spread of the radiation. We may perhaps consider this flat region as just an enlarged "picture" of the original beam diameter D . In this sense we can expect the α_J values to hold for any value of beam diameter D , e.g., for an infinite, plane, monodirectional source. In this special case there would be no difference any more between current density and transmission measurement. Again we face the problem of formulating proper similitude laws for such measurements. Obviously the detector diameter should be much smaller than s , and be of the same order as the source diameter D . The acceptance angle must be specified as well and should approach 180 degrees.

We must also remember if the logarithmic region of the attenuation curve is expressed as in equation 7, the value J_0 depends

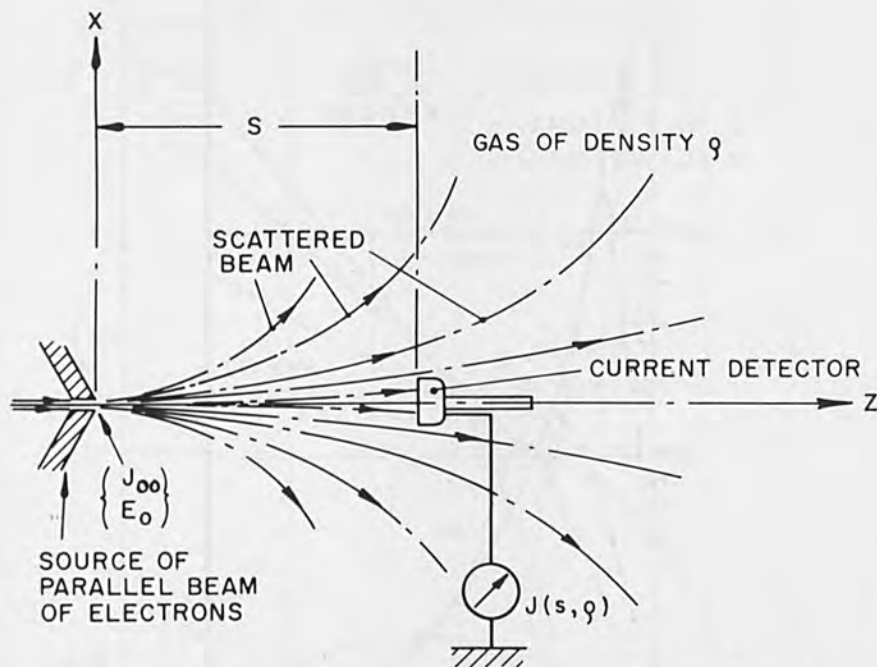


Fig. 38. Geometry when measuring current density along the original beam axis in a thick, homogeneous absorber.

not only on the source strength, but also on the detector diameter and source diameter and is therefore not identical with J_{00} at the source. (It has not been checked whether a unified description is possible by referring all currents to unit area.) This does not alter the values measured for αJ .

The detector must show a minimum of backscatter and therefore either be of carbon or beryllium, or a deep Faraday cage must be used. Its potential must be within about 0.05 V of the local space-charge potential, if any. Otherwise it will collect ions. This can be checked by measuring the Langmuir probe characteristic of this detector.

Figures 40 to 42 show measurements of $J(\rho s)$ made by Finckewirth [72] with the apparatus built by the writer. A Faraday cage with 2-mm opening and approximately 120 degrees acceptance angle was used. At the bottom of it a ZnS crystal was placed and its fluorescence intensity $H(\rho s)$ measured by a photomultiplier. (H is given in relative units only.)

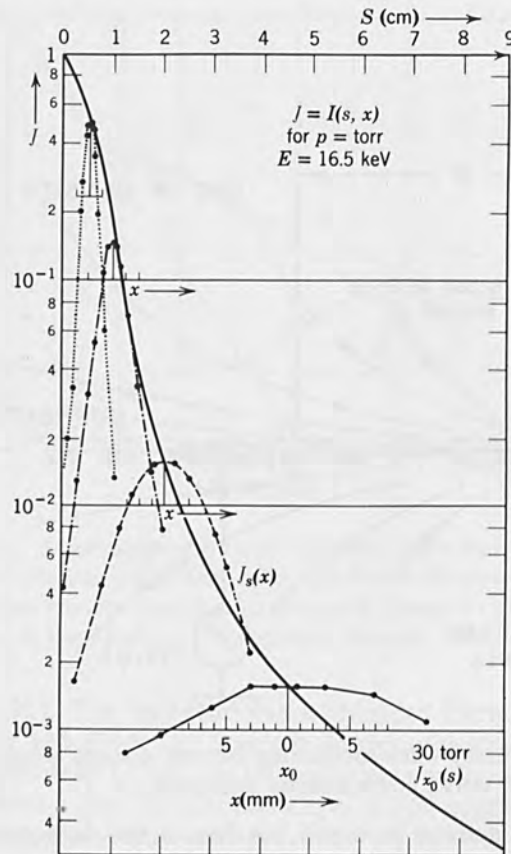


Fig. 39. Current density J along z and $\chi \perp z$, in air of 30 torr [11].

In these measurements the density ρ as well as the distance s was varied. This gave when $\log J$ versus (ρs) was plotted identical curves within the accuracy of the measurements. This is remarkable insofar as (since ρ changes everywhere) it can be taken as proof that in the region of small s , where the $J(s)$ curve does not follow the exponential function, the same attenuation law holds and the recorded deviations in $J(s)$ are simply a result of not recognized violations of the measuring conditions, or of changes in the measuring conditions which may be quite unavoidable.

In Fig. 41 the curve $H(\rho s)$ is steeper than the one $J(\rho s)$, because

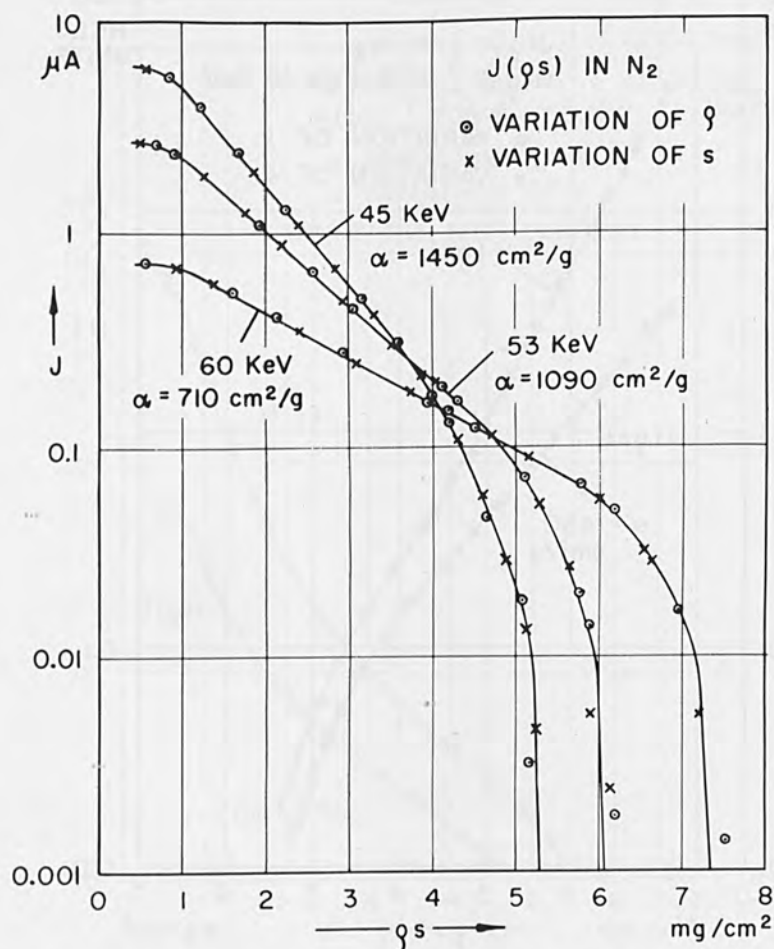


Fig. 40. Current density J along z measured with variation of ρ as well as s ; adapted from [72].

the acceptance angle of the H detector is smaller and H is proportional to energy rather than to current. The steeper portion for small values of ρs in Fig. 42 is believed to be due to a heating effect of the very much higher primary beam current on the gas. More measurements with a systematic variation of all parameters would be very desirable.

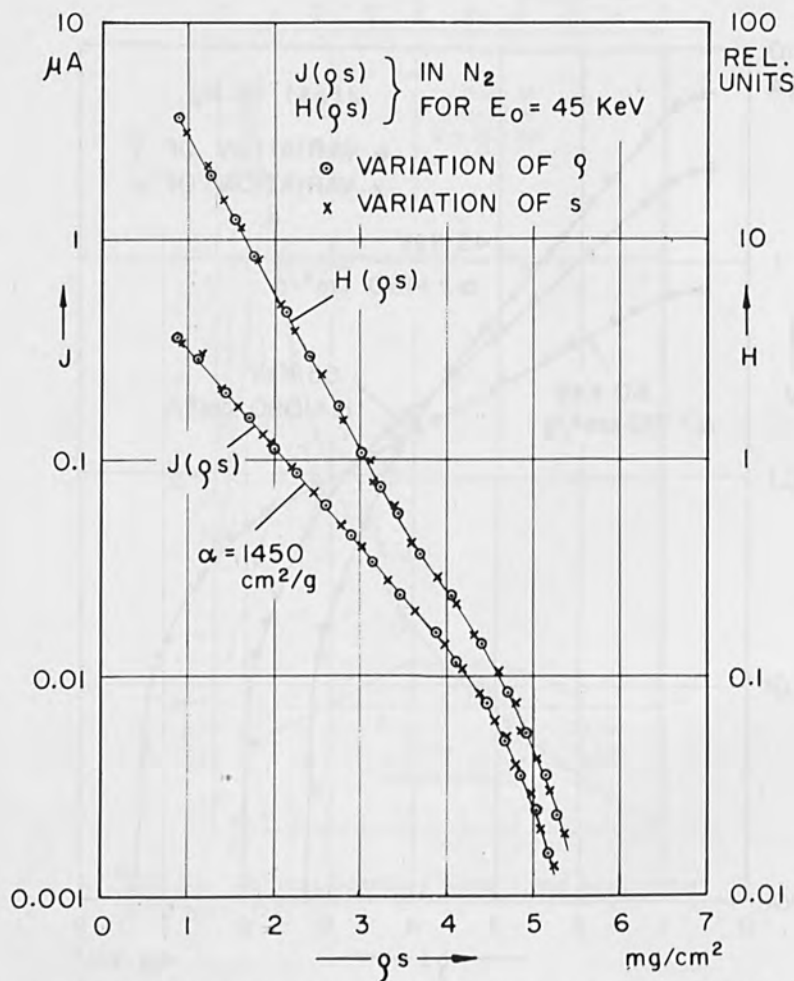


Fig. 41. Current density J and luminosity H of a scintillator as function of ρs ; adapted from [72].

Exponential Attenuation Laws and Practical Range R_J ; Compatibility of Range Law and Exponential Law

In two cases we found exponential attenuation laws to hold exactly over more than one decade of current or current density. These laws are not only, as is sometimes said, an approximation.

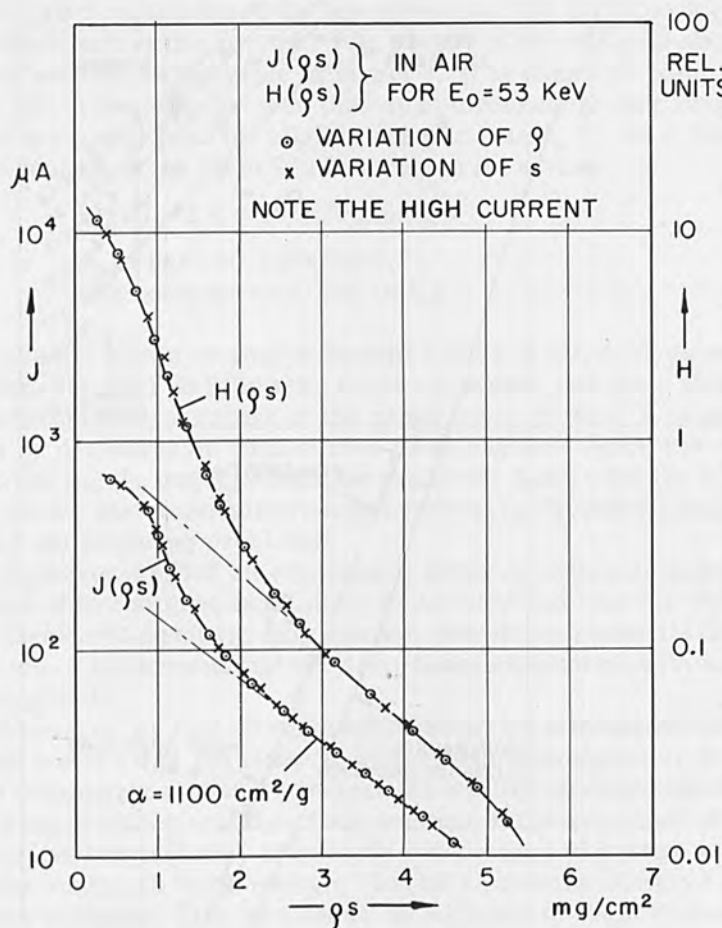
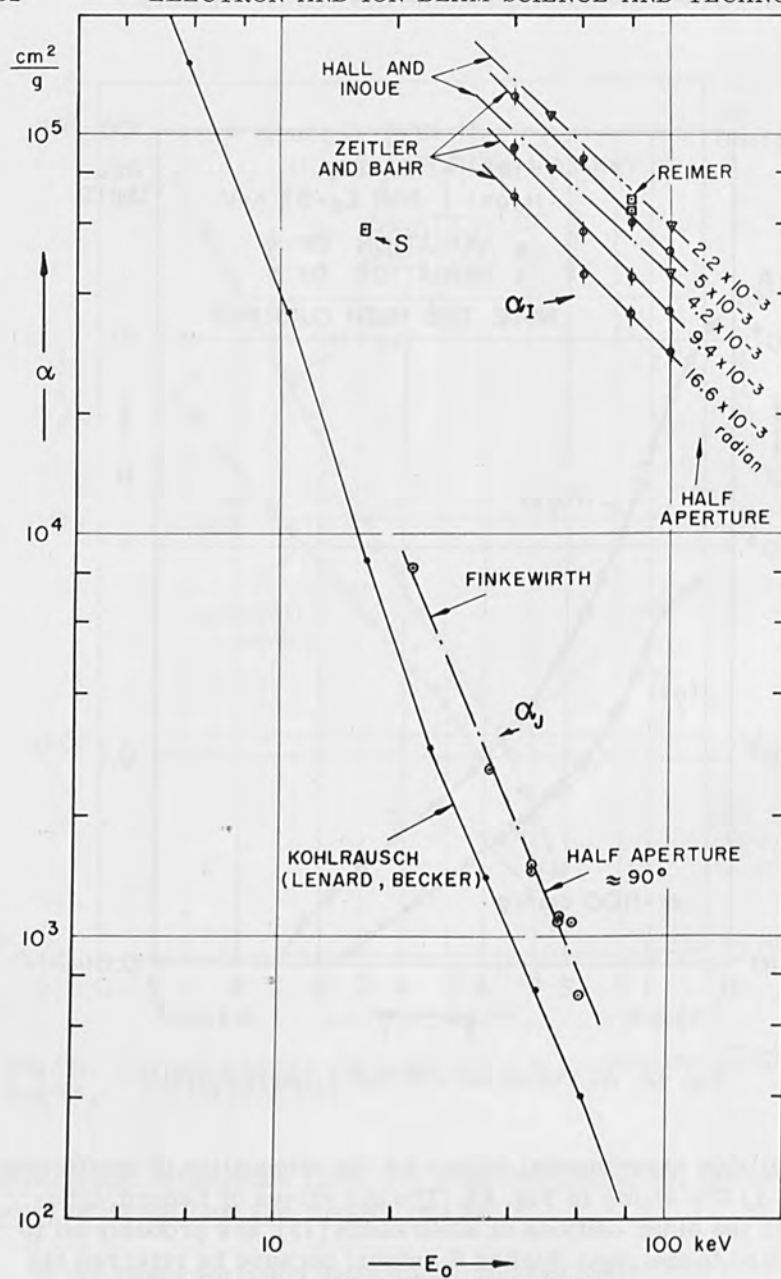


Fig. 42. As Fig. 41 but with high primary current.

Available experimental values for the attenuation of coefficients α_I and α_J are shown in Fig. 43. The old values of Lenard, tabulated in the older editions of Kohlrausch [73], are probably all to be shifted to the right (higher E values) because he referred his measurements to a "mean energy" $E < E_0$, which is only vaguely defined.

The values of α_I for $\delta \rightarrow 0$ and of α_J for a flat detector with 180 degrees acceptance angle must be considered to be extreme cases.



BEAM ATTENUATION COEFFICIENT α
AS FUNCTION OF ELECTRON ENERGY E_0

Fig. 43. Summary of beam attenuation coefficients measured by various investigators under various conditions α_I refers to conditions of Fig. 34, α_J to Fig. 38. Data from [3], [4], [7], [71], [72], [73].

For all other conditions the α value, if one can be found or defined, must lie in between, as for instance the lower α_I values for $\delta = 16.6 \times 10^{-3}$ radian show. On the other hand the α_J value for the steepest part of the curve of Fig. 39 is $6 \times 10^4 \text{ cm}^2/\text{g}$ or about ten times as high as the value at larger s . It is shown as point S in Fig. 43. It can also be seen that α_I is inversely proportional to E_0 , whereas $\alpha_J(E_0)$ falls off slightly steeper than E_0^{-2} . As a first approximation in the 20 to 200 keV region let us use

$$\alpha_J(E_0) = 2.4 \times 10^6 E_0^{-2} \text{ cm}^2/\text{g} \quad (8)$$

(E_0 in keV; $20 < 200 \text{ keV}$;
only accurate to $\pm 15\%$ in E_0)

It is not known to what minimum values of I/I_0 such curves as in Fig. 35 could be followed. It is well known, however, that J/J_0 measurements terminate at the range limit. In fact, a range $R_J(E_0)$ may be defined from such curves as in Fig. 40. Again the relations between R_E , R_I , and R_J should be explored. Let us for the time being ignore the minor differences between R_E , R_I , and R_J and consider the following problem:

Whenever we find an exponential decay or attenuation law we expect it to continue indefinitely to smaller and smaller values of the measured quantity. How can we, therefore, reconcile the existence of the exponential electron-beam attenuation with a definite range limit?

Whenever we find an exponential decay or attenuation law we must assume that the elementary process (absorption or scatter act) is constant or has a constant probability or cross section.

If we consider now the cross sections for energy loss of electrons, we see that they are nearly independent of energy as long as the energy is "high enough," but they increase steeply towards lower energies. This was shown by example in Figs. 11 and 12. We could therefore say that scattering leads to the exponential attenuation law and energy loss leads to its breakdown. The shape of the function J/J_0 near the range limit may therefore contain information about excitation and ionization thresholds, etc. This has not yet been studied.

The fact that in spite of energy loss we find a constant attenuation coefficient with increasing absorber thickness is not self-evident. But somehow, increased scattering inward, as well as outward, compensates for the lost energy; probably the same mechanism leads to the universal depth dose distribution and transmission curves. A detailed explanation will probably have to

await the experiment suggested later in this paper.

In some instances exponential absorption curves have also been found in simple transmission measurements. This is to be expected whenever the transmitted current becomes proportional to the current density in z direction. And this is the case when the mean scattering angle reaches 45 degrees ($z \approx R/2$). All such experiments would need a critical appraisal as to all "scaling parameters" involved, or perhaps changing if one factor, such as the absorber thickness, is varied.

Another aspect, which shall just be mentioned, concerns the relation between the current density and the local dose or expended energy. The measured current through a unit area perpendicular to the z axis on the z axis (looking toward the source and shielded from the rear) may be very small, but may in fact be the small z component of two larger currents flowing essentially in the $+$ and $-X$ direction. Therefore the energy flux through this area is not in any simple way related to the current.

The limit of the exponential attenuation law, equation 7, due to the range-energy law, equation 1, can be stated explicitly as a function of either E_0 , R , or α .

$$\text{We get} \quad \ln (J/J_0)_{\min} = -\alpha R = -11 E_0^{-0.25} \quad (9)$$

$$\text{or, since} \quad E_0^2 = (2.4 \times 10^{-6}/\alpha) :$$

$$\ln (J/J_0)_{\min} = -1.76 \alpha^{0.125} \quad (10)$$

$$\text{or, since} \quad E_0^{1.75} \doteq (R/4.57 \times 10^{-6}) :$$

$$\ln (J/J_0)_{\min} = -1.9 R^{-0.143} \quad (11)$$

$$(E_0 \text{ in keV; } \alpha \text{ in cm}^2/\text{g; } R \text{ in g/cm}^2)$$

The remarkable result is that with an increase in energy and range the length of the exponential attenuation region is reduced, or conversely, with an increase in the value of the attenuation coefficient the length of this region increases. The effect is quite drastic; we find

$E_0 = 6 \text{ keV}$	$(J/J_0)_{\min} = 1 : 1120$
60 keV	$1 : 50$
600 keV	$1 : 9.2$

For R at energies above 100 keV we should actually use equation 2 instead of 1. But equation 8 for α is not very accurate either, and we will not argue numerical values here. In any graph such as in Figs. 44 and 47, the $(J/J_0)_{\min}$ values can simply be found

from the intersection of the $\alpha_J(E_0)$ and $R(E_0)$ lines.

The physical meaning of all this seems to be that at higher energies the scattering loses in importance as compared with the ener-

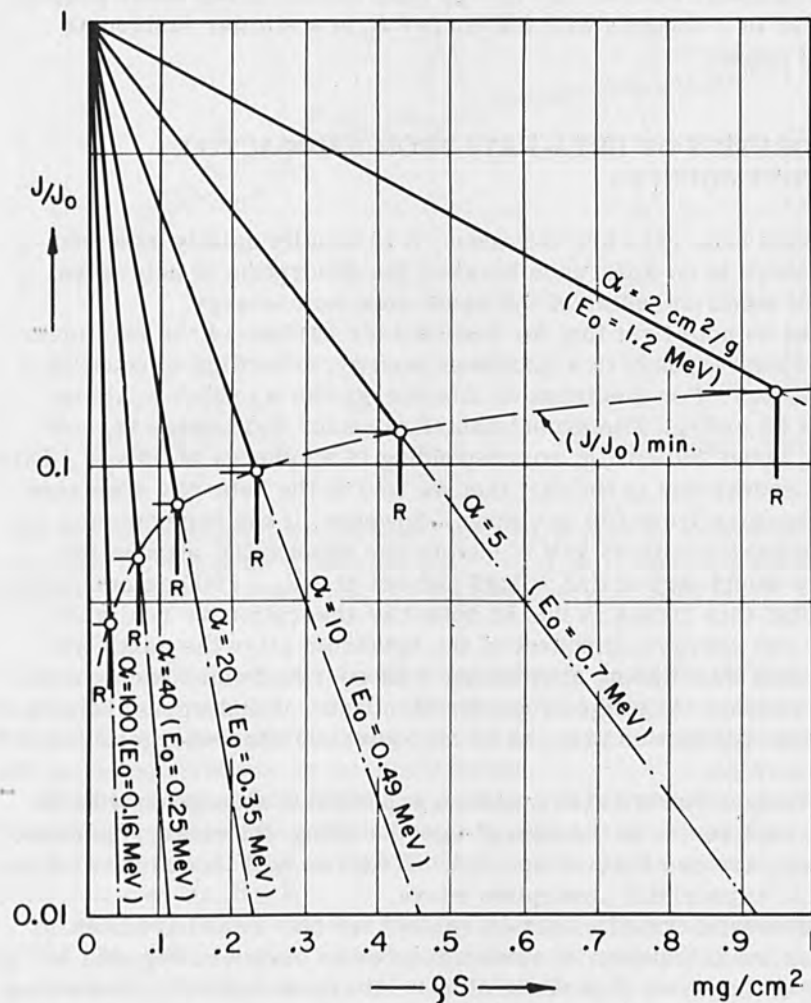


Fig. 44. Relationship between attenuation curves and range R . None of the exponential absorption curves can be followed beyond the limit (J/J_0) which is determined by $\alpha(E_0)$ and $R(E_0)$. Numerical values are approximate only.

gy loss. The relation, equation 9, between α , R , and E_0 has a definite significance in connection with the scatter-absorption of monoenergetic electron beams. Certain similar, empirical relations for beta-ray absorption and range [74], [75] have no such significance but, at best, tell us something about the beta-emission spectrum. It is an interesting question, for which there is presently no answer, whether the energy distribution in any plane perpendicular to z changes with the energy E_0 in a similar fashion as $\alpha(E_0)$ does.

ABSORPTION OF BETA-RAYS FROM RADIOACTIVE ISOTOPE SOURCES

Since beta-rays are electrons, it is usually quietly assumed that there is no difference between the absorption of beta-rays and of electron beams of the same maximum energy.

Let us note first that for instance for carbon-14 the maximum range corresponds to a maximum energy, according to equation 2, of 150 keV. For electrons of this energy the α_J value would be about 95 cm²/g. The experimental value for C-14 betas is, however, about 280 cm²/g, corresponding to an energy of 95 keV. This is of course due to the fact that we find in the beta-ray spectrum all energies from 150 keV down. However, if the experimental α_J value were due to 95 keV electrons the exponential attenuation curve would stop at $J/J_0 = 1:35$ and not at $J/J_0 \approx 1500$ as one finds.

What this means is that in beta-ray absorption we really absorb one energy component of the spectrum after the next. We may call this "fading attenuation." Of course, before each component reaches its range or penetration limit, it undergoes the same scatter-attenuation process as an equivalent monoenergetic beam of electrons.

The fact that we also measure exponential absorption with the beta-rays is due to the energy spectra being, let us say, "smooth." If there are two unequal spectra mixed, one will indeed not find a simple exponential absorption curve.

Attenuation coefficients (α values) for beta rays have been measured as function of atomic number as shown in Fig. 45. In view of the above it is doubtful to relate these values to scattering (to which α_J and α_I are certainly related). The α values for beta-rays from isotopes reflect rather on the dependence of R on the atomic number. In the older literature the statement is often made "if there were any appreciable diminution in the velocities of the

rays on passing through matter there should be a marked increase in the coefficient of absorption as the thickness of matter traversed was increased." The situation is not this simple. There are the changes in the directional spectrum of the electrons in addition to the energy loss.

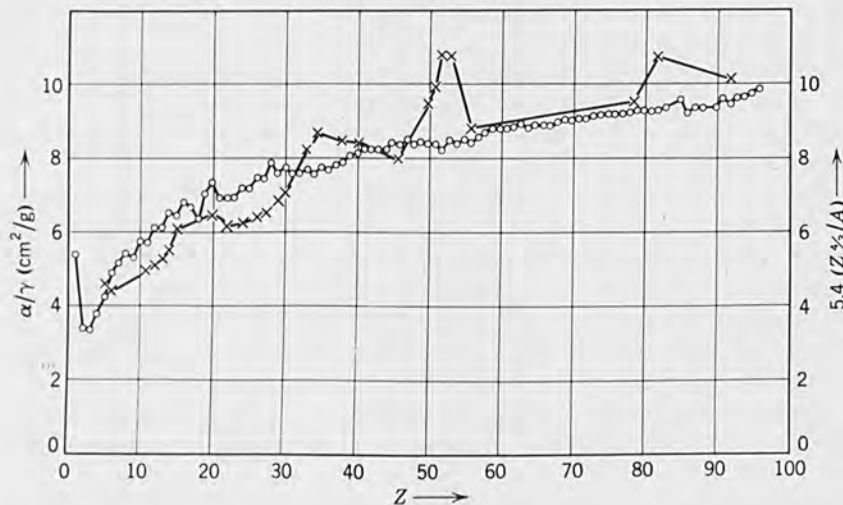


Fig. 45. Mass absorption coefficients for beta-rays of UX measured in a fractional transmission experiment by Crowther [76] as function of atomic number. O gives the coefficient $Z^{4/3}/A$ from theory of Lenz [43].

A much more detailed study of the subject would be indicated to produce a complete picture. But this is not the place and time for it. These few remarks may mainly serve as a warning against too naive experiments or interpretations.

Experimental values for the maximum ranges and attenuation coefficients, taken from the literature, regardless of how they were measured, are given in Fig. 47 for most of the commonly used beta-ray emitters.

Since the α values for the "fading attenuation" are determined by the beta-source spectrum and the range-energy relation for its components, the dependence on atomic number Z must be expected to differ from this dependence for α_J . The nonexponential early part of beta-absorption curves for light elements, as often reported, may however be due to a violation in similarity conditions for the geometry of the experiments.

GRAPHICAL REPRESENTATION OF ATTENUATION AND RANGE DATA

It seems worthwhile to discuss the graphical presentation of the data on electron-beam penetration in a special section. Properly made graphs which show several related quantities at once have the very useful property of being self-checking and revealing discrepancies in the data.

Values for the attenuation coefficients as function of energy, with other quantities shown as parameters, have already been presented in Fig. 43. Values for range as function of energy are shown in Fig. 46.

For the presentation of the exponential function it is customary to use semilogarithmic coordinates as in Fig. 44. Only a limited number of data can be accumulated in one such sheet without overcrowding the presentation. Using instead log log vs. log coordinates makes the presentation much more useful. This shifts the point $J_0 = 1$, $\rho s = 0$ to infinity, and makes parallel lines of the fan of lines in Fig. 44.

The exponential function is presented in this new fashion in Fig. 47. (This presentation is, of course, independent of the meaning of J , α , ρ , and s .) It was devised originally as an aid for the design of gas-density gauges. Parallel to the ρs scale, a scale with the energy needed for penetration is drawn, using equation 1 up to 100 keV and equation 2 from 0.1 to 3 meV (at 100 keV the slight discrepancy between the two equations is indicated). The J scale on the far left holds for the special case of $J_0 = 10,000$, which may be either a current or a count rate of particles. The scale $\sigma(J)/J$ gives the so-called coefficient of variance or statistical error for these specific values of J , if J is a particle count (some further comments below). Parallel to the α scale, for which the diagonal lines of the net are drawn, the related values $E_0(\alpha, J)$ are marked using equation 8. This may not be very accurate at very low and very high values of E_0 , but better data are not available.

In this general net for exponential attenuation some measured curves (Figs. 40 to 42) are replotted (marked 60N/53L/35L). The α value for some beta-ray spectra are marked, or rather the range of these values found in the literature. Measured points for Sr-90 [77], Kr-85 [78], and P-32 [79] beta-rays are also shown.

The relationship between $(J/J_0)_{\min}$ and R , α , and E_0 , as expressed in equation 9, appears in this diagram as a straight line. For $E_0 > 100$ keV where equation 9 is no longer accurate, the $(J/J_0)_{\min}$ values can be found as the intersection of the $\alpha(E_0)$ with $R(E_0)$ lines,

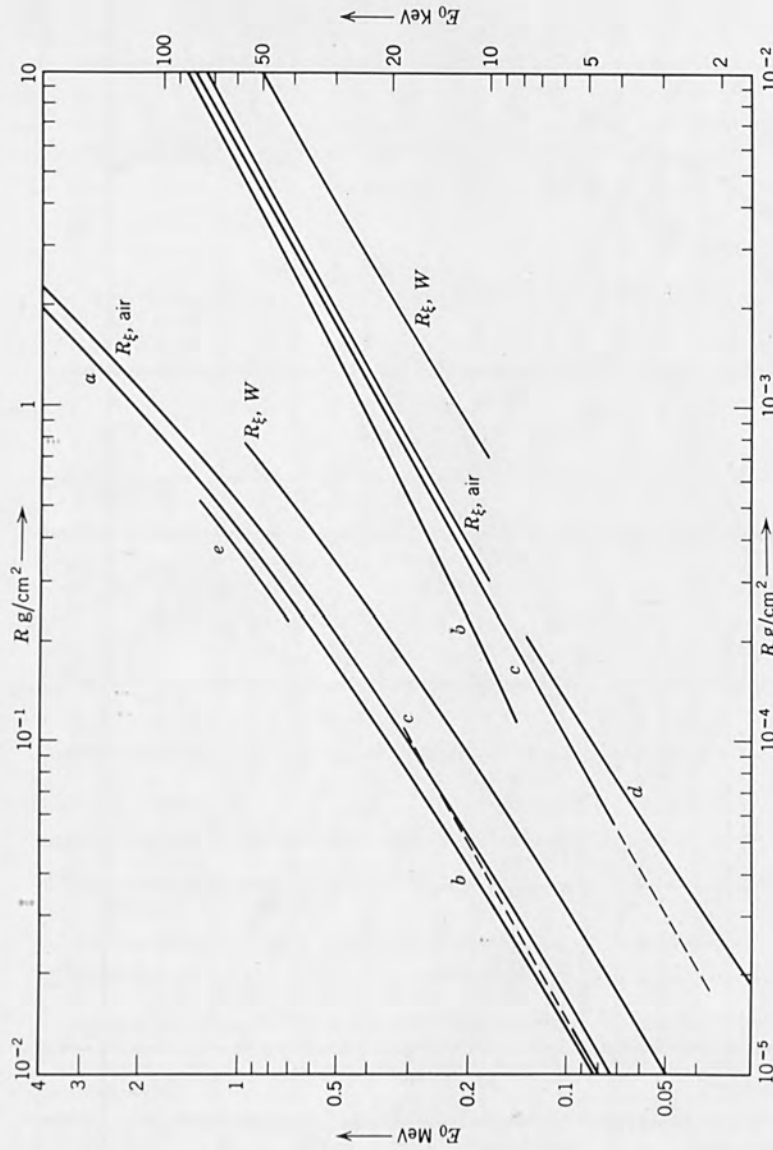


Fig. 46. Range-energy relations for electrons. (a) $R = 0.530 E_0 - 0.106 \text{ g/cm}^2$. (b) $R = 0.412 E_0^{(1.265-0.0954 \ln E_0)} \text{ g/cm}^2$. (c) $R_E = 0.813 E_0^{1.75} \text{ g/cm}^2$. (E_0 in MeV). (d) Kanter (Fig. 26). (e) Values of Horszowski [67]. a and b from Katz and Penfold [58], c from Gruen [26] R_E from Nelms [49].

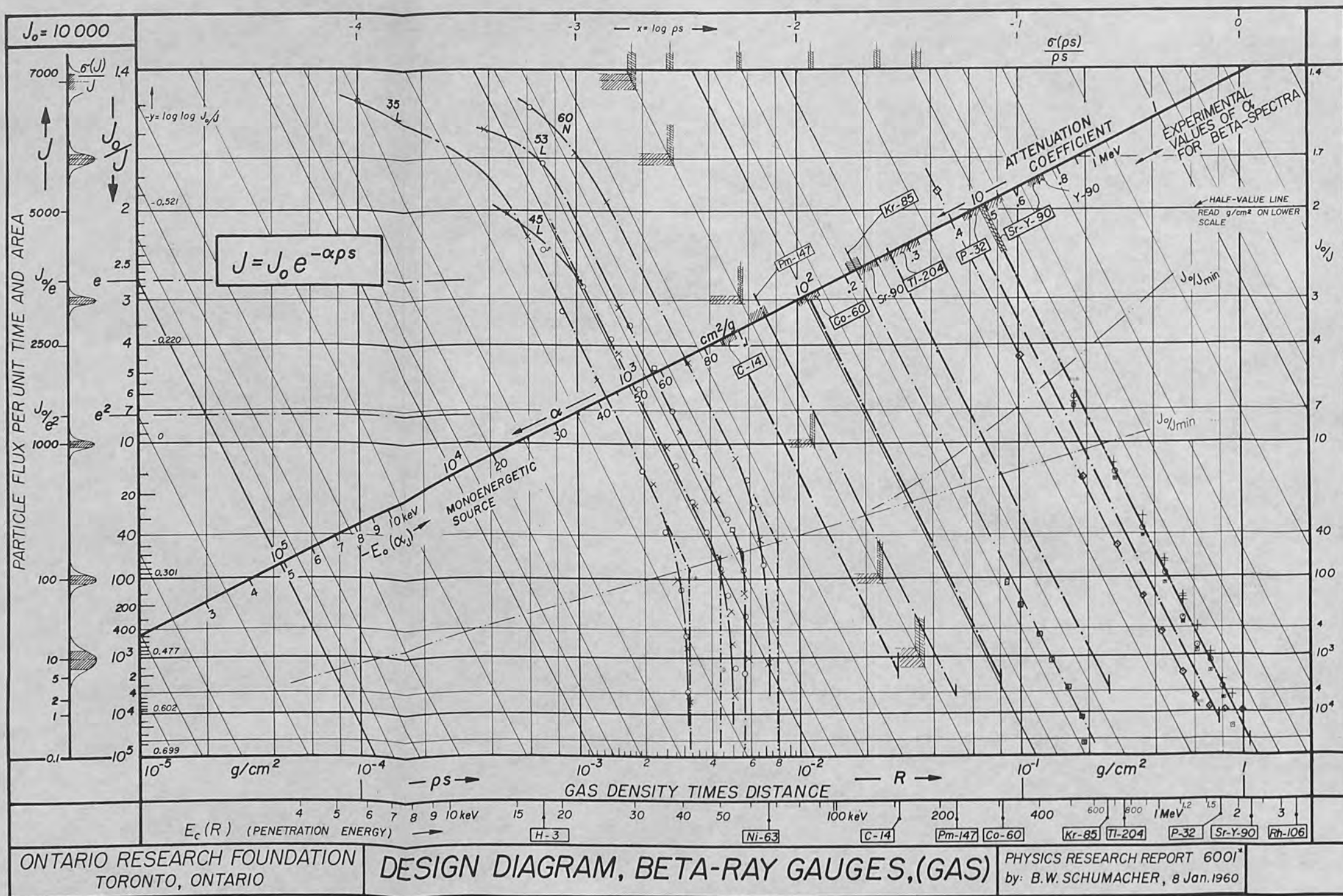


Fig. 47. Log-log graph of exponential attenuation function with additional scale for range and penetration (critical) energy. Numerical values of α for monoenergetic electrons and empirical values α for beta-rays.

as also shown. It is clearly evident how the exponential absorption of beta-rays proceeds to much lower values than that of monoenergetic beams. Note how the termination of the exponential curves for monoenergetic beams (60 N, etc.) falls on the line $(J/J_0)_{\min}$. (When making these measurements Finckewirth was not aware of the relationship, equation 9.) The shape of the termination is of course not given by the graph and should be investigated by more accurate measurements. The dependence of all parameters on atomic number Z should be re-examined. A plot as in Fig. 47 allows us to check the compatibility of all numerical factors entering equations 1 or 2, 8, and 9. The $(J/J_0)_{\min}$ line may be particularly sensitive to atomic number.

When measuring exponential absorption by particle counting methods it is not always realized that the greatest accuracy is obtained not for $J/J_0 = 1/e$, but for $J/J_0 = 1/e^2$, where the coefficient of variance, if expressed in terms of ρs or α rather than in terms of J , has a minimum [22], [80].

BACKSCATTERING

Several times, backscattering has been mentioned in passing. For completeness of this review let us look at the available quantitative data.

Backscatter can be understood from a look at a "slowing down collision series," Fig. 48, which can be treated mathematically [81].

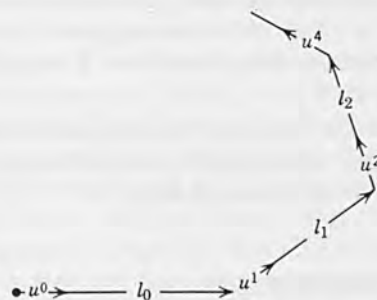


Fig. 48. Schematic diagram of a slowing down collision series; Leibfried [81].

as also shown. It is clearly evident how the exponential absorption of beta-rays proceeds to much lower values than that of monoenergetic beams. Note how the termination of the exponential curves for monoenergetic beams (60 N, etc.) falls on the line $(J/J_0)_{\min}$. (When making these measurements Finckewirth was not aware of the relationship, equation 9.) The shape of the termination is of course not given by the graph and should be investigated by more accurate measurements. The dependence of all parameters on atomic number Z should be re-examined. A plot as in Fig. 47 allows us to check the compatibility of all numerical factors entering equations 1 or 2, 8, and 9. The $(J/J_0)_{\min}$ line may be particularly sensitive to atomic number.

When measuring exponential absorption by particle counting methods it is not always realized that the greatest accuracy is obtained not for $J/J_0 = 1/e$, but for $J/J_0 = 1/e^2$, where the coefficient of variance, if expressed in terms of ρs or α rather than in terms of J , has a minimum [22], [80].

BACKSCATTERING

Several times, backscattering has been mentioned in passing. For completeness of this review let us look at the available quantitative data.

Backscatter can be understood from a look at a "slowing down collision series," Fig. 48, which can be treated mathematically [81].

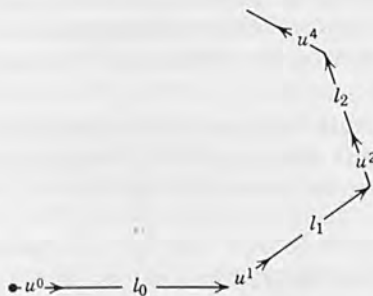


Fig. 48. Schematic diagram of a slowing down collision series; Leibfried [81].

On the average, each successive scattering angle increases and each free path decreases. That this leads to complete path reversal is typical for electrons. The effect is reduced at higher energies (where electron scattering approaches ion scattering) in keeping with similar findings for the scatter attenuation, as previously discussed.

Quantitative values for backscatter ratios and energy spectra of the backscattered electrons are shown in Figs. 49 to 51 [82], [83]. Directional spectra have been measured [84] and the low-energy range was particularly investigated [86], [25].

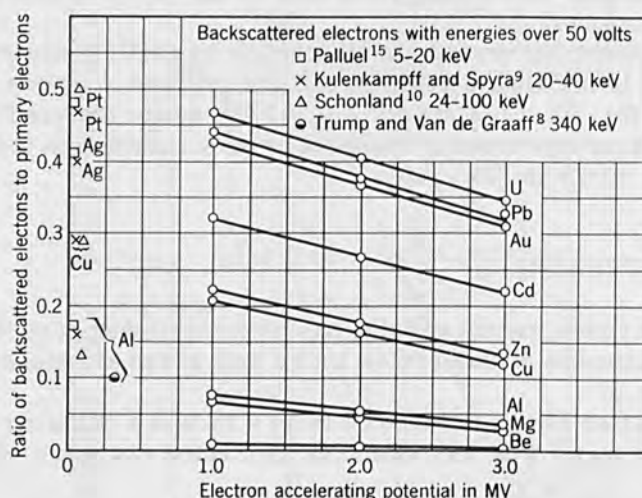


Fig. 49. Ratio of backscattered electrons to primary electrons normally incident with energies from 1 to 3 MeV on various target metals; from Wright and Trump [82].

If a beam of electrons impinges on targets of different atomic number located in a gas atmosphere, the difference in backscattering can readily be seen as shown in Fig. 52

SUGGESTED MEASUREMENTS

It was mentioned in several places how incomplete our quantitative knowledge of electron penetration still is, and how difficult it is to derive certain parameters from the customary experiments.

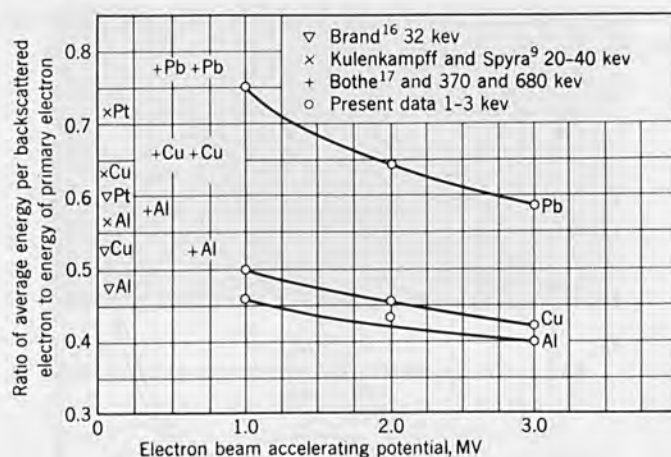


Fig. 50. Ratio of average energy per backscattered electron to energy of primary electron plotted for electrons normally incident on Al, Cu, and Pb with energies of 1 to 3 MeV; from Wright and Trump [82].

Twelve years ago I thought of the following experiment for measuring the directional energy spectrum $\Sigma_{\vec{p}}(E, r, z)$ at each point in the scattering field, and Gruen [27] has made a preliminary analysis of the connection of current density, local energy dissipation, and other quantities—measurable by “integrating” detectors—with this differential spectrum. Due to external circumstances nothing more has become of these beginnings. Nevertheless the experiment would still be worth doing and the quantitative results very useful. It is therefore suggested here.

As shown in Fig. 53 a monoenergetic electron beam is fired into a gas target, the density of which is adjusted so that the range becomes approximately $R = 50$ cm. This large scattering field is little disturbed by a detector probe of, say, 2 cm in length and 0.5 in diameter located at any point (X, Z) . This probe can be built as a little “telescope” with small entrance diameter and small angular aperture, as shown in Fig. 54. Two apertures spaced about 1 cm apart, and the space between evacuated by differential pumping, will perform this “telescope” function. Current can be measured by a Faraday cup and electron energy by a proportional counter (scintillator, or solid-state detector, or gas-filled detector). By variations over (X, Z) and the probe angles (Θ, Φ) , the spectrum $\Sigma_{\vec{p}}$ can be obtained with good resolution. The dependence on atomic

number can be obtained since gases like H_2 , Ne, Kr, Xe, and Hg vapor can be used as targets.

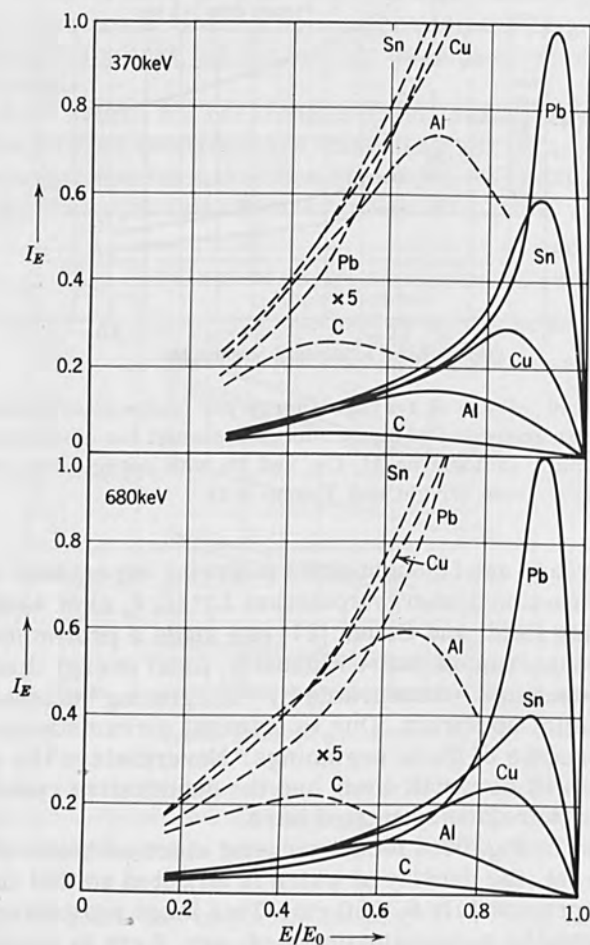


Fig. 51. Energy spectrum of backscattered electrons for different targets; same primary beam current in each case; angle of incidence 20° to 35° , angle of observation $50^\circ \pm 15^\circ$ against normal of target plane; Bothe [83].



Fig. 52. Backscatter made visible in an atmosphere of 1 torr N_2 ; electron beam of 45 keV, $60 \mu A$; target: C (top), Al (middle), Pb (bottom) [85].

REFERENCES

1. David L. Anderson "The Discovery of the Electron," Van Nostrand, 1964.
2. L. Marton and L. I. Schiff, *J. Appl. Phys.* **12**, 759 (1941).
3. C. E. Hall, *J. Biophys. Biochem. Cytol.* **1**, 1 (1955).
4. C. E. Hall and T. Inoue, *J. Appl. Phys.* **28**, 1346 (1957).
5. V. E. Cosslett, *J. Roy. Microsc. Soc.* **78**, 1-17 (1959), and **78**, 18-25 (1959).
6. R. E. Burge and N. R. Sylvester, *J. Biophys. Biochem. Cytol.* **8**, 1 (1960).
7. E. Zeitler and F. Bahr, *J. Appl. Phys.* **33**, 847-853 (1962).
8. R. Castain and J. Descamps, *J. Phys. Radium* **16**, 304 (1955).
9. R. Kriegler and B. W. Schumacher, *Plating*, April 1960.
10. B. W. Schumacher and S. S. Mitra, *Electr. Reliability and Microminiaturization* **1**, 321-331 (1962).
11. E. Schopper and B. Schumacher, *Z. Naturforsch.* **6a**, 700-705 (1951).
12. B. Schumacher, *Ann. Phys.* **13**, 404-420 (1953).
13. B. Schumacher *Optik* **10** 116-131 (1953).
14. A. E. Gruen, E. Schopper, and B. Schumacher, *J. Appl. Phys.* **24**, 1527 (1953).
15. A. E. Gruen, E. Schopper, and B. Schumacher, *Z. Angew. Phys.* **6**, 198 (1954).
16. B. W. Schumacher and E. O. Gadamer, *Can. J. Phys.* **36**, 659-671 (1958).

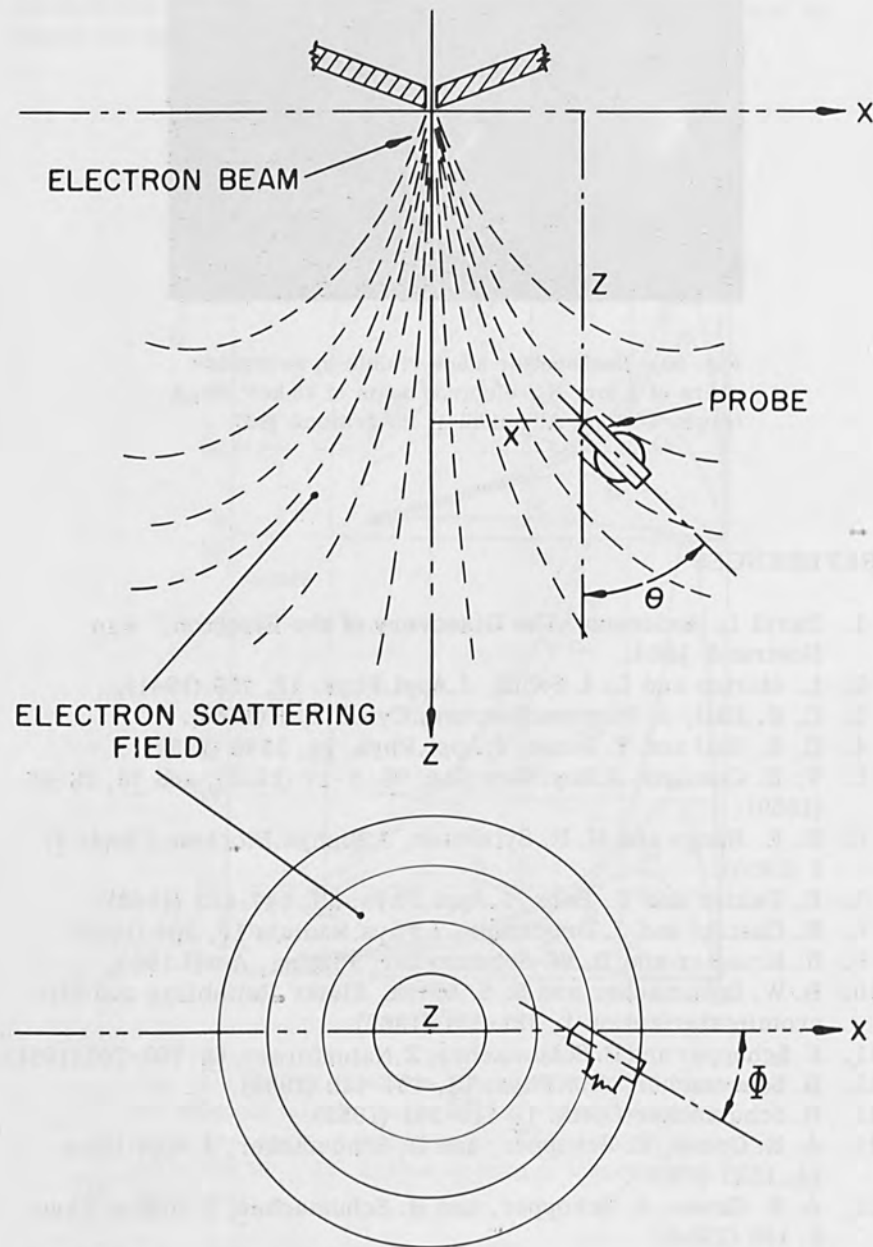


Fig. 53. Suggested measurement of the directional and energy spectrum in an extended scattering field (gas, density adjusted to give $R_E \approx 50$ cm) with a small probe.

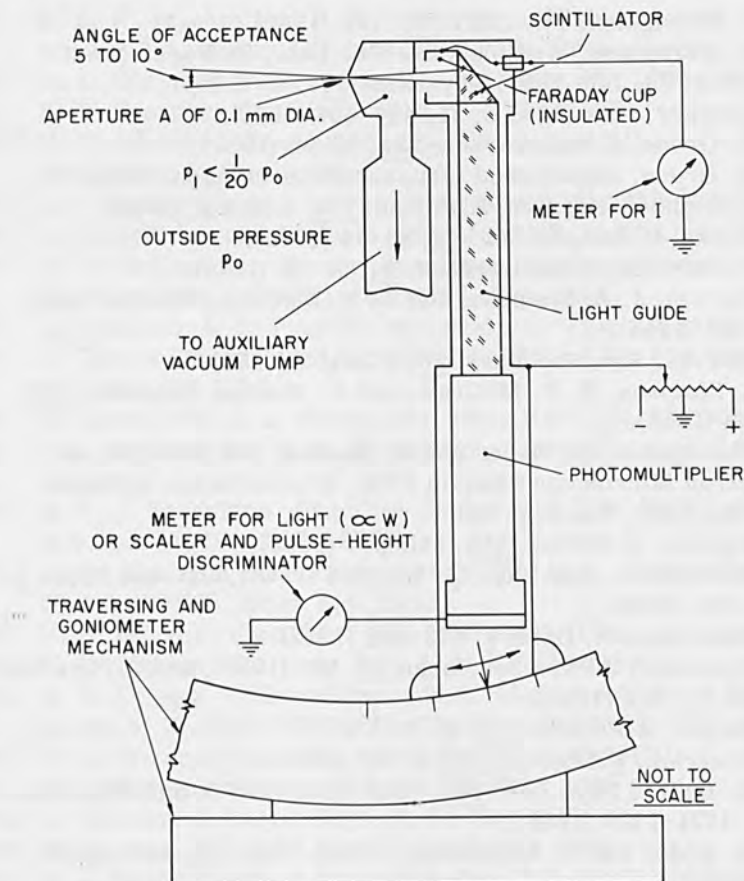


Fig. 54. Probe of Fig. 53 with differentially pumped apertures and current as well as energy sensitive detector; schematic.

17. B. W. Schumacher, *Nucleonics* **18**, 106-114 (1960).
18. H. Boersch, *Ann.Phys.* **26**, 631 (1936).
19. L. Marton, D. C. Schubert, and S. R. Mielczarek, *J.Appl.Phys.* **27**, 419 (1956).
20. D. Venable and D. E. Kaplan, *J.Appl.Phys.* **26**, 539 (1955).
21. S. S. Mitra, ORF Phys.Res.Report 6204, May 1962.
B. W. Schumacher, ORF Res.Report 6303, Feb. 1963.
22. B. W. Schumacher, *Symp.Transact.*, 4th Symp. on Temperature, Reinhold, New York, 1962, Vol. 3, Part 2.
23. H. R. Falckenberg and B. W. Schumacher, *Can.J.Phys.* **40**, 1521-1527 (1962).

- 24a E. J. Sternglass, *Phys.Rev.* 76, 189 (1949).
- 24b E. J. Sternglass, Westinghouse Res.Lab., Sc.Paper No. 6-94410-2-P9, July 1957, *Phys.Rev.* 95, 345 (1954).
- 25. K. Gentner, *Ann.Phys.* 5, 31, 407-424 (1938).
- 26. A. E. Gruen, *Z.Naturforsch.* 12a, 89-95 (1957).
- 27. A. E. Gruen, unpublished manuscript, Hechingen, 1954.
- 28. K. Jost and J. Kessler, *Z.Physik.* 176, 126-142 (1963).
- 29. W. Bothe, *Z.Naturforsch.* 4a, 88-94 (1949).
- 30. B. V. Borries, *Z.Naturforsch.* 4a, 51-70 (1949).
- 31. L. Marton, J. A. Simpson, and T. F. McCraw, *Rev.Sci.Instr.* 26, 855 (1955).
- 32. L. Marton, *Rev.Mod.Phys.* 28, 172-183 (1956).
- 33. J. A. Simpson, T. F. McCraw, and L. Marton, *Phys.Rev.* 104, 64-68 (1956).
- 34. M. Horstmann, G. Meyer and H. Raether, 4th Int.Conf. on Electron Microscopy, Berlin 1958, Transactions, Springer, Berlin, 1960, Vol. I, p. 290.
- 35. J. Kessler, *Z.Physik.* 155, 350-367 (1959).
- 36. G. Ruthemann, *Ann.Phys.* 2, 113-134 (1948), and *Ann.Phys.* 2, 135-146 (1948).
- 37. G. Moellenstedt, *Optik* 9, 473-480 (1952).
- 38. H. Watanabe, *J.Phys.Soc.Japan* 10, 321 (1955), and *J.Phys.Soc. Japan* 11, 112 (1956).
- 39. G. Meyer, *Z.Physik.* 148, 61-71 (1957).
- 40. W. Dietrick, *Z.Physik.* 152, 87-97 (1958).
- 41. L. B. Leder, *Phys.Rev.* 107, 1569-1578 (1957), and *Phys.Rev.* 103, 1721-1726 (1956).
- 42. L. B. Leder and J. A. Suddeth, *J.Appl.Phys.* 31, 1422-1426 (1960).
- 43a F. Lenz, *Z.Naturforsch.* 9a, 185-204 (1954).
- 43b H. Wyrwich and Fr. Lenz, *Z.Naturforsch.* 13a, 515-523 (1958).
- 44. N. H. Sarkhar, *J.Appl.Phys.* 34, 2379 (1963).
- 45. W. T. Scott, *Rev.Mod.Phys.* 35, 231-313 (1963).
- 46. L. Marton, J. A. Simpson, H. A. Fowler, and N. Swanson, *Phys.Rev.* 126, 182-192 (1962).
- 47. -- (anonymous) *Chem.Eng.*, Jan. 1954, p. 278.
- 48. H. Bethe, *Ann.Phys.* 5, 325 (1930), and *Handbuch der Phys.* (Springer, Berlin) Vol. 24, 1933.
- 49. A. T. Nelms, "Energy Loss and Range of Electrons and Positrons," NBS Circular 577, 1956.
- 50. W. Brandt, "Energy Loss and Range of Charged Particles in Compounds," Research Report from the Radiation Physics Laboratory, E. I. Du Pont de Nours & Co., Wilmington, Del.

51. H. S. W. Massey and E. H. S. Burhop, "Electronic and Ionic Impact Phenomena," Oxford University Press, 1952.
52. G. L. Olde and E. Brannen, *Rev.Sci.Instr.* **30**, 1014-1016 (1959).
53. L. V. Spencer, *Phys.Rev.* **98**, 1597-1615 (1955).
54. L. V. Spencer, "Energy Dissipation by Fast Electrons," NBS Monograph 1, Sept. 1959.
55. L. V. Spencer and J. Coyne, *Phys.Rev.* **128**, 2230-2237 (1962).
56. L. V. Spencer and U. Fano, *Phys.Rev.* **93**, 1172-1181 (1954).
57. R. T. McGinnies, "Energy Spectrum Resulting from Electron Slowing Down," NBS Circular 597, Feb. 1959.
58. L. Katz and A. S. Penfold, *Rev.Mod.Phys.* **24**, 28-44 (1952).
59. H. Kanter, *Phys.Rev.* **121**, 461-471 (1961), **121**, 677-681 (1961); **121**, 681-684 (1961).
60. H. Kanter and E. J. Sternglass, *Phys.Rev.* **126**, 620-626 (1962).
61. J. E. Holliday and E. J. Sternglass, *J.Appl.Phys.* **30**, 1428-2431 (1959).
62. B. F. J. Schonland, *Proc.Roy.Soc. A*, **104**, 235 (1923); *Proc. Roy.Soc. A*, **108**, 187 (1925).
63. Japan Electron Optics Laboratory Co., Ltd., Report No. XA 63-1 and JEOL News **1**, 6 (1963).
64. H. H. Seliger, *Phys.Rev.* **100**, 1029-1037 (1955).
65. A. F. Makhov, *Fit.Tver.Tela* **2**, 2161-2184 (1960).
66. B. N. C. Agu, T. Burdette, and E. Matsukawa, *Proc.Phys.Soc. (London)* **72**, 201-206 (1958); **72**, 727-732 (1958).
67. S. M. Horszowski, *J.Sci.Instr.* **40**, 532-534 (1963).
68. P. S. Rudolph, *Science* **135**, March 1962, title page.
- 69a. L. Reimer, *Z.Angew.Phys.* **9**, 34-38 (1957).
- 69b. L. Reimer, *Z.Angew.Phys.* **13**, 432-434 (1961).
70. R. D. Heidenreich, *J.Appl.Phys.* **33**, 2321-2333 (1962).
71. P. Lenard, *Wied.Ann.Phys. u. Chem.* **51**, 225-267 (1894); "Quantitatives ueber Kathodenstrahlen" Winter Verlag, Heidelberg, 1918.
72. K. Finckewirth, Diplomarbeit, Stuttgart 1953, unpublished.
73. F. Kohlrausch, *Praktische Physik*, 18th ed., Teubner, Berlin, 1944.
74. M. Forro, *Phys.Rev.* **92**, 931-933 (1953), and *Z.Phys.* **138**, 441-448 (1954).
75. L. Daddi and V. D'Angelo, *J.Appl.Radiation and Isotopes* **14**, 97-103 (1963).
76. J. A. Crowther, *Phil.Mag.* **12**, 379 (1906).
77. B. W. Schumacher, W. Wojcik, and R. C. Zavitz, *J.Appl. Radiation and Isotopes* **13**, 123-129 (1962).
78. B. W. Schumacher and U. Thiede, unpublished.

79. S. A. Husain and J. L. Putman, Proc.Phys.Soc. A, 70, 304 (1957)
80. B. W. Schumacher, ORF Phys.Res.Report 6001, Jan. 1960.
81. G. Liebfried, J.Appl.Phys. 33, 1933-1943 (1962).
82. K. A. Wright and J. G. Trump, J.Appl.Phys. 33, 687-690 (1962).
83. W. Bothe, Z.Naturforsch. 4a, 542-545 (1949).
84. J. M. Kuschner and G. W. Der-Schwarz, 4th Int.Conf. on Electron Microscopy, Berlin 1958, Transactions, Springer, Berlin, 1960, Vol. 1, p. 222.
85. B. W. Schumacher, Can.J.Phys. 40, 376 (1962).
86. J. E. Holliday and E. J. Sternglass, J.Appl.Phys. 28, 1189-1193 (1957).

FUNDAMENTALS OF THERMIONIC ENERGY CONVERSION[†]

INTRODUCTION

Energy derived from a heat source can be converted directly to electrical energy by the proper application of the principles of thermionic electron emission. To utilize this method of energy conversion, three elements are indispensable. First, electrons must be emitted from a conducting surface maintained at a high temperature. Second, the electron current must be carried by means of some transport mechanism across a gap to a collector. And third, an electron collector must be present which is maintained at a considerably lower temperature than the emitter and has a low enough work function in comparison with that of the emitter to permit the device to deliver power to an external circuit.

The device described in these most general terms is a thermionic diode. It is not to be implied that the thermionic device which is finally shown to have the greatest utility as an energy converter will be a simple diode. At the outset, stress must be placed on ultimate objectives from an overall systems point of view. For example, one user may wish to have a converter which operates at the maximum possible efficiency, whereas another application may call for a converter which delivers the maximum amount of power per unit weight of the entire system. It is almost certain that the construction which gives the maximum power per unit weight will not be the same construction as the one that gives the maximum efficiency.

It is always the ambition of a true scientist to utilize basic knowledge in order to accomplish a specific objective. The indispensable background of information which here is classified as

*Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Mass.

[†]This work supported in part by the U.S. Army, Navy, and Air Force under Contract DA36-039-AMC-03200(E). This paper was one of the four plenary presentations on the opening session of the conference.

"fundamental" really relates to at least three basic scientific areas. One may be identified as the fundamental physics of "microscopic" phenomena. This term is used for lack of a better one to indicate the need for detailed information concerning the physical processes involved in the actual thermionic emission of electrons from a surface. The transport phenomena depend on detailed information concerning ion generation, electron and ion space charge, and other properties associated with general concepts of plasma generation and maintenance. The work function of the collector is again a microscopic quantity in that knowledge concerning the means by which the most suitable collector work function can be maintained is needed. In contrast to these considerations are the "macroscopic" aspects of the problem.

The designer of a converter needs to make detailed studies of the heat and electrical conduction of materials. He needs to design macroscopic configurations of these materials to minimize the heat losses and minimize the losses in power due to the ohmic resistance of the electrical conductors that serve as terminals of this energy converter. Radiation losses must be minimized and these depend on macroscopic considerations and "engineering" design. Still a third area of comparable importance has to do with the "material" problems. Long life of the device may be of extreme importance and efficiency and power per unit weight may have to be sacrificed in order to select materials that will not disintegrate or evaporate under the conditions that are dictated by the need to have extreme reliability for long periods of time.

Important though these last two areas involving fundamentals are, this discussion will be directed exclusively to the fundamentals related to the "microscopic" phenomena. Even to deal with this area comprehensively would require a book instead of an article; therefore, this treatise will simply highlight some of the problems and indicate the approach that forms the basis of extensive research programs now directed toward the achievement of energy conversion through thermionics.

AN INTERRELATION BETWEEN COMPONENT PARTS

In order to bring forward the interrelation between the components of a thermionic energy converter, Fig. 1 has been prepared. It first calls attention to the fact that the difference between the heat energy put in and the heat energy taken out is the only heat available for conversion directly to electrical power. Since this converter is thermodynamically a heat engine, the Carnot efficiency

defined by $(T_1 - T_2)/T_1$ sets an absolute upper limit to the efficiency of the device. It goes without saying that this efficiency can never be attained in any practical structure. Electrical power can always be defined as the product of current and voltage. The diagram of

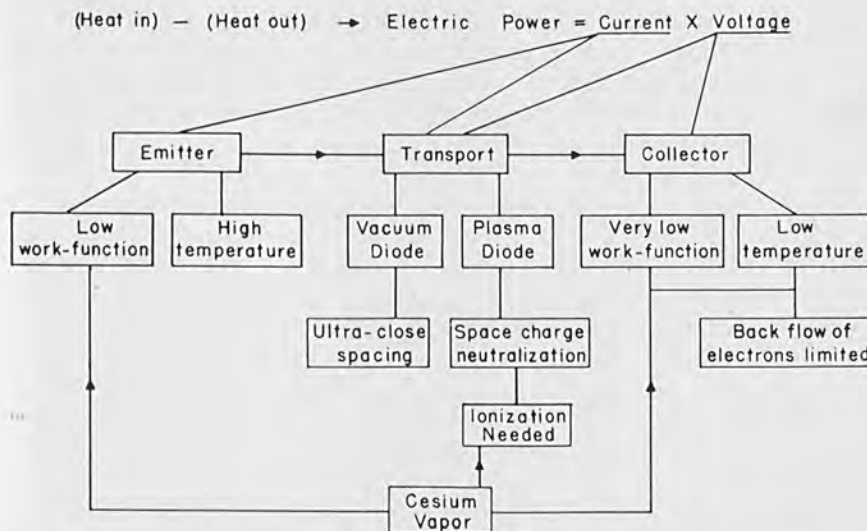


Fig. 1. Block diagram to show interrelation between elements of a thermionic converter.

Fig. 1 shows that the emitter and the transport mechanism predominate in the determination of the available current. The transport and the collector properties predominantly determine the voltage available. To attain the desired current density from the emitter, it must have a suitably low work function and operate at a relatively high temperature. Unless ultra-close spacing is used, electrons cannot be transported efficiently over to the collector unless positive ions are provided to neutralize the electron space charge. Unless the collector work function is appreciably less than the work function of the emitter, the output voltage when the device is operating under the condition of maximum power will be extremely low. In fact, it will be the voltage equivalent of temperature which can be computed directly as $\bar{V} = 8.616 \times 10^{-5} T_1$. At 2000°K , this voltage is 0.17 V. To achieve a high-power density, therefore, the current would have to be very large. If the difference between the emitter work function and the collector work function is 1.0 eV,

then the available voltage could approach 1.2 V to give six times the power for the same current.

To operate at such a high temperature at 2000°K calls for a very refractory material such as tungsten or rhenium. Either of these materials alone would yield a current density of less than 0.01 amp/cm². Such a low-current density would be hopelessly small for a practical device. The fundamental studies of Langmuir and Taylor [1] established the fact that the work function of a refractory material can be reduced so greatly in the presence of a cesium film maintained by the equilibrium between condensation and evaporation to give an emission current density at the specified temperature higher than 10 amp/cm².

In schematic, the diagram of Fig. 1 calls attention to the importance of having cesium present in the converter in order to establish the required low work function for the emitter and provide for an adequately great thermionic emission current density. Clearly, if cesium will reduce the work function of the emitter, it will also reduce that of the collector. Studies to be mentioned here show that this reduction does in fact take place by the suitable choice of materials and temperatures so that an emitter-to-collector work-function difference of at least 1 eV can be maintained as desired.

Cesium also has the property of being easily ionized. The ionization procedure depends on a combination of fundamental mechanisms. An important one is the production of ions at the emitter surface by thermal ionization. The anticipated yield of ions is well approximated by the Langmuir-Saha [2] theory. Evidence is strong that under suitably chosen operational conditions many more ions are produced by electron collisions with the cesium atoms in the interelectrode space. Generally speaking, this is not accomplished by the simple impact of an energetic electron with a neutral cesium atom. Cumulative processes are clearly evident and the fundamentals, although under present investigation, are not clearly established.

THERMIONIC EMISSION IN THE PRESENCE OF CESIUM

Most specimens of tungsten are fabricated from polycrystalline material. Before extensive heat treatment, the crystals are very small, but the maintenance of the specimen at a high temperature results in recrystallization and the formation of much larger crystals. In order to achieve stability in the operation of thermionic

converters, part of the processing should involve the maintenance of the emitter at a high temperature and for a sufficiently long period of time so that the major part of the recrystallization will have been completed.

Studies made of single crystals of tungsten reveal that the true thermionic work function is very dependent on the exposed crystallographic direction [3]. The surface characterized by the 110 direction has a very high work function of at least 5.3 eV and holds a cesium layer at a higher equilibrium density than any of the other surfaces. Other tungsten areas have work functions as low as 4.3 eV. In spite of this nonuniformity in the properties of the various crystallographic faces of tungsten, the quantitative results that describe the electron emission, current density as a function of the emitter temperature and the cesium bath temperature, have been in remarkably good agreement even though the specimens used for those studies have had different heat treatment and geometrical configuration [4].

It is important to the efficiency of a thermionic converter to have a very uniform work function over the surface as it is operated. It is not easy to determine that the emitter work function is uniform and a number of researches are now in progress designed to evaluate the nonuniformity associated with individual specimens operated under conditions that simulate, to some extent at least, ones likely to be found in an actual converter.

For design purposes, it is important to have available the best knowledge concerning the thermionic properties of refractory materials; therefore, the available data on tungsten used to form the chart [5] shown in Fig. 2. The independent variables are the emitter temperature and the cesium bath temperature, both expressed in degrees Kelvin. Associated with these two temperatures, an observer can determine an electron emission current density under a wide range of conditions. The Richardson-type formula can be solved in terms of the emitter temperature and the observed current density to give the effective work function. This equation is

$$\phi_1 = \bar{V} (14.0 + \ln T_1 - \ln J_0) \quad (1)$$

It is essentially by this means that the map of tungsten-cesium properties in Fig. 2 is made. The solid lines establish then a correlation between cesium temperature and emitter temperature to accomplish a specific effective work function. The dotted lines that form "S" curves represent the emission capability expressed in amp/cm². Across this chart, in the general region of the work function 2.8 to

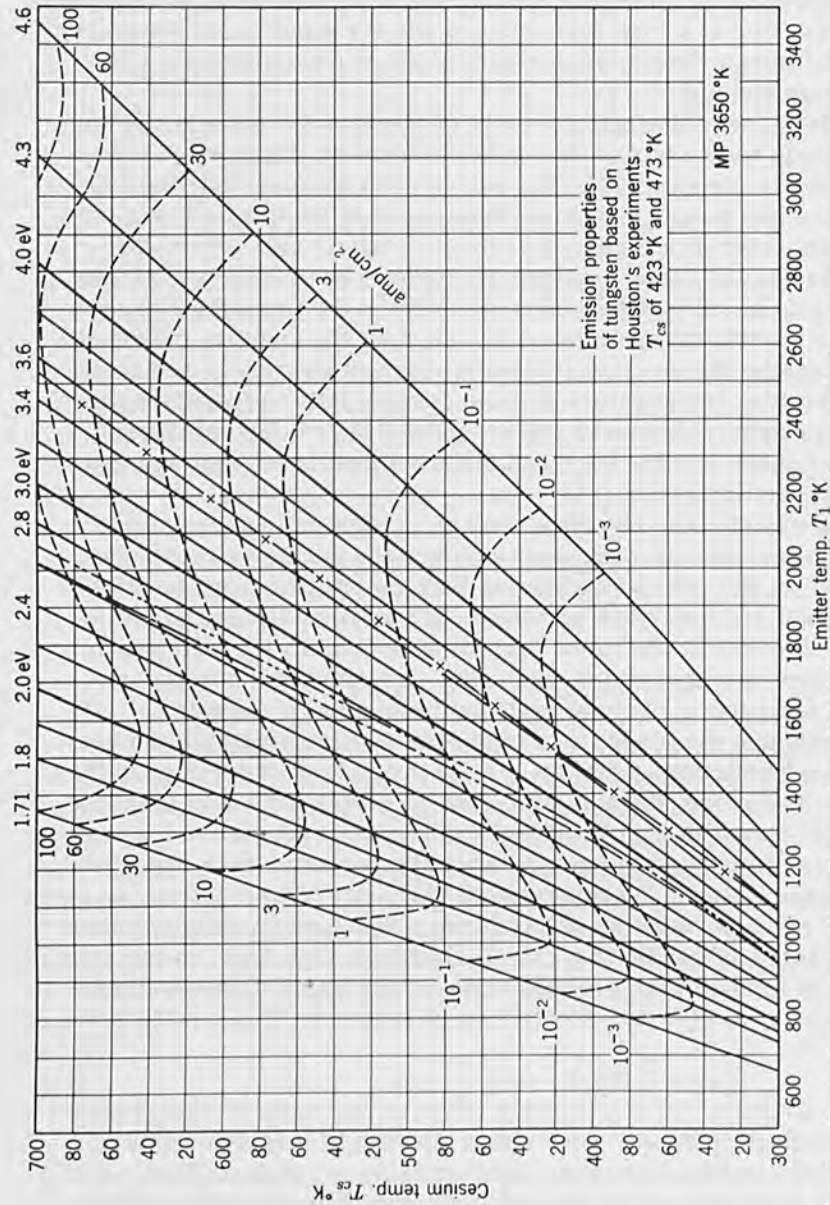


Fig. 2. Emission properties of tungsten. Solid lines: work function related to emitter temperature and cesium temperature. Dotted lines: current density in amp/cm². Double dot-dash line: demarcation between electron-rich sheath on the left and ion-rich sheath on the right as given by Langmuir-Saha equation. Plus-dash line: demarcation between stable and unstable region. Ions to the right of line must be returned to the emitter to obtain stable results.

2.6 eV, are some lines that divide the chart into two important regions. The region to the left of these lines is one identified as being "electron rich," whereas the one to the right of these lines is "ion rich." Thus, in the absence of ions being generated in the space between the emitter and the collector, an electron space-charge sheath depresses electron emission for the electron-rich region, whereas on the right part of the diagram, an ion sheath accelerates the electrons into the interelectrode space and suppresses the available ion production.

If the zero of the cesium temperature and the zero of the emitter temperature had not been suppressed in the layout of the coordinate scales, it would be seen that the lines tend to go through the origin of the coordinate system. It is this tendency that encourages some observers to maintain that the work function is dependent only on the ratio (T_1/T_{Cs}). The fact that the lines do not go through the origin exactly, demonstrates that this assumed dependency on the ratio is simply a useful approximation.

Some data are available that describe the properties of molybdenum and tantalum in a similar manner [5]. The properties of rhenium are being investigated, but the details are not as yet published. Because of the dearth of well-documented information concerning the emission properties of potentially good thermionic emitters, it is still impossible to identify the emitter material most likely to be used. Not only will it be necessary to discover the most favorable emitter material, but it will be equally important to discover the best way to process this material in order to obtain the desired uniformity.

DESIRABLE COLLECTOR PROPERTIES

Some of the desirable collector properties may be enumerated:

- (1) The collector work function should be as low as possible consistent with the overall systems requirements. This point will be discussed later.
- (2) The collector work function should be uniform.
- (3) The collector work function should not change with time.
- (4) The collector work function should not be dependent on the flow of electron current to it.
- (5) If the collector work function is lowered by the presence of a complex molecular structure, the film must have a negligible electrical resistance.

The collector work function is dependent on the structure of the

base material, the density of the partial monomolecular layer of cesium, and on the temperature. In certain applications, specifically related to the direct conversion of heat to electricity in outer space, it will be desirable to operate the collector at a relatively high temperature in order to minimize the weight of the radiator used to discharge the excess heat. This is one of the factors that makes it exceedingly difficult to generalize concerning the most desirable work-function properties of the collector.

ANALYSIS OF VOLTAGE CURRENT CURVES

One of the first steps that must be taken to evaluate a thermionic converter design depends on the acquisition of a voltage-current curve. For any particular test vehicle, such a curve depends on controllable parameters such as emitter temperature, cesium bath temperature, collector temperature, and spacing. Uncontrolled parameters often include the detailed surface structure of the emitter and the collector, and the presence of spurious discharges that may possibly be initiated between other elements of the test vehicle such as the surrounding walls, the emitter or collector support, and other elements that may be present in a particular device. In order to acquire information concerning fundamental mechanisms active in determining a voltage-current characteristic, it is necessary to devise a test vehicle which minimizes spurious phenomena. Such a test vehicle has been under investigation by Breitwieser [6] and has yielded valuable information. His studies have not been confined to the limited region in the voltage-current curve that corresponds to power conversion. In his studies, the collector potential may be made quite negative with reference to the Fermi level of the emitter. Under this condition at close spacing, the ion-current yield may be observed and thanks to the presence of supporting guard rings, leakage currents may be minimized.

Two typical voltage-current curves are shown in Fig. 3. One applies to the very close spacing of $10\ \mu$ and the other to the larger spacing of $460\ \mu$. Data may be analyzed in terms of the Schottky [7] theory as it relates to ion production. Figure 4 is presented as illustrative of these results. At the close spacing of $10\ \mu$, the observed data follow close to the theoretical line associated with that small spacing. At the larger spacings, the figure shows lines of steeper slopes than those observed at close spacing, and these indicate directly that the Schottky theory is not applicable within the range of applied voltage available before sputtering effects and other interferences set in.

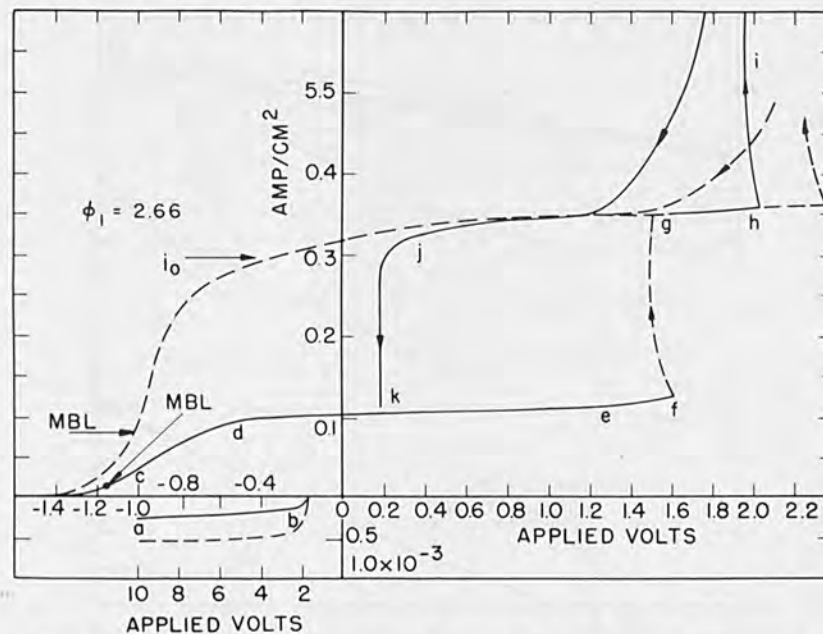


Fig. 3. Two voltage-current curves taken by Breitwieser. Emitter temperature 1500°K, cesium condensation 500°K. Solid-line spacing: 460 μ . Dash line spacing: 10 μ . Note change in voltage scale for ion currents shown negative. Current density anticipated from chart of Fig. 2 is i_0 . MBL limit of Maxwell-Boltzmann distribution. Passive mode range d to e : quasi saturation due to limiting barrier. Ignition at f . Range g to h : ignited mode before arc mode at i . Ignited mode returned toward passive mode j to k . Line may be retraced if k remains 5 per cent above passive mode line.

The analysis of the ion-current data, combined with possible electron emission from the collector, serves as the means of separating out the electron current itself from the total observed current in the voltage-current characteristics. The plot of the logarithm of the current density as a function of the dimensionless parameter $(V/\bar{V})$ yields additional valuable information. Four lines typical of these results are shown in Fig. 5. This means of displaying the data yields straight lines over the range in current density that corresponds to the Maxwell-Boltzmann electron energy distribution. The theoretical slope of these lines is such that for a shift in $(V/\bar{V})$ of 2.3 the electron current density changes one decade. The observed results yielded this slope quite accurately

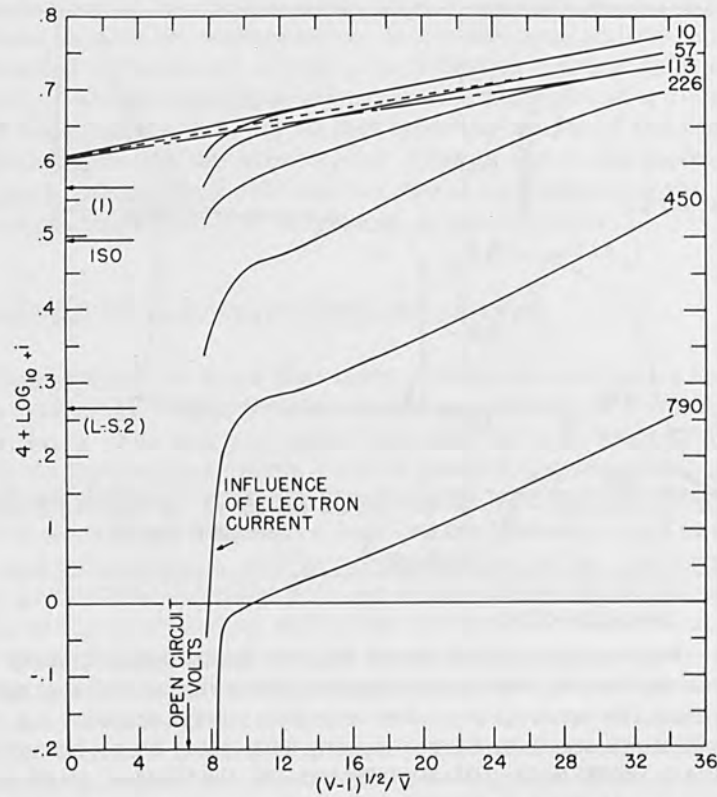


Fig. 4. Ion currents plotted on the basis of the Schottky mirror-image theory for ion current increase with applied voltage. Six spacings shown from 10 to 790 μ . (L-S . 2): anticipated ion current based on Langmuir-Saha theory. ISO: random current in isothermal diode. (1): Langmuir-Saha equation with statistical weight of 2 omitted.

up to the Maxwell-Boltzmann limit after which the slope became less than that required by theory. This deviation from theory is a direct indication of the presence of some space-charge effect. At the very close spacing of 10 μ , the knowledge of the current density at a particular value on the Boltzmann line permits the accurate calculation of the effective work function of the collector as given by the following formula:

$$\phi_2 = \bar{V} (14.0 + 2 \ln T_1 - \ln J_v) + V_{app} \quad (2)$$

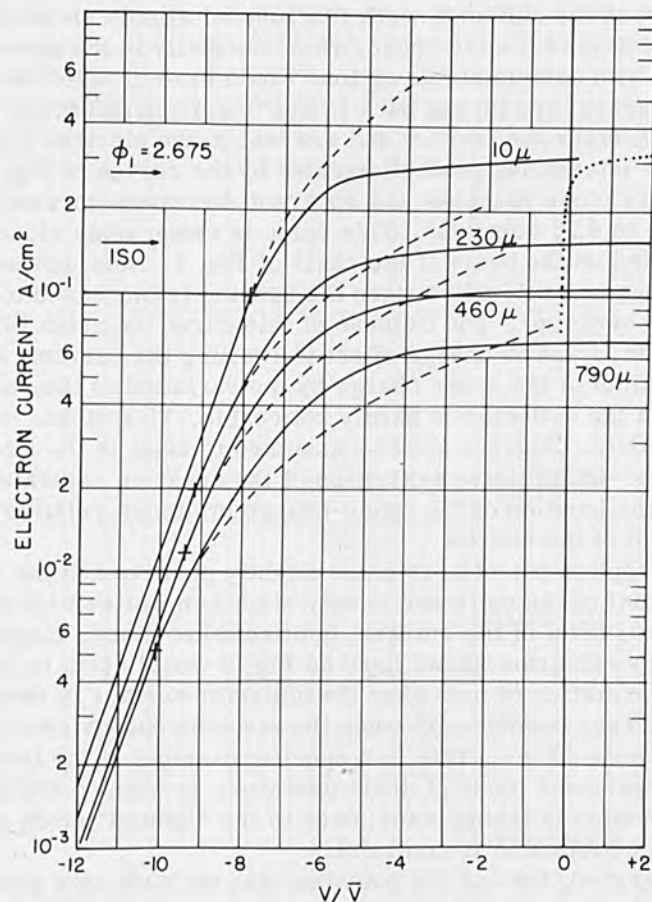


Fig. 5. Electron currents plotted according to Maxwell-Boltzmann energy distribution analysis. Emitter 1500°K , cesium 500°K . Limit of Maxwell-Boltzmann range shown by +. Dashed lines: space-charge theory applied to an ion-free electron emitter. Dotted line: return from ignited mode toward passive mode for the spacing 790μ . ISO: random current density expected in an isothermal diode.

Figure 5 would seem to indicate that the collector work function increases with the spacing even though its temperature and the other controllable parameters, namely, the emitter temperature and the cesium temperature, are held constant. It is our opinion that this displacement of the Boltzmann line is not a true indication

of a change in the collector work function but simply shows that a certain fraction of the electrons, which normally in the absence of collisions with cesium atoms or ions would have crossed the inter-electrode space, are turned back in the "electron-retarding" field and thus re-enter the emitter and are lost to the electron collector.

Another interesting point illustrated by the curves of Fig. 5 is that, at very close spacings, the apparent saturation current is very close to 0.28 amp/cm^2 . This value is remarkably close to that expected on the basis of the chart of Fig. 2. Note, however, that the apparent saturation when the spacing is increased to 790μ is $7 \times 10^{-2} \text{ amp/cm}^2$. The flatness of this curve indicates two things: first, a space-charge effect is limiting the current; second, the modulation of the space charge by the variation of the surface potential of the collector is hardly noticeable. This means that a complex motive diagram must be assumed to exist in the interelectrode space. Quantitative evaluation of the apparent saturation establishes the location of the space-charge minimum relative to the Fermi level of the emitter.

As the applied potential is made slightly positive and the surface potential of the collector is only slightly negative with respect to the Fermi level of the emitter, ignition takes place. Inspection of the motive diagram shown here as Fig. 6 would seem to indicate that this generation of ions near the collector surface is established at a very copious rate when the electron energy available barely exceeds 2.6 eV. This is a specific example of the transfer from the "passive" mode of diode operation, in which a negligible space ionization is taking place, over to the "ignited" mode for which space ionization is dominant.

After ignition, the applied potential may be made less positive and the current observed remains practically equal to the emission capability of the emitter, as illustrated by the dotted line of Fig. 5. Very close to zero applied voltage, it is evident that the current falls very sharply. In spite of the steepness of this curve, it is retraceable as long as the minimum current transferred across the diode does not become less than $7.5 \times 10^{-2} \text{ amp/cm}^2$. The ignited mode is lost when the electron current transmitted across the diode approaches within 5 per cent of the current observed while it was still operating in the passive mode.

It has been the objective of this discussion to indicate that valuable information concerning thermionic diode operation can be acquired as the result of the detailed study of the voltage current curve. To obtain this information, it is necessary to study over a very wide range of applied potentials, so that the various compo-

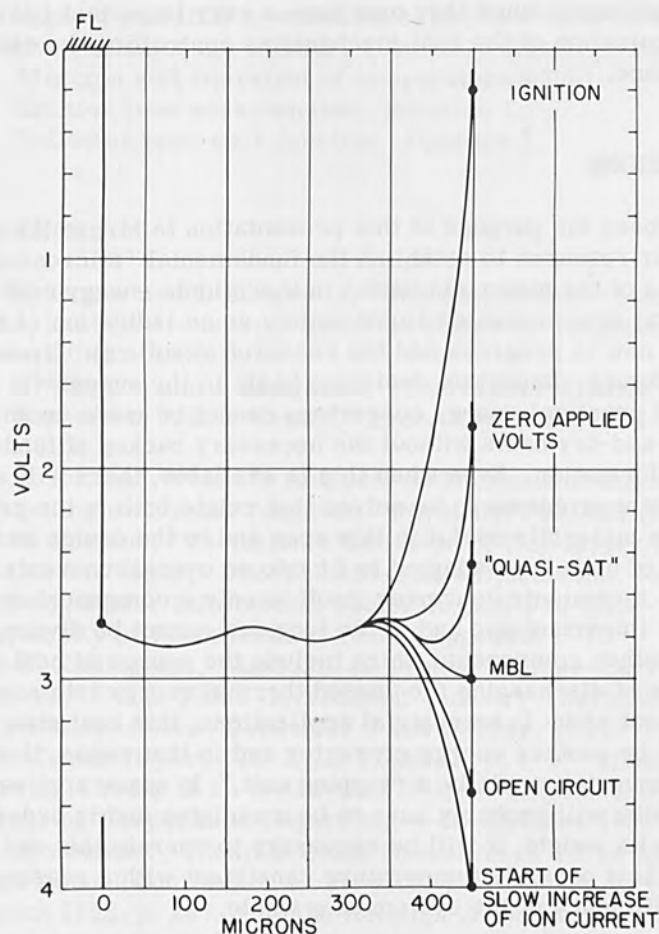


Fig. 6. Hypothetical motive diagrams applicable to spacing of 450μ , temperatures 1500°K and 500°K . Surface potential of the emitter relative to its Fermi level shown by left-hand dot. Collector surface potentials shown for six different values of applied potential. Collector work-function 1.8 eV . Note intermediate space-charge barriers not easily modulated. Important changes in the collector sheath are shown.

nents of current can be separated. It is not easy to make this separation in a completely unambiguous manner. Small details should not be overlooked since they may have a very important bearing on the determination of the real mechanisms controlling the converter performance.

CONCLUSIONS

It has been the purpose of this presentation to stress the need for further research to establish the fundamental "microscopic" properties of the materials useful in thermionic energy converters. The factual data presented serve mainly as an indication of the researches now in progress and the research results anticipated in the near future. Important decisions basic to the successful development of practical energy converters cannot be made on anything but a cut-and-try basis without the necessary backup of fundamental information. Even when this is available, there will still be important problems to be solved that relate both to the properties of the materials useful in this area and to the design and construction of units best adapted to fit into an overall converter system. The thermionic converter itself is only a component even though an important one and in the long run cannot be divorced from the other components which include the source of heat and the means of discharging the unused thermal energy into some form of heat sink. In terrestrial applications, this heat sink might very well be another energy converter and in that sense, the thermionic converter would be a "topping unit." In space applications, the heat sink will probably have to be a radiator and in order to minimize its weight, it will be necessary to operate this radiator at the highest possible temperature consistent with a reasonable overall efficiency of the system as a whole.

GLOSSARY OF SYMBOLS

k	Boltzmann's constant 1.380×10^{-23} joule/°K.
J_0	Electron current density at zero field in amp/m ² . Equation 1.
J_v	Current density in amp/m ² at any applied potential V_{app} on the Boltzmann line. Equation 2.
q	Electron charge 1.602×10^{-19} C.
T_1	Temperature of emitter in °K. Equation 1.

T_2	Temperature of collector.
T_{Cs}	Cesium reservoir temperature °K.
V_{app}	Applied potential to establish collector Fermi level relative to that of the emitter. Generally negative. Equation 2.
$\bar{V}$	Electron volt equivalent of temperature (kT/q).
ϕ_1	Emitter true work function. Equation 1.
ϕ_2	Collector true work function. Equation 2.

REFERENCES

1. J. B. Taylor and I. Langmuir, "The Evaporation of Atoms, Ions, and Electrons from Caesium Films on Tungsten," *Phys. Rev.* **44**, 423 (1933).
2. K. H. Kingdon and I. Langmuir, "Thermionic Effects Caused by Vapours of Alkali Metals," *Proc. Roy. Soc. (London)* **A107**, 61 (1925).
3. A. R. Hutson, "Velocity Analysis of Thermionic Emission from Single-Crystal Tungsten," *Phys. Rev.* **98**, 889 (1955).
4. J. M. Houston, "Thermionic Emission of Refractory Metals in Cesium Vapor," *Proc. Round-Table Discussion*, June 1 and 2, 1961, Power Information Center Report PIC-ELE-TI 3/3, University of Pennsylvania, Phila., Appendix F-1.
5. W. B. Nottingham, "Analysis of Typical Voltage-Current Curves," *Proc. Third Government-Industry Thermionic Round-Table Discussions*, December 6 and 7, 1961, Power Information Center Report PIC-ELE-TI 209/1, p. 1-1. "Sheath and Plasma Theory of an Isothermal Diode," Thermo Electron Engineering Corporation Report No. 4-62, October 1962.
6. R. Breitwieser, "Cesium Diode Operation in Three Modes," 23rd Annual Conference on Physical Electronics, M.I.T., March 1963, p. 267. "On the Relation of Ion and Electron Emission to Diode Diagnostics," Thermionic Conversion Specialist Conference, Gatlinburg, Tenn. October 1963, p. 17.
7. W. B. Nottingham, "Cesium Ion Production in a Strong Ion Accelerating Field," 23rd Annual Conference on Physical Electronics, M.I.T., March 1963, p. 281.

VERIFICATION AND USE OF HERRMANN'S OPTICAL THEORY OF THERMAL VELOCITY EFFECTS IN ELECTRON BEAMS IN THE LOW-PERVEANCE REGIME

I. INTRODUCTION

The magnetic focusing fields required to confine highly compressed, low-perveance electron beams are generally estimated from a formula given by Herrmann [1] in his "optical theory of thermal velocity effects in cylindrical electron beams." The beams to which this theory applies find application in traveling-wave tubes operating at millimeter wavelengths in which the delicate rf structure is readily damaged even by moderate amounts of beam interception. Unfortunately, the accuracy with which Herrmann's theory is obeyed is not known, since no quantitative investigations of the theory have been reported. In view of the importance of Herrmann's theory in the design of millimeter traveling-wave tubes, such an investigation was carried out in the course of the development of a certain millimeter-wave tube on a beam of perveance 0.015×10^{-6} .

The results of this study show that the Herrmann theory is obeyed with a good degree of accuracy. In view of this good agreement, the theory may be used in this perveance regime to draw meaningful conclusions about the conditions under which beam transmission is optimized with the least expenditure of magnetic field. The results of this investigation are presented in this paper in addition to those of the experimental study.

Various aspects of Herrmann's theory are given in Section II.

*Electron Dynamics Laboratory, Hughes Research Laboratories, Malibu, Calif.

These form the basis of the experimental investigations which are reported in Section III.

II. THEORETICAL IMPLICATIONS OF HERRMANN'S THEORY

A. Aspects of Herrmann's Theory

It is necessary to make frequent reference to Herrmann's paper and we therefore present below various modified aspects of his work.

The paraxial ray equation in the complex notation in which

$$u = x + iy = r \exp(i\theta) \quad (1)$$

has the form

$$\ddot{u} - 2i\omega_\ell \dot{u} - \left[\frac{\eta \bar{\rho}}{2\epsilon_0} + \frac{\eta}{2} \frac{\partial^2 \Phi}{\partial z^2} + iz \frac{\partial \omega_\ell}{\partial z} \right] u = 0 \quad [\text{H}(2)]^1$$

$$\ddot{z} + \eta \frac{\partial \Phi}{\partial z} = 0 \quad (3)$$

with time t as the independent variable.

Here the Larmor frequency ω_ℓ is given by

$$\omega_\ell = \frac{\eta B_z}{2} \quad (4)$$

B_z and Φ are the axial components of the magnetic field and the electric potential, respectively, both measured on the axis of symmetry, and $\bar{\rho}$ is an average value of the space charge density at every cross section.

Equation 2 is transformed to the Larmor coordinate frame which rotates about the axis with angular velocity ω_ℓ by the substitution

$$u_\ell = u \exp\left(-i \int_0^t \omega_\ell dt\right) = r \exp\left[-i\left(\int_0^t \omega_\ell dt - \theta\right)\right] \quad (5)$$

The transformed equation is

¹Equation numbers preceded by an H correspond to equations in Herrmann's paper.

$$\ddot{u}_\ell + \left[\omega_\ell^2 - \frac{\eta \bar{\rho}}{2\epsilon_0} - \frac{\eta}{2} \frac{\partial^2 \Phi}{\partial z^2} \right] u_\ell = 0 \quad (6)$$

(H7)

This equation is solved by Herrmann for a typical boundary electron which leaves the cathode edge at radius r_c with an initial transverse velocity $\sqrt{2kT/m}$ which is representative of the Maxwellian distribution of emission. The symbol U_ℓ is used for this electron.

By using the property of superposition possessed by the linear differential equation 6, Herrmann is able to express the motion of this representative edge electron as the vector sum of the paths of two electrons: one is a "nonthermal" electron designated $u_{\ell e}$, which leaves the edge of the cathode with zero initial transverse velocity; the other is a "thermal" electron $u_{\ell \sigma}$, which leaves the axis of the cathode with an initial transverse velocity $\sqrt{2kT/m}$.

$u_{\ell e}$ and $u_{\ell \sigma}$ vary with distance along the axis, and it is possible to show that cathode images are obtained at the positions at which $u_{\ell \sigma}$ vanishes. Under certain conditions $u_{\ell e}$ may cut the axis; at these "crossover" positions the beam takes on a Gaussian shape.

The quantities $u_{\ell e}$ and $u_{\ell \sigma}$ may be expressed in terms of X_ℓ and Y_ℓ , the x - y coordinates of the representative edge electron in the Larmor frame, by

$$u_{\ell e} = X_\ell - \frac{i\omega_{\ell c} r_c}{[\omega_{\ell c}^2 r_c^2 + 2kT/m]^{1/2}} Y_\ell$$

$$u_{\ell \sigma} = \frac{i\sqrt{2kT/m}}{\sqrt{2} [\omega_{\ell c}^2 r_c^2 + 2kT/m]^{1/2}} Y_\ell \quad (7)$$

(H26)

where $\omega_{\ell c}$ is the value of ω_ℓ at the cathode.

The moduli of $u_{\ell e}$ and $u_{\ell \sigma}$ are the quantities r_e and σ , respectively, as defined by Cutler and Hines [2]. These authors show that each value of the ratio r_e/σ is associated with a characteristic current density distribution which is obtainable from graphs in their paper. Their paper also gives as a function of r_e/σ the percentage of the total beam current found within any given radius of the beam. Hence once $|u_{\ell e}|$ and $|u_{\ell \sigma}|$ have been obtained as a function of distance along the axis, a complete description of the shape of the thermal beam can be given.

The evaluation of U_ℓ , and hence of $u_{\ell e}$ and $u_{\ell \sigma}$, requires a knowledge of the behavior of $\bar{\rho}$ which in itself depends on r_e and σ .

Herrmann assumes that ρ is uniformly distributed across an effective beam radius R such that

$$\bar{\rho} = I / \pi R^2 \dot{z} \quad (8)$$

(H22)

where I is the total beam current. R is a function of r_e and σ and is assumed to be given by

$$R^2 = r_e^2 + 2\sigma^2 = U_\ell \bar{U}_\ell \quad (9)$$

(H23)

where $\bar{U}_\ell$ is the complex conjugate of U_ℓ .

In solving equation 6 it is convenient to replace t by z as the independent variable at this stage. Integration of equation 3 with a mean initial velocity of emission in the z direction of $\sqrt{2kT/m}$ gives

$$\dot{z} = \sqrt{2\eta\Phi(1 + kT/m\eta\Phi)} \quad (10)$$

Substitution for $\bar{\rho}$ from equation 8 and transformation to cylindrical coordinates leads to the following equations for R and θ

$$\begin{aligned} R'' + \frac{\Phi'}{2\Phi(1 + kT/m\eta\Phi)} R' - \left\{ \frac{\Phi''}{4\Phi(1 + kT/m\eta\Phi)} \right. \\ \left. + \frac{P}{4\pi\epsilon_0\sqrt{2\eta}(1 + kT/m\eta\Phi)^{3/2}R^2} \right. \\ \left. - \frac{\omega_\ell^2}{2\eta\Phi(1 + kT/m\eta\Phi)} \left[1 - \left(\frac{\omega_\ell^2 r_c^2 + 2kT/m}{\omega_\ell^2} \right) \frac{r_c^2}{R^4} \right] \right\} R = 0 \end{aligned} \quad (11)$$

$$\theta - \int_{\text{cathode}}^z \frac{\omega_\ell}{\sqrt{2\eta\Phi(1 + kT/m\eta\Phi)}} dz =$$

$$\int_{\text{cathode}}^z \frac{r_c}{R^2} \left[\frac{\omega_\ell^2 r_c^2 + 2kT/m}{2\eta\Phi(1 + kT/m\eta\Phi)} \right]^{1/2} dz \quad (12)$$

Here P is the perveance of the beam. The form of the angular momentum term

$$[\omega_{lc}^2 r_c^2 + 2kT/m] r_c^2$$

is due to an assumption by Herrmann. X_ℓ and Y_ℓ are found by equating real and imaginary parts in

$$X_\ell + iY_\ell = U_\ell = R \exp \left\{ -i \int_{\text{cathode}}^z \frac{r_c}{R^2} \left[\frac{\omega_{lc}^2 r_c^2 + 2kT/m}{2\eta \Phi (1 + kT/m \eta \Phi)} \right]^{1/2} dz \right\} \quad (13)$$

Equations 11 and 12 can in general only be solved by numerical methods for arbitrary variations of Φ and ω_ℓ . A computational scheme involving approximations to the potential variation which is appropriate to low-perveance beams was used in the comparison between theory and experiment of Section III. This scheme is described in the Appendix where a table of the parameters used in the calculations is also found.

It is possible, however, to draw certain conclusions by inspecting the above equations and solving them in the special case where Φ and ω_ℓ are constant. In this case a stable radius R_o may be obtained by proper injection of the beam into the uniform field region with the initial condition $R = R_o$, $dR_o/dz = 0$. The radius R_o is given by

$$R_o = R_{BR} \left\{ \frac{1}{2} + \frac{1}{2} \left[1 + 4 \left(\frac{\omega_{lc}^2 r_c^4}{\omega_\ell^2 R_{BR}^4} + \frac{2kTr_c^2}{m\omega_\ell^2 R_{BR}^4} \right) \right]^{1/2} \right\}^{1/2} \quad (14)$$

where the Brillouin radius R_{BR} is given by

$$R_{BR}^2 = \frac{\sqrt{2} P \Phi}{\pi \epsilon_0 \eta^{3/2} B^2} \quad (15)$$

The small axial temperature correction is neglected here and in subsequent expressions. Its introduction into equations 11 and 12 is necessary to prevent the singular behavior of these equations near the cathode. Herrmann defines a "Brillouin field" B_{bo} as

$$B_{bo}^2 = \frac{R_{BR}^2}{R_o^2} B^2 \quad (16)$$

which, by suitable rearrangement of equation 12, may be used to rewrite equation 13 as

$$X_{\ell} + iY_{\ell} = R_o \exp \left(-i \left[\frac{\omega_{\ell}}{\sqrt{2\eta\Phi}} \left(1 - \frac{B_{bo}^2}{B^2} \right)^{1/2} + \delta \right] \right). \quad (17)$$

δ is here a phase angle.

Small perturbations in the entrance conditions to the uniform region lead to a periodic ripple on R_o and cause the following modification to equation 17

$$X_{\ell} + iY_{\ell} = R_o [1 + a \cos(\phi z + \mu)] \exp[-i(\psi z + \delta)]$$

where

$$\phi = \frac{2\omega_{\ell}}{\sqrt{2\eta\Phi}} \left[1 - \frac{1}{2} \frac{B_{bo}^2}{B^2} \right]^{1/2} \quad (18)$$

and

$$\psi = \frac{\omega_{\ell}}{\sqrt{2\eta\Phi}} \left[1 - \frac{B_{bo}^2}{B^2} \right]^{1/2} \quad (19)$$

A small perturbation to ψ , which on the average is zero, has here been neglected.

The following expressions are now obtained for r_e and σ

$$r_e = R_o [1 + a \cos(\phi z + \gamma)] \left\{ \cos^2 \psi z + \frac{\omega_{\ell c}^2 r_c^2}{[\omega_{\ell c}^2 r_c^2 + 2kT/m]} \sin^2 \psi z \right\}^{1/2} \quad (20)$$

$$\sigma = \frac{R_o}{\sqrt{2}} [1 + a \cos(\phi z + \gamma)] \left\{ \frac{2kT/m}{[\omega_{\ell c}^2 r_c^2 + 2kT/m]} \sin^2 \psi z \right\}^{1/2} \quad (21)$$

where γ accounts for the relative phase between μ and σ . Cathode images occur where σ vanishes and are therefore separated by a distance λ_I , given by

$$\lambda_I = \pi/\psi \quad (22)$$

B. Theoretical Conclusions from Herrmann's Theory

(1). The Effects of Geometric and Voltage Scaling. It is often necessary to conduct the experimental investigations of electron

beams on scaled up models and at reduced voltages; it is therefore important to know the scaling laws.

Let us consider a geometric scaling under the conditions that voltage and perveance remain constant. Equations 11 and 12 or 13 are unaltered when the dimensions R , r_c , and z are all multiplied by a factor M if the magnetic field is everywhere multiplied by a factor of $1/M$. The shape of the thermal beam is therefore unaltered provided the ratio $\omega_\ell R$ is kept constant.

In the absence of thermal velocities identical trajectories are obtained from equations 11 and 12 or 13 on voltage scaling if the ratio ω_ℓ^2/Φ is kept constant. The inclusion of the temperature terms makes voltage scaling impossible; in practice the presence of a large temperature term may lead to sufficiently different trajectories to make experimentally found conditions which lead to minimum beam interception at one voltage invalid for a greatly different voltage.

(2). The Conditions for optimum Beam Transmission. At each value of z the effective beam radius R has an associated specific value of r_e/σ which determines the beam contour. The relationship between R and the "beam edge," defined by a radius R_x which encloses a fraction x of the total beam current, is obtained from an equation of the form

$$R_x = f(r_e/\sigma, x) \sigma \quad (23)$$

where σ is a function of R . Here $f(r_e/\sigma, x)$ is one of a set of functions tabulated by Cutler and Hines [2]. Using equations 7 and 13, R_x becomes

$$R_x = f(r_e/\sigma, x) R \left| \sin \int_{\text{cathode}}^x \frac{r_c}{R^2} \left[\frac{\omega_\ell^2 r_c^2 + 2 kT/m}{2 \eta \Phi} \right]^{1/2} dz \right| \quad (24)$$

The problem of optimizing beam transmission therefore reduces to the problem of minimizing the excursion of R_x for a given magnetic field. Since r_e/σ is a function of $\omega_\ell r_c$ and T as well as of the local value of ω_ℓ , each beam must in general be treated on its own merit; it is possible, however, to discuss the often encountered special case of the beam in a uniform field—with and without flux at the cathode.

If the beam is injected into the uniform magnetic field under the proper conditions to obtain a stable value R_0 for the effective

beam radius, and if flux is excluded from the cathode, a characteristic current density variation between cathode images is obtained which is independent of the operating voltage. This follows from equations 20 and 21 which for $\omega_{\ell c} = 0$ give r_e/σ as

$$r_e/\sigma = \sqrt{2} |\cot \psi_z| \quad (25)$$

A set of theoretical current density profiles between two cathode images which corresponds to a beam with no flux in the cathode is shown in Fig. 1. Since the profiles bear a constant relationship to each other, the relative variation of current density on

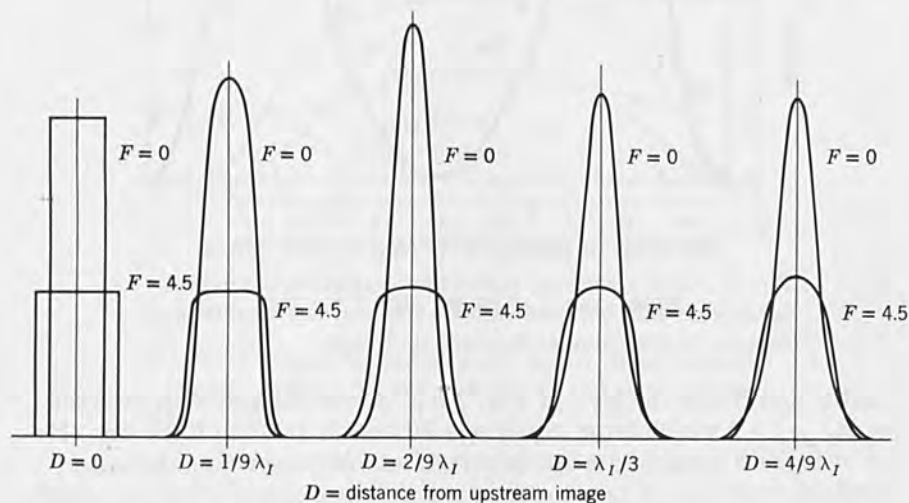


Fig. 1. Theoretical current density profiles between cathode images showing the effect of flux ($F \neq 0$) in the cathode under otherwise identical conditions.

the axis of symmetry is likewise independent of voltage and has the form shown in Fig. 2. Although r_e/σ is not a function of voltage, λ_I is, and increases without limit as the voltage is raised. Consequently, at the higher voltages the current density profiles change with distance sufficiently slowly that they appear constant.

The axial variation of R_x between images is shown in Fig. 3 for two values of x , $x = 0.995$, and $x = 0.95$. These curves are in essence a different presentation of the data shown in Fig. 7 of

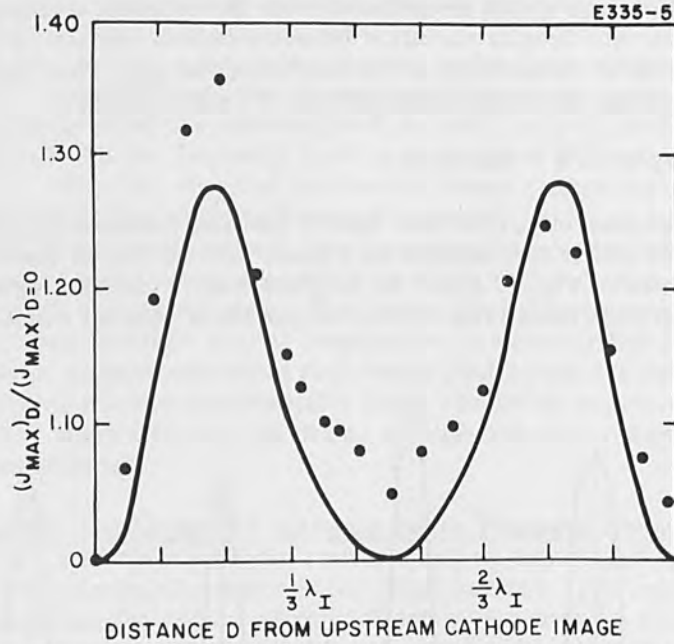


Fig. 2. Graph showing theoretical variation of current density on axis between cathode images, normalized with respect to the value at the cathode image.

Cutler and Hines' paper. In Fig. 3 R_x is normalized with respect to $(R_0)_{\omega_{lc} = 0}$ which from equations 20 and 21 is seen to be the value of R_x at a cathode image (where $\sigma = 0$) for the case $\omega_{lc} = 0$, $x = 1.00$.

The maximum values of R_x in Fig. 3 occur for the minimum value of r_e/σ ; since the presence of the ω_{lc} term in equation 20 prevents r_e/σ from becoming zero it appears that cathode flux may be of benefit in reducing the excursion of the beam edge. From equation 14 one finds that R_0 increases with ω_{lc} and although $f(r_e/\sigma, x)$ in equation 24 decreases with increasing ω_{lc} , it is in general not possible to predict whether or not the product of $R_0 f(r_e/\sigma, x)$ decreases with increasing ω_{lc} . In the specific example plotted in Fig. 3 in which the flux factor F defined by

$$F = \frac{m\omega_{lc}^2 r_c^2}{2kT} \quad (26)$$

has the value 4.5, the maximum value of the radius enclosing 99.5%

of the current decreases by about 10%. On the other hand, the maximum value of the radius enclosing 95% of the current increases very slightly. The corresponding beam profiles are plotted in Fig. 1 and, as is well known, they show that flux in the cathode produces a beam with a more uniform cross section.

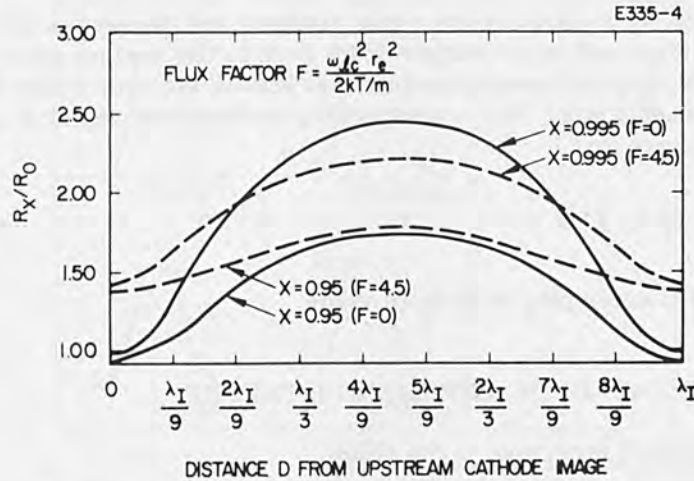


Fig. 3. Theoretical variation in radius R_x containing fraction x of total beam current with and without flux in the cathode.

Inspection of Fig. 3 raises the question of the benefit to be gained by introducing a suitable amount of ripple in order to counteract the spread of the beam edge between the cathode images. The answer to this question depends on the parameters of the given tube. One finds from equations 18 and 19, that the ripple wavelength λ_R does not correspond exactly to the separation between cathode images λ_I . Hence, the region over which the beam scallops inward moves with respect to the portion to be depressed until, after a series of cycles, the opposite effect is achieved. The distance Δz_f over which the scalloping has a favorable effect may be calculated from the distance Δz in which the scallop comes exactly out of phase with a particular beam profile. We have that

$$\begin{aligned}\psi \Delta z &= n\pi \\ \phi \Delta z &= (2n+1)\pi\end{aligned}$$

From equation 19 we then find that

$$\Delta z = \frac{\frac{\pi}{2} \sqrt{\frac{2\eta\Phi}{B^2}}}{\left\{ \left[1 - \frac{1}{2} \frac{B_{b0}^2}{B^2} \right]^{1/2} - \left[1 - \frac{B_{b0}^2}{B^2} \right]^{1/2} \right\}} \quad (27)$$

The scalloping is effective over this distance and decreases the beam by an amount which ranges from zero to the scallop amplitude a . To insure a benefit the decrease should not drop below a value of roughly $a/2$. The corresponding confinement region Δz_f must therefore be

$$\Delta z_f \leq \frac{1}{2} \Delta z \quad (28)$$

if controlled scalloping is to be of value.

III. VERIFICATION OF HERRMANN'S THEORY

A. Existence of Structure in the Beam

The theory predicts the existence of a well-defined structure at all locations of the beam. Ashkin [3] has observed a persisting structure in exceedingly low-perveance beams at several times the Brillouin value of the magnetic field. Beam studies by Brewer [4] and Gilmour [5] of high-perveance beams ($P > 1.0 \times 10^{-6} \text{ A/V}^{3/2}$) confined by fields of approximately the Brillouin value have failed, however, to reveal the characteristic structure of Fig. 1. In the cases [4] where a structure is observed, it is attributed to the existence of a separate "translaminar" stream which passes through the otherwise laminar main stream of electrons. Beam profiles with approximately the predicted structure have been observed by Palmer [6]. These profiles fade rapidly with increasing distance from the gun.

In the present series of experiments a well-defined structure was observed which degenerated only slowly with increasing distance. Figure 4 shows a series of profiles at 8.0 kV between the first and second cathode image. Figure 5 shows successive cathode images at this voltage; only a small degree of fading can be observed.

Figure 5 includes cathode images and crossovers at lower

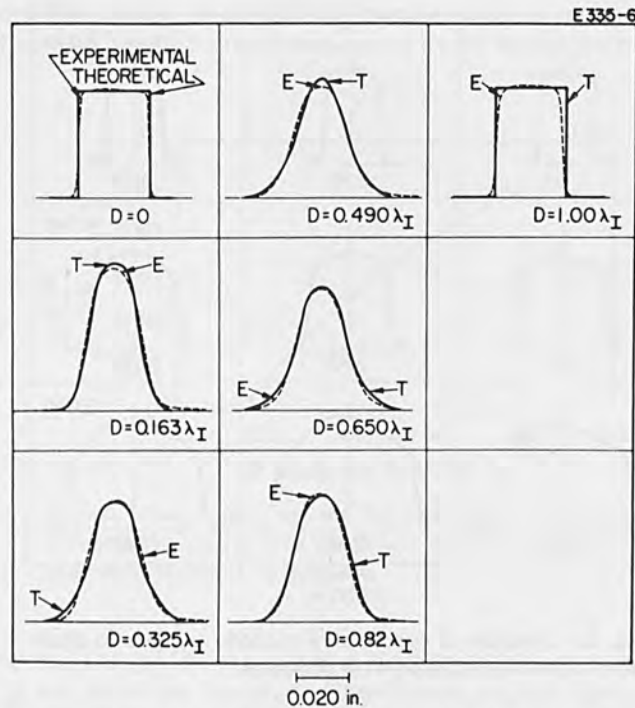


Fig. 4. Comparison between the computed and the measured current density profiles between first and second cathode image with entrance conditions adjusted to make ripple amplitude $a = 0$.

beam voltages. With reduced beam voltage the sharpness of the images is seen to decrease. The reason for the degradation has not been investigated. However, there is evidence to suggest that it is owing to emission from the gap between the focus electrode and the cathode.

B. Effect of Voltage Scaling

Several series of profiles were taken at beam voltages ranging from 2.0 to 8.0 kV. In order to compare the results at the various voltages, conditions were adjusted in each experiment to minimize the ripple on R_0 in the region of uniform magnetic field. This condition was judged to have been achieved when the current density variation on the beam axis between cathode images took the form shown in Fig. 2. At 8.0 kV a close approximation to this

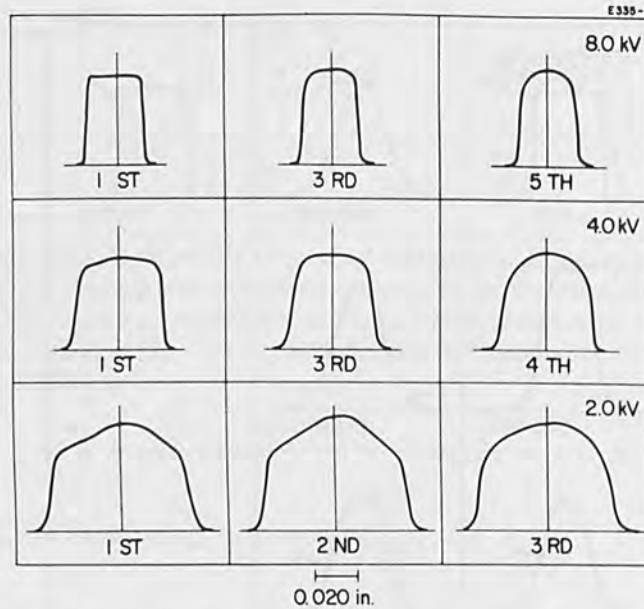


Fig. 5. Display of successive cathode images to show their slow degradation with distance.

curve could be achieved by successive adjustments on the gun-pole-piece distance and of the magnitude of the magnetic field. A comparison between the theoretical and the best experimental variation is also shown in Fig. 2. At the lower beam voltages an insufficient amount of axial adjustment of the gun was available to obtain the correct gun-pole-piece spacing, and only the magnetic field could be varied to produce the required axial current density variation. However, for a small degree of rippling the adjustment of the magnetic field is nearly independent of the error in the gun placement; the magnetic field so found should therefore be very close to the value required to produce a ripple-free beam at the various voltages. The values of the magnetic field were therefore used to investigate the variation of ω_p^2/Φ with voltage. Figure 6 shows the marked departure with voltage of the parameter ω_p^2/Φ from a constant value which is due to thermal velocity effects.

C. Comparison between Predicted and Measured Current Density Profiles and Their Location

The comparison of the theoretical and experimentally obtained current density profiles at 8.0 kV in Fig. 4 shows the good agree-

ment between them. A numerical comparison is more easily made by plotting the predicted and measured axial variation of $R_{1/2}$ and

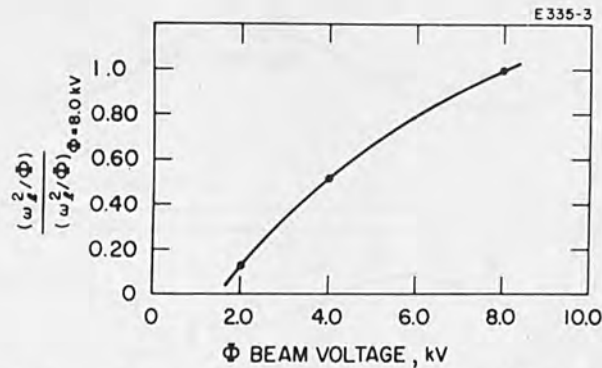


Fig. 6. Graph showing the departure of ω_c^2 / Φ from a constant value due to variation in effects associated with thermal velocities.

$R_{1/10}$, the radii at which the current density has dropped to 1/2 and 1/10th of the value on the axis. These radii rather than, for example, $R_{99.5}$ and R_{95} , were chosen for purposes of comparison since a considerable amount of numerical integration is involved in obtaining the latter. The theoretical relationship between the various radii $R, R_{1/2}, R_{1/10}, R_{95}$, and $R_{99.5}$ may be obtained from Fig. 7, plotted in this case for the beam at 4.0 kV. The theoretical and experimental values of $R_{1/2}$ and $R_{1/10}$ at 2.0, 4.0, and 8.0 kV are compared in Fig. 8.

The percentage errors ϵ between a computed and a theoretical radius defined by

$$\epsilon = \frac{|(R_x)_{\text{experimental}} - (R_x)_{\text{computed}}|}{(R_x)_{\text{computed}}} \times 100 \quad (29)$$

are compared for various voltages in Table I.

Apart from any errors introduced into the theory by approximations to the potential variation, these error figures include, of course, the usual small experimental errors due to inaccuracies in the measurement of the voltage, the solenoid current, and the beam temperature. Also included are errors associated with the

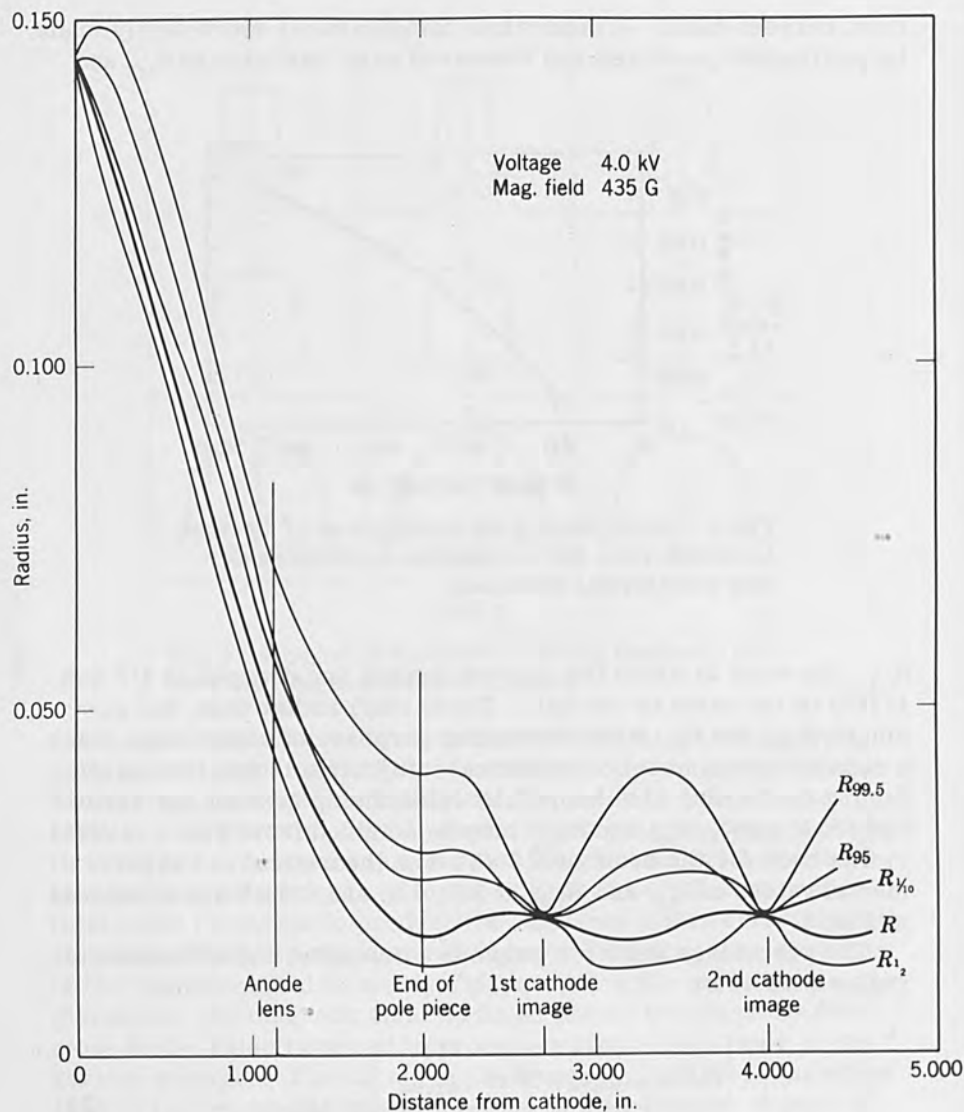


Fig. 7. Theoretical relationship between radii R , $R_{99.5}$, R_{95} , $R_{1/2}$, and $R_{1/10}$.

inertia of the pen of the $x - y$ recorder and the broadening of the profiles due to the finite diameter of 0.002 in. of the scanning aperture. The experiments were conducted in a vacuum of $\cong 8 \times 10^{-7}$ torr; errors due to collision processes were therefore held to be negligibly small.

Quite appreciable errors in the measurement of the beam voltage, of the cathode temperature, and of the value of the uniform

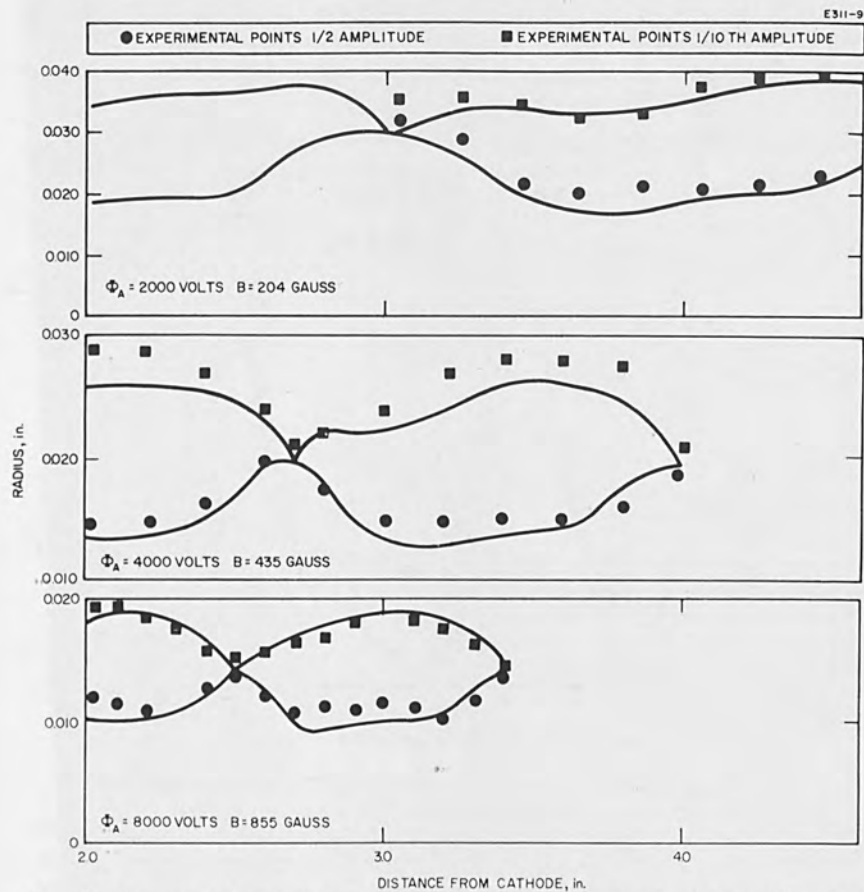


Fig. 8. Comparison between measured and theoretical values of $R_{1/2}$ and $R_{1/10}$ at various voltages.

Table I

Percentage Difference between Computed and Measured
Values of $R_{1/2}$ and $R_{1/10}$

Voltage, kV	2.0		4.0		8.0	
	Amplitude					
	1/2	1/10	1/2	1/10	1/2	1/10
Maximum ϵ	30	24	12	14.5	20	8.0
Mean ϵ	21	7.6	6	10.4	5.5	3.5

magnetic field have only small effects as may be seen from Fig. 9. This figure compares the measured values of $R_{1/2}$ at 4 kV with those computed with 5% variations in the above parameters. Imper-

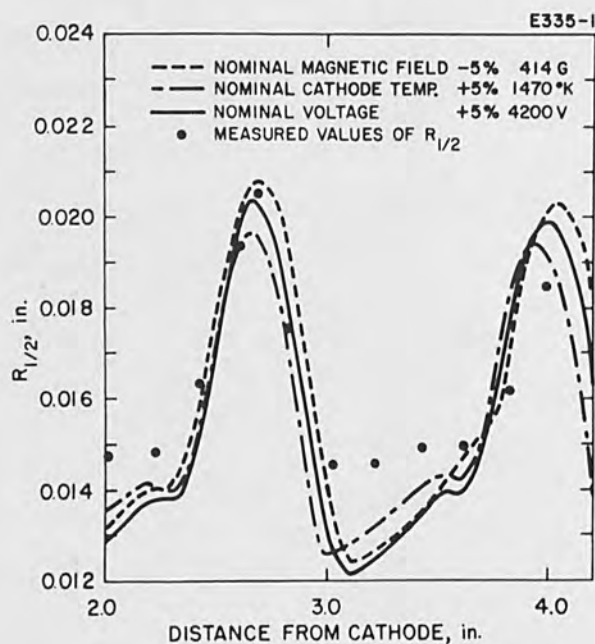


Fig. 9. Graph showing the effect on $R_{1/2}$ of computed departures of the magnetic field, the beam voltage, and the cathode temperature.

fections in the gun lead to significant discrepancies between theory and experiment; this point is further discussed below in Section III-D.

The positions at which $R_{1/2}$ and $R_{1/10}$ have the value R are cathode images. The predicted and measured locations of the cathode images at the above voltages are compared in Table II. The values are seen to be exceedingly close.

D. Effect of Errors Associated with the Electron Gun

Some of the discrepancies between the theoretical and experimental beam profiles are due to the neglect of higher order terms in the calculations. One important source of such errors arises from the calculation of electric fields which are based on the as-

sumption of a uniform current density distribution across the beam. For a gun which is emitting uniformly, such a distribution is (to a good approximation) obtained at a cathode image; although this distribution degenerates into a Gaussian distribution between images, it remains reasonably uniform over almost a third of the distance between images. The resulting departure of the electrons from their paraxial paths would therefore appear to be small, and the distribution of electrons should be close to that of the Cutler-Hines curves which are computed for an initially uniform current density distribution.

Table II
Comparison of Measured and Computed
Positions of Cathode Images

Voltage, kV		2.0		4.0		8.0	
Cathode Image No.	Comp.	Distance from Cathode, inches					
		Meas.	Comp.	Meas.	Comp.	Meas.	
1	3.145	3.05	2.710	2.720	2.530	2.520	
2	4.980	4.950	4.00	4.035	3.440	3.440	
3	6.940	6.850	5.270	5.340	4.360	4.325	
4			6.560	6.650	5.270	5.215	
5					6.220	6.180	

In the case of a nonuniformly emitting electron gun the nonuniformity results not only in an additional perturbation of the trajectories but it also modifies all of the Cutler-Hines curves. Quite small errors in the placement of the cathode with respect to the focus electrode cause appreciable variations in emission. The effect of such an error, an axial displacement of the cathode from its correct position, is shown in Fig. 10. Apart from causing a highly nonuniform current density distribution across the cathode, the displacement permitted emission from the side of the cathode to take place. This gave rise to a definite translaminar stream which may be seen to propagate through the main stream.

One possible explanation of why cathode images are not always observed is furnished by Fig. 10. The figure shows the extent to which the sharp edges of the cathode image have been rounded off, making them similar in appearance to the beam near the cross-overs.

E279-20

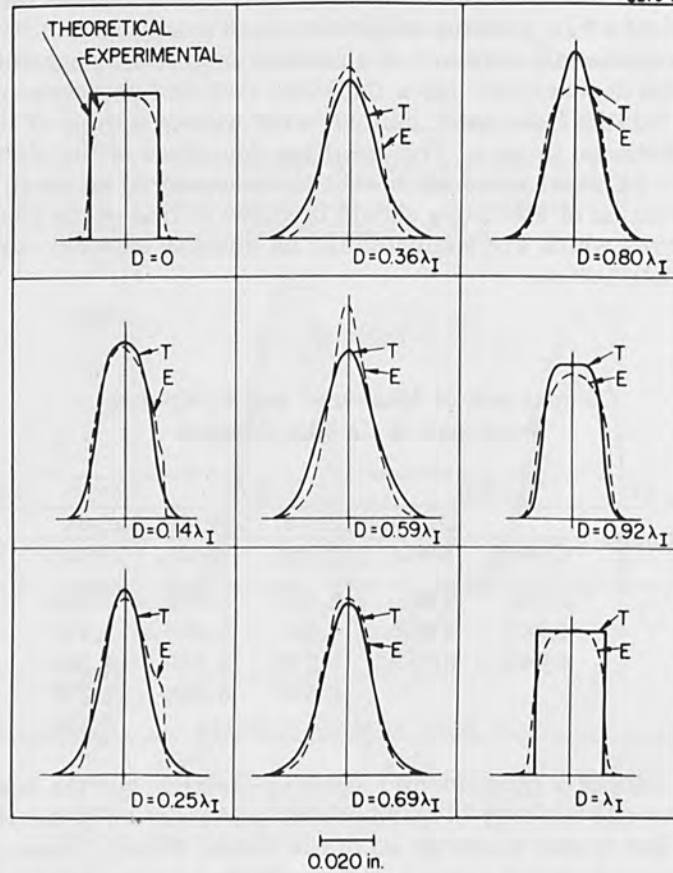


Fig. 10. Comparison between the computed and the measured current density profiles between first and second cathode image.

IV. CONCLUSIONS

The results of an experimental verification of Herrmann's optical theory of thermal velocity effects conducted on a beam of perveance 0.015×10^{-6} amp/ $V^{3/2}$ have been presented. Good agreement between theory and experiment has been noted. The theoretically predicted cathode images appear extremely close to their predicted positions. The cross section of the beam of these positions and elsewhere is also close to that prescribed by theory.

The profiles depend critically on the uniformity of emission. The beam from the same gun with nonuniform emission produced by axial misalignment of the focus electrode shows a marked departure from the predicted profiles.

The structure of the beam fades slowly with distance, an effect which the theory does not predict. The fading is sufficiently slow for the theory to be applied to the problem of optimizing the transmission of thermal beams through long drift tubes. In contrast to the laminar theory, conditions exist under which flux in the cathode can lead to higher beam transmission; in addition, the length of the drift tube is a parameter which affects the optimization of beam transmission.

APPENDIX--APPROXIMATE METHOD OF OBTAINING THE FIELDS FOR THE SOLUTION OF THE PARAXIAL SPACE CHARGE RAY EQUATION

The electric potential and the magnetic field on the axis of symmetry need to be given for the computation of equations 11 and 12. For the results given in Section III an experimentally obtained magnetic field variation was used which was supplied to the digital computer in tabular form. A smooth function was generated from these data by means of a second degree polynomial interpolation routine. The electric potential was given in analytic form and was calculated by separating the flow into the three regions of Fig. 11. In region (1), the accelerating region of the gun, the variation was assumed to be the same as that in a spherical diode and of the form

$$\Phi = \Phi_A \left(\frac{\alpha}{\alpha_A} \right)^{4/3} . \quad (\text{A.1})$$

Here α is the appropriate Langmuire [7] function and the subscript A denotes values of the anode. The curved cathode surface implies that the r_e and σ trajectories have different initial axial positions; this fact was ignored in the computation. In the drift region (3) the potential was set equal to Φ_A . Region (2) was assumed to be occupied by an infinitely thin electron lens which altered the slope of R but left all other quantities unchanged. The focal length F of this lens was computed from the Davisson-Calbick [8] thin lens formula.

E311-6

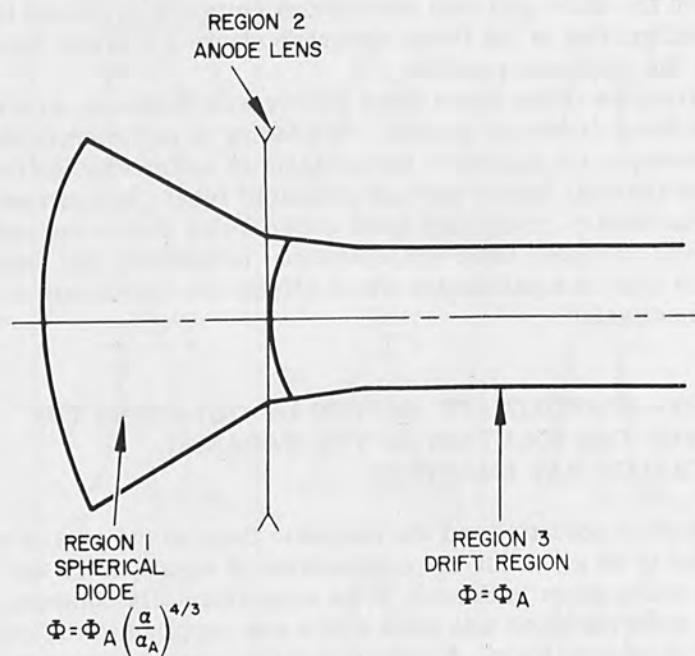


Fig. 11. Diagram showing the separation of the flow into three regions for computational purposes.

$$\frac{1}{F} = K \left(\frac{\Phi'}{4\Phi} \right). \quad (\text{A.2})$$

The factor K which does not appear in the original formula is a lens correction factor which could be supplied to the computer. Shown below are the values of K and other parameters used in the computation of the theoretical results given in Section III.

Radius of cathode sphere	1.660 in.
Radius of anode sphere	0.500 in.
Radius of cathode	0.145 in.
Semiangle of convergence	0.087 rad
Temperature of cathode	1400°K
Radius of pole piece hold	0.062 in.
Distance from cathode to end of pole piece	1.995 in.
Anode lens correction factor K	1.025
Magnetic field at the cathode	0.0 G

REFERENCES

1. G. Herrmann, J. Appl. Phys. 29, 127-138, February 1958.
2. C. C. Cutler and M. E. Hines, Proc. IRE 43, 307-315, March 1955.
3. A. Ashkin, J. Appl. Phys. 29, 1594-1604, November 1958.
4. G. R. Brewer, J. Appl. Phys. 30, 1022-1038, July 1959.
5. A. S. Gilmour, Jr., "A Beam Tester for Studying the Characteristics of D-C and Velocity-Modulated Electron Beams," Cornell University School of Electrical Engineering, Res. Report EE 495, 1961.
6. J. L. Palmer, "Effects of Thermal Velocities in Magnetically Focused Electron Beams" (University of California, Berkeley), ASD Tech. Report 61-125, 1961.
7. I. Langmuir and K. B. Blodgett, Phys. Rev. 24, 49-59, July 1924.
8. J. C. Davisson and J. C. Calbick, Phys. Rev. 38, 585, August 1, 1931; and Phys. Rev. 42, 580; November 15, 1932.

EXPERIMENTAL INVESTIGATION OF THE STRUCTURE OF HIGH-POWER-DENSITY ELECTRON BEAMS

INTRODUCTION

A beam generated in an electron gun is defined by the following three quantities: (1) the position of the crossover on the system axis, (2) the current density distribution in the crossover plane, and (3) the solid angle distribution of the beam current coming from a surface element of the crossover. If the crossover C_0 , as shown on Fig. 1, is projected through a focusing lens in a focal point C_1 , knowledge of all three quantities is necessary for evaluating the current density distribution at that point, if the spherical aberrations of the lens must be considered. In such a system, the angular distribution of the beam current will be derived, for the sake of simplicity, from the current density distribution in the plane of an aperture diaphragm Ap . It must not be forgotten, however, that the space angle distribution of the current j_a , calculated in such a way will always depend to some extent on the position of the diaphragm.

We introduce the following definition for the part of the beam current coming from the central part of the crossover of radius r :

$$\frac{I_r(r)}{I_{tot}} = \frac{\int_0^r j_{cr}(r) 2\pi r dr}{\int_0^\infty j_{cr}(r) 2\pi r dr} = \psi(r) \quad (1)$$

Similarly, we get for the part of the beam current remaining inside the angle α :

$$\frac{I_a(\alpha)}{I_{tot}} = \frac{\int_0^\alpha j_a(\alpha) 2\pi \alpha d\alpha}{\int_0^\infty j_a(\alpha) 2\pi \alpha d\alpha} = \psi_a(\alpha) \quad (2)$$

*Department for Industrial Research, Swiss Federal Institute of Technology (ETH), Zurich, Switzerland.

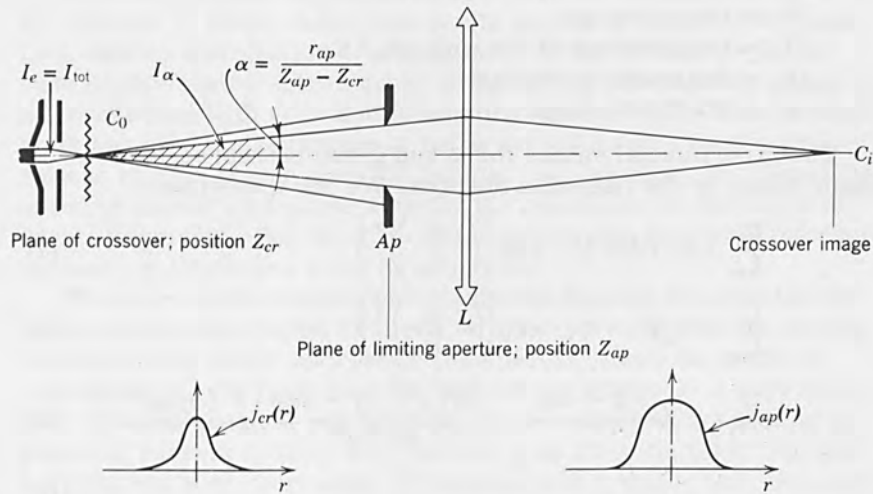


Fig. 1. Definition of the characteristic parameters of an electron beam coming from an electron gun.

Therefore, the beam current $I_{r,a}$ emitted by the area πr^2 of the crossover into the space angle $\pi \alpha^2$ can be expressed thus:

$$I_{r,a} = I_e \psi_r(r) \cdot \psi_a(\alpha) \quad (3)$$

We then get the following relation for the mean brilliancy of the crossover, usually designated in electron optics as the "Richtstrahlwert" $K_{r,a}$:

$$K_{r,a} = \frac{I}{\Delta F \cdot \Delta \Omega} = I_e \frac{\psi_r(r) \cdot \psi_a(\alpha)}{\pi^2 r^2 \alpha^2} \quad (4)$$

If r and α are decreased until zero, the maximum "Richtstrahlwert," $K_{\max}$ is obtained:

$$K_{\max} = \lim_{\substack{r \rightarrow 0 \\ \alpha \rightarrow 0}} K_{r,a} \quad (5)$$

For $K_{\max}$ the theory of beam generation gives a simple relation [1], [2]:

$$K_{\max} = \frac{j_e}{\pi} \frac{eV}{kT} \quad (6)$$

where j_e = emission current density of the cathode

V = beam voltage

T = temperature of the cathode, °K

k = Boltzmann's constant

e = electron charge

The experimental values for j_r and j_a can be represented in many cases by the Gaussian function. We can then state:

$$\frac{I_r}{I_{\text{tot}}} = \psi_r(r) = 1 - \exp\left\{\left(-\frac{r}{r_n}\right)^2\right\} \quad (7)$$

$$\frac{I_a}{I_{\text{tot}}} = \psi_a(\alpha) = 1 - \exp\left\{\left(-\frac{\alpha}{\alpha_n}\right)^2\right\} \quad (8)$$

$$K_{r,a} = I_e \frac{[1 - \exp\{- (r/r_n)^2\}][1 - \exp\{-(\alpha/\alpha_n)^2\}]}{\pi^2 r^2 \alpha^2} \quad (9)$$

$$K_{\text{max}} = \frac{I_e}{\pi^2 r_n^2 \alpha_n^2} \quad (10)$$

If, instead of r_n and α_n , the half-height widths of the experimental graphs $r_{1/2}$ and $\alpha_{1/2}$ are introduced, then:

$$K_{\text{max}} = 0.049 \frac{I_e}{r_{1/2}^2 \alpha_{1/2}^2} \quad (11)$$

We shall return later to the comparison of the experimental and theoretical "Richtstrahlwert" calculated from equations 11 and 6.

EXPERIMENTAL INVESTIGATION METHOD

The main difficulty in the experimental investigation of high-power electron beams lies in the measurement of the current density distribution in the crossover, where beam power densities can reach several megawatts per cm². There is a possibility of solving this problem by projecting the crossover, electron optically, with great magnification onto the measuring diaphragm, getting in this way a much smaller current density in that image plane. This way, high thermal stress of the measuring diaphragm can be avoided. But it is not always possible to do so; the crossover position is often subject to considerable changes. The position of a virtual crossover can appear far behind the cathode plane; on the other hand, the crossover in telefocus electron guns is located very far in front of the cathode.

A greatly magnifying electron optical system cannot practically be realized in these cases, and one is forced to measure the current density distribution either directly in the crossover (in the case of telefocus electron guns) or in the electron optical image of the crossover with very small magnification ($M \approx 1$). This arrangement has also the advantage of being very similar to some applications of high-power electron beams, e.g., welding, and therefore of being able to reproduce directly the conditions of operation of such equipments. But the difficulties of a strong, thermally stressed measuring diaphragm must be overcome.

The exact measurement of any current density distribution in an electron beam can be realized in principle only with measuring diaphragms of small aperture. The aperture must be small in comparison to the local current density variation. It is well known that the measurement can be made, in the case of a rotation symmetrical current density distribution, with an (infinitely) long and very narrow slot diaphragm by moving such a diaphragm laterally through the beam and recording the beam current passing through the diaphragm aperture.

The calculation of the actual current density distribution from this scanning current in general cases is somewhat time-consuming [3], [4].

As has been already mentioned, the current density distribution in a crossover can be reproduced in many cases by a Gaussian function of the form:

$$j_{cr} = j_{\max} \exp \left\{ - \left(\frac{r}{r_n} \right)^2 \right\} \quad (12)$$

Under these conditions the scanning current i_s is given by the same function:

$$i_s = i_{\max} \exp \left\{ - \left(\frac{r}{r_n} \right)^2 \right\} \quad (13)$$

r being the distance between the measuring diaphragm and the beam axis. We can read the value of the width of the current density distribution directly from the scanning diagram $i_s(r)$.¹

¹ This is only correct if the width of the measuring slot is very small compared to r_n . The exact equation for the scanning current $i_s(r)$ is:

$$i_s(r) / (i_s)_{\max} = \frac{1}{\Phi\left(\frac{s}{2r_n}\right)} \left\{ \Phi\left(\frac{r+s/2}{r_n}\right) - \Phi\left(\frac{r-s/2}{r_n}\right) \right\}$$

where Φ is the error function of the argument in brackets. For $s/r_n = 1/10$, the scanning current calculated according to this formula corresponds to three decimal places to the value calculated from the Gaussian function according to equation 13.

The quantity r_n gives the radius at which the current density has decreased to the fraction $1/e = 0.368$ of the maximum current density in the center. It is more usual to give the half-height width $r_{1/2}$ or $d_{1/2} = 2r_{1/2}$. Then 50% of the entire beam current is located inside $d_{1/2}$, and almost the entire beam current inside $2d_{1/2}$. There follows:

$$r_n = 1.2r_{1/2} = 0.6d_{1/2}$$

In the following, we shall always start from the Gaussian current density distribution and use the half-height width $d_{1/2}$, although these diagrams will not always have the character of Gaussian curves. We are aware that our values for the focal spot diameter $d_{1/2}$ contain a partial uncertainty, and we shall return to it later.

The thermal stress is also very great in the case of slot diaphragms, and the scanning of the beam must be very fast. The measuring system we use is shown in principle on Fig. 2. The crossover of other than telefocus electron guns is projected onto the measuring diaphragm with a magnetic lens. In the rest position, the beam is deflected by the current in a first coil of the magnetic deflecting element into a hole in a water-cooled massive copper holder, and at the same time is defocused by an iron-free auxiliary magnetic lens. In this way, a beam power of more than 3 kW can be dissipated without trouble in the copper-target.

An alternating current of 50 cps flows through a second coil of the deflecting element; the beam is thus wobbled perpendicularly to the slot direction of the measuring diaphragm with an amplitude of about 1.5 mm. For the registration of the scanning diagram, both the constant deflecting current and the defocusing current in the auxiliary lens are switched off for about 0.3 sec by a time relay. The 50 cps deflecting current remains switched on. The beam springs onto the measuring diaphragm and is further wobbled with 50 cps. The current through the 40μ wide slot diaphragm is collected by a Faraday cage and the voltage drop at a measuring resistor is lead to the vertical plates of a cathode-ray oscillograph, whereas the horizontal deflection is derived from the same 50 cps voltage source as the wobbling current. The correct phase between the oscillograph deflection and the wobbling current is achieved with a resistance capacitor circuit; the 50 cps stray fields in the beam path of the electron beam can also be compensated in this way. The undisturbed scanning diagram appears on the oscillograph screen and is photographed.

The deflecting scale for the horizontal deflection is needed for the evaluation; consequently, the measuring diaphragm is mounted

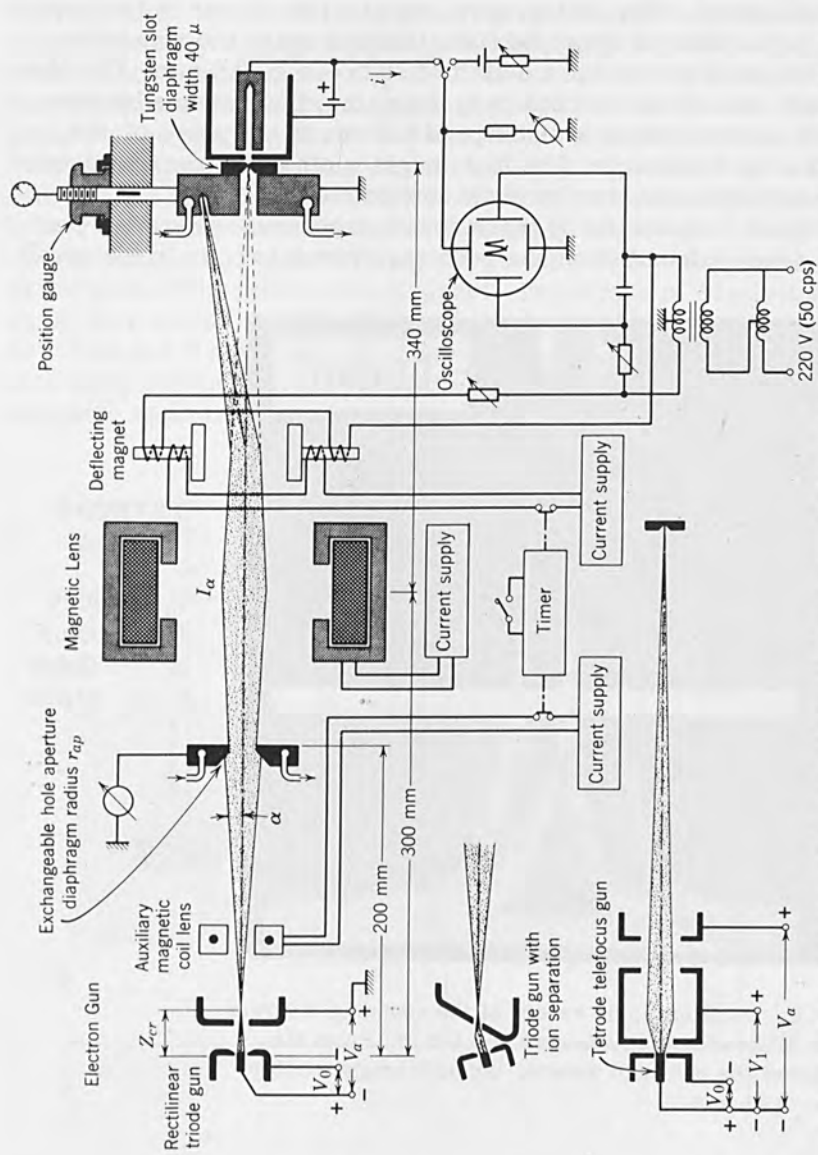
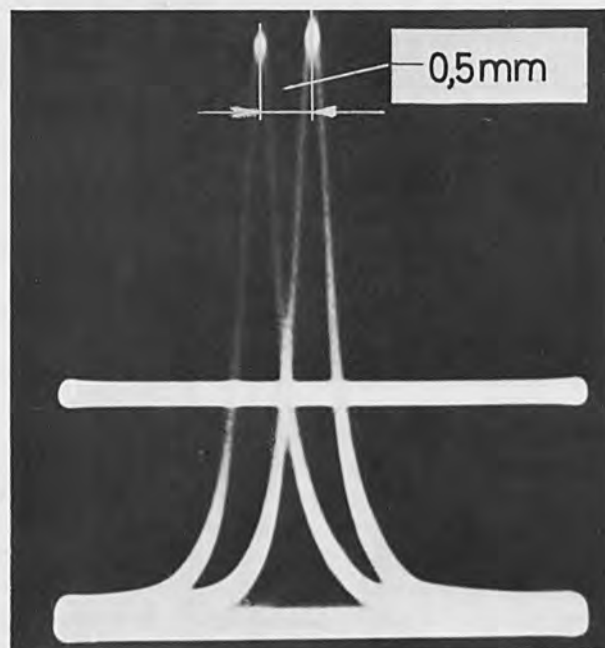


Fig. 2. Schematic diagram of the experimental arrangement for measuring the current density distribution in high-power electron beams.

so as to be very finely adjustable perpendicular to the slot direction. The displacement can be measured very accurately with a position gauge. The photographic registration occurs in two separate expositions of the same film, between which the measuring diaphragm is moved for a definite distance, e.g., 0.5 mm. The photograph then shows two scanning diagrams, the distance between which corresponds to a distance of 0.5 mm in the plane of the measuring diaphragm. The half-height width of the current density distribution can then be given immediately.

Figure 3 shows one of these double exposures, where the current density distribution has been registered directly in the cross-



TETRODE SYSTEM

$$\begin{aligned} V_a &= 20\text{kV} \\ I_b &= 100\text{mA} \\ d_c &= 2\text{mm} \\ S &= 50\mu\text{m} \end{aligned}$$

Fig. 3. Oscillographic record of the scanning diagram with calibration displacement of 0.5 mm. From the diagram one can read directly the half-height width to $d_{1/2} = 0.41\text{ mm}$.

over of a telefocus electron gun at a distance of 330 mm from the cathode. The main magnetic lens had been removed, but otherwise the measuring installation was as described above. The

beam currents was 130 mA at an accelerating voltage of 20 kV.

The second horizontal line in the photograph reproduces the vertical deflection scale; it is generated by an additional exposure when a definite constant current is sent through the measuring resistor from a battery (see Fig. 2). The current calibration is needed when the scanning diagram is integrated for control. To complete the photograph, a fourth exposure gives the zero axis. This short-time measuring circuitry allows registration of current density distributions with a maximum power density of about 3 MW/cm^2 without damage to the measuring diaphragm. The latter consists of two massive tungsten plates which are pressed onto the back of the water-cooled target. Two strips of 40μ tungsten sheet keep them a definite distance apart, yielding a slot aperture 40μ wide and 5 mm long. The edges on the beam side are provided with steep facets to avoid thermal damage. Figure 4 shows the complete measuring diaphragm system.

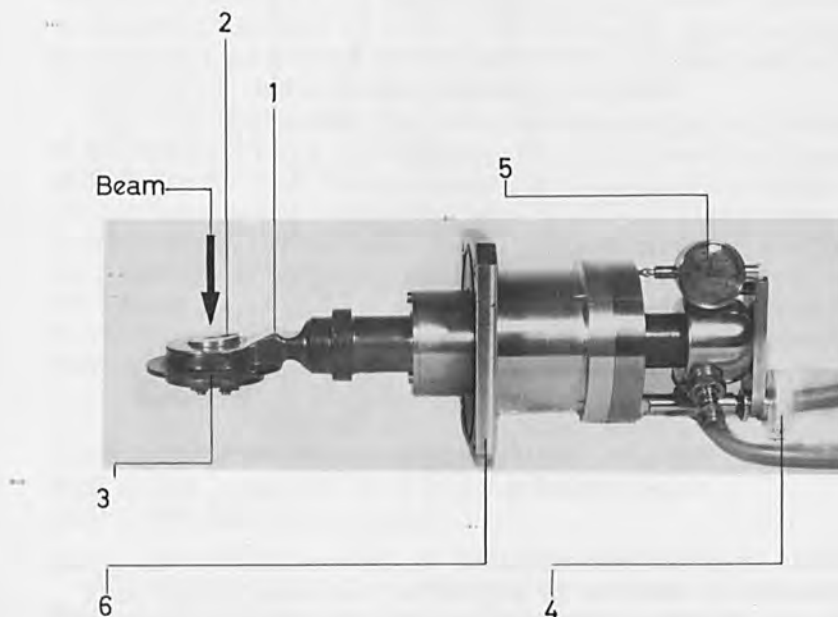


Fig. 4. Photograph of the measuring aperture system. (1) water-cooled holder. (2) Hole for electron beam in rest position. (3) Tungsten plates of the measuring slot diaphragm. (4) Displacement screw. (5) Position gauge. (6) Vacuum flange.

For the determination of the second characteristic value of the beam, i.e., the space angle distribution of the beam current, a water-cooled circular diaphragm of variable radius is introduced in the beam path (Fig. 2). The system of the slot diaphragm is replaced by a Faraday cage; the beam current reaching this cage is registered in function of the emission current for diaphragm diameters of 1, 2.5, 5, 7.5, 10, 12.5, 15, 17.5 and 20 mm with a x - y recorder. Figure 5 shows the recorded diagram.

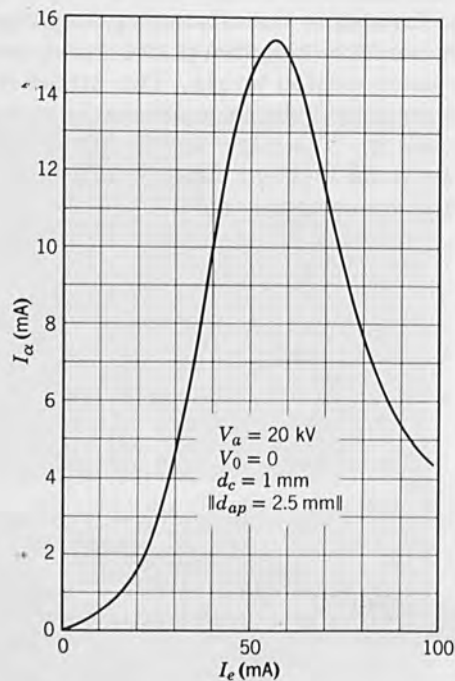


Fig. 5. $I_\alpha - I_e$ diagram as recorded with an x - y recorder.

A third important measurement, the determination of the crossover position, is achieved by registering the shade projection of an aperture diaphragm of 5-mm diameter. For this purpose, the Faraday cage is replaced by a fluorescent screen and the magnetic lens is switched off. The fluorescent material is spread onto a water-cooled copper plate, which is marked with a quadratic grating. The shade projection of the diaphragm on the screen is photographed and the position of the projection center, i.e., of the crossover, is

calculated from this photograph and the radius of the aperture diaphragm. Moreover, the crossover position is also calculated from the focal length of the focusing lens from the first experiment. We shall compare the two results in the following sections.

MEASUREMENT OF THE CURRENT DENSITY DISTRIBUTION IN THE CROSSOVER OF A TELEFOCUS GUN OF TETRODE TYPE IN FUNCTION OF THE BEAM CURRENT

As a first example of the investigation made with the measuring system described above, we would like to mention some results of the measurement of current density distribution directly in the crossover of a telefocus electron gun of the tetrode type. These investigations will be described more fully elsewhere. Figure 6 shows a series of oscillographic recordings for various beam currents. The accelerating voltage is 20 kV and the radius of the cathode 2 mm. The scanning diagrams shown can be well represented by Gaussian functions according to equation 12, so that a characterization of the current density distribution is justifiable by using the half-height width of these scanning diagrams.

The half-height width $d_{1/2}$ is represented on Fig. 9 in function of the beam current I . The voltage of the first anode is changed to achieve the focusing of the beam on the measuring diaphragm, and the variation of this voltage can also be seen on Fig. 9. The half-height width at 150 mA is 0.3 mm; thus, a maximum power density (in the center of the crossover) of about 3 MW/cm² was achieved for a beam power of 3 kW. That is approximately the power limit at which the equipment described here can be operated without fear of destroying the measuring diaphragm.

PROPERTIES OF THE ELECTRON BEAM IN FUNCTION OF THE BEAM CURRENT IN A TRIODE ELECTRON GUN WITH ION SEPARATION

As a second example, a beam coming from an electron gun shall be characterized according to the points mentioned at the beginning. The electron gun is of the triode type and built according to the principle of ion separation described elsewhere [3], [5]. The beam properties described in the following remain valid for a triode electron gun of conventional structure.

First, Fig. 10 shows the position of the crossover in function

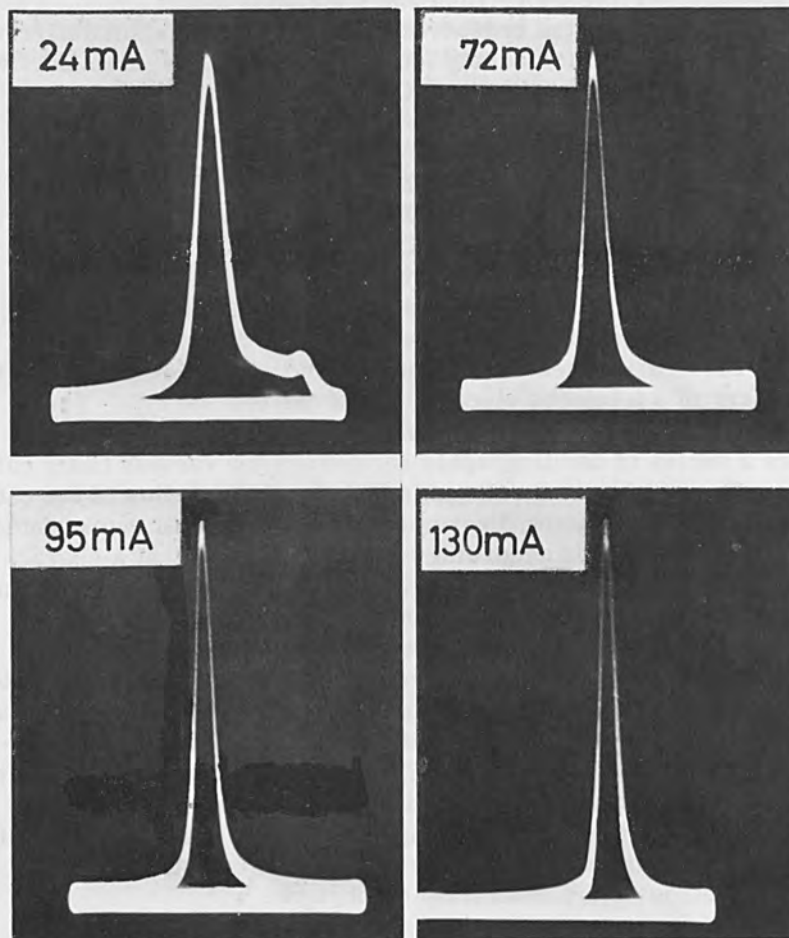


Fig. 6. A series of scanning diagrams giving the current density distribution directly in the crossover of a telefocus electron gun of tetrode type. The beam currents are indicated on the diagrams.

$V_{a''} = 20\text{kV}$, $d_c = 1.5\text{ mm}$, $Z_{cr} = 330\text{ mm}$.

of the emission current. The Wehnelt cylinder is kept constantly at the cathode potential. The position of the crossover is referred to the position of the cathode as origin. The solid line is obtained from the point projection of the aperture diaphragm, the dotted line represents the values calculated from the focusing current of the magnetic lens. The crossover position for 1 mA emission current,

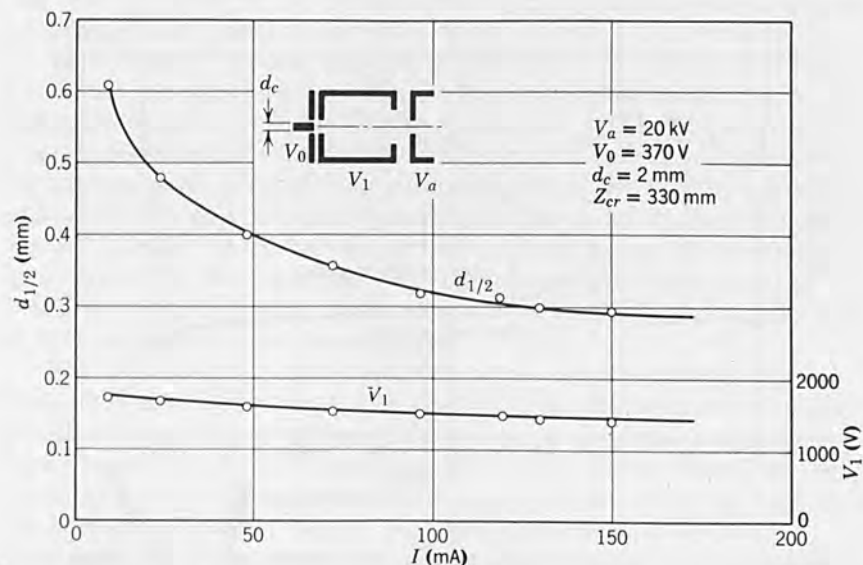


Fig. 7. Half-height width $d_{1/2}$ of the current density distribution in the cross-over of a telefocus electron gun of tetrode type as a function of beam current I . The focusing of the beam at a distance of 330 mm from the cathode occurs by adjusting the first anode voltage. The change of this voltage V_1 is also given in the diagram.

calculated from the point projection, was used as standard for focal length calibration. We can see from Fig. 10 that the crossover moves rather extensively when the emission current is changed. Two reasons can be given for this alternating movement: first, the position of the emitting surface, relative to the Wehnelt cylinder, is changed by the stronger heating of the cathode, and thereby influences the potential field of the cathode. This axial translation of the emitting surface z_c , referring to the position at an emitting current of 1 mA, is also shown on Fig. 10; at 100 mA, this displacement amounts to about 60μ . Since stronger cathode heating also causes the Wehnelt cylinder to expand, the relative displacement of the emitting surface in respect to the Wehnelt cylinder will be smaller still.

The second effect is the space charge in the electron beam which also influences the potential field in the electron gun, the influence being greatest immediately in front of the cathode. The emission current of 100 mA corresponds to an emission current density of

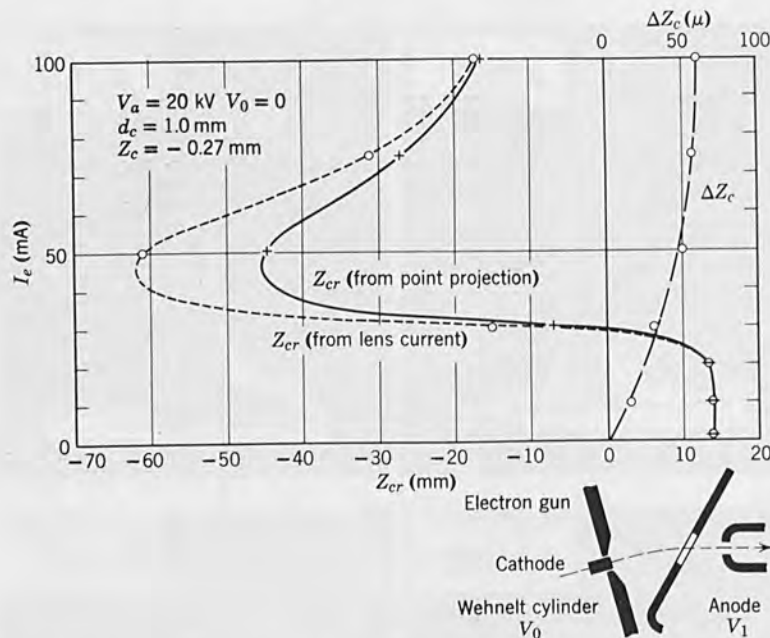


Fig. 8. Crossover position Z_{cr} as a function of the emission current I_e in a triode electron gun. Full line gives Z_{cr} as calculated from the point projection experiment, dotted line gives Z_{cr} as calculated from the focusing current of the magnetic lens. ΔZ_c = relative displacement of the emission surface.

about 13 amp/cm². The space charge density in front of the cathode is therefore very high. The compensation of the negative electron charge with positive ions is very slight, since the latter are eliminated very fast from the beam region. For the moment, no satisfactory explanation can be given for the form of the z_c - I_e curve on Fig. 10. Work is in progress to measure the potential distribution in a resistance network for the rotation-symmetrical case and to calculate from it the electron optical properties of the electron gun.

The discrepancy between the coordinates for the crossover, obtained from the point projection, and the values given by the lens formula of the magnetic lens, is remarkable. The point projection gives the true position of the (real or virtual) crossover; the discrepancy mentioned means that the magnetic lens must be more weakly excited for the projection of the crossover on the measuring diaphragm than would be necessary according to the geometri-

cal lens formula. It can be seen from Fig. 10 that the discrepancy is greatest at a beam current of about 50 mA. At an emission current of about 50 mA, the aperture angle of the beam is at its minimum, as will be shown later. The space charge density in the beam becomes very high. The repulsing forces of the electron charge between the lens and the measuring diaphragm might be noticed in spite of the strong compensation of the negative space charge by the positive ions generated in the beam (pressure: about 2×10^{-5} torr). The beam would then be spread, the focus moving away from the lens; the magnetic lens would have to be excited more strongly to bring it back onto the measuring diaphragm. But in fact we observe just the opposite.

The explanation of our observation can only be correct if we assume that the electron space charge is overcompensated by the positive ions. This overcompensation can come from the copper vapor beam that we are observing when the electron beam springs from the rest position to the measuring aperture over the edge of the hole in the copper target. An overcompensation of electron space charge of the same kind can be observed during a welding operation.

The results of the measurements of the angle distribution of the beam current are shown on Fig. 11. The solid lines represent the adjusted Gaussian curves according to equation 8. The r_n values of these adjusted curves are also given in the diagrams. The Gaussian distribution is valid with good approximation for the nucleus of the beam. For $I_e = 75$ mA the agreement is very good for the whole of the beam. For $I_e < 75$ mA, there is a beam current excess for higher angle values, and for $I_e > 75$ mA, there is a beam current shortage in respect to the Gaussian distribution. No well-defined explanation can be given for this, but it seems that this behavior of the beam is connected with lens aberrations of the electron gun. We shall return to this later.

We can already see on Fig. 11 that the beam has its smallest aperture angle for $I_e = 50$ mA ($\alpha_n = 6.8 \times 10^{-3}$ rad, as against 28.2×10^{-3} rad for 100 mA). The space charge density in the beam is proportional to the expression I_e/α_n^2 ; therefore the space charge density in the 50 mA beam is about nine and a half times greater than in the 100 mA beam. This has been already mentioned above.

Figure 10 shows the half-height angle of the beam $\alpha_{1/2}$ and the half-height width of the current density distribution in the crossover $d_{1/2}$ in function of the emission current I_e . The values for $d_{1/2}$ were obtained from the scanning diagrams registered in the crossover image plane and recalculated for the crossover plane according to the prevailing imaging scale M (which is dependent on z_{cy}).

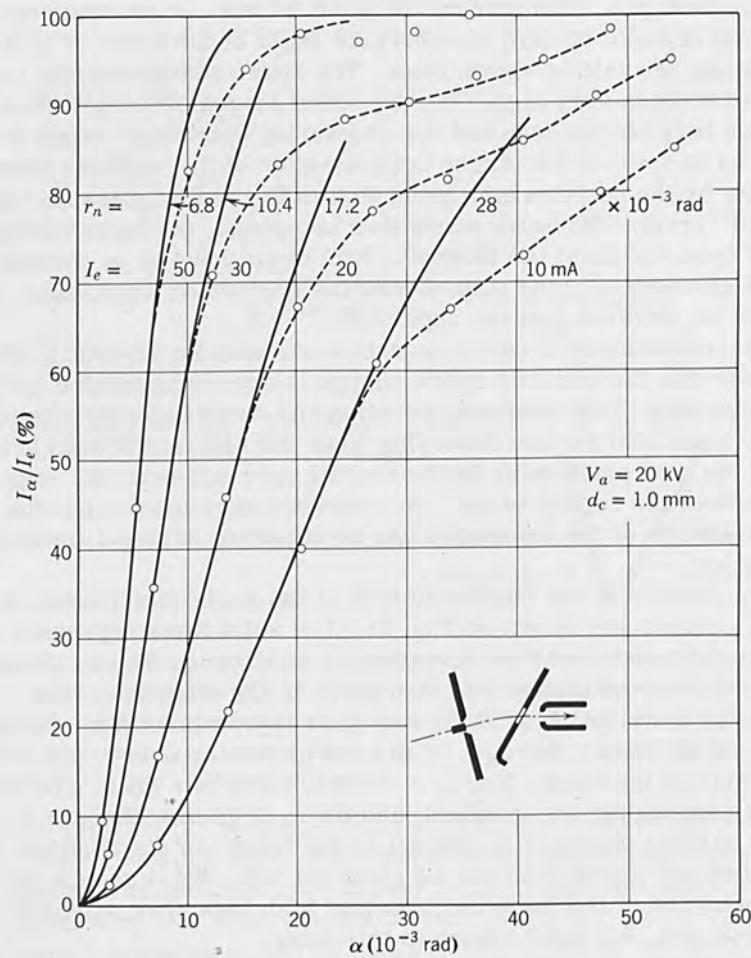


Fig. 9a. Angular distribution of the beam current for various emission currents I_e . Circles give measuring points, the full lines represent the adjusted Gaussian curves with indicated r_n .

The coordinates of z_{cy} have been included in Fig. 10. The parallelism of the three curves is remarkable; the $d_{1/2}$ -graph has a maximum where the $\alpha_{1/2}$ -graph has a minimum. In theory, the product $d_{1/2} \cdot \alpha_{1/2}$ should, in the first approximation, be independent from I_e , but this is obviously not the case. The theoretical maximum

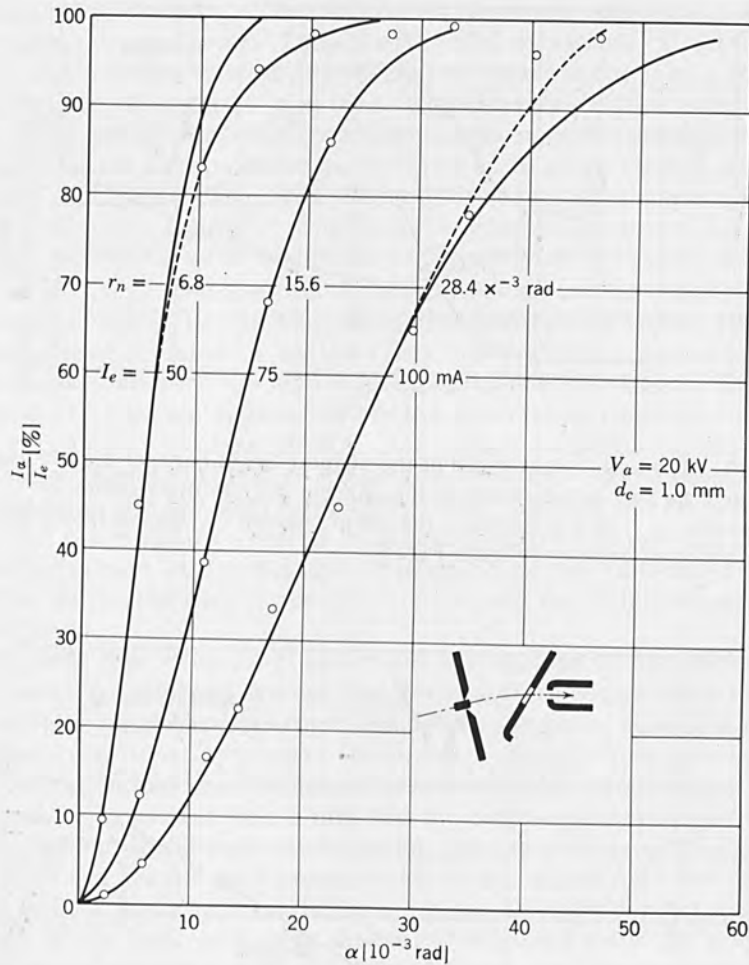


Fig. 9b. Angular distribution of the beam current for various emission currents I_e . Circles give measuring points, the full lines represent the adjusted Gaussian curves with indicated r_n .

"Richtstrahlwert" $(K_{\max})_{th}$ was calculated according to equation 6 and drawn as a solid curve on Fig. 11 in function of the emission current I_e . For this calculation the cathode current density was obtained from the cathode diameter and the emission current, considering the distribution of the current density in the emitting sur-

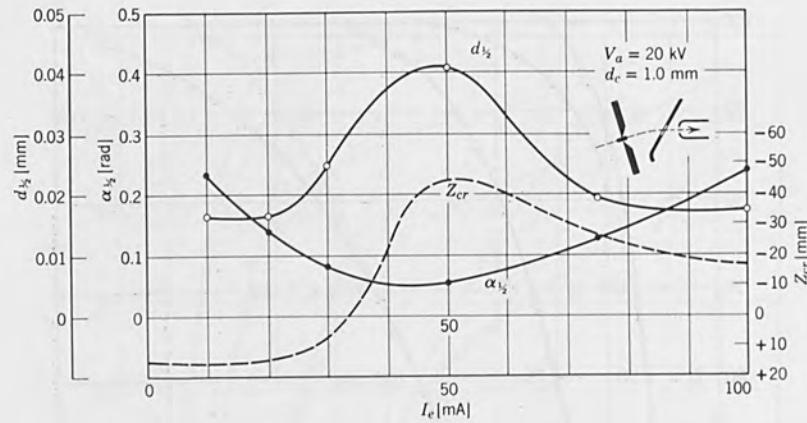


Fig. 10. The half-height width of the angular beam current distribution, $\alpha_{1/2}$, and the half-height width of the current density distribution in crossover, $d_{1/2}$, as a function of the beam current I_e . Dotted line gives the position of crossover from Fig. 8.

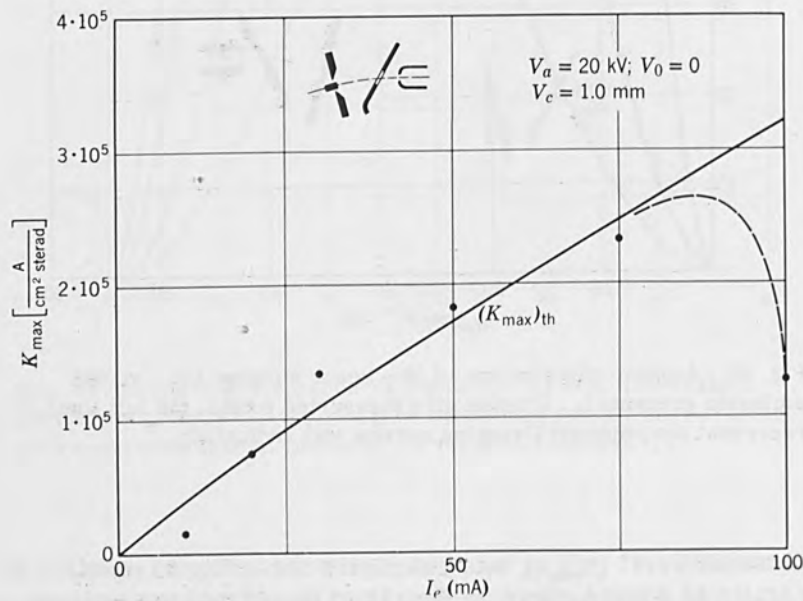


Fig. 11. Comparison of the experimental "Richtstrahlwert" $(K_{\max})_{\text{exp}}$ with the theoretical "Richtstrahlwert" $(K_{\max})_{\text{th}}$.

face as being constant. The cathode temperature was measured by an optical pyrometer. The experimental maximum "Richtstrahlwert" was calculated from the measured values for $d_{1/2}$ and $\alpha_{1/2}$ according to equation 11, and these experimental data are shown in Fig. 11 by points. There is good agreement between the experimental results and the theoretical curve up to about 90 mA, as one can see from this graph. The theoretical curve is an upper limit for the "Richtstrahlwert." That some experimental points lie over the theoretical curve is due mostly to experimental errors in obtaining $(K_{\max})_{\text{exp}}$. The use of average emission current density for calculation of $(K_{\max})_{\text{th}}$ can also falsify the theoretical value. Nevertheless the conclusion is justified that in the electron gun arrangement mentioned here the achieved experimental maximum "Richtstrahlwert" reaches approximately the theoretical limit for emission currents up to about 90 mA. For higher emission currents the maximum "Richtstrahlwert" drops very rapidly. On the other hand we find from the I-V characteristics of the gun that the transition from the temperature-limited emission to space-charge-limited emission occurs in this region. This space charge in front of the cathode must also have a great influence onto the "Richtstrahlwert" of the beam.

To study the influence of spherical aberrations of the magnetic lens, measurements of the current density distribution were made for various aperture diaphragm diameters; to make sure of a more complete rotational symmetry in the beam, the electron gun with ion separation has been replaced by a rectilinear electron gun. The cathode diameter was 1 mm and the emission current 10 mA. Measurement of the angle distribution yielded a value of $(\alpha_{1/2})_0 = 25 \times 10^{-3}$ rad for the half-height width on the object side and of $(\alpha_{1/2})_i = 20.4 \times 10^{-3}$ rad on the image side. Current density distributions in the focal spot were measured with this beam for aperture diaphragm diameters of 20, 15, 10 and 5 mm, and the scanning diagrams are shown in Fig. 12. It can already be seen that the beam diameter decreases with the decrease of diaphragm diameter. The half-height widths $(d_{1/2})_i$ and the tenth-height widths $(d_{1/10})_i$, calculated from these scanning diagrams, are shown on Fig. 13; the decrease in beam diameter is small but definite. Therefore, the spherical aberrations of the magnetic lens are influent in this beam. The angular distribution of the beam current for a 50-mA beam is very much smaller ($(\alpha_{1/2})_i \approx 7.3 \times 10^{-3}$ rad), the influence of the aperture diaphragm on the beam diameter is very weak; since in that case 100% of the beam current is inside the aperture angle of about 20×10^{-3} rad, the influence of the spherical aberrations of the

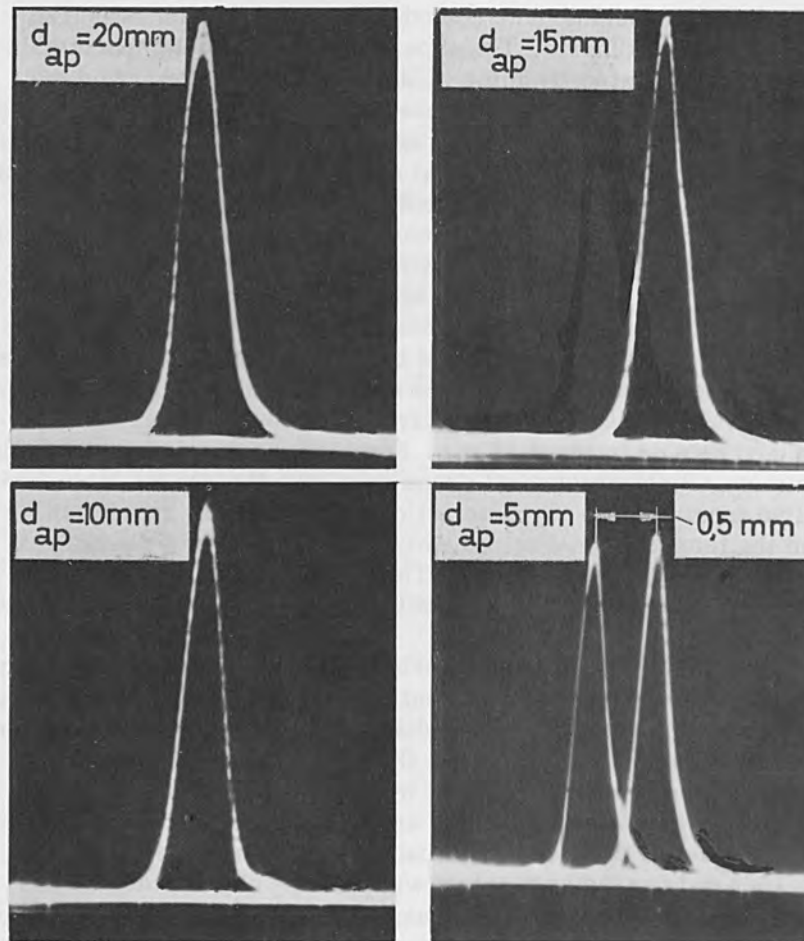


Fig. 12. Scanning diagrams for various diameters of the aperture diaphragm with constant emission current. Rectilinear triode electron gun: $V_a = 20\text{ kV}$, $V_0 = 0$, $d_c = 1\text{ mm}$, $I_e = 10\text{ mA}$, spherical aberration coefficient $C_s^* = (M + 1)^4 C_{s\infty} = 6400\text{ mm}$.

magnetic lens should be negligible ($C_s = 6400\text{ mm}$ for $M = 1$). Decreasing the diaphragm diameter eliminates not only the influence of the spherical aberration of the magnetic lens, but also the spherical aberration of the electron gun. Therefore, according to our observations, the spherical aberration of the electron gun is important in the case of a 50-mA beam.

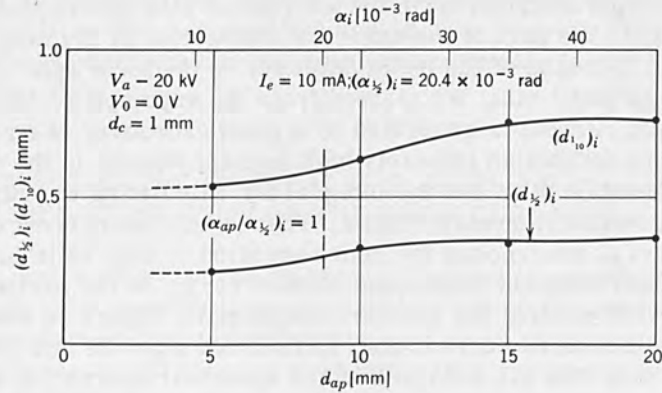


Fig. 13. Diameter $(d_{1/2})_i$ (half-height width) of the electron beam at the focus as a function of the aperture diaphragm diameter d_{ap} for a 10-mA electron beam. (Rectilinear triode electron gun.)

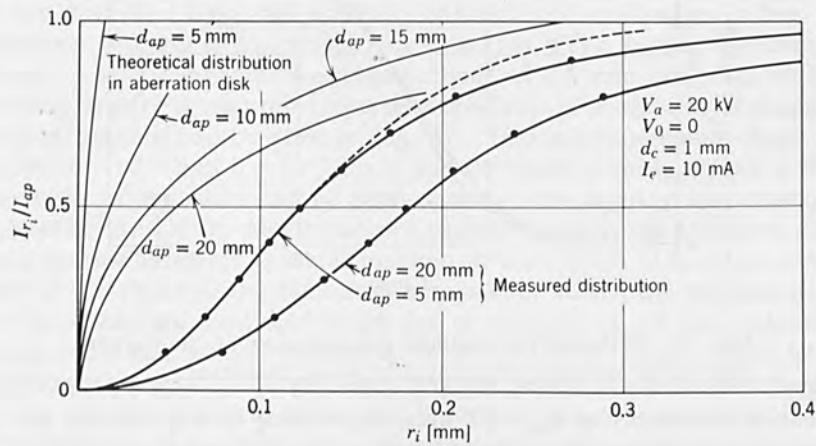


Fig. 14. Comparison of the calculated current distribution in the aberration disk due to spherical aberration with the measured current distribution in the focal spot: I_{r_i} = beam current inside the circle of radius r_i in the image plane of the crossover; I_{ap} = beam current passing the aperture diaphragm of diameter d_{ap} .

The current distribution in the aberration disk was calculated to investigate the part of the spherical aberration of the magnetic lens in the increase of the beam diameter in the focal spot in the case of a 10-mA beam. We started from the assumption that the entire beam current is generated by a point crossover of no extension. There follows an infinitely high current density in the center of the aberration disk, but the integral for calculating the current inside the radius r_i remains finite. That is why the current distribution $I_{r_i} : I_{ap}$ was chosen for representation in Fig. 16 in function of r_i for four different diaphragm diameters; I_{ap} is the partial beam current passing the aperture diaphragm. Figure 16 also shows the measured current distributions for $d_{ap} = 20$ and 5 mm. It can be seen that the influence of the spherical aberration with $d_{ap} = 20$ mm must be effective. The change from $d_{ap} = 20$ mm to $d_{ap} = 15$ mm does not diminish the current distribution in the aberration disk very much. We have noted, in Fig. 15, that the beam diameter in the focal spot remains almost constant, and only de-

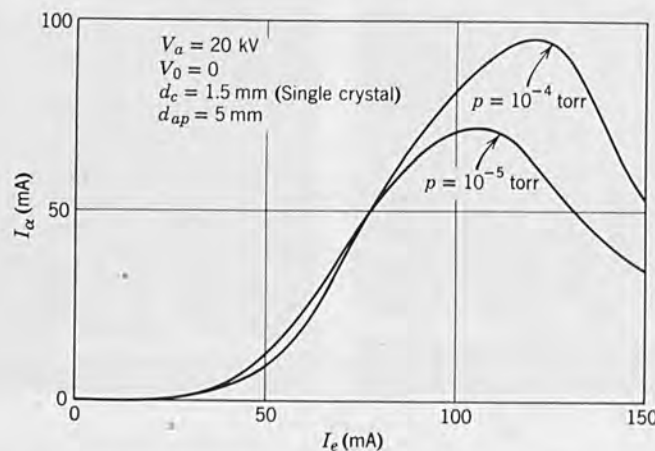


Fig. 15. Effect of the residual gas pressure on the current passing an aperture diaphragm of 5-mm diameter.

creases noticeably at $d_{ap} = 10$ mm. According to Fig. 16, the influence of the aberration disk should not be noticeable at all for $d_{ap} = 5$ mm. The Gaussian curve, adjusted for this case, has been included as dotted line; the measured distribution differs from the Gaussian curve, and we still find a relatively high part of the current in the tail of the curve. But since the spherical aberration of the magnetic lens has been eliminated, this deviation must be ex-

plained by the spherical aberration of the electron gun, which is still effective. The astigmatism should also be participating since the scanning diagram was not quite symmetrical, as can be seen from Fig. 14.

INFLUENCE OF THE RESIDUAL GAS PRESSURE ON THE ANGULAR DISTRIBUTION OF THE BEAM CURRENT

It is well known that the electron charge causes a spreading of the beam. The positive ions generated by the collision of electrons with the residual gas molecules are capable of compensating entirely the electron charge in the beam; if the gas pressure is too high, a positive space charge can develop, which leads to an additional focusing effect. (The result is self-focusing rays known as "Knoten" rays, or "Faden" rays [6]). We have investigated in our experimental system the influence of the vacuum on the electron beam. We are able to influence independently the vacuum in the electron gun and in the measuring chamber since both chambers are evacuated by separate diffusion pumps and are only connected by a narrow channel [7].

We shall mention, as an example, only one measurement: the dependence of the beam current through an aperture diaphragm of 5 mm diameter from the emitting current; the pressures in the electron gun and in the measuring chamber were kept at the same level. A single-crystal tungsten cathode of 1.5 mm diameter was used to ensure a regular electron emission. The results of these measurements can be seen in Fig. 15. Two measurements were made, for 10^{-5} and for 10^{-4} torr, and the considerable effect of the increase in pressure can be seen readily: the angular distribution becomes narrower. More current passes through the aperture diaphragm. The effect is most marked in the region of $I_e \approx 120$ mA; this is the value of the emission current for which the angular distribution has its minimum width (as in the case of a 1 mm cathode for $I_e \approx 50$ mA, as has already been shown). Therefore, there is a considerable space charge density in the beam. This space charge of the electrons seems to be still insufficiently compensated by positive ions at $p = 10^{-5}$ torr. The repulsive forces of the electrons are effective and spread the beam. If the pressure is increased to 10^{-4} torr, ten times more positive ions are generated and they largely compensate the electron charge. The repulsive forces become weaker and the beam narrows. In the case of weaker emitting currents, the spreading of the beam is so small that an in-

crease in pressure from 10^{-5} torr to 10^{-4} torr has no effect. The graphs even cross, i.e., the increase in pressure causes a small spreading of the beam. This could be explained by the scattering of the electrons by residual gas molecules. Increase in pressure widens somewhat the current density distribution in the image of the crossover. This may derive from the decrease in aperture angle described above if we assume that $r_n \cdot \alpha_n$ is a constant. The increase in focal spot diameter has a maximum at $I_e \approx 120$ mA.

We have described mainly the experimental method that we used and have shown some examples of the possibilities of application of this method. Investigation results are described more fully in another paper.

These investigations were supported in part by the firm Balzers A.G. für Hochvakuumtechnik und dünne Schichten, to which we wish to express our thanks. We also wish to thank Messrs. E. Naef, A. Jordi, and M. Jordi for their help in the realization of the measurements, as well as Mr. A. Meier who prepared the cathodes. Our thanks also go to professor E. Baumann, director of the Department of Industrial Research, who made this work possible and gave permission for its publication. And we should like to thank Mr. J. B. Ansermoz for his assistance in the translation into English.

REFERENCES

1. D. B. Langmuir, Proc. IRE **25**, 977 (1937).
2. J. R. Pierce, J. Appl. Phys. **10**, 715 (1939).
3. E. B. Bas, Optik **12**, 377 (1955).
4. H. J. Harker, J. Appl. Phys. **28**, 1354 (1957).
5. E. B. Bas, G. Cremosnik, and H. Lerch, 1961 Transactions of the Eighth Vacuum Symposium and Second International Congress, Vol. 2, p. 817, Pergamon Press. See also Schweiz. Archiv. f. Angew. Wissensch. Technik, **27**, 55 (1961).
6. E. B. Bas, B. Cremosnik, H. Lerch, O. Winkler, and O. Gmeiner, "A 30-kV Electron-Beam Welding Machine with Indirectly Heated Tungsten Cathode," p. 734 of this book. See also Ref. 3 and E. B. Bas, G. Cremosnik, H. Lerch. 1961 Transactions of the Eighth Vacuum Symposium and Second International Congress, Vol. 2, p. 817, Pergamon Press.
7. E. Brücke and A. Recknagel, Electronengeräte, p. 241, Springer Verlag, Berlin, 1941; see also I. B. Jonson, Phys. Rev. **17**, 420 (1920).

APPENDIX

Distribution of the beam current in the aberration disk due to the spherical aberration assuming a Gaussian angular distribution of the beam current.

To get the current density distribution in the image plane of the crossover, we have to calculate the isophotes in this plane considering the following items: current density distribution at the crossover (e.g., Gaussian distribution), angular current density distribution of each surface element of the crossover, and the spherical aberration of the imaging lens. It is possible in principle to proceed in this way, but it is a complicated and time-consuming task. To get an idea of the effect of spherical aberration of the imaging lens we have chosen here simplified considerations. We assume that the whole beam current is coming from a central point of the crossover with an angular current density distribution j_{ai} . (Subscripts i in the following mean that we consider all quantities on the image side of the lens.) We ask now for the current density distribution in the aberration disk in the image plane and consider a surface element $\Delta F_i = 2\pi r_i dr_i$ at a distance r_i from the axis. The beam current comes to this element from a solid angle element $\Delta\Omega_i = 2\pi\alpha_i d\alpha_i$ and amounts to:

$$dI_{a_i} = j_{a_i}(\alpha_i) 2\pi\alpha_i d\alpha_i \quad (1)$$

For the current density in the image plane we can then write:

$$j_{r_i}(r_i) = j_{a_i}(\alpha_i) \frac{\alpha_i}{r_i} \frac{d\alpha_i}{dr_i} \quad (2)$$

r_i and α_i are coupled together with the aberration formula:

$$r_i = C_s^* \alpha_i^3 \quad (3)$$

$$C_s^* = (M+1)^4 C_s^\infty \quad (4)$$

Here C_s^∞ indicates the coefficient of spherical aberration for very high magnification or demagnification (i.e., $M \rightarrow 0$). From equation 3 we get now:

$$\alpha_i = (C_s^*)^{-1/3} r_i^{1/3} \quad (5)$$

$$\frac{d\alpha_i}{dr_i} = \frac{1}{3} (C_s^*)^{-1/3} r_i^{-2/3} \quad (6)$$

Putting equations 5 and 6 into equation 2 gives:

$$j_{r_i}(r_i) = j_{a_i}(\alpha_i) \frac{1}{3} (C_s^*)^{-2/3} r_i^{-4/3} \quad (7)$$

If we assume a Gaussian angular current density distribution:

$$j_{a_i}(\alpha_i) = \frac{I_e}{\pi \alpha_{n_i}^2} \exp \left\{ - \left(\frac{\alpha_i}{\alpha_{n_i}} \right)^2 \right\} \quad (8)$$

it follows then from equation 7:

$$j_{r_i}(r_i) = I_e \frac{1}{3\pi \alpha_{n_i}^2} (C_s^*)^{-2/3} r_i^{-4/3} \exp \left\{ - \frac{r_i^{2/3}}{(C_s^*)^{2/3} \alpha_{n_i}^2} \right\} \quad (9)$$

As we see from equation 9, the current density reaches infinity on the axis, which is plausible from our assumption of a point crossover. It is, therefore, reasonable to use the current distribution L_{r_i} instead of j_{r_i} . L_{r_i} means the current of the area πr_i^2 , for which we get, by integration of equation 9,

$$\frac{L_{r_i}}{I_e} = 1 - \exp \left\{ - \frac{r_i^{2/3}}{(C_s^*)^{2/3} \alpha_{n_i}^2} \right\} \quad (10)$$

Similarly to the Gaussian distribution we now define here also a radius r_{n_i} for which $L_{r_i}/I_e = 1 - e$. It follows then:

$$r_{n_i} = C_s^* \alpha_{n_i}^3 \quad (11)$$

It can be easily shown that this equation is also true for the half-height radius $(r_{1/2})_i$ (corresponding to $L_{r_i}/I_e = 1/2$) if we replace α_{n_i} by $(\alpha_{1/2})_i$:

$$(r_{1/2})_i = C_s^* (\alpha_{1/2})_i^3 \quad (12)$$

In the case where the electron beam is limited by an aperture diaphragm, with I_{ap} indicating the beam current passing the diaphragm and $(\alpha_{ap})_i$ indicating the beam angle defined by this aperture diaphragm, we get from equation 10 the following relation for the beam current distribution in the image plane:

$$\frac{L_{r_i}(r_i)}{I_{ap}} = \frac{1 - \exp \left\{ - \frac{r_i^{2/3}}{(C_s^*)^{2/3} \alpha_{n_i}^2} \right\}}{1 - \exp \left\{ - \frac{(\alpha_{ap})_i^2}{(\alpha_n)_i^2} \right\}} \quad (13)$$

This equation is used to calculate a set of curves with d_{ap} as parameter and with $(\alpha_{1/2})_i = 20.4 \times 10^{-3}$ rad, $C_s^* = 6400$ mm. These curves are reproduced in Fig. 14.

ENERGETIC ELECTRON BEAMS AND COLD CATHODE BOUNDARY CONDITIONS

INTRODUCTION

When the cathode of a glow discharge is a hollow cylinder and the pressure is properly adjusted, the negative glow will completely fill the hollow region of the cathode. This discharge is called a hollow cathode discharge (HCD) or Schüller discharge. The electron density in the negative glow is orders of magnitude higher than that obtainable for a normal glow discharge for the same current density. The classical HCD has been used as a source of high-intensity radiation by spectroscopists for over a half a century. Recently a perforated wall, or screen-mesh, hollow cathode has been found to produce a well-collimated electron beam (van Paassen et al., 1962). The beam-producing discharge or electron-beam mode discharge (EBMD) is a cold cathode phenomena which operates at pressures usually associated with the abnormal glow discharge. The beam produced by the mode is energetic (0.5 to 100 kV) and extremely well collimated.

The recent interest in electron-beam production by gas discharges was stimulated by the reduced vacuum requirement afforded by the use of these electron sources when used in welding and melting systems.

Screen-mesh cathodes used to produce electron beams operating in the EBMD are simple in construction and not affected by gas poisoning. These cathodes can typically operate at pressures as high as 1 torr for indefinite periods of time without destruction of their emission properties. Spherical cathodes with concave shaping in the vicinity of the dominant aperture or emitter were

*Presently with the Electrical Engineering Dept., George Washington University, Washington, D. C.

†The Martin Company, Baltimore, Md.

constructed giving increased total power delivered to the beam in a particular range of pressure operation. Experimental voltage equations for the EBMD for various gas-cathode material combinations all have the same form, $I = C_1 (V/C_2)^{C_3} P^{-C_4}$, where the values of the constants C_1 , C_2 , C_3 , and C_4 depend upon the particular filling gas, cathode material, and the screen-mesh, optical transmission factor.

The beam is produced when electrons created interior to the cathode cavity are emitted from a dominant aperture. Experimentally, it has been found that an aperture must be at least three times the diameter of the screen-mesh hole size used in the cathode construction. After leaving the cathode electrons interact with gas molecules exterior to the cathode surface and produce positive ions. These positive ions re-enter the cathode after acceleration in the exterior field region to produce additional electrons and ions in the cathode cavity, and the process is repeated.

A mesh, hollow cathode was designed to allow microwave diagnosis of the cavity region. A cylindrical cathode is designed to function both as a EBMD cathode and a microwave resonant cavity. A microwave probing signal is used to determine the shift in resonance frequency of the cavity due to the presence of the ionization. The resulting shift is a measure of the average electron density. Measurements are obtained as a function of applied voltage, discharge current, and filling gas pressure.

GENERAL DESCRIPTION

The electron-beam mode discharge is a high-impedance, cold cathode discharge characterized by the production of one or more well-defined electron beams. A cylindrical, perforated wall, hollow cathode structure formed from fine mesh wire screening is used to produce the electron-beam mode discharge. This cathode is installed in a vacuum chamber whose pressure can be maintained at a fixed value between 1 and 1000 microns Hg. An anode is also placed in this vacuum chamber at some distance from the cathode. The placement of the anode must be outside the region normally occupied by the cathode dark space.

With this experimental apparatus we can easily obtain the normal hollow cathode glow discharge over the pressure and voltage ranges normally associated with solid-walled, hollow cathode structure (Fig. 1). In addition to this it is observed that in the low-

er portion of the voltage-pressure domain of the classical hollow cathode discharge there is a second, stable, high-impedance discharge mode which is markedly different from the hollow cathode

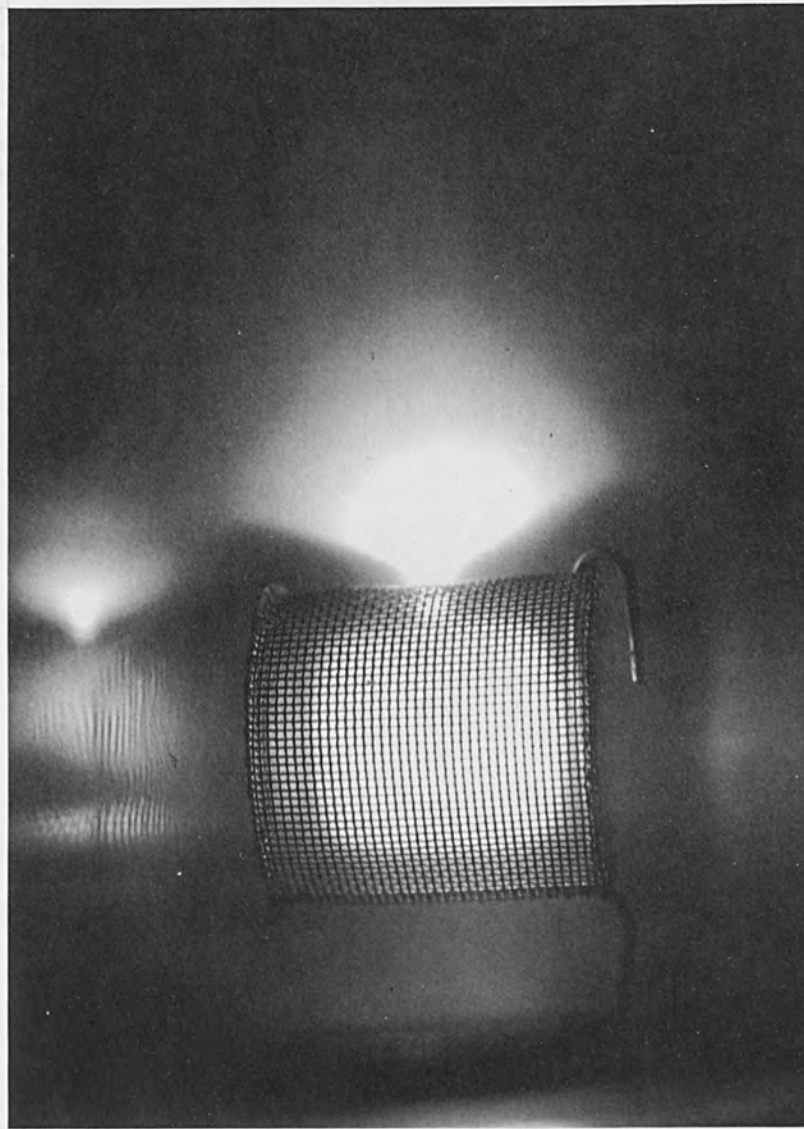


Fig. 1. Cylindrical screen-mesh cathode operating in the hollow cathode discharge mode.

mode (Fig. 2). Over the entire range of pressure and voltage in which the high-impedance electron-beam mode operates, the

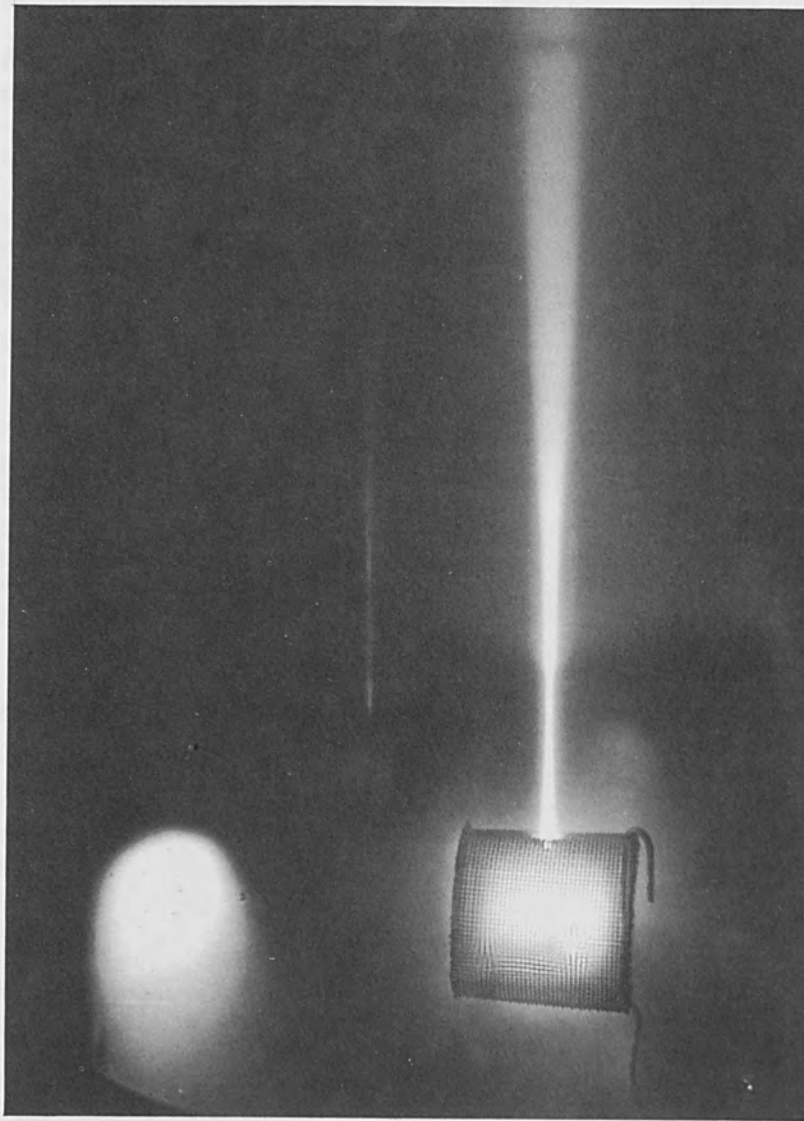


Fig. 2. Cylindrical screen-mesh cathode operating in the electron-beam mode discharge.

classical hollow cathode mode will also operate. Both of these modes are stable over this pressure and voltage range, the mode being determined by the initial conditions. Excitation of the gas in the test chamber, by an rf source, will cause a transition from the high-impedance mode to the hollow cathode mode. A momentary interruption of cathode potential will switch the discharge back to the high-impedance mode.

The subsequent cathodes used to produce the electron beam mode discharge are simple spheres 5 cm in diameter with 1-cm diameter apertures (Fig. 3).

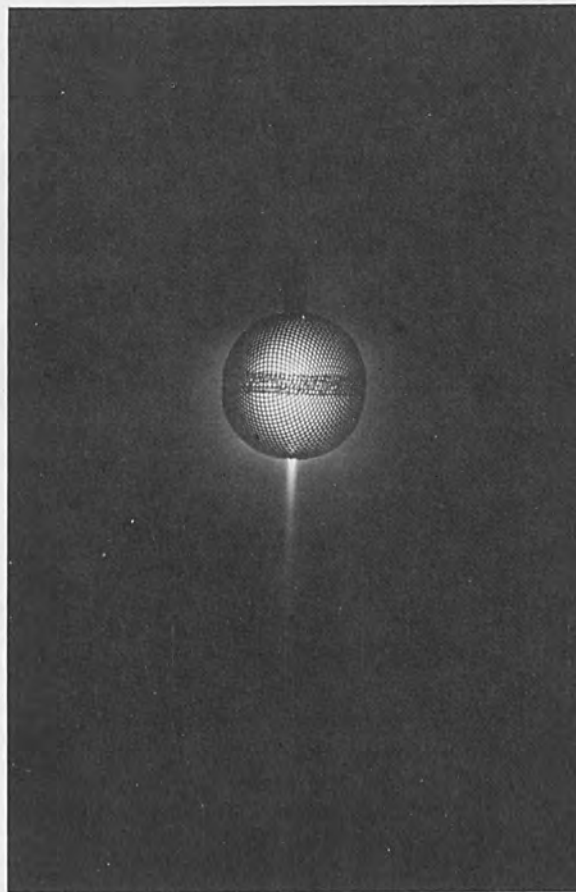


Fig. 3. Spherical screen-mesh cathode operating in the electron-beam mode discharge.

Dispersing the return flow of positive ions over a larger portion of the cathode surface, and thereby reducing the positive ion current density, permits the electron-beam mode to operate at very high power levels. A diverging electrostatic lens is used to reduce the positive ion current density in the vicinity of the cathode aperture. Two such electrostatic lenses were devised which work well, both independently and in combination with each other.

Figure 4 is a photograph of four cathode configuration studies in which a concave area surrounding the dominant aperture and a ring focusing element were used. The resulting cathode characteristics are summarized in Table I.

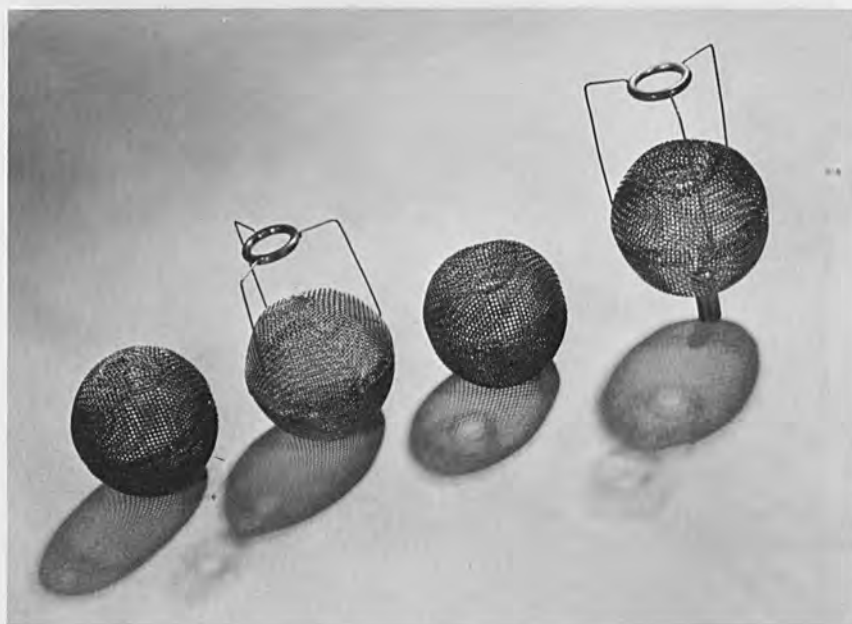


Fig. 4. Spherical cathode configuration (1 to r) simple sphere, sphere with ring, dimpled sphere, dimpled sphere with ring.

THE SPHERICAL CATHODE SURFACE EMITTER

The spherical cathode surface electron emitter has been studied in great detail because of the well-defined boundary conditions for returning positive ions. In this section we shall consider the

Table I

The Characteristics of Four Cathode Geometries Studied

Cathode	Pressure Range	Power	Beam Characteristics	Comments
Simple sphere	5-300 microns Hg	75 W	Variable focus	Strongly pressure and voltage sensitive
Sphere with ring	20-50 microns Hg	2 kW	Focused by ring potential changes	Less sensitive to pressure changes
Dimpled	2-50 microns Hg	2 kW	Focused by pressure changes	Strongly pressure and voltage sensitive
Dimpled sphere with ring	2-20 microns Hg	5 kW	Variable focus, usually short	Comparatively insensitive to small voltage and pressure variations

simple spherical cathode without a dominant aperture. The resulting analysis of the cathode surface yields the effect of variation of the screen-mesh transparency of the cathode surface upon the discharge characteristics.

In the physical model of the spherical solid-surface cathode, electrons are emitted from the cathode as a result of ion bombardment. γ_i electrons are emitted for every ion which strikes the cathode surface. The electrons are accelerated by the cathode-anode potential drop and obtain sufficient energy to produce ionization by collision with gas atoms. Near the cathode surface, little of the current is carried by electrons, the major portion is carried by positive ions. Using this model, the current density near the solid cathode surface is given by:

$$J = J_c^+ (1 + \gamma_i) \quad (\text{for solid surface cathode}) \quad (1)$$

where J_c^+ = the ion current density at the cathode.

The surface of a spherical screen-mesh cathode can be considered as two distinct areas: (1) the solid area created by the wire mesh and, (2) the holes or mesh openings. Figure 5 is a schematic

comparison of the mechanisms which take place for a solid and a screen-mesh cathode. If it is assumed that the mesh openings produce small perturbations on the electric field near the cathode, it follows that J_c^+ for the screen cathode will be a constant over the cathode surface.

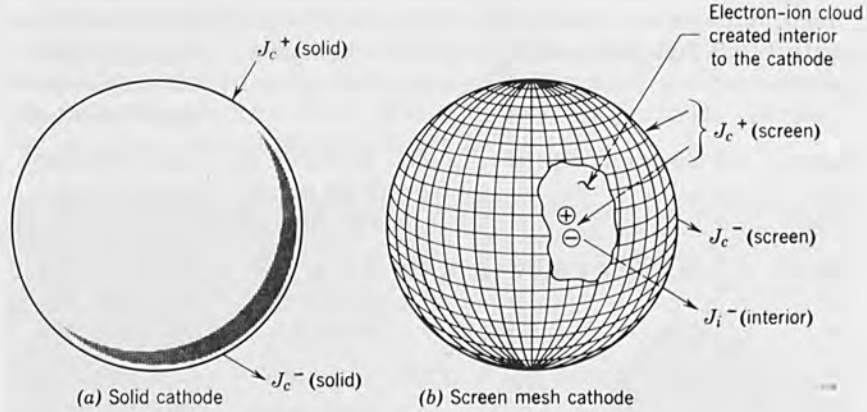


Fig. 5. Schematic comparison of mechanism for solid- and screen-mesh spherical cathodes.

We can then write

$$J_c^+ (\text{screen}) = J_c^+ (\text{solid}) + \Delta J_c^+ \quad (2)$$

where ΔJ_c^+ = the change in ion current due to the presence of the perforations in the mesh sphere.

The current density over the solid surface of the screen-mesh sphere can be written using equation 1.

$$J_s = \{1 + \gamma_i\} \{J_c^+ + \Delta J_c^+\} \quad (3)$$

If A = area of a solid surface cathode, the same diameter screen-mesh cathode can be divided into two regions, A_1 = area of the hole surface and $(A - A_1)$ = area of the solid surface. Using equation 3, the current due to the solid portion of the surface can be written

$$I_{\text{mesh}} = \{A - A_1\} J_s \quad (4)$$

The current due to the electrons which are produced "internally" to the screen mesh cathode and are emitted through the holes in the screen can be written

$$I_{\text{holes}} = A_1 J_i = A_1 \{1 + C_{vp}\} \{J_c^+ (\text{solid}) + \Delta J_c^+\} \quad (5)$$

where

J_i = current density in each opening of the screen due to the bombardment of the interior of the cathode by ion returning to the cathode.

C_{vp} = a factor which relates the effect of V (voltage drop the ions have obtained before producing ionization in the interior region) and P (pressure at which the discharge is operating).

The total current to the screen-mesh cathode can be written

$$I (\text{total}) = I (\text{mesh}) + I (\text{holes}) \quad (6)$$

Substituting into equation 6 from equations 3, 4, and 5, and dividing by

$$I (\text{total solid sphere}) = A \{1 + \gamma_i\} J_c^+ (\text{solid}) \quad (7)$$

we obtain

$$\frac{I (\text{total mesh ball})}{I (\text{total solid ball})} = \left\{ 1 - \frac{A_1}{A} + \frac{A_1}{A} \left(\frac{1 + C_{vp}}{1 + \gamma_i} \right) \right\} \left\{ 1 + \frac{\Delta J_c^+}{J_c^+} \right\} \quad (8)$$

where A_1/A = the optical transparency factor of the screen mesh.

Equation 8 gives the ratio of mesh current to solid ball current as a function of the optical screen-mesh transparency and the factor C_{vp} .

Figures 6 through 10 are experimentally determined, volt-ampere curves for 5-cm diameter stainless steel, type 304 cathode balls operating in argon gas.

The ratio of $I (\text{mesh})/I (\text{solid})$ can be formed from this experimental data at fixed pressure and voltage.

In equation 8, the factors ΔJ_c^+ and C_{vp} are unknown. As we change the cathode from a solid sphere to a perforated sphere, the hole areas become sources of electron emission (C_{vp}) and the electrons from these new sources of electron emission create increased numbers of positive ions (ΔJ_c^+). They can be expressed as follows:

$$\frac{1 + C_{vp}}{1 + \gamma_i} = \beta \quad (9)$$

$$\frac{\Delta J_c^+}{J_c^+} = \Delta \quad (10)$$

where the factor β gives the ratio of hole emission to solid-surface emission and Δ the fractional change in positive ion current returning to the cathode.

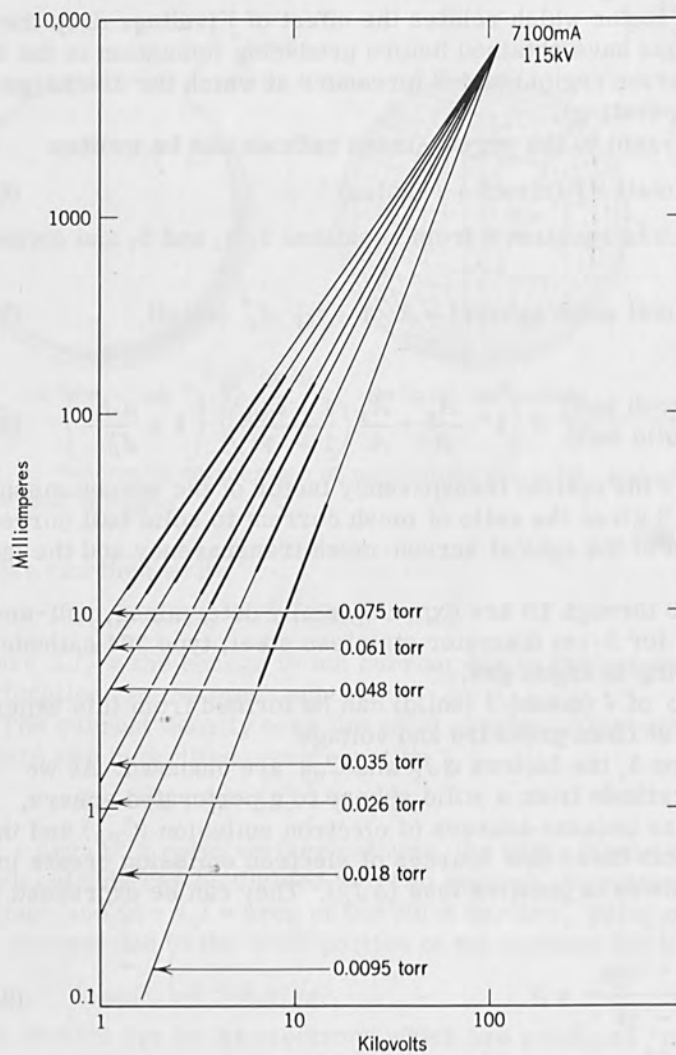


Fig. 6. Volt-ampere curves of a 5-cm diameter, 304 stainless steel solid-sphere cathode in argon.

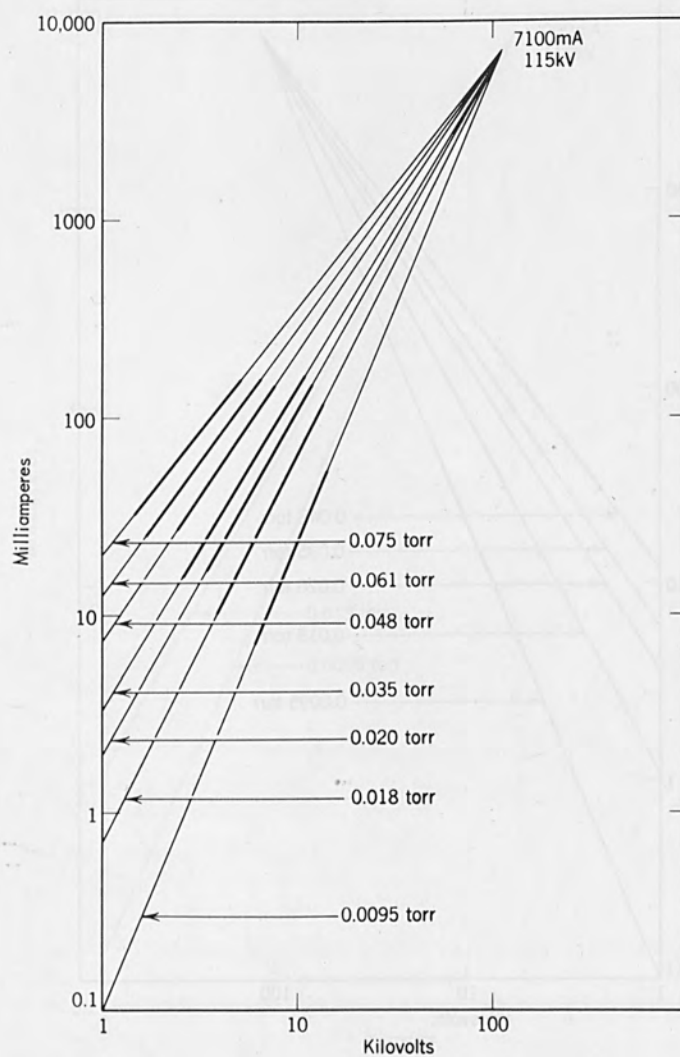


Fig. 7. Volt-ampere curves of a 5-cm diameter, 304 stainless steel mesh sphere cathode (30.2% transparency) in argon.

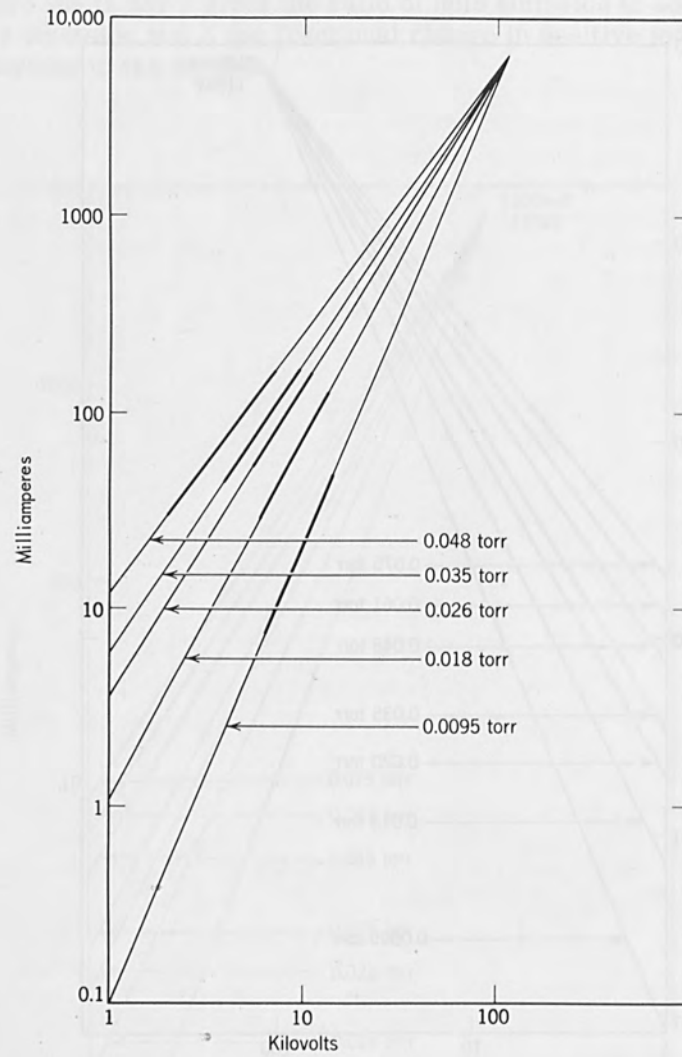


Fig. 8. Volt-ampere curves of a 5-cm diameter, 304 stainless steel mesh sphere cathode (37.9% transparency) in argon.

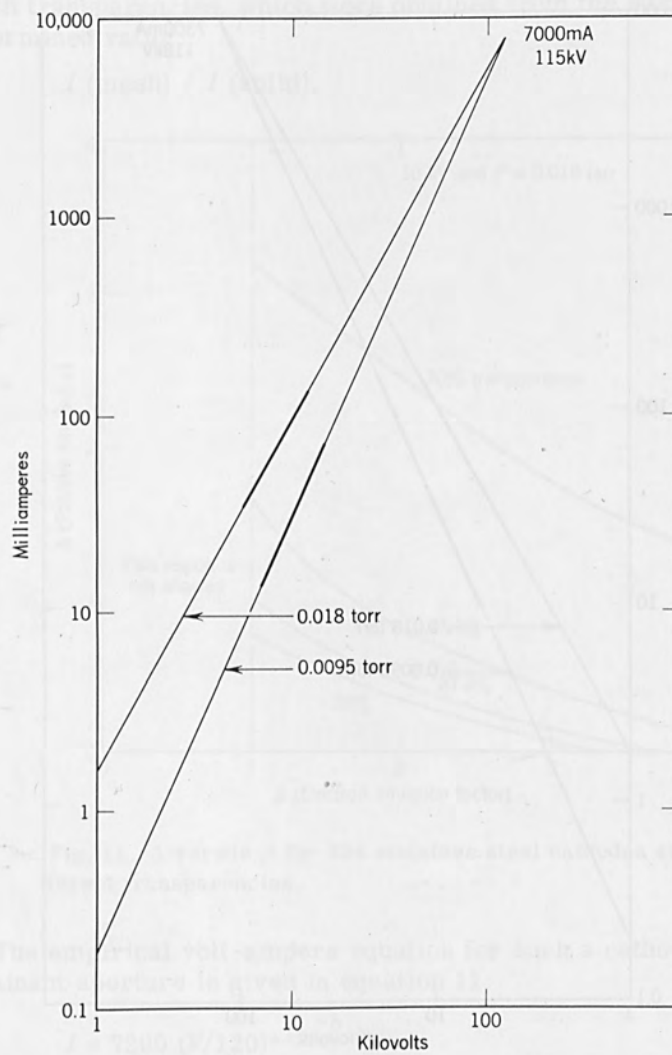


Fig. 9. Volt-ampere curves of a 5-cm diameter, 304 stainless steel mesh sphere cathode (47.4% transparency).

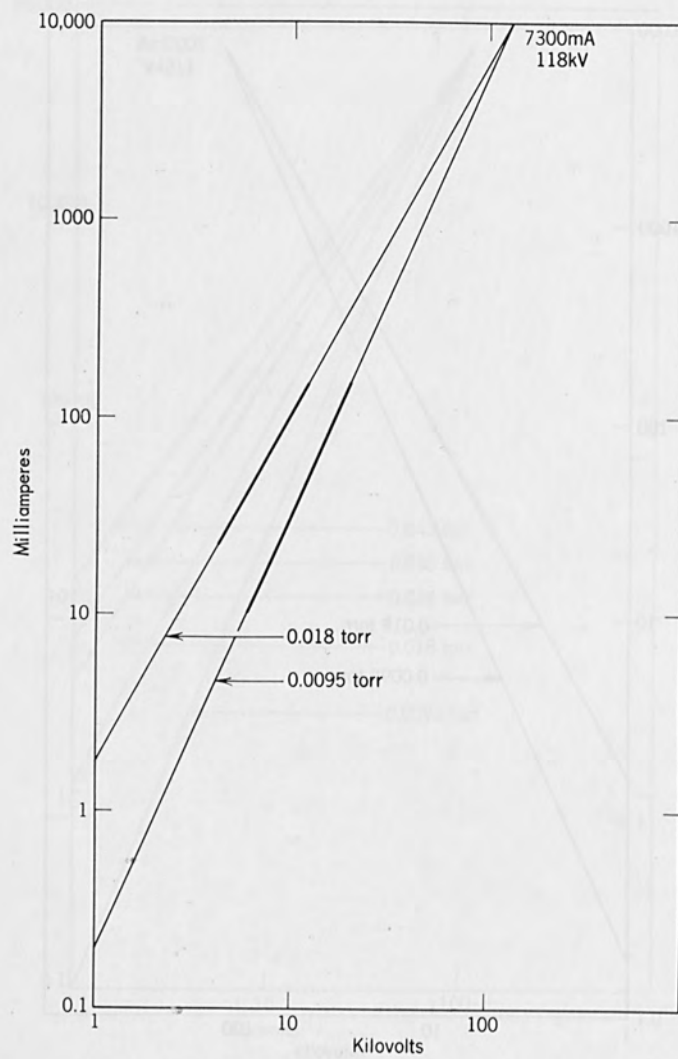


Fig. 10. Volt-ampere curves of a 5-cm diameter, 304 stainless steel mesh sphere cathode (50.8% transparency).

Figures 11 and 12 are plots of Δ versus β for differing screen-mesh transparencies, which were obtained from the empirically determined ratio:

$$I(\text{mesh}) / I(\text{solid}).$$

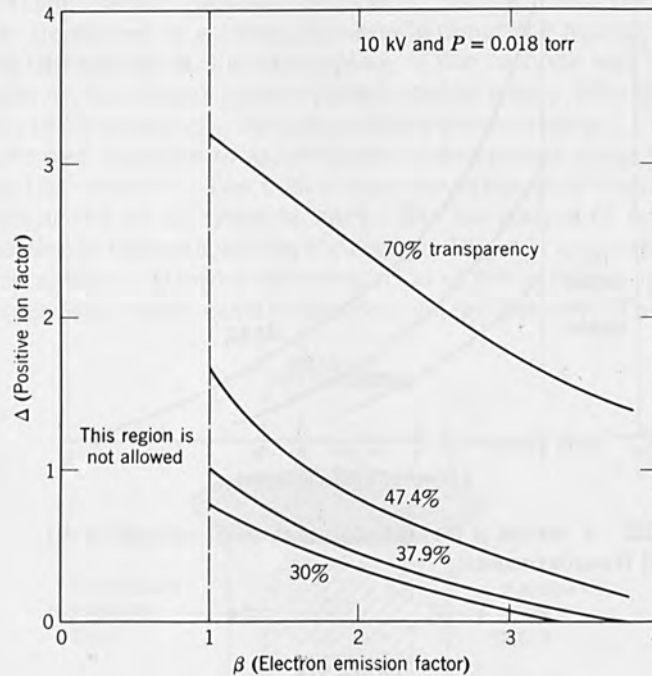


Fig. 11. Δ versus β for 304 stainless steel cathodes at different transparencies.

The empirical volt-ampere equation for such a cathode with no dominant aperture is given in equation 11

$$I = 7200 (V/120)^{4.8} P^{-b} \quad (11)$$

where V is in kilovolts, I is in milliamperes, P is in microns Hg, and b is a factor depending on screen-mesh transparency. Table II gives the factor b as a function of A_1/A for 304 stainless cathode operation in argon.

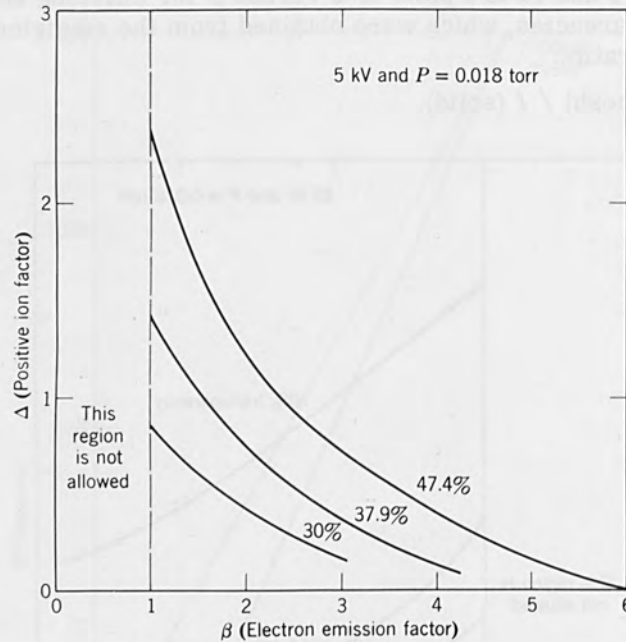


Fig. 12. Δ versus β for 304 stainless steel cathode at different transparencies.

Table II*

A_1/A	b
0.000	0.282
0.302	0.312
0.379	0.330
0.474	0.344
0.508	0.345

*van Paassen, et al., 1964.

THE PLANAR CATHODE SURFACE EMITTER

Experiments using parallel plate geometries with low-mesh transparencies ($0 < A_1/A < .30$) have been carried out in order to

more closely approximate the electric field distribution. A decrease in hole size is necessary so that its dimensions will not be comparable to the dark space dimensions. In addition, the smaller the screen-mesh holes, the less chance of exciting the HCD. The screen mesh was constructed to allow for continuous variation in transparency under vacuum conditions. This allowed the screen mesh to be varied at a fixed pressure without the necessity of opening the system to the atmosphere. The cathode was formed from two 4-mil copper sheets photo-etched with 0.025-in. square holes in their surfaces. The two sheets were arranged one above the other and mounted to allow relative movement along the diagonal of the squares. The result was a continuously variable screen mesh from 28% to 2% transparency. The utilization of microwave diagnostics in the cathode cavity region, Fig. 13, and volt-ampere equation analysis allowed determination of the electron hole current dependency upon cavity electron charge density. The micro-

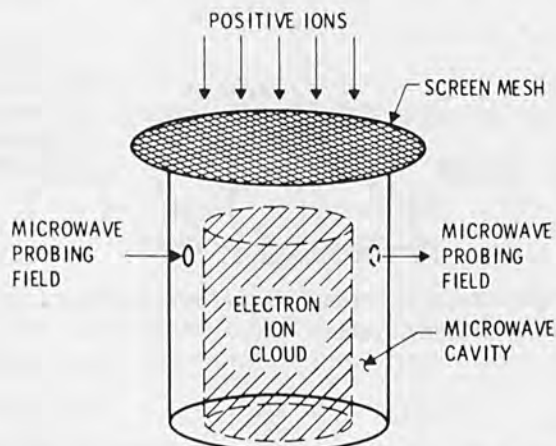


Fig. 13. Diagram of the combination discharge and microwave test cavity.

wave cavity was a right circular cylinder and excited by coupling holes to resonate in the TE_{011} mode. This mode is ideal for measurement of high-density plasmas in narrow columns. If a uniform electron density is assumed to fill the cathode cavity region because of ion bombardment of the exterior cathode surface, the shift in resonance frequency owing to free electrons (ρ) in the cathode cavity is given by:

$$\rho = 213 \Delta f \text{ electrons/cm}^3 \quad (12)$$

where Δf = the shift in resonance frequencies of the microwave cavity due to the electron-ion cloud (mcs)

Microwave energy is supplied to the test cavity by a microwave signal generator. Figure 14 is a block diagram showing the experimental arrangement of the resonant test cavity and microwave measuring system. The shift in frequency is measured by comparing the resonant frequency of the test cavity with that of a reference wave meter. Standard microwave vacuum windows are used to isolate the test cavity from the atmosphere.

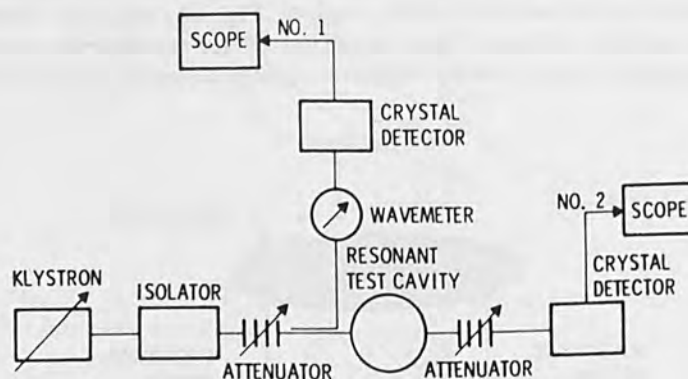


Fig. 14. Experimental arrangement of resonant test cavity and microwave measuring system.

The data was obtained at a particular pressure of interest using a constant voltage power supply and continuously varying the screen-mesh transparency while monitoring the tube current. The resulting data was taken for *He*, *Ne*, and *Ar*. These gases were chosen due to the wide range of ionization cross-sections which they offer. The data reveals a linear variation of $I(\text{mesh})/I(\text{solid})$ over a wide range of transparency. We can therefore write

$$\frac{I(\text{mesh})}{I(\text{solid})} = 1 + \frac{b}{a} \frac{A_1}{A} \quad (13)$$

The ratio $I(\text{mesh})/I(\text{solid})$ for the low-transparency mesh screen, given in equation 8, can be written

$$\frac{I(\text{mesh})}{I(\text{solid})} = 1 - \frac{A_1}{A} + \frac{A_1}{A} \left(\frac{1+C_{vp}}{1+\gamma_i} + \frac{\Delta J_c^+}{J_c^+} \left[1 - \frac{A_1}{A} + \frac{A_1}{A} \left(\frac{1+C_{vp}}{1+\gamma_i} \right) \right] \right) \quad (14)$$

$$\frac{I(\text{mesh})}{I(\text{solid})} = \left(1 + \frac{\Delta J_c^+}{J_c^+} \right) + \left[\frac{A_1}{A} - \left(1 + \frac{J_v^+}{J_c^+} \right) \right] + \left(\frac{1+C_{vp}}{1+\gamma_i} \right) \left\{ 1 + \frac{\Delta J_c^+}{J_c^+} \right\} \quad (15)$$

Comparing equations 13 and 15 we see that at low transparencies we can approximate:

$$\frac{\Delta J_c^+}{J_c^+} \ll 1 \quad (16)$$

The increased current at low values of transparency is therefore caused by increased hole electron emission and not increased positive ion current. Substituting equation 16 into equation 15 and equating to equation 13 we obtain

$$\begin{aligned} \frac{1+C_{vp}}{1+\gamma_i} - 1 &= \frac{b}{a} \\ \frac{1+C_{vp}}{1+\gamma_i} &= \frac{b}{a} + 1 \end{aligned} \quad (17)$$

The application of equation 17 to the experimental data obtained for *Ne*, *He* and *Ar* is shown in Table III.

Table III

Voltage	0.110 torr	0.131 torr	0.230 torr	0.028 torr	0.034 torr	0.039 torr	0.042 torr	0.013 torr	0.022 torr
4.2 kV	2.54	2.21		2.95	2.80	3.15		3.07	3.88
4.0 kV								3.09	3.89
3.8 kV	2.79	2.21		2.75	2.74	3.12	3.51	3.15	3.68
3.6 kV								3.38	3.78
3.4 kV	3.23	2.17	2.37	3.51	2.87	3.06	3.81	3.46	3.69
3.2 kV								3.84	3.95
3.0 kV	4.17	2.50	2.64		2.65	3.11	3.85	4.20	4.19

Solving equation 17 for the ratio C_{vp}/γ_i , we obtain

$$\frac{C_{vp}}{\gamma_i} = \frac{(b/a)}{\gamma_i} + \left(\frac{b}{a} + 1 \right) \quad (18)$$

Substituting for a typical value of (b/a) from Table III of 2, equation 17 becomes

$$\frac{C_{vp}}{\gamma_i} = \frac{2}{\gamma_i} + 3 \quad (19)$$

If values of γ_i range from 0.01 to 1 for *He*, *Ne*, and *Ar* for positive ion bombardment of the C_u cathode surface, the calculated values of C_{vp} range from five to twenty times those of secondary emission by bombardment of the exterior solid cathode surface.

Using 3 as a typical value of β from the Tables, and evaluating the Δ versus β curve for all screen-mesh cathodes given in Figs. 11 and 12, results in Table IV for low screen-mesh transparencies.

Table IV

Data for 5-cm 304 Stainless Steel Screen-Mesh Spheres

Percentage of Transmission	Tube Voltage	Pressure	β	Δ
30	10 kV	0.018 torr	3	0.1
30	5 kV	0.018 torr	3	0.2

Estimates for the increase in ion current from the screen-mesh cathode from Table IV are from 10 to 20% for a 30% transparent screen mesh.

Figures 15 through 18 are graphs of the total screen-mesh current and resonance frequency versus transparency for the planar screen-mesh cathode surface and microwave cavity combination. The linearity of the current curves is consistent with the previously considered data and theory of low-transparency electron emission. The component parts of the screen-mesh current, the hole current and the solid-surface current, are shown on these graphs.

The plot of solid-mesh current was obtained by taking the solid-surface current, i.e., $A_1/A = 0$ and applying

$$I_{(\text{mesh})} = I_{(\text{solid})} \left(1 - \frac{A_1}{A} \right) \quad (20)$$

The total hole current is calculated by subtracting the current given by equation 20 from the experimental expression for the total mesh current at a given transparency. The resulting equation for the hole current can be written as follows:

$$I_{(\text{hole})}(\text{total}) = C_1 T \quad (21)$$

The values of the constant C_1 for argon gas at 0.021 torr are given in Table V.

Table V

Tube Voltage	C_1 (Ma/% Transparency/100)
6.0 kV	3.2
4.5 kV	1.55
4.0 kV	1.12

From the observed variation of shift in resonance frequency of the microwave cavity and the total hole current variation with screen-mesh transparency, the relation between the electron density interior to the cathode and the electron hole current can be derived. The expression for the shift in resonance versus transparency can be obtained from Fig. 18.

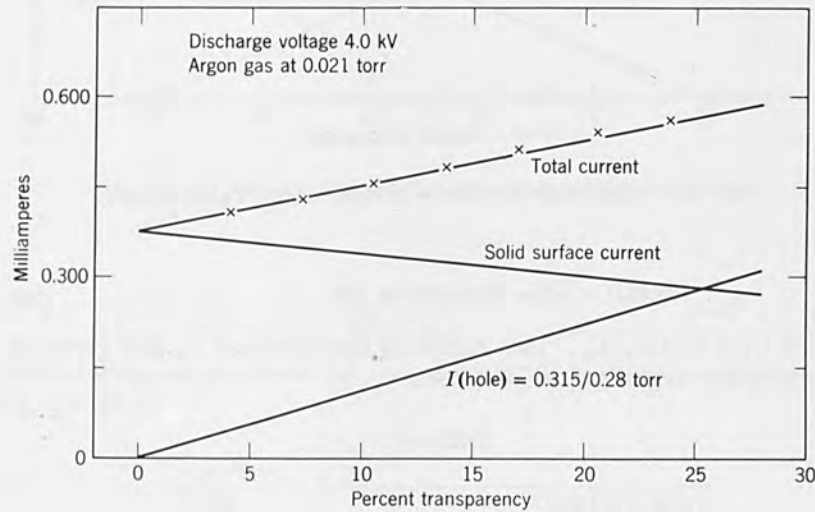


Fig. 15. Tube current versus screen-mesh transparency.

$$\Delta f = C_2 T \quad (22)$$

where C_2 = constant depending upon the tube voltage.

Substituting equation 22 into equation 12 and combining with equation 21, we obtain

$$\rho = C_3 I_{(\text{hole})}(\text{total}) \text{ electrons/cm}^3, \text{ for}$$

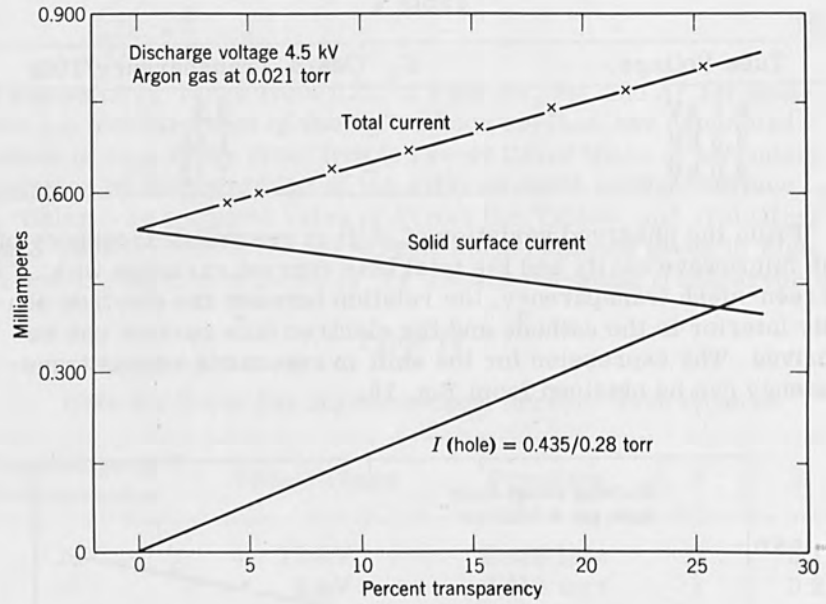


Fig. 16. Tube current versus screen-mesh transparency.

$$I_{(\text{hole})}(\text{total}) = \text{hole current in Ma} \quad (23)$$

where $C_3 = 213 C_2 / C_1$. The values of the constant C_3 are given in Table VI for argon gas at .021 torr.

Table VI

Tube Voltage	C_3
6.0 kV _s	8.62×10^9
4.5 kV	7.35×10^9
4.0 kV	5.42×10^9

The total hole current can be written

$$I_{(\text{hole})}(\text{total}) = I_{(\text{hole})}(\text{ion}) + I_{(\text{hole})}(\text{electron}) \quad (24)$$

where

$$I_{(\text{hole})}(\text{ion}) = A_1 J_C^+ \quad (25)$$

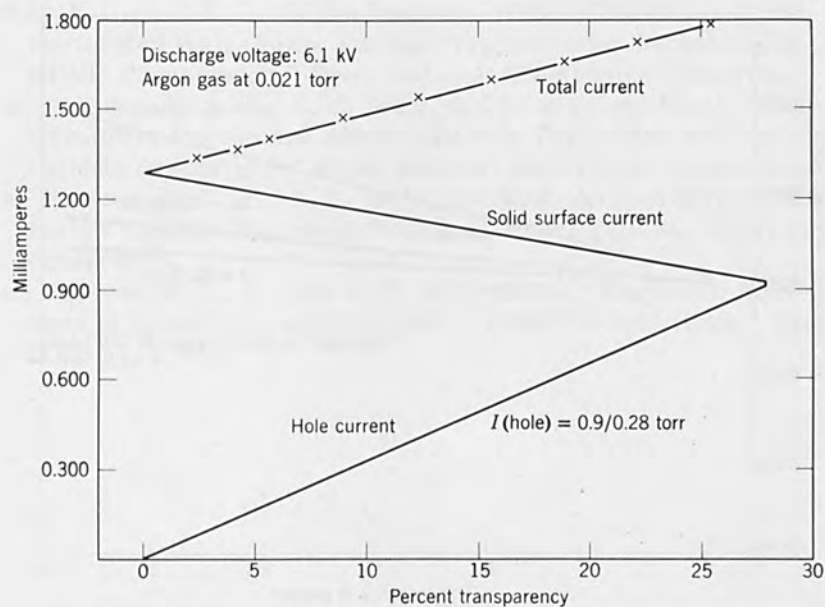


Fig. 17. Tube current versus screen-mesh transparency.

$$I_{(\text{hole})} (\text{electron}) = A_1 J_c^+ C_{vp} \quad (26)$$

Substituting into equation 24 the value for $I_{(\text{hole})}(\text{total})$ from equation 23, we obtain

$$\frac{\rho}{C_3} = C_{vp} A_1 J_c^+ \left\{ \frac{1+C_{vp}}{C_{vp}} \right\} \quad (27)$$

Solving for the electron hole current, we obtain

$$I_{(\text{hole})} (\text{electron}) = \frac{\rho}{C_3} \left\{ \frac{C_{vp}}{1+C_{vp}} \right\} \quad (28)$$

REFERENCES

- Boring, K. L., and L. H. Stauffer (1963), "A New-Nonthermionic Electron Gun," Proc. National Electronics Conference, Vol. 19, p. 535.
Fowler, R. D., and G. E. Gibson (1934), "The Production of Intense Beams of Positive Ions," Phys. Rev. 46, 1075.

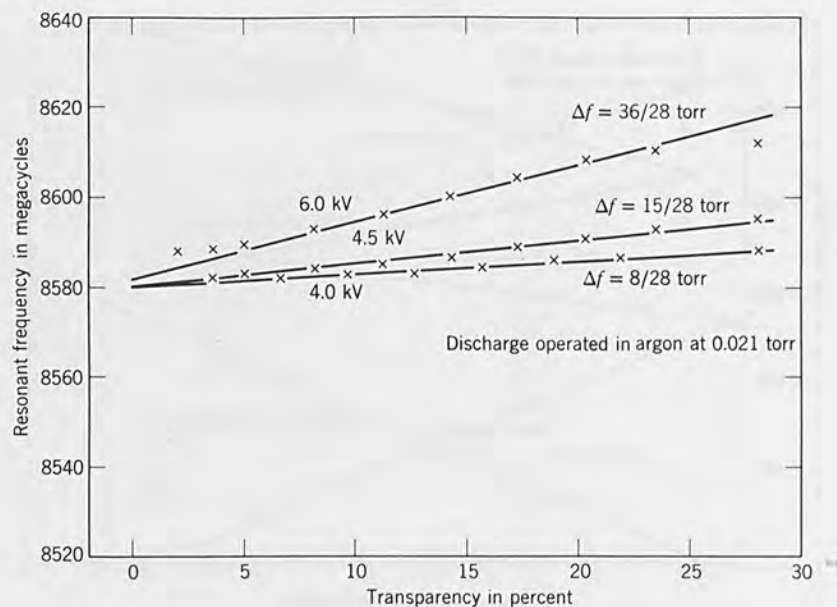


Fig. 18. Microwave resonance frequency versus screen-mesh transparency.

- Harrison, E. R. (1959), "Emission of Relatively High-Energy Ions from Low-Voltage Arcs," *Nature*, **184**, 245.
- Harrison, E. R., and R. H. Dawson (1958), "Apparatus for Producing Plasma Beams," *J. Electronics and Control* **5**, 29.
- Hernqvist, J. D. (June, 1960), "High-Voltage Electron Extraction from an Arc-Discharge Plasma," *RCA Rev.* **21**, 170.
- Lidsky, L. M., S. D. Rothleder, D. J. Rose, S. Yoshikawa, C. Michelson, and R. J. Markin (1962), "Highly Ionized Hollow Cathode Discharge," *J. Appl. Phys.* **33**, 2490.
- Little, P. F., and A. von Engel (1954), "The Hollow-Cathode Effect and the Theory of Glow Discharges," *Proc. Phys. Soc. (London)* **A224**, 209.
- Kofoed, M. J., and H. V. Cleva (1962), "Bursting of Electron Beams with Electron Trapping," *J. Electronics and Control* **13**, 525.
- Livingston, R. S., and R. J. Jones (1954), "High-Intensity Ion Source for Cyclotrons," *Rev. Sci. Instr.* **25**, 6, 552.
- Muly, E. C. and J. Litton (1964), "Recent Developments in Electron Beam Mode Discharge Diagnostics," Twenty-Fourth Conference on Physical Electronics, M.I.T.

- Muly, E. C., and H. L. L. van Paassen (1963), "The Effect of the Perforated Wall Hollow Cathode Transmission Factors upon the EBMD Phenomenon," Proc. National Electronics Conference.
- van Paassen, H. L. L., E. C. Muly, and R. J. Allen (1962), "Electron Beam Phenomena Associated with Perforated Wall Hollow Cathode Discharges," Proc. National Electronics Conference.
- van Paassen, H. L. L., E. C. Muly, and R. J. Allen (1962), "Cold Hollow Cathode Discharge Welding," Proc. National Electronics Conference.
- van Paassen, H. L. L., and E. C. Muly (1964), "Empirical Equations of Spherical Cathodes used in EBMD Experiments," Proc. IEEE 52, 3, 316.

MECHANISM OF HIGH-POWER-DENSITY ELECTRON-BEAM PENETRATION IN METAL†

INTRODUCTION

With high-power-density electron beams it is possible to penetrate solids or liquids several orders of magnitude deeper than their classical range. For example, a beam of electrons of only 150-keV energy and approximately 0.3-mm diameter penetrates a steel plate of 2.5-cm thickness within 50 msec (see Fig. 1 and Fig. 1 in Ref. 1). Recently, even 10.0 cm penetration was achieved. The startling fact is that after the beam had been interrupted no hole could be found, although the electrons could be detected on the other side of the plate at energies not much different from their initial value at the entrance. Only the grain structure of the affected zone had been changed and even in the case of stainless steel, the mass density had been slightly decreased, but the plate is found to be vacuum tight. The penetration depth achieved is about one thousand times larger than the classical penetration depth $x_{\max}$.¹ From Whiddington's law [2] $x_{\max}$ can be calculated by

*Hartford Graduate Center, Rensselaer Polytechnic Institute of Connecticut, East Windsor Hill, Conn.

†A slightly shorter version of this paper appeared in the *Journal of Applied Physics*, July 1964. The author acknowledges the permission granted by the *Journal of Applied Physics* to have the paper included in this book. Portions of this work were performed with facilities at Philips Electronic Instruments, Mt. Vernon, N.Y., Union Carbide Nuclear Co., Tuxedo, N.Y. and at different divisions of United Aircraft Corporation.

¹ $x_{\max}$ is defined as the path length of the electrons at the end of which the most probable velocity becomes zero.

$$x_{\max} = a \frac{A}{Z\rho} V^2 \quad (1)$$

in which a is a constant, A the atomic weight, Z the atomic number, ρ the mass density, and V the acceleration voltage of the electron beam before entering the material. This formula becomes for iron, using practical units,

$$x_{\max} = 3.0 \times 10^{-9} V^2 \quad (1a)$$

where $x_{\max}$ is in microns (μ), if V is entered in volts. This function is plotted in Fig. 2 and shows that electrons of 100 keV lost their energy after reaching a depth of approximately 30μ (≈ 1 mil), a depth which is much less than high-power-density electron beams can penetrate. Figure 2 can also be used for nickel, chromium, copper, aluminum, tungsten, tantalum, and lead, applying the scale factors as shown in the table in Fig. 2.

From this discrepancy it must be concluded that the material had substantially lowered its density along the electron-beam path during the penetration process. The electron beam must have transformed part of the material in low-density vapor along its path.

When electrons impinge on material they are stopped after having passed through a certain thickness; therefore, they give up their kinetic energy. From classical considerations of collision processes, this transfer cannot be direct to the lattice atoms of mass M , but first to the lattice electrons of mass m , since the ratio r of transferred energy and initial energy before collision is

$$r = \frac{2m}{(m+M)^2} \approx \frac{2m}{M} \quad (2)$$

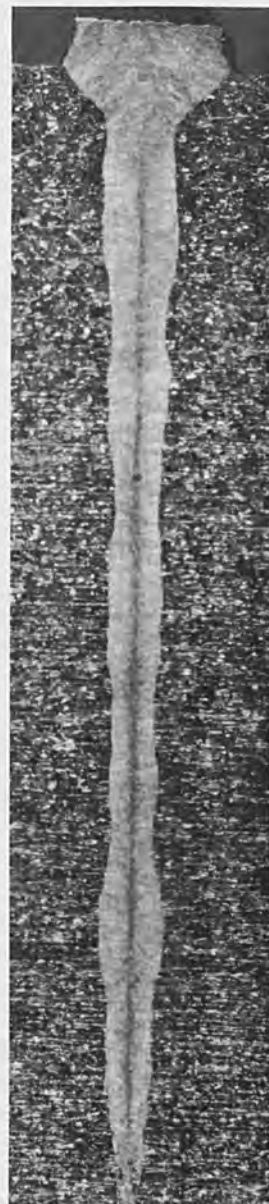


Fig. 1. Electron-beam weld pass in stainless steel type 302, 1-cm thick at $V = 150$ kV and $i = 9$ mA

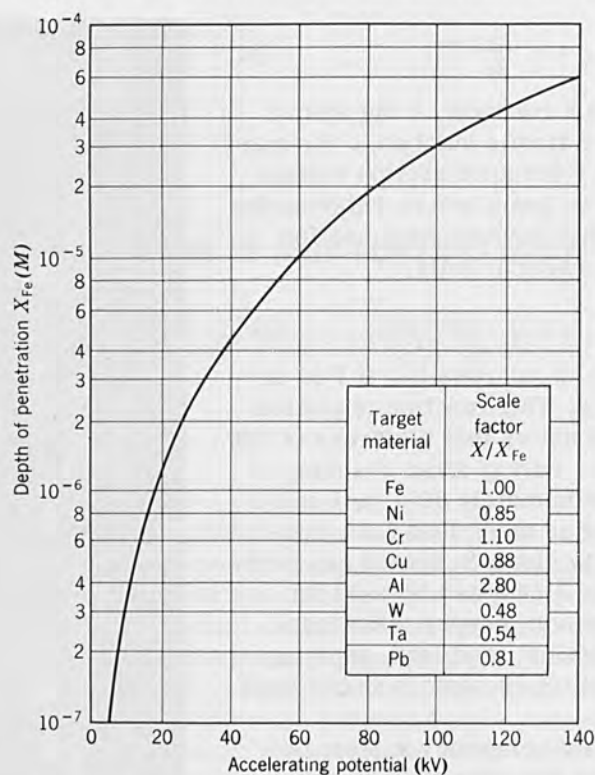


Fig. 2. Penetration depth of electrons in a constant density medium as a function of acceleration voltage.

For iron r is only 2×10^{-5} , i.e., only 0.002% of the kinetic energy of the electrons would go directly to the lattice. Therefore, practically all direct transfer will occur with the lattice electrons. Most of this transfer will result in higher vibrational energy. The lattice electrons then partially transmit their increased vibrational energy to the total lattice. The amplitude of the lattice vibrations is therefore increased which means that the material can reach a very high temperature. The material melts and even evaporates or sublimates directly. This evaporation permits the deep penetration of electrons. Such deep penetrations as observed with high-power-density electron beams can only be achieved by evaporation of a fine channel within the material. Whether this material leaves the workpiece or whether some stays in and increases the density at its boundaries has to be investigated. It seems to be unlikely

that the material which had been in the volume occupied by the electron beam during the process had all left the channel completely and then returned, since it cannot be explained how it could come back through the narrow long channel (less than 1/3 mm in diameter), whose walls are relatively cold. Mass density measurements before and after the process confirm this, i.e., in stainless steel, for example, a decrease of approximately 2% was observed.

Extensive electron probe X-ray microanalysis and radioactive tracing techniques showed that the electron-beam affected material went through a complete mixing all along the electron-beam zone. This was found by inserting a thin cobalt sheet and, in another experiment, a radioactive metal sheet, between two plates of iron; the electron beam then penetrated normal to the plane of entrance. A metallurgical cut through the plane passing through the center line of the electron-beam path was scanned, so that the degree of intermixing could be measured by the two techniques mentioned.

High-speed cinematographs taken of the electron-beam process in steel indicated that during most of the time the surface is closed by a liquid metal film and that it ruptures at certain instances for a very short period. Using measurements of the rate of evaporation from the surface, the surface temperature, i.e., the temperature of the thin liquid metal film covering the hole, was determined. The temperature at the bottom of the hole was calculated from an equation set up for the process. Also the evaluation of the diffusion equation for the tracing experiments yields the same order of magnitude for the bottom temperature.

MEASUREMENTS

Mass Density

Mass density measurements at two different specimens were carried out, one at pure iron, the other at stainless steel (type 302). As to be expected the density, ρ_a , of the electron-beam affected zone of volume V_a was always lower than that, ρ_u , of the unaffected part of volume V_u . Assuming that during the process the mass density of the solid and liquid part of the electron-beam affected zone was equal or even somewhat larger than that of the unaffected part before the process, there should have been an "empty" space of volume V_e available for the electron beam. If we assume, as a first approximation, V_e as being a cylinder of diameter d_e and $V_a = V_u$ cyl-

inders of diameters d , which could be estimated from metallurgical cuts, d_e can easily be calculated from the density measurements and from the 10% precise estimates of d by using the following simple relations:

$$\begin{aligned} (V_u - V_e) \rho_u &= V_a \rho_a \\ d_e &\approx d \left(1 - \frac{\rho_a}{\rho_u} \right)^{1/2} \end{aligned} \quad (3)$$

The measurements and results are listed in Table I. The decrease in density due to probable oxidation in the relatively poor vacuum

Table I

Change of Mass Density in Electron-Beam Affected Zone

	ρ_u [g cm ⁻³]	ρ_a [g cm ⁻³]	ρ_a/ρ_u	d [cm]	d_e [cm]
Pure Iron	7.879	7.587	0.963	0.12	2.3×10^{-2}
Stainless Steel	7.921	7.828	0.012	0.13	1.4×10^{-2}

of 10^{-4} to 10^{-5} torr was found to be negligible. The electron-beam diameter as calculated from the electron-optical characteristics of the electron-beam machine is somewhat larger than equation 3 would permit for the practically empty electron-beam volume. However, the values of d_e lie in the range of the electron-beam diameter of 0.005 to 0.010 in. ($= 1.3 \times 10^{-2}$ to 2.5×10^{-2} cm). The small differences are within the precision of the measurements and the calculations of the electron-beam machine characteristics. It is also feasible to assume that the electrons might be "squeezed" through the somewhat narrower channel like a gas under pressure.

These simple considerations make it clear that it does not seem necessary to assume that material has to flow back. This is more evident, since during the process there might be a temporary slight increase in density due to radial pressure which pushes the material against the internal wall of the electron-beam channel.

Temperature of Electron-Beam Affected Zone during Process

Since the electron-beam process is very fast and therefore does not reach an equilibrium, it is doubtful whether one can talk about temperatures in their true sense. But one can, to a first approxi-

mation, assume at least two temperatures, one that exists at the surface, T_f , of the thin foil that covers the hole and bursts open only during very short intervals (less than 1/2 msec), the other temperature, T_b , that exists at the bottom of the channel where the electron beam impinges and transfers most of its energy to the workpiece. Figure 3 shows evidence of a thin film covering the channel; this photograph had been taken by Brasch and Lange, while



Fig. 3. Blister formation by subsurface heating (Brasch and Lange [3]).

an electron beam hit the surface of a metal for a very short time. Two blisters are visible here, one showing a slight crack, the other already ruptured as a result of internal pressure.

Surface Temperature, T_f . Since the heated surface spot is very small, measurements of its temperature did not seem feasible with a pyrometer. Also thermocouples would have introduced material problems and serious errors. By determining the amount of vapor leaving unit area during unit time, the temperature of the surface can be calculated from simple formulas of the kinetic theory of gases, assuming that the vapor density is not too high. The number v of vapor atoms, that leave the heated area reduced to unity and

during unit time is given by

$$V = \frac{1}{4} \tau \bar{c}_v N_v \quad (4)$$

$\bar{c}_v$ being the mean thermal velocity of the vapor atoms and N_v the density; τ is the transmission coefficient which is $\tau \leq 1$ and takes into consideration the fact that in a dense vapor a portion of the vapor atoms are scattered back by its own vapor cloud which it has to penetrate. However, for most cases described in this paper, it is very close to unity. Assuming $\tau = 1$ might yield somewhat lower temperatures than really exist at the surface. After substituting for $\bar{c}_v$ and N_v their relations to temperature and pressure, equation 4, expressed in total amount of mass evaporated per unit time and per unit area reads

$$R_{ev} = \left(\frac{M_v}{2\pi k N} \right)^{1/2} \frac{p_v}{T^{1/2}} \quad (5)$$

M_v molecular weight, k Boltzmann's constant, N Avogadro's number, p_v vapor pressure at temperature T_v of surface. In practical units for iron this can be written as follows:

$$R_{ev-Fe} = 4.36 p_v T_v^{-1/2}$$

where R_{ev-Fe} is in $\text{g cm}^{-2} \text{sec}^{-1}$, if p_v is introduced as torr and T_v in $^{\circ}\text{K}$.

This relation is plotted in Fig. 4 for iron. For the purpose of the measurement in this paper, the ratio $p/T^{1/2}$ is also plotted as a function of temperature using the recent data as compiled by R. E. Honig [4].

In order to collect the vapor, a hollow half-sphere (diameter 4 in.), see Fig. 5, of pure copper sheet was placed on the work piece; a small hole of 0.2-cm diameter at the top allowed the electron beam to pass. Two measurements were performed, one using the Zeiss Electron Beam Welder and the other using the Hamilton Standard-Zeiss Electron Beam Welder with somewhat higher power density. The conditions and results of the measurements are listed in Table II. The temperatures were determined from Fig. 4, using the right-side ordinate.

Since the curve of Fig. 4 is rather steep at the range of the surface temperature, the results do not differ very much over a relatively wide range of evaporation rates. Therefore, the indicated surface temperatures of 2770 and 2800 $^{\circ}\text{K}$, respectively, should be quite accurate. The two determinations differ by 1%, which is surprising for this high temperature, and the deviation has the correct

sign, since the higher one refers to a higher power density of the electron beam.

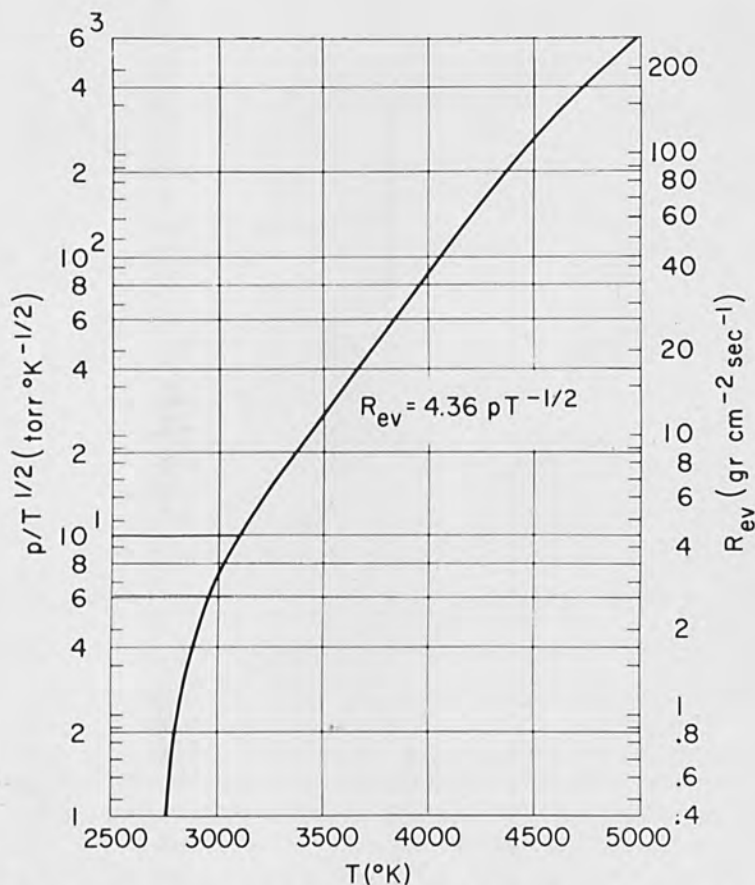


Fig. 4. Evaporation rate of iron as a function of vapor pressure and surface temperature.

Bottom Temperature, T_b . Figures 6 and 7 depict a few photo frames of high-speed cinematographs (2500 frames per second). The time scale starts arbitrarily at a randomly picked moment of surface rupture. At three instances on the photos the rupture of the film can be observed, at 72 msec, 145.4 msec, and 222.0 msec. On the photographs the effect of rupturing cannot be observed as readily as on the actual movie. The probability that the occurrence of a rupture might have been missed on the frames, i.e., might

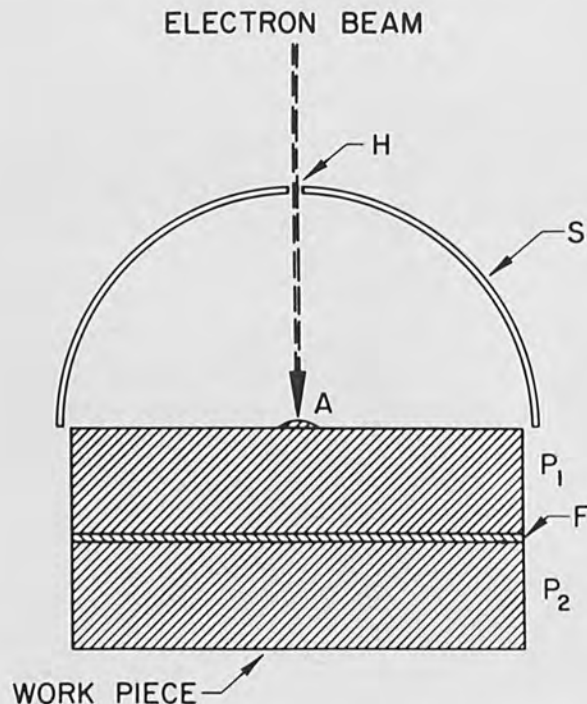


Fig. 5. Schematics of vapor collection and tracing experiments.

have occurred just between two frame exposures is very small, otherwise the intervals would not be equally long between the ruptures, approximately 72-77 msec in this particular example.

The surface film must be very thin ($< 1\mu$, because the range of 100-keV electrons is 30μ); all attempts to measure or determine theoretically its thickness from the energy balance failed. The reason for this is that the energy dissipated by scattering of electrons within this film is negligible. Therefore, it does not play a significant role in the energy balance of the electron-beam process. But it certainly will account for the wave shape of the longitudinal cross sections of electron-beam welds in a plane of the electron-beam axis (see, for example, Fig. 1). Also, it will partly explain the fact that the bottom temperature, T_b , of the electron-beam channel is much higher than the surface temperature, T_f . The periodical ruptures of the film will release the vapor pressure built up underneath its surface. The metal vapor density within the

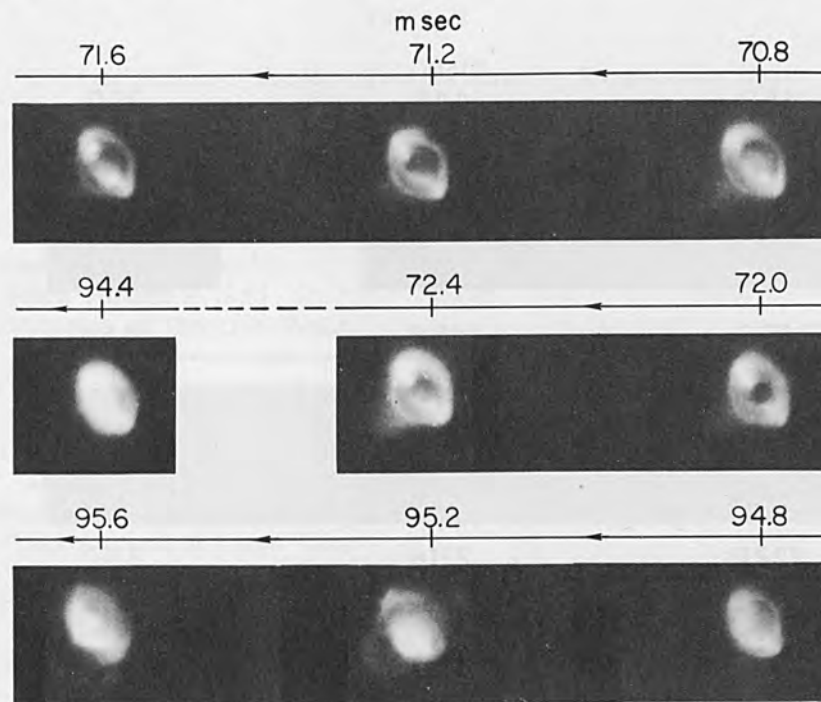


Fig. 6. High-speed cinematographs of electron-beam welding process. Time scale starts arbitrarily at randomly picked moment of rupture of liquid film. (70.8 to 95.6 msec, rupture at 72.0 msec.)

electron-beam channel will fluctuate between a very low value $\rho_{\min}$ and a critical maximum value ρ_{cr} . When the vapor density is at its maximum, the electron beam will scatter the most and therefore widen the channel. Irregularities are due to impurities in the lattice and at the boundary surface which holds the film closed by surface tension.

Because of the high-density electron stream, a high degree of ionization of vapor atoms will exist and, therefore, a "gas-focusing" effect will occur in addition to the scattering and reflections from the channel walls due to the forced flow of electrons. This effect has been known from the early days of cathode ray tubes in which a small amount of inert gas was introduced in order to focus the

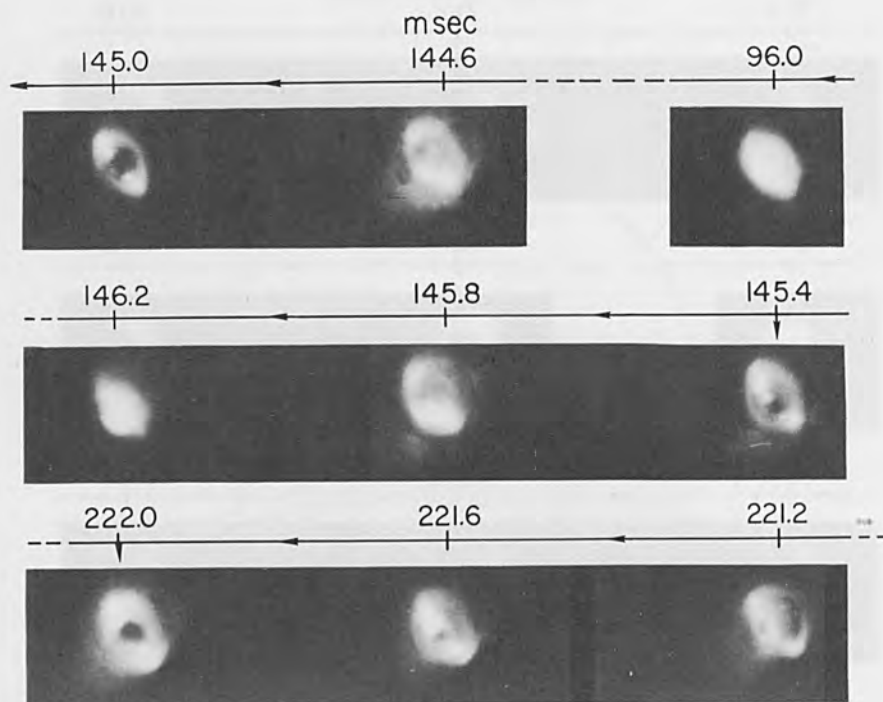


Fig. 7. Continuation of Fig. 6, range 96.0 to 222.0 msec. Ruptures at 145.4 and 222.0 msec.

electron beam. It is due to the radial diffusion of electrons which is much faster than that of the heavier ions; thus, a kind of electrostatic lens is formed. Although some emphasis had been given to this focusing effect [5], and although it had subsequently been stressed as the predominant cause [6] for the focusing effect and for the wave-shaped longitudinal section [7], it alone cannot describe this mechanism, especially how the electron beam remains in a narrow channel, despite the fact that it enters the surface in focus under an aperture angle of almost 5 degrees. Under extreme conditions, the electrons can be considered as a gas that is forced through a narrow channel; reflections at the walls and scattering are the major effects that can explain this mechanism. In the case of gas-focusing, one would expect quite uniform waves in the longi-

Table II

Determination of Surface Temperature by Evaporation Rate

	Experiment I (Zeiss Machine)	Experiment II (Hamilton-Zeiss Machine)
Beam Current i [mA]	11.5	8
Beam Voltage V [kV]	125	120
Diameter of Electron Beam d_e [cm]	0.02	0.013
Diameter of Electron-Beam Affected Area d_a [cm]	0.08	0.04
Power Density of Electron Beam at Workpiece D_d [Watt meter ⁻²]	5×10^{10}	7×10^{10}
Duration of Process t [sec]	23.0	45.0
Total Amount of Vapor Con- densed G [mg]	58.0	44.4
Evaporation Rate R_{ev-Fe} [g cm ⁻² sec ⁻¹]	0.5	0.79
Temperature T_f [°K]	2770	2800

tudinal section of the weld and not as many irregularities as can be seen on the left part of the photograph of Fig. 1, shown in reference [1]. Only scattering can be affected by irregularities, i.e., impurities, in the crystal structure as can be seen from this example.

As already pointed out, the density of the vapor within the channel covered by a thin metal film of time-variable thickness x changes between a value $\rho_{\min}$ (practically zero) and ρ_{cr} . When ρ_{cr} is reached, a critical pressure p_{cr} has built up under the liquid metal film that exerts a force F_{cr} which just begins to exceed the surface tension of the metal skin held by its periphery to the bulk of the work piece. Equating these two forces should yield, to a first approximation, the critical pressure p_{cr} that causes the liquid metal foil to rupture.

$$p_{cr} \frac{\pi}{4} d_f^2 = \gamma \pi d_f \quad (6)$$

$$p_{cr} = 4 \gamma / d_f$$

γ is the coefficient of surface tension and d_f the diameter of the liquid film at the surface, which should be closer to the diameter of the electron beam than to that of the electron-beam affected zone.

There exist a few measured values of γ in the literature [8], [9]. For iron, the most recent value of γ was taken from measurements by Namba and Isobe [9]: $\gamma = 1510 \text{ dynes cm}^{-1} = 1.5 \text{ kg sec}^{-2}$.

Assuming $d_f = 3.0 \times 10^{-4}$ meters (a value estimated 15% higher than the electron-beam diameter as calculated from the electron-optical characteristics of the electron-beam machine),

$$p_{cr} = 2 \times 10^4 \text{ Nm}^{-2} = 150 \text{ torr} ,$$

which corresponds to a vapor atom density of

$$N_v = \frac{p_b}{kT_b} = 2.7 \times 10^{17} \text{ cm}^{-3}$$

where T_b was taken to be approximately 5300°K as calculated later. Therefore, a maximum mass density of $\rho_{cr} = 2.5 \times 10^{-5} \text{ g cm}^{-3}$ is to be expected, which means that on a path length of 1 cm, less than 1% of the initial electron-beam energy (for example, 100 keV) will be lost in the vapor [10].

DETERMINATION OF BOTTOM TEMPERATURE, T_b

After each rupture of the liquid metal skin at the surface, the metal film is formed again, since the pressure inside the channel is lowered considerably below the critical value that corresponds to the surface tension. But the formation will soon arrive at a quasi-steady state of uniform maximum thickness after it ruptures. The net rate of condensation and evaporation will soon become zero; the liquid film will neither increase nor decrease any more. Any tendency to increase in thickness would cause an increase in scattering within the film with an accompanying increase of vaporization. At the film there are essentially three terms of condensation and evaporation. As shown in Fig. 8, V_1 is the rate at which vapor atoms arrive from the bottom of the channel and condense at the inside surface of the film, V_2 is the rate at which vapor atoms leave the film towards the inside of the channel with downward velocity components, and V_3 is the rate at which vapor atoms leave the film to the outside of the workpiece. Since the film is very thin, its temperature should be the same at the upper side and at the lower side; therefore we can write $V_2 = V_3$. V_3 had al-

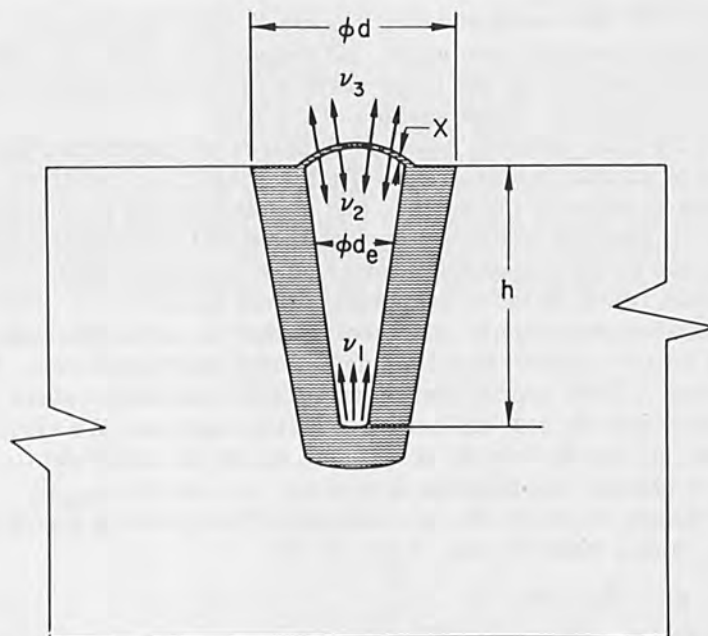


Fig. 8. Condensation and evaporation at liquid metal film.

ready been measured and served to determine the surface temperature T_f . The heat needed for the evaporation rate $V_2 = V_3$ comes partly from the scattering of electrons within the foil and partly from the conduction and radiation originating from the bottom of the electron-beam channel where most of the electron-beam energy is dissipated. According to Whiddington's law [2] at $V_0 = 100$ kV, even in a relatively thick iron foil of $10,000 \text{ \AA} = 1 \mu$, only 1.5% of the total power is dissipated.

For the calculation of the maximum bottom temperature, it is assumed that, in first-order approximation, a steady state of evaporation and deposition will be reached at the film before it ruptures. This assumption is justified by two facts.

1. The maximum film thickness must be very small, otherwise the electron beam could not perform the work of such deep penetration and could not have been detected with such little loss of energy after passing through as much as 5 cm of steel.

2. The period during which the channel is closed by the film is longer than the duration of penetration.

These two facts lead to the conclusion that most of the material

escaping from the electron-beam volume V_e must have gone through the following three steps: (a) evaporation from its original site, (b) "deposition" at the inner surface of the film, and (c) re-evaporation from the outer surface of the film.

Since the time interval between ruptures is longer than the duration of normal penetration, while the beam is in relative movement parallel to the surface and maintaining its perpendicular impact, the film would become too thick and its temperature too high due to the increasing scattering of electrons. This, in turn, would result in more evaporation from the film, i.e., the film would become thinner and "cooler" due to less scattering; thus the build-up of the film thickness would start again, etc. This means that in first approximation an equilibrium temperature of the surface will be reached before the film ruptures. We arrive, therefore, at a quasi-steady state when the liquid metal skin ceases to change its thickness with time, i.e., $dx/dt = 0$.

The change of thickness, dx , consists of two parts, a positive one, dx_1 , and a negative one, dx_2 :

$$dx = dx_1 + dx_2 \quad (7)$$

dx_1 is a function of the bottom temperature, T_b , and its corresponding vapor pressure, p_b , whereas dx_2 is a function of the film temperature, T_f , and its corresponding vapor pressure, p_f . dx_2 is negative since it concerns the evaporation from above and underneath the thin foil; it is therefore:

$$dx_2 = - \frac{1}{\rho_e} (V_2 + V_3) m_v dt \quad (8)$$

where ρ_e is the mass density of the liquid iron and $V_2 = V_3$ are given in number of vapor atoms leaving unit area during unit time. By using equation 5, equation 7 can also be written as:

$$\frac{dx}{dt} = \frac{1}{\rho_e} \left(\frac{M_v}{2 \pi k N} \right)^{1/2} \left(\alpha \frac{p_b}{T_b^{1/2}} - 2 \frac{p_f}{T_f^{1/2}} \right) \quad (9)$$

where α is the factor that takes into account the geometry of the channel while assuming that the accommodation coefficient is one. For the steady state where $dx/dt = 0$, we arrive at a simple relation between T_b and T_f :

$$\alpha p_b T^{-1/2} - 2 p_f T^{-1/2} = 0 \quad (10)$$

α can be calculated applying the cosine law [11]. At a given diam-

eter d_e of the electron-beam channel, it decreases with the square of the depth h of this channel.

For any given electron-beam penetration depth, h , and surface temperature, T_f , it is possible to determine the bottom temperature, T_b , by applying the vapor pressure curves of Honig [4], for example, and using the relations as plotted on the curves of Fig. 4. Of course, equations 9 and 10 are applicable only for the case where $h \gg d_e$ which was the main assumption for the approximate calculation of α from the cosine law. Figure 9 represents a family of curves from which one can read the temperature T_b for dif-

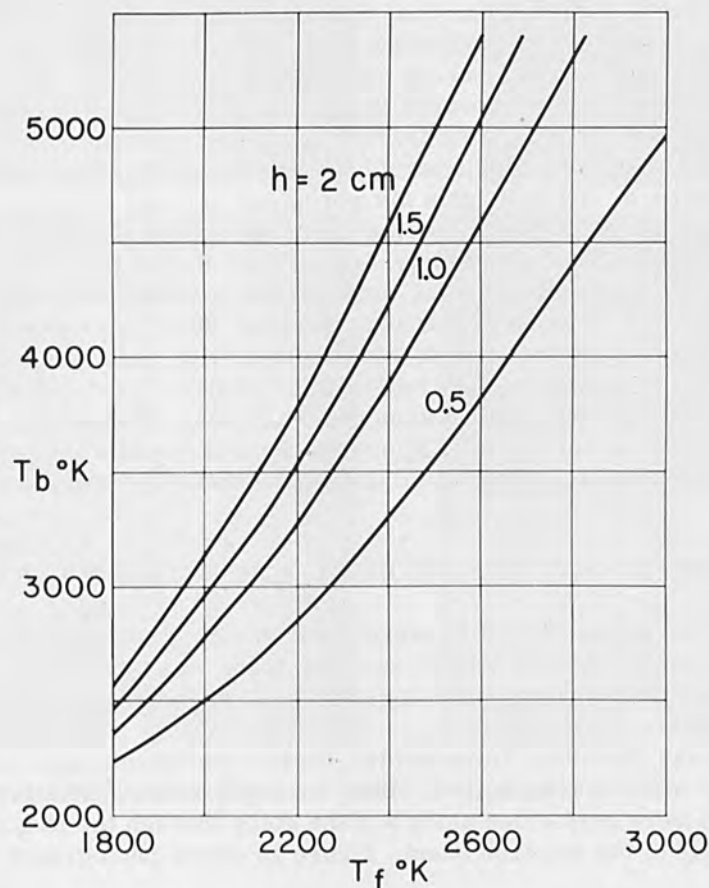


Fig. 9. Bottom temperature T_b as a function of surface temperature T_f at different penetration depths h for pure iron.

ferent surface temperatures T_f at various penetration depths, h , as parameter. The curves have been calculated using equation 10. For the two measurements of the surface temperatures as indicated above, viz., $T_f = 2770^\circ\text{K}$ and $T_f = 2800^\circ\text{K}$, we obtain at a depth of 1 cm and a width of 10 mil ($= 2.54 \times 10^{-4}$ meter) for the electron-beam channel a temperature at the bottom $T_b = 5200^\circ\text{K}$, and $T_b = 5300^\circ\text{K}$, respectively. This means that the bottom is approximately 2500°C hotter at a depth of 1 cm than the surface film itself.

TRACING EXPERIMENTS

In order to determine in what manner the material dislocates within the electron-beam affected zone, two different types of experiments were carried out. In one type, a foil of dissimilar metal F (Fig. 5) was pressed between two plates, P_1 and P_2 , of the same metal. In another type, a radioactive layer was introduced, so that the whole workpiece consisted of only one metal. A hollow copper half-sphere, S , covered again the workpiece in order to collect on the inside the metal vapor. At the top of the sphere a hole, H , of approximately 2 mm in diameter was drilled so that the electron beam could pass and reach the workpiece at A without any influence from the Cu sphere. For the two plates 99.8% pure iron was used. Spectroanalysis showed that the impurities consisted mainly of copper, ($1 < 0.1\%$ Cu, $2 < 0.04\%$ Ni, $3 < 0.03\%$ C, $4 < 0.004\%$ Mn, $5 < 0.001\%$ Cr). No cobalt traces could be found. Therefore, cobalt was chosen for the foil, F ; also because it has similar properties of diffusion, evaporation, and conduction.

EXPERIMENTS OF TYPE I (COBALT-FOIL BETWEEN PLATES)

The two plates, P_1 and P_2 , were equal in size, 2-in. square and 1/4-in. thick. The foil had the same area and was 0.5-mm thick. The three pieces were bolted together by 8 machine bolts 3/16 in. in diameter.

With the Hamilton-Zeiss welder, three experiments were carried out² with this workpiece. After the experiments, metallurgical cuts were performed along a plane going through the longitudinal axis of the electron beam. Figure 10 shows photographs of

²Courtesy of J. W. Meier, Hamilton Standard Division of United Aircraft Corporation, Windsor Locks, Conn.

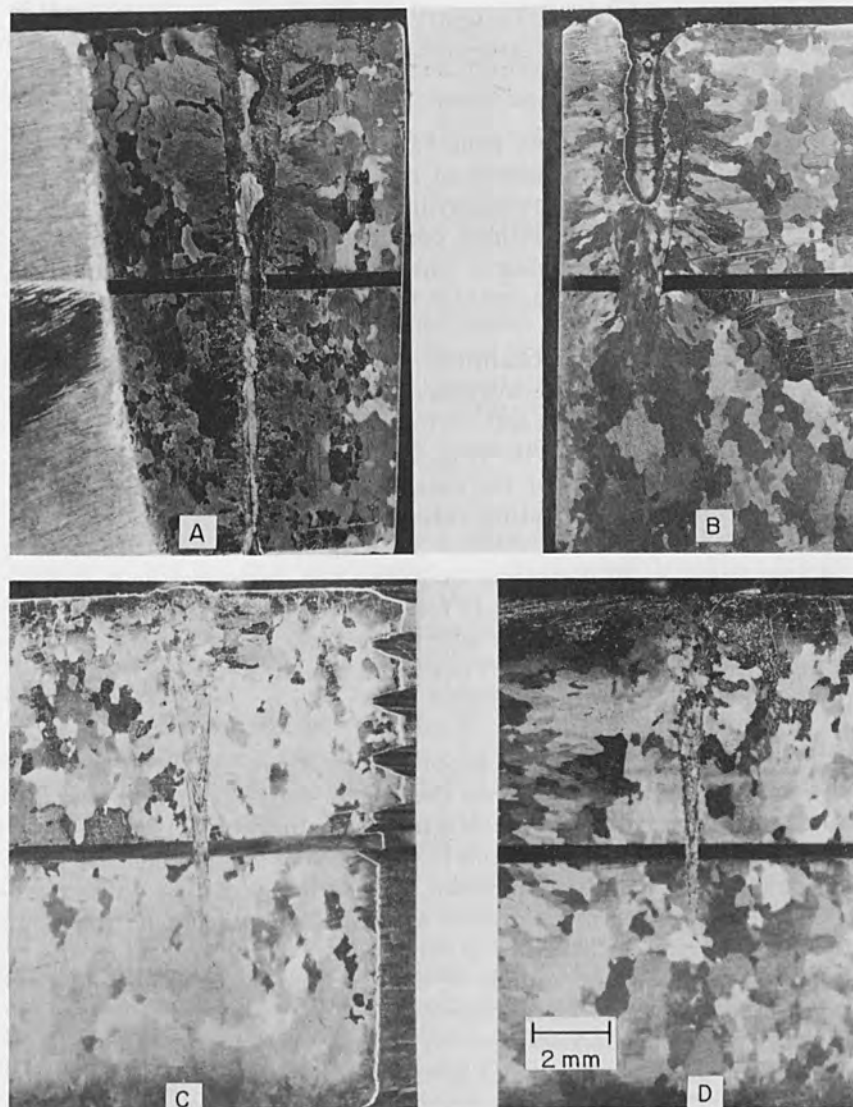


Fig. 10. Photographs of penetration (7 x magnification).

these cuts that were etched slightly. The tapered electron-beam affected zone is clearly distinguishable by the different grain structure.

Experiment 1. Complete penetration with a beam current of 10 mA and acceleration voltage of 120 kV (Fig. 10, photo A); no investigations were performed with this piece, since part of the vapor left the workpiece without being collected at the opposite side of the copper half-sphere. The power density was obviously too high for this thickness, so that the channel remained open.

Experiment 2. With a beam current of 8 mA and an acceleration voltage of 120 kV, the workpiece was not penetrated completely, although the beam was left on for approximately 45 sec which caused a considerable cavity (Fig. 10, photo B). This experiment was mainly carried out for the determination of the surface temperature from the evaporation rate.

Experiment 3. At a current of 10 mA and acceleration voltage of 120 kV, the beam stayed only for approximately 110 msec at one spot, since the workpiece was moved in a plane normal to the beam with a speed of 2.14 mm per second in a circle with a diameter of 24 mm (Fig. 10, photo C).

The total amount of metal vapor per unit penetration depth of the electron beam condensed on the copper half-spheres in experiment 3, excluding the amount originating from the cavity, was less than twice as much as collected in experiment 2, although the volume covered by the electron beam at approximately the same penetration depth in both experiments was almost one hundred times more in experiment 3 than in experiment 2. This means that the material occupying the volume of the electron beam in the workpiece stays within the workpiece and recondenses at the inside wall of the channel opposite the direction of the moving electron beam. Only such an amount of metal vapor that corresponds to a volume slightly more than the volume of the electron beam itself leaves the workpiece.

An attempt was made to determine the amount of cobalt vapor condensed at the half-sphere; but the methods available did not allow better than 25% precision for that small total amount. However, the relative cobalt content in the condensed area was approximately 1% of the total, i.e., less than was found in all cross sections of the weld zones. As will be seen later, even at the surface

of the workpiece, 4% cobalt could be detected. The difference between the cobalt concentration in the condensed film at the copper sphere and within the electron-beam affected zone is mainly due to the lower evaporation rate of cobalt as compared with that of iron at the surface temperature and due to losses by reflection from the Cu sphere.

COBALT DISTRIBUTION IN ELECTRON-BEAM AFFECTED ZONE

In order to find the cobalt distribution in the electron-beam affected zone, the metallurgical cuts, as shown in the photographs of Fig. 10, were scanned by electron probe X-ray microanalyzers. The equipment used was from Cambridge Instruments, England, and another developed by Philips Electronic Instruments, Mt. Vernon, N.Y.

In order to cross-check, the two instruments were partly used for the same sample. They did not show any difference in their results within the precision range (which is about 10^{-12} to 10^{-13} g absolute). The electron probe of the Philips instrument had a finer spot, approximately 0.5μ , than that of the British instrument which had a probe diameter of approximately 1μ . The former had a beam current of 0.2 to $0.3 \mu\text{A}$, whereas the latter had a somewhat higher beam current of $0.35 \mu\text{A}$. The acceleration voltage used in the Philips instrument was 30 kV and that in the Cambridge instrument 25 kV . With the British instrument, pictures (approximately 200 times magnification) of the scanned area either from the emitted X-rays or from the reflected electrons could be taken. The $K_{\alpha 5}$ -line of cobalt was used for the analysis. As 100% reference points the counting rates were taken of the X-rays received from the pure cobalt foil hit by the electron probe. In Fig. 11 are plotted the point counts as measured with the British instrument along the center line of the electron-beam affected zone. Directing the electron probe on the pure cobalt surface produced in the scintillation counter of the British instrument, the counting rate of $r_{Co} = 27,270.1 \text{ cps}$; it showed a background rate of $r_b = 415.3 \text{ cps}$ when directed on the pure iron surface. In this experiment the electron beam had not moved (Fig. 10, photo B). The counts were started at the surface of the electron-beam affected zone, i.e., at the bottom of the cavity as seen in phot B of Fig. 10. The cobalt distribution is almost constant at approximately 4% all along the center line. The location where the cobalt sheet had been before the process is indicated.

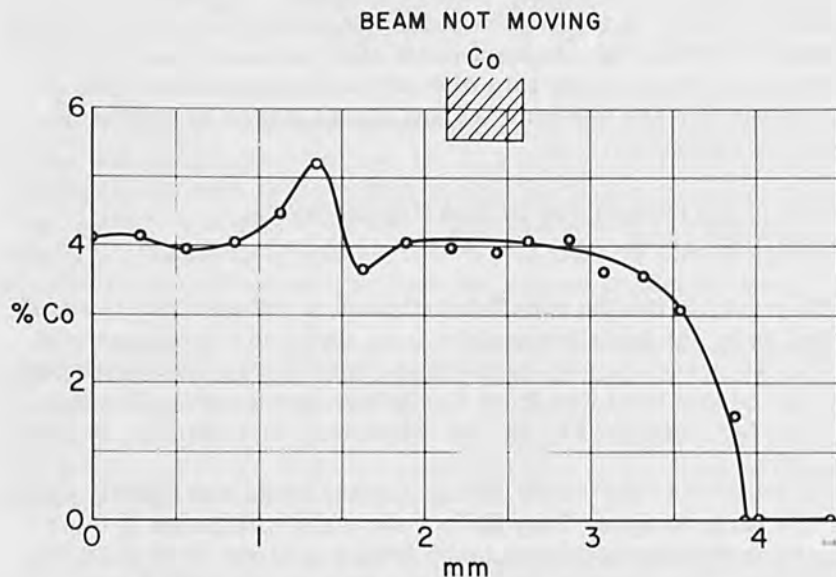


Fig. 11. Point counts on center line of electron-beam affected zone. Experiment No. 2 beam not moving.

The distribution of the cobalt content in the neighborhood of the cobalt sheet can also be seen in the photographs of Fig. 12. These photographs show the area of the cut where the electron-beam affected zone passes through the cobalt sheet. The three photos on the left of Fig. 12 show the X-ray emission of the area scanned with the electron probe, while the three photos on the right are exposures of the reflected electrons taken of the same area under an angle different from the Bragg angle.³ Several scanings across the electron-beam affected zone perpendicular to the center line of the electron beam were performed; the path of such a scanning is indicated as white lines on the photos of Fig. 12; each path length took 20 min for the scanning. The counting rate scanning along this path is shown as a typical example for a recorded scanning in Fig. 13. The steep concentration gradient at the boundary is surprising. Over a path length less than 30μ the concentration of cobalt drops from 100% down to approximately 5%. Almost as steep is the gradient at the cross sections far away from the vicinity of

³ The diamond-shaped impression marks served for identification of location.

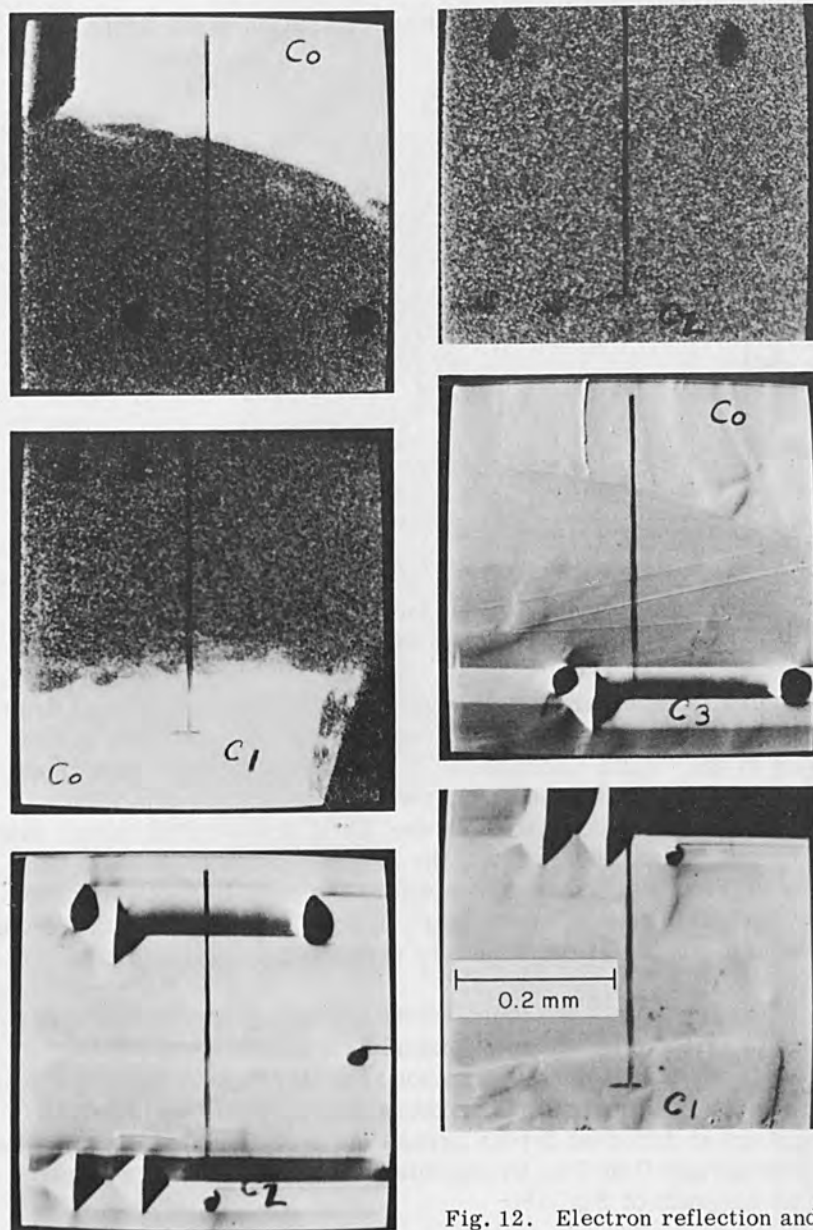


Fig. 12. Electron reflection and x-ray emission photographs of electron-beam affected zone with cobalt content in vicinity of cobalt sheet.

the cobalt sheet where the cobalt concentration of 4% drops down to zero.

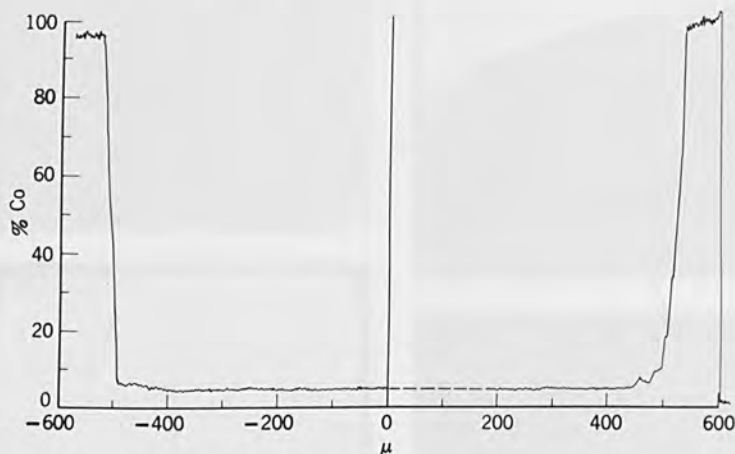


Fig. 13. Typical example for a recorded counting rate scanning of X-rays as adjusted for 100% cobalt.

Figure 14 summarizes all results in a three-dimensional drawing. The center line of the electron beam is marked here by the 0-line of the "width-coordinate" (x -coordinate); the y -coordinate indicates the depth of the measured point, and the z -coordinate gives the cobalt content in per cent. Even at the points where originally there was 100% cobalt, the percentage drops down to the same value of 4%. The position where the cobalt sheet was originally is clearly visible where there is no "floor" in the drawing and the cobalt content at the boundary exceeds the scale and goes up to 100%.

Similar results were found where the workpiece moved with a speed of 12.8 cm per minute, experiment 3 (photograph B of Fig. 10); only the cobalt content was not as uniform as in the fixed beam experiment. Near the surface, there was only 2.5% Co content. Scannings at different depths across the metallurgical cut as shown on Photograph C of Fig. 10 are summarized in the three-dimensional diagram of Fig. 15.

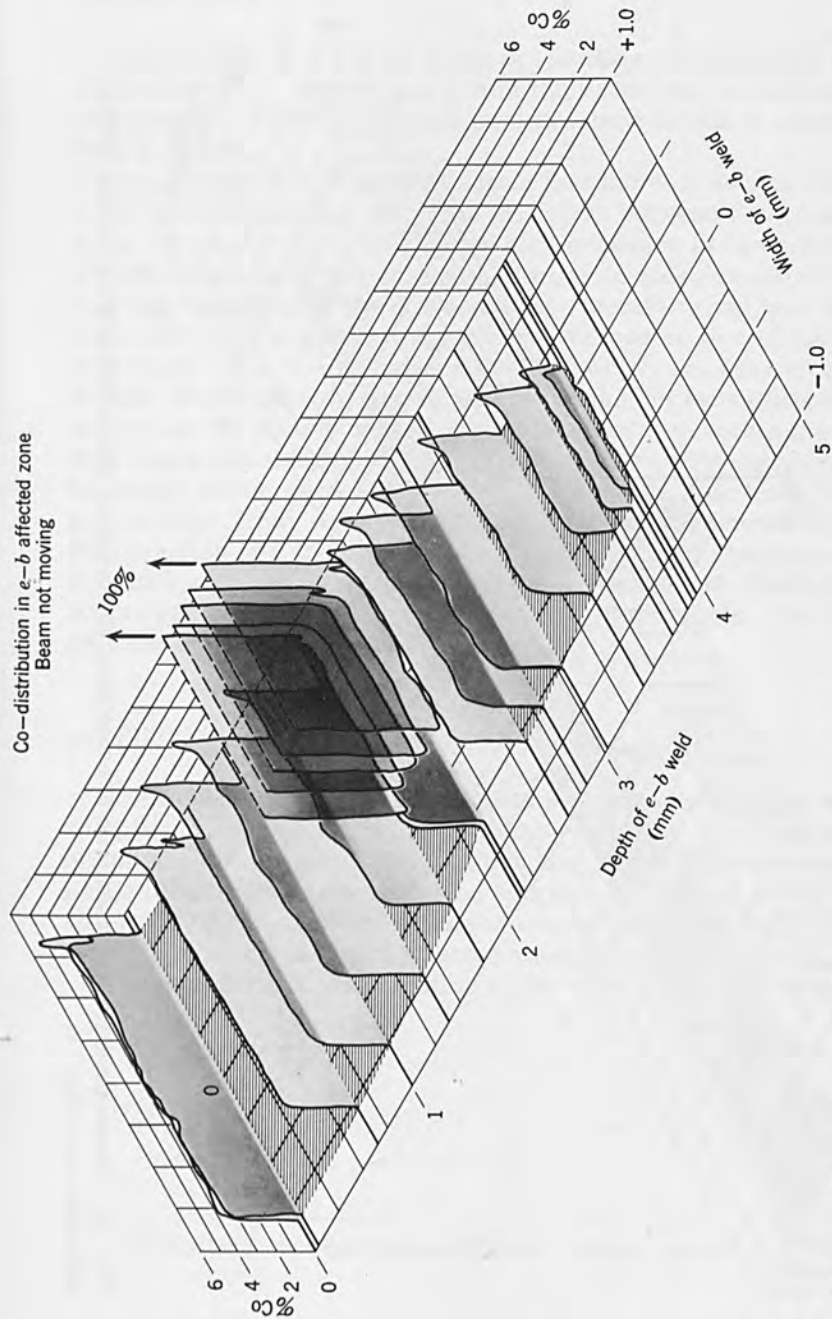


Fig. 14. Three-dimensional presentation of electron probe scanings at workpiece treated with fixed electron beam. x-Coordinate: width in electron-beam affected zone; y-coordinate: depth of the measured points; z-coordinate: percentage of cobalt.

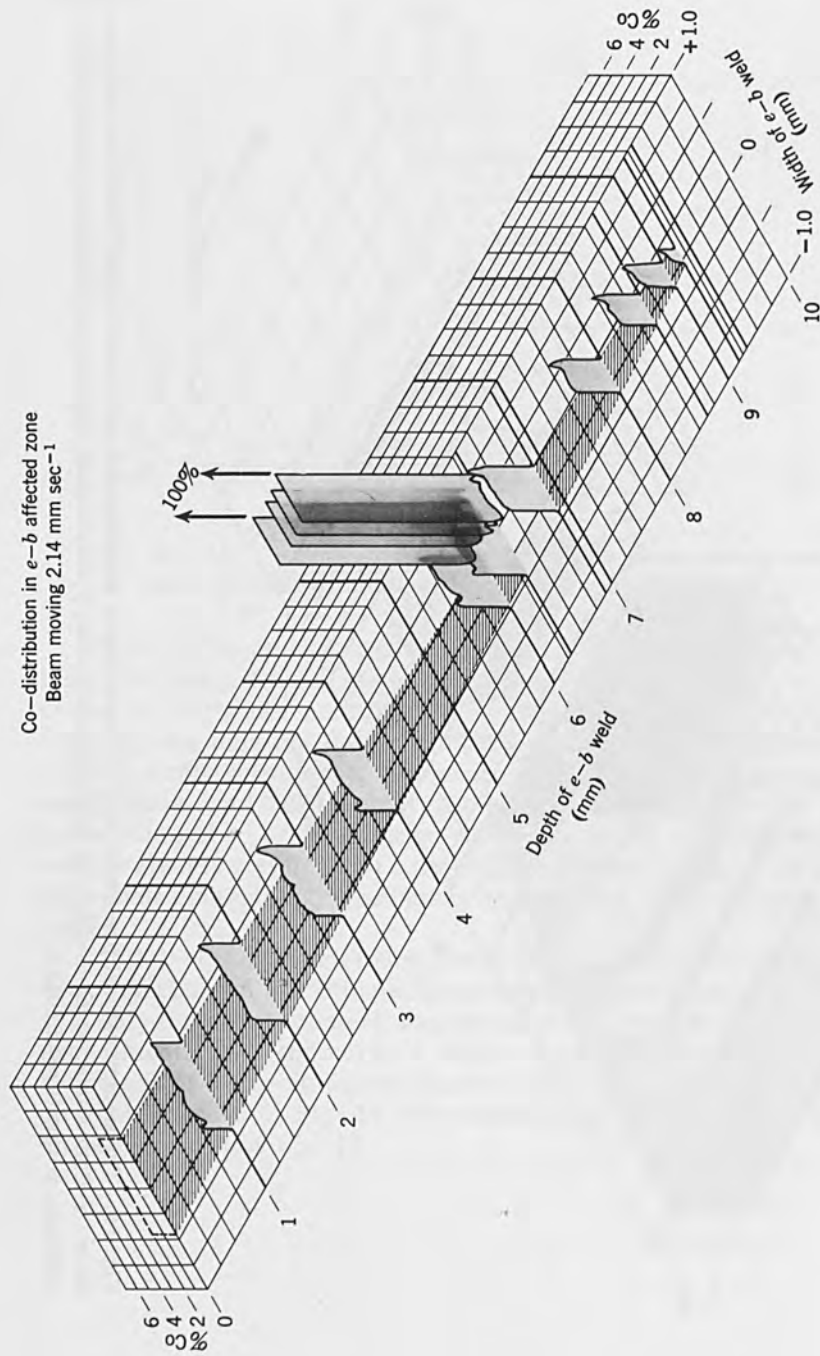


Fig. 15. Three-dimensional presentation of experiment at workpiece with electron-beam moving, same co-ordinates as in Fig. 14.

EXPERIMENTS OF TYPE II (HOMOGENEOUS METAL, RADIOACTIVE FOIL)

An iron foil of the same type iron as described before with the dimensions 2-in. square and 1-mm thick had been irradiated with neutrons in a 5-MW swimming pool reactor⁴ so that it reached 500 μ C of Fe₅₉.

Preliminary experiments showed that the best way to bond such a foil without changing the grain structure between two iron plates (2 in. $\times$ 2 in. $\times$ 1/4 in.) was under high pressure in vacuum at 800°C for about 10 hours. Experiments similar to those as performed with the cobalt sheet were carried out. Metallurgical cuts were made through a plane passing through the center line of the electron beam. They were exposed to films which are sensitive to β -rays. Densitometer measurements of the degree of blackening determine the distribution of the radioactive iron within the electron-beam affected zone. They confirm the results obtained with the cobalt sheet, but were less precise and are, therefore, not reported here. They were mainly carried out to determine whether the insertion of a dissimilar metal by mechanical clamping would simulate sufficiently well a homogeneous metal and whether microscopic gas occlusion at the interface of foil and plates would have affected the experiments.

DIFFUSION ANALYSIS

The fast mixing of the two metals in the electron-beam affected zone is only possible at very high temperatures. The laws of diffusion should be applicable. Unfortunately, there are not many reliable measurements of diffusion "activation" energy which would allow one to calculate the diffusion coefficient at the high temperature of the electron-beam affected zone during the process.

The one-dimensional solution of the equation of diffusion:

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial x^2} \quad (11)$$

is given by:

$$\frac{n}{n_0} = (\pi Dt)^{-1/2} e^{-x^2/4Dt} \quad (12)$$

⁴Irradiations were performed at Union Carbide Nuclear Co., Tuxedo, N. Y.

where n_0 is the initial surface concentration at the interphase and n the concentration of the diffusing material at a depth h after a time t . D is the diffusion coefficient. Its dependence on the temperature is given by:

$$D = D_0 e^{-q/kT} \quad (13)$$

where q is the "activation energy" per atom and k Boltzmann's constant. For the system C-Fe (carbon-iron) as given by Jost [12], $q = 2.54 \times 10^{-19}$ J, and $D_0 = 4.9 \times 10^{-1}$ cm² sec⁻¹.

At 5000°K the diffusion coefficient for such a system would be $D_{5000} = 1.2 \times 10^{-2}$ cm² sec⁻¹. Entering these values into the solution of equation 12 and assuming $n/n_0 = 0.05$ cm⁻¹ and $t = 1$ sec, h would be 0.7 cm. It is likely that the system C-Fe can be compared with the experiments performed with Co-Fe. The calculated values are of the same order of magnitude as those measured.

DISCUSSION AND SUMMARY

The experimental and theoretical investigations of the deep penetration of high-power-density electron beams have shown that the effect can be explained in a purely classical way. Electrons are stopped in the material and transfer their kinetic energy to heat by increasing the amplitude of the vibrational energy of the lattice electrons which in turn transmit this energy to the whole lattice. The heat brings the material along the path of the electron beam to melting and successively to evaporation. Initially, one channel is made free from material by this process and it is not filled in again, but the material decreases its mass density and thus in many cases closes the one channel with its own mass by "expanding," i.e., decreasing the mass density and changing the grain structure.

In the case of a moving beam, there is initially a relatively large amount of material that evaporates and escapes to the outside of the workpiece. Subsequently, evaporated material, for providing the volume of low-enough density for the electron beam to pass through, condenses backwards at the inside walls of the channels previously covered by the electron beam. Within a fraction of a second, a practically complete intermixing of the metal takes place.

In many cases a thin surface film covers the electron-beam channel. This film is so thin that it is transparent for the elec-

trons, i.e., there is practically no heat dissipated within this film. But it makes the process economical by shielding the much hotter spot at the bottom of the channel: direct radiation through this foil is not possible, on the contrary, it will reflect the radiation which, in this case of approximately 5000°K, has its peak in the middle of the visible light spectrum. Only when the vapor pressure is built up high enough, so that it exceeds the surface tension, will the surface rupture, but immediately close again. The liquid walls are prevented from collapsing by this pressure and the reaction forces that the evaporation vapor atoms will exert to the surface.

The reaction forces, as already calculated previously [13], are much larger than the pressure that the electrons themselves might exert due to their momentum. But before the vapor pressure inside the electron-beam channel reaches the critical value to exceed the surface tension, the thin film will have reached quasi-steady thickness. This fact makes it possible to calculate, from the evaporation rates and the geometry of the channel, the bottom temperature, which was found to be approximately 2500°C higher than the surface temperature, when applying power densities of the order of 5×10^{10} W per square meter. Analysis of the fast diffusion within the electron-beam affected zone by the application of electron probe X-ray microanalysis and radioactive tracing techniques confirms the high temperature difference.

ACKNOWLEDGMENTS

In portions of the experimental work, the author was most kindly assisted by C. T. Brown and W. Lasko, of United Aircraft Corporation Research Laboratories, East Hartford, Conn.; by R. Dills, of Advanced Materials Research Laboratories of Pratt & Whitney Aircraft Division of United Aircraft Corporation, North Haven, Conn.; and by J. D. Rogers, of North American Philips Electronic Instruments Division, Mt. Vernon, N.Y.

This work was partially supported by Philips Electronics Instruments and United Aircraft Corporation.

REFERENCES

1. H. Schwarz, *J. Appl. Phys.* **33**, 3464 (1962).
2. R. Whiddington, *Proc. Roy. Soc. (London)* **A 89**, 554 (1914).
3. J. Dosse and G. Mierdel, "Der elektrische Strom im Hoch-

- vakuum und in Gasen," Leipzig, 1945, p. 46.
4. R. E. Honig, RCA Rev. 23, 567 (1962).
 5. H. Schwarz, 1961 Trans. Second International Congress on Vacuum Science and Technology, Pergamon Press, New York, 1962, Vol. II, p. 699.
 6. J. W. Meier, *Ibid.*, p. 670.
 7. Cecil E. Hall, Massachusetts Institute of Technology, private communication, 1961.
 8. J. D'Ans and E. Lax, "Taschenbuch f. Chemiker und Physiker," Springer Verlag, Berlin, 1943, p. 1001.
 9. S. Namba and T. Isobe, Proc. 4th Symposium on Electron Beam Technology, R. Bakish (Ed.), Boston, 1962, p. 95.
 10. F. Kohlrausch, "Praktische Physik," Teubner Verlag, Leipzig, 1944, Vol. 2, p. 534.
 11. See, for example, L. Holland, "Vacuum Deposition of Thin Films," Chapman & Hall, London, 1958, p. 143.
 12. See, for example, W. Jost, "Diffusion in Solids, Liquids, Gases," Academic Press, New York, 1960.
 13. H. Schwarz, Rev. Sci. Instr. 33, 688 (1962).

ELECTRON AND ION BEAMS IN MICROELECTRONICS

Section II

ELECTRON AND ION BEAMS
IN MICROELECTRONICS

From the work of the past few years, the fully automated multichamber systems for the deposition of thin films and passive elements, we can see the progress of the technology. These systems through the many control tasks including scanning, microprobe, and microphoto. Some of the early work has been reviewed by Moore (1962) and Wells (1967), while the years 1963 and 1964 have seen a considerable number of contributions on the subject. Many of them will be found in the references of the papers in this issue. To appreciate the growth that has taken place in the last few years, we should only compare mentally the simple work accelerated gun with some of the systems used in the papers to follow. These contributions present new studies and show achievements and trends on both sides of the Pacific. The reader is also advised to consult the proceedings of the 1964 Symposium on Electron Beam Techniques for Microelectronics for additional up-to-date information.

Today both the field of electron and microelectronic components and the utilization of electron and ion beams for microelectronic applications are in a state of flux. It is a period of dynamic change, which in my opinion will need three to five years for stabilization and for the firming up of designs and types of machines that will be required to meet the constantly increasing demands for device production machinery. This will certainly super-

1. vacuum and its gases," *Phys. Rev.* 1948, p. 46.
2. R. E. Hoag, *NCA Rev.* 42, 207 (1953).
3. H. Schwarz, 1961 *Trans. Second International Congress on Vacuum Science and Technology*, Pergamon Press, New York, 1962, Vol. II, p. 659.
4. J. W. Meier, *ibid.*, p. 674.
5. Cecil S. Hall, *Massachusetts Institute of Technology*, private communication, 1961.
6. J. D. Auer and E. Lee, "Techniques of Chemistry and Physics," Springer Verlag, Berlin, 1963, p. 1001.
7. S. Harabara and F. Isobe, *Proc. 4th Symposium on Electron Beam Technology, G. Tabor (Ed.)*, Berlin, 1962, p. 95.
8. F. Kohlrausch, "Praktische Physik," Teubner Verlag, Leipzig, 1954, Vol. 2, p. 534.
9. See, for example, L. Holts, "Vacuum Deposition of Thin Films," *Trans. 2nd Int. Symp.* 1962, p. 244.
10. See, for example, J. W. Lee, "Electron Beam Evaporation," *Trans. 2nd Int. Symp.* 1962, p. 244.
11. H. Schwarz, *Trans. 2nd Int. Symp.* 1962, p. 244.

ELECTRON AND ION BEAMS

IN MICROELECTRONICS

ELECTRON AND ION BEAMS IN MICROELECTRONICS

Today there is hardly a laboratory in the electronics industry of the United States or in other countries possessing advanced technologies which does not use an electron-beam heat source for at least some application. By nature of these concerns' diverse aims and areas of interest electron beams are utilized in a wide range of systems and lead to a variety of products. Predominant in all these must be the utilization of electron beams for evaporation tasks; a survey of the literature beginning with O'Brien and Skinner (1933) will assert this.

From the work-accelerated filament gun to the fully automated multichamber system capable of producing both active and passive elements, we can see the beam transition from a pure thermal source through the many control tasks including scanning microscopy and microprobe. Some of the early work has been reviewed by Moore (1962) and Wells (1962), while the years 1963 and 1964 have seen a considerable number of communications on the subject. Many of them will be found in the references of the papers to follow. To appreciate the growth that has taken place in the last few years, we should only compare mentally the simple work-accelerated gun with some of the systems used in the papers to follow. These communications present new studies and show achievements and trends on both sides of the Pacific. The reader is also advised to obtain the proceedings of the 1964 Symposium on Electron Beam Techniques for Microelectronics for additional up-to-date information.

Today both the field of electronic and microelectronic components and the utilization of electron and ion beams for microelectronic applications are in a state of flux. It is a period of dynamic change, which in my opinion will need some three to five years for stabilization and for the firming up of designs and types of machines that will be best suited to meet the constantly increasing demands for device production machinery. This will certainly super-

cede decisions on type of devices and device specifications of units to be produced. These production systems will be complex machines that will contain electron beams to perform thermal tasks such as evaporation, micromachining, and localized heat treating. They will utilize scanning, and emission microscopes and microphones as integral system component parts. They will further use beams for performance of nonthermal tasks not unlike some of those discussed in this section. These systems will have various degrees of automation and they will be operational both in high and ultrahigh vacuums and they could either be single or multichamber installations. Some of them are certain to use lasers also as complementary tools for performance of the many known and unknown requisite tasks for future microelectronic device production.

One thing that can be asserted is that we will see an unprecedented growth in the utilization of electron and ion beams as production and control tools for the microelectronics industry of tomorrow. The considerable capital equipment requirements for such component production systems indicate convincingly that in the future these systems could be acquired only by those who can meet these capital needs, namely the large and well financed electronics corporations.

Editor

REFERENCES

- Moore, D.W., 1962, in "Introduction to Electron Beam Technology," R. Bakish, Ed., John Wiley and Sons, New York, p. 382.
O'Brian, H. M., and H. W. B. Skinner, Phys. Rev. **44**, 602 (1933).
Proc. Symposium on Electron Beam Techniques for Microelectronics, July 6-8, 1964. Ministry of Aviation, Royal Radar Establishment, Malvern, Worcestershire, England (to be published).
Wells, O., 1962, in "Introduction to Electron Beam Technology," R. Bakish Ed., John Wiley and Sons, New York, p. 354.

MICROMACHINING BY SPUTTERING THROUGH A MASK OF CONTAMINATION LAID DOWN BY AN ELECTRON BEAM

When a surface is bombarded with an electron beam in a normal high-vacuum system, hydrocarbon vapors remaining in the vacuum are polymerized at the point of impact of the beam. This phenomenon is known in electron microscopy as contamination. The contaminant material has a large molecular structure and, therefore, a low sputtering rate. If placed on the surface of a material having a faster sputtering rate, it will protect selectively the areas which it covers from erosion by an ion beam. The selectivity of this erosion can be controlled to the same accuracy as the contamination can be placed on the surface.

All the following experiments were carried out in the specimen chamber of a high-resolution scanning electron microscope with an electron probe width of 100 Å and a measured point to point resolution of 150 Å. (For scanning electron microscope see K. C. A. Smith and C. W. Oatley, *Brit. J. Appl. Phys.* 1955, 6, p. 391.) The probe of the microscope was used to lay down the contamination, and the microscope itself was used to follow the machining process. Although the probe is only 100 Å in diameter the contamination spreads to a diameter of about 500 Å before a layer is formed that is thick enough to adequately protect the underlying surface from sputtering. This 500 Å represents the machining accuracy of the technique.

Lines of contamination were laid down on a single crystal of aluminum and the crystal bombarded with 5-kV argon ions at a current density of 0.5 mA/cm² for 3 min. Walls formed under-

*Engineering Laboratory, Cambridge University, England.

neath the contamination which were $600 \text{ \AA}$ thick and 0.3μ high.

The next step was an attempt to form a transmission grid by laying down the contamination in the form of a lattice on a $1000 \text{ \AA}$ aluminum film and then to sputter away completely the unprotected film. The film was supported on a standard transmission, electron microscope specimen grid. This proved unsuccessful since with the lowest ion-beam current density the film still buckled when it had become appreciably thinned.

A thin film on a solid substrate was used next. It is possible with the scanning electron microscope, by forming the picture with the secondary electrons excited by the primary high-energy scanned beam, to obtain contrast between substances having different secondary emission coefficients. For the purposes of this experiment, a $2000 \text{ \AA}$ gold film was evaporated through a microscope specimen grid on to a single crystal slice of silicon. Gold has a higher coefficient of secondary emission than silicon and appears brighter when viewed by the microscope. The specimen therefore is seen as a dark grid on a light background [see Fig. 1a(E)]. As the gold was removed by sputtering this contrast decreased [see Figs. 1a(F) and (G)] and eventually disappeared, in the area of interest, when all the gold had been removed [see Fig. 1a(H)]. The contamination, seen as dark lines, was laid down by leaving the electron probe scanning over each line separately for 45 sec. The ion beam was positioned so that only the outer edge of its profile bombarded the area under study. The gold was first sputtered away from the area shown at the bottom of Figs. 1a(E) to (H). Figures 1a(B) and 1b(B) show the specimen when the gold had been partly removed in the region of the contamination lines leaving gold ridges on the thinned gold film. Figures 1a(C) and 1b(C) show the same area after further bombardment. Practically all the gold had been sputtered away except where the contamination protected it, and arrays of gold ridges on a silicon base remained.

Because the gold film was uneven not all the unwanted material was removed at the same time. The further erosion required to complete this removal has the unfortunate result of thinning the desired structure as well. The contamination used to form the lattice, as opposed to the raster of lines, was thicker because the line over which the probe was scanned was shorter and the time of formation for each line the same. This explains why in Figs. 1a(D) and 1b(D), after 5 min bombardment, the lattice was still complete, while the raster of lines had itself begun to sputter away. At this stage all the contamination must have been removed from the raster and probably

from the lattice as well, leaving only ridges of gold. The ion beam was incident at 45 degrees to the surface in the direction shown in Fig. 1a(E) which explains why the ridges in the transverse direction on the micrographs appear narrower; they are viewed end-on, while those vertically oriented are viewed partly from their sides.

To investigate the situation when the conducting film to be micromachined is on an insulating substrate, a 4000 Å gold film was evaporated on to a glass slide. By forming the scanning picture with the signal obtained from the high-energy electrons reflected from the specimen charging can be overcome and the microscope used to examine insulators. Contamination was laid down as before [see Fig. 2a(A)], and Figs. 2a(B), (C), and (D) follow the process of machining through its various stages of surface removal. The vertically oriented bars in Fig. 2a(C) are surrounded by an island of material which probably results from the shielding effect of the contamination laid down while the bars themselves were being examined with the complete raster of the scan. Again the ion beam was incident at 45 degrees to the surface.

Figures 2b(A), (B), and (C) were taken at the same stages of machining as Figs. 2a(B), (C), and (D) and show the area surrounding a piece of dust on the surface close to the contamination lines. The current density of the ion beam striking both areas is the same. The height of the step between the shielded material underneath the piece of dust and the adjacent bombarded area is a measure of the amount of surface removed. Figures 2b(D) and (E) show this step at higher magnification after 7 min and 13 min of ion bombardment. Its height was 4000 Å in both cases, which is the thickness of the original gold film within the accuracy of the measurement. It was assumed, therefore, that all the gold had been removed after 7 min and that then the surface erosion slowed down considerably. This probably results from the glass charging up to the ion-beam potential and therefore the beam reaching the surface with zero energy. This phenomenon, occurring when an insulating substrate is used, acts as a useful automatic stop to the etching process and the charging becomes an advantage rather than a disadvantage as might have been expected.

Figure 3 shows a cross micromachined on a copper film. The width of the vertical bar is about 500 Å which represents the best machining accuracy so far obtained with the technique. The height of the bars above the surface is approximately 2000 Å. This is measured from the angle of impact of the ion beam and the difference in apparent width of the two bars.

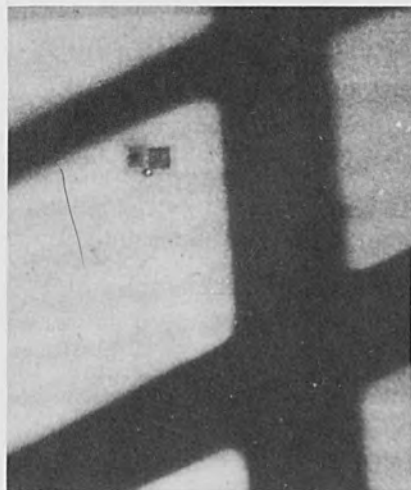
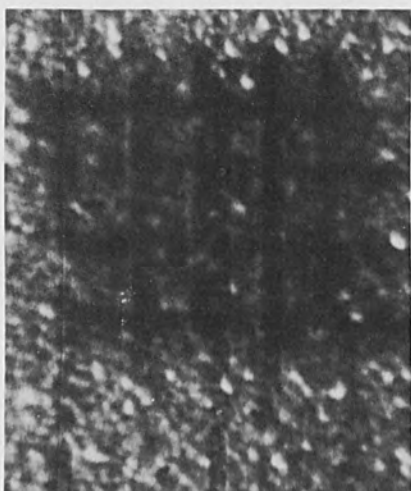
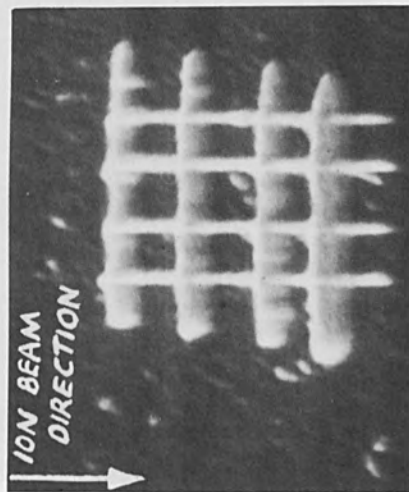
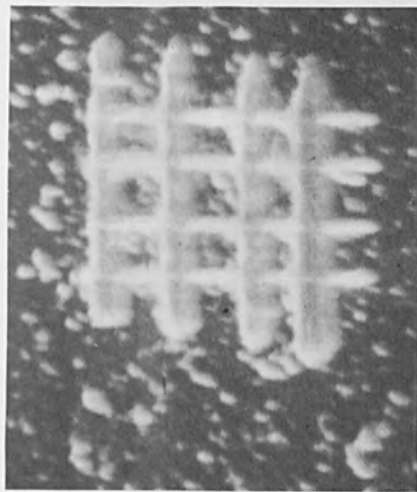
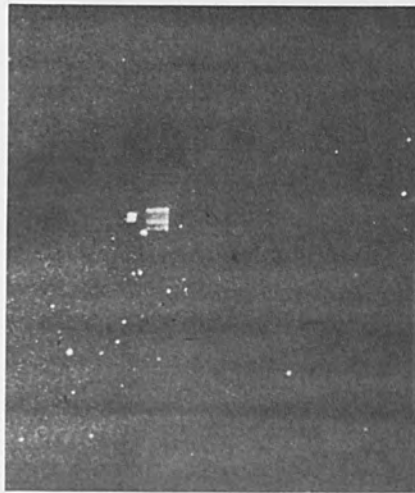
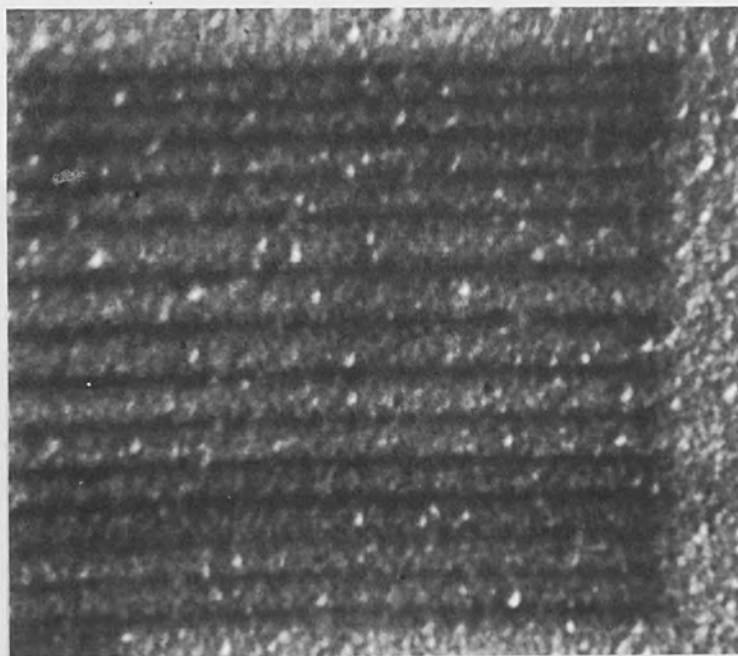
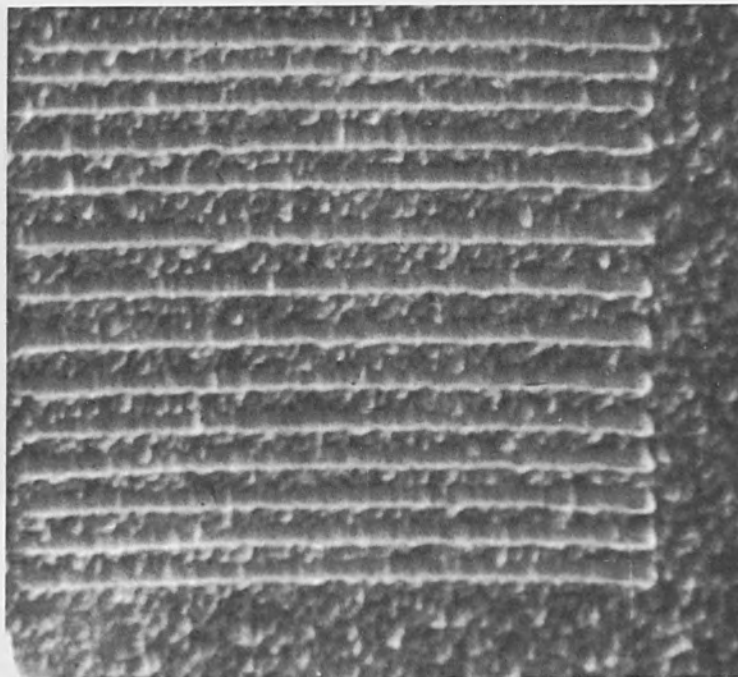


Fig. 1a. Argon ion bombardment of a gold film on a silicon substrate. (A) Shows the lattice of contamination lines laid down with the probe of the scanning electron microscope.







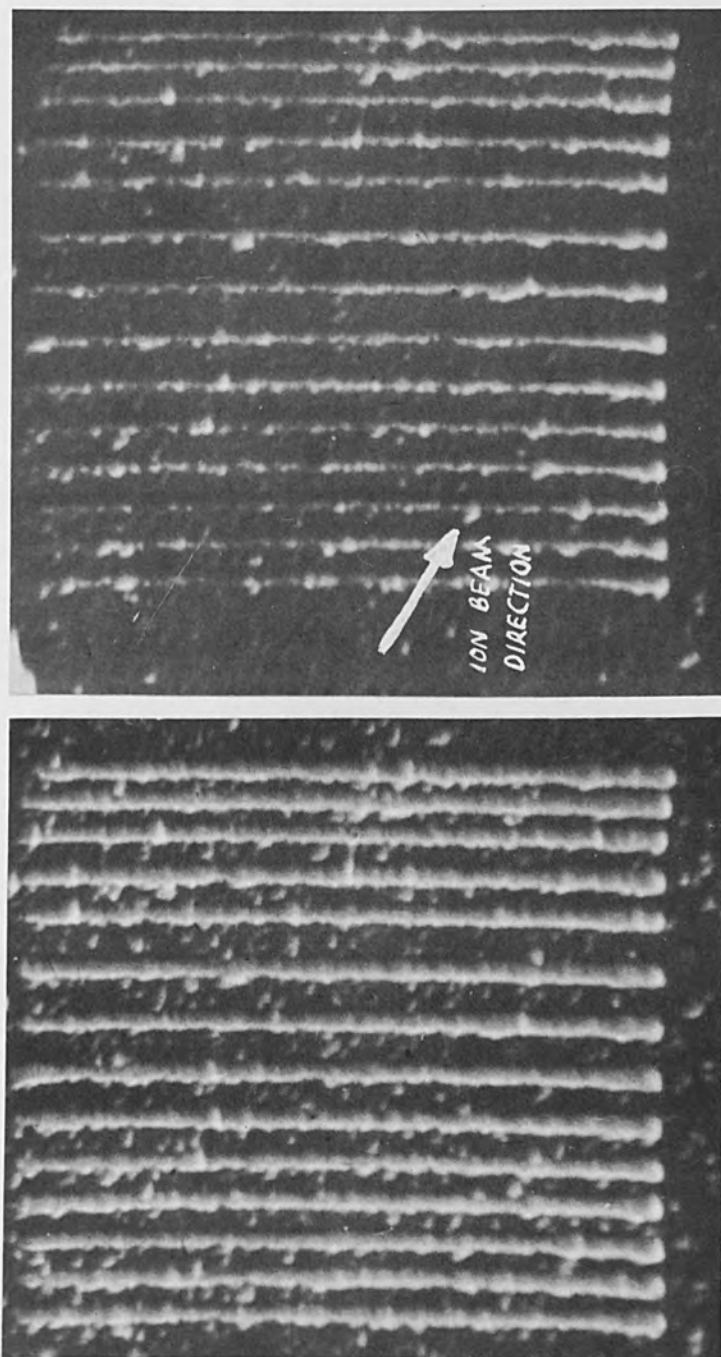
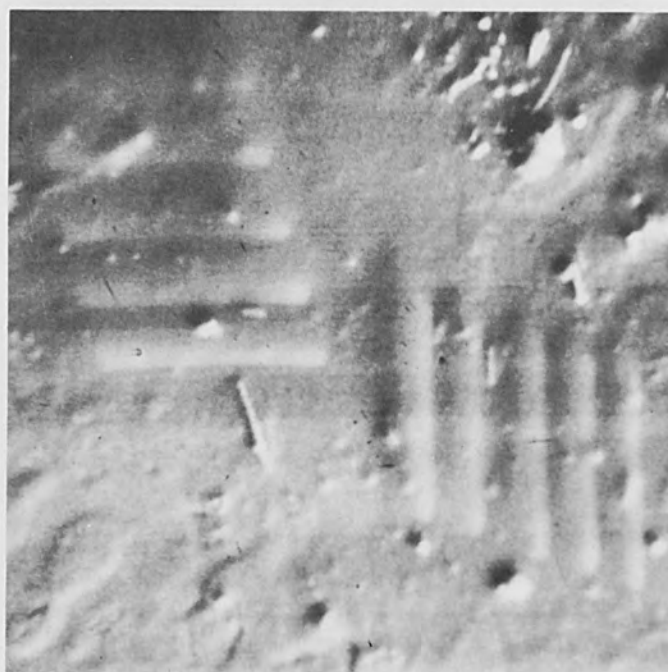
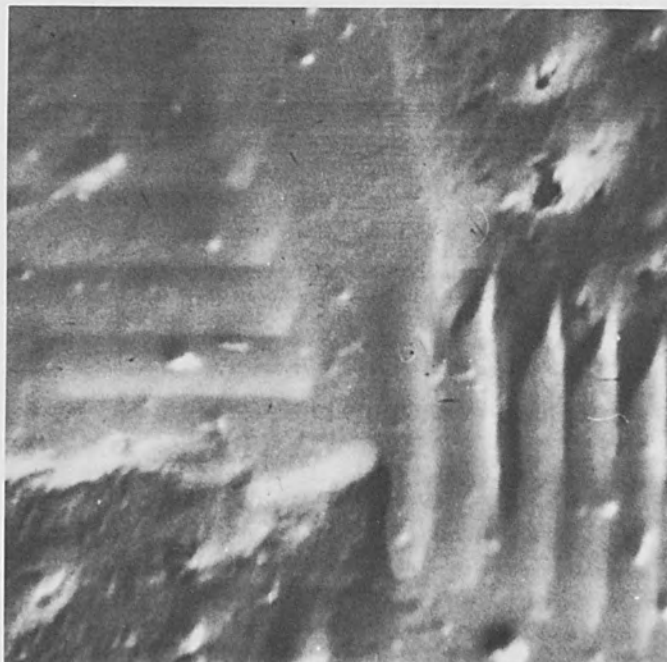


Fig. 1b. Argon ion bombardment of a gold film on a glass substrate. (A) shows the lines of contamination laid down with the probe of the scanning electron microscope.



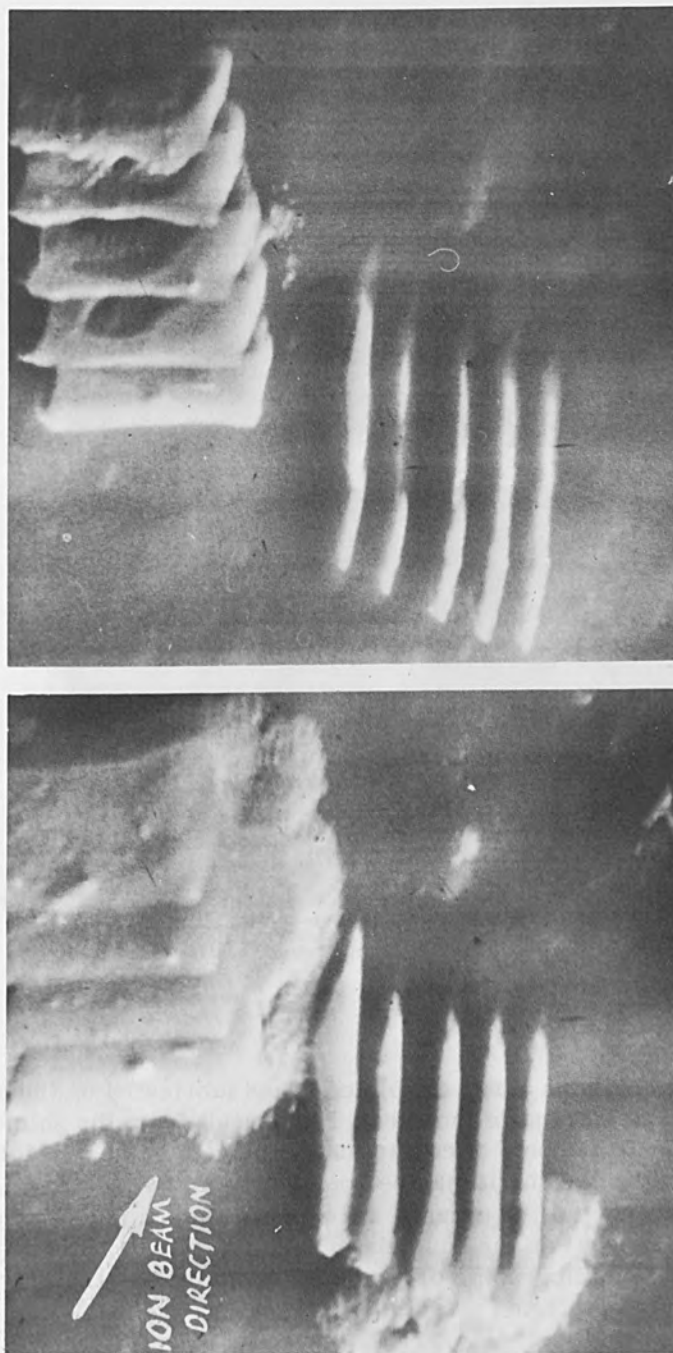


Fig. 2a. Argon ion bombardment of a gold film on a glass substrate. (A) shows the lines of contamination laid down with the probe of the scanning electron microscope.

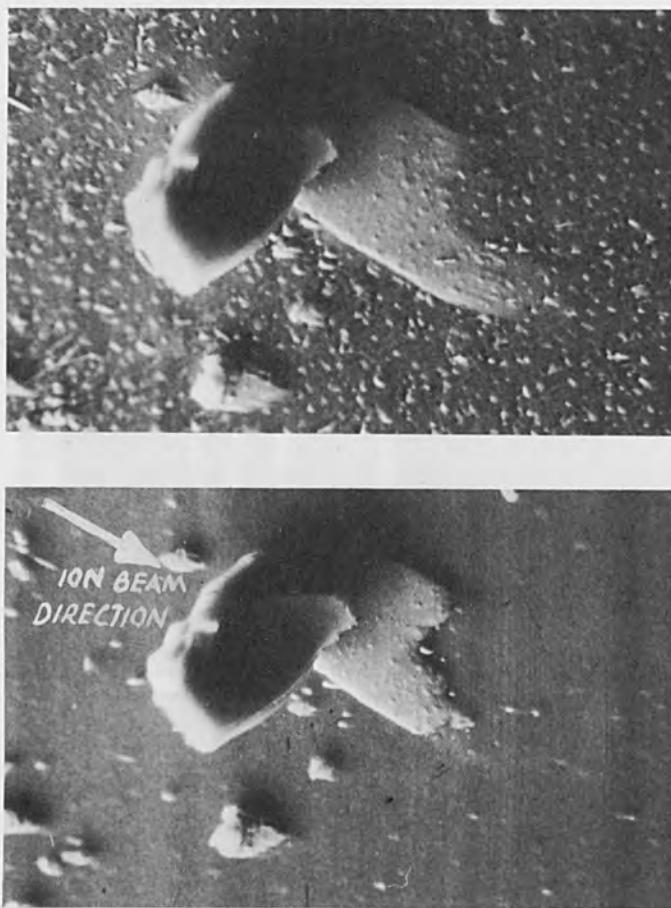


Fig. 2b. Argon ion bombardment of a gold film on a glass substrate.

Figure 4 shows an array of pointed cones fabricated on the surface of a single crystal of tungsten. They result from the shielding effect of single spots of contamination placed on the surface with the electron beam. It may be possible with this technique, if the cones were taller, to make a field emission cathode. By using a single crystal as the base material it would then be easy to control the crystallographic orientation of the tips and therefore their preferred emission directions.



CONCLUSION

The technique of sputtering through a mask of contamination has proved itself readily applicable to machining on a microscopic scale. A particular advantage when conductors are being fabricated is that they can be made taller than they are wide. Therefore the lateral packing for a given conductivity can be high. The vertical

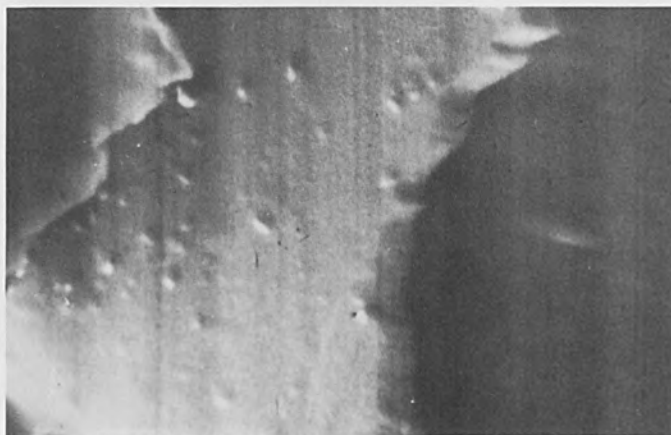


Fig. 3. A cross, micromachined by argon ion bombardment of a copper film.

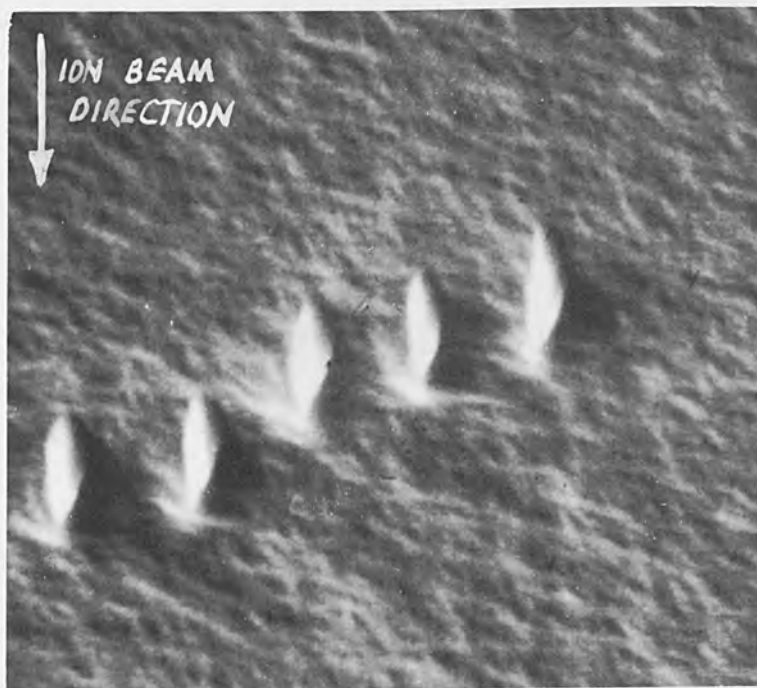


Fig. 4. Pointed cones micromachined on a crystal of tungsten.

extension of the conductors is not likely to be of consequence in miniature circuitry applications as the substrate needs to be of macroscopic thickness to be self-supporting. Another application which would lend itself to this process is the recording of information in a permanent or erasible form. Generally the technique would be made more powerful still if a form of contamination was found with an even slower sputtering rate than that already tried. All these possibilities are being further investigated.

This work forms part of a continuing program of research into and development and applications of the scanning electron microscope which was initiated in the Engineering Laboratory at Cambridge University by Professor C. W. Oatley in 1948. Thanks are due to the atomic Weapons Research Establishment, Aldermaston, for

providing the grant under which this particular work was carried out.

SOME OBSERVATION ON THE FORMATION OF TRACKS IN THIN, SUPPORTED METAL FILMS PRODUCED BY AN ELECTRON BEAM

INTRODUCTION

Considerable interest has arisen over the past decade in the use of electron beams for operating on materials. This follows the early work on electron guns and focusing systems by Stigerwald (1960), and their subsequent development (Schleich and others) of electron-beam machines. At the same time the many and diverse possible uses of electron beams have become the focus of interest and have been discussed in detail (e.g., Shoulders and Peters, 1961).

The recent advent of "micro miniaturization" would appear to be a particularly suitable field for electron-beam operation and as a result the building of electron-beam machines has multiplied over the last three years. However, the use of these machines is still mainly confined to the laboratory because their design is not sufficiently advanced to incorporate accurate control of the various functions, and also because the machining operation is only just beginning to be understood. In the meantime, rapid strides have been made in the development of competitive techniques based on photochemical methods (etchineering) and spark machining. These are inherently less sophisticated than the electron-beam techniques and have a more widespread industrial use. It is felt that the industrial application of electron-beam micro-engineering is only likely to succeed if it can be shown

*Vickers Research, Ltd., Sunning Hill, Ascot, Berkshire, England.

that this technique can perform required tasks either better or more economically, or can perform tasks which cannot be carried out in any other way.

A particular application which would appear suitable is the cutting of delineation tracks in thin, supported metal films. The aim at present in the manufacture of thin-film resistor networks is to steer away from the use of deposition masks; the shaping of circuits using electron-beam cutting would appear an important application, particularly for refractory metals which present some difficulty in (wet) chemical processing. Another application which seems promising, and which is being examined in a number of laboratories, is the formation of active devices by very localized heating and alloying of materials, induced by a short pulse of electrons. An example is the formation of tunnel diodes in GaAs.

The experiments described here are concerned with the manner of formation of tracks in thin metal films of the order of 1000 Å, supported on a substrate of thermally poorly conducting material such as silica or glass.

APPARATUS

An experimental two-stage electron-beam machine was developed and is shown schematically in Fig. 1. The design theory of this machine has been adequately described (Einstein et al., 1963, El Kareh, 1963). Briefly, the machine which operates up to 60 kV produces electron spots from 2 to 70 μ in diameter containing 0.7 to 10 watts power. The specimen can be traversed mechanically and the spot also deflected magnetically. Operation can be viewed directly with an internal microscope having $\times 125$ magnification. Several possible combinations of lens excitations leading to different size spots are shown in Fig. 2. These combinations range from the use of either the upper or lower lens singly, to combinations of the two lenses in which intermediate images (real or virtual) are formed by the upper lens. In the present series of experiments only two spot sizes were employed: 2 μ diameter using arrangement (a) and 15 μ diameter using arrangement (d).

The deflector coils were connected to a sweep circuit which caused the spot to be deflected at a chosen instant at a constant speed so as to form a single track. The sweep speed could be varied and as a rule a number of adjacent tracks were cut at successively increased speed into the sample.

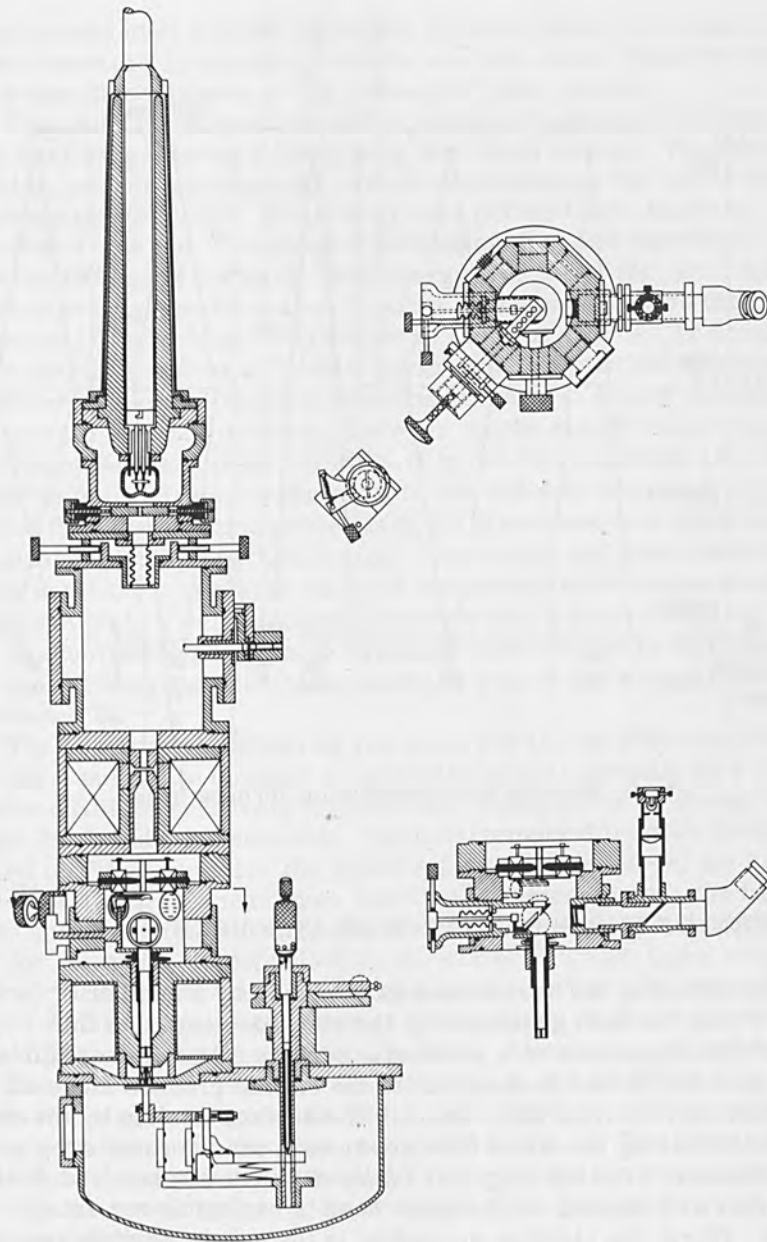


Fig. 1. Schematic of a two-stage electron beam machine developed for this study.

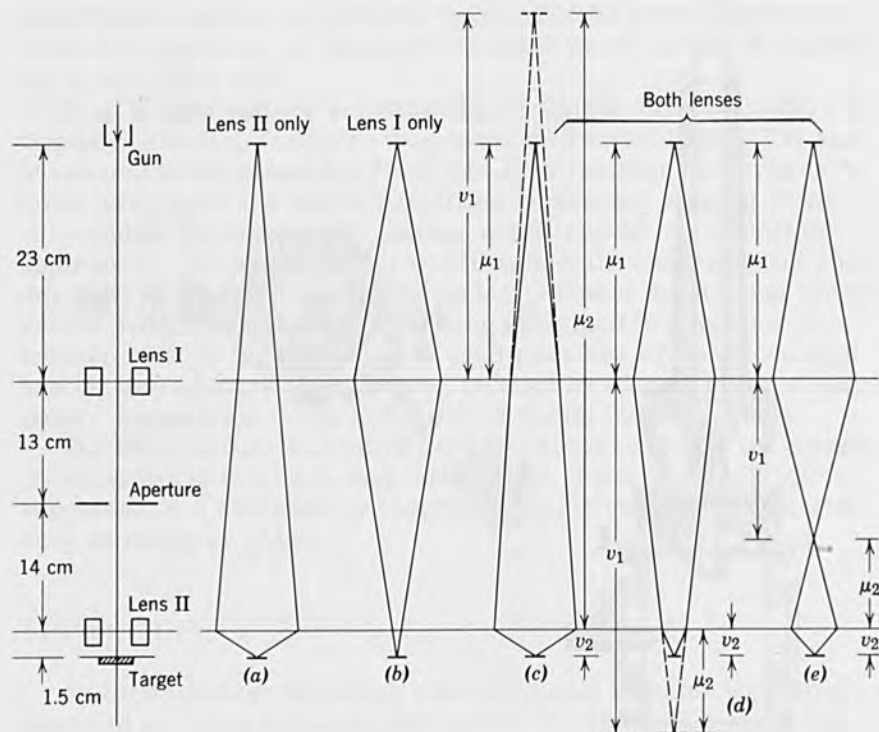


Fig. 2. Possible arrangements for forming image.

HEATING OF FILMS; NATURE OF HEAT SOURCE

The nature of the heat source is governed by a number of factors. First, the heat generated by the electron beam in a thin metal film supported on a substrate, such as silica, is negligible; the substrate in fact is essential to the cutting process since all the heat derives from this.¹ Second, the surface cooling by thermal conduction along the metal film produces a considerable drop in temperature, even for very thin films of only a few hundred Å; the heat lost by radiation on the other hand is negligible for small spots. Third, the electron spreading in the substrate disperses the

¹A good demonstration of this was the inability of a 60-kV beam to melt or cut a thin, unsupported 300 Å thick Al_2O_3 film, electron energy loss in this being only approximately 500 eV (Ω 50 μW power).

heat source over a fairly extensive volume; there is not much purpose therefore in operating with an electron-beam diameter much less than the diameter of the "effective" heat source.

Electrons penetrate into matter, their integrated path length (or total range) being a function of the initial energy. The paths tend to become increasingly curved or random as the electrons lose energy (Fig. 3a). The energy loss per unit path length is derived from the Thomason-Whiddington law and increases toward the end of the path. This energy loss along the path together with knowledge of the spatial location of the paths allows the computation of the volume distribution of the heat source. To compute this exactly requires a "Monte Carlo" type calculation involving knowledge of the electron-scattering processes; a very good approximation to path location, however, is the simple model suggested by Archard (1961) in which (Fig. 3b) the electrons are considered to penetrate the material to the diffusion center, from which they then radiate outwards in all directions to a spherical boundary to complete their range. This model has been used by Vine and Lauria (1963) to compute the steady-state temperature distribution in a semi-infinite substrate as a result of heating by an incident electron beam of Gaussian current density distribution. (A typical heat distribution is shown in Fig. 3f for a semi-infinite medium).

The conditions considered are those for (1) the substrate only, (2) for a substrate covered completely, and (3) partially by a thin surface film of relatively high thermal conductivity as shown in Figs. 3c, d, and e, respectively. The axial temperature distribution is given in Fig. 4, and, for the silica substrate, rises from the surface downward to a maximum near the diffusion center; the figure also shows the temperature distribution for several thicknesses of, for example, tin deposited on the surface. These films reduce the surface temperature by a considerable amount (for example, a 350 Å film reduces the surface temperature at the center to two-fifths of that without a film, a 700 Å film to one-eighth, etc.).

The surface temperature distributions in the presence of a complete surface coverage of tin 700 Å thick and also with a circular hole of radius r , in this film, are given in Fig. 5. The object of performing the latter computations is to estimate very approximately the temperature of the substrate when part of the metal film has disappeared.

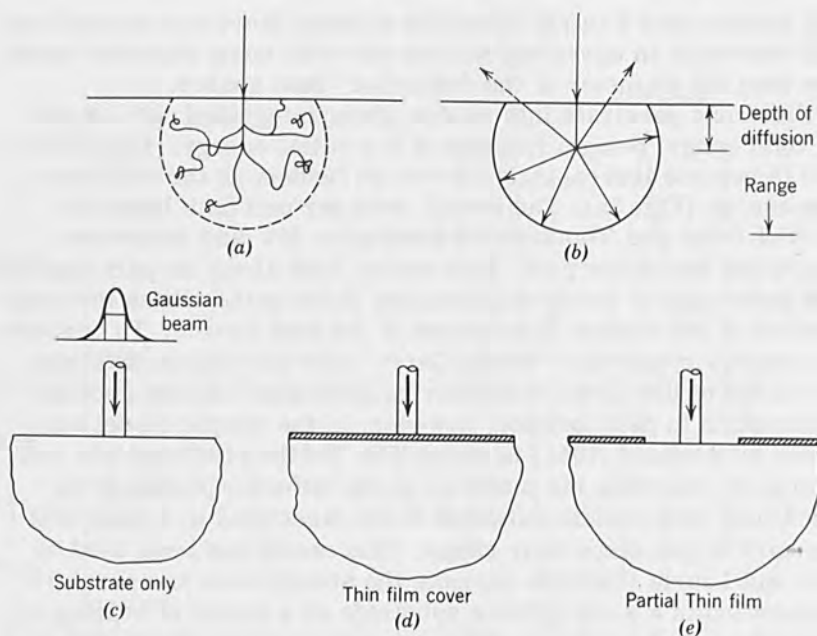


Fig. 3. Situations which can arise when beam impacts on target.

Traveling Beam

The transient thermal problem associated with the cutting action of a traveling beam is most complicated if electron penetration and the cooling effect of the surface film are to be taken into account. The simple cases considered by Pittaway (1961) cannot really be applied in the present instance; useful information, though only very approximate, can be obtained by considering the "thermal time constant." This is given by Oosterkamp (1948) and also by Vine (1963) as

$$r \Omega \frac{2.5 R^2 \rho s}{K}$$

and is the time to attain two-thirds of the steady-state temperature at a distance R from a point source of constant heat supply in an infinite medium of density ρ , specific heat s , and thermal conductivity K .

If reasonable assumptions are made concerning the effective location of the heat source in the medium, and the distance R from

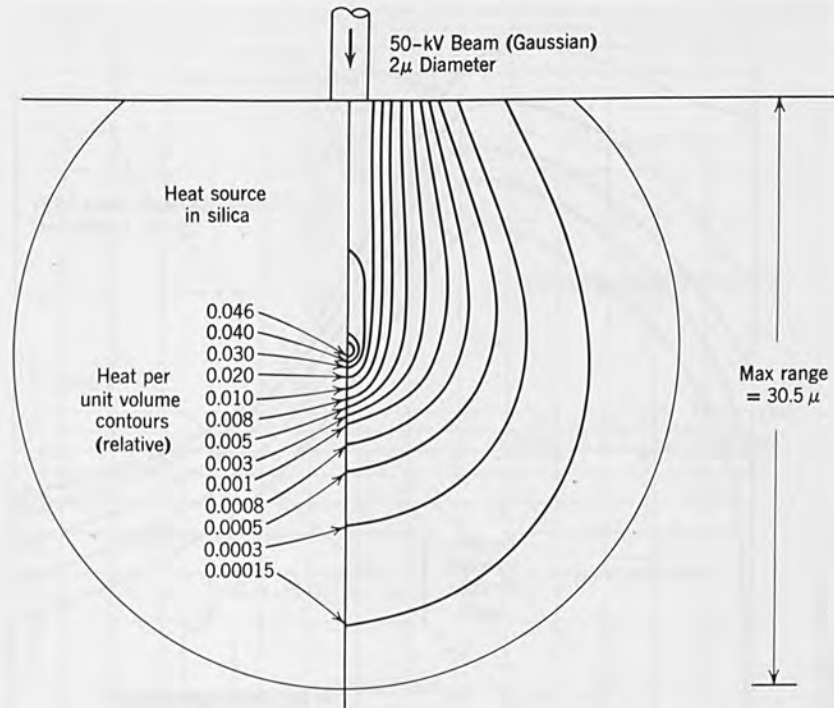


Fig. 3f. Situations which can arise when beam impacts on target.

this to the surface, and, if the effective time of irradiation under the moving spot is equated to the "time per spot width," then the calculated temperature rise is found to account well for observed effects. Thus, for instance, the maximum spot velocity for which "cuts" in silica can just be produced yield a calculated surface temperature of 1600° C which coincides with the softening temperature of silica.

TRACK CUTTING

The manner in which the metal film "disappears" from the surface in tracks ranging anything from 3 to some 100's of microns in width depends to some extent on the nature of the metal. In general it appears that metals with a low melting point (to circa 1500°C) are removed more readily than the high melting point metals.

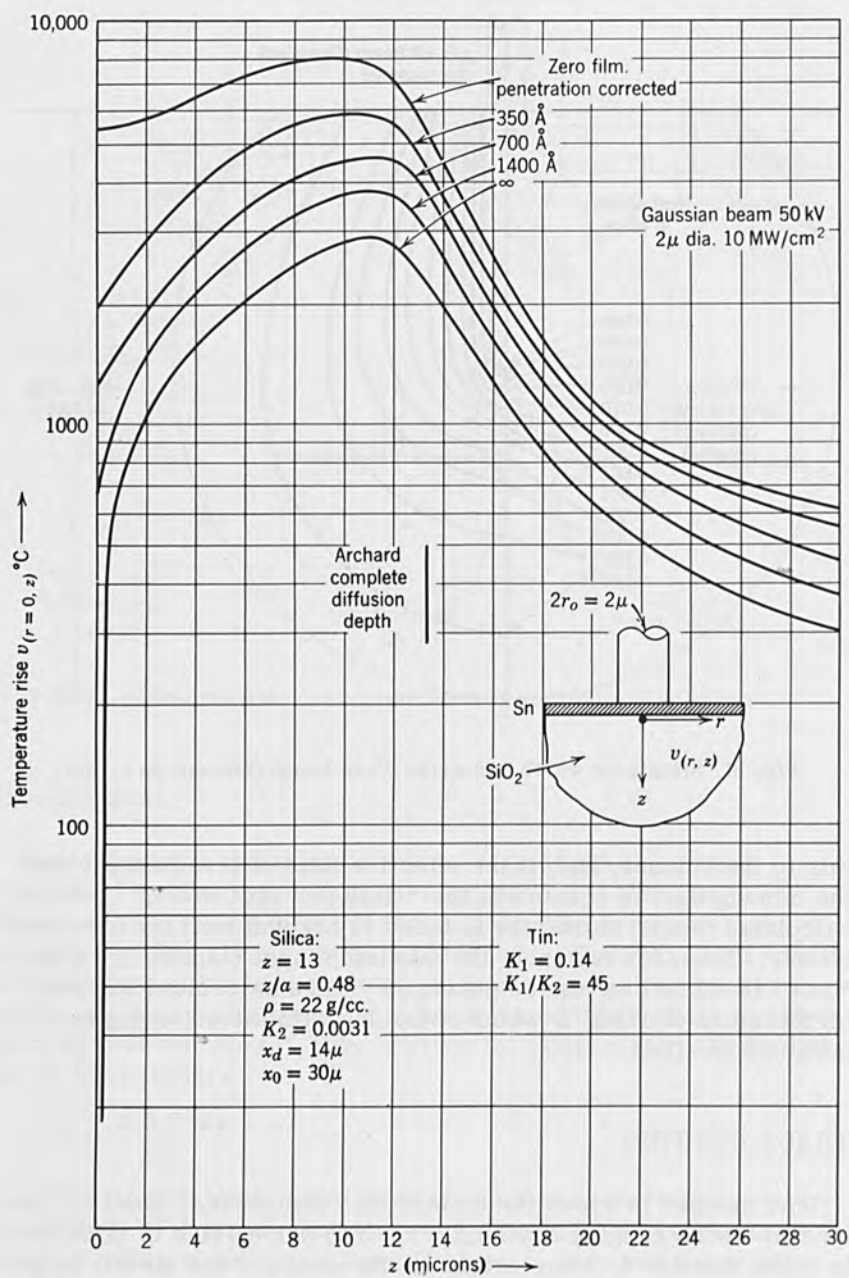


Fig. 4. Steady-state axial temperature distribution in the presence of a surface film of tin.

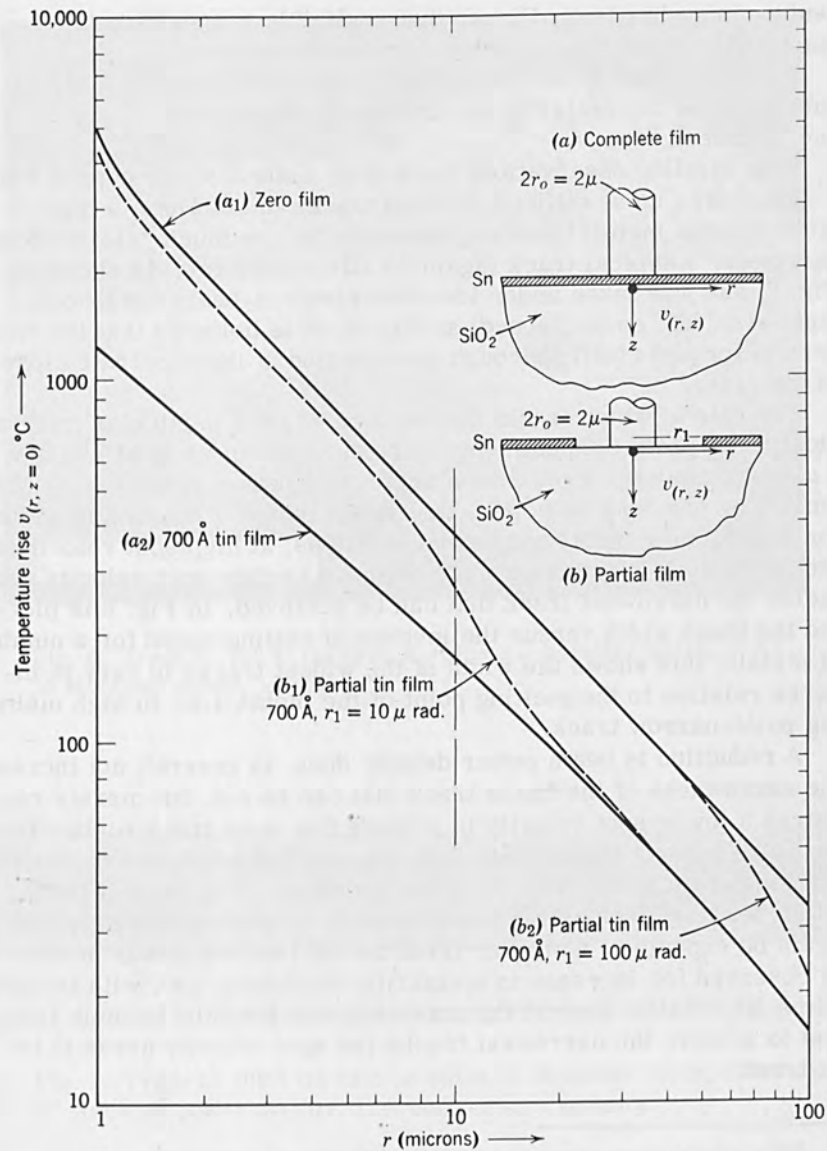


Fig. 5. Steady-state surface temperature distribution in the presence of a partial surface film of tin. Beam: 50 kV, 2μ diameter, 10 MW/cm^2 (Gaussian).

Evaporation only plays a minor part in the metal disappearance; the surface temperature, even at a short distance from the beam

center, is so low (e.g., Fig. 5) that negligible evaporation takes place in the short time in which tracks are formed.²

A typical track is shown in Fig. 6; the metal is tin on a silica substrate. In the center of the track is a groove some 5 μ wide and 20 μ deep.

Very similar observations have been made also for copper films.

It appears more difficult to form tracks in tantalum, a high melting point metal; however, these tracks are much "cleaner" and narrower. A typical track (again on silica substrate) is shown in Fig. 7, and was taken under identical electron-beam conditions (spot size, kV, power, speed) as Fig. 6. It is believed that the track here is formed chiefly through the eruption of the substrate close to the beam.

The rate at which tracks can be formed is of particular interest. Most of the initial experiments were carried out at 60 kV using a 2 μ spot containing a maximum achievable power density of 25 MW/cm². For low-spot velocities the tracks tend to a maximum width for a particular metal and beam conditions; at high spot velocities the beam fails to produce a cut beyond a certain spot velocity and yields the narrowest track that can be achieved. In Fig. 8 is plotted the track width versus the inverse of cutting speed for a number of metals; this shows the width of the widest tracks to vary in inverse relation to the melting point of the metal, i.e., in high melting point-narrow track.

A reduction in beam power density does, in general, not increase the narrowness of the finest track that can be cut, but merely requires a lower spot velocity to achieve the same track width. This would be expected since both spot velocity and beam power control the temperature distribution in the substrate. The largest track width becomes less as the power density is lowered which would again be expected. A similar trend as for lowered power density is observed for increase in metal film thickness, i.e., with thicker films (at constant power) the maximum track widths become less and to achieve the narrowest tracks the spot velocity needs to be reduced.

² The evaporation rate is given by the Dushman formula:

$$x = 5.85 \times 10^{-2} \frac{p}{\rho} \sqrt{M/T} \text{ cm/sec}$$

where p = vapor pressure in mm Hg

ρ = density in g/cc

M = molecular weight

T = °K

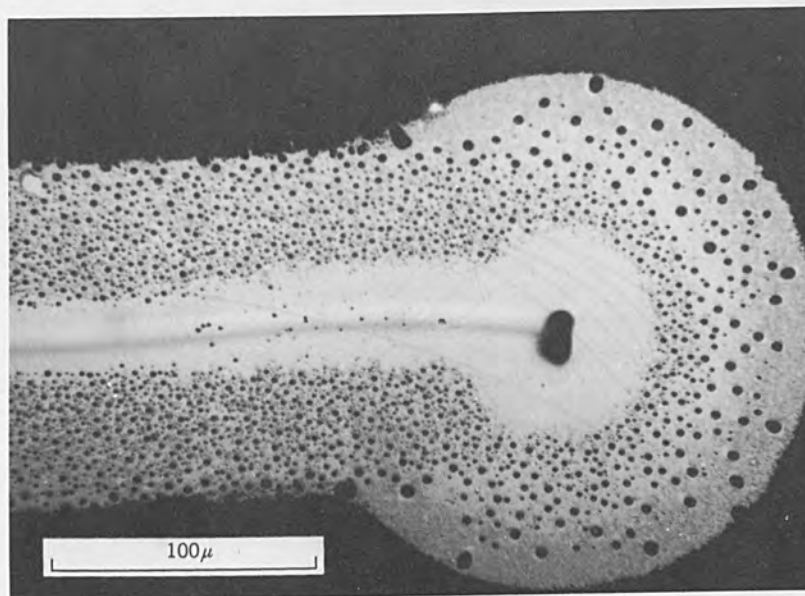


Fig. 6. Trade in 700 Å tin film of silica 2 μ diameter beam, 60 kV, 10 MW/cm² power density.

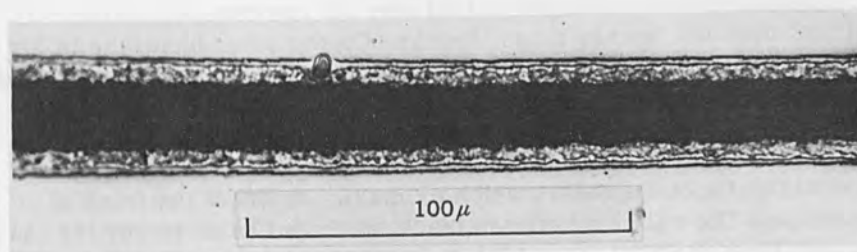


Fig. 7. Trade in 700 Å tin film on silica 2 μ diameter beam, 60 kV, 10 MW/cm² power density, 1.21 cm/sec spot velocity.

Larger Spot Size and Lower Voltage

As anticipated, an increase in the spot size to a diameter comparable with the "dimension" of the heat source (15 μ at 60 kV in

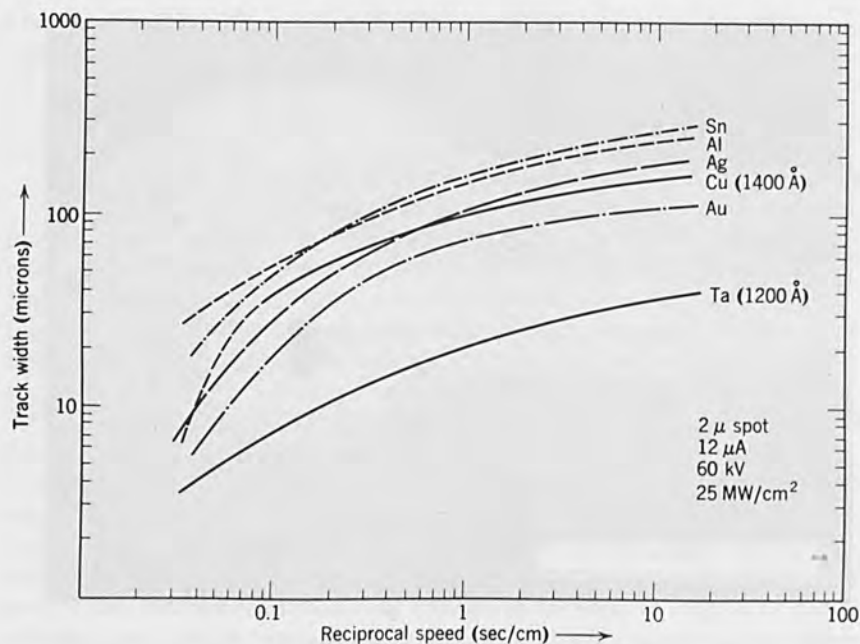


Fig. 8. Several metals 1700 Å thick on silica.

silica) does not lead to larger tracks. On the other hand, the increased available power enables tracks to be cut at a greater speed. Also note that the minimum track width (for example, in tantalum at 60 kV, see Fig. 9) can be less than the spot diameter; again this follows since it is the nature of the heat source in the substrate, and not the spot diameter, which primarily controls the track dimension. The relation between track width and beam power for tantalum films at 60 kV for a 15 μ diameter is shown in Fig. 9; track width varies as (beam power)^{0.85}.

At a lower beam voltage the electron range (x_0) in the substrate is less ($x_0 \propto V^2$) and the heat source contracts and moves closer to the surface. The available beam power in a given spot diameter also becomes less (power $\propto V^2$). Not many data have as yet been collected or calculations performed for lower voltages, except to establish experimentally the relation between power and track width for the same tantalum samples. These results are shown in Fig. 10 and again give the relation: track width $\propto$ (beam power)^{0.85}.

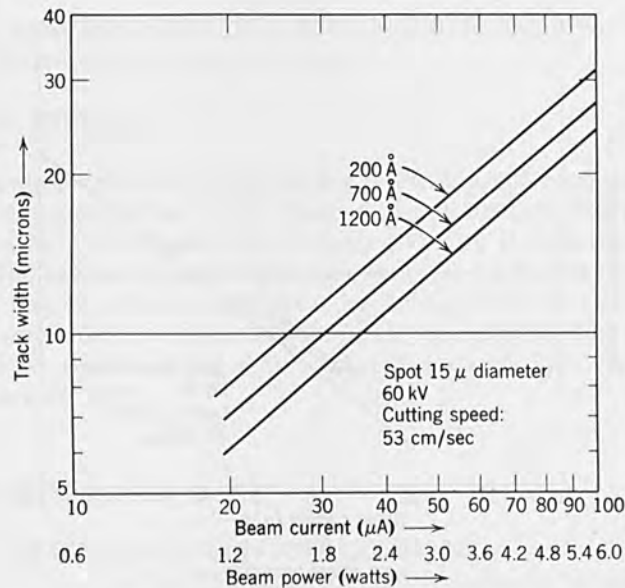


Fig. 9. Tantalum on silica: large spot.

The distinction between the results for different film thickness is not as marked here as with the 60-kV beam energy; the maximum cutting speed to achieve the minimum track width of approximately 15 μ is also lower.

It is of interest to consider the time required to cut a resistance maze of the type envisaged to form part of a "thin-film circuit," in, say, a film tantalum. Thus if the resistance is cut on a former 1/2 cm square (power dissipation approximately 0.2 watt) and the delineation tracks are separated by 100 μ , the total track length would be $\frac{5000}{100} \times 0.5 = 25$ cm and with an achievable cutting rate of 25 cm/sec would require 1 sec to cut. The total resistance of such a circuit would be $\rho_s \times b/a$ where ρ_s = sheet resistance (ohms/square) and b/a the aspect ratio of the resistance track. Thus total resistance at 10 Ω /square film resistance is $10 \times \frac{25}{100 \times 10^{-4}} = 25$ K Ω .

If the delineation tracks were 30 μ apart instead of 100 μ , the cutting time would be 3 seconds and the overall resistance 200 K Ω .

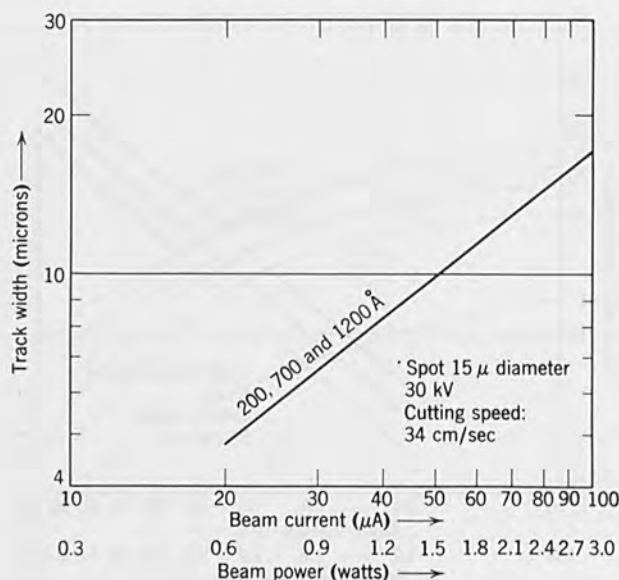


Fig. 10. Tantalum on silica: large spot.

CONCLUSIONS

This paper has described some of the initial work carried out with a simple two-stage experimental electron-beam machine. The nature of the heat source has been discussed and calculated temperature distributions show the considerable substrate cooling caused by a thin surface metal film.

In the formation of tracks, metal evaporation plays only a minor role; track cutting rates have been measured and it appears that the case of metal removal is linked inversely with the melting point of the metal. Only a few metals have so far been tried and of course much experimental conformation is still required. The effects of other factors such as film adhesion, substrate wetting, and deposition conditions in general on the film-removal process also require further study.

In general the beam diameter should be chosen to coincide with the dimension of the heat source. The ultimate choice of beam voltage will be determined by the power requirement and will, no doubt, depend on the specific task to be performed and the required speed.

It has been shown that it should be possible to cut resistive circuits in times of the order of 1 sec.

ACKNOWLEDGMENTS

The author gratefully acknowledges the helpful cooperation of many colleagues at the A.E.I. Research Laboratory, Harlow, and, in particular, the valuable assistance given by P.J. Simmons. Most of the computational work was carried out by Messrs. J. Vine and J.N. Lauria. Thanks are also due to the Directors of the A.E.I. Laboratories and to the Admiralty for permission to publish. The work was carried out at the A.E.I. Research Laboratory Harlow during 1963.

REFERENCES

- Archard, G.D., *J. Appl. Phys.* 32, 1505 (1961).
Crawford, C.K., in "Introduction to Electron Beam Technology," R. Bakish, Ed., Chapter 11, John Wiley and Sons, New York, 1962.
Dushman, S., "Scientific Foundation of Vacuum Technique," second edition, J.A. Lafferty, Ed., John Wiley and Sons, New York, 1962.
Einstein, P.A., *Proc. National Conference on Solid Circuits and Microminiaturization*, West Ham, London, June 1963.
Einstein, P.A., D.R. Harvey, and P.J. Simmons, *J. Sci. Instr.*, December 1963.
El Kareh, A.B., *RCA Rev.*, March 1963.
Pittaway, L.G., *Proc. 5th Annual Electron Beam Symposium*, Alloyd General Corp., Cambridge, Mass., 1961.
Schleich, F., *Proc. 4th International Congress on Electron Microscopy*, Springer Verlag, Berlin, 1960.
Shoulders, K.R., "Research in Microelectronics Using Electron Beam Activated Machining Techniques," Interim Scientific Report 1, Contract No. 2887(00), Stanford Research Inst., Menlo Park, Calif., Sept 1960. (Technical Services, Dept. of Commerce publication P.B. 171027.) U.S. Govt. Printing Office, Washington, D.C.
Shoulders, K.R., and D.W. Peters, "Application of Electron Beam Machining Techniques to Semiconductors," Ditto AD 259547.
Steigerwald, K.H., *Proc. 4th International Congress on Electron Microscopy*, Springer Verlag, Berlin, 1960.
Vine, J., and J.N. Lauria, private communications, 1963.

MICROFORMATION OF FILAMENTS

INTRODUCTION

When an electron beam impinges on a substrate in a demountable high-vacuum system a layer forms on the substrate in the region of the point of impact. This effect has been known for many years especially to electron microscopists who refer to the phenomenon as contamination.

Ennos (1953, 1954) studied contamination and concluded (1) that the mechanism was electron-beam polymerization of hydrocarbon molecules adsorbed to the substrate, (2) that replenishment of the adsorbed layer took place from the vapor phase, and (3) that the chief sources of the hydrocarbon molecules were diffusion pump oil, rubber gaskets, and greasy surfaces.

Christy (1960) studied contamination due to silicone diffusion pump oil and showed that the rate of growth of the polymerized film is dependent upon both the electron flux and the silicone molecule flux. When the latter is high enough to maintain a continuous adsorbed layer on the surface; the rate of growth depends only on the electron-current density.

This paper is concerned with certain contamination growths that have been observed in a scanning electron microscope.

THE SCANNING ELECTRON MICROSCOPE

The principle of the scanning electron microscope has been described by previous authors (e.g., O. C. Wells, Alloyd Conference, 1961) and will be only briefly described. The electron optical system focuses an electron probe onto the specimen. The probe

*Engineering Laboratory, Cambridge University, England.

is scanned in a rectangular raster and the electrons emerging from the specimen are used to build up a picture point by point on a C.R.T. screen. The magnification is given by the ratio of the scan amplitude on the C.R.T. screen to that on the specimen and the resolution largely depends on the probe width. The microscope normally works in the reflection mode and the signal is then largely derived from the low-energy secondary electrons. However, when measuring probe width d and probe current i , it is more convenient to work in transmission using an opaque edge as a test specimen and an earthed, metalized, scintillator, placed behind the specimen, as an electron collector. In all the micrographs shown the specimen was a silver grid with 1500 meshes per inch. The microscope used was developed at the Cambridge University Engineering Laboratory, and a sectional view of the column is shown in Fig. 1. With this column it was possible to obtain $d = 80$ atomic units (A.U.), $i = 4 \times 10^{-13}$ amp at $V = 15$ kV, and $d = 50$ Å.U. with $i = 8 \times 10^{-13}$ amp at $V = 30$ kV. The last set of figures represents a current density of 4 amp/cm² but the total power input is only 2.4×10^{-11} W.

CONTAMINATION EFFECTS

When the probe was focused onto a grid edge and moved slowly away from the edge, an unsupported filament of contamination formed in the path of the beam (Pease, 1964). The original vacuum was far from clean and had an overall pressure of 3×10^{-5} torr (measured on a Bayard Alpert gauge).

Such a filament is shown in Fig. 2. Fig. 2a shows the original grid edge; the mottling in the clear region is due to shot noise on the incident beam. In Fig. 2b, two portions of the grid have been joined and some attempts made to form the letter 'P'; the growth, however, tended to be unstable. In Fig. 2c V has been increased to 30 kV and the relative absence of shot noise due to the increased i is evident; the contamination is also more transparent to the 30 kV electrons. In Fig. 2d the letter 'P' has been completed. The small growth, arrowed in Fig. 2b, is about 100 Å.U. wide and may be the smallest object yet deliberately formed in a known position.

The rates of growth of filaments can be estimated from the dimensions of a filament formed at a known probe velocity. Under the above vacuum conditions and with $V = 30$ kV, $i = 4 \times 10^{-13}$ amp, a probe moving at 300 A.U./sec formed a filament 150 A.U. thick and 200 A.U. deep (estimated from the transparency of the

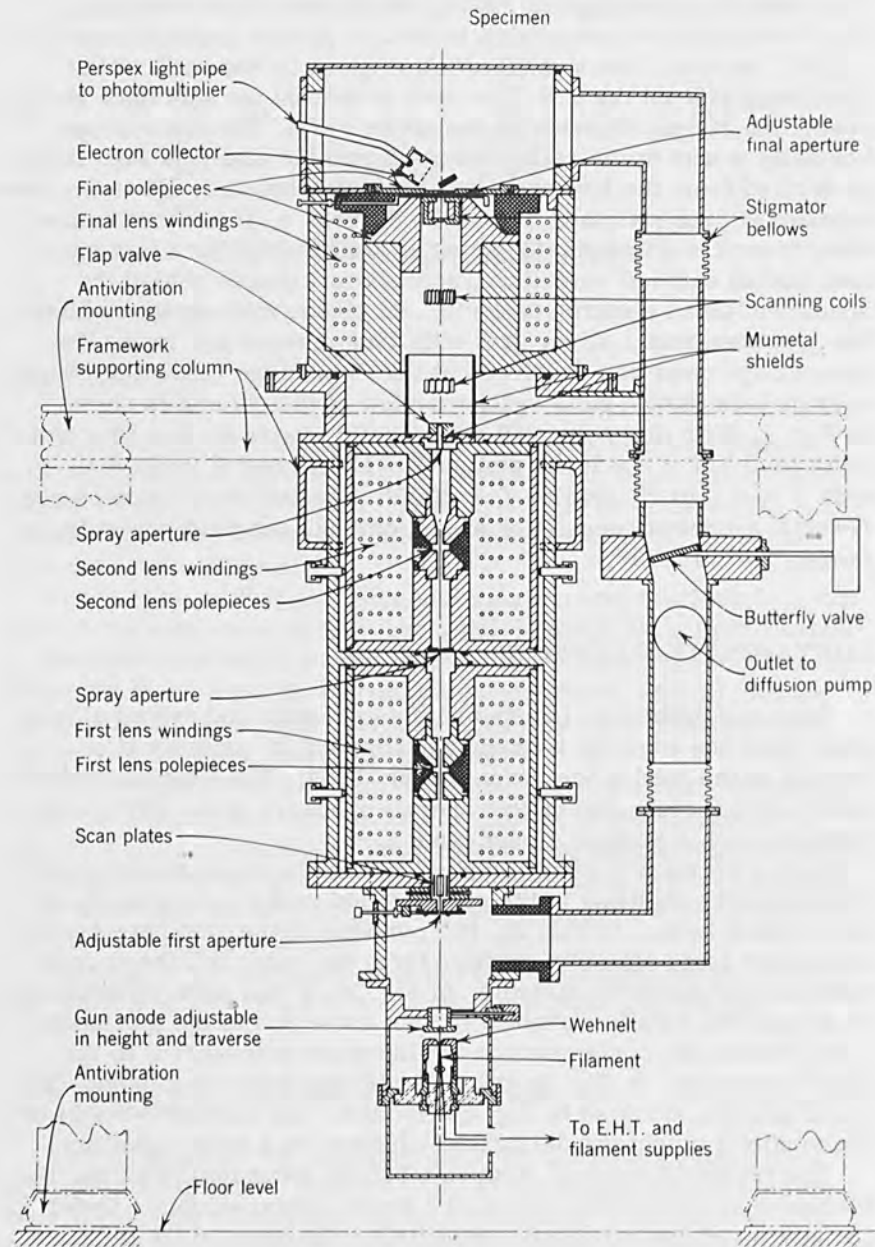


Fig. 1. Sectional view of scanning electron microscope number 5.

filament), hence the volume growth rate = $300 \times 200 \times 150 \times 10^{-24}$ cc/sec = 10^{-17} cc/sec; alternatively this could be expressed as 2.5×10^{-5} cc/coulomb.

Ennos (1953,1954), using a somewhat similar vacuum system, found that the rate of film growth increased to 2 A.U./sec at an electron current density, i /amp, of 0.1 a/cm², but did not increase appreciably as i /amp was increased further. His results represent a maximum growth rate of 2×10^{-20} cc/sec for $i = 4 \times 10^{-13}$ a or 2×10^{-7} cc/coulomb; this is considerably slower than the growth rate achieved in the present experiment. This might be due to (1) substrate heating due to the high overall power input in Ennos' apparatus, (2) polymerization being caused by secondary electrons emerging from many surfaces of the filament in the present system, and (3) a much dirtier vacuum than Ennos' being used at present.

THE EFFECT OF INTRODUCING GASES INTO THE SPECIMEN REGION

Later experiments were aimed at improving the growth rate and stability of the filaments. Bradley and Hammes (1963) measured the growth rates for polymer films formed in a glow discharge in atmospheres of a wide variety of monomers. Much higher growth rates were reported than for electron-beam contamination; typical values range from 10^{-3} gm/coulomb for acetylene to 5×10^{-3} gms/coulomb for acrylonitrile. A selection of these were used to form filaments in the scanning electron microscope.

First the overall vacuum in the microscope was cleaned up and reduced to 2×10^{-6} torr (nominal) and the growth rates of the contamination filaments was thereby reduced by a factor of 3 to 8×10^{-6} cc/coulomb. The monomer molecules were then admitted to the specimen via a needle valve and capillary tube whose orifice was about 1 cm from the specimen. A Bayard Alpert gauge was placed some 10 cms further away.

The first set of experiments was carried out with the following monomers: butadiene, acetylene, anilene, and acrylonitrile and with $i = 8 \times 10^{-13}$ amp. In no case was the growth rate of filaments significantly increased on admitting the gases to give an overall pressure of 5×10^{-6} torr. With acetylene admitted a filament of aspect ratio 1:20 and width 500 Å.U. was grown which suggested an increased stability but, on viewing, the filament buckled (Fig. 3a)

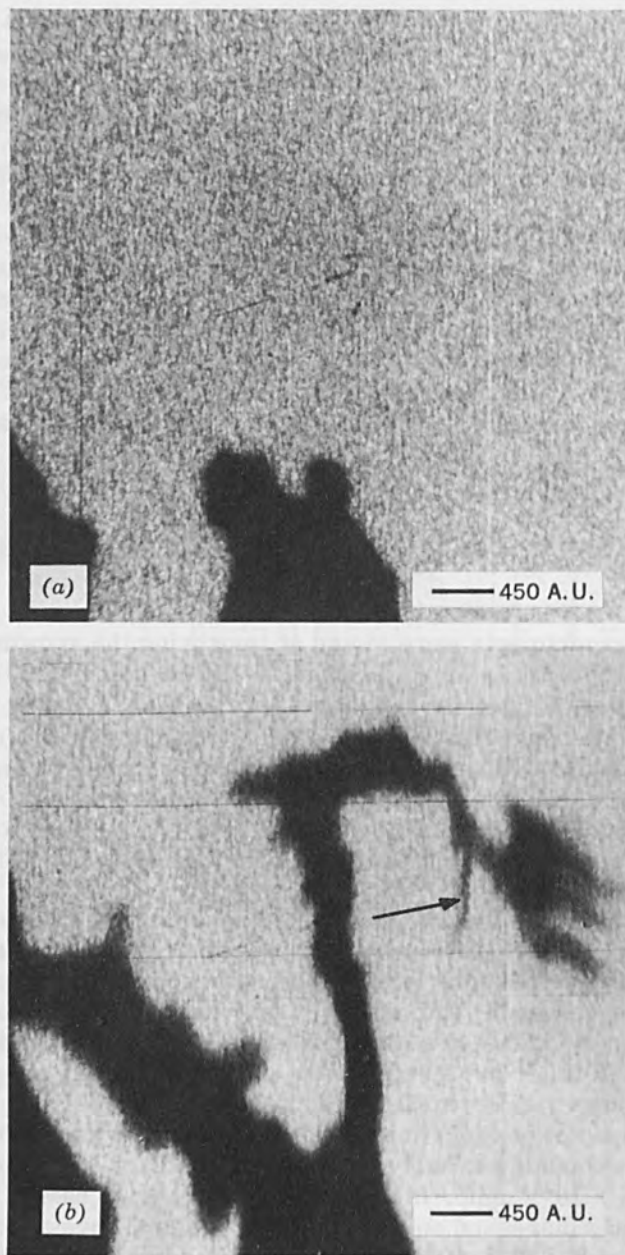
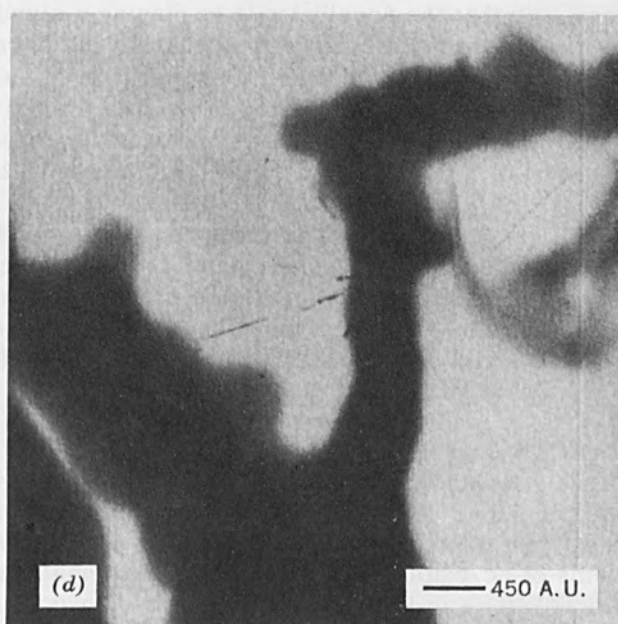
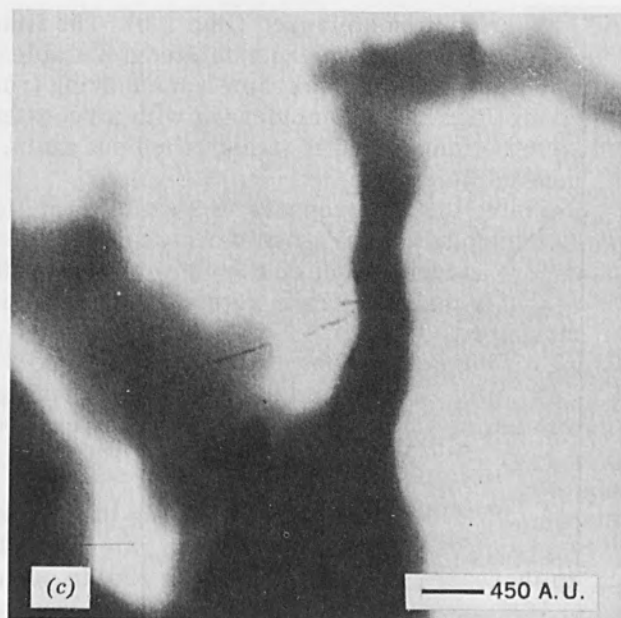


Fig. 2. Contamination formed filaments grown in the path of the beam. (a) 450 A.U. (b) 400 A.U. (c) 450 A.U. (d) 400 A.U.



and after further irradiation collapsed (Fig. 3 b). The filament grown in an acrylonitrile atmosphere exhibited a bistable characteristic; when viewed with a single frame scan moving from right to left it curled up (Fig. 4a), but on viewing with a repeated frame scan moving from bottom to top, it straightened out again. This process was repeatable.

Neither increasing the gas pressure to 2×10^{-5} torr (ion gauge reading), nor the introduction of a liquid air-cooled baffle into the specimen chamber, had any effect on the growth rates of the filaments. This suggests that the growth rate was solely limited by the electron current i .

By switching off the second lens, i was increased to 3×10^{-10} amp and d to 700 A.U. and the growth rates of filaments grown in atmospheres containing (1) no additional gases, (2) acrylonitrile, and (3) aniline were increased considerably and some are shown in Fig. 5.

Filament A (width 2500 Å.) was grown during the admission of acrylonitrile and represents a volume growth rate of 2×10^{-16} cc/sec, i.e., an increase of 20X; although the growth per coulomb has reduced to 10^{-6} cc/coulomb. The acrylonitrile was cut off and filament B formed. The growth rate was reduced to 10^{-16} cc/sec, suggesting that the acrylonitrile is playing a significant part. It was also found that at this higher current the introduction of a liquid air-cooled baffle reduced the growth rate of the filaments as well as reducing the indicated pressure, with no gases admitted, to 10^{-6} torr. Filament C was grown under these conditions and the growth rate, for the same i , is now only 2.4×10^{-17} cc/sec.

Filament D was grown under the same conditions but with air being admitted to the specimen. The rate of growth is unchanged which is surprising in view of Pashley's results (1962), in which the admission of air into a comparable vacuum system resulted in a reduced rate of contamination. This might again be explained by the low overall power input, hence the small temperature rise, of the specimen in the present experiment.

Acrylonitrile was admitted to an indicated pressure of 10^{-5} torr during the formation another filament (not shown). The growth rate has now risen to 4×10^{-17} cc/sec which, although just significant, indicates that a given pressure of acrylonitrile is much less efficient (in terms of cc/coulomb of polymerized material) than the residual hydrocarbons in the normal microscope vacuum. This suggests that the hydrocarbon molecules have a much higher "sticking coefficient"; it might also explain why the much higher

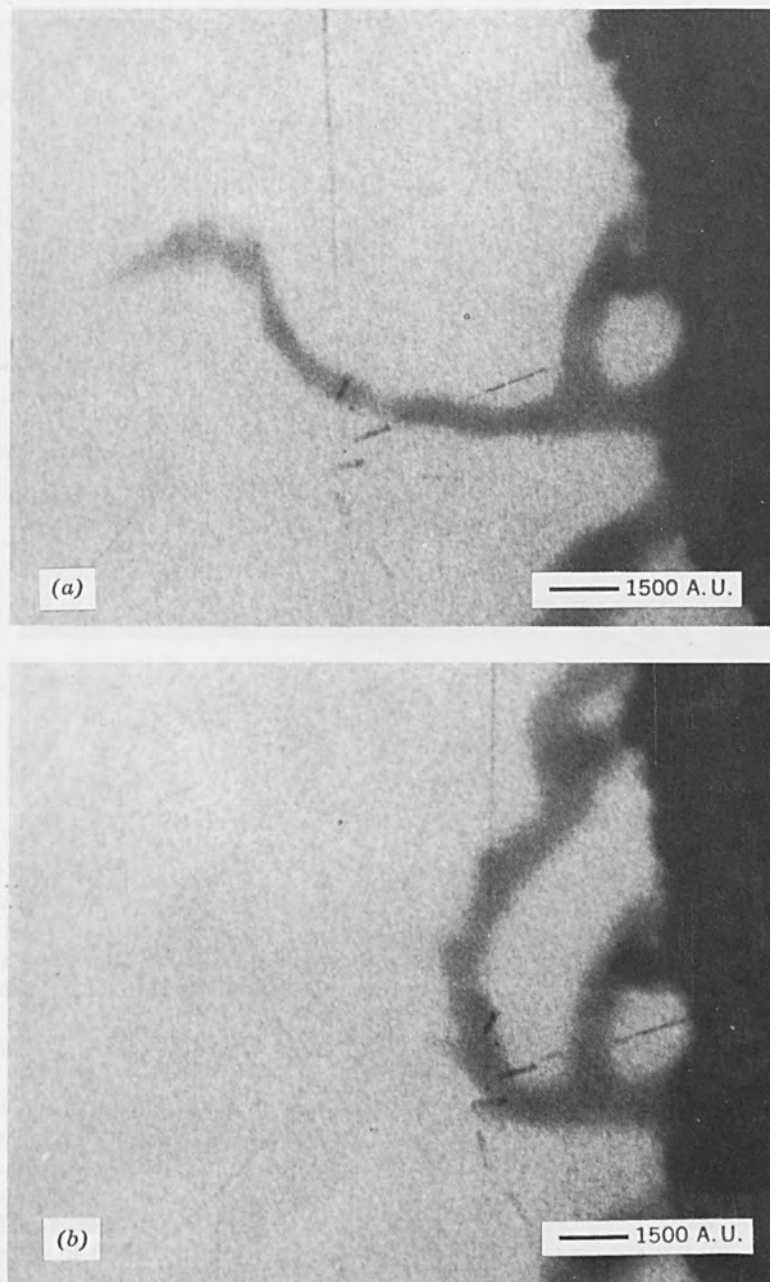


Fig. 3. Filament grown with acetylene in the system.

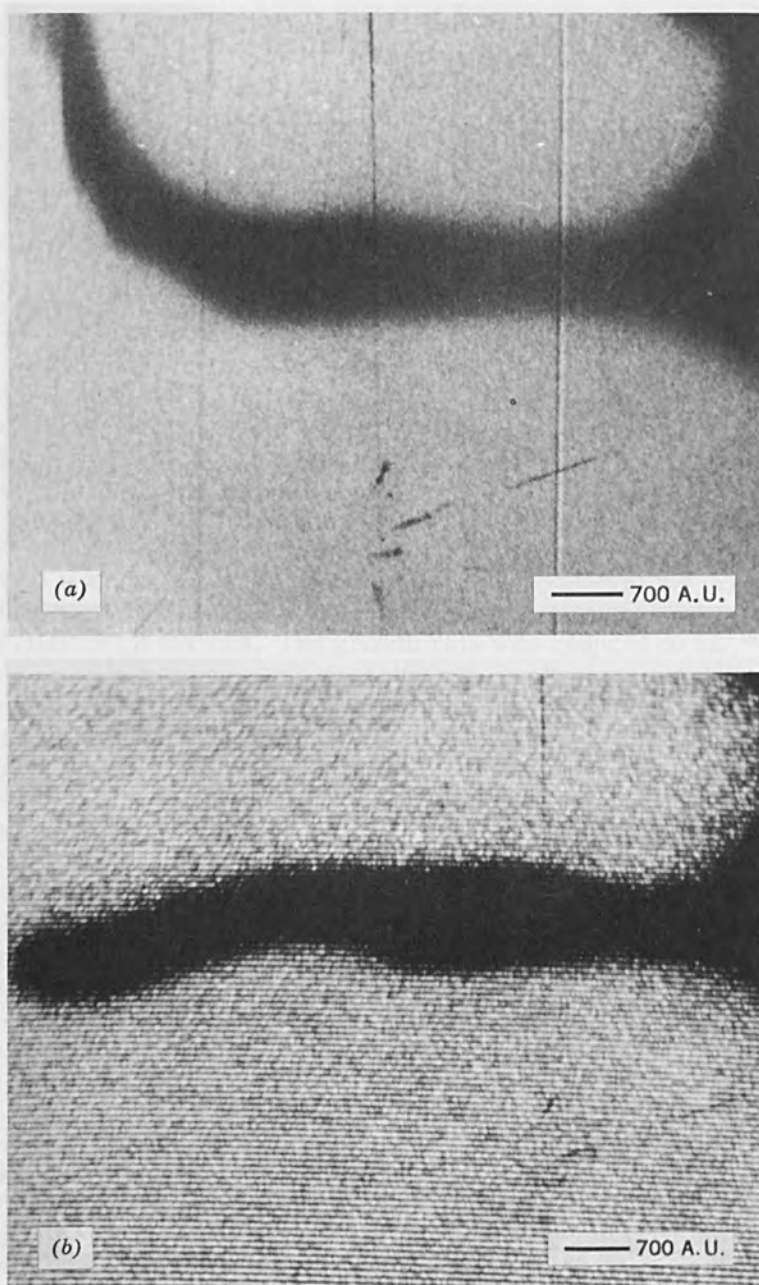


Fig. 4. Filament grown in acrylonitrile containing atmosphere.

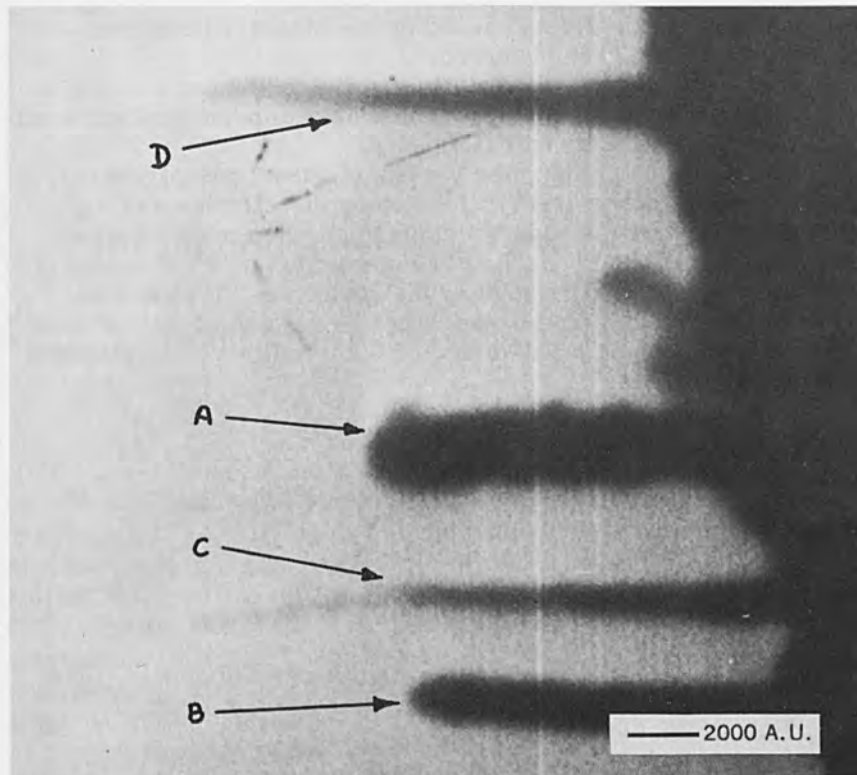


Fig. 5. Filaments grown under current of 3×10^{-10} amp.

rates of polymerization were observed in the glow discharge apparatus of Bradley and Hammes who used gas pressures of about 1 mm Hg but current densities of only a few mA/cm². The admission of aniline to the specimen chamber made no significant difference to the growth rate of filaments.

CONCLUSIONS AND FUTURE WORK

It has been shown possible to grow unsupported filaments in free space using a fine electron probe to polymerize low-pressure gas. The filaments can be as small as 100 Å wide and with a typical aspect ratio of 1:10.

The rate of growth of contamination (forming the filaments) has been up to one hundred times faster than has previously been reported for comparable systems.

Larger filaments, of about 0.2 μ diameter, have also been grown using probes with greater current. These filaments tend to be more

stable and no undue effects caused by the increased electron - beam heating have been observed.

Preliminary experiments on the growth of filaments during the admission of various monomers have failed to promote significantly faster growth or stronger filaments.

Future work will extend the variety of gases, gas pressures, and electron currents used. If it appears that filaments of high-electrical conductivity σ are being formed, then measurements of σ will be made by connecting two electrodes with a filament of known dimensions and measuring the resistance. It should be pointed out, however, that even if σ can be as high as $10^{-3} \Omega^{-1}\text{cm}^{-1}$, a filament of length 1μ and diameter 0.1μ will have a resistance of $10^9 \Omega$.

REFERENCES

- Bradley, A.J., and J.P. Hammes, *J. Electrochem. Soc.* **110**, 15, 1963.
Christy, R.W., *J. Appl. Phys.* **9**, 1681 1960.
Ennos, A.E., *Brit J. Appl. Phys.*, **4**, 101, 1953.
Ennos, A.E., *Brit. J. Appl. Phys.*, **5**, 27, 1954.
Pashley, D.W., *Phil. Mag.*, 1962.
Pease, R.F.W., Ph.D. dissertation, Cambridge University, 1964.
Wells, O.C., *Proc. Third Symp. on Electron Beam Tech.*, R. Bakish Ed. 1961, Boston, Mass.

THE FABRICATION OF ELECTRONIC COMPONENTS USING LOW-ENERGY ELECTRON BEAMS†

The ever-increasing volume and complexity of electronic gear in military installations and vehicles has provided impetus for the development of a variety of microelectronic fabrication techniques so that electronic circuitry may be manufactured with high reliability, lower weight and volume per unit function, and at low cost. The micromodule concept was a natural outcome of such requirements followed by an extension of the state-of-the-art such that complete circuit functions could be fabricated using monolithic semiconductor blocks.

A consideration of microcircuit requirements established that a general technique for fabricating passive networks without the use of masking techniques would be of general value by providing a higher component density, possibly higher manufacturing yields, and the generation of special low production volume circuits at lower cost. Such a technique would be even more valuable if it had a potentiality of being utilized to fabricate active components.

A general approach for fabricating microcircuits without the use of masking involves the utilization of an electron beam which may be precisely controlled with respect to beam diameter and position and which is capable of being swept over the surface of a substrate in order to trace out component areas. High-power-density electron beams may be utilized to thermally etch conductors, etc. This approach, however, does not yield the highest resolution

*Physics Department, Horizons Incorporated, Cleveland, Ohio.

†This research was supported by the Electronics Branch, Manufacturing Technology Division, Air Force Materials Laboratory, under Contract No. AF33 (600)-41705.

due to spreading in the processed area size because of the thermal conductivity of the substrate.

The decomposition of an adsorbed vapor in a vacuum system by an electron beam was first observed by electron microscopists. These investigators found that over a short period of time samples under investigation would become contaminated with a thin film of carbon whose formation was attributed to the electron-beam cross-linking of diffusion pump oil which had backstreamed into the system. During 1959, investigators at Horizons established that, by choosing the proper vapor to be introduced into the vacuum system, it was possible to generate both dielectric and resistive films utilizing the electron beam irradiation of a substrate. Continuous investigations resulted in the development of techniques for forming individual passive components and for combining the components to form specific circuit functions.

The fabrication of electronic components by this technique involves the interaction of a low-energy electron beam with molecules adsorbed on the surface of a substrate. The electron disrupts the valency electronic structure of the adsorbed molecule which may then result in a polymerization or condensation process or in a molecular decomposition.

The process is effected by introducing a vapor into an evacuated chamber so that the vapor pressure of the introduced gas is on the order of 10^{-4} torr. Upon introducing this vapor, it is found to condense onto all of the surfaces within the chamber that are below a certain critical temperature; the surface coverage generally being on the order of 1 monolayer. The surface is then swept by an electron beam. The action of the impinging electron beam involves the breaking of the bonding electrons or the removal of a bonding electron with the formation of an ion or free radical at the surface. This bond provides an adsorption site for another molecule and the process is repeated. In this manner a defect-free film is built up on a surface.

It might be anticipated that the film growth rate is dependent upon both the electron-beam current density and the rate at which molecules impinge upon the surface. Two limiting cases exist. The first involves an electron-beam current density much larger than the rate of molecular impingement so that the growth rate is limited by the molecular arrival rate. The second involves a molecular impingement rate much higher than the electron impingement rate so that the growth rate is controlled by the electron beam current density. As anticipated, it was found that the film deposition rate is proportional to the electron beam current density.

A detailed description of the growth rate dynamics has been worked out by Christy.¹

Figure 1 is a diagrammatic view of the mechanical arrangement of the vacuum system and electron gun. Mercury diffusion pumps and liquid nitrogen cold traps are employed in order to eliminate film contamination by backstreaming pump fluids. The work chamber may be pumped down to 5×10^{-5} torr in five minutes and reaches an ultimate pressure of 2×10^{-7} torr in three hours. The top of the chamber is removable for easy access to the substrate table. Provisions for admitting different vapors are provided through the use of servo-controlled variable leaks which maintain a predetermined pressure. An evaporator with automatic control for providing

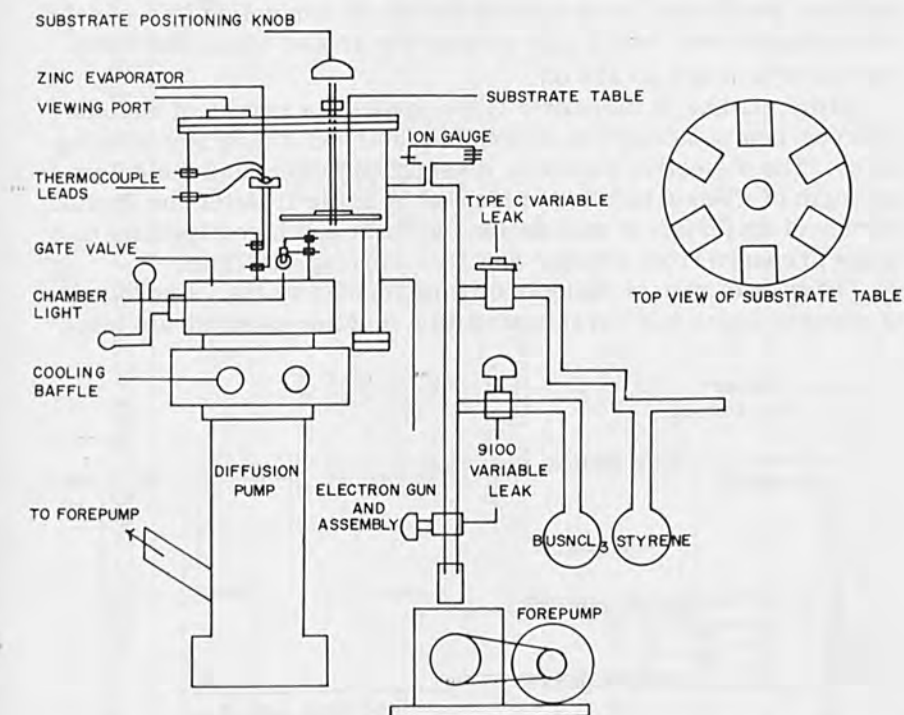


Fig. 1. A diagrammatic view of the mechanical arrangement of the vacuum system and electron gun.

¹R. W. Christy, J. Appl. Phys. 31, 1680 (1960).

zinc vapor is also contained in the chamber. The substrates are positioned in the substrate table above the electron gun.

The electron gun shown in Fig. 2 is a modification of the SE 200 V Vidicon mount made by Superior Electronics. These modifications include: (1) enlarging the Grid No. 1 aperture to 8 mils; (2) replacing the oxide cathode with a tungsten hairpin filament; (3) placing a 1000 mesh screen over Grids 3 and 4; (4) adding a flat plate in the plane of the screen; and (5) adding an anode approximately 1/2 in. from the screen. This modified gun provides a 10-mil beam diameter with a maximum beam current density of 3 mA/cm². In addition, the energy of the electrons may be varied from 50 to 1600 eV. Electromagnetic deflection is employed with precision positioning and sweep electronics. The deflection of the electron beam over predetermined areas is controlled by a punch card programmer which also selects the proper vapor and turns the electron beam on and off.

Initial studies of dielectric films involved a survey of various hydrocarbon monomers to determine their suitability for forming films. The dielectric constant, dissipation factor, and dielectric strength of these films are presented in Table I. After the initial survey of materials it was decided to limit our investigations to films prepared from styrene and Dow Corning 510 fluid.

The growth rate of dielectric films formed by the cathodolysis of styrene vapor was investigated as a function of substrate tem-

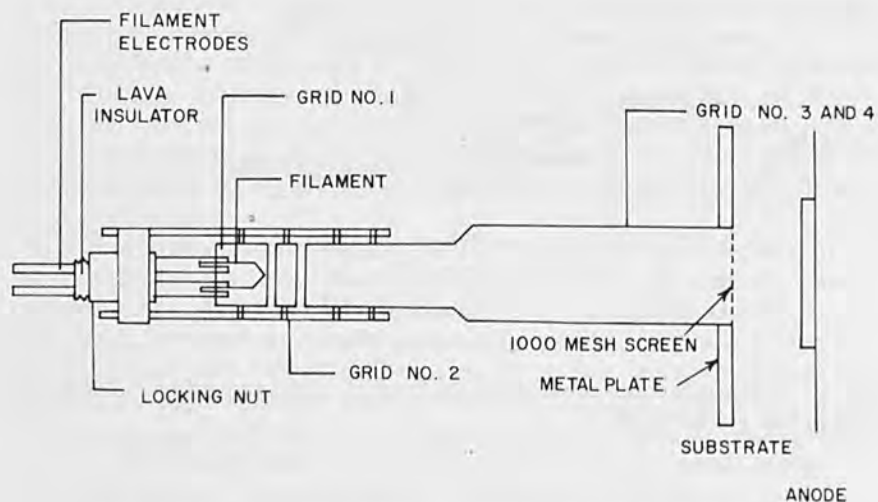


Fig. 2. Modified vidicon electron gun (reduced about 30%).

TABLE I
DIELECTRIC CHARACTERISTICS OF VARIOUS THIN FILMS

Film	$C(\mu f) / \text{cm}^2$	D	Thickness $\text{\AA}$	Breakdown Field (V/cm)
Styrene -				
Acrylonitrile	455×10^3	0.034	400	--
Nitrobenzene	392×10^3	0.093	700	2×10^6
Benzonitrile	520×10^3	0.037	700	1.7×10^6
Phenyl Isocyanate	640×10^3	0.027	475	2.5×10^6
Styrene	236×10^3	0.018	700	1.5×10^6
Acetylene	22×10^3	0.007	600	...
2-propyne-1-ol	All shorted	...	400	...
Methane	No film formed	...	...	...
Ethane	"	...	...	...
Methylmethacrylate *	14×10^3	...	...	...
Dicyclopentadene *	20×10^3	...	...	...
Cyclohexanol *	13×10^3	...	...	...
Vinylacetate *	Shorted	...	...	...
Dow-Corning 510	150×10^3	0.04	600	18×10^6
Silicone Fluid				

*Early work in a different system.

perature which was varied between 243°K and 423°K, being held constant for any one film deposition. The results of this variation in temperature are shown in Fig. 3, where the total film thickness is plotted versus the reciprocal temperature in degrees Kelvin. Film thickness was determined with a microscope fitted with an interference objective. The slope of the curve represents an activation energy of 6.8 K calories per mole, indicative of van der Waals type bonding between the adsorbed molecule and the surface.

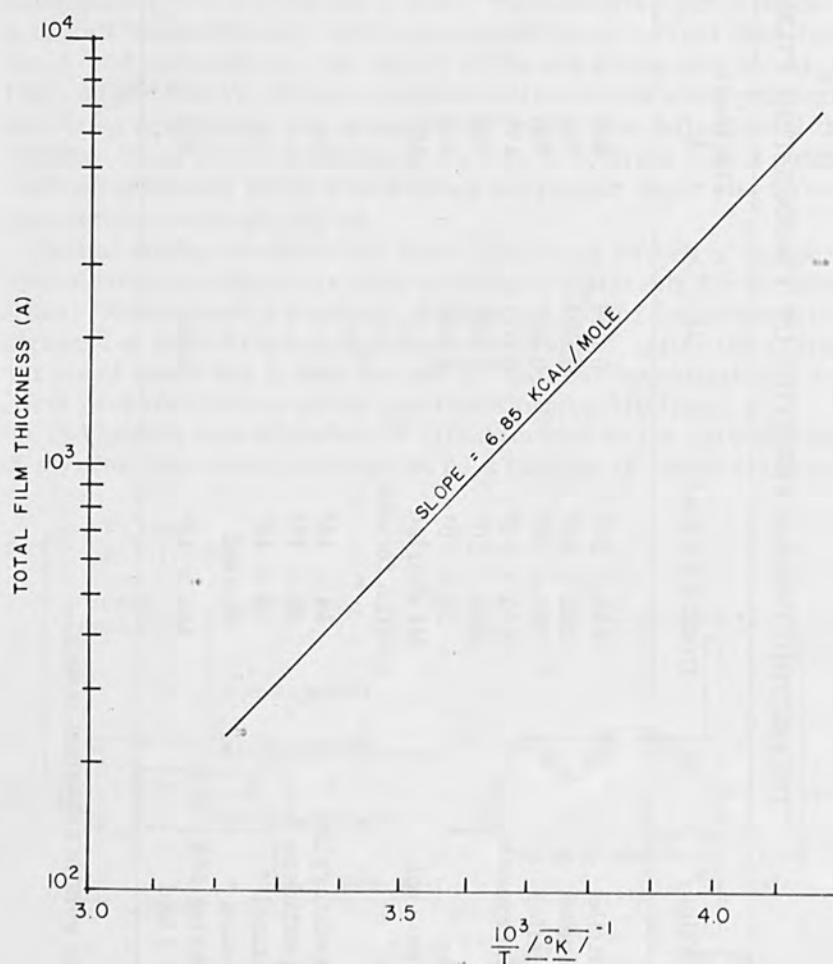


Fig. 3. Total thickness of deposited film after twenty minutes of a standard deposition at various temperatures.

The dielectric characteristics of these films were investigated as a function of frequency, film thickness, and substrate electrode.

Figure 4 is a plot of the capacitance and dissipation factor versus frequency. The dispersion at 40 kc is probably due to a reduction of the dipolar moment of the polymer film.

Measurements have shown that the dielectric constant is large compared to the bulk value of polystyrene. As a consequence, an investigation was carried out to determine the value of the dielectric constant of the film as a function of thickness. The results of this investigation are shown in Fig. 5. The plot of K/t (capacity) versus thickness indicates that the capacity is independent of thickness for film thicknesses up to 1000 Å. For thicknesses greater than 1000 Å the dielectric constant approaches the bulk value of 4.5.

Initially it was suspected that perhaps the material used for forming the electrodes, in this case aluminum, was contributing to this effect. A series of experiments were conducted to determine the effect of substrate material on the dielectric constant. The results of this investigation are tabulated in Table II. Here it

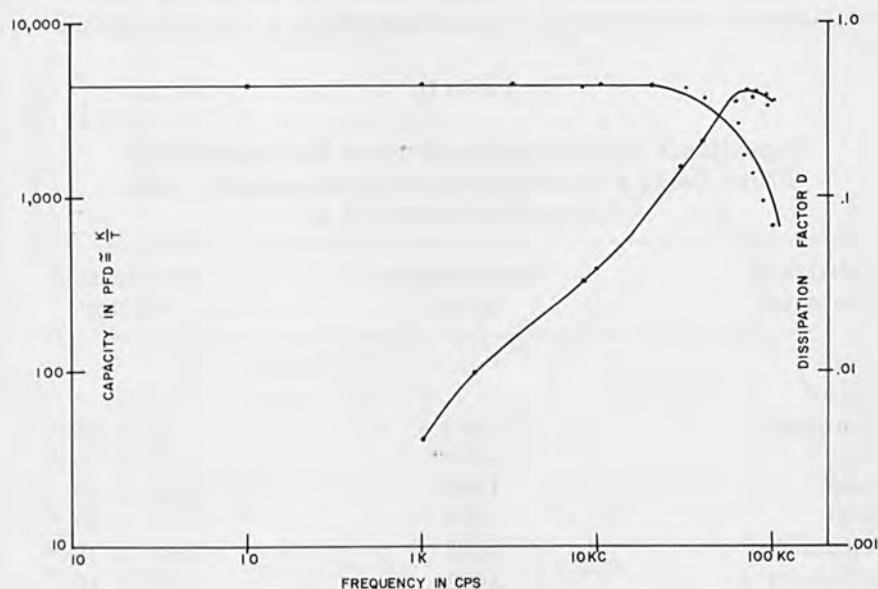


Fig. 4. Dispersion curve for a thin-film styrene dielectric capacitor.

can be seen that the electrode material did not significantly alter the dielectric constant of the thin films. However, the dissipation factor was affected.

A similar investigation was carried out using Dow Corning 510 as a monomer and it was found that both the dielectric constant and dissipation factor were independent of frequency to the maximum measuring frequency of 100 kc.

An investigation of electrode materials was also conducted with capacitors fabricated with Dow Corning 510 material. Again the dielectric constant and the dissipation were relatively independent of the electrode material but the incidence of pinholes in the dielectric was substantially higher when gold was used than when materials that were more easily oxidized were used.

It is apparent from these studies that the electron beam acts as a powerful polymerization agent. Indeed, from an examination of Table I, it can be observed that the action of an electron beam is to form polymer films from starting compounds which are not generally of the monomer class. Also, it appears that all of the polymer films formed are highly cross-linked, i.e., the dielectric film consists of one giant molecule. This is evidenced by the lack of thermoplastic nature of the dielectric films as well as their resistance to dissolution at room temperature in common labora-

Table II
Capacitance and Dissipation Factors for Polystyrene
Films Using Various Substrate Materials and a Gold
Counterelectrode at 1 kc

Substrate Material	Capacitance ($\mu\mu f$)	Dissipation Factor
Gold		Short
Silver		"
Aluminum	1400	3.30×10^{-2}
Tin	1458	0.27×10^{-2}
Lead	1965	1.95×10^{-2}
Zinc	1351	3.10×10^{-2}
Antimony *	3086	2.60×10^{-2}
Antimony †	1400	2.20×10^{-2}

*Capacitor formed with an antimony counterelectrode.

†Capacitor formed with a bismuth counterelectrode.

tory reagents, including strong acids, bases, and organic solvents.

Typical capacitors are fabricated of these polymer films at low substrate temperatures in a period of approximately 5 min. With a dielectric thickness in the range of 600 to 1000 Å a capacity per unit area of 1600 pF/sq mm is obtained with dissipation factors of 0.01 to 0.03 and a dielectric breakdown voltage between 8 and 12 V. The adherence of the dielectric film to the electrodes is excellent. In some cases some peeling is observed with films prepared at low substrate temperatures for film thicknesses of several thousand angstroms.

A large group of compounds were surveyed for use as resistive elements. Lowest resistance films were formed by the electron-beam decomposition of tungsten hexycarbonyl or germanium tetrani-tride. The organo-metallics and metal halides investigated did not

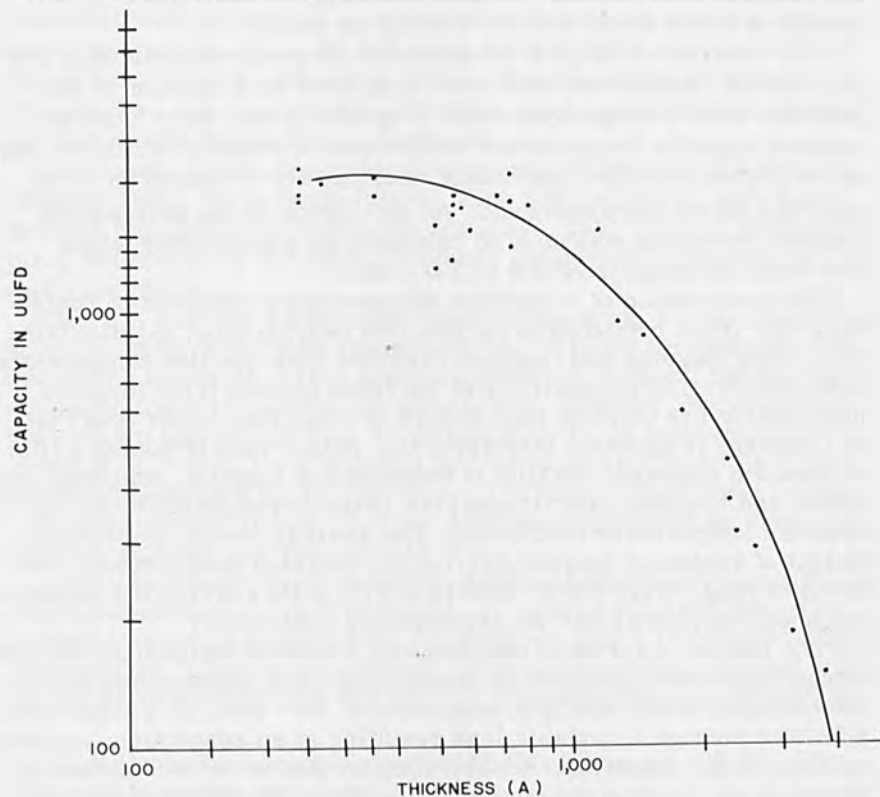


Fig. 5. K/t (capacity) vs. thickness for al-styrene-al capacitors (constant area of 0.64 mm^2).

produce satisfactory resistive films. These compounds, in general, do not decompose completely but form a metallo-organic polymer film. As a consequence, the resistivity of the films formed from the organo-metallics is on the order of $10^9 \Omega / \text{sq}$ or greater. In the case of halogen compounds it is felt that the reactive halogen radicals immediately recombined with either the original molecule or a neighboring molecule. Resistivities ranging from 10^7 to $10^{10} \Omega / \text{sq}$ were recorded for this class of compound.

One obvious requirement of a metal compound suitable for utilization with this process is that the compound have a vapor pressure of 10^{-4} torr or greater at room temperature. A further requirement is that the compound should be relatively inert chemically and that the by-products formed during the decomposition of the compound should also be relatively inert. Compounds containing halogen, double bonds, or reactive oxygen result in the formation of films which are not metallic in nature.

The resistances of film formed from tungsten hexycarbonyl and germanium tetranitride have been measured as a function of temperature over a range from -28°C to $+90^\circ \text{C}$ and were found to possess negative temperature coefficients of resistance. In the case of the germanium film, activation energies for conductivity from 0.25 to 0.39 eV were observed. An estimation of the germanium resistor thickness was used to calculate the conductivity which was found to range from 0.6 to $3 \Omega \cdot \text{cm}$.

The observation of a negative temperature coefficient of resistance for films prepared using tungsten hexycarbonyl is surprising since both tungsten and tungsten carbides have positive temperature coefficients. The conductivity of the films formed from tungsten hexycarbonyl is three to four orders of magnitude lower than that of tungsten. It appears, therefore, that rather than obtaining a film of metallic tungsten, the film is composed of tungsten, tungsten carbides, and tungsten oxycarbides; the latter is responsible for the negative temperature coefficient. The average temperature coefficient of resistors fabricated from the tungsten hexycarbonyl was found to range from 900 to 3000 ppm with little correlation between the sheet resistivity and the temperature coefficient.

The resistors formed from tungsten hexycarbonyl and germanium tetranitride were prepared by positioning clean glass substrates in the chamber which was then evacuated to 10^{-7} torr. The vapor was admitted through a variable leak resulting in an adsorption onto the surface of the substrate. Adsorbed molecules were decomposed by sweeping an electron beam over the surface. Resistances ranging from $130 \Omega / \text{sq}$ to $530 \text{ k}\Omega / \text{sq}$ have been manufactured using tungsten hexycarbonyl and resistors ranging from $1 \text{ M}\Omega / \text{sq}$ to $10 \text{ M}\Omega / \text{sq}$ have been fabricated using germanium tetranitride.

Conducting films have been fabricated by a low-vapor-pressure zinc evaporation onto a surface presensitized by electron bombardment in the presence of butyltin trichloride.

It is known that, at low-vapor pressures, the condensation of evaporated metals will not take place on a glass surface unless the glass is held below a critical temperature or there is present on the surface some substance which acts as a nucleating agent for the condensation.² Langmuir explains this as a condensation-evaporation phenomenon³ rather than one of reflection. Thus, below a certain vapor pressure, for substrate temperatures above the critical temperature, the impinging atom re-evaporates from the glass before other atoms are deposited adjacent to it. If the substrate is cooled below the critical temperature, or if the vapor pressure of the metal is raised above a certain critical level, other atoms are deposited adjacent to the first one. It required more work to remove a metal atom from another metal atom than to remove a single atom from the glass surface, so that once these groups are formed they act as nuclei for the growth of metal crystals on the glass surface. These views are supported by electron microscope studies carried out by Sennett, McLauchlan, and Scott.⁴

For most metals, the critical temperature is several hundred degrees above room temperature. Zinc and cadmium, both possessing low-adsorption energies for glass, have critical temperatures below 300° K and therefore were employed to form conducting thin-film paths.

Butyltin trichloride has been used in our work as the nucleating agent because of the high reproducibility of films formed with this nucleating material. Other metallo-organics or metal halides have also been found to function as sensitizers, however. Experiments have shown that a very thin, invisible layer of tin compound (on the order of 20 Å thick) decomposed by cathodolysis is sufficient to nucleate the formation of zinc films. Optimum conditions involve a 20-sec scanning time at 2 to 5 μ A beam current and an indicated BuSnCl_3 pressure of about 5×10^{-4} mm.

Zinc deposition onto the sensitized glass is effected by vaporizing chunk zinc contained in a carbon boat. Temperature is monitored by a thermocouple in contact with the massive zinc. The evaporation temperature is somewhat critical, best results being obtained at a temperature between 300 and 350° C. The deposition is difficult to

²L. Holland, *The Vacuum Deposition of Thin Films*, pp. 199-205, John Wiley and Sons, New York (1956).

³Irving Langmuir, *Proc. Natl. Acad. Sci.*, **3**, pp. 141-147 (1917).

⁴R. S. Sennett, T. A. McLauchlan, and G. D. Scott, *Can. J. Phys.*, **30**, 370-372 (1952).

control at temperatures in excess of 350° while the entire slide is coated nonselectively as the zinc temperature approaches 400° .

Conductors and resistors with sheet resistances of $2\ \Omega$ to about $20\ \Omega$ per square are easily prepared by this method.

In initial attempts to deposit zinc, the primary goal was to lay down as heavy a layer of zinc as possible. Using BuSnCl_3 as a nucleating agent an optimum exposure time to the electron beam was determined. The electron gun electrical parameters and the evaporation parameters were held constant and only the exposure time to the electron beam was varied. The results of this experiment are shown in Fig. 6. It appears that reproducibility is best when substrates are processed without breaking vacuum between the nucleation and evaporation steps.

A group of 180 degrees distributed phase-shift networks were fabricated using the described process. The circuit diagram for the 180 degrees phase-shift network, shown in Fig. 7, is equivalent to a transmission line without any series inductance or parallel conductance. To obtain 180 degrees phase shift in a distributed circuit the following conditions must be met:

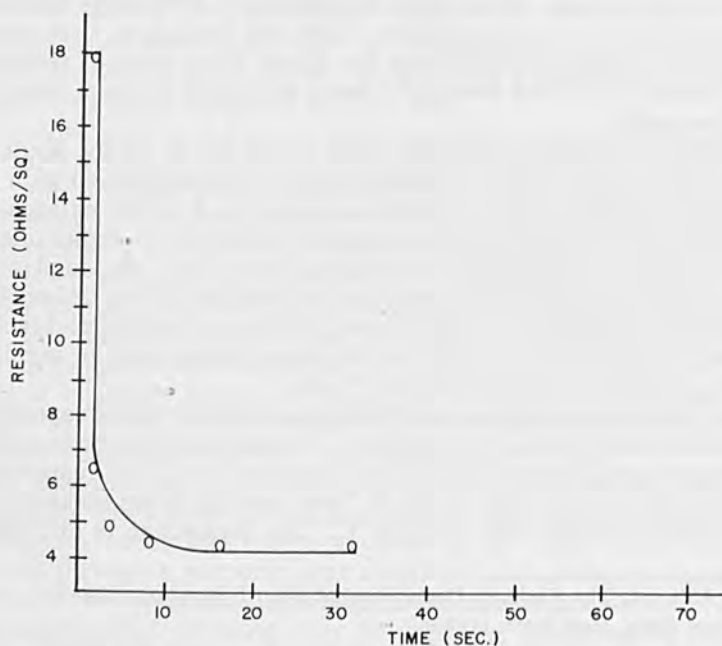


Fig. 6. Beam exposure time vs. resistance for fixed substrate temperature and zinc development time.

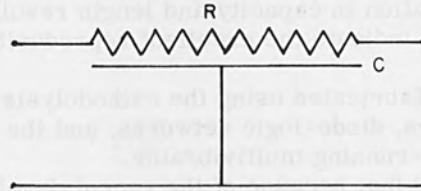


Fig. 7. Circuit diagram — 180° phase-shift network.

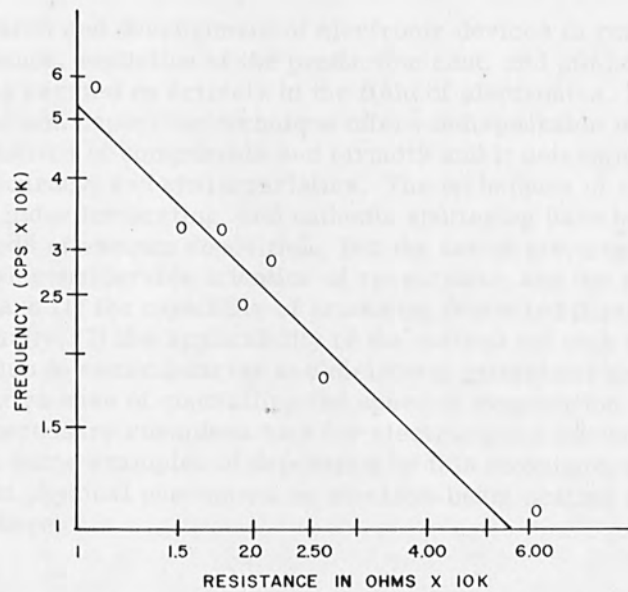


Fig. 8. Frequency vs. resistance for phase-shift networks.

where $fRC = \pi$
 f is the 180 degrees phase-shift frequency
 R is the resistance
 C is the capacity.

A graph of frequency versus resistance with C equal to 5700 pF is shown in Fig. 8. If the capacity and length of the distributed circuit were constant, all of the sample points would have been located

on this line. Variation in capacity and length resulted in the deviations shown which indicate the excellent reproducibility of the process.

Other circuits fabricated using the cathodolysis technique included notch filters, diode-logic networks, and the passive components for a free-running multivibrator.

It is anticipated that because of the recent developments in insulated-gate field-effect transistors fabricated from evaporated thin-film semiconductors and the trend towards micro-power circuitry that the cathodolysis technique will become of increasing value for the generation of microelectronic systems.

VACUUM DEPOSITION BY MEANS OF ELECTRON BEAM

INTRODUCTION

Research and development of electronic devices to realize high performance, reduction of the production cost, and miniaturization are being carried on actively in the field of electronics. In this field, vacuum deposition technique offers indispensable means for the production of components and circuits and it determines their yield, accuracy, and characteristics. The techniques of resistance heating, induction heating, and cathodic sputtering have been used as methods of vacuum deposition. But the use of electron beams has drawn considerable attention of recent date, and the reasons for this are (1) the capability of producing deposited films of extreme purity; (2) the applicability of the method not only to metallic but also to semiconductor and dielectric materials; and (3) the comparative ease of controlling the speed of evaporation.

The necessary considerations for electron guns for vacuum deposition, some examples of deposition by this technique, and some important physical phenomena on electron-beam heating are described here.

2. THE VACUUM DEPOSITION APPARATUS

2.1 A. Electron Gun Design.

There are many kinds of electron guns which can be used for beam generation; the Steigerwald gun, the Bricka and Bruck gun, the work-accelerated gun, and the Pierce gun are but a few.

Theoretically, any kind of gun is satisfactory if it produces an electron beam which has sufficient power for the evaporation of

*Fujitsu Limited, Tokyo, Japan.

the materials of interest. For practical use, however, electron guns which are especially designed for deposition are preferable.

The main items to consider in design are:

Power requirements, which are determined by the nature of the deposition materials, the necessary evaporation speeds, the distance from source to substrate, and the method of containment of the deposition materials. Some examples are shown in Table 1.

Table I

Example of Necessary Power for Evaporation

Materials	Ceramic Crucible *	Water-Cooled Copper Bed	Rate of Evaporation
Al	300 Watts	> 1.2 Kwatt	5 mg/sec
Cr	300 "	0.4 "	2 "
Ni-Cr	200 "	0.6 "	0.5 "
Au	350 "	> 1.5 "	5 "
Ta	—	> 1.5 "	0.5 "

*The crucible will be explained in Section 3.1.

The evaporation rates shown here are considered practical for 20 cm to 30 cm deposition distance (the distance between the evaporation source and the deposition substrate).

Five hundred watts of power may be sufficient if it is possible to use ceramic crucibles, while 2000 watts will be needed when using a water-cooled copper crucible; these are conservative estimates.

(2) Anode Voltage. Considering possible discharge problems between electrodes in the gun and in the power leads, the radiation hazard, and the price of the power supply, the anode voltage should be kept to as low as possible. Anode voltage less than 20 kV is desirable considering the above factors.

(3) Electron Emission Source. Because of problems with the emitter cathodes' life under the experimental conditions encountered, W, Ta, and Re, are predominantly used at the present stage.

Further bombarded solid cathodes are preferred to filamentary cathodes, even though the equipment becomes somewhat more complicated as a consequence.

(4) Control of the Electron Beam. The general methods for controlling electron-beam power are the following.

(a) Through Adjustment of Anode Voltage. Generally speaking, it is difficult to adjust the anode voltage while the deposition is in progress even though the adjustment of the anode voltage at the preliminary stage of the deposition operation is rather easy.

(b) Through Adjustment of the Applied Voltage relative to the Wehnelt Electrode. This method is considered to be most practical. The ratio of the anode voltage V_A to the Wehnelt electrode voltage V_{GC} is important, and determined by the relative position of the Wehnelt electrode to the cathode and anode, and the shape of the Wehnelt electrode.

In order to change only the electron beam power without influencing the path of the electron beam, it is necessary to pulse the anode voltage in a square wave form, or to apply the cutoff bias voltage to the Wehnelt electrode and then impress the normal voltage of the square wave pulse. One can thus control the average power by the pulse width or the frequency.

(c) The Spot Size of the Electron Beam. The practical spot size of the electron gun for machining is below 20μ in diameter and this is achieved by complex electron optical systems.

In the case of the electron guns for evaporation, the minimum spot diameter of 1~2 mm is good enough. It is thus advisable to simplify gun design as much as possible.

In electron guns for vacuum deposition, it is far more important to keep the power density inside the spot uniform, even though the spot diameter must be expanded to 10 mm.

2.1 B. Practical Example of the Electron Gun

The Pierce gun was chosen for our vacuum deposition apparatus. The electrodes of this gun, the Wehnelt electrode, cathode, and its holder and filament are all constructed in one unit as shown in Fig. 1. The anode belongs to water-cooled outer cylinder and the anode potential is at ground potential. The gun had the following specifications:

1. Anode voltage: 5 - 10 kV.
2. Maximum beam current: 150 mA. More than 95% of this current is located within a 5 mm diameter.

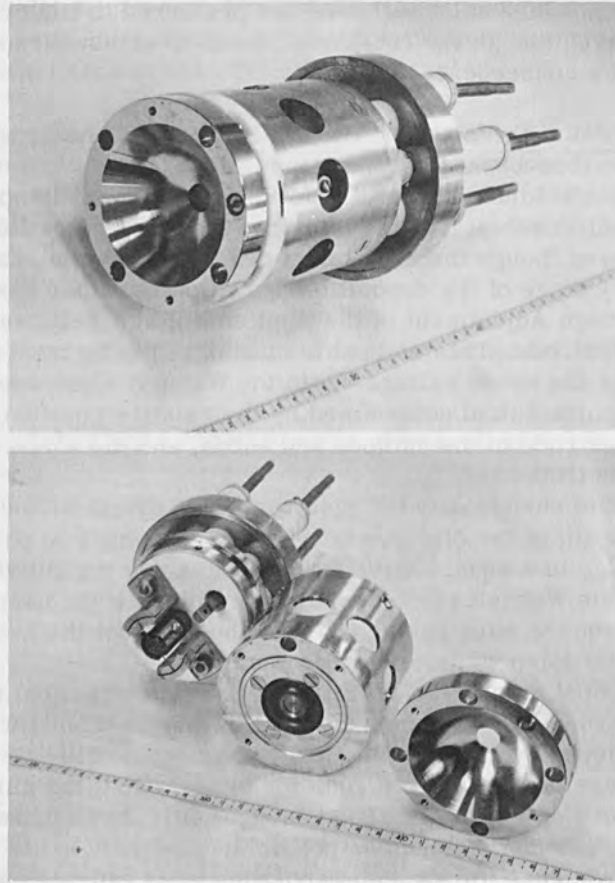


Fig. 1. The Pierce gun for vacuum deposition.

3. Beam spot size : 2 to 10 mm in diameter.
4. Focal length : $25 \text{ cm} \pm 10 \text{ cm}$.
5. Deflection range: within a circle of 10-mm diameter.
6. Control: the beam power is controllable and is capable of square wave modulation by the voltage to the Wehnelt electrode.

2.2. The Vacuum Deposition Apparatus

Figure 2 shows the deposition apparatus installed in the vacuum chamber, with the condenser lens applied to the aforementioned electron gun. The vacuum chamber is made of stainless steel and

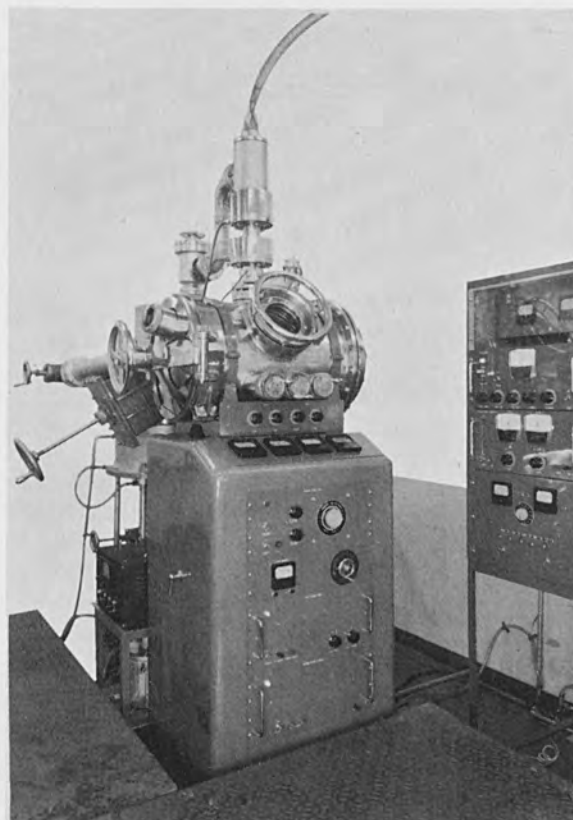


Fig. 2. The vacuum deposition apparatus.

is 40 cm in diameter and 60 cm in length. The material turret, which can hold six kinds of deposition materials, the mask turret, which can fix six kinds of deposition masks, and the substrate turret, which can hold twenty-four sheets of substrates, are driven by three handles installed in the body of the vacuum chamber. The evacuation system of this apparatus consists of an oil rotary pump with the exhaust speed of 1800 l/min, a 10 in. oil diffusion pump and a cold trap. It takes approximately 15 min to achieve vacuums of the order of 10^{-6} torr.

EVAPORATION STUDIES

3.1. Relation between Incident Electron-Beam Power and Rate of Evaporation

The rate of evaporation is given by the following equation.

$$G = P \cdot \left(\frac{M}{2\pi R_0} \right)^{1/2} \quad (3.1)$$

where G = rate of evaporation $\text{g} \cdot \text{cm}^{-2} \cdot \text{sec}^{-1}$

P = vapor pressure in microbars

M = gram - molecular weight

R_0 = gas constant ($8.3146 \cdot 10^7$ ergs $\cdot \text{deg}^{-1} \text{ k} \cdot \text{g} \cdot \text{mole}^{-1}$)

P in (3.1) is given by

$$P = \alpha \cdot \exp(\Delta F/R_0 T) \quad (3.2)$$

where α = const. (1.0133×10^6), ΔF = free energy in $\text{Cal} \cdot \text{g} - \text{mole}^{-1}$. From (3.1) and (3.2), we obtain the relations

$$G = \alpha \cdot \exp(-\Delta F/R_0 T) \cdot \left(\frac{M}{2\pi R_0} \right)^{1/2} \quad (3.3)$$

Equation 3.3 shows that rate of evaporation is only temperature dependent for a given material. When the material is heated by the constant power (W) of the electron beam, and the system is in a state of equilibrium, its temperature (T) is essentially determined by heat loss due to vaporization, conduction from the material holder, and radiation from the material and its holder. In general, the relation between W and T is very complicated; and the calculated temperature of the evaporation surface is much different from temperature which is measured by a pyrometer. Therefore, the relation between rate of evaporation and incident power must be determined experimentally. Experiment shows that relation between G and T under equilibrium conditions is given very closely by equation 3.4, provided the vapor pressure is in the range of 10^{-3} to 10 torr.

$$G = C \cdot W^n \quad (3.4)$$

c and n in equation 3.4 are determined by the nature of the material and the holding method of the material.

Representative examples of n are shown in Figs. 3 through 5. In the case of Fig. 3, Al is evaporated from a zirconium oxide crucible. It is cylindrical in shape and has 12 mm diameter, it is 12 mm deep, and 1 mm thick. Fig. 3 shows that value of n is very

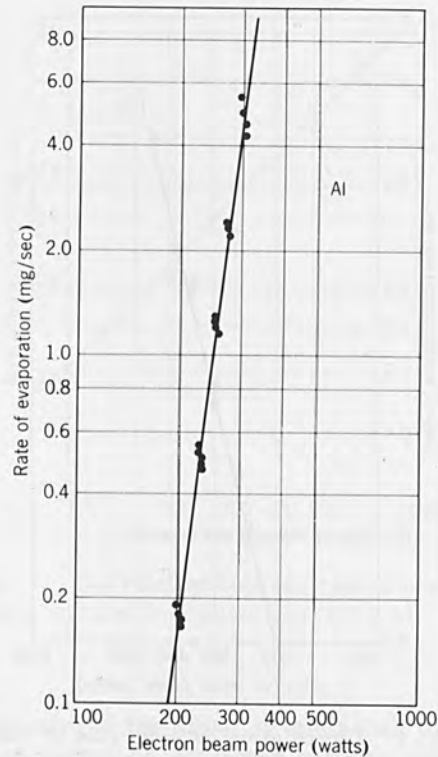


Fig. 3. The relation between sets of evaporation and electron-beam power for Al.

close to 8. In Fig. 4, Ni-Cr is evaporated from the same type of ceramic crucible as in Fig. 3, but this crucible is 8 mm in diameter, 8 mm deep, and 15 mm thick. Figure 4 shows that the value of n is very close to 4. In Fig. 5, Al is evaporated from a conical basket made of tungsten wire, it has a 10 mm top diameter and is 10 mm deep. Figure 5 shows that value of n is very close to 1. It is very clear from Figs. 3 to 5 that value of n is dependent on the nature of the material and its holding method.

3.2. Control of Evaporation Rate

As shown in Figs. 3 to 5, the relation between the incident beam power and rate of evaporation is so sensitive that even a slight

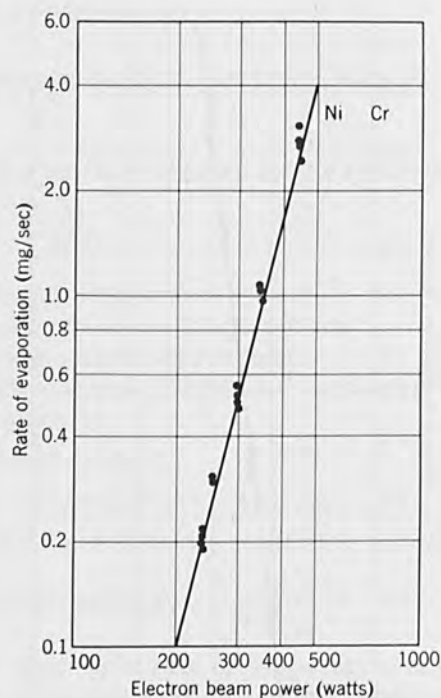


Fig. 4. The relation between rate of evaporation and electron-beam power for Ni-Cr.

power variation influences rate of evaporation to a great extent. Therefore it is necessary to use a feedback circuit control between the vapor pressure monitor and the electron gun in order to deposit at constant vapor pressure. The control system used is shown in Fig. 6. The vapor pressure monitor in itself can be also used as a criterion of the vacuum in the system. It is a modified ionization gauge.

As Fig. 6 clearly shows, the output from the monitor is amplified; by this means the voltage applied to the Wehnelt electrode is controlled and the electron-beam power is controlled so that the vapor pressure may be kept constant. The relation between the voltage of the Wehnelt electrode and the beam power is shown in Fig. 7. Examples of vacuum deposition, where constant vapor pressure is kept by the vapor pressure controller, are shown in Figs. 8 and 9. If we desire more accurate film thickness, desorbed gas from the material and its holder must be kept in negligible quantity

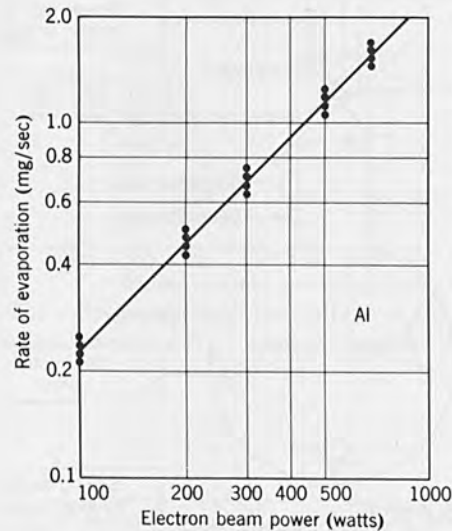


Fig. 5. The relation between rate of evaporation and electron-beam power for Al.

compared with quantity of the vapor of material, and furthermore, the temperature of deposited substrate surface must be kept constant. A monitor for alloys and compounds needs a mass spectrometer, since the partial pressure of each of the material constituents must be measured.

4. IMPORTANT PHYSICAL PHENOMENA PERTAINING TO ELECTRON-BEAM DEPOSITION

4.1. Heating Phenomena by Electron Beam

When incident electrons with high energy are given to the surface of the materials, some are reflected near the surface, but most of them penetrate into materials and lose their energy. In this case, the lost energy is spent for actuations of the lattice vibration and of the electrons inside the band or plasma vibrations. The kinetic energy of the electron is converted to heat in the following process, and the incident electrons vibrate the lattice directly. The plasma vibration is actuated, most of the energy is con-

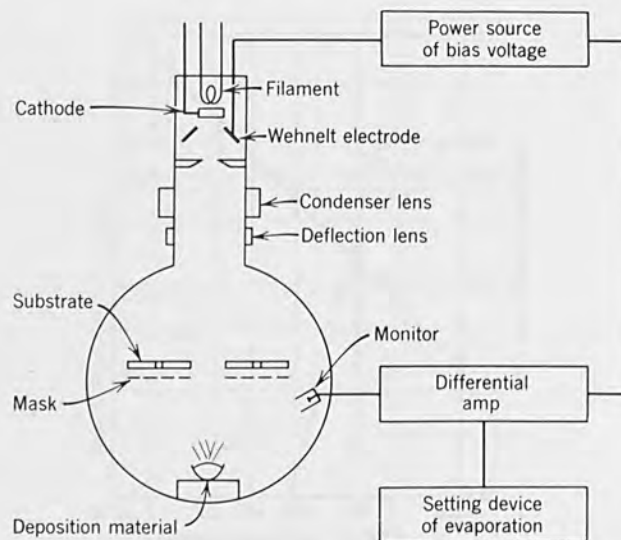


Fig. 6. The schematic diagram of the evaporation control system.

verted to the energy of the random vibration of electron, and the multiple action of these processes is seen.

The detailed process of the conversion of kinetic energy of the electrons to heat is not analyzed satisfactorily theoretically, since certain problems still remain to be solved. However, the general review is to be given to heat-generating locations which are considered to be important.

When the electron with energy V penetrates in the material, the ratio of the lost average energy $-dV/dx$ has been experimentally studied, using the thin film in most cases instead of bulk since the thin film case is simpler. x is the distance from the surface to the location of the electron.

The decay of the penetrating electron energy $V_p(x)$, is given approximately in the following formula:

$$-dV_p(x)/dx = A/V_p(x) \quad (4.1)$$

A is a constant determined by a material. When formula 4.1 is solved with $V_p(x) = V_p(0)$ at the surface of the material, $x = 0$, the following formula is obtained.

$$V_p^2(x) = V_p^2(0) - 2A \quad (4.2)$$

The formula 4.2 is known as Thomason-Whiddington law. The val-

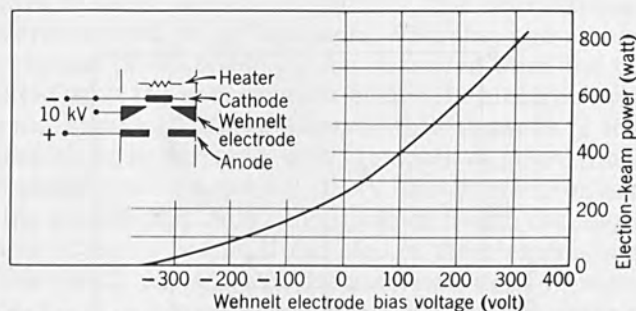


Fig. 7. The relation between the voltage of the Wehnelt electrode and beam power at anode voltage of 10 kV.

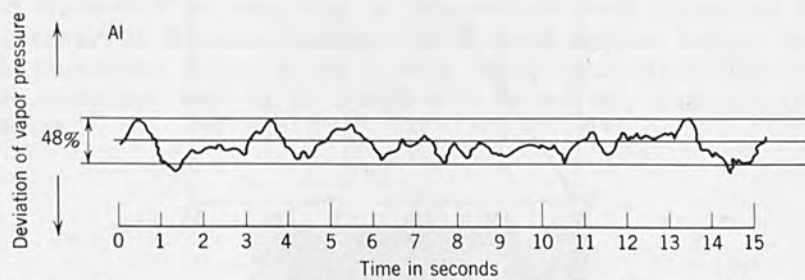


Fig. 8. Example of controlled vacuum deposition. Fluctuation of the vapor pressure is within $\pm 2.5\%$.

Table II

Whiddington's Constant
 $a = 2A$

Material	$a \times 10^{-12}$ (eV) ² /cm	$a/\rho \times 10^{-12}$
Be	0.75	0.39
Al	1.1	0.41
Cu	3.6	0.40
Ag	4.2	0.40
Au	8.9	0.46

Note: ρ in the table 2 is specific density.

ues of A in formula 4.1 is as given in Table 2 by Terrill.

When the range of incident electron for x_0 is taken, the following formula is obtained as $V_p(x) = 0$ with $x = x_0$:

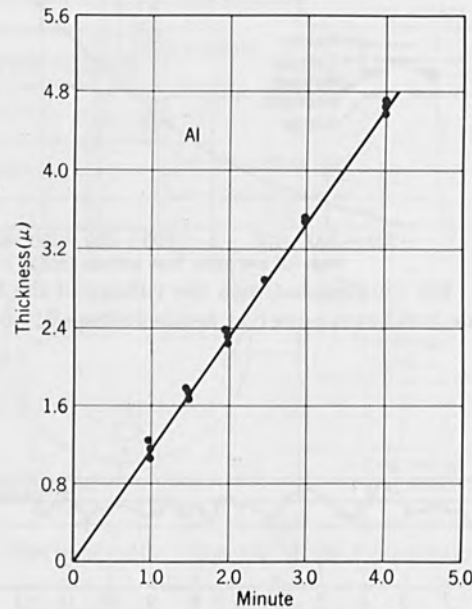


Fig. 9. Example of controlled vacuum deposition. The relation between deposition time and film thickness is almost linear.

$$V_p o^2 = 2Ax_0 \quad (4.3)$$

Therefore, formula 4.2 is indicated as follows

$$V_p^2(x) = V_p o^2 \left(1 - \frac{x}{x_0}\right) \quad (4.4)$$

$$\text{or } V_{po}/V_p(x) = 1/(1 - x/x_0)^{1/2} \quad (4.4)$$

This shows that most of the electron energy is spent toward the end of the range. Strictly speaking, it is considered that the temperature becomes maximum in a certain depth inside the material without the temperature of incident surface getting up to maximum. This phenomenon becomes remarkable when the pulse modulation of the narrow square waves is given to the electron beam.

In regard to these numerical matters, the calculations and studies were carried on by Taniguchi, Vibrans, and others. However the similar phenomenon can be observed when the power is concentrated upon the narrow area even with electron beam of direct current. Figure 10 shows electron micrographs of the deposition surface of Al evaporated with the spots of approximately 1 mm diameter which were made by a 10-kV 250-W electron beam. In Fig. 11, the photograph shows the spots of 5-mm diameter made by the same electron beam. If the size of the electron beam spot is made too small, particles of the material come to scatter actively, which was observed through the eye-window during deposition. The main reason for this phenomenon is considered to be that they come to be boiled owing to the maximum temperature under the surface of the materials. In the electron-beam deposition, it is necessary to deposit moderate size spots of electron beams, since the deposition surface is likely to be rough, as mentioned above, if the deposition material is heated with excessively small electron beams.



Fig. 10. Photograph of Al surface which is deposited on glass substrate by a narrow-shaped electron beam, 20,000X.



Fig. 11. Photograph of Al surface which is deposited on glass substrate by a wide-shaped electron beam. Power of electron beam is the same as Fig. 10, 20,000X.

4.2 Influence of Backscattering Electrons

When accelerated an electron beam bombards the material surface, and secondary electron emission in a broad sense is noticed. As this secondary electron was classified, by Rudbery, it is divided into the following three electrons: (a) Primary electron reflected elastically, (b) primary electron reflected inelastically, (c) the true secondary electron. Near the border line of (b) and (c) above, it is impossible to distinguish these two electrons exactly. However, in case the primary electron energy is of more than several hundred eV, the energy of the true secondary electron is to be less than 50 eV, and thus is discriminated from (b) in general. (a) and (b) are not distinguished, but the secondary electrons with more than 50 eV energy is, as a whole, regarded as a backscattering electron.

Therefore all the secondary electrons classified by energy are as follows:

$$\delta_{\text{tot}} = \delta_{\text{tru}} + \eta \quad (4.5)$$

In this formula

$$\begin{aligned} \delta_{\text{tot}} &= \text{yield of all the secondary electrons} \\ \delta_{\text{tru}} &= \text{yield of the true secondary electron} \\ \eta &= \text{coefficient of the backscattering electron} \end{aligned}$$

The electron beam used for deposition, being usually accelerated up to more than several thousand volts, the energetic influence of the true secondary electron is negligible. The higher the accelerated voltage becomes, the more the loss caused by radiation of X-rays becomes. As a result, the energy of the electron beam which is actually used for heating decreases to that extent. However, in the case of 10 kV, the radiation loss is smaller by 3 orders than that of the collision loss, which can be neglected. The comparison of the energy loss by collision with the electromagnetic wave radiation loss is roughly indicated by the following formula:

$$\frac{\text{Radiation loss}}{\text{Collision loss}} = \frac{V(\text{MeV}) \cdot Z}{800} \quad (4.6)$$

where

$$\begin{aligned} V &= \text{the electron-beam accelerated voltage indicated by MeV} \\ Z &= \text{atomic number} \end{aligned}$$

This fact proves that almost all the incident electron-beam energy is converted to heat when the incident electron beam penetrates the material and does not emerge from it.

However, the heating efficiency will decrease because the energy taken out of the deposition materials by scattering electrons becomes a large amount, when the backscattering coefficient η is large, and its average energy is also large. Therefore it is important to know the value of η and its energy distribution when the accelerated voltage of the incident electron beam is high. In regard to the values of the backscattering coefficient η , Sternglass [9], Pallnel [10], and Stahberg have studied the cases of incident electron of less than 20 KeV; and Trump, Van de Graaff, and Bethe have respectively studied cases where the incident electron is more than 20 KeV. These values are shown in Fig. 12. The phenomena of the backscattering electron are studied in order to explain the

value of η [13], [14]. The energy spectra of the backscattering electron is as shown in Figs. 13 and 14.

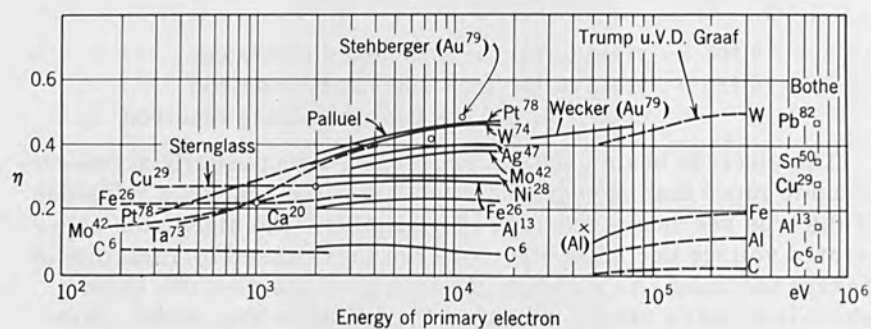


Fig. 12. The relation between the coefficient of backscattering electron and energy of the primary electron as the parameter of materials.

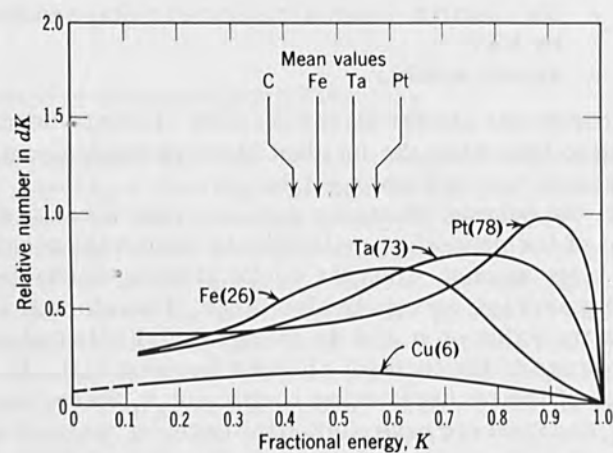


Fig. 13. Energy spectra of backscattered electrons for C, Fe, and Pt in energy ranging from 0.5 ~ 2 keV.

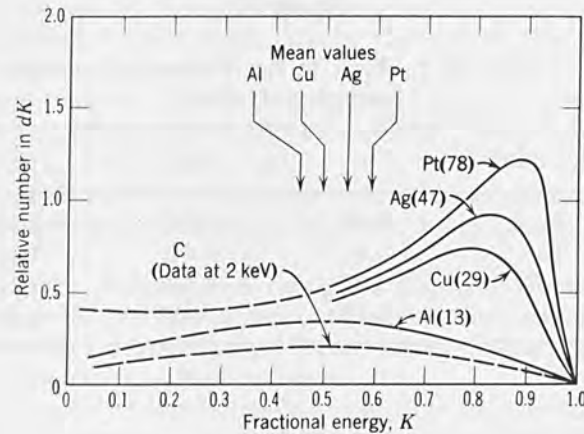


Fig. 14. Energy spectra by backscattered electrons for Al, Cu, Ag, and Pt at 32 keV.

Finding out by these energy spectra that the average fractional energy K is in proportion to atomic number Z is almost linear, Sternglas introduced the following formula as the energy of the incident electrons can be applied with the accuracy of $\pm 5\%$ in the range of 0.2 - 32 keV.

$$\bar{K} = 0.45 + 2 \times 10^{-3} Z \quad (4.7)$$

In the case of the vacuum deposition of Au, Ta, Ni-Cr, and Al are carried with the electron beam of 10 kV accelerated voltage, the backscattering coefficient η , the fraction average energy K and energy S ($\eta \times \bar{K}$), which is carried away by the backscattering electrons, are shown in Table III. η , K , S of Table III, as is clear from the definition, are all the values for the primary incident electrons. It is not difficult to shoot more than 95% of electron beam energy from the electron gun at the focusing area. But it follows that large amounts of energy are carried away by the backscattering electrons as shown in Table III.

The energy carried away by the secondary electrons reaches approximately 30%, for example, in case of the deposition of Au. This energy is likely to cause overheating of the deposition mechanism, the vacuum chamber, and the deposited substrate. This is not to be desired at the deposition, and moreover it has an unfavorable influence on the characteristics of the deposition film, so that the counter measures of the backscattering electrons are very important.

Table III

Values of η , K , S , at the Primary Electron
Energy of 10 KeV

Material	η	$\bar{K}$	S
Au	0.48	0.61	0.29
Ta	0.47	0.60	0.28
Ni-Cr	0.30	0.50	0.15
Al	0.15	0.49	0.074

5. CONCLUSION

As stated at the beginning of this paper, the electron beam has many advantages, and it is believed that application of this method will receive ever wider acceptance. In connection with the growth of acceptance of electron beam evaporation, it is considered to be necessary that the method of holding of the deposition materials as well as electron guns for deposition be studied in greater depth. Improvements in deposition monitor devices should also be made in order to improve the accuracy of the deposits. We must also consider the influence of the backscattering electron.

6. ACKNOWLEDGMENT

The author wishes to express his appreciation to Mr. Yazaki, chief of the Research Department, and to Mr. Nagata, Section Chief, Fujitsu Ltd., for their helpful comments.

REFERENCES

1. A.B. El Kareh, "Design of A High Voltage, High Power Density Electron Beam," Proc. 5th Symp. on Electron Beam Technology, Boston, Mass. 17.
2. J.R. Morley, "Electron Gun Design and Cathode Limitation," Proc. 3rd Symp. on Electron Beam Technology, Boston, Mass. 26.

3. E. Sugata and M. Terade, "A Study on Laminarity of Electron Stream," Tech. Rept., Osaka University, Vol. 12, No. 518.
4. E. Sugata and M. Terada "Analytical Design of Electron Gun with Temperature Limited Cathode," Tech Rept. Osaka University, Vol. 12, No. 495.
5. K. Yamagishi, "Trial Vacuum Deposition Apparatus Using Electron Beam," Commt. E.B. Tech. Japan, Rept. No. 38A-61.
6. J.R. Pierce, "Theory and Design of Electron Beam."
7. N. Taniguchi and S. Maezawa, "Temperature Analysis of Electron Beam Machining," Proc. 5th Symp. on Electron Beam Technology, Boston, Mass., 35.
8. G.E. Vibrans "Theoretical Temperature Rise in Materials to Electron Beam," Proc. 5th Symp. on Electron Beam Technology, Boston, Mass. 124.
9. E.J. Sternglass, Phys. Rev. 95, 345.
10. P.C.R. Pallnel, C.R. Acad. Scir Paris 225, 383.
11. J.G. Trump and R.J. van deGraaf, J. Appl. Phys. 18, 327.
12. R. Kollath, "Handbuch der Physik," XXI, 232.
13. T.E. Everhart, J. Appl. Phys., 31, 1483.
14. G.D. Archard, J. Appl. Phys., 32, 1505.

AUTOMATIC CONTROL OF ELECTRON-BEAM MICROMACHINING

INTRODUCTION

Recently, the industrial applications of electron-beam heating, such as melting, evaporation, welding, drilling, and cutting of materials, have developed rapidly. But, when it comes to the micromachining by focusing the beam in a very small area for fine processing of work material, full automatization of its control is desired to make it truly practical. In this paper a newly developed automatic control system is described. By the use of this system the micromachining can be fully automated by the program prerecorded on a magnetic tape.

THE APPARATUS

The apparatus consists of three main units: the electron-beam micromachining unit, the electron-beam control unit, and the digital-control unit of the workpiece carriage.

Figure 1 shows a block diagram of the apparatus. The following items as information are recorded on the magnetic tape:

1. Information on the control of the workpiece carriage:
 - (a) Processing speeds in X, Y directions.
 - (b) Processing distances in X, Y directions.
 - (c) Processing direction.
2. Information on the condition of the electron beam:
 - (a) Pulsative or not.
 - (b) Scanning of the electron beam.
 - (c) Duration of the beam illumination.
 - (d) Drill: the electron beam to be cut-off at the instance of perforation.

*The Institute of Physical and Chemical Research, Komagome Bunkyo-Ku Tokyo, Japan.

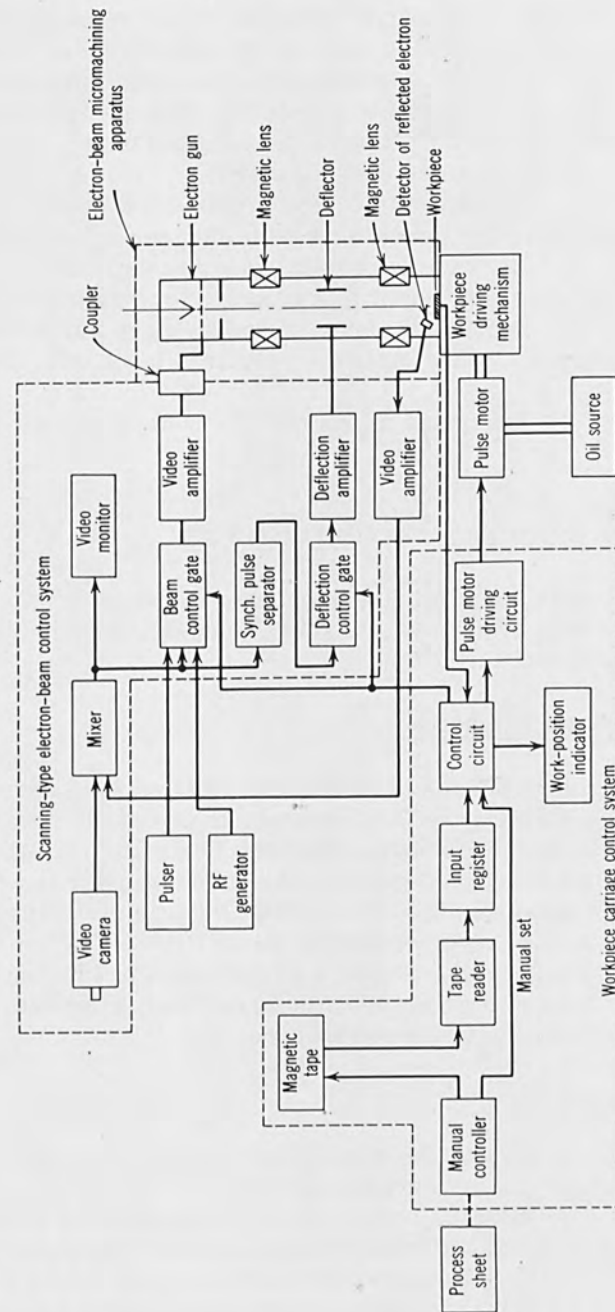


Fig. 1. Block diagram of electron-beam micromachining apparatus.

During the processing of the material, these items of information on the magnetic tape are read out by a tape reader, memorized in the input-register successively, and sent to proper channels from the control circuit to direct the movement of the workpiece carriage and the condition of the electron beam. The control circuit can be operated also manually.

For instance, the following can be processed by this system: (1) cutting of fine slits or patterns with electron beam fixed and carriage moved; (2) cutting of rectangular slots, surface treatment and zone melting of thin film by electron-beam deflection and carriage movement; and (3) engraving of pattern and surface treatment by ITV controlled electron beam with fixed carriage.

A photo of the apparatus minus the ITV control unit is shown in Figure 2.

ELECTRON-BEAM MICROMACHINING UNIT

This unit consists of an electron-beam chamber and a machining chamber both evacuated to 1×10^{-5} mm Hg. Figure 3 shows the unit in diagram form.

Electron-Beam Chamber [1]

The electron-beam chamber is made up of an electron gun, the first focusing magnetic lens, stigmator, electrostatic deflection plates, and the second focusing magnetic lens. The electron beam from a tungsten hairpin filament of 0.2-mm diameter is accelerated by 50 kV and focused to about 50 to 100 μ in diameter on the workpiece with the power density of about $10^8 - 10^4$ W/cm².

The focal length of the second magnetic lens is 120 mm which provides an ample space for machining and fitting out with observation accessories over the workpiece.

Machining Chamber

As shown in Fig. 4, the machining chamber contains the workpiece which can be moved in *X*, *Y* directions by a running mechanism, the optical accessories for observation of the workpiece, and the detector of reflected electrons for the scanning-electron microscope to observe the surface of the workpiece. The movement of the carriage, on which the workpiece is fixed, is operated from outside of the vacuum chamber as shown in Figure 5,

which enabled its accurate automatic control of the digital system and at the same time the minimization of the chamber space to save the evacuation time. In the figure, the sealing between the carriage and the evacuated chamber is by an O-ring, but metal-to-metal sliding contact with oil film in between can also do it.

Two pulse motors drive the workpiece carriage, one in X direction, the other in Y direction, with speeds from 5.12 mm/sec



Fig. 2. Electron-beam micromachining apparatus.

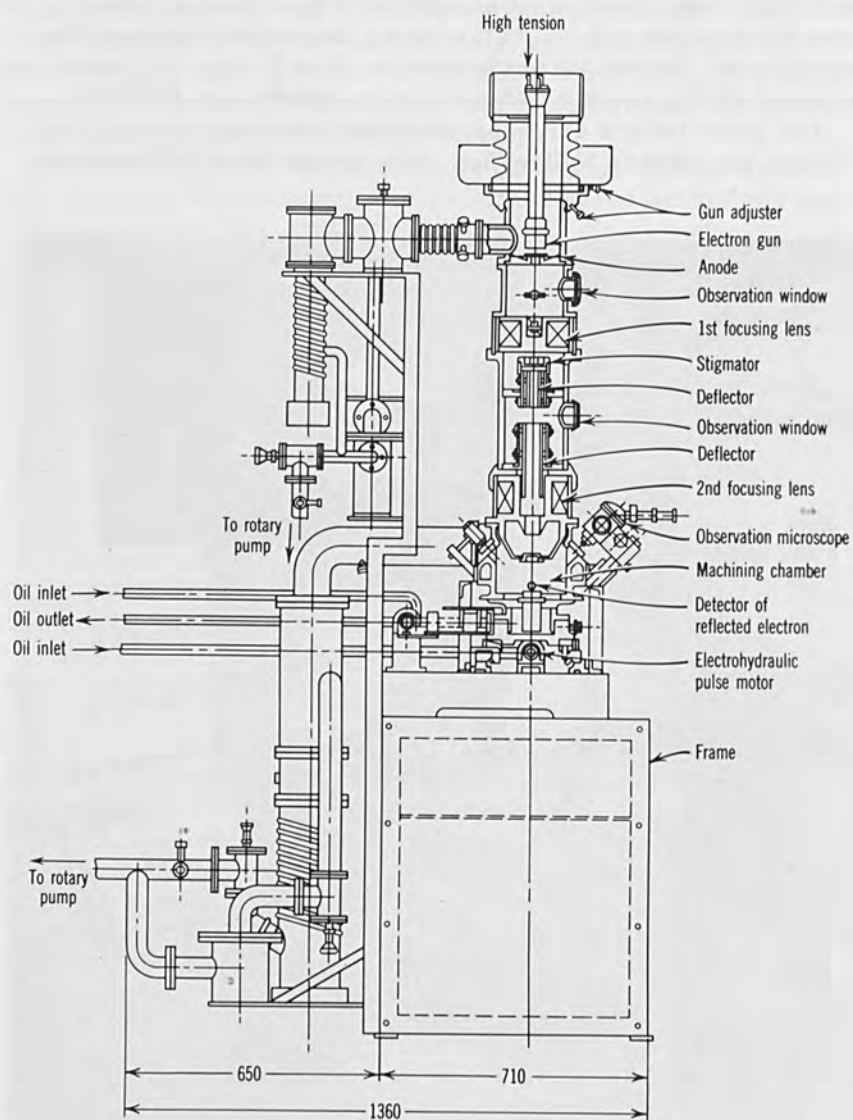


Fig. 3. Construction diagram of electron-beam micromachining unit.

to 1.25×10^{-4} mm/sec, every pulse to the motor effecting the carriage movement of 10μ .

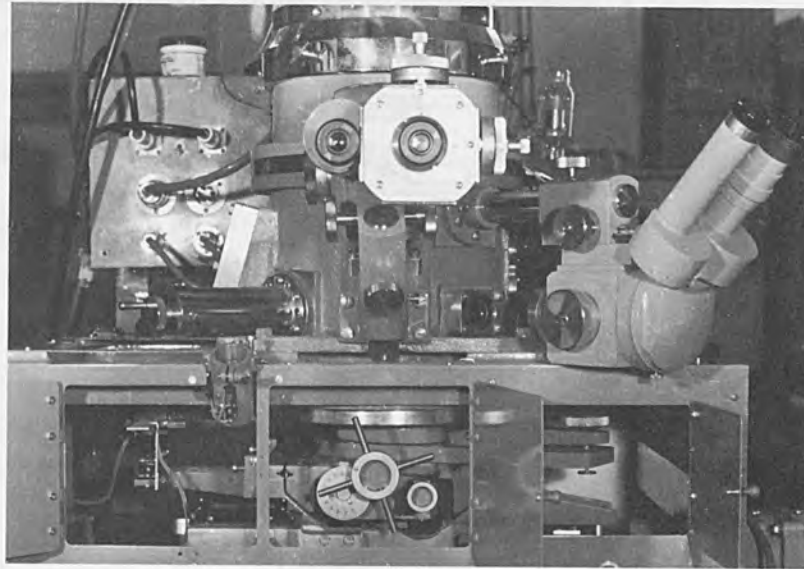


Fig. 4. Electron-beam micromachining unit less electron-beam chamber.

THE ELECTRON-BEAM CONTROL UNIT

The most outstanding feature of the electron-beam machining is that the electron beam can be very easily controlled as follows by the gate signal from the control circuit to the gate circuit as shown in Fig. 6:

1. The beam gate controls the beam bombardment interval.
2. The dc gate controls the dc intensity of electron beam.
3. The pulse gate controls the pulsatory operation of electron beam.
4. The video gate is used when the electron beam is ITV operated.
5. The deflection gate controls the electron beam deflection (Fig. 1).

DC Operation

The electron beam is cutoff by the supply of a negative voltage to the control grid of the electron gun. By the dc gate, RF carrier is transmitted, amplified to a suitable voltage, delivered at -50

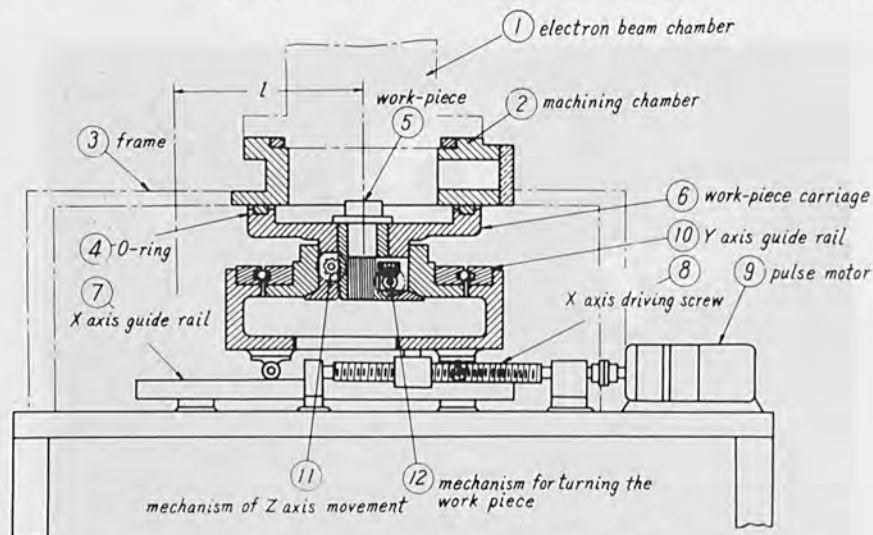


Fig. 5. Construction diagram of machining chamber with driving mechanism of carriage.

kV level by the RF transformer and rectified to supply a dc voltage to the control grid.

In an apparatus used previously [1], the RF carrier system was applied to supply a pulse voltage to the grid by using the pulsed RF carrier.

Pulse Operation

As is well known, pulsatory operation of electron beam is effective in reducing undesired heating and melting around the processed point. Such operation can be realized by supplying successive positive pulses to the control grid whereby the electron beam impinges upon the workpiece only for the pulse duration.

ITV Operation

As shown in Fig. 7 the scanning-type control system was devised on the principle of television system with ITV camera and the scanning-electron microscope. The deflection of the electron beam of the machine is synchronized with that of the electron beam of the video camera, and the intensity of the electron beam is modified by the video signal.

A large black and white pattern, viewed by the video camera, is transferred onto the workpiece as a reduced image, which is the pattern for processing. The reduction rate of the pattern is

set by the voltage applied to the deflection plate. A 14-in. TV monitor is used to observe the pattern to be machined.

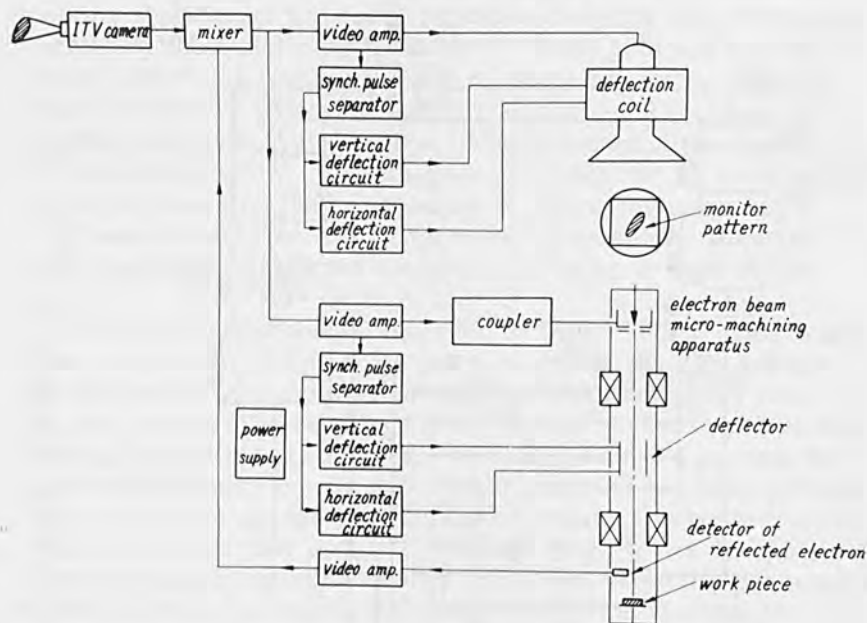


Fig. 6. Scanning control system for electron-beam micromachining.

The scanning-electron microscope is used to set the position on the workpiece where it is to be processed by overlapping the microscope image over the video camera image in the field of the video monitor.

The maximum deflection of the electron beam on the work-piece is about $6 \times 6 \text{ mm}^2$ in area, and the resolution of the pattern is about 350 lines. As the spot size of the electron beam is about 50μ , this resolution is sufficient for the spot to cover the whole area. A new electron beam chamber with the spot size less than 10μ with increased scanning lines is now being developed.

Since a commercial ITV camera of vertical and horizontal frequencies of 30 cps and 8 kc, respectively, is used in the present work, the video signal has the bandwidth of about 2 Mc. To transfer the video signal onto the grid with high fidelity, the coupler must have the bandwidth of 2 Mc. As the capacitance in the gun system is about $60 \mu\mu\text{f}$ and the cutoff frequency of the amplifier

is given $f_n = 1/6.6 RC$, the output impedance of the video amplifier was designed to be about $1.5 \text{ k}\Omega$.

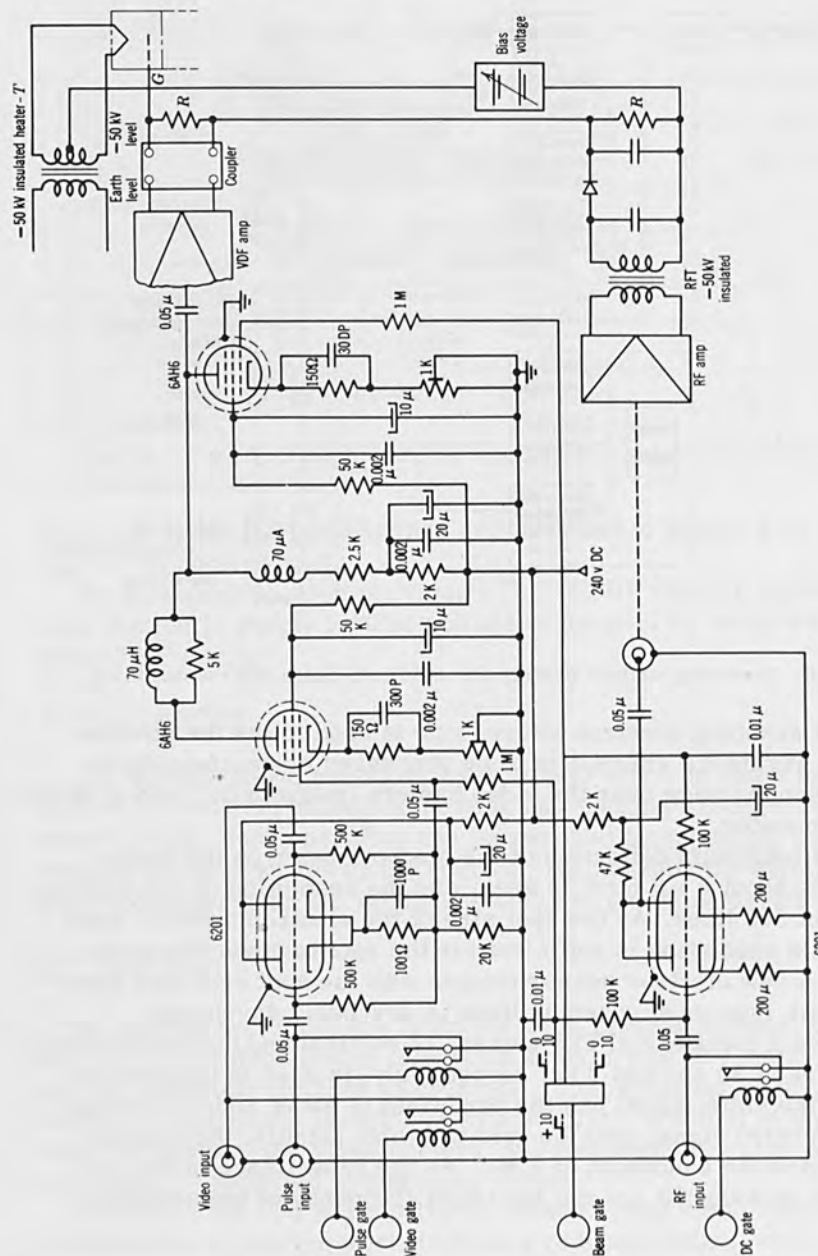


Fig. 7. Circuit diagram of electron-beam controlled gate.

Coupler

To control the electron beam in the range from the optimum processing condition to the cutoff condition, the grid must be biased negatively about 200 volts to cathode. As the grid and the cathode are at -50 kV , the level of the control signals must be raised to 50 kV . There are various coupling methods, such as transformer-coupling and condenser-coupling between the amplifier and the grid at high potential. Use of a pulse transformer [2] insulated for 50 kV is an excellent method for obtaining the pulse beam with many advantages, but is not suited to obtain the required wide bandwidth. Hence the condenser-coupling is used in this work.

During the experiments, however, when the high-tension breakdown occurred in the electron-beam chamber, the high voltage pulsed back to the amplifier through the coupling, which often caused trouble. The couplings mentioned above cause accidental pulsing back of high tension to low tension side. To prevent this undesirable occurrence, a new coupling method has been devised by the use of an optical coupling transformer. The method of optical coupling shown in block diagram (Fig. 8) has been devised, in which the only possible energy transfer is from the light source to the photocell and not in the reverse direction whereby the anomalous voltage caused by the high-tension breakdown is perfectly arrested in the light path of the optical coupling.

The light beam can be modulated by the video signal with a Kerr cell light modulator (Fig. 8a), or better with a semiconductor laser (Fig. 8b), the light source of which can be modulated by the video signal current through the laser. This method with a GaAs laser is known in the optical transmission system of television. In the case of the pulse control of the electron beam, the pulse-operated GaAs laser is most suited.

As the detector, AgCs photocell or silicon photodiode of high-speed response of about 10^{-8} sec is used.

THE WORKPIECE CONTROL UNIT

This unit is for the workpiece to move accurately to X and Y directions with given speeds and for given distances following the information from the magnetic tape.

The speed of the workpiece carriage can be changed in thirteen steps from $2^9 \times 10 \mu\text{sec}$ (5.12 mm/sec) to $2^{-3} \times 10 \mu\text{sec}$ ($1.25 \times 10^{-4}\text{ mm/sec}$), which are effected by the frequency of the feed

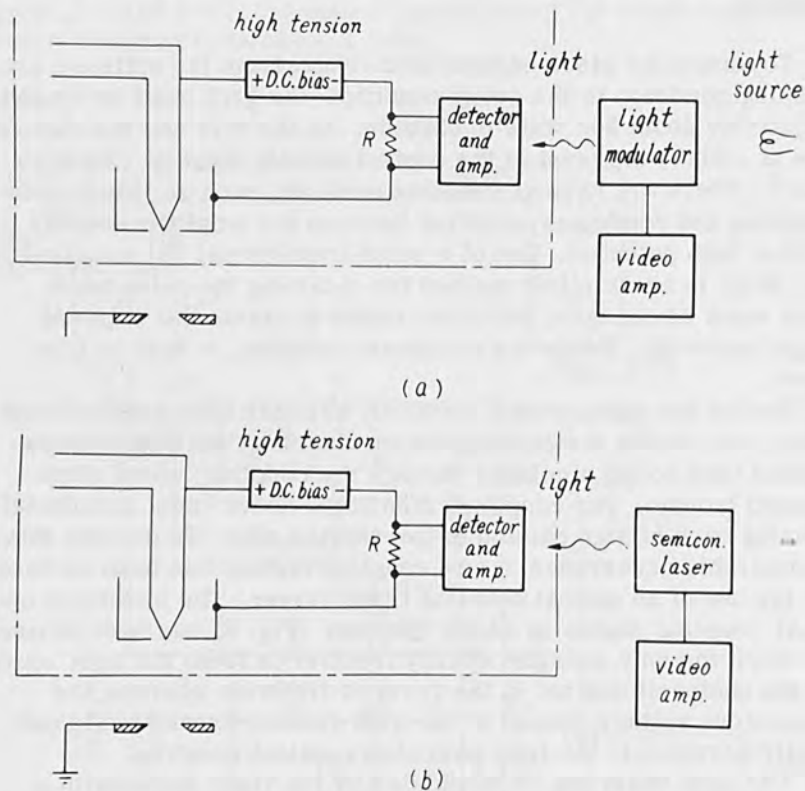


Fig. 8. Block diagram of optical coupling method.

pulse to the pulse motor. Feed pulses are made by stepping-down of pulses from a standard quartz oscillator of 2048 cps. Maximum speed of the carriage is 5.12 mm/sec by pulses of 512 cps frequency to the pulse motor. X, Y coordinates of the carriage are indicated on Decatron array by counting the feed pulse to the pulse motor with the unit of 10μ .

EXAMPLES OF PROCESSING

Drilling

Figure 9 shows holes perforated in a quartz plate by a dc electron beam of 50 kV, $50 \mu\text{A}$. In (a) the electron beam was cutoff as soon as the hole was perforated; the processing time was 15 sec. In (b) the electron beam was cutoff before the perforation.

For transparent materials such as quartz, the residual stress

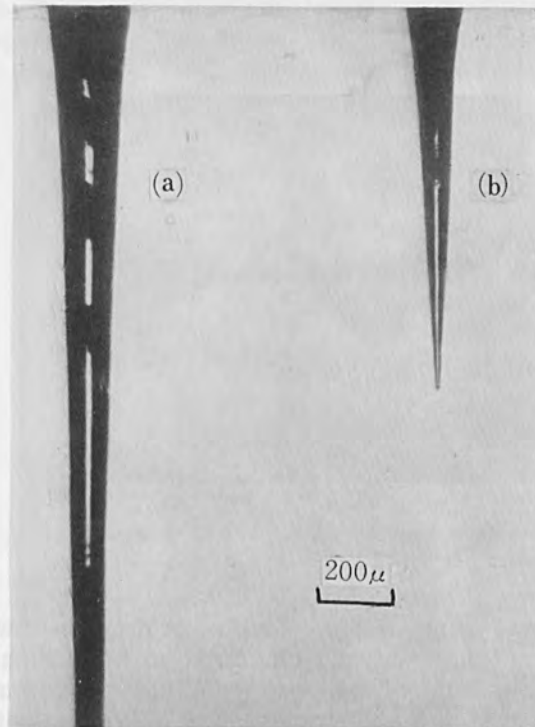


Fig. 9. Electron-beam perforation in 2-mm thick quartz. Beam intensity 50 kV, 50 A, dc.

in the material caused by the heat of processing can be estimated by the photoelastic method. Figure 10 shows an example of the photoelastic pattern of quartz with perforation in the center, and from the pattern the residual stress is estimated at about 5 kg/mm^2 .

Figure 11 shows an example of drilling in ferrite, which is easily processed by the electron beam.

Cutting

For cutting, the workpiece is moved under the fixed dc electron beam.

Figure 12 is an etched view of cross sections of the cut on a stainless steel plate by the electron beam of 50 kV, $300 \mu\text{A}$, and the carriage movement of $25 \mu\text{sec}$. The width of the cut is

independent of the speed of the carriage, but the depth of the cut is proportional to the inverse square root of the carriage speed.

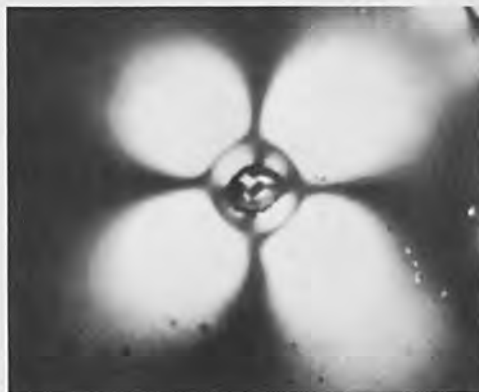


Fig. 10. Photoelastic pattern of quartz with perforation.

Figures 13 and 14 are some examples of processed workpieces by the carriage control method. At corners the angles are a little rounded because of the backlash of the running mechanism and the deformation of the O-ring. Figure 13 is a cut pattern on a deposited titanium film on glass substrate by the electron beam of 50 kV, $0.5 \mu\text{A}$ and the carriage movement of 0.32 mm/sec. It is clear that this method can be applied to fine cutting of thin-film circuit elements. Figure 14 is a cut pattern on a stainless steel plate by the electron beam of 50 kV, $120 \mu\text{A}$, and the carriage movement of 0.08 mm/sec.

ITV Controlled Processing

The ITV controlled electron beam makes the processing of complicated shapes possible. The cutting of fine patterns on metal thin films requires only a few seconds by the scanning-electron beam with 50 kV, $20 \mu\text{A}$, and the scanning area of 6-mm square.

Figure 15 is an example of a pattern cut on a stainless steel with the electron beam of 50 kV, $80 \mu\text{A}$, and 20 sec illumination. In this case, the horizontal and vertical scanning are 8 kc and 30 cps, respectively, and the reduction rate of the processing pattern from the mother pattern is 1/200.

In comparison with the original pattern, the cut pattern is



Fig. 11. Electron-beam perforation in 2-mm thick ferrite.

distorted and lacks in uniformity; the distortion is caused by the nonlinear characteristic of the deflection voltage for scanning the electron beams of the video camera and the machine, and the lack of uniformity is caused by the lack of uniformity of illumination on the mother pattern and by the relation of the scanning direction to the pattern.

When the scanning crosses the pattern line perpendicularly, the processing is not effective in comparison with the scanning parallel to the line, as is clearly demonstrated by cutting a specially shaped pattern shown in Fig. 16a. If the scanning is along the arrow A direction, the processed pattern becomes as shown in Figure 16b. Therefore, to obtain a uniform processing

of this pattern, the scanning must be along the arrow B direction. In the scanning-type processing, which is a special characteristic of the electron-beam processing, the scanning direction should be given a careful consideration.

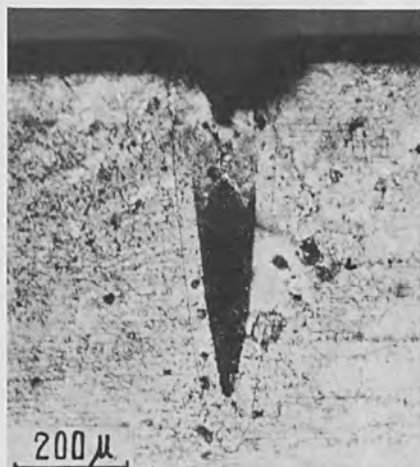


Fig. 12. Etched cross section of cut on stainless steel beam intensity: 50 kV, 300 μ A. Carriage speed 25 μ sec.

Surface Treatment and Zone Melting

The deflection of the electron beam and the carriage movement are both used for surface treatment of materials and zone melting of thin films. For example, vacuum evaporated Ge films on tungsten plate, which are almost amorphous, can be locally crystallized by the zone melting. Figure 17 is a photograph of evaporated Ge film zone-melted by electron-beam heating. It was found that the zone-melted film becomes partly single crystal. As is well known, the evaporated Ge films is of *p* type even if it is made from *n* type germanium, but after the zone melting it becomes *n* type.

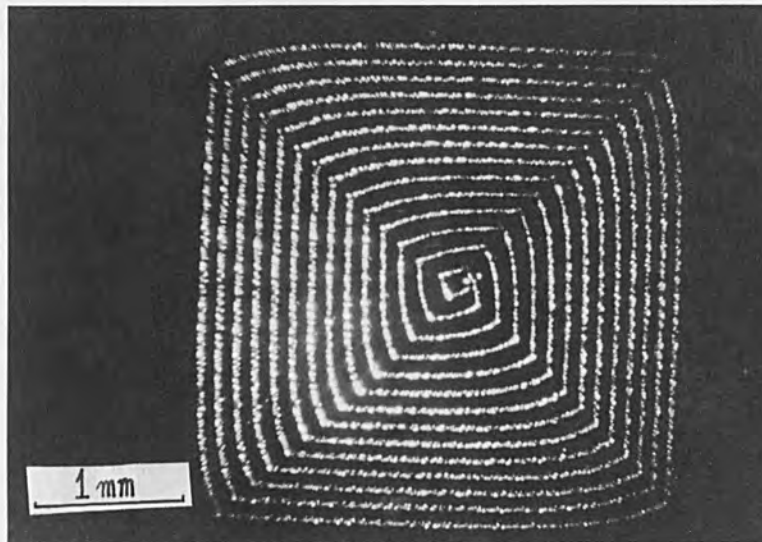


Fig. 13. Cut pattern on Ti film. Beam intensity: 50 kV, $0.5 \mu\text{A}$. Carriage speed 0.32 mm/sec.

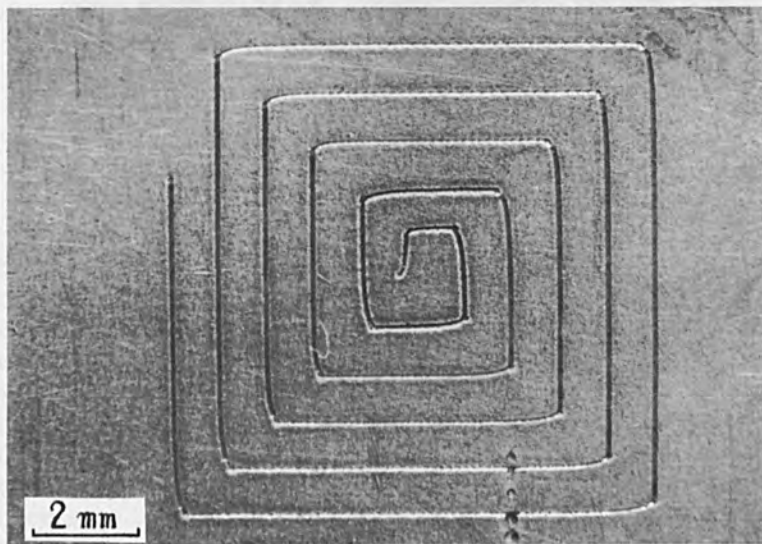


Fig. 14. Cut pattern on stainless steel. Beam intensity: 50 kV, $120 \mu\text{A}$. Carriage speed: 0.8 mm/sec.

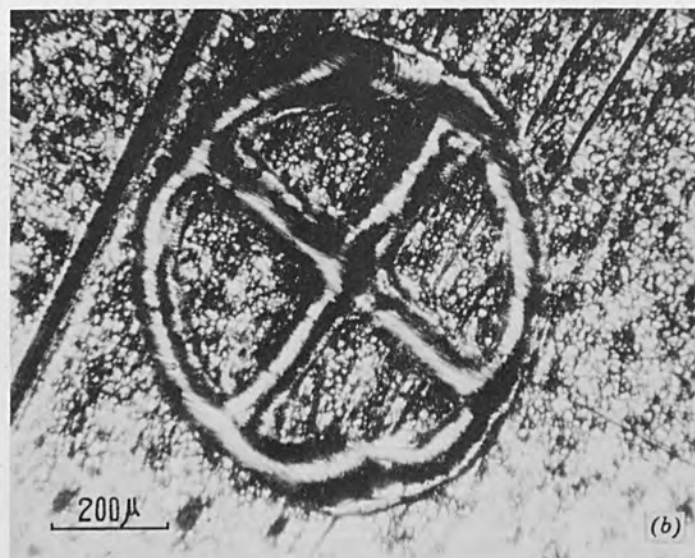
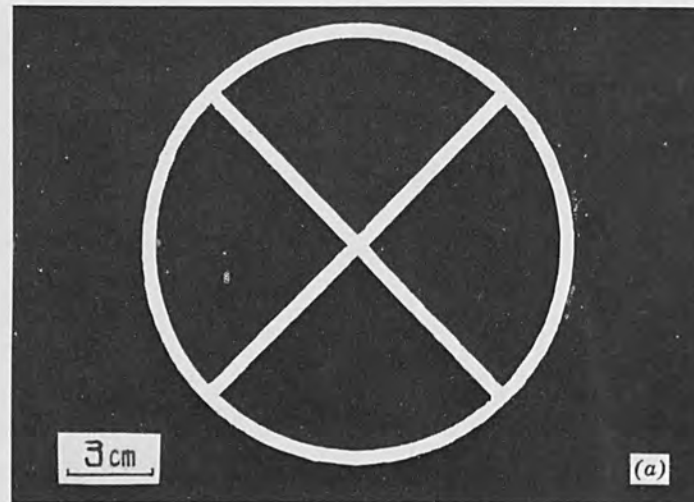


Fig. 15. Cut pattern on stainless steel by ITV controlled processing: (a) mother pattern, (b) cut pattern, reduction rate: 1/2000.

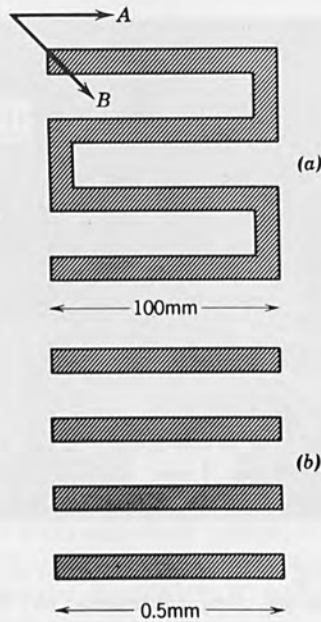


Fig. 16. Schematic representation of effect of ITV scanning: (a) mother pattern, (b) cut pattern by scanning of direction A.

CONCLUSION

The electron-beam micromachining was automated by adopting the scanning-type control system of the electron beam and the digital-control system for the workpiece carriage. The electron-beam machine with these control systems was used for drilling of fine holes, cutting of complicated patterns, and also for surface treatment of materials, zone melting of thin films, and scanning processing of materials. Some examples of these processing confirmed that the micromachining could be used for fine processing of materials. But, it is still experimental and has to be improved upon in several aspects; the spot size of the electron beam should be of the order of microns, and the carriage movement should be more exact.

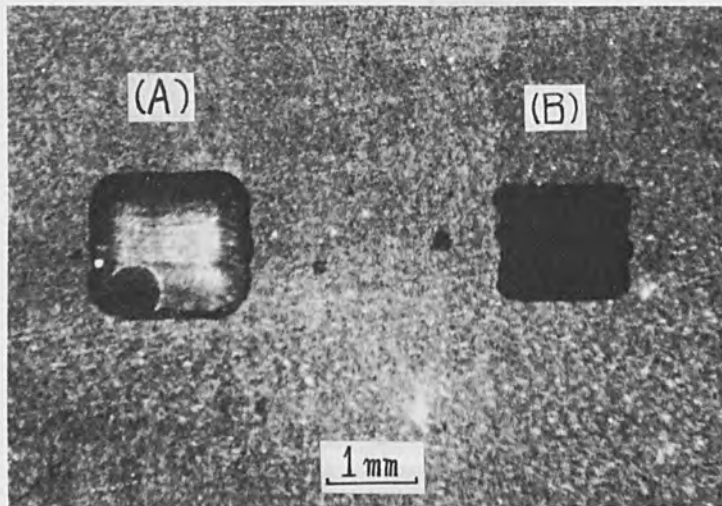


Fig. 17. Zone melting of Ge evaporated film on tungsten plate by electron-beam heating. Beam intensity: (A) 50 kV, 40μ A. (B) 50 kV, 10μ A.

ACKNOWLEDGMENTS

The authors wish to express their thanks to Dr. Y. Suge, chief of the laboratory, for his kind advice and also to members of the Committee of Electron Beam Science and Technology of Japan for valuable discussions.

REFERENCES

1. S. Namba, Proc. 4th Symp. on Electron Beam Technology (Alloyd Corporation), 304(1962).
2. W. Optiz, Proc. 2nd Symp. on Electron Beam Technology (Alloyd Corporation), 32(1960).

*Hirokazu Kimura, Hifumi Tamura,
Kazutoshi Ikegami, and Kiyoshi Sakashita**

FABRICATION OF MICROELEMENT RESISTORS BY ELECTRON BEAM AND THEIR CHARACTERISTICS

INTRODUCTION

The use of electron beams for cutting grooves in metal films for the production of microelement resistors is well known. The process has many advantages of high production rate, adaptability to different resistance, flexibility to pattern design, and high accuracy of resistance.

In this paper, the conditions most suitable for cutting grooves in metal films and the electrical properties of many resistors produced by this method are reported. A special laboratory instrument is designed, in which a simple controlling circuit of electron beam for cutting the normalized pattern, an electron lens excited by a permanent magnet, and a successive feed mechanism of elements are used. The disadvantages of high capital investment and elaborate operation are improved.

SUITABLE CONDITIONS FOR CUTTING

Nickel-chrome films with thickness of a few thousand angstroms on ceramic substrate and that on Fotoceram substrate are used. The elements with low resistance between $100 \Omega/\text{cm}^2$ and $200 \Omega\text{cm}^2$ are employed, since the cutting conditions depend upon the thickness of metal films and irregularities on the surface of substrate. The size of substrate is $8 \text{ mm} \times 8 \text{ mm}$ and that

*Hitachi Central Research Laboratory, Kokubunki, Tokyo, Japan, and Hitachi Research Laboratory, Hitachi, Ibaragi, Japan.

of metal film is $5\text{ mm} \times 5\text{ mm}$. Typical microelement resistors are shown in Figure 1; (a) is cut by a diamond-scribing process and (b) by an electron-beam process.

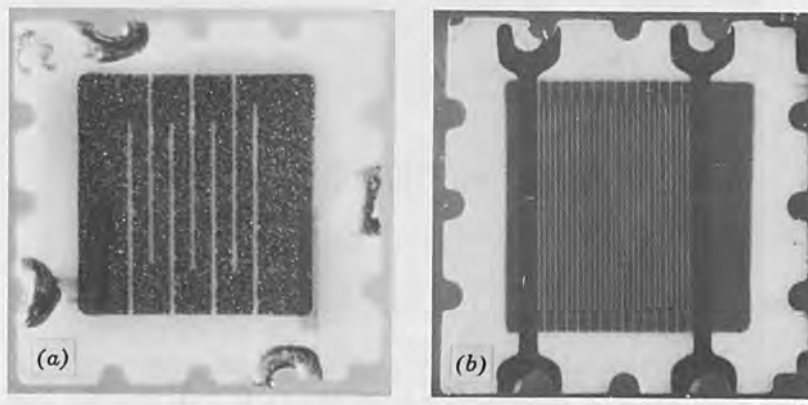


Fig. 1. Typical microelement resistors. (a) By diamond scribing process. (b) By electron beam process.

The higher scanning speed of electron beam and lower accelerating voltage are desirable for production. The relations between beam-scanning speed and minimum beam current for cutting are measured, the parameter being the accelerating voltage. The examples are shown in Figure 2, in which the accelerating voltage of 30 kV, the beam-scanning speed of 30 mm/sec, and the different beam current are used. Figure 3 shows the results of experiments. In the case of ceramic substrate, the good results are obtained under the conditions of accelerating voltage of 30 kV, scanning speed of 30 mm/sec, beam current of $15\mu\text{A}$, and groove width of more than 10μ . In this experiment, the direct current heating of tungsten filament in electron gun is always used, because in the case of alternating current heating the grooves in metal films are cut discontinuously, owing to the ripple in the beam current emitted from the filament. For Fotoceram substrate, the accelerating voltage of 10 kV is enough.

Under the higher accelerating voltage cracks in the substrate are frequently recognized by the electron microscopy. Figure 4 shows an example.

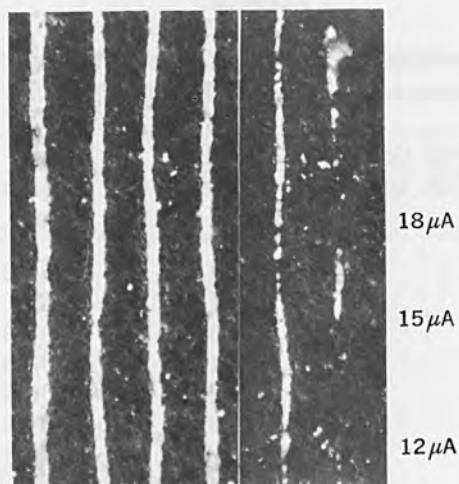


Fig. 2. Measurement of minimum beam current under the constant accelerating voltage (30 kV) and constant scanning speed (30 mm/sec).

ELECTRICAL PROPERTIES

The electrical properties of a few hundred elements produced by the electron-beam process are examined. The low-temperature coefficient and noise voltage of less than $1 \mu V/V$ are obtained and the change of resistance is less than 1% after the life test of 7000 hours. The typical examples are shown in Table I.

The width of the groove affects the quality of the element. The elements with grooves of less than 10μ width have frequently inferior qualities caused by the pinholes and irregularities of metal films on the ceramic substrate. The greater the number of grooves, the worse the qualities of elements become. In the case of metal films on the ceramic substrate, about 10% of elements with grooves less than 20, and 50% of the elements with grooves more than 30, show the inferior properties. Table II shows these results. In the case of Fotoceram substrate, the good results are usually obtained.

Table I

Electrical Properties of MicroElement
Resistors Produced by Electron Beam

Number of Grooves	Cutting Conditions		Initial Resistance Ω	Final Resistance $k\Omega$	Noise Volt. $\mu V/V$
	Acc.Volt. kV	Beam Curr. μA			
36	30	10	91.4	293.2	0.13
35	30	15	104	242	1.45
35	30	15	302	695.9	1.7
34	30	15	113	270.3	0.16
33	30	15	223	636.4	0.39
33	30	10	322	954.8	0.55
27	30	10	70	96.1	2.85
27	30	10	107	122	0.32
28	30	10	335	997	0.48
24	30	11	720	1146	0.48
23	30	15	470	1041	0.48
22	30	11	1290	2350	0.54
22	30	15	450	1049	0.48
18	30	15	740	1047	0.48
16	30	13	1080	1093	0.48
10	30	15	280	68.2	0.10
10	30	15	215	50.4	0.14
10	30	15	282	65.9	0.13
9	30	15	248	51.8	0.16
9	30	15	288	55.7	1.30
8	30	15	274	52.4	0.16
7	30	15	330	51.7	0.18
7	30	15	259	51.3	0.16
12*	30	5	164	38.1	0.15
12*	30	5	144	32.9	0.19
12*	30	5	72	15.3	0.20

Scanning speed = 1 - 3 mm/sec. Substrate = Ceramic
(*Fotoceram).

Table II

Relations Between Number of Grooves and
Inferior Quality Rate

Number of Grooves	More than 30 Lines	25-29 Lines	20-25 Lines	15-19 Lines	Less than 14 Lines
Inferior quality rate	55%	45%	18%	14%	4%

Substrate = ceramic.

A NEW INSTRUMENT FOR FABRICATION

A special laboratory instrument for fabrication is newly designed. The main column of the instrument is composed of an electron gun with a tungsten filament of hairpin type, an electron lens of double-gap type excited by a permanent magnet, and a working chamber. The cross section of the instrument is shown in Fig. 5.

The minimum spot size of electron beam on the element is about 10μ when the tungsten filament of 0.15-mm diameter is used. The maximum current density on the element is about 10 amp cm^2 when the lens aperture of 1 mm is used.

The twenty-four elements are set on a rotating disk in the working chamber, three of which are always left in vacuum. They are fed from outside at atmosphere pressure to the vacuum chamber of 10^{-4} mm Hg successively, and after working they are taken out continuously.

ELECTRON-BEAM SCANNING SYSTEM

The electron beam is scanned automatically by a beam-deflecting system and the normalized pattern on the metal film is engraved. The schematic diagram of beam scanning system is shown in Fig. 6. Figure 7 shows the wave forms applied to the grid of electron gun and X- and Y-axis deflectors of the instrument, respectively. In Fig. 8, an element of normalized pattern and a model for illustration are shown.

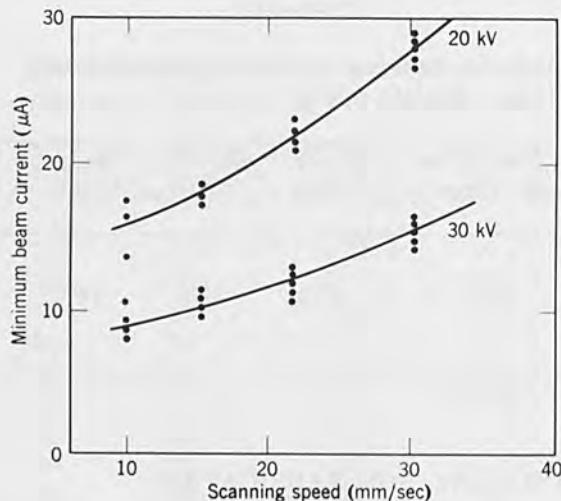


Fig. 3. Relations between minimum beam current and scanning speed, the parameter being accelerating voltage.

In Figure 6, a resistor R is set in a working chamber of instrument, and terminals T_1 and T_2 of the resistor are connected to detector circuit Det . When switch Sw is on, gate G is opened. The rectangular waves generated from a flip-flop circuit FF , which is excited by an original pulse generator PG , are applied to an integral circuit INT . The deflecting voltage of triangular wave form generated from the integral circuit INT is adjusted through the control circuit XC and is applied to the X -axis deflector of the instrument, the wave form of which is shown in Fig. 7a. The original pulse signals are also applied to a step-voltage generator RD and the step voltage generated from it is adjusted through the control circuit YC and is applied to the Y -axis deflector, the wave form of which is shown in Fig. 7b.

The scanned electron beam should be turned off at the point C_2 in Fig. 8, for fear that the electron beam may cut the metal film completely. For this purpose, the negative pulse voltages generated from the original pulse generator PG are applied to the grid of electron gun through a delay circuit DR , the wave form of which is shown in Fig. 7c.

When the resistance reaches the desired value, the electron beam is turned off. The wave form of the voltage applied to

the grid for this purpose is shown in Fig. 7d.

Figure 9 shows a few typical examples produced by this scanning system, in which the accelerating voltage of 30 kV, beam current of 15 μ A, and beam scanning speed of 30 mm/sec are used.



Fig. 4. An electron micrograph of a part of a groove cut by electron beam (accelerating voltage: 50 kV, ac heating).

CONCLUSION AND ACKNOWLEDGMENT

The suitable conditions of electron-beam process and electrical properties of resistors are examined. A special laboratory instrument for fabrication of microelement resistors is designed, which has desirable features of low cost, easy operation, and simple maintenance.

The authors gratefully acknowledge the helpful discussions with Mr. Y. Kondo of Hitachi Central Research Laboratory.

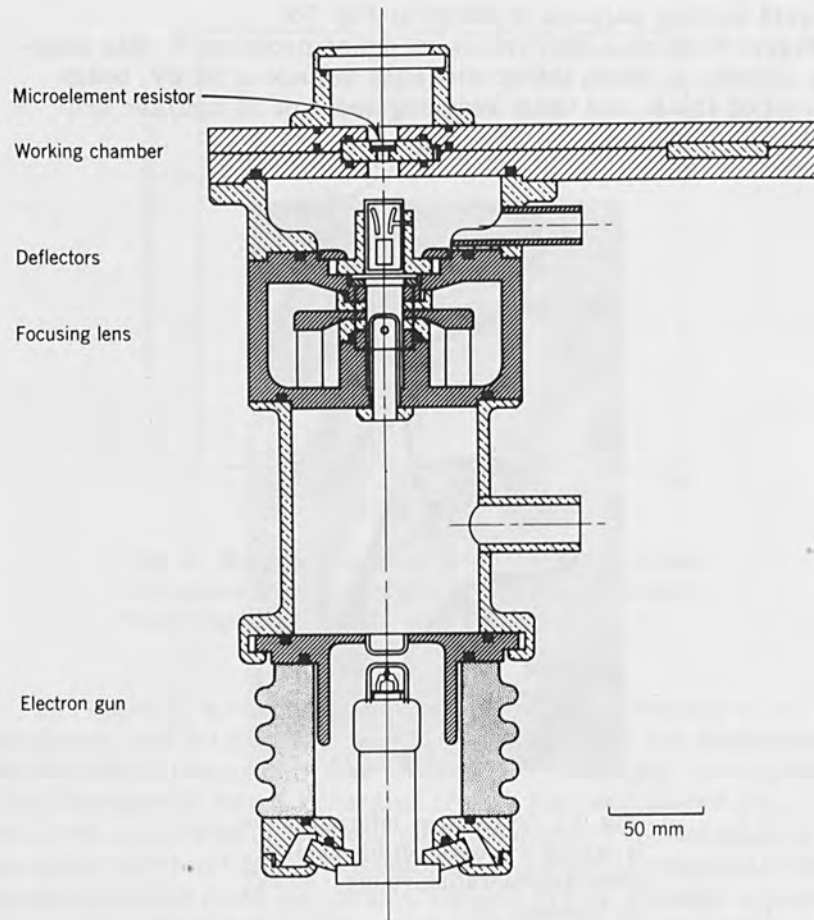


Fig. 5. Cross section of a new instrument.

REFERENCES

1. W. Opitz, Proc. 2nd Symp. on Electron Beam Processes, 32 (1960).
2. R.C.Langford, Proc. Electron Components Conference, 66 (1960).
3. K.H.Steigerwald, Proc. 3rd Symp. on Electron Beam Technology, 269 (1961).

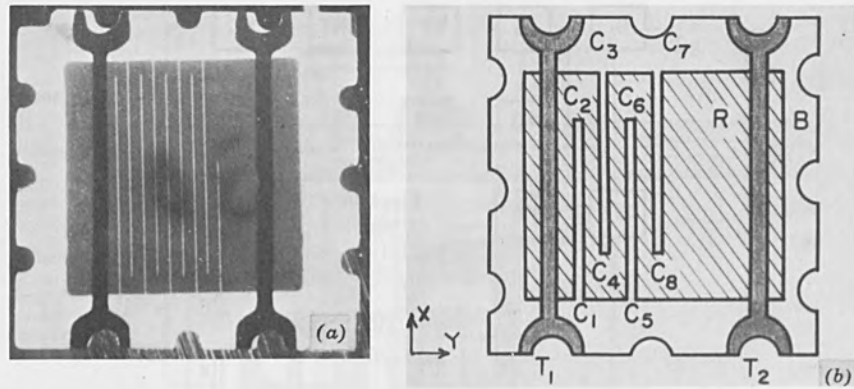


Fig. 8. (a) Microelement resistor produced by electron-beam process. (b) Model of microelement resistor.

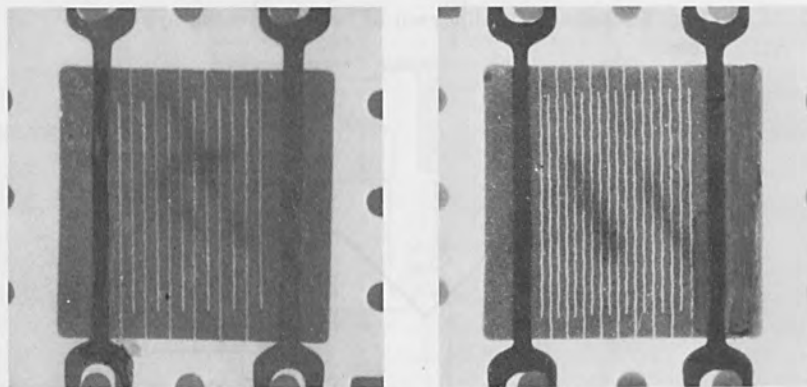


Fig. 9. Examples produced by electron-beam scanning system.

AUTOMATION OF ELECTRON-BEAM PROCESSES FOR MICROELECTRONICS

INTRODUCTION

Electron beams have unique processing and analytical characteristics which are particularly well suited for the fabrication and assembly of microminiaturized electronic systems. Until recently, the majority of the technology was confined prevalently to applied research and development; however, at present, electron-beam processes are being transferred from laboratory to manufacturing environments.

The unique resolution, speed, precision, and maneuverability characteristics of electron beams in vacuum offer many interesting possibilities for increased reliability, economy of fabrication, and volume production through automation. Transition from the laboratory to manufacturing has proceeded more slowly than expected in view of the promised rewards. Until recently, available machines were either of the analytical type, and therefore required extensive modification, or the apparatus lacked the precision, ruggedness, and beam controls requisite for microfabrication.

The progressive decrease in size of microminiaturized components and circuits coupled with the increase in the number of closely spaced inter-connections has added considerable impetus to programs directed toward (1) a better understanding of the basic electron-beam process, (2) design and construction of versatile

* Electronics Department, Hamilton Standard Division, United Aircraft Corporation, Broad Brook, Conn.

† Electronics Branch, Manufacturing Technology Division, A. F. Material Laboratory, Wright-Patterson AFB, Ohio.

‡ Electronics Department, Hamilton Standard Division, United Aircraft Corporation, Broad Brook, Conn.

electron beam apparatus with high resolution and automatic controls, and (3) development of specific techniques for the fabrication and assembly of microminiaturized circuits and devices. Equipment manufacturers are responding to the demands of the industry and providing the required apparatus.

PROPERTIES OF ELECTRON BEAM

Because of some unique properties, electron beams are well suited for use as fabrication and analytical tools. Most important of these, particularly in applications other than thermal processes, is the high resolution and long depth of field obtainable with short wavelength, high-energy electrons. Although machines are not available at present, there is no fundamental theoretical reason why electron-beam equipment for thermal processes should not have the same capabilities as electron microscopes. A second useful characteristic of electron beams is their extreme maneuverability. Electron beams are especially amenable to control and can, therefore, be positioned rapidly and accurately in space and time. This feature makes the apparatus attractive for application to automatic processes. A third factor, important in thermal processes, is the exceedingly high energies which may be transferred from beam-generating equipment to minute volumes of a workpiece. It is not uncommon to obtain beams with power densities of 10^9 watts/cm². Because of the high scattering cross section of electrons in solids, a high-energy transfer efficiency may be attained in a thin layer of material. Electron beams can also be used in a nonthermal mode to catalyze chemical reactions, thus selectively creating or destroying films of one material upon another. Also, due to the charged particle nature and short wavelength of the beam, it may be caused to reflect or diffract from a material. Consequently surface state analysis of materials, as well as the constitution and surface topography, may also be analyzed.

An important attribute of electron beams, which makes them well suited for the processing of microcircuits, is the feasibility of performing the fabrication and analytical functions simultaneously. This factor offers possibilities in improving the reliability of the device and yield of the fabrication processes.

To summarize the properties of electron beams, it may be stated that their unique characteristics permit application as:

1. A means of working material with high resolution, either as a source for the efficient transfer of kinetic to thermal energy, or as a catalyst for chemical dissociations or recombinations.

2. As a method for obtaining information to be used in process quality control by transfer of energy into radiative emissions such as X rays or secondary electrons, or by diffraction or reflection of the primary beam.
3. As a means which combines the properties above and is used to coordinate one high-resolution process with another.

ELECTRON-BEAM PROCESSES

Electron-beam techniques are generally divided into two categories: thermal and nonthermal. Thermal applications are those which rely on transfer of electron kinetic energy to thermal energy within the material. The resulting temperatures are sufficiently high to melt or vaporize the solid, or to alloy or diffuse foreign materials into it. Such thermal processes are useful in fusion welding of leads, brazing of coated lead materials, formation of diffused and alloy junctions, and thermal microetching by evaporation (subtractive process). Localized heating is achieved with stationary microbeams whereas area heating may be obtained by high-speed scanning of an electron beam. In an alternate approach, the entire workpiece may be heated with a low-energy density beam.

Nonthermal applications are those in which the beam is used to activate a chemical reaction. The latter may involve decomposition of an adsorbed layer on a surface, reaction of the surface material with an ambient gas, polymerization of an absorbed material, or activation of a photosensitive material.

NONTHERMAL PROCESSES

Electron-Beam Assisted Chemical Processes

The technical feasibility of nonthermal electron beam procedures other than analysis has been demonstrated by a number of researchers [1], [2], [3], but application of the techniques to micro-miniaturization is proceeding slowly. This may be the result of the requirement for more sophisticated equipment or complexity of the processes. A number of techniques are cited to demonstrate the potential of this technology.

Steigerwald [1] reports that in electron-beam drilling of holes in diamonds, it was necessary to raise the average temperature to the order of 3000° C. He reports that holes can be drilled without

raising the average temperature above 300° C if oxygen is admitted into the area where the electron beam impinges on the diamond. At this temperature, carbon will combine with oxygen to form CO₂ and CO.

Buck and Shoulders [2] and Shoulders [3] report on a process known as electron-beam activated micromachining. The method is similar to the photo-resist technique except for the higher resolution obtainable with electron imaging. In the process, an electron image is projected to the surface of a workpiece covered with a layer of a chemical such as triphenylsilanol. This chemical has the property that a latent image can be produced under electron-beam bombardment. The beam-bombarded layer is converted to silica by heating to 200° C. A gaseous or other etching technique is used to remove the unprotected parts of the workpiece. A resolution of better than 0.1 μ has been demonstrated.

Mollenstedt and Speidel [4] have shown the use of cellulose nitrate films in an electron microscope to record beam widths on the order of hundreds of Angstrom units. Cellulose nitrate decomposes and carbonizes under electron beam bombardment and results in a local reduction of film thickness. The latent image can be printed by standard electron microscope shadowing techniques. They also report on other methods of depositing hydrocarbon films by decomposition with electron beams.

Other electron-beam activated reactions, for example, the decomposition of tetraethylorthosilicate, have been presented by Buck and Shoulders [2] and the decomposition of diffusion pump oil to form insulating layers has been described by Christy [5]. The preparation of thin films of tin and lead by decomposition by electron-beam bombardment of tetrabutyl-tin, tetramethyl-tin, and tetraethyl-lead have been described by Baker and Morris [6].

The feasibility of polymerizing photoresist films by electron-beam exposure has been known for some time. This technique holds high promise of being applied to the production of devices at an early date. It is reported [7] that electron-beam radiation is effective in hardening of KPR films following exposure to an optical pattern and removal of unpolymerized portions.

¹Kodak Photo Resist.

ELECTRON BEAMS AS ANALYTICAL TOOLS

Electron-Beam Microprobe

In 1949, Castaing and Guinier [8] developed a unique analytical tool, the electron-beam microprobe, which makes it possible to obtain the chemical analysis of very small areas (about 1μ in diameter) on the surface of an inorganic sample. The operation of the electron-beam probe relies on the fact that when a specimen is irradiated with a finely focused beam, typically 1μ in diameter, an X-ray spectrum is produced which consists principally of the characteristic radiations of the elements present. The emitted radiation can then be analyzed with an X-ray diffractometer to determine which elements are present from their characteristic wavelengths. The relative intensities of the characteristic radiations are a measure of the concentration of the elements present. Qualitative and quantitative analysis can be obtained of volumes as small as 2 cubic microns with detectable limits for a given element as low as 10^{-14} grams [9]. Thus, the technique is extremely useful in determining which elements are present on the specimen surface as well as detecting local variations in their concentrations after such treatments as localized melting, annealing, diffusion, and oxidation.

Electron Imaging (Scanning-Electron Microscopy)

In this application, an electron probe is made to scan over the object surface in the form of a television raster with the help of beam-deflection elements. The secondary electrons emitted by the object are collected and the current is amplified and directed as an image signal to a television tube which writes its raster synchronously with the electron-probe deflection. The screen then shows a television image of the object surface. The various scanned zones have different powers of secondary emission and this contributes to the image contrast; however, the surface topography is the more important factor. Electron imaging can be coupled with the scanning microprobe output for any one element, thus yielding a composition profile coupled with surface morphology. This is accomplished by the use of long-persistence phosphor screens which retain an image long enough to allow superposition of the concentration profile line scan.

It has also been shown [10] that application of a reverse bias to the emitter junction renders the p - n junction visible. The remarkable features of these experiments are delineation of the junction

and extreme definition of details. The latter is due to the large depth of field of electron beams.

The electron beam also generates a photovoltage across $p-n$ junctions in the device being scanned [11]. This photovoltage signal may be mixed with the normal secondary electron video signal to produce micrographs wherein the edges of the junctions are clearly defined even if the latter is covered by vacuum deposited aluminum conductors.

THERMAL PROCESSES

The ability of electron beams to transfer energy and cause changes in the state of matter have been known for a long time. Early cathode ray tubes were often destroyed by the melting of an internal element or impingement of the electron beam upon the glass envelope. These effects were not very useful but did suggest the use of electron beams as efficient means for the conversion of kinetic to thermal energy. In the past five years, the practical utilization of electron beams as thermal sources has received considerable impetus with the development of electron optical systems that permit the construction of versatile machine tools such as the Hamilton Zeiss electron beam equipment.

The initial applications of electron-beam thermal processes have been concerned with welding of structural members. Almost any combination of metallurgically compatible metals and alloys can be now welded on a routine basis by this technique without degradation of the base metal properties. Materials which could not be joined by the previously available techniques have been electron beam welded into useful structures.

In electronics, the initial applications of electron-beam processes have been in the interconnection of microcomponents and circuits as well as the trimming of thin film passive components to value [12], [13]. The earliest production operation in the electronics industry involved utilization of the process for hermetic encapsulation. In Fig. 1, a group of enclosures are shown which have been joined by the electron-beam process. A typical microstructure is given in Fig. 2.

All enclosure materials of interest to the device industry have been joined by the technique. These include kovar, gold-plated kovar, nickel, gold-plated nickel, copper, copper-clad kovar, as well as many others. The technique appears to be useful for encapsulations involving special problems, such as low thermal input or special

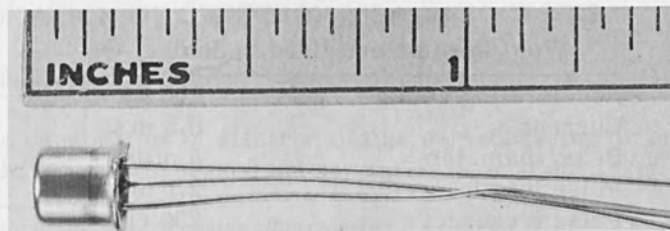
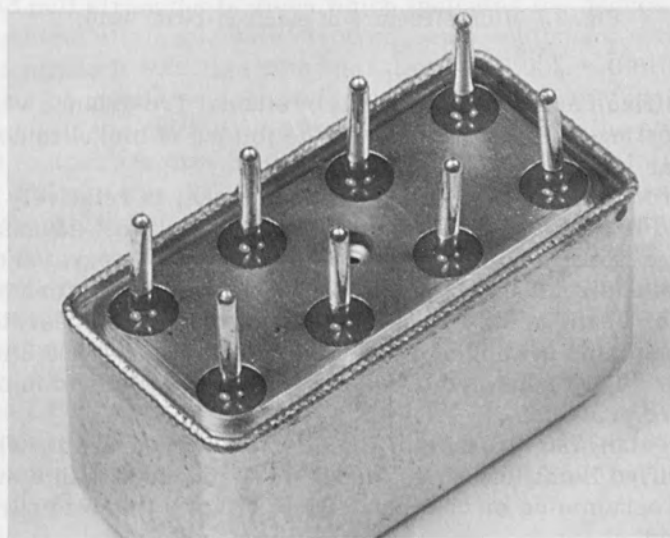
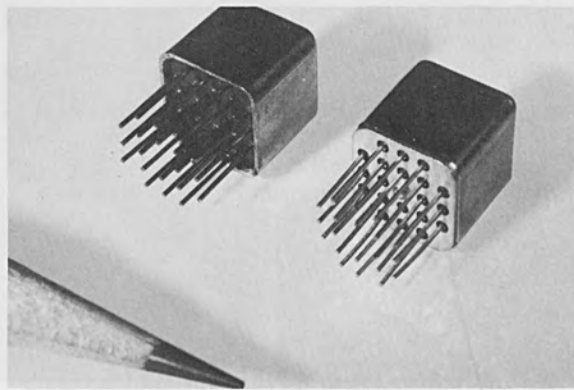


Fig. 1. Electronic devices hemetically sealed by electron-beam joining.



Fig. 2. Microstructure of electron-beam weld.

shapes difficult or impossible to join by standard resistance welding. An interesting application involves the joining of nickel to copper-clad kovar headers.

Electron-beam equipment, presently in use, is relatively slow compared to standard capping techniques and is most advantageously used for special situations, such as sealing of integrated circuit flat packs and rectangular packages, Fig. 1. The joint formed may be a lap or seam weld. Production apparatus has been developed which completes sealing of rectangular packages 0.4×0.8 in. in less than 30 sec; however, welding speeds of 300 in./min or higher are practical.

The rectangular package in Fig. 1 was electron-beam welded in a modified Hamilton-Zeiss Model W1-2 provided with a mechanical programming mechanism. Table I shows the weld parameters used.

Table I
Weld Parameters Used in Study

Accelerating voltage	100 kV
Current	0.5 mA
Beam diameter	5 mils
Pulse length	2.1 msec
Pulse frequency	200 cps
Translation speed	45 in./min

The electron-beam welding of leads for the interconnection packaging of microcircuits is now out of the development stage and moving into pilot production. As shown in Fig. 3, very high joint densities are easily achieved by the process. The assembly in the illustration includes a number of 0.002×0.010 in. OFHC copper ribbons electron-beam welded on 0.025 in. centers to vacuum deposited-nickel electroplated terminations on 0.020-in. thick alumina substrates. A pilot production line which utilizes electron-beam microwelding and sealing techniques is now in an advanced state of development.²

A series of electron-beam microwelding studies carried out under sponsorship of the United States Army Electronics Research and Development Laboratory during the past three years by Hamilton Standard Division, United Aircraft, has demonstrated the reliability of electron-beam microjoining.³ In Fig. 4, a distribution of weld pull strengths is given which indicates the degree of process control attainable with electron-beam equipment without feedback control. It was also demonstrated that 0.002×0.010 in. OFHC copper ribbons electron beam welded to electroplated nickel on molybdenum metallized alumina would have a failure rate equivalent to no more than 2 per 100,000 joints at a 90% confidence level. Failure was defined as a joint strength of less than 200 grams when the weldment was pulled in shear at room temperature. An additional factor of importance in manufacturing is yield. The data accumulated during these studies, as well as the fabrication of many microassemblies, indicate that structure containing 800 joints may be assembled with a yield of greater than 95%. It is interesting to recall that this kind of reproducibility is achieved without feedback control other than normal load and line regulation which is standard in high-voltage supplies. This gives some insight into the potential high process reliability to be expected from electron-beam processes.

AUTOMATION OF ELECTRON-BEAM PROCESSES

Introduction

The capabilities of electron beams as processing or measuring tools have been discussed briefly and it is concluded that the tech-

²Supported in part under contract No. DA 36-039 AMC-03620(E).

³Contracts Nos. DA 36-039 SC-85347, DA 36-039 SC-87301, and DA 36-039 SC-89176.

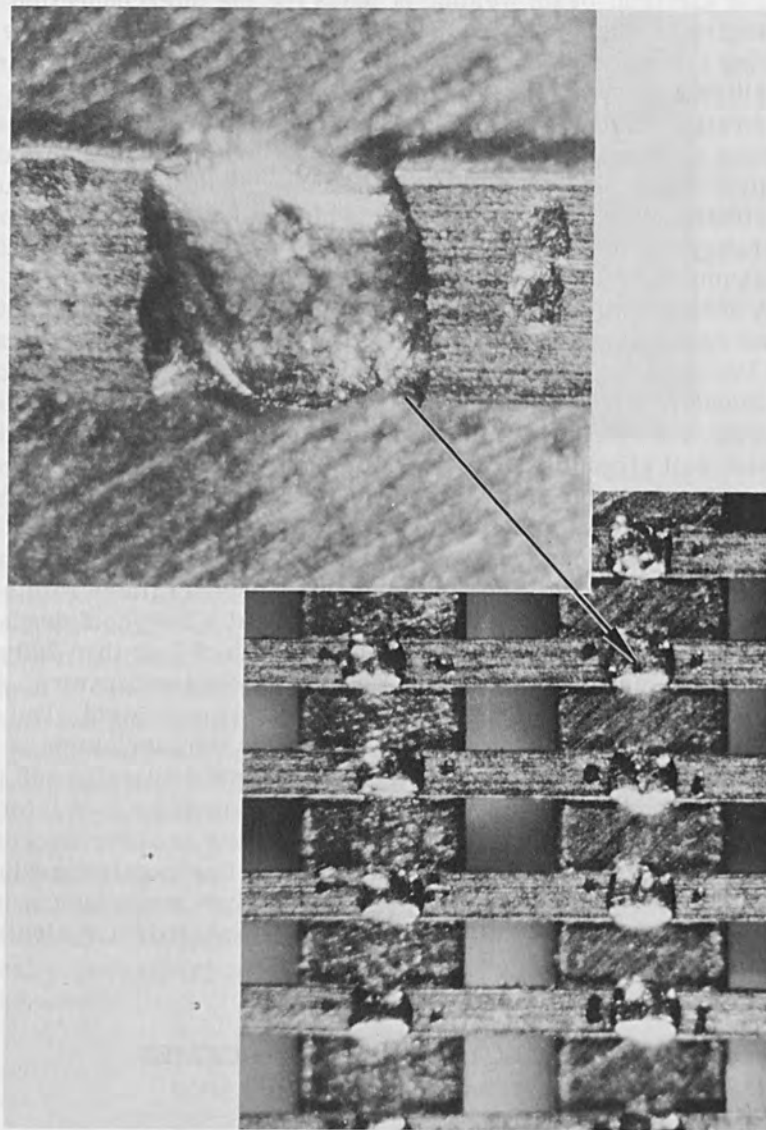


Fig. 3. Copper ribbons (0.002×0.010 in.) electron-beam welded to metallized ceramic wafers.

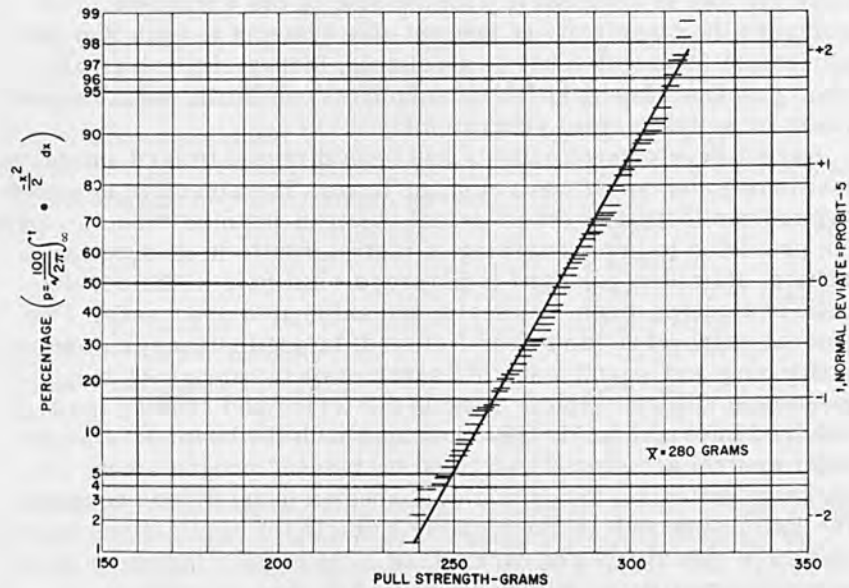


Fig. 4. Joint strength distribution of electron-beam microwelds.

nology has broad application to the microelectronics industry. Various equipment has been used in the development work. High-voltage apparatus, 50 to 150 kV and above; low voltage, about 20 kV average for processing beams; high- and low-energy densities, 10^6 to 10^{10} watts/cm² for working tools, are found to be useful. One factor is implied for all equipment: adequate controls.

The required controls can be categorized in four groups: beam-generation electronics; beam focusing and deflection system; pulse on-off control; and mechanical positioning or auxiliary functions. Beam-generating systems, including the electron optics and electronic supplies, have been the object of considerable development activity for many years. Accordingly, the precision beams needed for analyzing or processing have been available for some time, that is, the long-term stability, load and line regulated power supplies, as well as beam emitters of extended lifetime are sufficiently well developed to permit adoption of electron-beam technology into production environments.

The adoption of electron-beam processes for the processing of

microminiaturized circuits and components is a logical approach since the tool is compatible with the size of the workpiece. Furthermore the complexity of present-day systems is such that the fabrication technique must be a reliable, high-yield, extremely rapid process. Electron-beam techniques can fulfill these requirements by suitable beam programming.

Recent development activity has been directed toward automatic positioning and on-off control of the beam. The objective is advantageous application of the principal electron-beam attributes: speed and precision to the processing of microcircuits in an automated fashion. Hamilton Standard is playing a vital role in the development of electron-beam automatic and automated processes. Two general approaches have been followed: In one instance, automatic positioning and on-off control is achieved by means of a high-speed tape reader which is capable of storing and reading out the required information. In the other approach the beam parameters under control are stored in a fixed or "wired" programmer, i.e., the beam deflection and gating go through a quasi-fixed sequence. The latter approach is particularly suitable for applications where repetitive operations are carried out. The former technique is extremely versatile and will be discussed in the next sections.

The electron-beam automation system, which has been developed at Hamilton Standard, comprises an electron-beam machine which has been provided with suitable beam and worktable programming controls.

The primary equipment of the automation system is an electron-beam machine which has been specifically developed for micro-miniaturization work. The unit, which is referred to as the CW-1, has both welding and cutting or etching capabilities. The machine is illustrated in Fig. 5 and the pertinent characteristics are shown in Table II. When required, the machine is capable of beam-power densities of the order of 10^{10} watts/in² at a beam diameter less than 0.001 in. Table positioning accuracy and beam location control are accurate to at least ± 0.001 in.

As shown schematically in Fig. 6, electrons are produced by heating a 0.010-in. tungsten hairpin filament to approximately 2530° C under vacuum. The flow of electrons from the cathode is controlled by a negative bias voltage. Acceleration of the electrons is provided by adjusting a high potential difference (up to 150 kV) between the cathode and the anode, which is at ground potential. A magnetic lens system focuses the electron stream to form a beam of high-energy density which is deflected by means of a set of coils positioned underneath the magnetic lens. By judicious selection of

Characteristics of Hamilton Standard Micro-Cutter-Welder, Model CW-1

Accelerating potential	50 - 150 kV
Beam current - steady-state/pulse mode	1.00 mA/15 mA max.
Beam diameter - at maximum power	0.001 in.
at lower power	0.0005 in.
Deflection	± 0.250 in. Electronic at high sweep rates
Beam positional accuracy	± 0.001 in.
Table positioning accuracy	0.001 in.
Vacuum	10 ⁻⁶ torr

In actual operation, beam deflection as well as table motion are

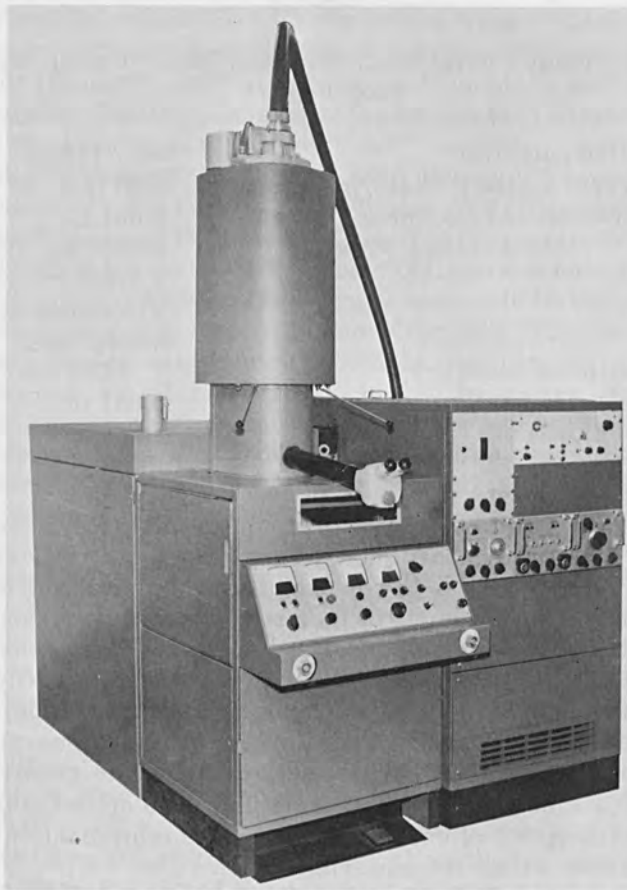


Fig. 5. CW-1 electron-beam machine for microelectronics.

used to carry out etching and microwelding operations over relatively large working areas. In general, table motion is used to accomplish macromotions, whereas beam deflection is suitable for precise operations within a scan area ± 0.200 in. of the optical axis of the electron beam. Larger deflections (± 0.500 in.) can be used with a lower degree of accuracy.

The worktable control system is a purchased semistandard, two-axis machine tool positioning control which is equipped with its own slow speed (20 characters per second) tape reader to provide a series of table positions as called for by the high-speed automation



1000

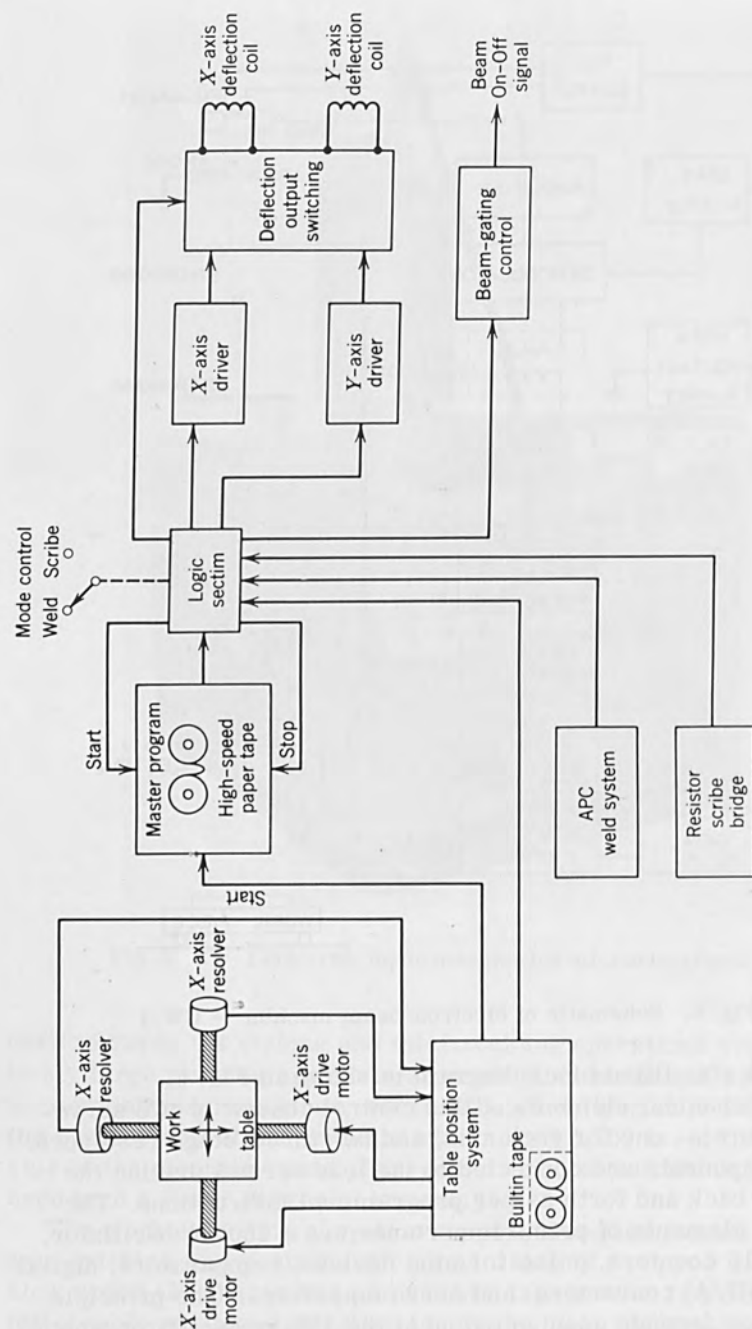


Fig. 7. Electron-beam automation system — block diagram.



Fig. 8. CW-1 electron-beam machine and automatic controls.

is first divided by 1000 using three decimal counters to produce a 250-cps signal. This signal is then synthesized into two two-phase voltages and is used to energize the three-stage (coarse, medium, and fine) resolver geared to each driver lead screw. Rotation of the lead screw produces varying phase differences that are applied to one input of the phase comparators. The other input to the comparators is obtained from secondary sets of decimal counters capable of being preset to number sequences set up on the tape program. The input to these counters is the same 250-kc oscillator driving the resolvers. In operation, when the phase difference created by the presettable divide by 10 counters differs from the phase difference produced by the resolvers, the comparator produces an error signal that when processed through a D/A converter and applied to the servo-amplifier causes the table driver motor to seek a null condition.

Program resolution to 0.0001 in. is possible with this machine,

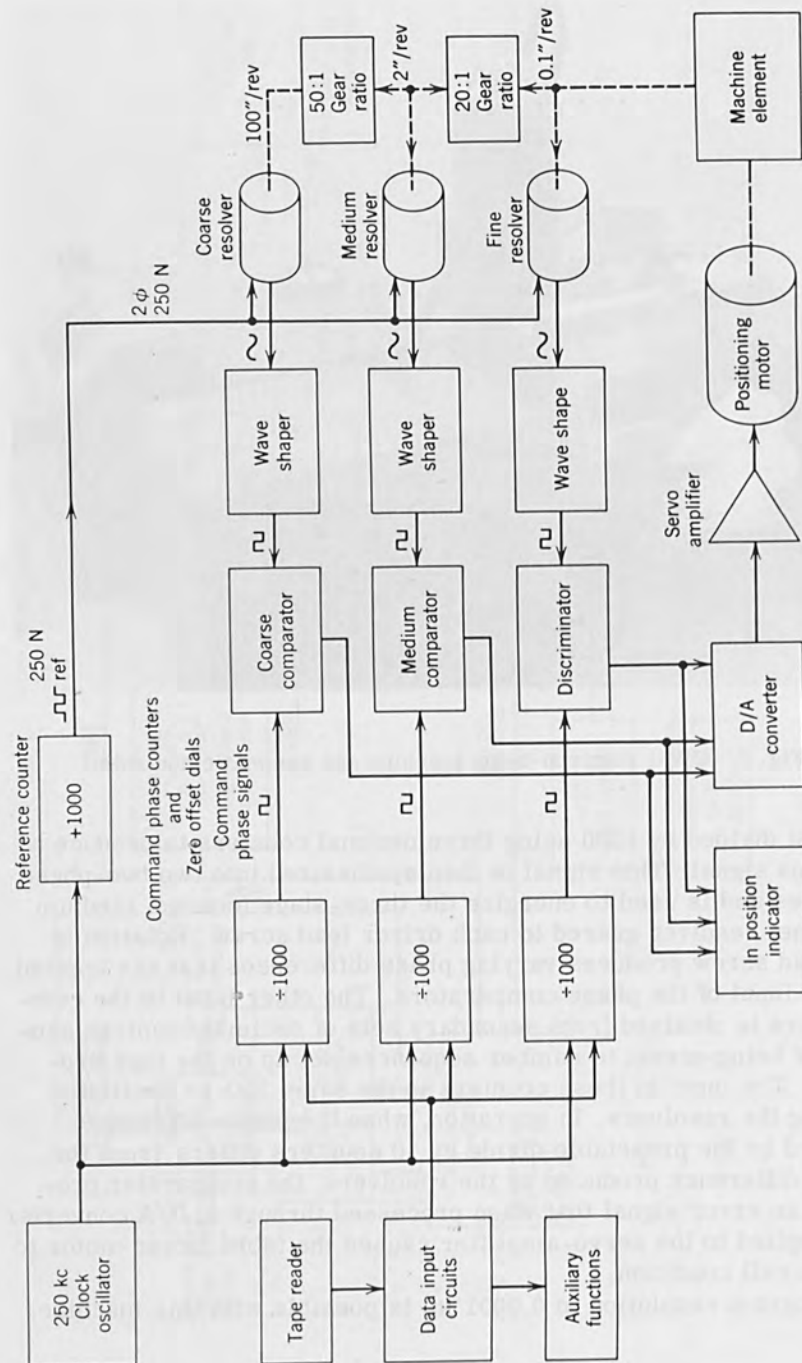


Fig. 9. Schematic of work table position control.

though in practice with reasonable mechanical implementation of lead screws, etc., actual positional accuracies of 0.001 inch are more realistic.

The electron-beam automation system comprises two separate programming subsystems which respectively control the scribing and microwelding processes. Both have been combined in one control system.

SUBTRACTIVE PROCESSES

One function accomplished with the equipment is automatic positioning and machining of discrete patterns. Formation of parallel staggered lines is accomplished by applying a current sawtooth generator to one deflection coil axis and a series of dc levels to the other. Control of the beam on-time in relation to the sawtooth component establishes the point on each line where the machine action starts and stops.

This process is used to scribe area thin-film resistors to value and also to define isolation of areas on metallized substrate to obtain discrete conductive paths.

The schematic in Fig. 10 illustrates the automation system with particular reference to the machining mode of operation. The process starts with an input to the table-positioning system to bring the specific area to be machined under the optical axis of the electron beam. When a working position condition has been reached, the table-positioning system stops and activates the high-speed tape reader. The latter immediately goes into a slewing mode, creating a series of alpha-numeric characters that pass from the input register into the sequencer, through to the data registers and the auxiliary switching functions. Three analog outputs are provided to accomplish the following functions: (1). Establish the value of dc deflection current, (2) Define the point in the sawtooth scan where beam-on occurs, (3) Define the point for beam-off. Each of these variables is programmed. In addition to the above primary functions, information is also stored to define the polarity of the dc deflection and on which axis the deflections are to take place.

A grid was chosen to "map" the area, Fig. 11, to be cut by the programmed beam. By plotting deflection current on one axis, and saw-tooth current on the other, it is possible to arrive at a basis for programming a system to fulfill the desired result. From inspection of Fig. 11, it is seen that a program possessing the format: deflection axis; deflection current, Sawtooth "beam-on" current;

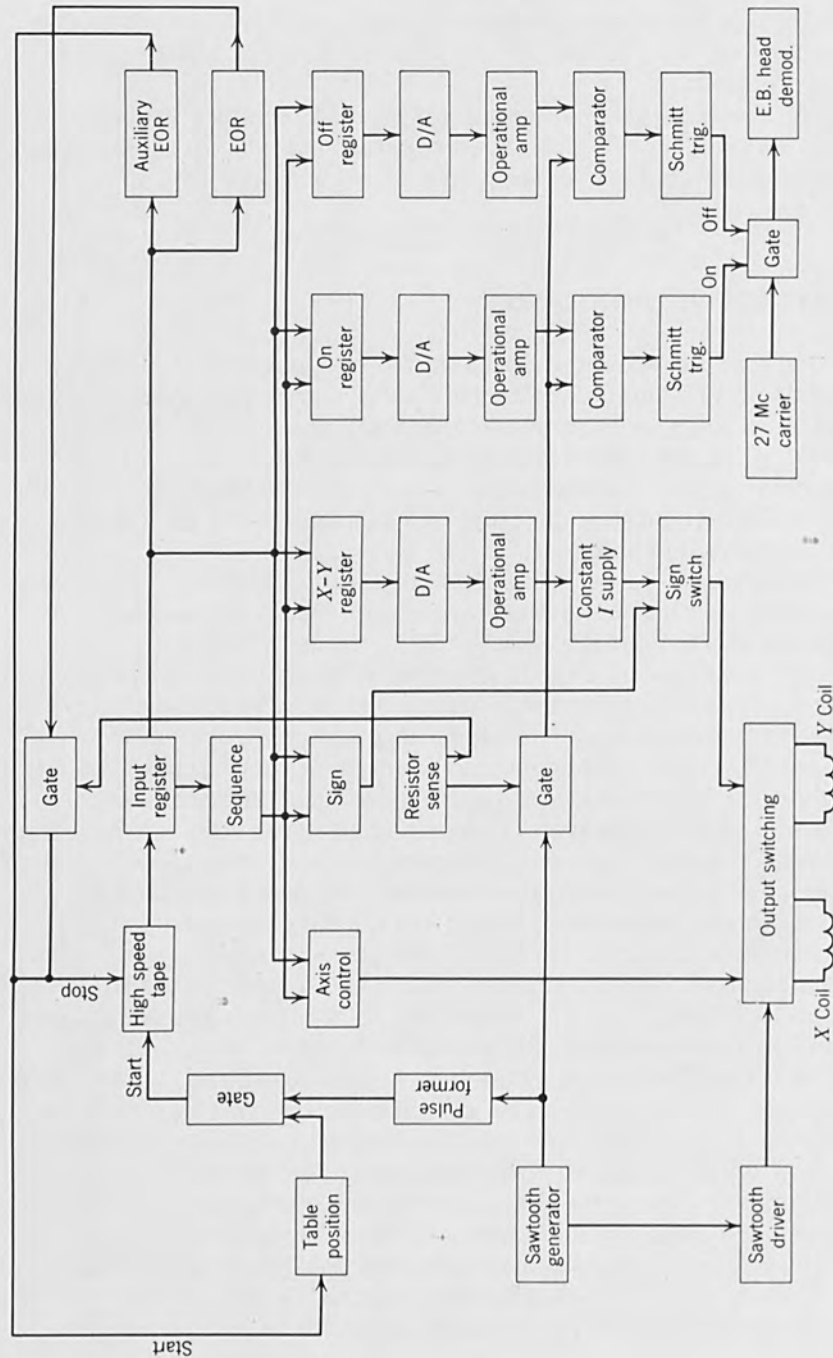


Fig. 10. Automation system — scribing mode — block diagram.

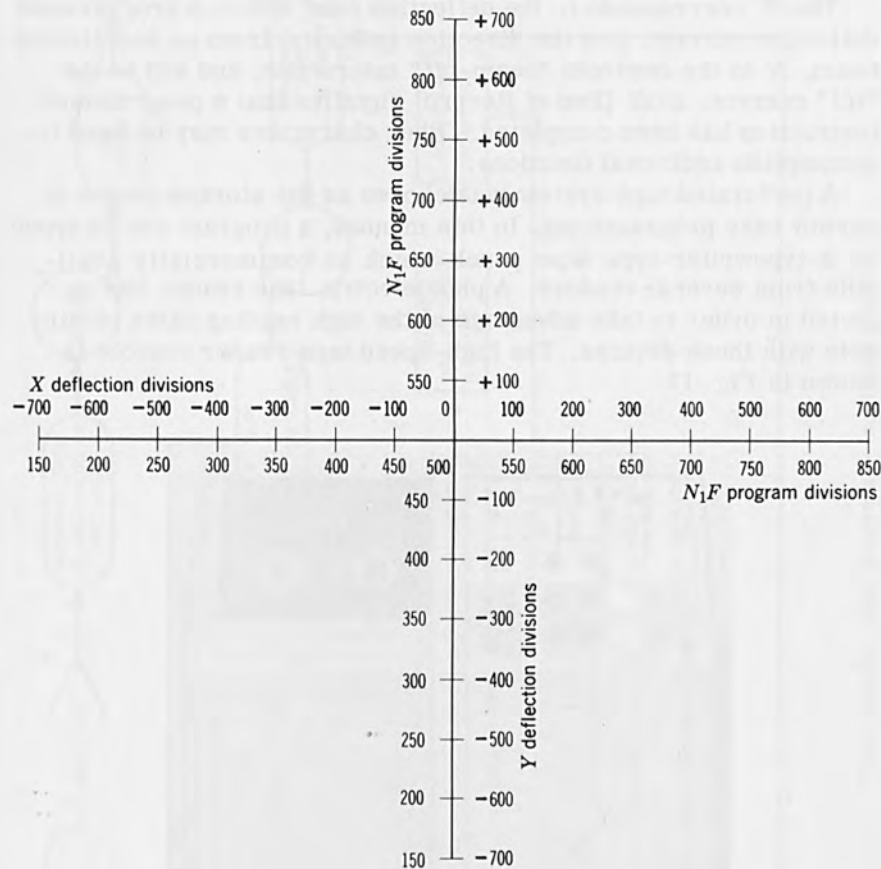


Fig. 11. Programming map for automatic scribing.

sawtooth "beam-off" current; would accomplish the desired result. Thus, a system possessing this capability was designed. A system using letter identifiers for the various programmed commands and *BCD* (binary-coded decimal) numerics was chosen as a system able to meet the accuracy requirements and yet be economical to build. A typical program instruction appears as:

X250 +, N 175 -, F 550 +, EOR

The *X* corresponds to the deflection axis, 250 to a programmed deflection current, + to the direction (polarity) from an undeflected beam, *N* to the sawtooth "beam-off" instruction, and 550 to the "off" current. *EOR* (End of Record) signifies that a programmed instruction has been completed. Other characters may be used to accomplish additional functions.

A perforated tape system was chosen as the storage device to permit easy programming. In this manner, a program can be typed on a typewriter-type tape punch which is commercially available from several vendors. A photoelectric tape reader was selected in order to take advantage of the high reading rates obtainable with these devices. The high-speed tape reader console is shown in Fig. 12.

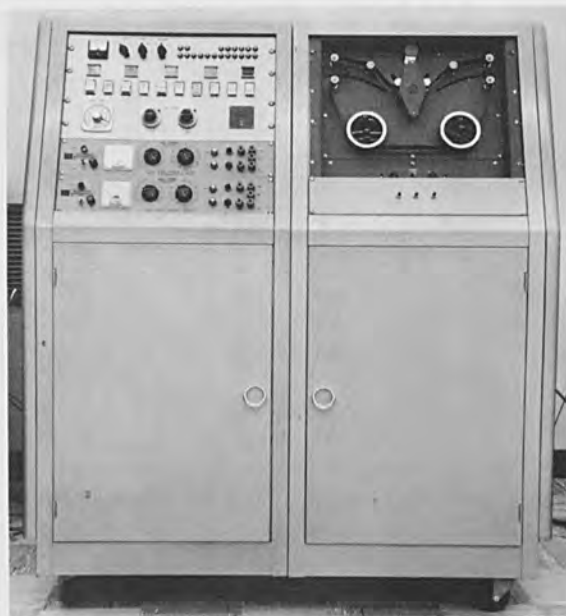


Fig. 12. High-speed tape reader console.

A flow diagram of the tape control system is shown in Fig. 13. Initially, after the power has been turned on and the system has been cleared, the tape is advanced to the beginning of its program.

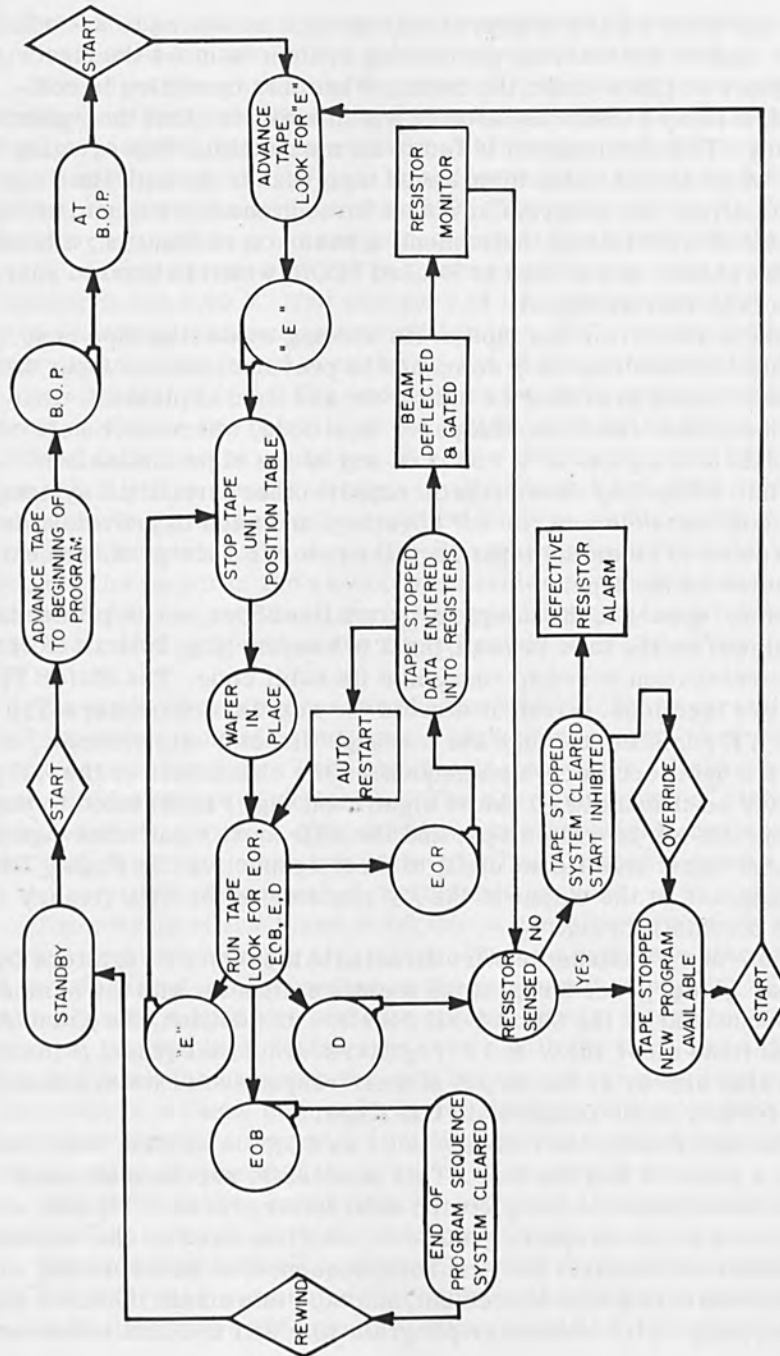


Fig. 13. Tape control flow diagram - scribing mode.

When the start switch is depressed, the tape advances to an "E", which signals the external positioning system to move the first workpiece in place under the beam. When this operation is completed, a relay closure is effected which indicates that the wafer is in place. This information is fed to an monostable, thus creating a pulse which is fed to the high-speed tape reader through the Tape Start Control. The program advances through one instruction, which consists of a deflection instruction, a beam-on instruction, a beam-off instruction, and an End of Record (*EOR*) which is used to energize a tape restart signal.

Data is read from the tape in its slewing mode (the tape runs free until an external stop command is provided) into an input data register, which provides the character and its complement. Programming uses the 8-bit EIA paper tape code, where each hole corresponds to a logical 1. A "one" on any of the eight channels is read into a flip flop where it is directed to other parts of the system. The eight channels are "or'ed" together, and used to provide a delayed pulse to clear the input register prior to receipt of the next character on the tape.

Referring back to the sample program line above, action proceeds as follows: as the tape is read, the *X* is sensed, (Fig. 10) by a *NOR* gate combination wired to recognize its 8-bit code. The *X* (or *Y*) does two functions. First, it directs the next four characters (250+) to the *XY* register, through the message director and secondly, it clears a 4-bit counter which sequences the characters to the *XY* register so that the *MSD* (Most significant digit) is directed to the appropriate set of 4 flip flops, and the *NSD* (next significant digit) and *LSD* (least significant digit) to their respective flip flops. Thus, data appears at the output of the *XY* register in the *BCD* (binary coded decimal) format.

The *N* and *F* instructions are directed to the *N* and *F* registers by similar gating procedures in the message director and the numbers are sequenced by the same 4-bit counter. In addition, the *N* and *F* instructions clear the *N* and *F* registers. Thus, the *N* and *F* instructions also appear at the output of their respective registers in a *BCD* format at the output of 12 flip flops.

The tape moves continuously until an *EOR* is sensed, which provides a pulse to halt the tape. This process is repeated as many times as necessary to complete the subtractive process. The data are sequenced by the sawtooth generator, which is used as the master clock source to control the scrubbing sequence, so that scribing will be inhibited during tape movement, and tape movement inhibited during scribing. A 16-character program line will use about 60msec.

Since a 20-cps sawtooth generator is employed, 2 sawtooth periods are reserved for data entry, and one is used for scribing.

The output of each of these registers (XY, N, F) is fed to a digital-to-analog converter, which converts the *BCD* number to an analog voltage proportional to the number. This output is fed to a summer amplifier which will give an output voltage from 0 to 9.99 V as the program varies from 0 to 999. The output of the XY D/A-summer is used to program a constant current power supply, which gives currents from 0 to 200 mA corresponding to voltage inputs between 0 and 9.99 V. The accuracy of the voltage setting of the D/A converter and summer amplifier is $\pm .25\%$. Actual load current changes measured over the period of an hour showed drift and changes within 0.05%. The programmed sign is used to energize a relay to change the polarity of the output current, so that a total of 400 mA deflection is possible. Another relay is energized by the output of a flip flop which indicates whether X or Y has been selected, and directs the output of the constant current supply to the X or Y coil, respectively. The same flip flop energizes another relay which directs the output of the sawtooth generator to the coil which is not connected to the constant current supply. Thus, a linear sweep is automatically provided on the opposite axis from the programmed deflection.

The sawtooth driver consists of a low-level Miller integrator-type circuit which is used to program a high-gain, closed-loop, constant current power supply. This combination produces a linearity of 3% over the full range of deflection. The use of this type of circuitry insures the absence of discontinuities in slope which may occur at the 0, 0 point with other type drivers. Thus, a constant velocity and a uniform rate of removal of material is assured.

The outputs of the N and F registers are also directed to D/A converters and summer amplifiers. These amplifiers also give a 0 to 9.99 output corresponding to the programmed number. The outputs of these amplifiers are then compared with the sweep sawtooth current (sensed by a resistor in series with the coil) and a pulse is created when the sawtooth exceeds the programmed voltage by 5 mV. The outputs of these comparison circuits are then gated into the set and clear inputs of the beam control flip flop to provide a level for the duration of time that the beam is to be turned on.

This level is then gated with the output of a pulse generator provided as an integral part of the electron-beam machine.

The pulse generator consists of a string of counters driven by a 1 Mc oscillator, connected with appropriate gating to produce pulse widths from 4 to 64 msec, at pulse repetition frequencies from

16 KC to 0.1 cps. A continuous beam is also available. By varying pulse-rate frequency and pulse width, the operator has a wide range of settings to use in controlling the beam intensity and, therefore, the rate and depth of material removal. In a typical operation, successive lines follow each other as the tape program dictates with each line activated by the last end-of-file mark together with the flyback transition of the 20-cycle sawtooth timing generator. Definition of conductive paths on any one area is accomplished by beam deflection according to tape program. In the case of resistors, scribing continues until the total wafer resistance equals a predetermined value. The value of the resistor being scribed is measured continuously by means of a bridge circuit adjusted to the value of resistance desired. When the bridge balances, a pulse is sent to the beam-control flip flop, clearing it, and inhibiting further scribing until the end of the program. The resistance monitor reacts with the beam control in $0.2 \mu\text{sec}$, insuring resistor accuracies within $\pm 1\%$. In order to permit automatic scribing of more than one resistor per substrate in conjunction with the resistance monitor, provisions have been incorporated into the automation system to provide a programmed restraint of the table positioning independent of the normal scribing information. This feature was implemented by means of a new decoder function that recognized the letter D punched on the tape at any point. This improved design of the auxiliary function separates the beginning of each resistor program from a mandatory table reposition command. The additional capability permits scribing of conductive patterns on large substrates by electron beam scribing in the deflection mode followed by table motion to the center of the next area (up to 1×1 in.) to be scribed. If a program is completed prior to the sensing of a resistor, an alarm light is illuminated, and the system is stopped from further action until an override switch is depressed. This enables the operator to correct any malfunction which may have caused the alarm. Thus, the use of these command functions enable the programmer at his option to cut multiple resistors on one wafer, or cut multiple resistors with a single automatic sequence, while maintaining the flexibility to stop after a single resistor. At the end of a series of programs, an *EOB* (End of Block) character is used to finally clear the system. The tape is then normally rewound to the starting point where either the sequence may be repeated or a new tape inserted.

In addition to scribing of discrete conductive paths, resistors and capacitors, the system may be used to drill fine holes on a grid matrix on metals as well as ceramics.

MICROWELDING PROCESSES

The system is also designed to carry out microwelding operations in an automated fashion. The inherent flexibility of the overall system permits microwelding in any one or combinations of the listed modes: (a). programmed worktable, (b). programmed electron-beam deflection, (c). combination of a and b, (d). adaptive positioning control (APC)⁴, and (e). combination of above approaches.

In the first technique the table is programmed to bring the elements to be joined under the electron-beam axis wherein the beam is activated and welding is carried out by preprogrammed beam gating and deflection. This method is relatively slow and does not take advantage of the inherent high-speed capabilities of electron-beam processes. In the second approach, a number of welds within an area is made by beam deflection, i.e., the beam is imparted a dc deflection, with a superimposed ac deflection program to assure a relatively large fusion zone.

Programmed microwelding proceeds as scribing except for a slight physical modification to the system. One of the deflection coils is disconnected so that the sawtooth generator does not cause the beam to sweep. Deflection is programmed in the other axis as before, but the sole purpose of the sawtooth generator is to provide a time reference for determining the length of the weld. The difference between the on (N) and off (F) current values can be equated to a percentage of the sweep times. Thus, a difference of 300 corresponds to a weld time of $.3 \times 50 \text{ msec} = 15 \text{ msec}$. A wider weld spot can be obtained by an additional modification, consisting of presenting an effective short to the sawtooth generator and applying an external sinewave to the other deflection coil. This "scrubs" the weld zone when the beam is turned on.

Welding by the APC technique uses many of the tape control and programming techniques discussed under scribing; however, some points will be repeated as they apply to the present discussion.

The concept around which the APC technique has been developed depends on the electron beam to perform a double function. The first function is locating the leads to be welded, whereas the second activity is welding of the lead to the substrate. Prior to fixturing and loading within the machine, a fluorescent dye is applied to the leads. In actual operation, a low-energy beam is scanned digitally along a preprogrammed pattern. At some point it strikes the lead in that zone, thus producing a light output, Fig. 14. The light is

⁴Developed under Air Force Contract No. AF 33(657)-8790.

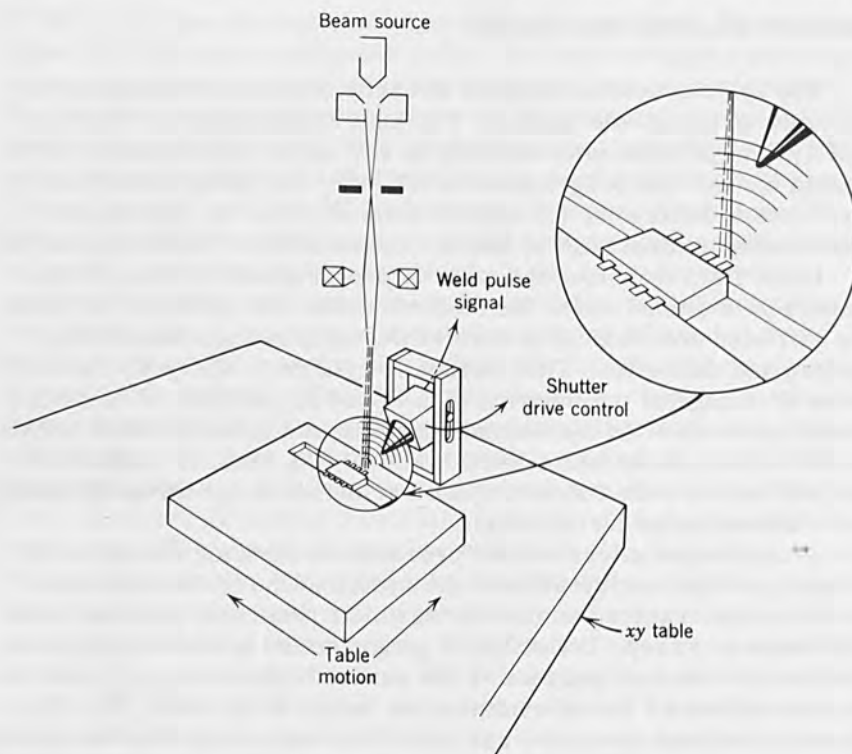


Fig. 14. Conceptual illustration of APC control operation.

sensed by a photo cell which immediately initiates a series of events to prepare for welding. A pulse is generated which triggers a flip flop controlling the tape unit, halting further data entry. Since the light output is first sensed at the edge of a pin, an additional amount of dc deflection is programmed to move the beam to its center. Next, an additional deflection component normal to the first is applied to assure that the weld will not take place on the extreme edge of the pin. Both of these dc offsets are adjustable up to 50 mA by means of panel controls and are switched in by means of a relay driven by the above flip flop. Welding of the leads is carried out by a pre-programmed ac deflection which is also switched in by the relay. This welding pattern is formed by a sine wave in one axis and a saw-tooth in the other, and is generated to produce or "scrub" a weld over an area up to 10 mA deflection larger than could be produced by a

stationary undeflected beam. The last operation prior to pin welding is a change in beam power from a scanning level to a welding level. At this point, the weld timer is activated to control total welding time. A light is attached to the timer which indicates during the duration of the weld. Upon completion of the weld cycle, the beam intensity reverts back to the scan level, the dc offset currents and ac deflection patterns are discontinued, and the high-speed tape runs to the next end of block in preparation for start of the succeeding pin-sensing pattern.

A block diagram, Fig. 15, is presented to show the implementation of the APC welding mode. In general, the operation is quite similar to scribing; however, the main difference is that dc deflection is used in both axes. In this instance, the tape reader fills two data registers which, in turn are translated to the D/A converters, current drivers, and into *X* and *Y* axis deflection. One set of coordinates after another is programmed in typical raster pattern to cover an area at least as large as the expected positional tolerances of the pins. Pin sensing and weld will, therefore, be expected to take place well before the entire raster has been scanned.

Typical time required per coordinate set during stepping is approximately 20 msec. Weld on-times can be adjusted throughout the range of 5 to 500 msec. DC offset and ac deflection currents can be varied from 0 to 25 mA in either axis. Test results of the analog circuits reported in the scribing description apply directly to the welding mode.

The flow diagram which describes the tape operation for welding is shown in Fig. 16. The tape unit is advanced to the Beginning of Program and after the Start switch is depressed, the character *E* is read which signals the table-positioning system to move the table, also under command of a tape reader.

After the table is in position, the scanning operation discussed above begins. As the tape runs, data on a program containing both *X* and *Y* deflection information enters. A 60-cps clock, formed from the line frequency, is enabled. Once started, the tape slews until it recognizes an *EOR*, which momentarily stops the tape until the next 60-cps start pulse. In the slewing mode, the tape runs at 400 cps, so the first location is reached after 10 characters have passed (both *X* and *Y* instructions) or in a 25 msec. Thereafter, each new location is reached in 12.5 msec until the end of a line (when an *EOR* is sensed). After the tape stops, a maximum of 16.6 msec (the period of the 60-cps clock) transpires until the next start pulse from the 60-cps clock. At this point, the operation is repeated. During the duration of the actual welding described below, the 60-cps clock

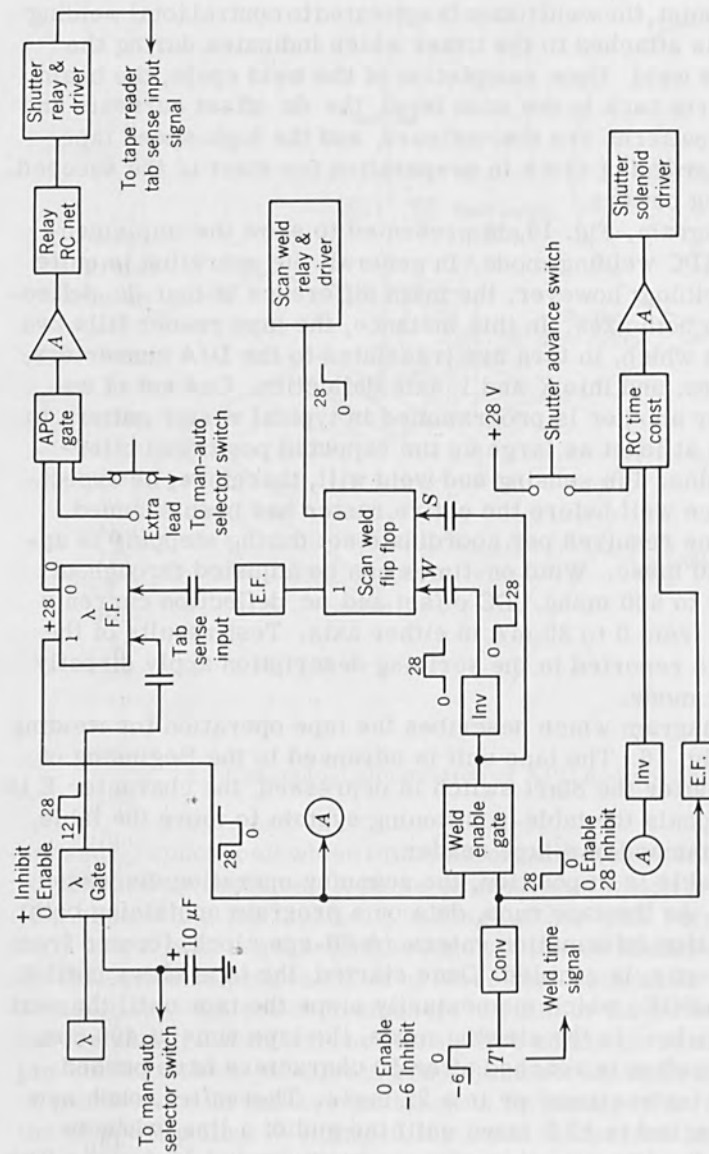


Fig. 15. Automatic positioning control (APC) for electron-beam welding — block diagram.

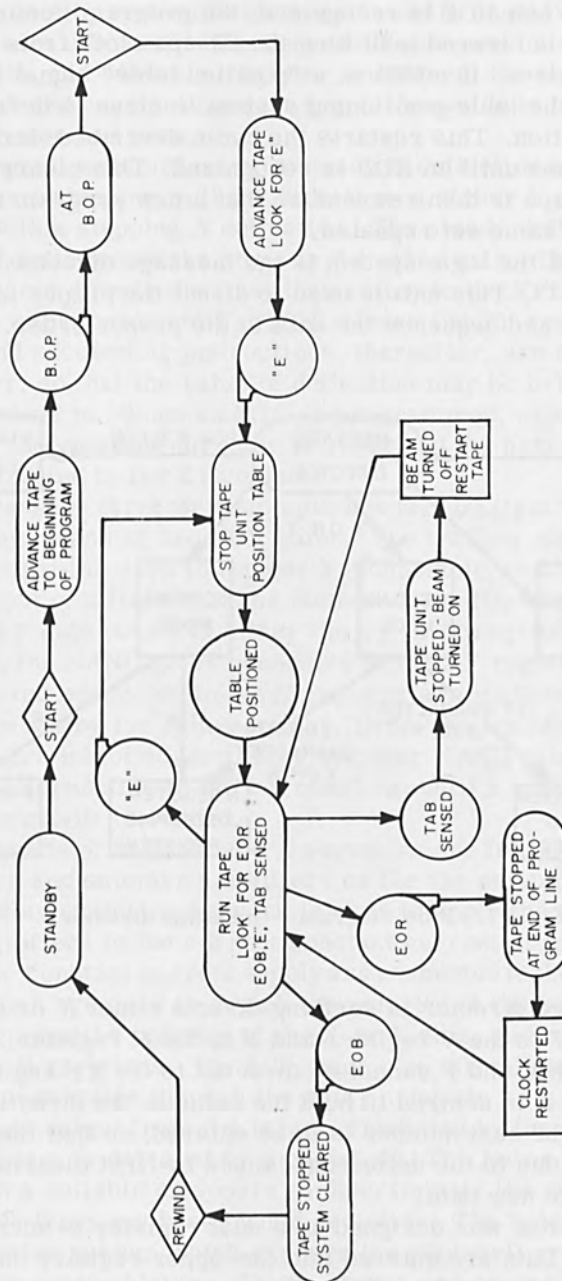


Fig. 16. Tape control flow diagram — APC welding.

is inhibited. When an *E* is recognized, the program is completed, and a flip flop is cleared inhibiting the 60-cps clock from sending future start pulses. In addition, a "position table" signal is generated, causing the table positioning system to move to the next programmed location. This restarts the cycle described, and the sequence continues until an *EOB* is recognized. This clears the system, and the tape is then rewound so that a new program may be inserted, or the same set repeated.

The heart of the logic system is the message director and sequencer, Fig. 17. This unit is used to direct the proper instruction to its register and sequence the data in the proper order.

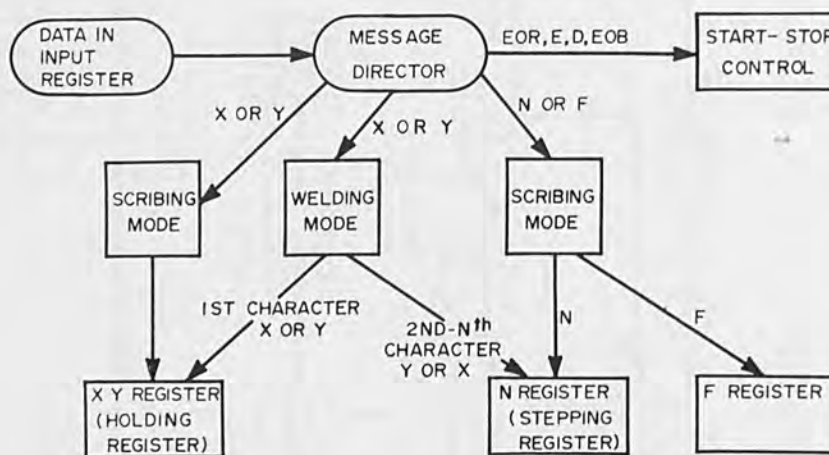


Fig. 17. Flow diagram — message director.

The message director in scribing directs either *X* or *Y* to the *XY* register, *N* to the *N* register and *F* to the *F* register; however, in welding both *X* and *Y* cannot be directed to the *XY* register. Furthermore, it is desired to hold the value in the direction to be stepped until the next number is to be entered, so that the beam will not skip a tab due to the deflection caused by first clearing and then re-entering the new data.

The *N* register was designed as a dual register to accomplish this purpose. Data are entered into the upper register one digit at a time as described above. When the sign bit appears, the data in the upper register (3-bit number) are gated simultaneously into the

lower register by means of a monostable and NOR gate arrangement. The previous information is cleared 10 μ sec previous to the arrival of new data, and while the register is cleared, the combined effect of the D/A and constant current supply response is to ignore that the N register has been reset.

In a typical weld program, a steady N deflection is desired, with a continuous stepping in the Y direction; a steady Y deflection is desired with a stepping X deflection. The steady deflection is programmed, followed by the steps in the other axis. Thus, the message director directs the first instruction to the XY register, which holds the information until an EOR clears the XY register. The second and succeeding instructions, thereafter, are directed to the N register, so that the value of deflection may be held until the new value is gated in. When an EOR is programmed, signifying the end of a line, the message director is reset and the next instruction will be directed to the XY register.

The message director accomplishes this by first "oring" both X and Y by means of two NOR gates. The trailing edge of the output of this gate is used to trigger a monostable which is applied to the set input of a flip flop. The outputs of the flip flop are used to gate the XY gate output to either the XY or N register. Prior to being set, the NAND gates connected to the XY register are enabled, and the gates connected to the N register are inhibited. When the flip flop is set by the delayed pulse, (from the training edge) the XY gates are inhibited, and the N register NAND gates are enabled. The flip flop remains in the set condition until a clear pulse is received when EOR is sensed.

The outputs of both XY and N registers are fed to the same D/A converters and summer amplifiers as for the scribing mode; however, a relay switching network is used to apply the outputs of these amplifiers to the correct constant current supply. The outputs of the constant current supply are connected to the coils through relays which are used to switch the polarity of the output current depending upon the polarity of the X or Y axis, and whether it is in the XY or N registers. The full range of ± 200 mA is possible from the current supplies through the relay network.

The light output from the target irradiated by the low-intensity electron beam is detected by a photocell. The latter has been provided with a suitable enclosure to discriminate the output of the target area from any background radiation. The holder is equipped with a shutter system which exposes the photocell sensing surface to the area scanned by the electron beam and covers it during the welding operation. The shutter protects the photocell from the high-

intensity light emanating from the weld zone and prevents any potential deposition of vaporized material on the photocell sensing surface.

Mechanically, the photocell assembly is mounted within the electron-beam machine with its sensing surface perpendicular to the fluorescent source, Fig. 18. A photodiode element has been

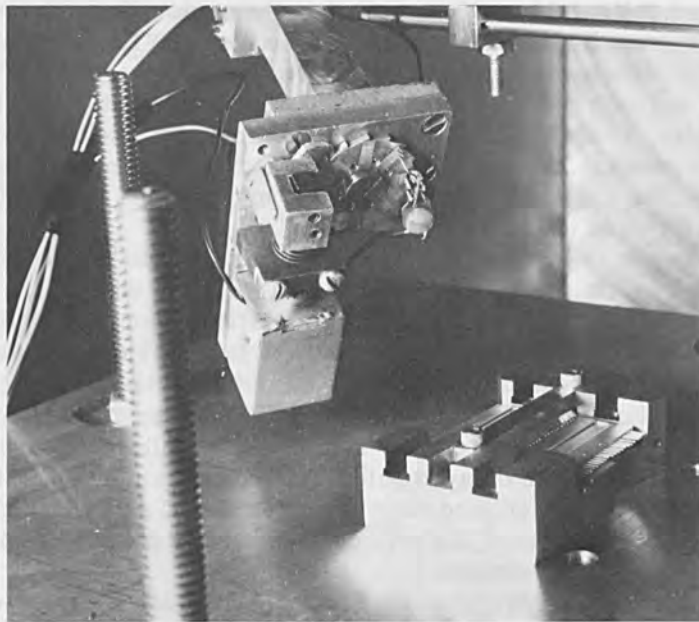


Fig. 18. Adaptive positioning control — photocell mounting and shielding.

selected which is particularly sensitive to the emitted radiation. Since the target is relatively small, for instance, a 0.010-in. wide ribbon, it is required that the photocell assembly be properly oriented relative to the target.

Photodiode response times have been determined and the measured rise times were from 0.005 to 0.030 sec, depending upon the intensity of the illumination reaching the photocell. A decay time of 1.5 sec was needed to afford complete recovery of initial dc test levels. Thus, the between weld time is approximately 1 sec to assure a suitable pulse.

Electronic Set-Up — APC

Since the photodiode is capable of delivering only low-current signals and in view of the fact that relatively high voltage are required for a useable output current level, a special matching network was designed and assembled. This permitted changing of the supply voltage level of operation and matching the $1\text{-M}\Omega$ load resistor necessary for good response time without loading the available current signal.

The change of photodiode current measured with a 90-V supply voltage across the photodiode in series with a megohm resistor is 40 mA maximum with an initial level from 4 to 12 mA due to background light absorption. The current is fed into a Darlington amplifier with a supply voltage of 90 V and impedance of $1\text{ M}\Omega$ to a source impedance of $100\text{ k}\Omega$. A discriminating type of network is used to eliminate the background noise level from the output signal delivered by the photocell system. Consequently, current signal level ranges from a minimum of approximately 15 mA to a maximum of 62 mA. Any level above 15 mA causes a step change at the output of the photocell driver system.

The signal current through the $100\text{ k}\Omega$ emitter resistor of the 90-V Darlington amplifier to the base of the following matching stage is limited with a series limiting resistor such that the voltage at the base of the following stage is less than the desired 28 V dc supply voltage used throughout the APC system. This change of supply voltages was decided upon to lower power dissipation; however, it does not affect the ratio of input-to-output signal except to limit it to the 28-V dc supply level.

The final design of the photocell driver system incorporates a 28-V Darlington amplifier with a $1\text{-k}\Omega$ emitter resistor to provide an output signal with a low-impedance source, and a $1\text{ M}\Omega$ input impedance to dc signal levels.

The output signal is fed into a differential amplifier which performs the function of increasing gain, providing for a dc level drift stability, and discrimination of the useable signal from the background noise level.

During actual runs, the operator obtains a null reading across the inputs of the differential amplifier by setting the reference control pot. A safety factor of -6 V is specified in the setting and any pulse obtained from the photocell driver in excess of the -6V dc level causes a step change of + 14 V. The latter photocell memory flip flop, closes the shutter over the photocell, and enables the APC gate to deliver a pin sensed signal to the associated output signal drivers.

APPLICATION OF AUTOMATED PROCESSES

The electron-beam machine (CW-1) coupled with the tape programmers and adaptive positioning control is a fully automatized system for the fabrication of microcomponents and their interconnection and packaging into circuits and equipments.

This system has been applied to the fabrication and assembly of an interconnection packaging structure⁵ upon which semiconductor as well as thin-film circuits are mounted and interconnected. The object of this study was the development of interconnection and joining techniques which do not unduly compromise the inherent reliability and packaging density of semi-conductor integrated circuits. The latter may be packaged in so-called "flat packs" with typical dimensions — $1/8 \times 1/4 \times 0.040$ in. — and also in cylindrical containers such as TO-5 and TO-18 cans. Economy of the assembly process and high-temperature performance of the package were additional desirable attributes.

The basic unit of the interconnection matrix-packaging system is a grooved composite ceramic wafer upon which the cased and uncased integrated circuits of various geometrics, and auxiliary passive elements, are attached in a manner which optimizes dissipation of thermal energy to the package exterior. Communication between integrated circuits, thin-film passive elements, and external circuitry is accomplished by means of a metallized horizontal and vertical multilayer interconnection matrix which permits multiple crossover networks.

A section of the interconnection matrix-packaging system is illustrated schematically in Fig. 19. A cutout view is presented to illustrate the approach employed to obtain crossovers and to attach terminal pins. The individual members which make up the multilayer structure are produced by a process similar to the following. The alumina or beryllia substrates are pressed, fired, and machined by methods well known to the ceramics industry. Special holes may be drilled by electron-beam techniques; however, generally standard patterns are used in order to reduce cost. The substrates are coated with an adherent film of chromium-gold on all surfaces, including the hole walls, by high-vacuum deposition of the conductive materials. Patterns are etched by electron-beam microthermal evaporation. Beam programming during etching is automatic and employs a tape control.

The ceramic substrates are illustrated in Fig. 20 and 21, re-

⁵ Developed under Air Force Contract No. AF 33(657)-8790.

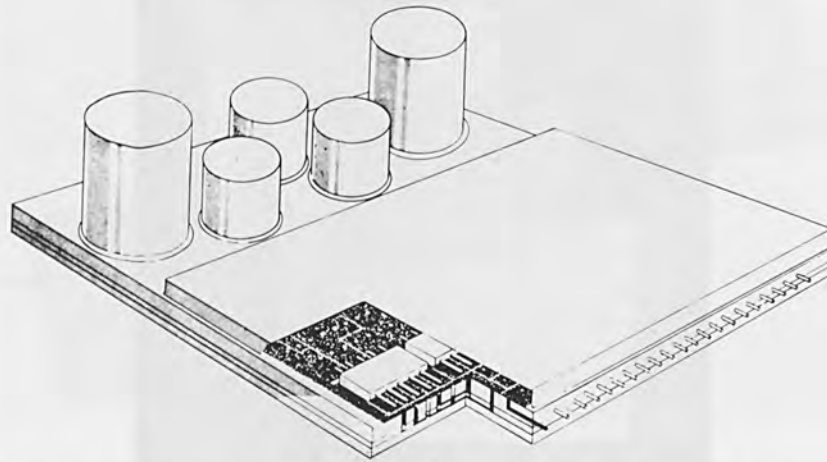


Fig. 19. Interconnection, packaging system, section of multilayer cross-over network with integrated circuits.

spectively, after machining and also after metallizing the etching [13]. The time required to define precisely the interconnection network by electron beam scribing is of the order of 10 sec. Typical machine parameters used for the process are as follows:

Accelerating voltage	130 kV
Current	7.5 mA
Beam diameter	1.0 mil
Pulse length	8 μ sec
Pulse frequency	16,000 cps
Translation speed	5 in./sec

After definition of the conductive pattern the substrates are stacked together and the vertical feedthroughs fabricated by a "vacuum casting technique [13]." An assembled multilayer structure is illustrated in Fig. 22.

Programmed electronbeam processes are employed to define both the conductive matrix on the metallized substrates and also to join the leads.

Interconnection of the cased integrated circuits and associated components is achieved by mounting of the packages within the grooves of the topmost substrate of the multilayer board followed

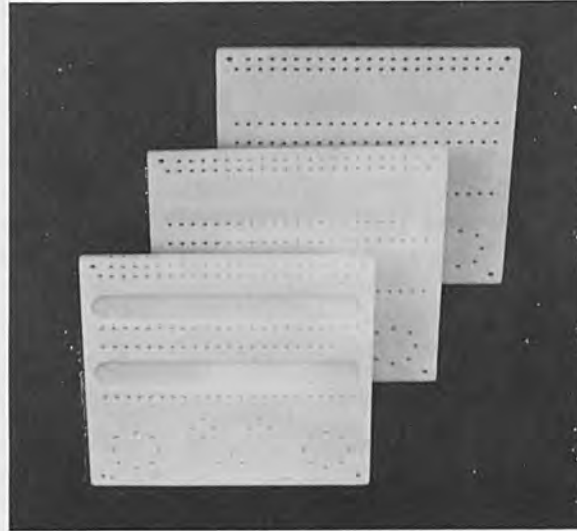


Fig. 20. Ceramic substrates after machining and firing.

by automatic or automated (APC) welding. Since only physical contact without any pressure is required between the leads and the metallized surface, a hold-down device has been developed for this purpose. A substrate with "flat packs" and lead hold-down fixture is shown in Fig. 23. The unit consists of a number of "comblike" members which are in contact with each lead thus assuring intimate contact with the metallized surface.

The multilayer boards with attached circuits packaged in "flat packs" are placed on a fixture which is mounted on the worktable within the electronbeam machine by means of locating dowel pins to provide repetitive and accurate positioning of the assemblies. The worktable motion is programmed to move to a pre-established or reference position under the optical axis of the electron-beam column. At this point electron-beam welding of the leads to the metallized substrate may be carried out by any of the previously described techniques. In the approach wherein microwelding is carried out by programmed worktable motion and beam deflection, the high-speed reader (master control) enables the worktable programmer which brings a reference lead of the first flat pack to be interconnected under the beam. The high-speed reader takes over

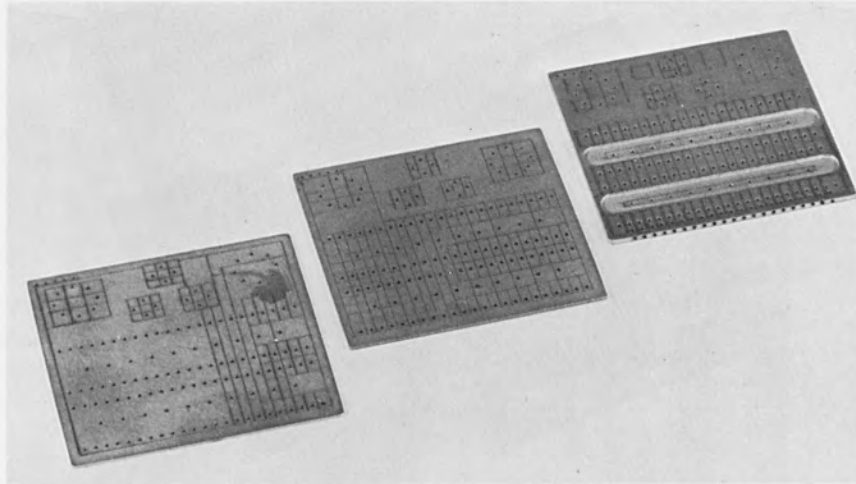


Fig. 21. Scribed patterns on metallized ceramic.

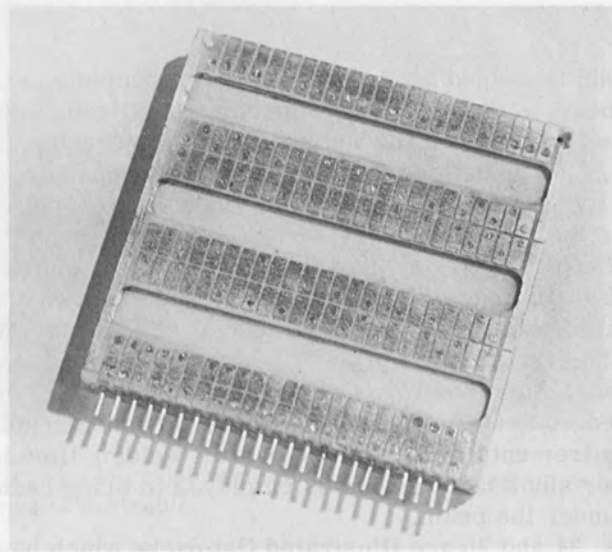


Fig. 22. Assembled multilayer board.

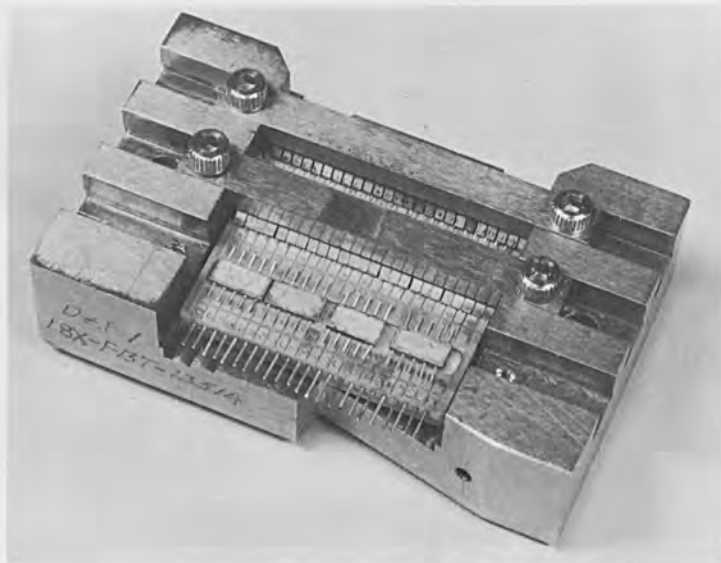


Fig. 23. Lead hold-down device.

and the lead is welded according to a preprogrammed schedule. Next the beam is dc deflected to the outermost lead on the same side of the flat pack and the weld sequence is repeated. The same technique, i.e., dc deflection followed by weld schedule, is employed to join all the leads of the flat pack. Subsequently the table is programmed to bring the reference lead of the next "flat pack" and the previous cycle is repeated. Typical weld times are of the order of 5 to 15 msec, whereas table motion requires about 2 sec, from point to point. As pointed out previously, this welding mode requires that the leads be precisely located on a grid matrix. Consequently, tight control on the lead spacing as well as fixturing is required. Use of the APC system lessens considerably the above requirements; however, the overall welding time is somewhat longer since table motion is employed to bring each sequential lead under the beam.

In Figs. 24 and 25 are illustrated flat packs which have been interconnected (a) to a simulated stainless steel plate, and (b) to a ceramic multilayer structure. Programmed microbonds have also been obtained with the APC system.

The automated electron-beam system capability is uniquely suited for the fabrication of thin-film resistors. Area thin-film

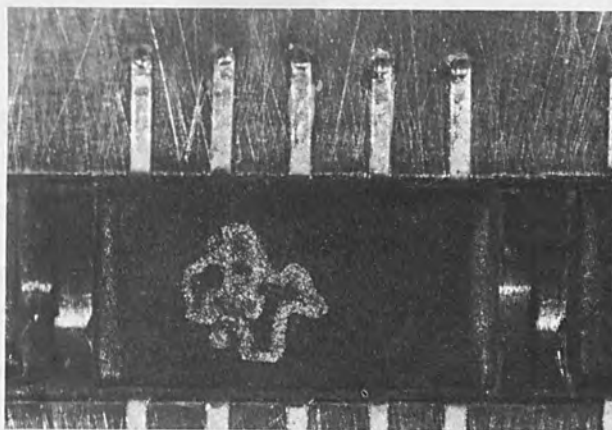
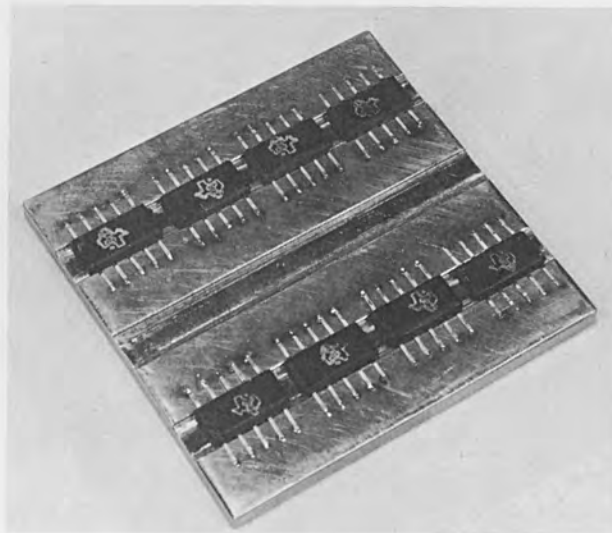


Fig. 24. Microjoints made by automatic programming of worktable.

resistive metals such as chromium are vacuum deposited on substrates provided with suitable terminations or conductive patterns. After deposition and stabilization of the resistor provided with a protective film, the element is loaded in a magazine which is inserted within the electron-beam machine. Definition and trimming

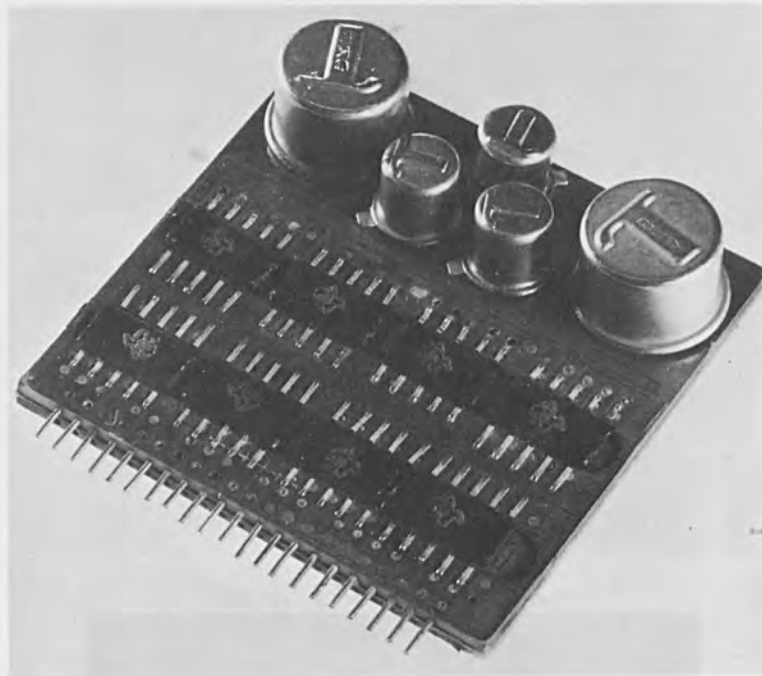


Fig. 25. Interconnection packaging system with welded integrated circuits.

of the resistive elements are carried out as follows: The wafer is mechanically transferred under the beam and the electron beam is programmed to subdivide the resistive area into the specified number of elements by scribing. Subsequently, each element is scribed to a preset value. The resistance is monitored during scribing and the process stopped when the specified value is reached. The operation of the resistance bridge was discussed previously. Typical scribing time/per resistive wafer is 2 sec. Electron-beam scribed thin-film resistors are illustrated in Fig. 26.

CONCLUSION

Electron-beam technology is rapidly evolving from development to production environments. This evolution is particularly evident

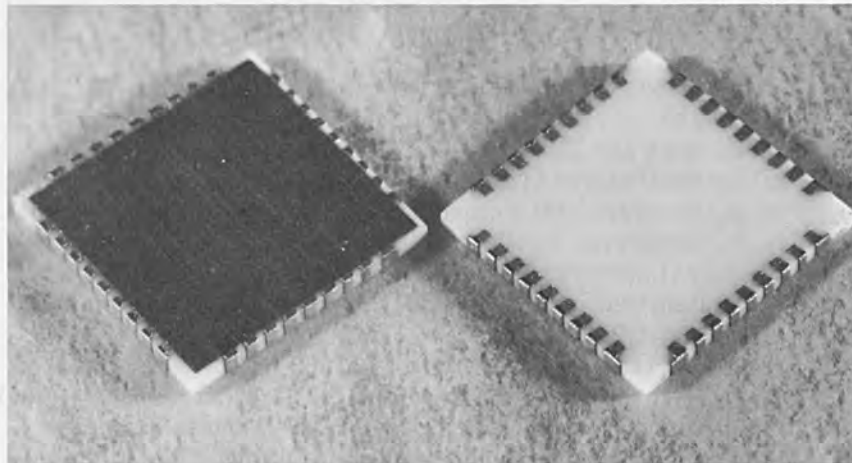


Fig. 26. Thin-film resistors prepared by programmed electron-beam scribing.

in the application of electron beams to the welding of structural parts.

Some strides have been made in the use of electron beams in microelectronics; however, large-scale applications await the development of automated processes.

Hamilton Standard Division, United Aircraft Corporation, is playing a vital role in the automation of electron-beam processes. One such system has been discussed and its application described as applied to the fabrication and assembly of microminiaturized circuits and equipments.

Acknowledgments

A considerable amount of the work described in this paper was carried out under Air Force Contract AF 33(657)-8790, Electronics Branch, Manufacturing Technology Division, AF Material Laboratory, Wright Patterson AFB, Ohio.

Acknowledgements are due to Messrs. K. O. Olson, R. J. Green, D. Spaulding, and P. O'Brien for their contribution in the experimental effort, and to Mr. Ullery for his technical guidance.

LIST OF REFERENCES

1. K. H. Steigerwald, Proc. 3rd Symp. on Electron Beam Tech, R. Bakish, Ed, p. 269 (1961).
2. D. A. Buck and Shoulders, K. R., "An Approach to Micro-miniature Printed Systems," Special Publication T-114, Proc. Eastern Joint Computer Conference (December, 1958).
3. K. R. Shoulders, Publication Number PB171027, Office of Technical Services (1960).
4. G. Mollenstedt, and R. Speidel, "Beitrag zur Microminaturization." Z. Angew. Phys. (1962).
5. R. W. Christy, J. Appl. Phys. 31, 1680 (1960).
6. A. G. Baker, and W. C. Morris, Rev. Sci. Instr. 32, 458 (1961).
7. P. J. Klass, "Electron Beam Technique Makes, Inspects Semiconductor Microcircuits," p. 80, Aviation Week, Sept. 23, 1963.
8. R. Castaing, and A. Guinier, Proc. International Conference on Electron Microscopy, Delft (1949).
9. R. E. Ogilvie, Proc. 3rd Symp. on Electron Beam Tech, R. Bakish, Ed., Boston, Mass. p. 322 (1961).
10. K. C. A. Smith, Pulp and Paper Magazine of Canada, Technical Section, p. 3, Dec., 1959.
11. P. J. Klass, "Electron Beam Technique Makes, Inspects Semiconductor Microcircuits," p. 80, Aviation Week, Sept. 23, 1963.
12. D. J. Garibotti, and W. V. Lane, "The Enhanced Micro-module -- A Disciplined Approach to Microcircuit Integration," presented at the 4th International Electronic Circuit Packaging Symp., Aug., 1963, Boulder, Colo.
13. D. J. Garibotti, and E. H. Miller, "Versatile Interconnection System for Integral Electronics," presented at the National Electronics Conference, Oct., 1963, Chicago, Ill.

ELECTRON BEAMS IN RECORDING AND INFORMATION STORAGE

Section III

ELECTRON BEAMS IN RECORDING AND INFORMATION STORAGE

Generalized growth of scientific and technological printed matter has led to a demand for more efficient means of recording and information storage. It is this requirement that is one of the prime movers for the development of electron beams for recording and information storage. This area of research has many excellent examples of progress. The first approach to electron beam recording and information storage with electron beams is the first, recording is effected through the thermally induced changes on the recording media by the electron beam. This approach has many limitations and it is not likely to see much growth in the future. The second, the so-called thermal approach, relies on chemical and physical changes induced by the electron beam on suitable materials. This includes the type investigated by Webb (1955) and Molenski (1959) involving development of evaporated metal thin film on organic substrates where the electron beam has already produced changes and it includes the thermoplastic approach developed by Glean (1959), where a charge pattern is frozen on the surface of a thermoplastic material; and last, the recently developed Apex method of recording on a special silver compound sensitive film which is essentially sensitive to electron radiation and virtually insensitive to light, Miller (1964).

In all the above cases, the information can be retrieved by optical or electron optical means, depending on the resolutions required. In general there are a considerable number of problems, including that of high cost with the direct electron beam record-

LIST OF REFERENCES

1. K. H. Maierwald, Proc. 3rd Symp. on Electron Beam Tech., V. Sakash, Ed. p. 263 (1963).
2. D. A. Book and Shaulders, K. H., "An Approach to Micro-miniature Printed Systems," Special Publication T-114, Proc. Eastern Joint Computer Conference (December, 1958).
3. K. R. Shaulders, Publication Number PB171027, Office of Technical Services (1960).
4. C. Moench and R. Spang, "Designing for Micro-miniaturization," *Z. Angew. Phys.* (1962).
5. R. W. Christy, *J. Appl. Phys.* 31, 1650 (1960).
6. A. G. Baker, and W. C. Morris, *Rev. Sci. Instr.* 32, 458 (1961).
7. E. J. Kane, "Electron Beam Technique Makes Progress in Semiconductor Microcircuits," p. 30, Aviation Week, Sept. 23, 1963.
8. B. Castaing, and A. Guinier, Proc. International Conference on Electron Microscopy, Delft (1963).
9. R. E. Tech, R. Haxius, *Proc. 3rd Symp. on Electron Beam Tech.*, V. Sakash, Ed. p. 263 (1963).
10. K. C. A. Smith, *Pulp and Paper Magazine of Canada*, Technical Section, 63, 10 (1963).
11. P. J. Kane, "Electron Beam Technique Makes Progress in Semiconductor Microcircuits," p. 30, Aviation Week, Sept. 23, 1963.
12. D. J. Caribelli, and W. V. Latta, "The Enhanced Micro-module - A Disciplined Approach to Microcircuit Integration," presented at the 14th International Electronic Circuit Packaging Symp., Aug. 1963, Boulder, Colo.
13. D. J. Caribelli, and E. H. Miller, "Versatile Interconnection System for Integral Electronics," presented at the National Electronics Conference, Oct., 1963, Chicago, Ill.

ELECTRON BEAMS IN RECORDING AND INFORMATION STORAGE

The unprecedented growth of scientific and technological printed material and the desire to record and withdraw any part of this information at will and with minimum effort continue to place a severe burden upon us. It is this requirement that is one of the prime movers for research and development into the utilization of electron beams for recording and information storage. A broad survey in this area of endeavor was made several years ago in an excellent presentation by Newberry (1962), who presented the many problems and possible solutions which have to be considered.

Generally speaking, two broad approaches have been utilized to effect recording and information storage with electron beams. In the first, recording is effected through the thermally induced changes on the recording media by the electron beam. This approach has severe resolution limitations and it is not likely to see much growth in the future. The second, the so-called athermal approach, relies on chemical and physical changes induced by the electron beam on suitable materials. This includes the type investigated by Wehe (1959) and Molenstedt (1960) needing development by evaporation of metal thin film on organic substrates where the electron beam has already produced changes and it includes the thermoplastic approach, developed by Glenn (1959), where a charge pattern is frozen on the surface of a thermoplastic material; and last, the recently developed Ampex method of recording on a special silver compound sensitized film which is essentially sensitive to electron radiation and virtually insensitive to light, Miller (1964).

In all the above cases, the information can be retrieved by optical or electron optical means, depending on the resolutions required. In general there are a considerable number of problems, including that of high cost with the diverse electron-beam record-

ing and information storage approaches. The overcoming of these is certain to bring about recording techniques with features surpassing the best magnetic recording techniques of today. By all available information, it also can be said that these techniques need not be excessive in cost.

The utilization of electron beams in recording and information storage systems is still two to five years removed from an accepted and commonly used approach. In closing these few introductory remarks, I would like to indicate that a number of industries will benefit from the eventual development and acceptance of electron beams in recording and information storage. The widest acceptance and growth is likely to take place in the video recording and reproduction field, military equipment and systems will follow closely thereafter.

Although several presentations on this topic were initially scheduled, difficulties with corporate release reduced the actual number to two, the papers of Everhardt and White and North. Although they provide us with much useful information, they do not cover the field comprehensively. It is hoped that in the 1966 Electron and Ion Beam Science and Technology conference this void of the first conference will be filled.

Editor

REFERENCES

- Glen, W., 1959, J. Appl. Phys. 30, 1870.
Miller, R., 1964, Aviation Week and Space Technology, April 13.
Newberry, S. P. 1962, Proc. Fourth Symp. Electron Beam Tech.,
R. Bakish, Ed., Boston, Mass., p. 81.
Mollenstedt, G., 1960, Phys. Blatter, 16, 192.
Wehe, H. G., 1958, Electron Beam Recording Bell System Monograph, No. 3576.

NONTHERMAL ELECTRON-BEAM RECORDING

INTRODUCTION

The amount of information in the modern world is increasing at a rapid rate. Much of this information is transmitted from place to place by electronic communication systems, is recorded in electrical form, and is processed by electronic means (such as digital computers). Electron beams, such as those in television picture tubes, have played an important part in the presentation of this information. In recent years, however, wide-band electronic recording has been accomplished primarily by magnetic means; magnetic tape has been a popular medium for serial recording, and magnetic toroidal cores have been widely used for rapid-access computer memories. One of the limitations in magnetic tape recording is the velocity of the recording tape head, which at a 5 Mc recording bandwidth approaches 90 miles per hour. The acceleration at the tip of this recording head is approximately 6000 g's, i.e., 6000 times greater than the acceleration of gravity at the surface of the earth. Under these conditions, large increases in recording speed are anticipated, and alternate methods of recording become attractive. Nonthermal electron-beam recording is one of the most attractive alternatives, because micron-diameter electron beams can be easily and rapidly deflected from spot to spot on any one of several different types of recording media, producing wide-bandwidth recordings with high packing densities.

Nonthermal electron-beam recording can be divided into two categories, "charge" recording and "energy" recording. In the first category, the information is related to the stored electrical charge, and in the second category, it is related to the energy dissipated in the storage media. Thermoplastic recording, as described by Glenn (1959), and electrostatic recording, as recently

*Department of Electrical Engineering, University of California, Berkeley, Calif.

described by Krittman and Inslee (1963), belong to the category of charge recording. Direct electron-beam recording on silver halide emulsions, as described by Tarnowski and Evans (1962) and Thornley, Brown, and Speth (1964), and electron-beam recording on thin collodion films, as described by Mollenstedt and Speidel (1960), are examples of energy recording. Reviews of electron-beam recording have been presented by Wehe (1961) and Glenn (1962).

There are several interdependent parameters in an electron-beam recording system. These parameters include the information recording rate, which is proportional to the system bandwidth, the dynamic range and signal-to-noise ratio, the information packing density, which depends upon both the beam and the chosen recording media, and the media sensitivity. The electron current, current density, spot diameter, and the number of spot diameters over which the beam can be deflected are important electron-optical parameters. This paper attempts to point out certain fundamental relationships among these parameters. These relationships are applicable to a wide variety of serial electron-beam recording methods, and they provide a basis of comparison for different methods. The important problem of information readout is not discussed in this paper, because different methods of electron-beam recording may employ very different methods of information readout, often not using electron beams, but using light. Thus the readout problem is somewhat more difficult to generalize.

There are also certain system requirements which may eliminate one or more possible methods of electron-beam recording, even though when judged on fundamental grounds these rejected methods may seem somewhat superior to the method chosen. For example, certain electron-beam recording media are not erasable and reusable. This consideration alone may eliminate them from consideration in a system in which the media must be reused several times.

BASIC CONSIDERATIONS

The maximum rate at which information can be recorded or can be transmitted over a channel is alternatively called the information recording rate or the channel capacity C , and for binary signals is measured in bits per second. Shannon (1949) has derived a general expression which gives the maximum value C may have for any physical channel recording or transmitting binary bits, namely,

$$C \leq B \log_2 \left[1 + \frac{S_p}{N_p} \right] \quad (1)$$

In this equation, B is the system bandwidth in cycles per second, S_p is the signal power, and N_p is the noise power. The equation points out that if one wishes to record with a given dynamic range (which is limited by the peak signal-to-noise ratio and the media characteristics), the recording speed is directly proportional to the bandwidth of the recording system. The channel capacity for analog recordings generally falls far short of the maximum value given by equation 1; however, for coded binary signals, the channel capacity is similar in form to Shannon's value. For example, for an average error rate of 10^{-5} , pulse code modulation requires about seven times the signal-to-noise power ratio to achieve the same channel capacity C as Shannon's ideal method of coding (see Schwartz, 1959).

It is customary in dealing with many sorts of information recording to deal with peak signal voltages or currents, which are here denoted by S , and with root mean square (RMS) noise, here denoted by N . In electron-beam recording the thermionic cathode, which is a random emitter, sets the maximum signal-to-noise ratio which can be obtained. The recording process can only reduce this maximum signal-to-noise ratio. The maximum number of electrons per information element, n , is a measure of the peak signal. The corresponding measure of the RMS noise N is the RMS deviation of n , which is $n^{1/2}$. The signal-to-noise ratio therefore may be expressed as

$$\frac{S}{N} \leq n^{1/2} = \left[\frac{i}{2eB} \right]^{1/2} \quad (2)$$

In the above equation, n has been set equal to the maximum primary beam current i , divided by the electronic charge, multiplied by the time required to record an information element, the so-called Nyquist interval, $1/2B$. The expression for signal-to-noise ratio which involves the beam current i , the electronic charge e , and the bandwidth B is also the ratio of the beam current i to the RMS shot current in the beam $(2eiB)^{1/2}$. Equation 2 is basic to all electron beam recording; the current i must be proportional to the bandwidth B for a given signal-to-noise ratio.

In wide-band recording, packing density is an important para-

meter. In the case of tape recording the electron beam is scanned in one direction only, while the tape is moved in the other direction. Figure 1 shows a schematic of a recording tape of width w and thickness t , moving with a velocity v . If p is the packing density in bits/cm², then the recording rate c in bits/second may be written.

$$C = p w v \quad (3)$$

This implies that for a given information recording rate C , the higher the packing density p , the slower will be the media velocity v ; similarly, the wider the media, the slower its velocity can be. The media width is often determined by the distance that the electron beam can be deflected while maintaining a given spot diameter. This value may be as high as 10,000 spot diameters deflection if dynamic focusing is used. In digital recording, the transverse accuracy with which the beam can be located may limit the media width. The maximum packing density is determined by either the media resolution, the electron beam spot diameter, or both.

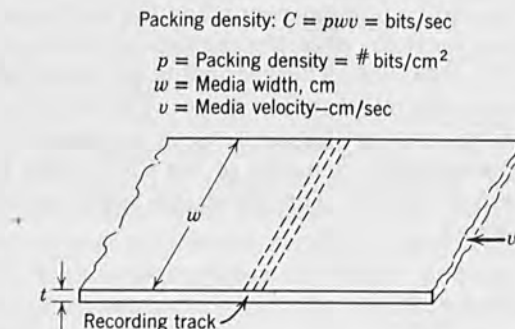


Fig. 1. Electron-beam recording tape, illustrating parameters discussed in text.

A detailed knowledge of electron penetration into matter, such as described by Schumacher (1964), is necessary to understand the mechanisms and limitations of electron-beam recording. The penetration in depth and the transverse scatter of the primary electrons determines the volume over which charge is deposited or energy is dissipated; this volume ultimately limits the maximum

packing density which can be obtained using electron-beam recording. The primary beam electrons penetrating into the solid lose energy through inelastic collisions; these collisions may excite bound electrons to higher bands in the solid or may ionize them completely, and result in an "excited" zone around the path of the penetrating electrons. Inelastic collisions with tightly bound electrons can cause large-angle scattering, as can collisions between the penetrating electrons and the nuclei of atoms in the material. A highly schematical diagram of typical electron trajectories in a solid target is drawn as Fig. 2. The maximum depth of

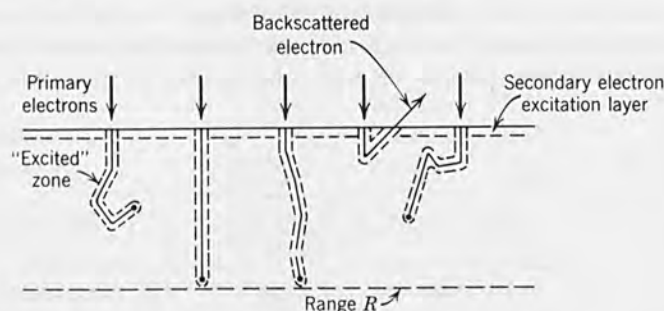


Fig. 2. Typical electron trajectories in a solid target.

penetration is the range R , which for a limited variation in the beam energy V can be written as

$$R = \frac{AV\gamma}{\rho} \quad (4)$$

In the above empirical equation, A and γ are constants which depend on the material, and ρ is the density of the target material. γ generally ranges between 1.3 and 1.8. Makhov (1960) has given values for A and γ for several different materials, and his values agree reasonably well with the work reported by Kanter (1960). Note that in Fig. 2 a thin layer at the surface has been denoted the secondary electron excitation layer because the excited electrons in this layer which escaped the target are the so-called "true" or low-energy secondary electrons. Backscattered electrons may originate from farther beneath the surface and are caused by one or more large-angle scatterings of the primary electrons; the

backscattered electrons also produce low-energy secondary electrons as they penetrate through the secondary electron excitation layer on their way to the surface. At beam voltages above a few hundred volts, it is believed the backscattered electrons and the secondaries they generate contribute a good deal to the secondary electron emission noise (see Everhart, Wells, and Oatley, 1960). This topic is discussed in more detail below.

The ultimate resolution of either charge or energy electron-beam recording is set by the scattering of electrons in the target material. Schematic diagrams showing the estimated energy dissipation vs. distance in thick and thin targets are given in Fig. 3. For beam diameters large compared to the range, the packing density is determined by the beam. For beam diameters small compared to the range, in thick targets the packing density is determined by the penetration effects, and in thin targets, the energy

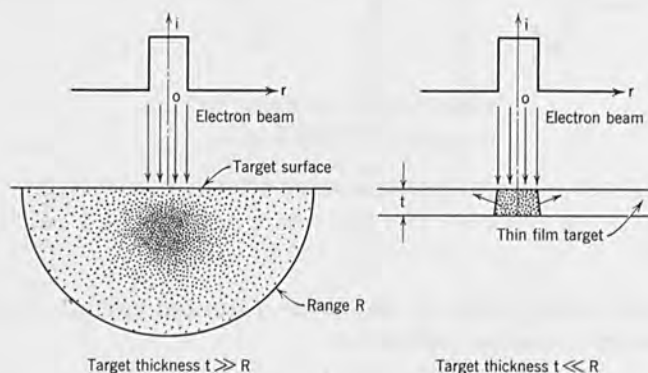


Fig. 3. Diagram. Energy dissipation versus distance.

is believed dissipated in the area approximately equal to the beam cross-sectional area (see Mollenstedt and Speidel, 1960). Thus in thin targets, a much higher resolution is expected, but at the expense of a lower sensitivity. Note also that for thick targets the distribution of stored charge density in the target will be determined by scattering; if no charge diffusion takes place, the stored charge should all be contained within the range R . The readout method will also affect the resolution (see, for example, Krittman, 1963).

For high-speed recording the number of electrons per unit area required to produce a given signal level is an important index of the media sensitivity; the lower the required number per unit area, the higher is the sensitivity. Table I compares representative methods of nonthermal electron-beam recording; the quoted resolutions and electrons per unit area apply to the same recording conditions. This table is less extensive, and differs slightly from one published earlier by Glenn (1962).

Table I
Comparison of Representative Electron-Beam
Recording Medias

Material (source)	Type	Beam Energy (volts)	Reso- lution (μ)	Sensi- tivity (elec- trons/cm ²)
Thermoplastic (Glenn)	[charge]	5-10 kV	4 μ	6 $\times 10$ in.
Silver halide (Tarnowski & Evans)	[energy]	20 kV 14 kV	1.5 μ <1.5 μ	2 $\times 10^{10}$ 4 $\times 10^{10}$
Polystyrene (Krittman & Inslee)	[charge]	300 V	$\sim 5 \mu$	$\sim 10^{11}$
Thin collodion film (Mollenstedt & Speidel)	[energy]	60 kV	$\sim 0.01 \mu$	$\sim 6 \times 10^{15}$
Photoresist (KPR) (Wells-Smith)	[energy]	10-20 kV	$\sim 1 \mu$	$\sim 10^{14}$

For a given signal-to-noise ratio and recording bandwidth, the sensitivity of the media must decrease as the packing density (resolution) increases. This relationship is well known for photographic emulsions; higher resolution emulsions require more light to produce a given density than do lower resolution (granier) emulsions. If equation 2 is set equal to a constant, then for a constant bandwidth, the current i must be constant. The current density is proportional to both current i and packing density p , and therefore will increase with p . To maintain the same signal-to-

noise ratio per information element, the sensitivity must therefore decrease so that the higher current density produces the same effective exposure (the same density in the case of photographic film).

The noise processes are somewhat different in the "charge" and "energy" categories of nonthermal electron beam recording. In the charge category, the signal is proportional to the stored charge, en_s , which is the primary incident charge en minus that lost through secondary emission δen . In the energy category, the signal is proportional to the number of "events" caused by the beam's energy dissipation. If the mean excitation energy per event is V_e , and the mean energy dissipated in the media per incident primary is V , then the mean number of events n_s due to n electrons striking the media is $n(V/V_e)$. The signal-to-noise ratio can also be computed for these cases; the results may be summarized as:

Charge	Energy
$\bar{n}_s = \bar{n} (1 - \bar{\delta})$	$n_s = \bar{n} \frac{V}{V_e}$

(5)

$$\frac{S}{N} = \bar{n}^{1/2} \frac{(1 - \bar{\delta})}{[\sigma_{\delta}^2 + (1 - \bar{\delta}^2)]^{1/2}} \quad \frac{S}{N} = \bar{n}^{1/2} \left[\frac{V}{V + V_e} \right]^{1/2} \quad (6)$$

In equation 6, $\bar{\delta}$ is the mean secondary emission coefficient, and σ_{δ}^2 is the variance of the secondary emission coefficient δ . In the special case when $\sigma_{\delta}^2 = \bar{\delta}$, which corresponds to random variations of δ about $\bar{\delta}$, the charge storage S/N reduces to

$$\frac{S}{N} = \bar{n}^{1/2} \left[\frac{|1 - \bar{\delta}|}{(\bar{\delta}^2 - \bar{\delta} + 1)^{1/2}} \right]$$

For "charge" recordings, it is important to use media for which $\bar{\delta} < 1$ or $\bar{\delta} > 1$ to avoid appreciable degradation in signal-to-noise ratio. For "energy" recordings, it is important that the mean energy dissipated in the media per incident primary electron V be much larger than the mean excitation energy V_e to avoid signal-to-noise degradation. Generally V is several kilovolts, whereas V_e is several tens of volts, so this condition is satisfied.

The most convenient criteria of the spacial resolution of an electron-beam recording is the sine-wave response introduced by Schade (1955). The sine-wave response may be calculated for the

electron beam knowing the current density distribution in the final spot, and may be calculated for the media knowing its resolution. The sine-wave response for the recorded signal τ is equal to the product of the beam and media sine-wave responses, and is always unity or less, generally decreasing as the recording frequency increases. This factor τ should multiply the peak signal $\bar{n}_s$ and the signal-to-noise ratio in equations 5 and 6, respectively for any frequency for which $\tau < 1$. Schade (1964) has reviewed the uses of sine-wave response for systems calculations. When this factor is included, the charge category S/N expression of equation 6 becomes very similar to the expression given by Krittman and Inslee (1963).

ELECTRON-OPTICAL CONSIDERATIONS

From the basic considerations presented above, it is apparent that the maximum possible current must be concentrated in a very small spot for wide-band, high packing density electron-beam recording. In addition, this spot of current must be deflected over the recording media both rapidly and with little spot degradation.

The current which can be focussed into a spot of given diameter is limited by the electron-beam brightness as discussed by Langmuir (1937) and by electron-optical aberrations. The aberrations which prove most troublesome for electron-beam recording are listed in Table II. Spherical aberration has so far proved impossible to eliminate from electron lenses. The spherical aberration

Table II

Aberrations Important in Electron-Beam Recording

Spherical	$d_s = 0.5 C'_s \alpha^3$	von Ardenne, p 39
Astigmatism	$d_a = Z_a \alpha$	von Ardenne, p 309
Deflection	$d_d = K_d \theta^2 \alpha$	von Ardenne, p 16

coefficient C_s is a function of lens focal length and geometry, and is generally tabulated for a point image at the lens focus. For a point image farther from the lens, a modified coefficient $C'_s = C_s(b/f)^4$ must be used, where b is the lens-to-image distance, and f is the lens focal length. The convergence angle at the image is α . Axial astigmatism is caused by nonsymmetries in the lens, either mechanical or magnetic in nature, and can be compensated with a stigmator. Deflection aberrations result from beam deflection, and

can be dynamically corrected by auxiliary cylindrical lenses. For several thousand spot diameter deflection, whereas much current as possible is desired in the electron spot, such dynamic correction is essential. Assuming that the astigmatism and deflection aberration can be corrected, the current in a spot of diameter d is limited by the brightness condition and spherical aberration to a value,

$$i = A j_0 e V_0 d^{8/3} / C_s^{2/3} k T \quad (7)$$

This equation has been derived by Smith (1962) among others. Here j_0 amps/cm² is the cathode current density, V_0 volts is the beam voltage, T °K the cathode temperature, e is the electronic charge, k is Boltzmann's constant, and A is a numerical constant (0.36 using Smith's derivation). In this expression d contains 80% of the beam current i . For a given spot size, the current can be increased only by increasing the cathode current density, the beam voltage, and by decreasing the cathode temperature and lens spherical aberration. It is often necessary to limit the convergence angle α to a value smaller than the optimum that produces i , to avoid excessive demands on the dynamic focusing. Thus equation 7 should be regarded as an upper limit which can be obtained with a thermionic cathode and electron lens; such a limit is useful to judge the effectiveness of any given system.

CONCLUSIONS

By using wide-band electron-beam recording, information may presently be recorded approximately one-to-two orders of magnitude faster than is possible using alternate methods. Information packing densities of approximately 10⁷ bits per cm² have been obtained using presently available media, but the fastest recording rates may not be possible with these packing densities. Resolutions corresponding to packing densities exceeding 10¹⁰ bits/cm² have been demonstrated on media which are much thinner than the electron range, but at much reduced sensitivity. Higher recording rates at these high packing densities depends primarily on the development of cathodes with much higher emission densities, so that much greater current densities are possible at the recording media. For wide-band recording, beams must be deflected without

loss of resolution, which implies dynamic focusing for deflections exceeding several hundred spot diameters. While there are still fundamental problems to solve, and much systems engineering remains before electron-beam recording becomes common, in a world where the amount and utilization of information is increasing exponentially, this system of recording which offers fundamental increases in speed and packing density would seem to have a bright future.

ACKNOWLEDGEMENTS

It is a pleasure to acknowledge helpful discussions concerning electron-beam recording with Emil Boblett, Reginal Lamb, Richard Thornley, and Kurt Wallace.

REFERENCES

1. T. E. Everhart, O. C. Wells, and C. W. Oatley, *J. Electronics and Control* **7**, 97 (1959).
2. W. E. Glenn, *J. Appl. Phys.* **30**, 1870 (1959).
3. W. E. Glenn, Chapter 14 of "Introduction to Electron Beam Technology," R. Bakish, Ed., John Wiley and Sons, New York (1962).
4. I. M. Krittman, *IEEE Trans. on Electron Devices* **10**, 404 (1963).
5. I. M. Krittman and J. A. Inslee, *RCA Review* **24**, 406 (1963).
6. A. F. Makhov, *Fiz. Tverd. Tela.* **2**, 2161 (1960).
7. G. Mollenstedt and R. Speidel, *Physikalische Blatter* **16**, 192 (1960).
8. O. H. Schade, *J. SMPTE* **64**, 593 (1955).
9. O. H. Schade, *J. SMPTE* **73**, 81 (1964).
10. B. Schumacher (1964), paper on electron penetration effects, see Section I, page 5.
11. M. Schwartz, "Information Transmission, Modulation, and Noise," McGraw-Hill Book Co., New York (1959).
12. C. E. Shannon, *Proc. IRE* **37**, 10 (1949).
13. K. C. A. Smith, *Encyclopedia of Microscopy* (G. L. Clark, Ed.), Reinhold, New York, p. 241 (1961).
14. A. A. Tarnowski and C. H. Evans, *J. SMPTE* **71**, 765 (1962).
15. R. F. M. Thornley, A. V. Brown, and A. J. Speth, *IEEE Trans. on Electronic Computers* **13**, (1964).
16. M. vonArdenne, "Tabellen der Elektronenphysik, Ionenphysik, und Uebermikroskopie," Band I, Deutscher Verlag der Wissenschaften, Berlin (1956).
17. H. G. Wehe, *Proc. Third Symp. on Electron Beam Technology* (R. Bakish, Ed.), 249 (1961).

AN ELECTRON-BEAM MEMORY UTILIZING SOME PROPERTIES OF POLYMER FILMS‡

INTRODUCTION

It has been known for some time that surfaces irradiated with electrons in the presence of organic vapors become covered with layers of contaminant. The process by which this contaminant forms was assumed by Buck and Shoulders (1) to be a surface reaction in which adsorbed gas was activated by the electron beam, the activated adsorbed species subsequently interacting with one another to form a solid layer. More detailed examination of the process by Haller and White (2) showed that the mechanism indeed proceeded through a step involving an adsorbed gas, but that the electron beam induced activity in the previously deposited polymer layer. The layer produced with butadiene gas was introduced into a vacuum chamber as a deliberate contaminant has been shown (3) from a comparison of dielectric properties to be similar to a sample of bulk polybutadiene. Since the polymer film is produced by a surface reaction, it might be anticipated that the formation would be selective in that polymers would form only where the electron beam struck the surface. This has been shown to be the case for films formed from butadiene or acrolein vapor (4).

It is suggested that this property of selective formation of a polymer film may be utilized in the storage of information on a metal plate. If the metal plate is imagined as being subdivided into a two-dimensional matrix of squares, then the presence of polymer film in a square could indicate a "1" and the absence of polymer

*Standard Telecommunication Laboratory, Harlow, Essex, England.

†Physics Department, Stanford University, Stanford, Calif.

‡This work was performed while both authors were employed at International Business Machines Research Center, Yorktown Hts., N.Y.

would indicate a "0." It is further suggested that the presence or absence of polymer film at any one position would be determined by the use of a simplified electron-scanning microscope similar to that described by Da Silva and White (5). This is similar in principle to the monoscope and uses an electron probe to scan the surface. The difference in signal when the probe scans a metal insulator interface is sufficient to permit detection of that interface.

The capacity of a memory such as that described would be limited only by the size of the electron beam. With present technologies, electron guns with beams of $10\text{-}\mu$ spot diameter are available. If then a spot of polymer $10\text{-}\mu$ in diameter is deposited in a square of $20\text{-}\mu$ side, a bit density of 2.5×10^5 bits/cm² is possible. Thus on a plate 40 cm^2 (2.5 in. along one edge) a total density of 10^7 would be possible.

The rate at which a memory of this type can be read may be estimated using the arguments developed by Zworykin and co-workers (6) and discussed by Da Silva and White (5). The time constant of the circuit comprising the load resistors is the important factor to be considered. A reasonable value for the capacitance would be 3pf which would correspond to an impedance of $15,000\Omega$ at 5 Mc. In such a case the load resistor may also be as large as $15,000\Omega$. In order to obtain a conservative estimate suppose a $10,000\Omega$ load resistor is used. An emission of 1 amp/cm² is well within the limit obtainable and this would correspond to approximately $1\text{ }\mu\text{A}$ in a $10\text{-}\mu$ spot. The voltage developed by this current in the load resistor is about 0.01 V, which is about thirty times the thermal noise voltage in the load resistor at 5 Mc. A reading rate of 5 Mc. appears therefore to be well within the capabilities of the method. The time required to scan the entire memory would then be 2 sec, and the access time, which is the time to scan along an edge of the memory to the correct row and then down the row, would be about 0.6 msec.

Obviously an important factor is the speed with which information may be written into the memory. Experimental measurements of the growth of polymer films on a polymer surface have indicated that growth rates up to $150\text{ }\text{\AA}/\text{sec}$ might be possible. It is anticipated that since adsorption of an organic vapor on a metal surface is probably faster than that on a polymer surface, the growth of the first few layers of polymer on a metal surface might proceed at a very much faster rate than that indicated above. The writing time still cannot be estimated with any accuracy, however, since it is not known what minimum thickness polymer layer is necessary before a measurable signal may be observed in the electron-

scanning microscope. The main object of this investigation was therefore to deposit polymer layers with certain measurable exposure times and to determine the manner in which the signal height observed in the electron-scanning microscope varied with exposure time of the surface to the electron beam. In this way a realistic experimental measurement may be obtained of the writing speed which is feasible for the proposed memory unit.

EXPERIMENTAL

The experimental arrangement was identical to that described by Da Silva and White (5) and is shown in Fig. 1. A 5 C PIA electron gun supplied by Superior Electronics or by RCA was mounted

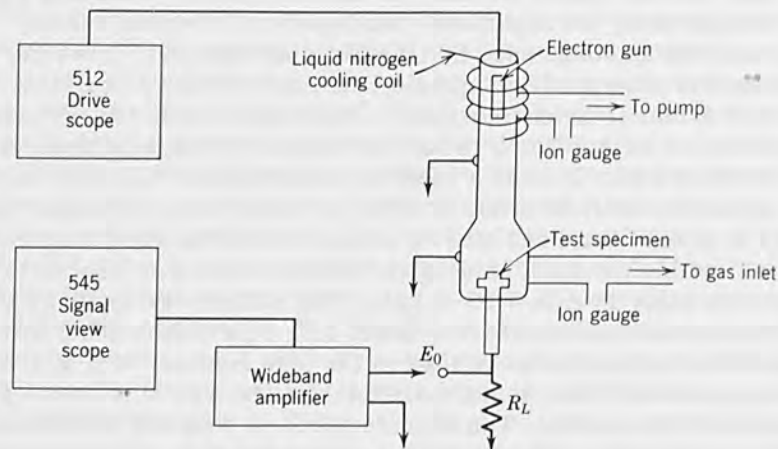


Fig. 1. Experimental system used.

in parallel with a Tektronix 512 oscilloscope, with the exception of the filament which was operated from a separate power supply. This arrangement was capable of producing a 0.010-in. diameter electron beam which had $10\text{-}\mu\text{A}$ beam current at an accelerating potential 3 kV. The target consisted of a lead film evaporated on to a glass slide which was mounted on an aluminum plate placed at right angles to the beam. The lead film made electrical contact with the aluminum plate through a silver paint electrode on the glass slide. A second glass slide coated with a phosphor material

was also mounted on the aluminum plate so that the electron beam could be accurately positioned and focussed. Aquadag electrodes were painted along the inside of the tube containing the electron gun and target, one to maintain the accelerating potential, and another, placed close to the target, was biased 22 1/2 V positive with respect to the target to facilitate the collection of secondaries. The whole assembly was evacuated with a 40 l/sec mercury diffusion pump, which was well trapped with two conventional liquid nitrogen traps between the pump and the electron gun tube, and also by means of a coil of copper tubing cooled with liquid nitrogen and placed around the outside of the glass tube in the immediate vicinity of the electron gun.

The pressure in the system was measured on an ionization gauge sealed into the wide tube connecting the diffusion pump to the tube containing the electron gun, an ultimate vacuum of 4×10^{-6} torr being obtainable. A second ionization gauge was placed on the tube through which the gas entered the system. In general, the pressures indicated by the two gauges were different, the higher pressure being indicated on the second gauge which was farther from the diffusion pump. When gas was admitted to the system, the pressure readings on the two gauges differed by about a factor of 10. This was thought to indicate that some differential pumping action was taking place and the pressure in the region of the electron gun was less than that in the region of the target.

A freshly evaporated lead film was placed in the system which was evacuated to its ultimate vacuum. The electron gun was turned on, and the beam focused and located in a suitable position on the phosphor slide which was placed immediately adjacent to the metal test film. The deflection plates controlling the horizontal scanning of the electron beam were connected to a Tektronix 545A cathode ray oscilloscope and the controls on this instrument were set so that the electron beam would scan a distance of 2 cm in specified times varying from 50 to 0.001 sec. The valve connecting the system to the gas reservoir was opened slightly until gauge no. 2 indicated a pressure in the range of 2×10^{-5} to 2×10^{-4} torr. When this had stabilized, the electron beam was positioned vertically on the phosphor close to the bottom end of the metal test slide and a single horizontal scan with the maximum scanning time was started. When the scan was completed, the vertical position of the electron beam was altered so that the next scan would be slightly above the first. The whole procedure was repeated until about seven horizontal scans had been made, one above the other, each with a different scanning time. The valve to the gas reservoir was closed

and the system evacuated to remove adsorbed gas for about 1 hour. The oscilloscope controls were then altered so that the accelerating potential was 1.5 kV and the electron beam scanned continuously in a vertical direction on the phosphor slide. The beam was moved manually in a horizontal direction until it was on the metal film. In a similar manner to that described by DaSilva and White (5), the anode current was monitored and displayed on a Tektronix 545A oscilloscope using a type L preamplifier. By this technique, it was possible to detect any insulating material which had been deposited on the metal film, and further to correlate the observed signal height to the scanning time. Since the length of the scan and the diameter of the electron-beam spot were known, it was thus possible to observe directly the signal height corresponding to particular writing times, and by observing the minimum signal height which could be adequately observed, to estimate the minimum writing time.

RESULTS

Initial experiments were performed using butadiene at a pressure of 5×10^{-5} torr and a mean current of $1 \mu\text{A}$. A typical pattern in which the various polymer strips were detected is shown in Fig. 2. The signal peaks representing the polymer strips are clearly

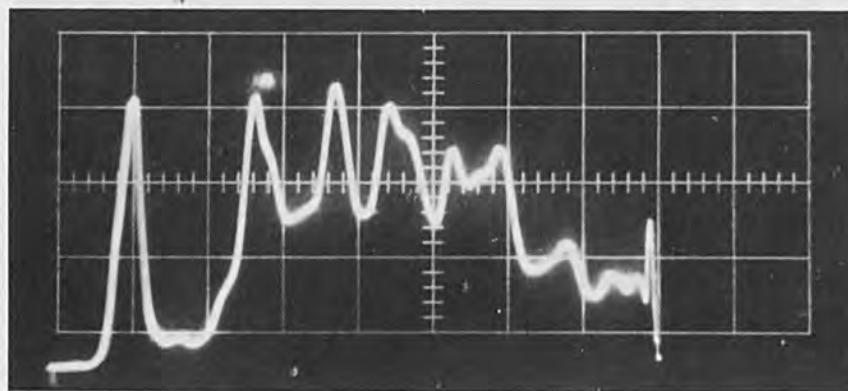


Fig. 2. Oscilloscope trace showing the presence of butadiene polymer strips on a lead film.

indicated as maxima above the signal corresponding to the metal background. The signal height may be represented as a function of writing speed as shown in Fig. 3. The minimum scanning time

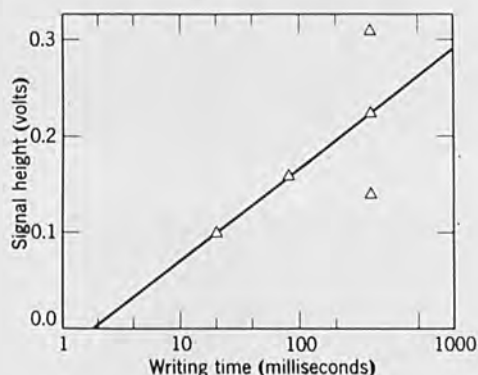


Fig. 3. Signal height (both positive and negative) as function of writing speed.

used (0.5 sec) corresponded to a 20 msec writing time and is quite clearly observable as signal of about 0.1 V. Figure 3 indicates a linearity between the signal height and the logarithm of the writing time. This is thought to be fortuitous and no significance is attached to this correlation.

It was anticipated that the rate of polymer formation might be very much dependent on the nature of monomer material used, so an alternative monomer, acrolein, which was known from other work to produce good polymer films was used in a second series of experiments.

The pressure of acrolein vapor was approximately 8×10^{-5} torr and the beam current was $5 \mu\text{A}$. The scanning rates were varied to correspond to writing times between 2000 and 0.2 msec. Figure 4a is a reproduction of the trace on the oscilloscope before the writing experiment, and Fig. 4b shows the same region, but with a slight expansion in the x direction, after deposition of polymer. It may be seen quite clearly that the position of the polymer films are characterized by signals superimposed on the background due to the metal film. Closer examination indicates that the signals corresponding to the higher writing times, on the right-hand side of Fig. 4b, are positive, or greater than background, while those corresponding to the lower writing times yield negative signals. The negative signals are shown a little more clearly in Fig. 5, in which

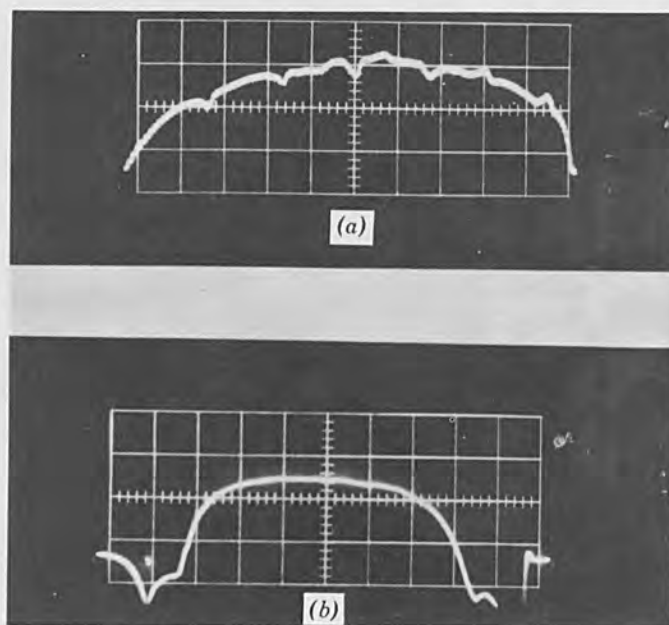


Fig. 4. (a) Oscilloscope trace of lead film before deposition of the polymer. (b) Oscilloscope trace of lead film after deposition of acrolein polymer film at various rates.

the section of the slide containing the polymer formed at the lowest writing times (5 msec and below) was examined in more detail. Figure 6 shows the result of plotting the signal height (both positive and negative) as a function of writing speed. Once again the signal height tends to be linearly dependent on the logarithm of the writing time, even though a negative signal is observed below 20 msec; this is discussed further below.

Previous work on rates of formation of polymer films by electron bombardment (2), (7) have indicated a dependence of rate of formation on both current and pressure. It appears likely, therefore, that the ultimate writing speed may be dependent to a large extent on both the pressure of active gas and on the current in the electron beam, and experiments using acrolein as the active monomer were performed to determine if this was the case. Horizontal scans were made with a fixed writing time (80 msec) and a fixed pressure of 8×10^{-5} torr, each scan being made with a different beam current. The signals resulting from a vertical scan of the polymer film so formed were measured and are shown in Fig. 7.

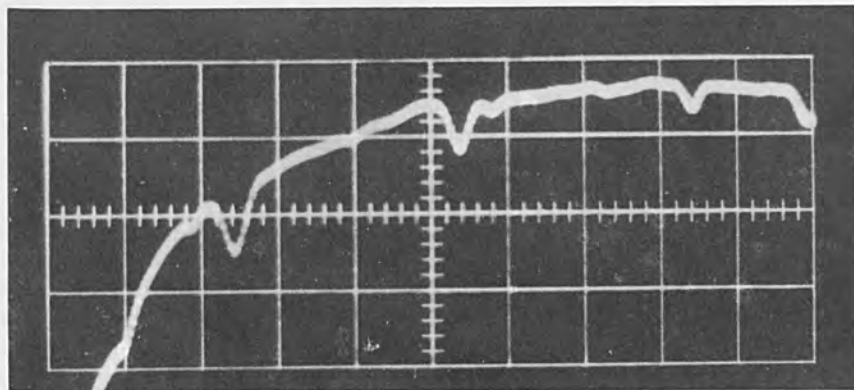


Fig. 5. Oscilloscope trace showing the presence of acrolein polymer strips on a lead film.

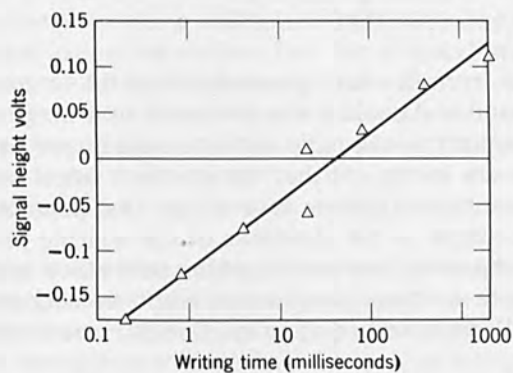


Fig. 6. Signal height as a function of writing speed.

Clearly a variation exists which is almost linear, and it may be concluded that the writing speed may be increased by increasing the current in the beam used to deposit polymer. In a similar manner, the dependence of writing speed on pressure was estimated by carrying out a series of horizontal scans with a fixed writing time (80 msec), and a fixed current $5 \mu\text{A}$ at pressures varying between 2×10^{-5} torr and 2×10^{-4} torr. The results of such an experiment is shown in Fig. 8, in which the signal height is plotted

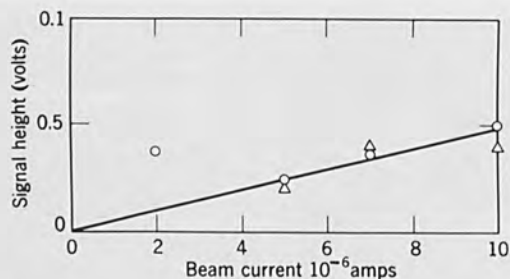


Fig. 7. Signal height resulting from vertical scan of polymer film.

as a function of pressure of active gas. Once again the signal height increases as the pressure is increased, indicating that the writing speed is proportional to the pressure of active gas.

DISCUSSION

It is obvious from the data presented that the technique proposed is basically sound and could form the basis of a large-scale memory. Such a memory would have definite advantages over those of a similar type now in use, in that the electron beam would be used to read the memory in addition to writing. One possible disadvantage of this technique is the slowness of the writing process, and it has been the purpose of the investigation described here to estimate the ultimate writing speed which might be obtainable with sufficiently well-designed equipment. Comparison of the results

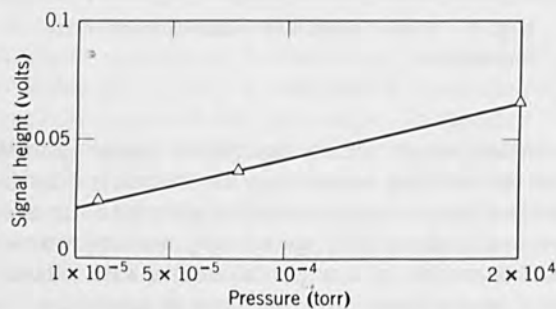


Fig. 8. Signal height as a function of active gas pressure.

obtained with acrolein to those obtained with butadiene indicate that under similar experimental conditions the rate of formation of polymer is quite dependent on the nature of the monomer. Although it is conceivable that a material better than acrolein may be found, all conclusions drawn in this paper are based on the results obtained using acrolein.

Figure 4a indicates that recognizable signals may be obtained with a writing time down to 0.2 msec. The presence of the signals at definite positions on the metal film corresponding to the positions in which the polymer films were deposited, and the fact that the signal height varied uniformly with writing time, would seem to confirm that the signals were real and corresponded to strips of polymer written on the metal film by the electron beam. The observation of a negative signal is rather unusual and deserves comment. As already explained by DaSilva and White (4), the anode current when the beam strikes an insulator is dependent on both the secondary emission coefficient and on the induced conductivity (if any) of the insulator. Under normal conditions, with no induced conductivity, the secondary emission coefficient for an insulator surface is generally greater than that for a metal surface, and this results in a greater flow of electrons from the target holder to the film surface when the electron beam scans an insulator. It was shown that as the insulating films approached 300 Å or less, the conductivity induced in the insulator film by the electron beam caused the signal height to increase as the film thickness decreased to 100 Å.

Obviously at some stage this effect must be reversed, and a region is to be anticipated in which signal height decreases as film thickness decreases. This appears to be the region in which the experimental measurements reported here are being performed. Logically the significance of a negative signal is that the area covered by the polymer layer has a secondary emission coefficient less than that of the surrounding metal, which is not conceivable. An alternative explanation may be advanced as a result of the rather dirty vacuum system used, and the fact already established in this laboratory that acrolein gas tends to corrode a lead film on prolonged contact (8). It is proposed that during the time which elapses between the writing and the reading operations (usually 1 1/2 to 2 hours), the unexposed metal film is corroded by the residual vapors in the vacuum system, that portion of metal film covered by polymer being protected. The result would be anticipated as shown schematically in Fig. 9. The signal from the corroded metal would be increased over that from the bare metal, and

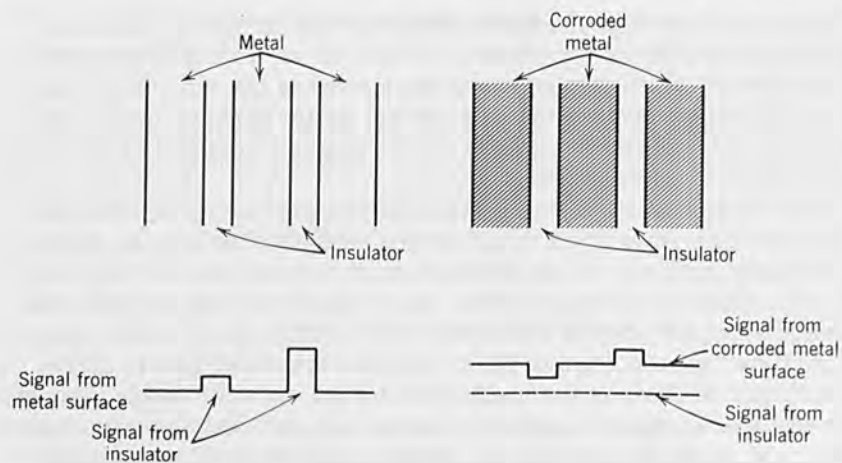


Fig. 9. Schematic representation of anticipated results.

it is conceivable that at some state the background signal might be greater than that from the thinner polymer films. Obviously under these conditions the signals corresponding to the thinner films would appear to be negative, but would still lie on a smooth curve as indicated by Fig. 6.

It seems, therefore, that a writing speed of 0.2 msec might be anticipated using acrolein gas at a pressure of 8×10^{-5} torr and a beam current of $5 \mu\text{A}$. Consider now, the result of altering both current and pressure to a limit imposed only by the emission from the cathode and the energy of the electrons. The diameter of the electron beam used in these experiments was 0.030 in. and the current was 5×10^{-6} A. This corresponds to a current density of 10^{-1} amp/cm². It is anticipated that an electron gun assembly might be obtained with a spot size of 10μ and with a beam current of 10 amp/cm². The data shown in Figs. 6 and 7 indicate that the writing time should increase almost linearly with increasing current. As a conservative estimate, therefore, if we assume a square root dependence, it should be possible to increase this writing speed by a factor of 10.

The upper limit of pressure of 2×10^{-4} torr was imposed by the susceptibility of the oxide cathode by poisoning, however, by construction of a differentially pumped system in which the cathode was maintained in a vacuum of 5×10^{-6} torr, it is conceivable that pressure of active gas as high as 10^{-2} torr might be achieved close to the substrate. Since the writing speed is observed to vary line-

arly with pressure of active gas, this should permit an increase in speed of at least one order of magnitude. It seems realistic therefore that the rate of deposition of a polymer film from acrolein vapor may be increased by at least two orders of magnitude. This would correspond to a writing time of 2×10^{-6} sec per spot.

In addition to increasing the rate of polymer formation, it would seem possible to effectively increase the writing time by use of materials for which the difference in secondary emission coefficients are greater than the difference between acrolein polymer film and lead. This possibility has not been explored and is not included in this evaluation.

As discussed so far, this technique is applicable solely to a read only memory, since no mention has been made of removal of the polymer film. Basically the problem of reversibility can be divided into two different areas, one in which the polymer layer may be coated with a layer of material with a similar secondary emission coefficient similar to the base metal, or one in which the polymer is completely removed from the substrate. The first of these two methods is not considered to be a practical solution, since obviously with repeated use the thickness of the metal-polymer-metal sandwich structure will become excessive and the reversibility will be limited.

The alternate method is complete erasure of the polymer spot. This is considered to be a quite feasible process and one of two different techniques might be used. A large number of polymers, of the methyl methacrylate type, may be degraded to the monomer vapor by simple heating in vacuum [9]. This could, of course, be accomplished using an electron beam as a source of heat and would have as the main disadvantage the possibility of interfering with any polymer material in the immediate vicinity. Alternatively, the polymer layer may be heated in an oxygen atmosphere and removed by a simple combustion process. This technique has been shown by Heide [10] to work well in removing layers of contaminant formed by electron-beam interaction with residual gases in the vacuum system.

CONCLUSION

It has been demonstrated that a new technique is available for construction of a large-scale memory (2.5×10^5 bits/cm²). Information may be written into and read from the memory with the same electron gun. The reading rate, using very simply circuitry,

may be 5 Mc or better, while the writing time has been demonstrated to be at least 0.2 msec/bit and might be anticipated to be two orders of magnitude faster with correctly designed system.

ACKNOWLEDGMENTS

The authors are indebted to Dr. H. L. Friedman for discussions in the initial stages of this work, and to Mr. E. M. DaSilva for the loan of much of the equipment used in this series of experiments.

REFERENCES

1. D. A. Buck and K. Shoulders, Proc. Eastern Joint Computer Conference, Philadelphia, p. 55 (1958).
2. I. Haller and P. White, submitted to J. Phys. Chem.
3. P. White, submitted to J. Electrochem. Soc.
4. E. M. DaSilva and P. White, unpublished work.
5. E. M. DaSilva and P. White, Proc. 8th National Vacuum Symp. p. 830, Pergamon Press, New York (1961).
6. V. K. Zworykin, J. Hillier, and R. L. Snyder, Bull. Am. Soc. for the Testing of Materials 117, 15 (1942).
7. R. W. Christy, J. Appl. Phys. 31, 1680 (1960).
8. L. V. Gregor, private communication.
9. H. W. Melville, Proc. Roy. Soc. (London) A, 163, 511 (1937).
10. H. G. Heide, Proc. 5th International Congress for Electron Microscopy, p. A 4, Academic Press, New York (1962).

ELECTRON BEAMS IN MATERIALS PROCESSING

Section IV

ELECTRON BEAMS IN MATERIALS PROCESSING

proposing the use of electron beams for melting. Related ideas are later seen in Tiede (1915) and his formula for making a variety of metals including some of the refractory and precious interest is seen in Noble, who conducted similar activities in the late 1930's. Hultgren and Pakshalis renewed interest in 1940's, and they discussed the use of electron beams for the melting of high melting point materials.

Activities of real consequence in melting and refining, however, began only in the late 1950's when Hugh Smith and C. A. Hall of Tennessee Metallurgical (Smith et al., 1958, 1959) announced their successful up-scaling of the electron-beam process for melting and refining. Preceding this work by several years, Davis, Calverley, and Lever (1957) had begun activities on electron beam zone refining and crystal growing.

Today, their seven short years later, these initial events and their achievements have been left far behind in a rapidly growing technology.

The two greatest contributions made by electron beams in this area of technology are the commercial realization of melting and refining of the refractory metals and the ability to grow crystals of these metals. Due to the fine distribution, materials like Tantalum and Columbian have virtually become commonplace, while as a consequence of the second contribution, we understand the

may be 5 Mev or better, while the working time has been demonstrated to be at least 0.2 sec and might be anticipated to be two orders of magnitude longer with specially designed systems.

ACKNOWLEDGMENTS

The authors are indebted to Dr. H. L. Friedland for assistance in the initial stages of this work and to Mr. R. M. Galt for the loan of much of the equipment used in this series of experiments.

REFERENCES

1. J. J. Van Turnhout, *Proc. 1961 Int. Conf. on Electron and Ion Beams*, Philadelphia, p. 55 (1961).
2. J. J. Van Turnhout and P. White, submitted to *J. Phys. Chem.*
3. P. White, submitted to *J. Electrochem. Soc.*
4. E. M. Galt and P. White, *Proc. 1961 National Vacuum Symp.*, p. 830, Pergamon Press, New York (1961).
5. V. K. Zvarozh, J. Miller, and R. L. Snyder, *Bull. Am. Soc. for the Testing of Materials*, 117, 15 (1942).
6. H. W. Christy, *J. Appl. Phys.*, 31, 1680 (1960).
7. L. V. Grogan, private communication.
8. H. W. Christy, *Proc. Roy. Soc. (London)*, A, 163, 511 (1937).
9. H. G. Bueche, *Proc. 5th International Congress for Electron Microscopy*, p. A-4, Academic Press, New York (1963).

ELECTRON BEAMS IN MATERIALS PROCESSING

This section considers recent developments in the utilization of electron beams as heat sources applied for the purposes of melting, refining, single crystal growing, and related tasks. Ignoring the accidental meeting of X-ray tube targets in the late 1890's, the formal application of electron beams to this area of technology probably began with the 1907 von Pirani patent (von Pirani, 1907) proposing the use of electron beams for melting. Related ideas are later seen in Tiede (1913) and his furnace for melting a variety of metals including some of the refractories and renewed interest is seen in Ruhle, who conducted similar activities in the late 1930's. Hultgren and Pakkaala resumed interest in 1940's, and they discuss the use of electron beams for the melting of high melting point materials.

Activities of real consequence in melting and refining, however, began only in the late 1950's when Hugh Smith and C. d'A. Hunt of Temescal Metallurgical (Smith, et al., 1958, 1959) announced their successful up-scaling of the electron-beam process for melting and refining. Preceding this work by several years, Davis, Calverly, and Lever (1957) had begun activities on electron beam, zone refining, and crystal growing.

Today, some seven short years later, these initial events and their achievements have been left far behind in a rapidly growing technology.

The two greatest contributions made by electron beams in this area of technology are the commercial realization of melting and refining of the refractory metals and the ability to grow crystals of these metals. Due to the first contribution, materials like Tantalum and Columbium have virtually become commonplace, while as a consequence of the second contribution, we understand the

flow and fracture characteristics of the refractories and the role of impurities in the process. A considerable number of organizations are involved in the continued commercial development and further perfection of the electron-beam process for melting, refining, and their control. Their efforts are directed toward new process development, existing process improvement, and equipment design and construction.

As the field has grown we have seen a continuous increase in furnace capacities, a cost reduction per pound of processed material, and in turn the economical processing of less sophisticated materials is becoming a reality.

We are beginning to see adoption of the electron-beam process to direct extractive metallurgy tasks on the one hand, while we continue research to evaluate the upper limits of purity of materials which adopt themselves to electron-beam heating, melting and refining.

Those interested in additional information on work published in the years preceding this conference are referred to Calverley (1959), Hetherington (1959), Bakish (1960, 1961, 1962, 1962 a), Mishler and Monroe (1962), Morley (1963), and the many references given by von Ardenne in the first presentation of this section.

Von Ardenne's presentation in a number of ways exceeds the scope of this section, but the bulk of the material is truly pertinent here. His plasma references are certainly of interest as a possible complementary materials processing tool. Von Ardenne's paper is also of interest because it acquaints us with work in the field in East Germany, work at a notable scientific and technological level. Patón and Movchan's paper from the Paton Institute, Kiev, also introduces to us thoughts from behind the Iron Curtain. The radial electron-beam heaters discussed are, to the best of my knowledge, a purely Soviet development with many virtues. Hayashi, Fiedler, and Accary and their associates from Japan, East Germany, and France, respectively, acquaint us with their recent experience in electron-beam melting of titanium, steel, and ceramics. These presentations discuss, essentially, applied and almost commercial tasks show the advanced stage and the contributions that electron beams can make. I regret that neither Temescal nor Stauffer-Heraeus (both pioneering corporations) could tell us at this conference about their recent work and hope that they will make contributions in the 1966 conference.

Hughes and his associates, Wilson and Picklesimer, Tedmon and Rose, and Rexer present to us their most recent work on single crystals, considering both problems of crystal growth and crystal

properties. The last two papers are interesting in the meaning of their content relative to the other material here. Doering and Shahinian discuss surface effects of electron bombardment heating, while Gillin presents us with an advanced electron-beam materials testing facility, another area where this heat source has found considerable growth.

In closing these few introductory remarks to this section let me say that I am confident that the future will see considerable additional expansion in the utilization of electron beams for these applications and this broader acceptance is certain to lead to additional per pound processing cost reduction for melting and refining tasks.

Editor

REFERENCES

- Bakish, R., Ed., 1960, Proc. 2nd Symp. on Electron Beam Technology, Boston, Mass.
- Bakish, R., Ed., 1961, Proc. 3rd Symp. on Electron Beam Technology, Boston, Mass.
- Bakish, R., Ed., 1962, "Introduction to Electron Beam Technology," John Wiley and Sons, New York.
- Bakish, R., Ed., 1962, Proc. 4th Symp. on Electron Beam Technology, Boston, Mass.
- Calverley, A., M. Davis, and R. F. Lever, 1957, J. Sci. Instr. 34, 142.
- Calverley, A., 1959, Proc. Symp. Elecelectron Bombardment, SERL Baldock Herts.
- Hetherington, J., Ed., 1959, Proc. 1st Symp. on Electron Beam Technology, Boston, Mass.
- Hultgren, R. and M. H. Pakaala, 1949, J. Appl. Phys. 11, 643.
- Mishler, H. W. and R. Monroe, 1962, DMIC Report 174, Battelle Memorial Institute.
- Morley, J., Ed., 1963, Proc. 5th Symp. on Electron Beam Technology, Boston, Mass.
- von Pirani, M., 1907, U.S. Patent No. 848,600.
- Smith, H., 1958, "Vacuum Metallurgy," R. Bunshah, Ed., Reinhold Publish Corp., New York.
- Smith, H. C. d'A and C. W. Hanks, 1959, J. Metals 2, 112.
- Tiede, E., 1913, Ber. Deut. Chem. Ges. 46, 2229.

SOME NEW CONTRIBUTIONS IN THE FIELD OF ELECTRON-BEAM AND PLASMA-BEAM TECHNOLOGY

INTRODUCTION

For a beginning I should like to present a comprehensive survey of the more recent investigations undertaken by our Dresden Research Institute in the field of charge carrier beam technology. These investigations have grown in the spiritual climate of a tradition whose beginnings date back to the time preceding the Second World War.

EFFORTS DATING FROM PRE-WAR PERIODS SAW THE ORIGIN OF PRESENT ENDEAVORS

Reminiscences of the development of these past periods shall be recalled to contribute to the knowledge of the historical foundation of the technology. In this connection I may be permitted to mention some of my own contributions in this field.

1. The development in 1928 of the triode electron gun, operating with accelerating voltages of some 1,000 V and beam currents up to 1 mA, incorporating a thermionic cathode and a control electrode negatively biased to such a degree that beam cross-over is obtained [1].

2. The development of the "flying spot scanner" operating with an electron-beam tube, in 1930 [2]. Note Fig. 1 showing the report in the New York Times of August 16, 1931.

*Research Institute, Weiser Hirsch, Dresden, East Germany.

†This paper was the opening presentation at the conference. It is placed as the first paper in this section, in keeping with the aims of the volume.

The New York Times.

Cathode radio television station on which Baron Manfred von Ardenne of Germany has been experimenting since 1928. The transmitter and receiver (inset) will be exhibited in a forthcoming Berlin Radio Exposition. The images are seen on the end of the tube in the square aperture of the receiver.

The Flying Spot Scanner

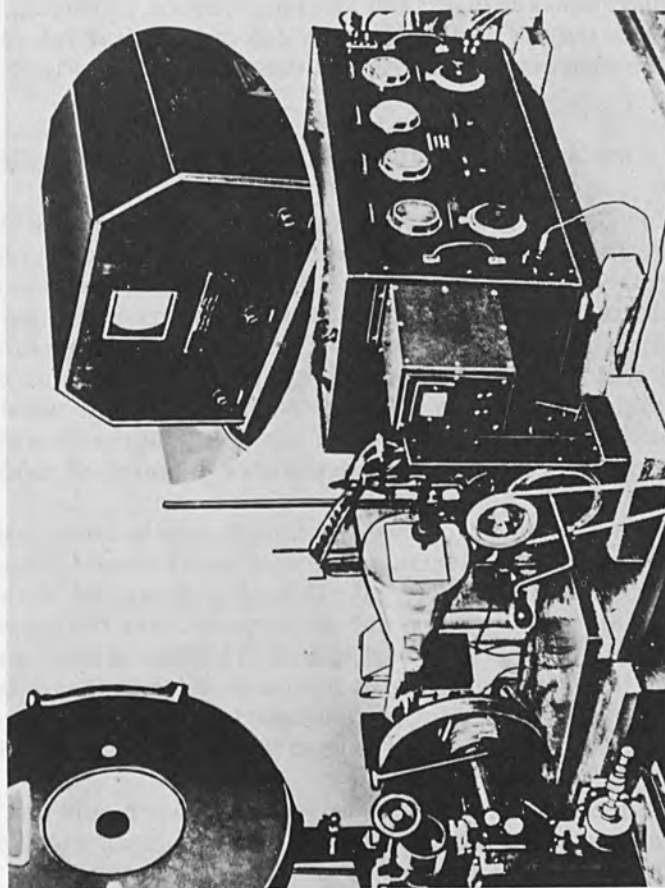


Fig. 1. Report on the Flying spot scanner published in the New York Times of August 16th, 1931.

3. The discovery in 1935 of the emission of negative ions from thermionic cathodes [3].

4. The electron microprobe and the electron-scanning microscope, developed in 1937 [4]. Note Fig. 2 illustrating the ray path, from the original.

5. The method of using the electron microprobe for micromanipulations, described in 1938 [5].

6. The proposal of the X-ray projection microscope made in 1939, and further detailed in 1940 [6]. Note illustrations in Fig. 3 from the original.

7. The development in 1939 of the ion microprobe and its application for micromanipulation work, e.g., drilling extremely fine holes and canals, obtaining point reactions, and similar operations [7]. Note illustration in Fig. 4 from the original.

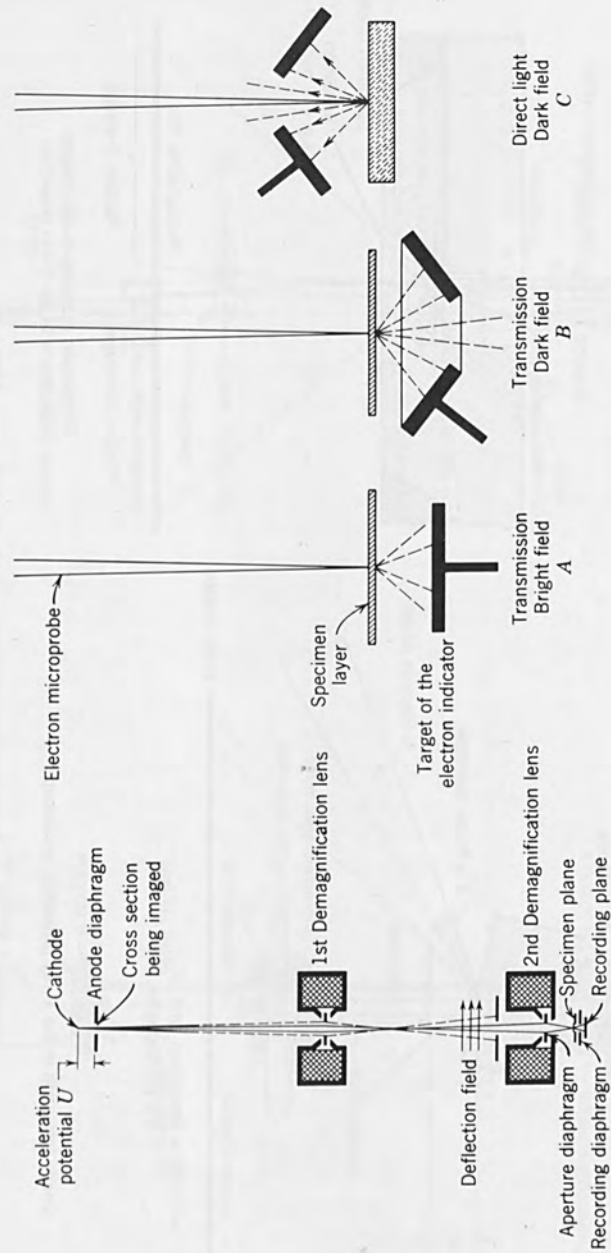
8. The development in 1944 of an apparatus for producing microscopes with the aid of demagnifying electron optical ray paths [8]. Note the cross-sectional view of the equipment in Fig. 5.

WORK IN THE FIELD OF ELECTRON-BEAM TECHNOLOGY

On the foundation provided by these earlier efforts, our Dresden Institute has in recent years concentrated its activity in the field of electron-beam technology on projects where the high energy of the electron beam is either utilized for micromachining of workpieces, or for heating processes. Thus, considerable effort has been directed to the development of universal apparatus for micromachining workpieces with electron beams, to the development of multichamber electron-beam furnaces and the development of high-vacuum electron-beam evaporation equipment on modern lines.

Micromachining using focused electron beams became possible with the development of electron probes of small diameter and high-power density in 1937 [4], [5]. The initial trials for the production of small holes with the aid of charge carrier beams were carried out, even at that time, using ions [7]. This basic work has today grown into its own extensive field with electron-beam thermal micromachining reaching the engineering stage, particularly through the work of Steigerwald [9] and the recent work of our institute [10], [11].

Although in our work in 1947, Steigerwald's work, and that of the research groups at Hamilton-Zeiss, and the Japan Electron Optic Laboratories, the cross-over of the electron gun was



Ray path in the electron scanning microscope

Schematic view of the different modes of illumination with the electron scanning microscope

Fig. 2. Electron microprobe and scanning electron microscope (von Ardenne, 1937).

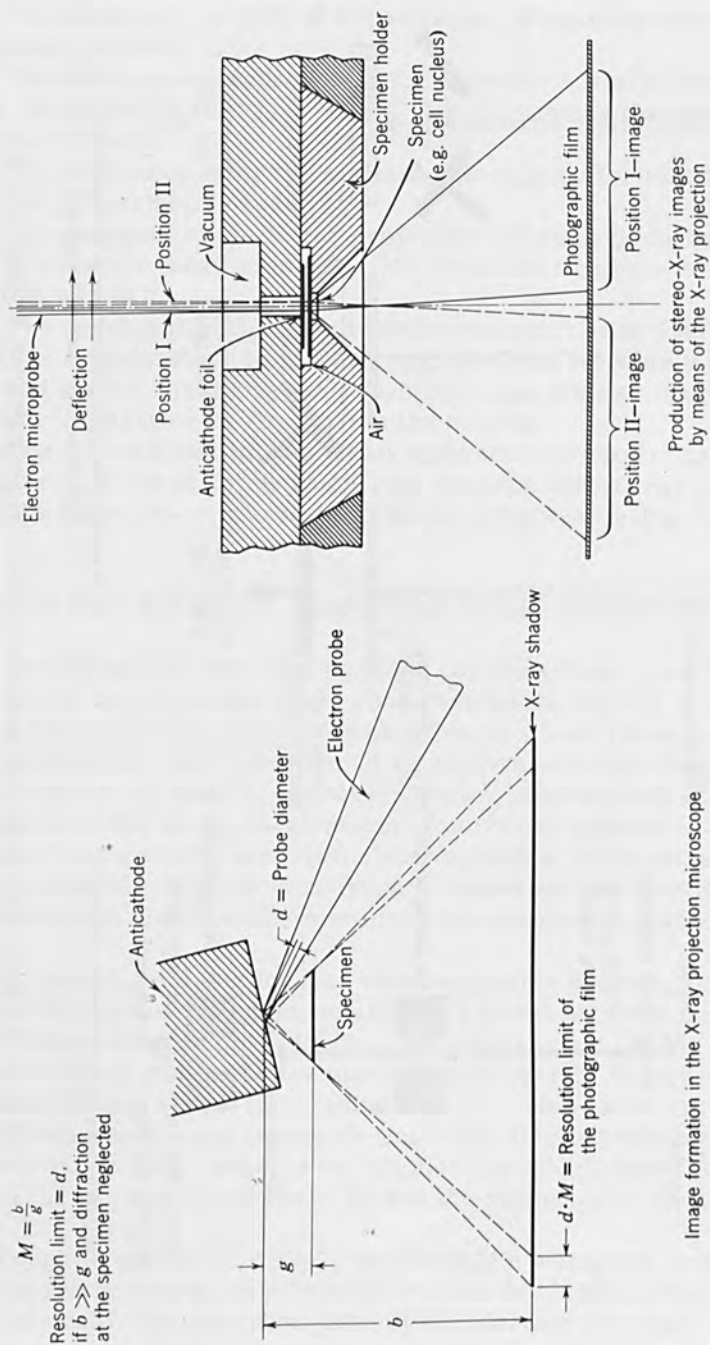
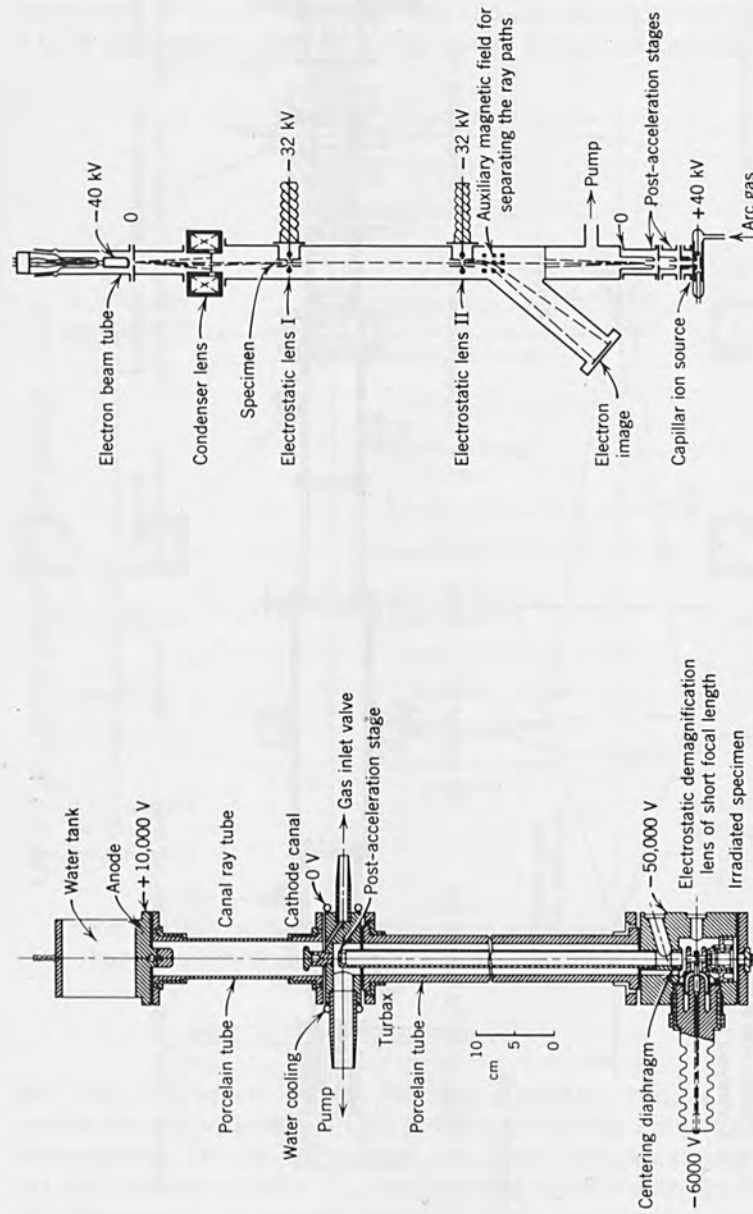


Fig. 3. X-ray projection microscope (von Ardenne, 1939).



Electrostatic electron microscope combined with an ion probe

Simple arrangement for producing the ion micro probe

Fig. 4. Ion microprobe and visually controlled micromanipulations with this probe (von Ardenne, 1939).

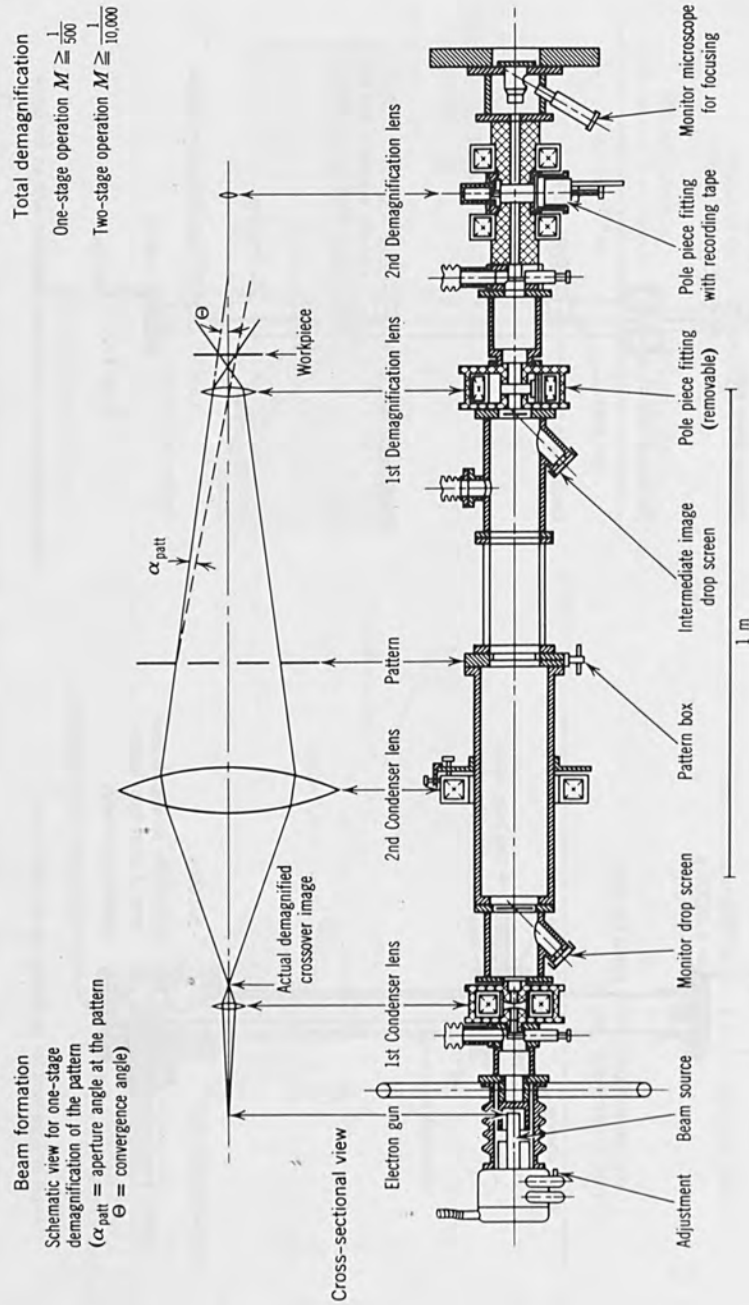


Fig. 5. Microscale equipment (von Ardenne, 1944).

directly imaged on the object by means of one magnetic lens, our institute has recently developed apparatus operating in the two-step mode with one matching lens and one demagnification lens, as Fig. 6 illustrates. The two-step image technique has the advantage

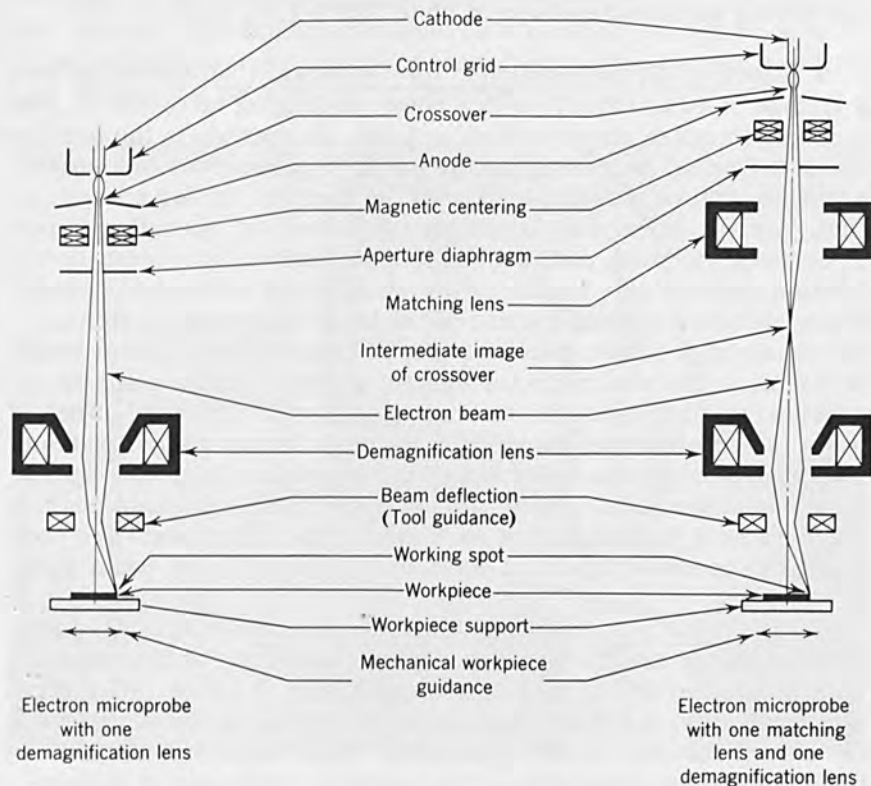


Fig. 6. Ray paths of electron-beam working units for thermal micromachining.

that with a fixed workpiece, the spot diameter, e.g., of 1 to 50 μm , can be varied with only slightly more additional electron optic components. Furthermore, with the latter arrangement, the spherical aberration constant C_o changes only a little with the variation spot diameter. From the aspect of power density this assumes considerable importance.

Three types of apparatus have been developed in our institute for the purpose of thermal micromachining:

1. Electron Probe Apparatus, specially suitable for thin-film and foil machining operating with a maximum voltage of 70 kV and having an average power of up to 350 W [10], [11].

2. Electron-Probe Apparatus for Universal Use, operating with a voltage of 50 to 150 kV, with a power averaging up to 150 W, and a maximum pulse energy reaching 2 kW. In addition to the familiar mode to control the energy on the workpiece by means of a pulse generator, this apparatus also features line scaling for energy-time control. According to special requirements, operation is possible using the beam path with or without control by a beam-deflection system, only requiring the exchange of some components. When operating without the use of the beam-deflecting system, extremely high-power densities can be obtained with a focal length of 7 mm. A mechanical motor control in the x - and y -plane is provided to afford specific machining also with this mode. The beam is centered with the aid of a magnetic centering coil system, which permits the displacement of the beam parallel to the aperture diaphragm, and the inclination of the beam at a point near the workpiece independent of each other. The adjustments are controlled by means of electron collectors inserted in the beam path.

3. Electron Optic Micromachining Apparatus for direct image of the inserted pattern on the workpiece, operating with a maximum voltage of 150 kV and an average power of 150 W. By means of a condenser, the beam cross-over is imaged on to the pattern (25 mm in diameter) in this apparatus. Time sequential and uniform convergent irradiation of the pattern is obtained by deflecting the beam in a line scanning mode. Electron optic means are utilized to project the image on the workpiece which has an area of some square millimeters.

A look into one of our laboratories for beam tools is shown in Fig. 7, with a 150-kV electron-probe apparatus seen in the foreground and the electron micromachining apparatus right behind it.

For many purposes of thin-film and foil machining, it has been our experience that a simplified apparatus is quite sufficient, e.g., analogous to our 70-kV apparatus. This applies for work where the thickness x of the material to be machined is smaller than the range S of the electrons in the workpiece. With thin-film machin-

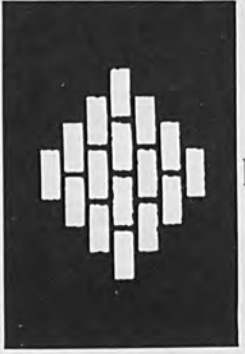
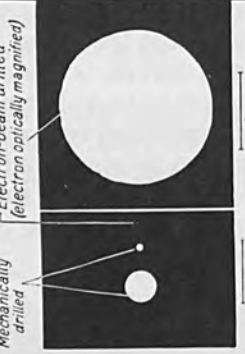
ing, the working speed is so high that the dwelling time of the beam at any point of the workpiece resulting from the beam control of the deflecting system is only 1 to 100 μsec , as is the case with pulse operation, and thus does not give rise to a thermal hazard for the immediate vicinity. This continuous current mode with omission of the pulse generator may also be employed if the thermal stress of the workpiece requires a reduction of total power. "Line pulse operation" on the processing face can be realized when line scaling during scanning is resorted to.



Fig. 7. View of one of the laboratories for electron beam tools with a 150-kV electron microprobe equipment (front) and an electron-optical 150 kV for micromachining of work pieces.

For complicated machining processes in the electron-probe apparatus, outside patterns are, for example, scanned by a flying spot scanner and thus utilized for machining control. An inserted pattern is imaged directly on the workpiece with the electron optic micromachining apparatus.

Three typical examples of thermal micromachining are shown in Fig. 8. It illustrates a machined micromodule resistor unit, a tantalum grid, and an electron-optic three-hole diaphragm. When

Electron-beam machining		Thin films	Foils	Bulk material
Electron range in material S_e Thickness of material x		≈ 1	$\approx 1 \approx \frac{d_f}{x}$	$\ll 1$
Image of product				
		Micro-module resistance	Frame-lock grid structure	Three-hole diaphragm
Product		Electron beam evaporating of thin film	Preliminary test. Machining of insets by scanning	For the objective of an electron microscope
Remarks		Chrome (20)–Nickel (80)	Tantalum	Platinum
Material				
Thickness of material x		$\approx 0,5$	5	100
Width of evaporated line		10	—	—
Resistance value before machining		200	—	—
Resistance value after machining		—	—	—
Width of strut		—	20	—
Spacing of scanned area		—	20	—
Diameter of hole D		—	—	20

d_f = Diameter of focus on work piece

Fig. 8. Typical results of the thermal micromachining with electron beams

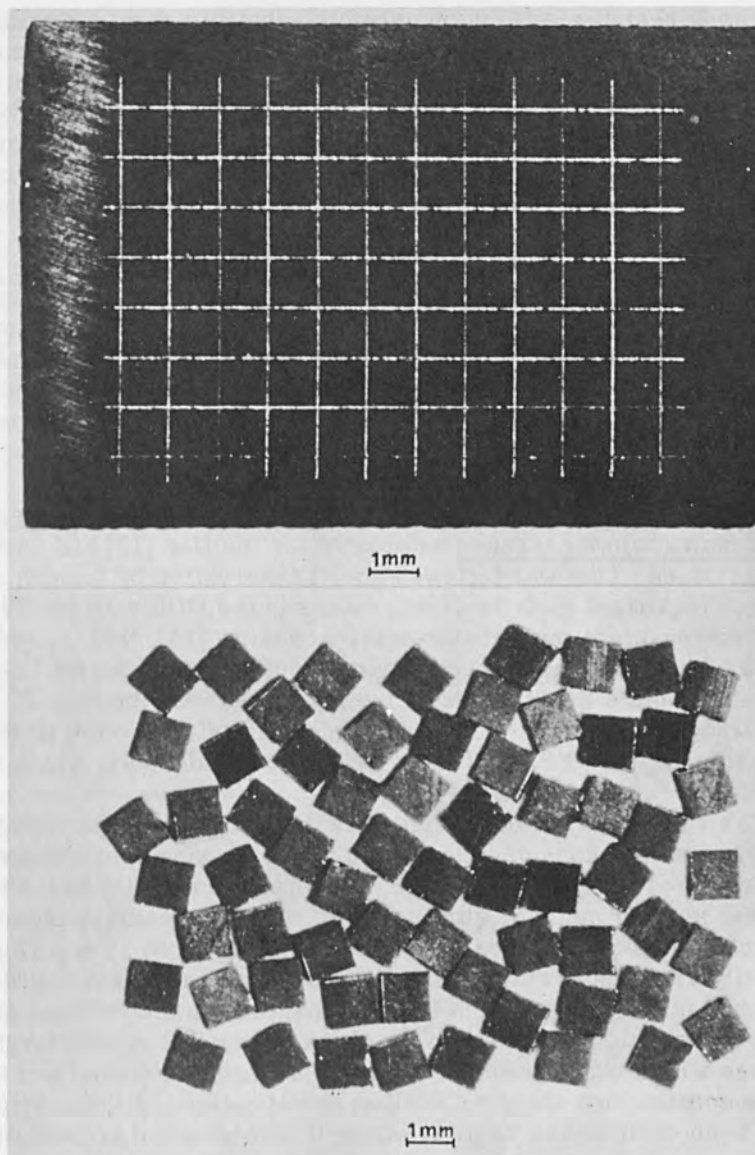


Fig. 8a. Ge single crystal, thickness 0.2 mm, diced by electron-beam machining cutting depth 0.1 mm, before breaking (left) and after breaking (right).

machining resistance films on ceramic substrate, e.g., a working speed of about 200 cm/sec could be obtained using beam currents of 200 μ A at the worked spot with continuous current operation and an electron energy of 60 keV. A working speed of 100 cm/sec at a beam current of 900 μ A was obtained when using 5- μ tantalum foil for the manufacture of a grid. Figure 8a shows a germanium single crystal, diced by electron-beam machining before breaking (left) and after breaking right).

Work on heating (melting and evaporation) of metals in high vacuum by means of focused electron beams began in our institute in 1953, operating with several kilowatts. Already the first concept [12] was characterized by a high-flow resistance separating the main furnace chamber and the electron gun chamber. For assuring adequate vacuum, even in the event of gaseous eruption from the heated metal in the main chamber, the electron gun chamber is separately evacuated (multichamber principle).

In 1959 we began the development of 45 kW and 60kW electron-beam multichamber furnace equipment for melting [13] and casting [14] work. This was followed shortly thereafter by furnishing several important metallurgical centers in the GDR with the 200-kW electron-beam multichamber furnace units [15], [16], shown in Fig. 9. Since then many metallurgical centers in the Soviet Union and the People's Republics have been supplied with the type 200-kW furnace units. In this connection, Fig. 10 offers a view of the assembly hall of VEB LEW Hans Beimler, Hennigsdorf, which now manufactures these furnaces.

With respect to the development in the field of vacuum metallurgy, scientific considerations resulted in passing over the stage of vacuum-arc methods [17], and we proceeded directly to the high-vacuum melting method, with the aid of electron beams, which is also utilized for large-scale steel refining. Figures 11 and 12 present illustrative drawings of our 1700 kW electron-beam multichamber furnace unit, which is being installed in the refining steel plant in Freital, near Dresden, to cite an example. A still larger furnace with a beam power of 6000 kW has been projected and is in the construction stage.* Detailed specifications of these furnaces have been compiled in Table I, where it will be noted immediately that for every type, including the large units, only a single two-pole electron gun is used. This and the high degree of reliability

*Although one does not deny the possibility of building a 6-MW furnace, the writer wonders as to the desirability to use such large power in a single gun installation (Ed.).

Table 1

Type (operation start)	Electron beam gun (W-Bulk cathode. Heating by electron bombardment)			Vacuum pumps (Furnace chamber final pressure 10 ⁻⁵ Torr)			Feeding	Ingot																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																		
	Max. beam power	Accelerating voltage (adjustable)	Beam current	Pervance	Gun type	Min. spot diameter			Max. spot diameter	Total pumping speed	Tube to diffusion pump	Pumping speed	Method	Diameter or edge length resp	Max. length	Max. diameter	Max. length	Max. weight																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																								
	kW	kV	A	AV ^{-3/2}		mm	mm	ls ⁻¹	Furnace chamber ls ⁻¹ (10 ⁻⁴ Torr)	mm	ls ⁻¹ kW ⁻¹	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm	mm

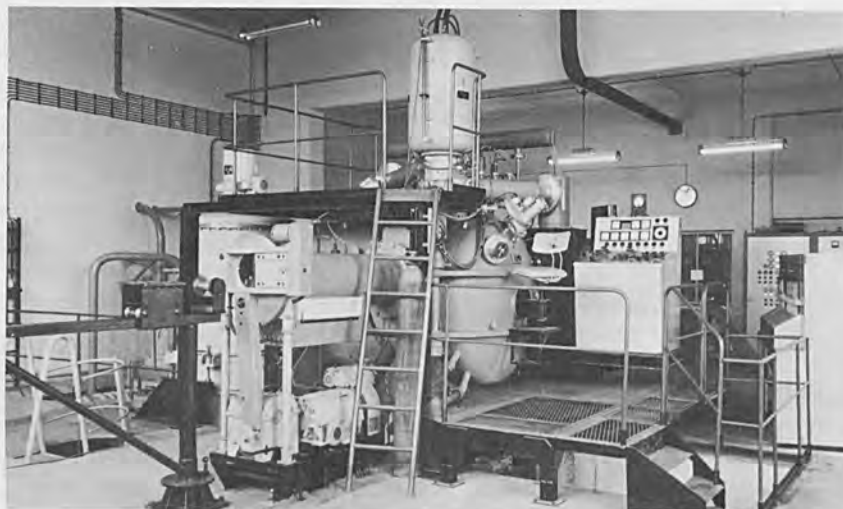


Fig. 9. View of a 200-kW electron-beam multichamber furnace for improving the properties of metals by remelting in high vacuum. In operation in the refined steel works Freital near Dresden.

of the gun assembly provide for particularly convenient operation. Continuous charging, the use of the continuous casting delivery, and the incorporation of vacuum locks, together with the above-mentioned features make for a high rate of production of the three large models, and provide ingots of excellent homogeneity.

Details concerning our ultra-high-vacuum electron-beam evaporation equipment [18] which is suitable for industrial production application, will be reported on elsewhere.

WORK IN THE FIELD OF ION-BEAM TECHNOLOGY

Since the earliest work, dating prior to the Second World War, we have been interested in the problem relating to ion sources. Out of this interest the duoplasmatron ion source [19] resulted in 1948. Its first industrial type [20], [21] is shown in Fig. 13. On continuous operation the ion emission current attains a value of $I_B = 1$ amp, operating with an emission opening diameter of $D_{em} = 1.4$ mm, using H_2 -gas charge and a 5-amp discharge current. Emission current density for ions amounts to $j_o = 65$ A/sq cm, and for electrons to $j_o = 500$ A/sq cm. The proportion of protons in the

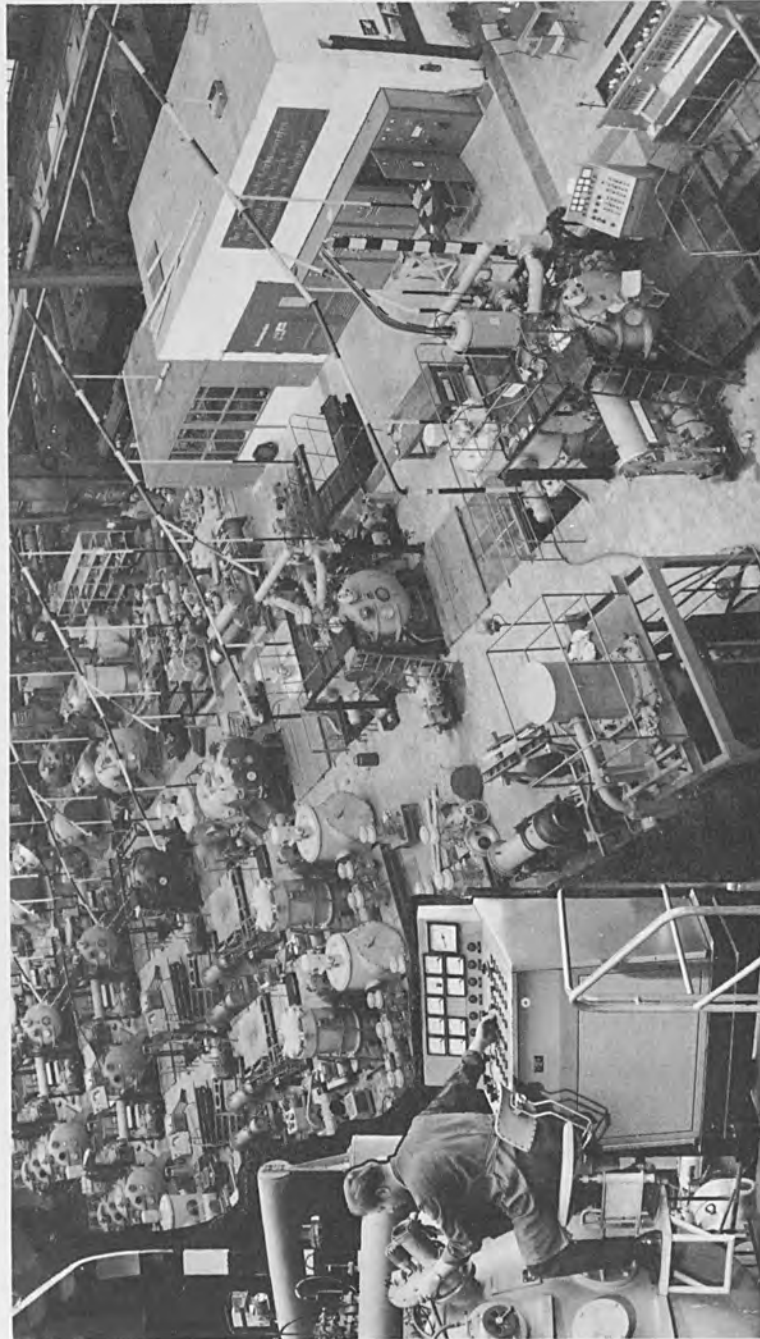


Fig. 10. View of the assembly shop for 200-kW electron-beam multichamber furnaces in the production works VEB Hans Beimler, Hennigsdorf. Summer 1963.

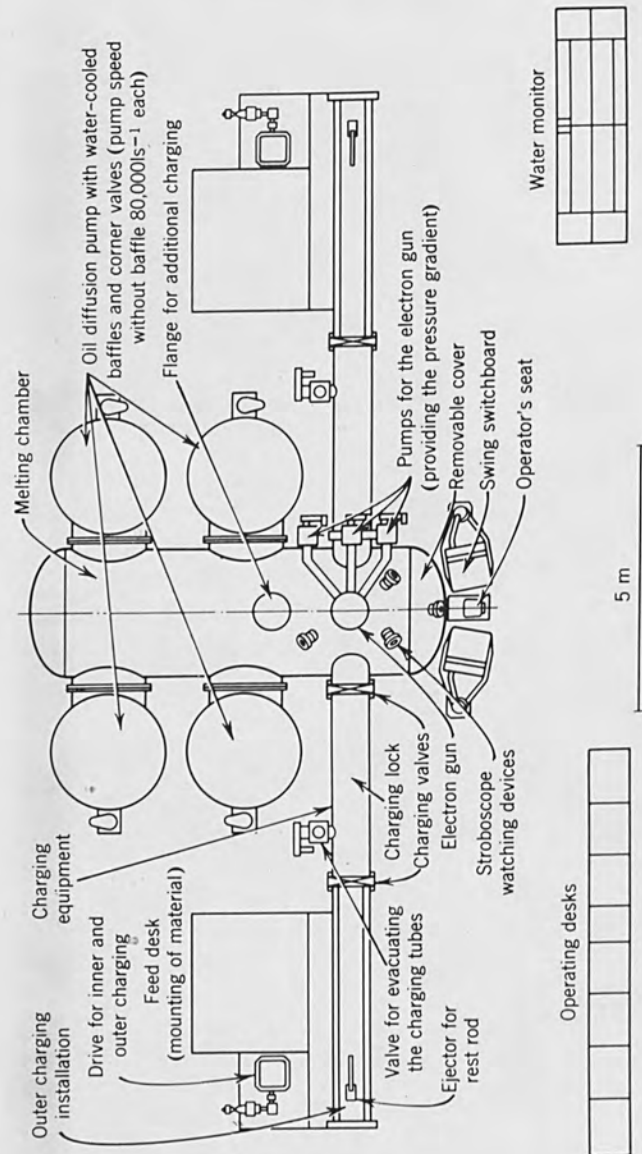


Fig. 11. Plan view of the 1700-kW electron-beam multichamber furnace.

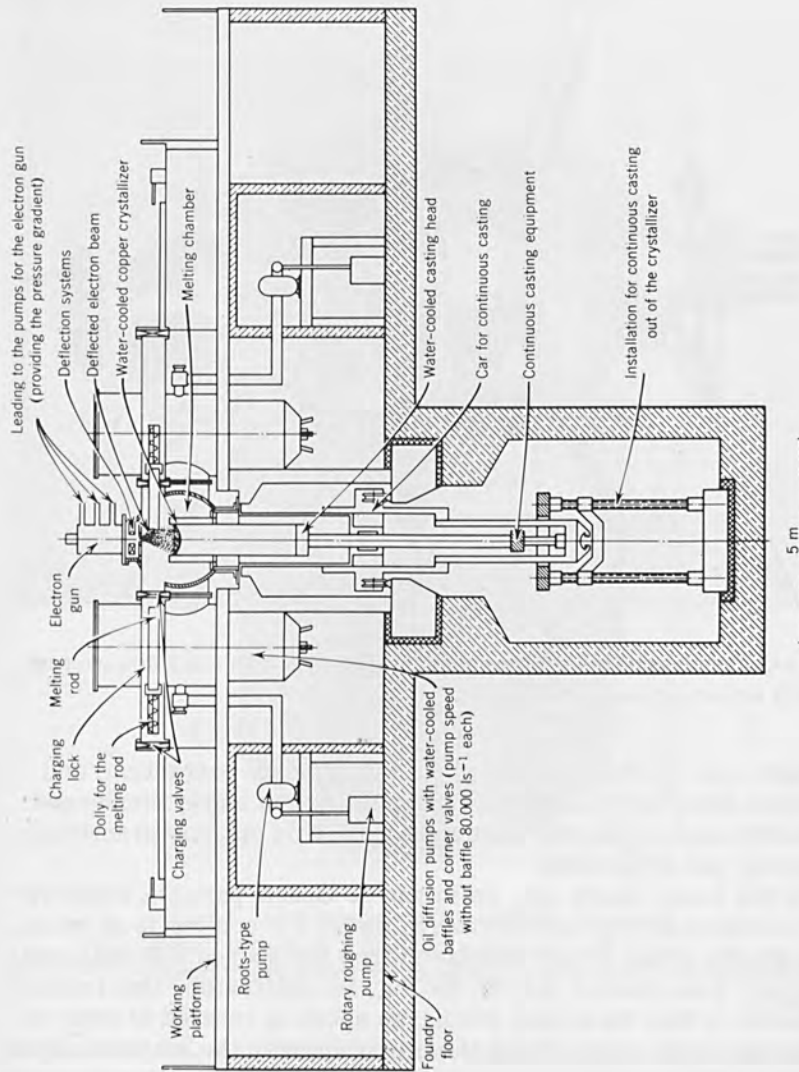


Fig. 12. Vertical cross section of the 1700-kW electron-beam multichamber furnace.

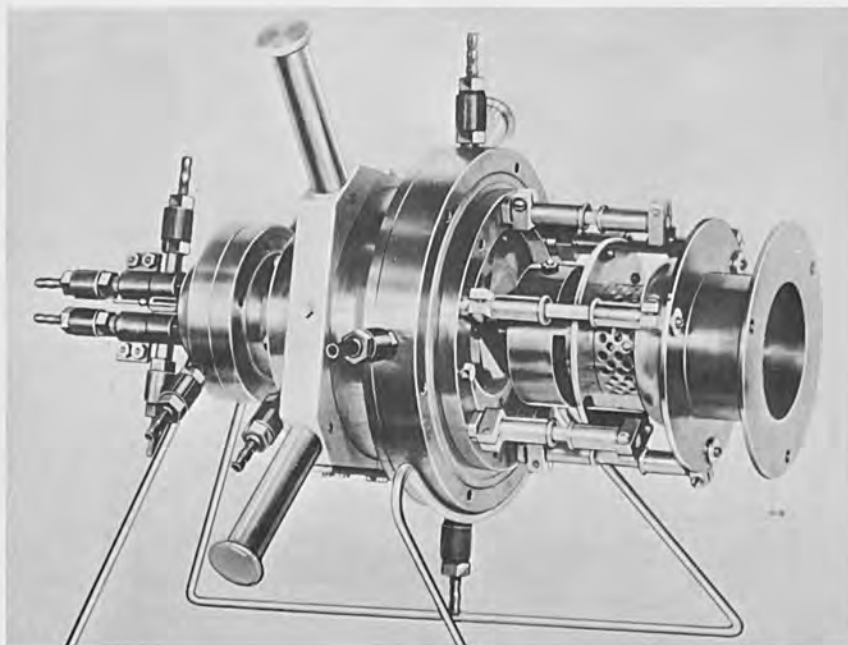


Fig. 13. Duoplasmatron ion-source with extreme high emission current density and an ionisation efficiency $\zeta \approx 1$.

hydrogen-ion current reaches about 60% with ion extraction, and ionization efficiency is practically 100%. In the highly condensed and highly ionized plasma before the emission opening practically no neutral gas is present.

For the small-scale use, as in van de Graaff particle accelerators and cascade-type accelerators, where air cooling is of value, the bakeable small design duoplasmatron ion source unit with metal gaskets was created in 1962, as Fig. 14 illustrates. One feature of the unit is that the actual discharge anode is located in front of the emission electrode. Thus the electrode with the emission opening can be given a potential exceeding that of the cathode potential only by about 20 V (potential of the intermediate electrode). As a result of this technique, an initial energy of about 100 eV is obtained for the ions flowing through the emission opening. Together with the opening diameters of the anode and the emission opening, this initial energy has been balanced in such a manner that a sputtering effect is created by the 100 eV ions, keeping emission

openings, even of narrow configuration, free of contamination for long periods.

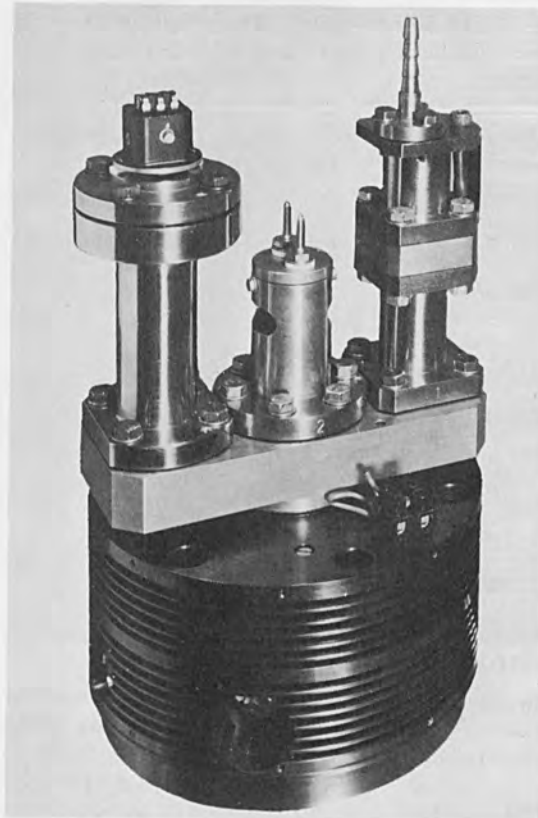


Fig. 14. Duoplasmatron ion-source for small accelerators as a bakable miniature unit with metal gaskets and air cooling.

The most important data of this ion source covering two designs with differing emission opening diameters, provided for two special purposes, have been compiled in Table II. In consideration of air-cooling aspects, the discharge power is kept low with the use of a discharge current of $I_A \leq 1$ amp. In consequence, ionization efficiency is not as high as this type of source can principally attain.

Since 1942 the problem of magnetic mass separation has been

Table II

Data of the Duoplasmatron Ion-Source
in Bakable Miniature Set-Up with
Metal Gaskets and Air-Cooling

Diameter of the emission hole	D_{em}	0,14 (Canal 1 = 0,12)	0,6	mm
Cathode		M-type (Ba-Sr-Ni-Powder-cathode)		
Cathode heating power	P_H	25		W
H ₂ -gas pressure	P	$4 \cdot 10^{-2}$		Torr
Discharge current	J_A	0,8	1,0	A
Discharge voltage	U_A	135	150	V
Initial energy of the ions	eU_{i_0}	≈ 100	≈ 100	eV
Ion beam current	J_B	0,08	6	mA
Potential of the ex- traction electrode	U_B	8	10	kV
Ionization efficien- cy	η	7	23	%
Leak rate	Q	$2 \cdot 10^{-4}$	$3,6 \cdot 10^{-3}$	Torr $1s^{-1}$
Ion economy			$3,5 \cdot 10^{-2}$	$mA W^{-1}$

included in our tasks. In this field the Dresden mass separator with small-size-magnet for high-mass transport was conceived in 1955 and reached practical construction a few years later. The design of the Dresden separator [22], [23] is shown on the photograph of Fig. 15. By incorporating a novel ion-optic path concept the separating magnet could be essentially reduced. For a large plant about 200 tons were required, with only 10 tons of the same steel quality or 20 tons using steel quality 2 being needed for the new unit of equivalent performance. It is characteristic for the utilized ion-optic beam path configuration that a beam crossover of the ion beam leaving the source is obtained in the 60 degrees separating magnetic field by the suitable curvature of the plasma

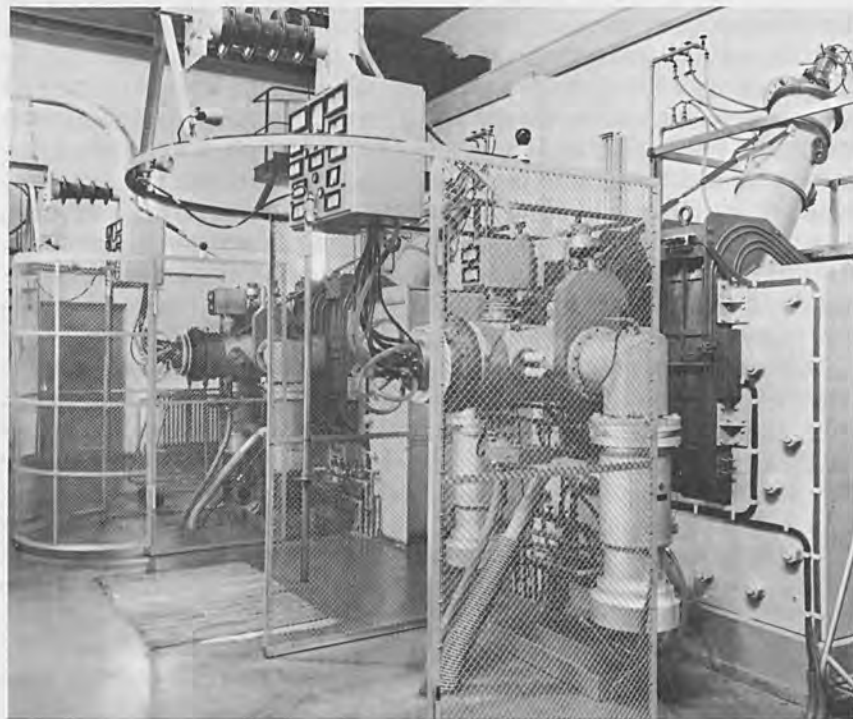


Fig. 15. Two electromagnetic mass separators for high mass transport having a magnet of small bulk (Manufacturer: VEB Vakutronik Dresden) and having been mounted at the Institute for Physical Separation of Substances.

column and the optical system of the ion source. This permitted the reduction of the gap between the magnetic poleshoe plates to $a_p = 7$ cm, and the magnetic separation field volume to 0.033 cu m.

A hot cathode vapor discharge source serves as ion source, operating with an axial auxiliary magnetic field, artificially increased cathode drop, and lateral ion extraction, and it is constructed in a specialized unit system assembly design. Thermal isolation of the substance vaporizing oven from the ionizing chamber block is a special feature, which prevents undefined additional heating of the vaporizing oven due to added energy supply from the ion source discharge and from returning parasitic electron currents. This provides for a very reliable function of the source, even when charged with easily vaporized substances.

The generation of gas from ion-beam initiated oil vapor cracking has been considerably restrained by severely reducing the

partial pressure of oil vapor in the beam chamber. In consequence, the evacuating system display for charging the ion source with metals, e.g., magnesium, could be brought down to a pumping speed of only 2000 liters/sec without the working pressure during operation rising above about $1.5 \cdot 10^{-5}$ torr. Total ion-beam current attained a level of up to 100 mA, operated with an accelerating voltage of 40 kV and with the source charged with magnesium, of which about 80% could be directed into the pockets of the collector. Separating data offered elsewhere [24] show that the enrichment attainable with the Dresden magnetic separator compares very well with the performance obtained with the familiar older plants for magnetic mass separation.

Intense investigations carried out since 1955 have given new impetus and direction to mass spectrographic research with molecule attachment mass spectrography. Fig. 16 shows the Dresden Molecule Mass Spectrograph [25], [26], [27] which was used to obtain the results given in subsequent lines.

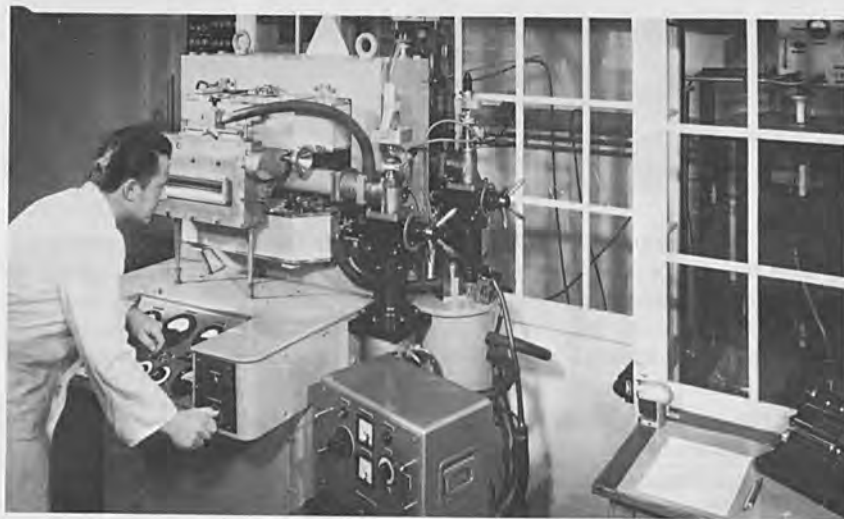


Fig. 16. The molecule mass spectrograph of Dresden with ionisation of polyatomic molecules by attacking electrons or negative ions of very low kinetic energy.

Samples which we have received from local establishments and from abroad for examination reached such an extent in 1963 that the development and manufacture of a simplified type of molecule attachment spectrograph unit seemed indicated for routing pur-

poses. In our molecule mass spectrographic work, molecule ionization is not effected with the conventional electron impact. Rather, we employ a cautious ionization of polyatomic molecules by attaching negative charge carriers with very low kinetic energy. Generally, it is possible to avoid the dissociation of the molecules when using the molecule attachment mass spectrography working with negative ions, which yields clear and simple spectrum presentation. Even when investigating mixtures of substances having differing molecular weight, confusion is avoided in most cases.

Compared with the ionization cross section, when generating positive ions by electron impact at an energy level of 100 eV, the average attachment cross section of attaching thermal-energy or nearly thermal-energy electrons to such types of molecules having an electron affinity sufficient for our purpose, is 2 to 6 orders of magnitudes lower. Frequently electron attachment molecule spectra with their very low intensity remained in obscurity in the past for this reason. With the advent of an electron attachment ion source of high working efficiency [25], [26], electron attachment mass spectrography became amenable to handling polyatomic molecules, offering convenient visual observation and photographic or oscilloscopic recording of spectra.

A significant extension of the scope of application of this kind of mass spectrography could be attained in 1962 with the introduction of the mechanism of negative ion attachment to test-vapor molecules [29]. By utilizing this mechanism, which could be verified by a marking mode using enriched stable isotopes, it becomes possible to analyze substances by attachment mass spectrography which it had been impossible to handle previously because their electron attachment cross section proved too small. Thus, utilizing the negative ion attachment mechanism the first molecule attachment, mass spectrum data could be obtained in 1962 of paraffines [29] and for example of oils [30].

In the illustrated Dresden unit, the ions leaving the emission slit of the ion source are accelerated by a voltage of 40 kV. The resulting ion beam, which is practically monoenergetic, is sharply focused in the center on the plane of the photoplate by means of the electrostatic cylindrical Einzel lens and the mass separating magnetic field. The aperture slit of the ion beam has been shaped in such a manner that adequate definition in depth is obtained to produce a sharp line across the entire recorded spectrum. Resolution of the unit practically assumes values between 2000 and 4000. Using an ion-image converter, most of the lines can be visually inspected before recording. As a special feature of the unit, a

changeover switch is provided permitting the recording of positive ions, aside from taking negative ion spectra. In addition, the unit affords qualitative analysis of the energy structure of the examined molecules by means of the mass spectrographic analysis of the progressive splitting of the molecules with thermal dissociation increased from recording to recording. Up to the present date, about 10,000 spectra have been obtained, of which some typical examples shall be shown.

Of these, Fig. 17 shows three spectrum records illustrating the increasing disintegration of the coronene molecule with rising discharge current at the ion source (rising plasma temperature). With a low discharge current of 10 mA, a normal spectrum (A) is obtained showing the most pronounced line at the parent peak 300. After previous hydrogen separation, a weak line at peak 315 appears due to ion attachment (O^-). When the discharge current is increased to 120 mA the separation of the hydrogen-atoms from the molecule begins, as shown on the next spectrum (B). The carbon structure is practically retained. The latter also disintegrates as illustrated on the spectrum (C), when the discharge current is further increased to 200 mA. These examples show the usefulness of this method for obtaining an indication of the bond structure in molecules.

Some of the commercially available oils for diffusion pumps have been analyzed, the molecular mass spectrum data of which are presented on Fig. 18. Oils of this kind are obtained from petroleum, partially by means of specialized distilling. They represent fractions ranging in mass value across 350 to 700. Despite the multiplicity of the components, the group formation at intervals of 14 mass units is evident. Our compilation shows the spectrum recordings of Diffelen Ultra (A) and of Diffoil (C). A comparison of the spectrum presentations (A) and (B) illustrates the alteration this kind of oil experiences because of cracking when exposed to air. During this event, fission products appear with lower molecular weight. Another spectrum (D) records the spectrum of polyphenylether (OS 124 of Monsanto Chemical Co.), which represents a vacuum oil used for very-high-vacuum diffusion pumps, showing four pronounced lines. The spectral line at peak 445 is 1 mass unit under the polyether line, having five benzene rings in the molecule. The distance between the other lines amounts to 92 mass units each, and results from the separation of the phenol groups.

In recent years, about 1000 natural substances have been examined with the Dresden method. Figure 19 shows an arbitrary selection, with spectrum A referring to Gibberallic-acid-methyl-

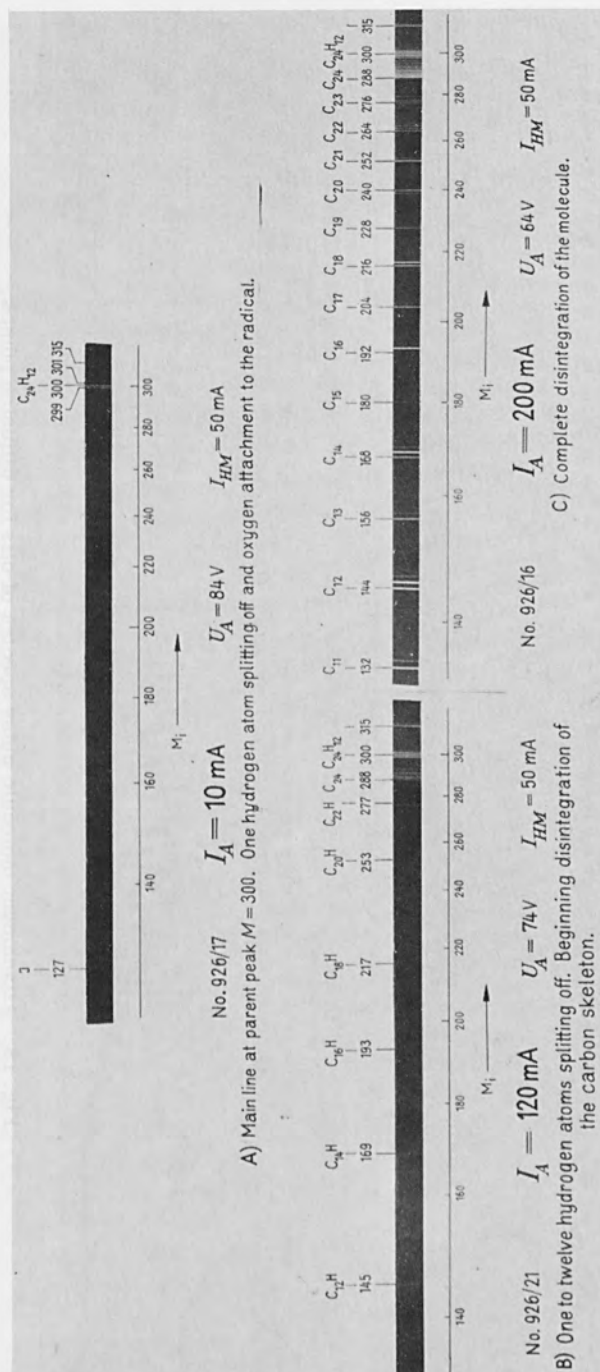


Fig. 17. The proceeding splitting of the coronene molecule with increasing discharge current.

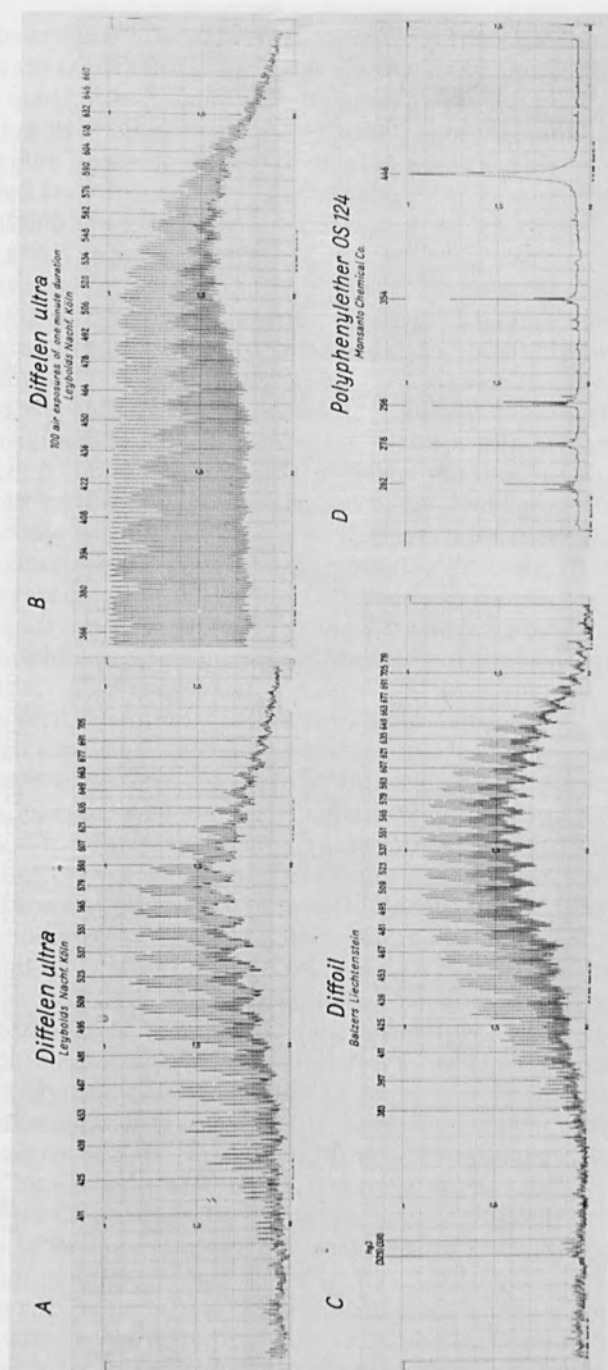


Fig. 18. Attachment molecule mass spectra of oils for diffusion pumps.

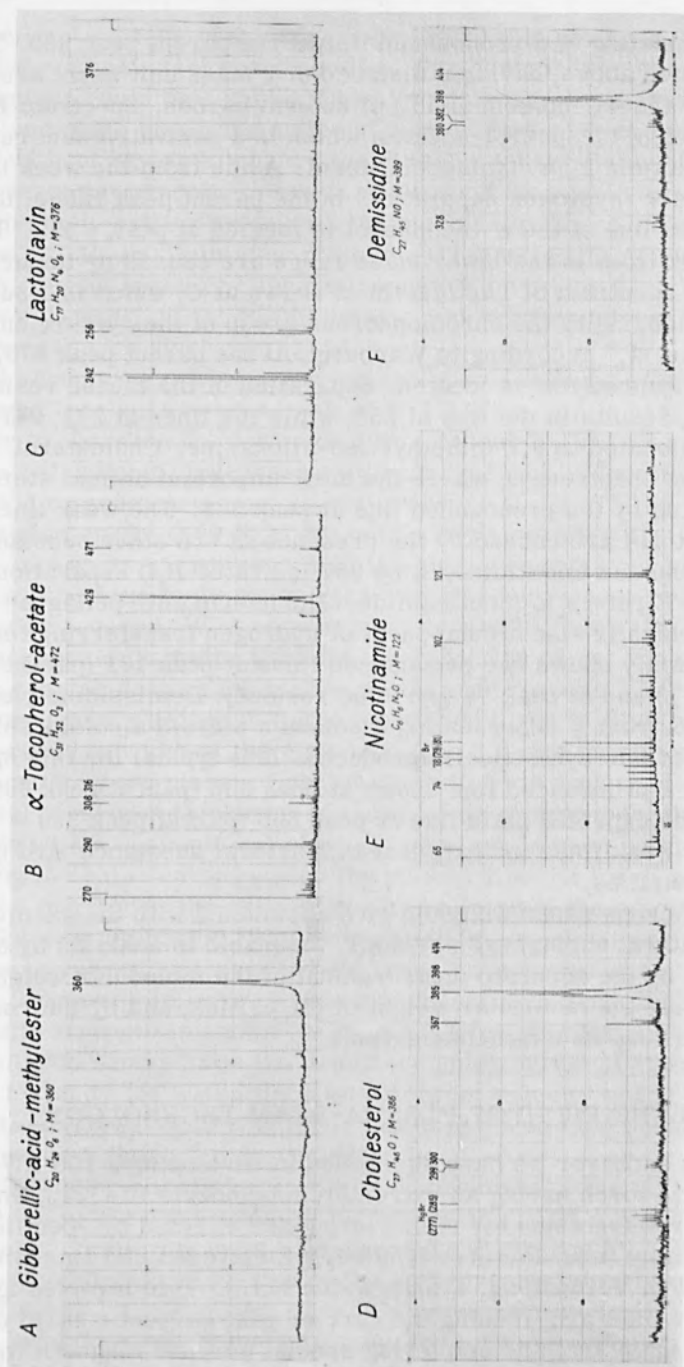


Fig. 19. Attachment molecule mass spectra of six natural substances.

ester, consisting of a pronounced line at the parent peak 360. The lines located above, having a distance of 1 mass unit each, are caused by the ^{13}C -content (1.1%) of natural carbon. Spectrum B is a record of α -tocopherol-acetate, which is a derivative and represents vitamin E in commercial form. Aside from the weak line at peak = 471 (hydrogen separation) in the parent peak range, the pronounced line of the α -tocopherol is located at peak = 429. The other weak lines in the lower mass range are caused by impurities. The spectrum of Lactoflavin is shown at C, which is a substance belonging to the chromophorous group of the "yellow enzyme of yeast," according to Warburg. At the parent peak 376, the most pronounced line is located. Separation of the ribose residue, from C2', results in the line at 256, while the lines at 242, 241 may be allocated to 6,7-dimethyl-iso-alloxazine. Cholesterol yielded the spectrum D, where the most important animal sterol is represented by the pronounced line at peak 385. The weak lines at 398 and at 414 are caused by the presence of two other substances, demissidine and tomatidine. Line 367 indicates H_2O separation. Spectrum E refers to nicotinamide. The human anti-pellagra-vitamin which is also a component of hydrogen transferring ferments, mainly shows two pronounced lines at peak 121 (parent peak 122-1) and at peak 78 (pyridine residue). Demissidine yielded the spectrum F representing a solanum steroid alcoholoid which has been of late synthetically produced. It is typical for this spectrum that a pronounced line shows at peak 396 (parent peak 399-3) besides the expected main line at peak 398 (parent peak 399 - 1). Again the weak lines indicate that this natural substance still has some impurities.

In numerous cases we could give assistance with the examination of natural substances not easily amenable to analysis by other methods, by the accurate measurement of the molecular weight, by detecting the molecular weight of impurities, and by information regarding structural indications.

WORK IN THE FIELD OF PLASMA-BEAM TECHNOLOGY

In this endeavor we have succeeded in developing a 100-kW plasma arc torch having an extremely long nozzle life [31], where the required functions for melt-cutting are effected by specific programming (manufacturer Kjellberg Elektroden und Maschinen B.m.b.H., in Verwaltung, Finsterwalde N.L.). This involves ignition of the pilot arc, feeding the carrier gas, current control of pilot and main arc, and employing special purpose magnetic valves in the unit and a control unit. This type of torch permits cutting

sheet metal packs of thicknesses up to 90 mm of chromium-nickel and structural steel sheeting [31]. Furthermore, an operating technique could be established which afforded plunge-cutting at any desired place of a plate material without hazard to the jet nozzle and without having to change the direction of the flame. This meets the essential preconditions for replacing autogenous torch cutting by the melt-cutting mode with the plasma-arc torch. This is proposed particularly in view of the very recently attained advances with respect to energy and gas consumption by the creation of our novel plasma fine-jet torch, using a fine nozzle and small volume high-pressure discharge [31].

Although the power density at the nozzle opening of conventional plasma torches amounts to about $5 \cdot 10^5$ W/sq cm, it became possible to increase the power density to about $2 \cdot 10^6$ W/sq cm with the small volume high-pressure discharge torch by specific measures, i.e., geometry, carrier gas pressure of about 10 kg/sq cm, gas composition (Ar O,60 H₂ O,40), and a conic nozzle with a diameter of 1 mm at its narrowest. With the higher energy concentration, the width of the melt-cut and the energy requirement could be materially reduced. With comparative separating work, the very small nozzle diameter permits the reduction of gas requirement to a quarter despite the higher discharge pressure. On Fig. 20 the improvement attained with the mentioned torch is illustrated with reference to width of the melt-cut. The 2.8 mm maximum width cut made by the plasma fine-jet torch (left) in a plate of chromium-nickel of 37-mm thickness is shown. For comparison, the melt-cut made with a conventional plasma-arc torch is seen on the right. Using the same cutting speed the plasma fine-jet torch requires only a quarter of the carrier gas and a third of the power.

Interesting possibilities in the field of hot machining reveal themselves with respect to heat transfer at power densities of 10^6 W/sq cm, and over. A machining test involving steel of high tensile strength is shown on Fig. 21. A feasible technique with this mode seems to be the use of the combination of atomizing of the material for which the high emergent velocity of the gas jet is particularly favorable with our last mentioned type of torch with subsequent finish turning in hot state.

Continued efforts for further improvement of the plasma cutting tool led to the conception of the plasma microjet torch with micro-nozzle and microvolume high-pressure discharge [31], [32]. The dense, high-energy plasma of the high-pressure discharge, which by a special arrangement only fills an extremely small volume, has been disposed immediately in front of the micronozzle which at its narrowest has a diameter between $D = 50$ and $800 \mu\text{m}$. The

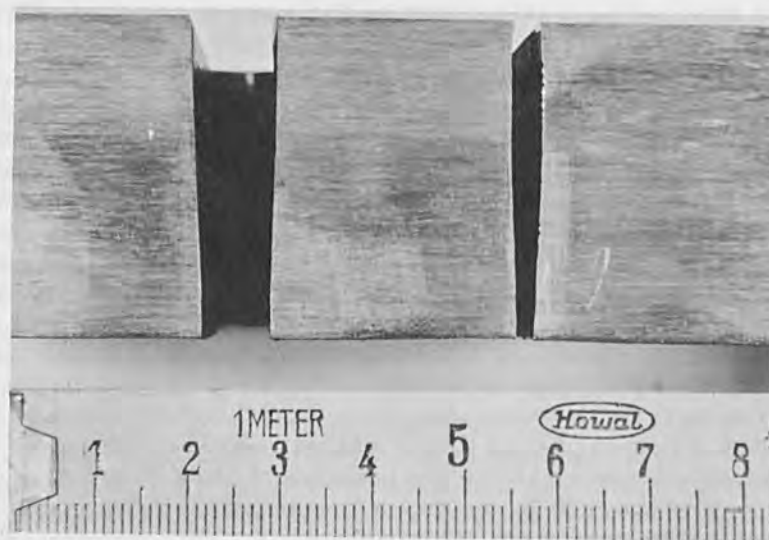


Fig. 20. Photo of a maximal 2.8-mm wide high-pressure plasma jet melting cut (left) in a 37-mm thick chrome-nickel alloy as compared with a maximal 9-mm wide standard plasma jet (right). Both jets operated with the same working speed $v = 0.22 \text{ m min}^{-1}$.

Left: High-pressure plasma jet $P = 13 \text{ kW}$; $I = 80 \text{ A}$; $U = 162 \text{ V}$;
 $Q_{Ar} = 0.72 \text{ m}^3 \text{ h}^{-1} \text{ (STP)}$ $Q_{H_2} = 0.50 \text{ m}^3 \text{ h}^{-1} \text{ (STP)}$

Right: Standard plasma jet $P = 39 \text{ kW}$; $I = 305 \text{ A}$; $U = 128 \text{ V}$;
 $Q_{Ar} = 3.4 \text{ m}^3 \text{ h}^{-1} \text{ (STP)}$ $Q_{H_2} = 2.0 \text{ m}^3 \text{ h}^{-1} \text{ (STP)}$

high pressure existing in the discharge chamber, together with the small size and the geometry of the nozzle, provide for the high-temperature plasma with supersonic speed as a fine and practically parallel jet. The auxiliary plasma of the continuously burning 100-W high-frequency arc ($5 \cdot 10^6 \text{ cps}$) permits the operation of the torch with accurately defined pulses. The non-retouched photograph of the flame of the plasma micro-jet torch is presented on Fig. 22. Some typical cuts and holes obtained with the plasma microjet torch have been photographed on Fig. 23, showing the results with continuous operation (60 to 500 W and $D = 100 \mu\text{m}$) and with pulse operation, respectively. In air and under protective gas, for example, holes down to $60\text{-}\mu\text{m}$ diameter and cuts up to $100 \mu\text{m}$ width could be produced even in very high melting material such as tungsten, tantalum and molybdenum (silicon, ceramics, quartz, and glass). One of the surprising phenomena is the conspicuously narrow zone of thermal stress in the vicinity of a fine cut.

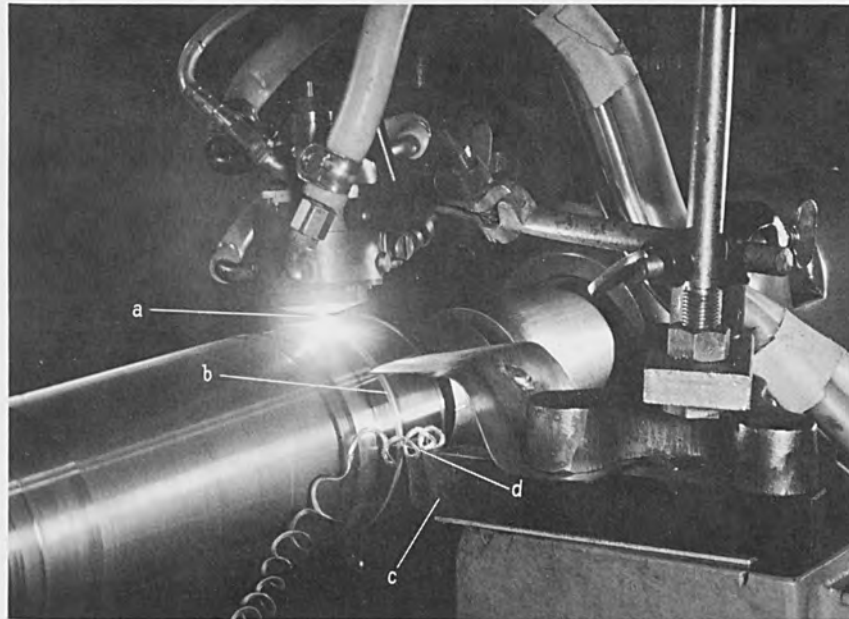


Fig. 21. Application of the plasma jet flame for hot chipping. The small volume highest pressure discharge causes the micro-heating of the chip forming zone (power 12.5 kW). (a) plasma flame, (b) softening material, (c) chisel, (d) red-hot chip.

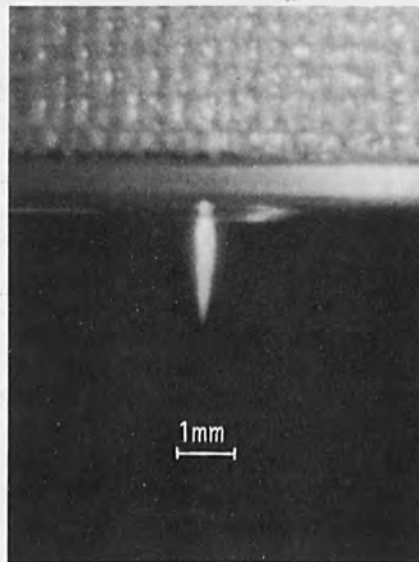


Fig. 22. Enlarged nonretouched photo of the high-temperature micro darting flame produced by the fine beam plasma jet (In the photo the flame core outshines the fine beam as a whole, thus a bigger flame diameter seems to be visible).

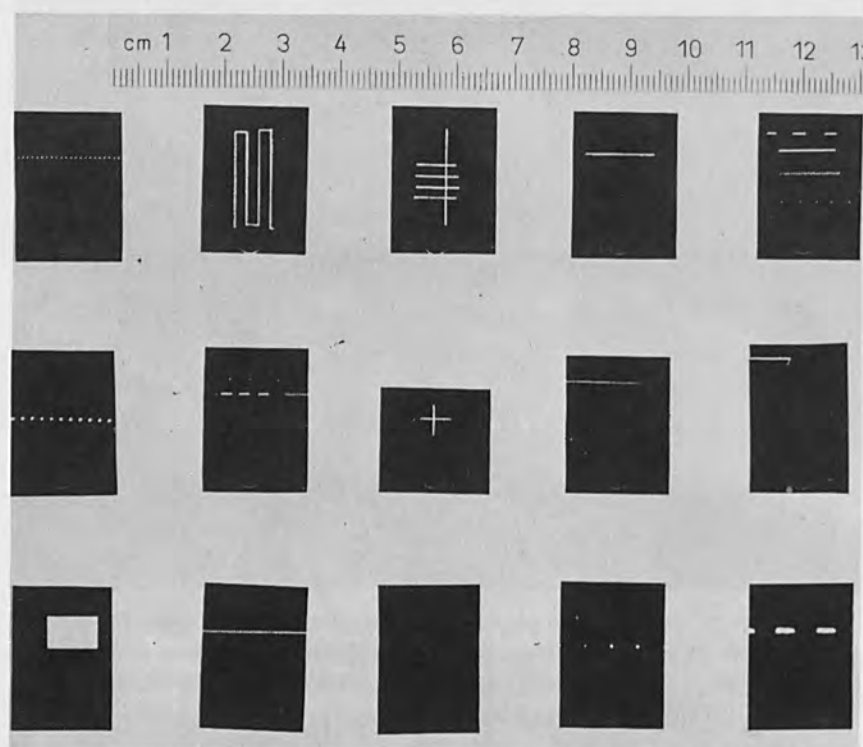


Fig. 23. Melting cuts and melting bores in various materials (tungsten, molybdenum, chrome nickel alloy, aluminum) of different thickness produced by means of the fine beam plasma jet.

REFERENCES

1. Neuer Glühkathoden-Oszillograph nach M. von Ardenne (New thermionic cathode-oscillograph acc. to M. von Ardenne), Mitt. E. Leybolds Nachf., Köln-Bayental, 1929.
2. M. von Ardenne, Ueber neue Fernsehsender und Fernsehempfänger mit Kathodenstrahlröhren. (On new television transmitters and receivers equipped with cathode-ray tubes), Fernsehen 2, 65, 1931.
3. M. von Ardenne, Negative Ionenstrahlen bei der Formierung von Hochvakuum-Elektronenstrahlröhren (Negative ion beams for the formation of high-vacuum electron beam valves), Arch. Elektrotech. 29, 731, 1935.

4. M. von Ardenne, Das Elektronen-Rastermikroskop (The electron scanning microscope) Z. Physik 109, 553, 1938.
5. M. von Ardenne, die verwendung der Elektronensonde für Mikromanipulationen (The use of electron probes for micro manipulations), Naturwiss. 26, 562, 1938.
3. M. von Ardenne, "Zur Leistungsfähigkeit des Elektronen-Schattenmikroskopes und über ein Röntgenstrahlen-Schattenmikroskop." (On the efficiency of electron shadow microscopes and x-ray shadow microscopes), Naturwiss. 27, 485, 1939; and "Elektronen-Uebermikroskopie," Springer, Berlin, 72 ff., 1940.
7. M. von Ardenne, Ueber eine Ionensonde grosser Schärfe (High definition focus ion probe), Z. techn. Phys. 20, 344, 1939 and Elektronen-Uebermikroskopie, Springer, Berlin, 162 ff., 1940.
8. M. von Ardenne, Mikroskalen-Apparatur (Micro-scale device), Nachrichtertechn. 10, 427, 1960.
9. K. H. Steigerwald, "Materialbearbeitung mit Elektronenstrahlen" (Material machining by means of electron beams), Verh. Int.Kongr.Elekt.Mikroskopie, Berlin, 1958, Springer, Berlin, 1960, p. 276; and Der Elektronenstrahl als Werkzeug (The electron beam as a tool) Elektrotech. Z. 81, 925, 1960; and Schweissen und Schneiden mit Elektronenstrahlen (Welding and cutting operations by means of electron beams), Schweissen und Schneiden, 12, 89, 1960.
10. M. von Ardenne, Elektronen- oder Ionenstrahlung als Mikrowerkzeug (Electron- or ion-beams as a micro-tool), Nachrichtentechn. 11, 339, 1961.
11. S. Schiller, S. K. Panzer, and U. Heisig, Der Elektronenstrahl als thermisches Mikrowerkzeug (The electron beam as a thermal micro-tool), Nachrichtentechn. 13, 128, 1963.
12. M. von Ardenne, "Tabellen der Elektronenphysik, Ionenphysik und Uebermikroskopie" (Tables of electron physics, ion physics and electron-microscopy), Dtsch. Verlag Wissenschaften, Berlin 1956, Vol. II, p. 640, IX, and p. 884, column "primary-electron impact in high-vacuum."
13. M. von Ardenne and S. Schiller, Ein 45-kW-Elektronenstrahl-Mehrkammerofen für das Schmelzen und Giessen beliebiger Metalle (45-kW-electron beam multi-chamber furnace for melting and casting various metals), Kernenergie 3, 507, 1960.
14. M. von Ardenne and H. Förster, Das Vakuumgiessen im Elektronenstrahl-Mehrkammerofen (Vacuum-casting processes in electron beam multi-chamber furnaces), Neue Hütte 8, 313, 1963.

15. M. von Ardenne, G. Jäger, and S. Schiller, Ein 200-kW-Elektronenstrahl-Mehrkammerofen (200-kW-electron beam multi-chamber furnace), *Neue Hütte* 8, 2, 1963.
16. H. Westmeyer, S. Schiller, and M. Godenschweg, Eine Beobachtungseinrichtung für Hochvakuumelektronenstrahl-Schmelzöfen (Observation device for high-vacuum electron beam furnaces), *Vakuum-Technik* 12, 17, 1963.
17. M. von Ardenne, Hochvakuumerschmelzen mit dem Dresdner Mehrkammervorgang (High vacuum melting with the Dresden multi-chamber process), *Freiberger Forschungs.-Hefte B* 68, 5, 1961.
18. M. von Ardenne, "Tabellen zur angewandten Physik" (Tables for applied physics), Dtsch. Verlag Wissenschaften, Berlin, 1964, Vol. II, p. 36 (main characteristics).
19. M. von Ardenne, 1948, "Ueber Plasmatron-Ionenquellen und einige ihrer Anwendungen" (On plasmatron-ion sources and some of its applications), Lecture held at Leipzig in 1956; and "Probleme des Plasma in Physik und Astronomie" (Problems concerning the plasma in physics and astronomy), Akademie Verlag, Berlin, 197, 1958.
20. M. von Ardenne, Das Duoplasmatron als Ionen- oder Elektronenspritze extrem hoher Emissionsstromdichte (Duoplasmatron as ion- or electron jet of extremely high emission current density), *Exper. Techn. Physik* 9, 227, 1961.
21. M. von Ardenne, "Tabellen zur angewandten Physik" (Tables for applied physics), loc.cit. 653, 655 ff., 661, 670.
22. M. von Ardenne, Brit.Pat. 795 766, May 10, 1955.
23. M. von Ardenne, G. Jäger, F. Köhlet, and G. Petter, Ueber einen magnetischen Massentrenner mit kleinem Magnetfeldvolumen (On a magnetic mass separator with small size magnet), *Kernenergie* 3, 1177, 1960.
24. M. von Ardenne, Ueber Aufbau und Eigenschaften des Dresdner magnetischen Isotopentrenners (Design and properties of the Dresden magnetic isotope separator), *Kernenergie* 5, 305, 1962.
25. M. von Ardenne, Der Dresdner Molekülspektrograph, ein neuartiger Präzisions-Massenspektrograph für negative hochmolekulare Ionen (Dresden molecular spectrograph—a new precision-type mass spectrograph for negative high-molecular ions), *Kernenergie* 1, 1029, 1958.
26. M. von Ardenne, Fortschritte in der Elektronenanlagerung-Massenspektrographie vielatomiger Moleküle (Developments in electron attachment mass spectrography of polyatomic molecules), *A. Angew. Phys.* 11, 121, 1959.

27. M. von Ardenne, K. Steinfeld, R. Tümmeler, and H. Westmeyer, Spektren anorganischer Substanzen als Basis der Massenskala beim EA-Massenspektrographen für vielatomige Moleküle (Spectra of anorganic substances as a base of the peak scale used in electron attachment mass spectrographs for polyatomic molecules), *Kernenergie* 2, 1094, 1959.
28. M. von Ardenne and K. Steinfeld, Ueber eine Elektronen-anlagerungs-Ionenquelle mit Fernhaltung des Testampfes von der Entladungskathode (On an electron attachment ion-source, excluding test vapors from the discharge cathode), *Kernenergie* 3, 717, 1960.
29. M. von Ardenne, K. Steinfeld, and R. Tümmeler, Erweiterung der EA-Massenspektrographischen Methode durch den Mechanismus der Anlagerung negativer Ionen an Moleküle (Extending the electron attachment mass spectrographic method by means of attaching negative ions to molecules), *Z. Physik. Chem.* 221, 240, 1962.
30. M. von Ardenne, K. Steinfeld, and R. Tümmeler, Molekül-massenspektrographische Untersuchungen von Diffusionspumpenölen (Molecule mass spectrographic investigation of diffusion pump oils), *Vakuum-Technik* 12, 135, 1963.
31. M. von Ardenne, L. Haase, R. Pochert, W. Roggenbuck, H. Wachtel, P. Wiese, Weitere Arbeiten zur Hochzüchtung und zum Einsatz des Plasmabrenners (Further efforts to improve and use the plasma-arc torches), *Technik* 18, 695, 1963.
32. M. von Ardenne, Metallurgie und angewandte Physik (Metallurgy and applied physics), *Technik* 18, 263, 1963.

ELECTRON-BEAM RADIAL HEATERS FOR MELTING OF METALS

Electron-beam heating sources in combination with high vacuum have created new directions in areas of metal production and treatment, this is generally termed electron-beam technology.

Several claims are made for these power sources, in particular the absence of an accelerating electric field in the melting area, the possibility of independent evacuation of the cathode-anode system, and a uniform distribution of energy in the heating zone of a given configuration.

The methods of beam generation, beam control, and gun distribution and design in a system are amenable to radical changes, depending on the particular application. In view of the relatively large number of technological problems, it is only natural that we should wish to develop some general approach to the creation of suitable heaters (electron guns).

At the E.O. Paton Electric Welding Institute of the Ukrainian Academy of Sciences, several types of electron-beam heaters have been developed, based on the electron gun forming a flat beam of electrons.

Use of the flat beam is expedient in many instances, particularly when it is necessary to heat large surfaces and to provide a heating zone of definite form. Undoubtedly, in these cases, the use of cylindrical beams is also possible, but the task is complicated because, in order to provide the same power, either higher accelerating voltage or more complex electron-optical systems are needed. Figure 1 shows the diagram of the electron gun based on a Pierce electron-optical system and forming a flat flux of electrons.

A directly heated tungsten filament serves as the emitter cathode. The use of a linear cathode simplifies considerably the elec-

*The E. O. Paton Electric Welding Institute of the Academy of Sciences of the Ukrainian Soviet Socialist Republic.

tron gun design. The cathode electrode of the Pierce design has the shape of a 135 degree dihedral angle in the vertex with the linear cathode placed in the cut-slot. The accelerating anode, in contradistinction to the first electrode, unlike the Pierce design systems repeats, the shape of the cathode electrode. The given accelerating anode shape is simple to manufacture and does not require elaborate tooling arrangements.

The considered deviation from the Pierce system (cathode of round section and accelerating anode of simple shape) leads to the formation of a wedge-shaped beam that meets in the gap between the cathode and the accelerating anode. Behind the acceler-

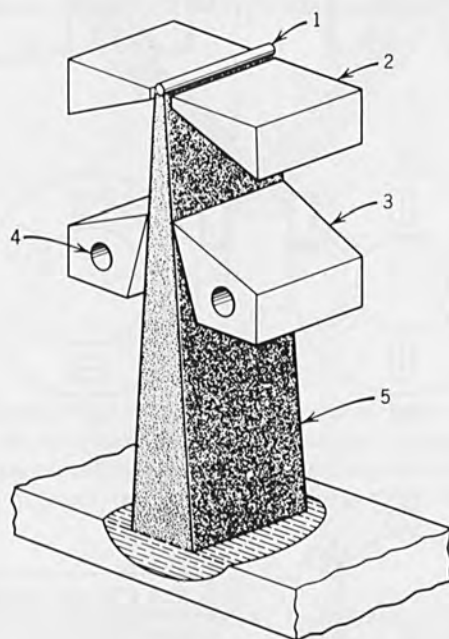


Fig. 1. Diagram of the electron gun forming flat beam of electrons. (1) Linear cathode; (2) cathode electrode; (3) accelerating anode; (4) channels of water cooling; (5) flat beam of electrons.

ating anode the beam becomes slightly divergent. For example, with the cathodes of 1-2-mm diameter and 10-to 15-kv acceleration voltage, the divergence angle does not exceed 8 to 10 degrees.

Where it is necessary to have a smaller divergence angle, the emitting surface of the cathode should be flat, and the accelerating anode should have the shape nearest to the calculated one.

The requirement of 10 to 15 kV as an accelerating voltage allows for the use of standard power sources, simple gun design and does not require special measures in terms of radiation protection.

Changing the space position of several guns (using linear elements), it is possible to change the configuration of the heating area and distribution of electron flux energy in these areas over wide limits.

Figure 2 gives some diagrams of space position of the linear elements. Horizontal position along the radius (a) and the circle (b); vertical position along the circle (c), and horizontal — along the line (d).

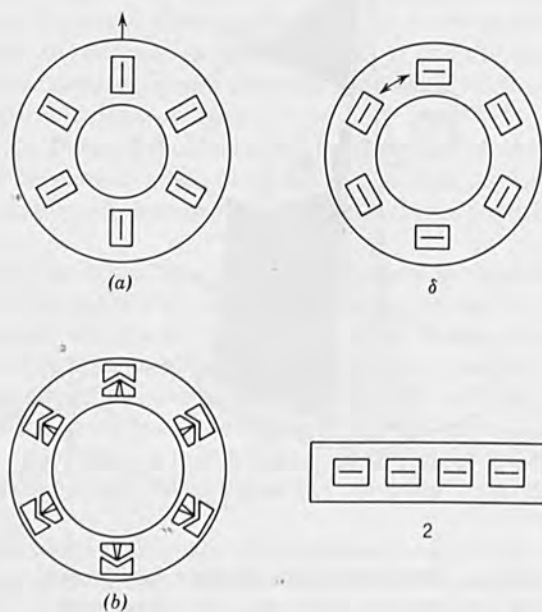


Fig. 2. Some diagrams of space position of individual electron guns (of linear elements).

Practical realization of the above-considered diagrams allows for the creation of a series of electron-beam heaters for specific technological tasks. The heaters are relatively simple and reliable in operation.

Figure 3 shows a diagram of the melting technique by means of the radial-heater system with horizontally placed linear elements (Fig. 2a).

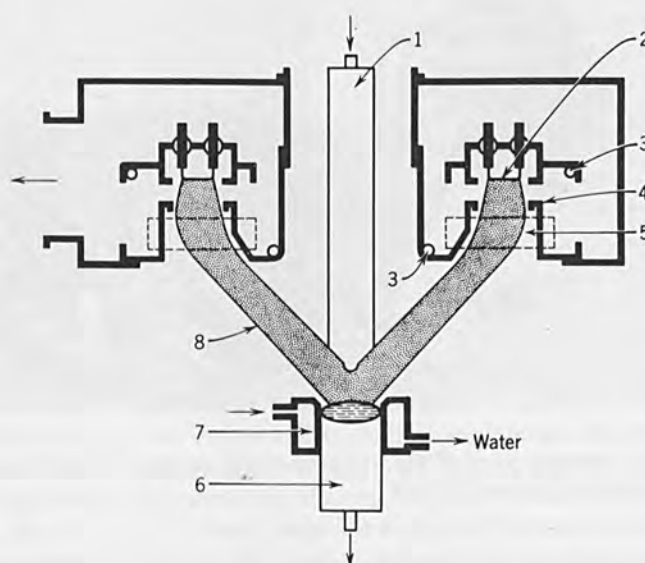


Fig. 3. Diagram of melting with the electron-beam radial heater. (1) Metal to be remelted; (2) cathode head; (3) channels of water cooling; (4) accelerating anode; (5) deflecting electromagnet; (6) ingot; (7) crystallizer; (8) electron beam.

The flat electron beam formed by individual elements of the heater with the deflecting electromagnets are directed into the water-cooled mold. Space position of the heater and the mold are so arranged that one part of the electron beam melts the metal bar, while another maintains the pool in the mold. Application of the deflecting magnets enables us to obtain the most desirable temperature distribution in the mold and at the same time prevents direct contact of metal vapors and spray on the cathode.

The melting area is connected to the heater chamber by means of narrow slit channels and enables us to maintain a separate pumping in each of these areas.

A 150 kW electron-beam radial heater is shown in Fig. 4. The working voltage is 10 to 15 kV.



Fig. 4. General view of the electron-beam radial heater for melting of metals, power 150 kW.

At present, the radial heaters of the above-mentioned type with 30, 80, 150, and 350 kW, and the corresponding plants for melting and refining of metals, have been built.

The plant feeding blocks consist of the standard units. All the plants are differentially pumped.

Figure 5 is a diagram of the electron-beam evaporator with vertical location of the two linear elements. By means of deflecting electromagnets, two flat electron beams are directed into the metal crucible with the metal to be evaporated. It is necessary to note that, by simultaneously changing the shape of the electromagnetic nozzles, it is possible to obtain convergence of the electron beam together with their deflection.

Figure 6 shows the final construction of an electron-beam evaporator with 20 to 30 kW power and working voltage of 10 to 15 kV. When necessary, the evaporator may have two independent systems

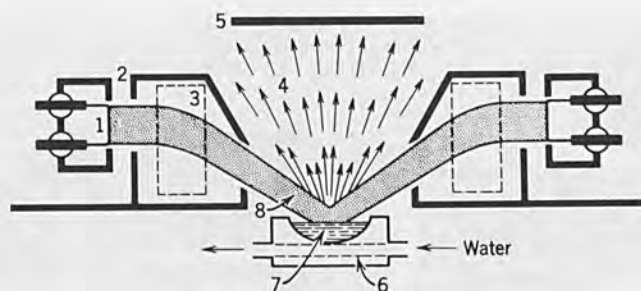


Fig. 5. Diagram of the electron-beam evaporator. (1) cathode head; (2) accelerating electromagnetic anode; (3) deflecting electromagnetic magnet; (4) vapor flow; (5) surface to be evaporated on; (6) water-cooled crucible; (7) material to be evaporated; (8) electron beam.

for pumping out, i.e., a system for the heating elements and a system for the working area of the evaporator.

On the basis of the vertical location of the linear elements, three- and four-beam evaporations have been created. Simultaneous or alternate evaporation of one, two, and four materials, depending on the specific task, may be obtained in such evaporators.

Horizontal location of the linear elements in accordance with the diagram of Fig. 2b enables us to obtain circular heating areas which are necessary, for example, for growing of monocrystals and for the production of hollow ingots (of pipe billets).

Figure 7 shows the arrangement for an electron-beam radial heater for growing monocrystals without crucible, in accordance with Chokhralsky's method.

The heater consists of six to eight linear elements (guns). Each element has the deflecting plate with the edge in the form of arc and screen. The deflecting plate is placed under the potential of the focusing electrode. The flat electron beam coming out of the anode slit is deflected, and simultaneously alters the cross-sectional shape. Electrostatic deflection of the beam and the presence of the screen exclude emitter vapors getting into fusion.

Figure 8 shows the arrangement for the production of hollow ingots, with the help of the radial electron-beam heater mentioned

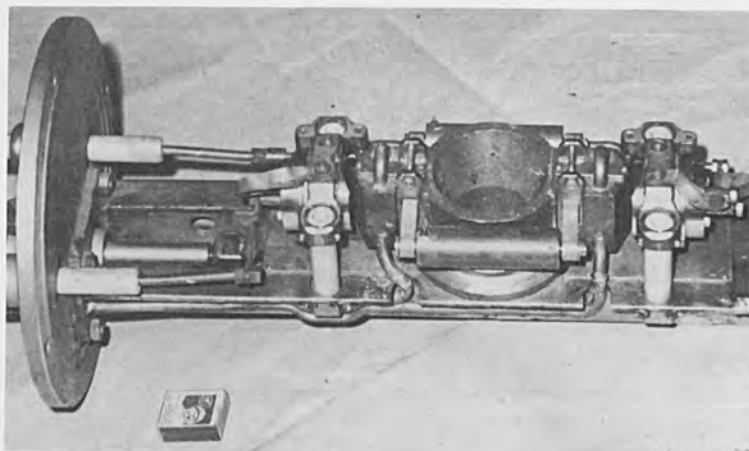


Fig. 6. General view of electron-beam evaporator, 25-30 kW power.

above. A circular heating area is created in the special water-cooled crucible. The crucible consists of the cylindrical water-cooled coupling rod and of the circular mould coaxial with this rod. In the circular gap between them, the hollow ingot forms in proportion to drawing out. The bars located around the coupling rod, granulated powder, or briquets fed from a special hopper unit may serve as initial charge.

Naturally the mentioned examples do not exhaust all the possibilities of the proposed approach to creation of universal electron-beam heaters. They could be added in the form of additional schemes for melting, casting and heat treating.

We considered it worthwhile to discuss at this stage only radial heaters which had been found practical for the solution of specific technological terms.

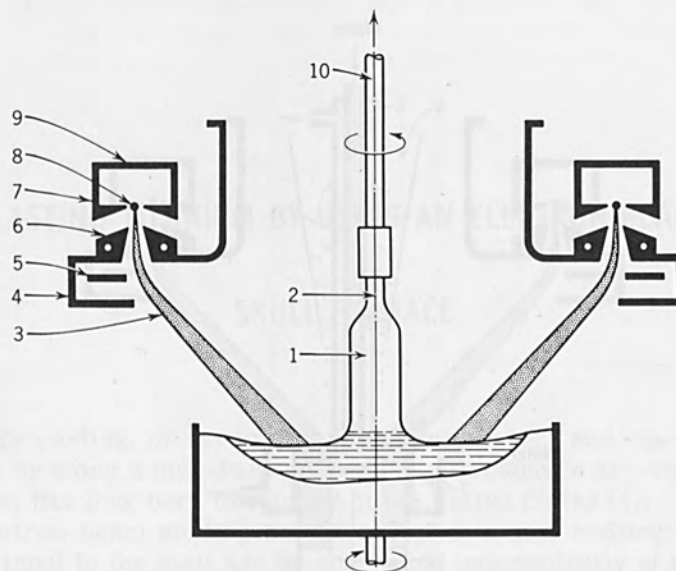


Fig. 7. Schematic of a crystal puller using electron-beam heating. (1) Monocrystal; (2) seed; (3) electron beam; (4) screen; (5) deflecting electrode; (6) accelerating anode; (7) electrode attached to the cathode; (8) linear cathode; (9) cathode head; (10) coupling rod.

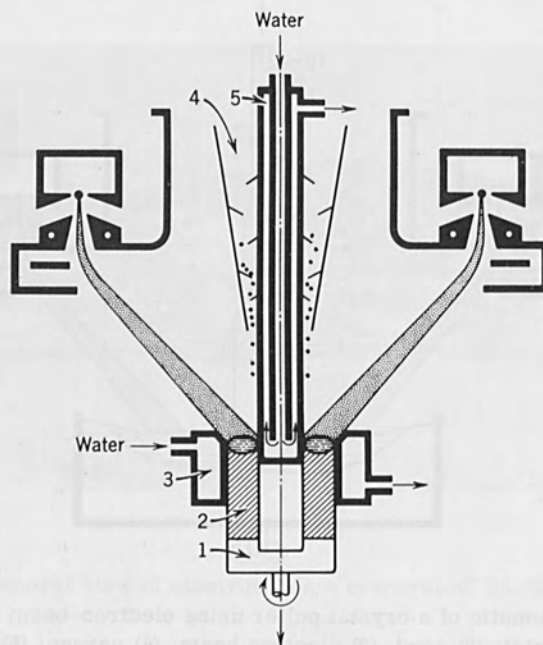


Fig. 8. Production of hollow ingots. (1) ingot base; (2) hollow ingot; (3) crystallizer; (4) hopper; (5) water-cooled coupling rod.

CASTING TITANIUM BY USING AN ELECTRON-BEAM SKULL FURNACE

Shape casting, or scrap recovery of refractory and reactive metals by using a high-vacuum or inert atmosphere arc-skull furnace, has long been developed in the United States [1].

Electron-beam melting has advantage over arc melting in that power input to the melt can be controlled independently of the rate of melting. This enables more super heat of the melt after prolonged purification and alloying period, if necessary. These are the very desirable features to be incorporated with a skull furnace. In this report, results obtained through more than 200 skull castings of titanium and zirconium in a specially designed 120-kw electron-beam skull furnace are shown.

FURNACE

A schematic drawing of the 120-kw electron-beam skull furnace is shown in Fig. 1. A vibration feeder for charging small pieces of scraps is also equipped with the furnace, though it is not shown in Fig. 1. The temperature rise of crucible cooling water showed that approximately 85 kW were transferred to the water in a stationary condition at the gun output of 120 kW. Approximately 28 kg of titanium charge were used as the target. The estimated heat radiation from the melt toward the furnace wall was 20-kW. A 22-in. 13,000 L/sec oil-diffusion pump maintains a vacuum of 10^{-5} torr range in the furnace throughout melting.

Estimated temperature of the pool measured by an optical pyrometer immediately after cutting off the beam are 1990° C and

*Japan Vacuum Engineering Co., Ltd. Yokohama, Japan.

2145°C at 90 kw and 120 kw output of the guns, respectively. A corresponding measurement with an 8000-amp consumable electrode vacuum-arc-skull furnace using the same crucible recorded 1900°C. Table I shows a series of pouring data obtained by initially charging 28.5 kg of total weight of titanium in the crucible. In each run, beam power was started at a low level and was grad-

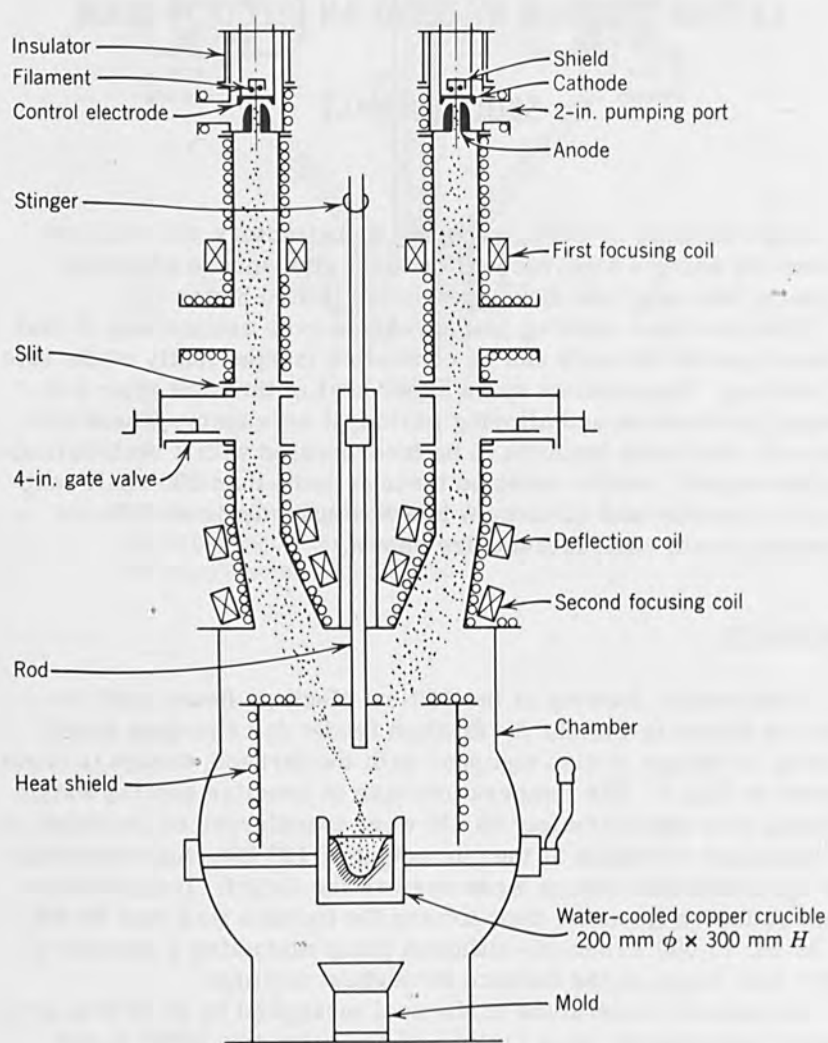


Fig. 1. Schematic drawing of the 120-kw electron-beam skull furnace.

Table I
Electron-Beam Skull-Casting Record (Titanium)

No.	Date	<u>A</u>		<u>B</u>		<u>C</u>		Total Power Consumed Kwhr	$\frac{C}{A+B}$ %
		Weight of Skull kg	Distance between Pool Surface and Lip mm	Weight of Newly charged Titanium kg	Poured Quantity kg	Loss kg	Weight of Skull After Pouring Cycle kg	Total Time for the Cycle min	
167	6-18-'63	23.7	150	4.8	4.2	0.4	23.9	30	14.8
168	6-19	23.9	140	4.6	4.3	0.4	23.8	26	15.1
169	6-19	23.8	140	4.7	4.3	0.3	23.9	27.5	15.1
170	6-20	23.9	140	4.6	4.2	0.6	23.7	29	14.8
171	6-20	23.7	140	4.8	4.0	0.4	24.1	23	14.0
172	6-21	24.1	135	4.4	3.7	0.4	24.4	23	13.0
173	6-24	24.4	140	4.1	4.2	0.4	23.9	43	14.8
174	6-24	23.9	140	4.6	4.2	0.6	23.7	27	14.8
Mean value		Initial skull	New + charge	= 28.5 kg		4.13			14.55

ually increased to 120 kW, where it was held for three minutes just before tilting the crucible. Weight of poured metal with 120 kW beam output, however, ranged from 3.6 to 8.9 kg for titanium, depending on shape and size of charge, process of material charging, and also on rate of feeding the power. Table II shows another series of pouring data for zirconium. Integrated power input to the melt has practically nothing to do with the poured quantity as shown in Fig. 2. For comparison, a tungsten electrode, inert at-

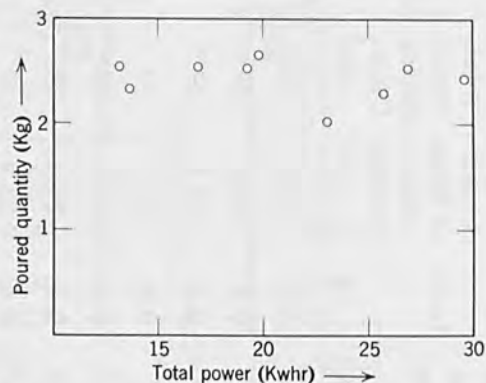


Fig. 2. Weight of poured titanium for integrated power input. Electron gun output — 90 kw constant.

mosphere arc-skull furnace in the author's laboratory pours 5.5 kg of zirconium at 3000 amp, 30 V with radiation reflector assembly over the crucible. Fluidity of the melt was checked by using a machined graphite mold. Table III shows the result with a similar data obtained by consumable electrode skull casting of titanium.

PROPERTIES OF CAST TITANIUM

Chemical analysis of the cast product is shown in Table IV. Starting materials for the electron-beam casting include low-grade sponge, untreated scraps from consumable electrode arc-skull cast product, and the scraps acid treated and washed with water. Mechanical properties of the electron-beam cast titanium are quite similar with that of the consumable electrode arc cast titanium as shown in Fig. 3 and Table V. Some data taken from arc-cast

Table II

Electron-Beam Skull-Casting Record (Zirconium)

No.	Gun Output kW	A		B		C		Total Time for the Cycle min	$\frac{C}{A + B}$ %
		Weight of Skull kg	Weight of Newly Charged Zirconium kg	Poured Quantity kg	Loss kg	Weight of Skull after Pouring kg			
3	75	23.4	8.7	3.7	0.2	28.2	23.0	11.5	
4	95	28.2	5.1	5.0	0.1	28.2	24.5	15.0	
5	110	28.2	5.6	5.4	0.2	28.2	27.0	16.0	
6	120	28.2	4.6	5.1	0.2	27.5	27.5	15.5	

Table III

Fluidity Test

Process	Material	Hydrostatic Pressure			Length of Cast Material cm	Hardness V.H.N. 20 kg	(B) / (A)
		Head Cm-Titanium (or Zirconium)					
Electron-beam skull	Unalloyed-titanium	Sieved raw scrap	Acid-treated scrap *	11.9	32.9	255	2.8
				13.0	79.0	179	6.1
		Sponge	JIS-2	12.5	74.4	182	6.0
			JIS. 1-2	12.0	73.2	189	6.1
				Sponge	8.0	59.2	159
Arc skull	Unalloyed-titanium	1st melted ingot, Made from sponge	JIS 1-2	8.2	36.8	156	4.5
				14.0	76.2	179	5.4
		Mold:		Machined Graphite 0.5" ϕ , 41.5" L		Hair-Pin Shaped Grooved, without Preheating and Horizontally placed.	
Output:		E.B.Skull, 120 kw. Arc Skull, 33 V.					

*HF 3%, HNO₃ 41% at 20°C, 10 min.

Table IV

Chemical Analysis

Impurity Component <i>w/o</i>	E. B. Skull Casting			Consutrode Skull	Arc Casting	
	Sponge		Acid-Treated Scrap Sieved Raw Scrap	Sponge		
	Starting Material	After Casting		Typical Start- ing Constrode	After Casting	
C	0.05	0.05	0.0245 0.0340	0.11	0.015	0.011
O		0.047	0.016 ?	0.04	0.094	0.106
H	0.05	0.0028	0.002	0.003	0.0032	0.0034
N	0.03	0.004	0.016	0.012	0.010	0.007
Fe	0.35	0.15	0.09 0.11	0.11	0.136	0.130
Hardness V.H.N. 20kg	185	182	171	221	174	160

Table V

Impact Test Data

Casting Method	Material			Charpy Impact Value (20°C) kg-m
Electron-beam skull casting	Unalloyed Ti	Sponge	JIS 1-2 *	11.68
			JIS 2 *	11.56
		Acid-treated scrap		9.2
		Sieved raw scrap		3.7
	Zirconium *			5.46
Arc-skull casting	Consutrode Unalloyed-Ti Made of JIS. 1-2			11.56

* Chemical Analysis (w/o)

Elements max	JIS 1-2	JIS 2	Zirconium
Fe	0.20	0.35	0.2
Cl	0.12	0.15	0.06
Mn	0.02	0.05	0.04
Mg	0.07	0.10	0.06
Si	0.03	0.04	—
N	0.02	0.03	0.01
C	0.03	0.05	0.06
H	0.005	0.05	—
Ti	Bal	Bal	—
Zr	—	—	Bal

Table VI
Corrosion Test Data

Condition			Corrosion Rate mm/year					
			Unalloyed-Ti					
			Arc Skull JIS. 1-2		Electron-Beam Skull		Stain- less Type 304	
Acid	Temp. °C	Time hr	Machined Graphite Mold	Rammed Carbon Mold	Sieved Raw Scrap	Acid Treated Scrap	Sponge JIS. 1-2	
			Wrought Products JIS. 1-2		Machined Graphite Mold	Rammed Carbon Mold		
H ₂ SO ₄ 35%	20~30	208	3.206	2.994	2.892	2.458		47.123
		96	2.227	2.425	1.294	1.368	2.044	57.64
HCl 30%	20~30	242	4.281	5.361	5.038	4.792		67.299
H ₃ PO ₄ 35%	20~30	192	0.0305	0.031	0.0298	0.0351		0.0170

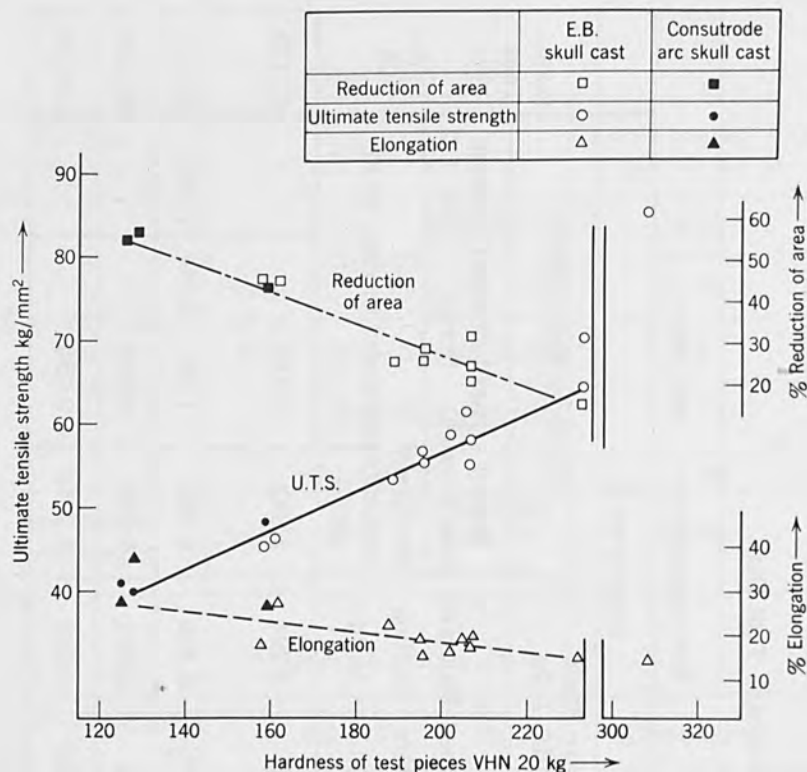


Fig. 3. Tensile test results versus hardness of test pieces.

samples obtained from best quality sponges commercially available in Japan (JIS 1-1) are also shown in Fig. 3. Figure 4 shows hardness near the surface of cast shape, which shows thin but definitely contaminated layer. Table VI shows corrosion test results. Cast materials made of raw scraps were corrosion resistant to the same degree as that of virgin sponge.

OPERATIONAL ADVANTAGES OF THE ELECTRON-BEAM PROCESS

Permanent Service Life of the Skull

The same skull has been used throughout the experiment. This

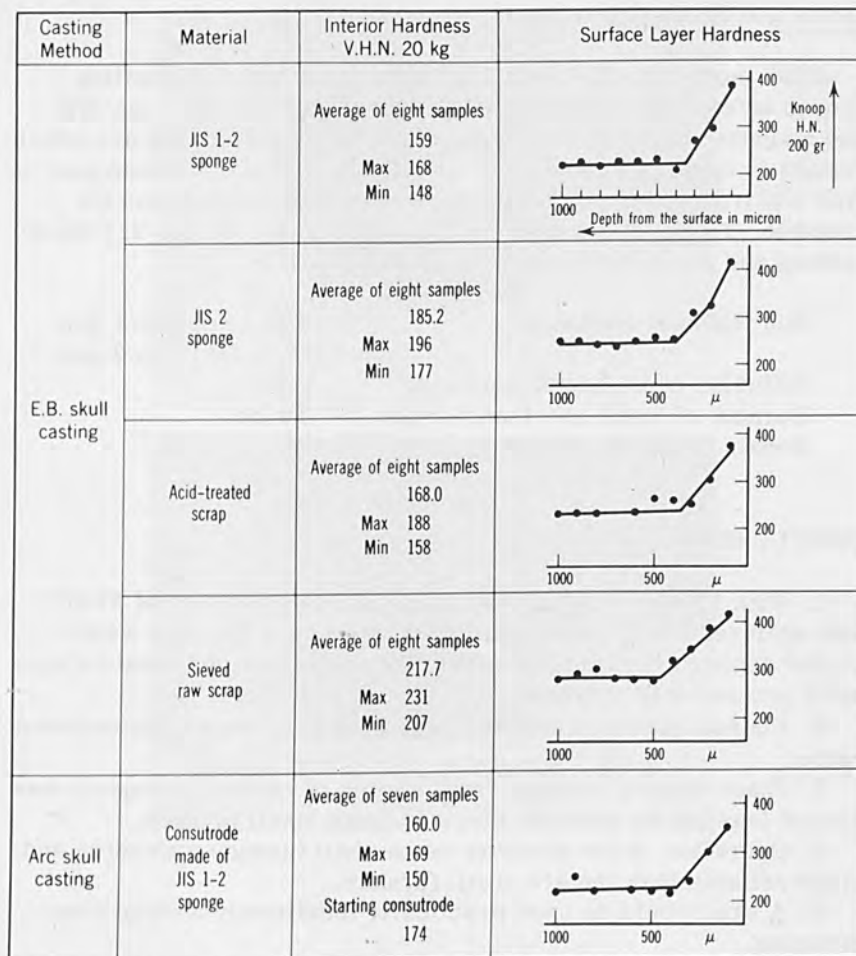


Fig. 4. Hardness of cast titanium.

verified possible construction of a semicontinuous electron-beam skull furnace. With the 8000-amp consumable electrode skull furnace in the author's laboratory, solid residue at the lip limited the semicontinuous operation. Permanent electrode-arc-skull furnace may permit both repair of the lip and charge of random geometry, but it requires refill of argon in the furnace and its operation is not so easy as an electron-beam furnace. Consumption of, and contamination by, electrodes may be another difficulty.

Safety and Stable Operation

After more than 200 pourings, electron-beam skull casting proved safer than that of arc. In the arc-skull furnace, electric and magnetic fields around the crucible often deflect the arc which results in breakage of the crucible. In the self-accelerated gun-type electron-beam skull furnace, there is no field around the crucible. Troubles experienced with the furnace during 223 skull casting and some tantalum ingot production are:

Gun filament exchange	Twice for No. 1 gun Once for No. 2 gun
Diffusion pump heater exchange	Once
Burnout of small autotransformer	Twice
Repair of feeding mechanism	Once

CONCLUSION

1. With 120-kW electron-beam, a pool temperature of 2145°C was achieved for titanium skull casting using a 200-mm water-cooled copper crucible. More fluidity of the melt than that by arc-skull process was obtained.
2. Poured quantity could be controlled to a reasonably constant value.
3. Shape casting starting from scraps of random geometry was proved feasible by using an electron-beam skull furnace.
4. Operation of the electron-beam skull furnace was safer and more reliable than the arc-skull furnace.
5. A skull could be used practically indefinitely without conditioning.
6. Properties of electron-beam cast products are quite similar to those of consutrode skull-cast materials.

The largest and probably the only defect of the electron-beam skull process will be the high initial cost of the furnace. Elimination of the need for electrode preparation, versatility of the process, and safety from the viewpoint of crucible damage are among the interesting features of the electron-beam process. Figure 5 shows an example of an electron-beam skull-cast product.

The authors wish to express their gratitude to Dr. R. F. Bunshah for his valuable comments on the manuscript. This research was conducted under the auspices of the Ministry of International Trade and Industry of Japanese Government.

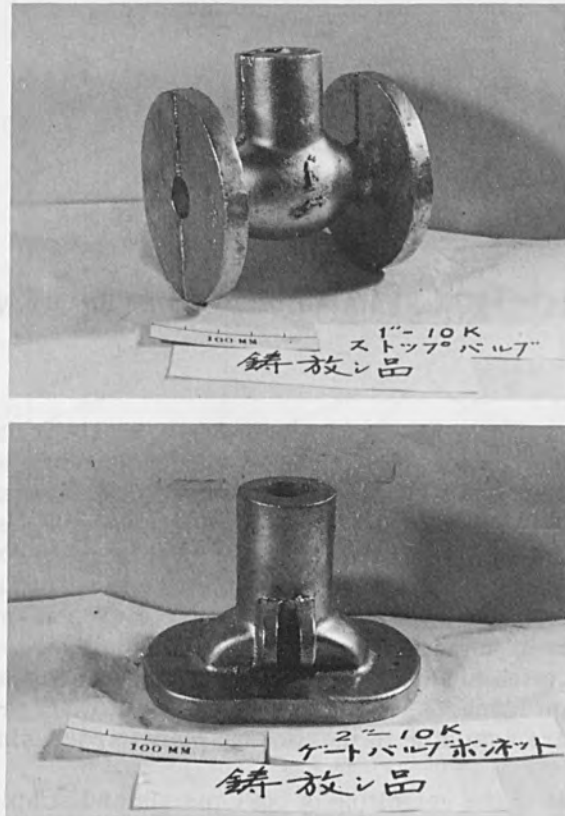


Fig. 5.

REFERENCES

1. S. L. Ausmus, F. W. Wood, and R. A. Beal, U.S. Bureau of Mines Reports of Investigations No. 5686.
2. R. W. Huber, E. J. Chapin, and I. R. Lane, Foundry, 87, 6, 88-93, 1959.
3. R. G. Hardy, Metal Industry, p. 202-206, Sept. 15, 1961.
4. A. E. LeMarche, Westinghouse Electrical Corp., Materials Manufacturing Dept., Report No. 2-CPFF-WG-78256-1-1-(3) 1959.
5. N. Beecher and J. L. Ham, 1961 Trans. 8th Vacuum Symposium, p. 737-743.

ON THE PROBLEM OF REMELTING STEEL IN AN ELECTRON-BEAM MULTICHAMBERED FURNACE

The steel industry has so far been able to meet virtually all consumer requirements which are pertinent to the properties of steel as a working material, but there have been a few exceptions. These requirements have arisen as a consequence of the material needs of atomic power plants and nuclear reactors, and the material needs of supersonic aircraft rocketry and space travel in general. These have led to the introduction of vacuum-metallurgical methods in the steel-producing industry on a large scale. Vacuum-induction furnaces and, in particular, vacuum-arc and electron-beam furnaces, have developed into efficient production units which are available to the modern plants processing stainless steels.

In contrast to the remelting of pure metals and simple alloy systems, high-vacuum processing in the remelting of steels, i.e., complicated multimaterial systems, involve desirable and undesirable reactions (Fig. 1). Figure 1 shows the change in the free energy of the most important metal oxides and CO as a function of temperature, and the stability of the metal as consequence thereof. The value of the free energy for the formation of CO increases with increase in temperature and decrease in pressure. A. M. Samarin [1] demonstrated the vaporization of the suboxides of Al in the system Al - Al_2O_3 and of Si in the system Si - SiO_2 considering the kinetics of the reaction. As a consequence of these reactions, we have the needed prerequisites for the production of well-deoxidized steels with a very low slag content.

The tendency of the principal steel alloy elements towards vaporization during remelting under high vacuum can be seen in a

*Freital, East Germany.

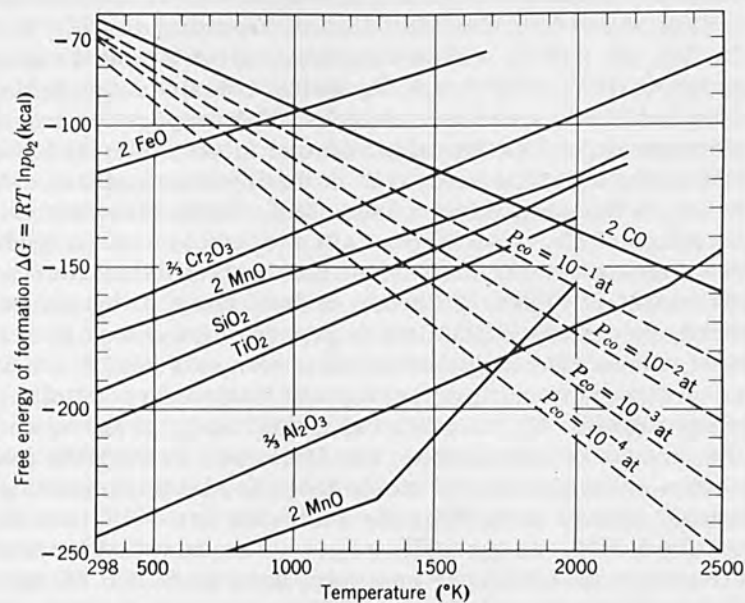


Fig. 1. Temperature dependence of free formation energy of various metal oxides and of carbon monoxide (3). (1) Free energy of formation. (2) Temperature.

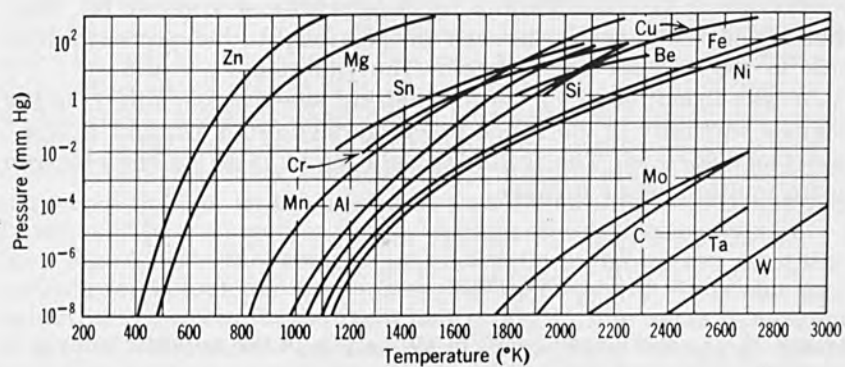


Fig. 2. Temperature dependence of vapor pressures of several metals (4). (1) Pressure, (2) temperature.

first approximation at least qualitatively in Fig. 2. If we look at the melting temperature of 1650°C , corresponding to 1923°K , then Sn, Zn, Mg, Sb, and Cu will be vaporized before Fe, at the working pressures of $10^{-3} - 10^{-4}$ mm Hg which are customary in the electron-beam multichamber furnaces. The evaporation losses of the elements depend on the initial content in the material to be remelted, on their partial pressures in the liquid and gaseous phases, and hence on the composition of the steel. These elements, with the exception of Cu, occur as so-called trace elements in highly alloyed stainless steels, corrosion- and heat-resistant steels, and in heat-resistant alloys. If certain critical composition limits are exceeded, they cause difficulties in processing and loss in certain physical values. The undesirable trace elements can be removed either by single or multiple melting and lead to very definite quality improvements. On the other hand, the loss by evaporation of Mn, Fe, and Cr is undesirable. The Mn losses in the EMO (electron-beam multichamber furnace), depending on the remelting conditions, amount to 40-60%, the Fe losses are 4-6%, and the Cr losses are 4-10%. As a result, we get a concentration increase in the elements with low-vapor pressure, such as Ni, Co, W, and Mo in the remelted material. We can make provisions for this through the chemical composition of the initial melt; the remelting of Mn-alloy steels in high vacuum is not recommended.

The reduction of the H_2 and N_2 content through remelting is important in improving the steel quality. We can get maximum H_2 content down to 0.5 ml in 100 G. The N_2 content in remelted steels amounts to 30-40 ppm, depending on the content of the nitride-forming elements Cr and Al.

According to investigations by Bunghardt and Vollmer [2], the melting performance is directly proportional to the current intensity in the vacuum-arc furnace. The equation $V_A = 0.6 \cdot I - b$ was found; here V_A is the melting speed in kG/min, I is the current intensity in the range investigated up to 6 kA, and b is a constant 0.05-0.55. The following relation is valid for the electron-beam multichamber furnace:

$$V_A = \frac{N_E}{e \cdot 60} \text{ kG/min}$$

where N_E is the power level in kW and e is the specific energy expenditure in kWh/kg. Accordingly, the melting speed increases with power increases. It drops when the power level is constant, while the specific energy expenditure increases. The specific

energy expenditure in kWh/kg becomes an important index for the melting process in the EMO because these furnaces are, as a rule, operated at maximum power levels.

Looking at this from the industrial and technical point of view, the material supply and feed rate thus determine the melting speed. Slow material feed means high specific energy expenditure, greater overheating of material to be melted, longer periods of time during which the melt is under a vacuum, and, in turn, intensive degassing. In other words, the melting process can be regulated within relatively wide limits, depending on the effects we want to achieve. The operation of our 200-kW EMO has revealed that the remelting of steel calls for a specific energy expenditure of 1.2 to 3.0 kWh/kg in order to create optimum conditions for the various brands of steel. The illustration once again shows an overall view of the furnace. The fully automatic controls for the feed of the material to be remelted are applied, alternately, from two sides, perpendicularly to the electron beam. As charge material we use cast billets with a diameter of 140 mm; their surface is steel-shot blasted or pickled. The dripping, molted (fused) steel is collected in a water-cooled copper mold and formed into a block with an average diameter of 230 mm and a length of 310 to 340 mm, with a weight of 110 to 120 kg.

Figure 3 shows a longitudinally separated block of 15 Cr₃ after etching with copper ammonium chloride. We can see the vertically aligned coarse grains with size and homogeneity determined by the specific energy inputs discussed in greater detail earlier. The ingot in Fig. 4 was melted with 2.5 kWh/kg. At 1.5 kWh/kg, the grains are finer and grow at an angle toward the mold wall, which is a consequence of the lesser degree of overheating of the melting material, and hence of the increased influence of the cooling by the pot wall and the formation of the melt sump. In this connection it is interesting to note the quality of the block surface which improves as the specific energy expenditure increases, and which decisively influences the success of hot-forming. On the other hand, we can achieve faultless ingot surfaces in pot melting also when the melting speed is high, on the provision that the melting bath is uniformly exposed to the electron beam. A magnetic deflection system (see Fig. 4, left side) enables us to distribute the energy over the melting feed stock and the molten pool by means of a circular pendulumlike motion. In the right half of Fig. 4, the focus movement of the 1200 kW radiator is sketched; this radiator belongs to one of the electron-beam multi-chamber furnaces in which we can produce forged blocks with diameter up to 1000 mm, and rolled shapes with a cross section of 380 × 420 mm.

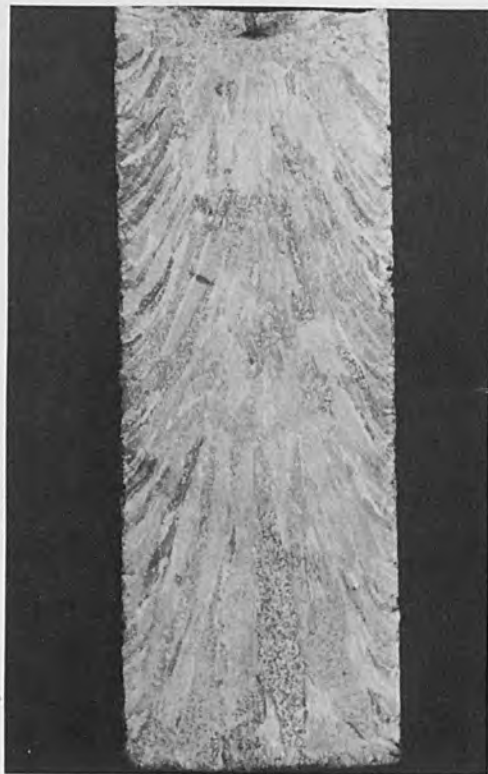


Figure 5.

properties in the cross-rolling detection reach 90 to 100% of the longitudinal values. Conversion points Ac_1 and Ac_3 as a rule are unchanged. Using hardness and fracture series, it was shown that the steel from the EMO is somewhat more sensitive to overheating than steel produced in the open.

Figure 6 shows a few steel types and their chemical composition before and after remelting in the EMO; they represent a part of our research effort on electron-beam-melted steels; we shall discuss the changes in their properties below (Fig. 6).

Consumer requirements for roller-bearing steel with extremely low slag content for the improvement of the fatigue resistance can be met with 100 Cr 6 (No. 3). This steel is used successfully for precision bearings, e.g., high-rpm grinding mandrels which run at 40,000 to 60,000 rpm; the running time here can be as much

No.	Stahlmarke	C	Si	Mn	P	S	Cu	Ni	Cr	W	Mo	V
1	210 Cr 46	A	2.05	0.31	0.48	0.025	0.007	—	11.39	—	—	—
		U	2.05	0.40	0.10	0.026	0.006	—	9.98	—	—	—
2.	142 WCrV13	A	1.38	0.27	0.34	0.014	0.006	—	0.36	2.85	—	0.27
		U	1.41	0.28	0.10	0.016	0.008	—	0.30	3.48	—	0.25
3.	100 Cr 6	A	1.00	0.27	0.27	0.020	0.006	—	1.52	—	—	—
		U	1.02	0.28	0.11	0.017	0.007	—	1.43	—	—	—
4.	40 NiCrMo15	A	0.44	0.25	0.42	0.022	0.011	3.64	1.29	—	0.24	—
		U	0.42	0.21	0.02	0.024	0.010	4.06	1.10	—	0.24	—
5.	38 CrMoV21.14	A	0.36	0.93	0.46	0.029	0.008	—	5.80	—	1.40	0.30
		U	0.36	0.82	0.10	0.026	0.006	—	5.50	—	1.66	0.32
6.	22 CrMo4	A	0.24	0.38	0.72	0.017	0.010	—	1.08	—	0.31	—
		U	0.24	0.38	0.15	0.017	0.008	—	0.94	—	0.34	—
7.	X 82WMo6.5	A	0.83	0.13	0.28	0.024	0.007	—	4.26	6.09	4.89	1.86
		U	0.79	0.14	0.17	0.016	0.005	—	4.14	6.34	4.98	1.80
8.	X 20Cr13	A	0.20	0.57	0.73	0.019	0.011	—	13.78	—	—	—
		U	0.21	0.48	0.24	0.020	0.010	—	13.08	—	—	—

Figure 6. Chemical composition of investigated steels from basic arc furnace and after remelting in 200 kW EMO. (1) number; (2) steel brand.

as 1200 to 1500 hours. Outstanding life qualities were also obtained in bearings in small gas turbines. Rollover tests on testing stands with 100 Cr 6 from the EMO revealed a life increase of 230% compared to the basic arc furnace steel. Another example for the use of vacuum-metled 100 Cr 6 would be the production of embossing dies for watch glasses, where even minimum surface flaws as a result of slag or liquation cause the embossing stamp and matrixes to be rejected. Such quality flaws could be completely eliminated by means of vacuum-cast steel.

In this connection we might take a look at Fig. 7, which shows the reduction in the oxygen content for steels 2, 3, and 7. We get final oxygen values of 20 ppm, with the exception of high-speed steel DMo 5, which, however, reveals a relatively high initial content.

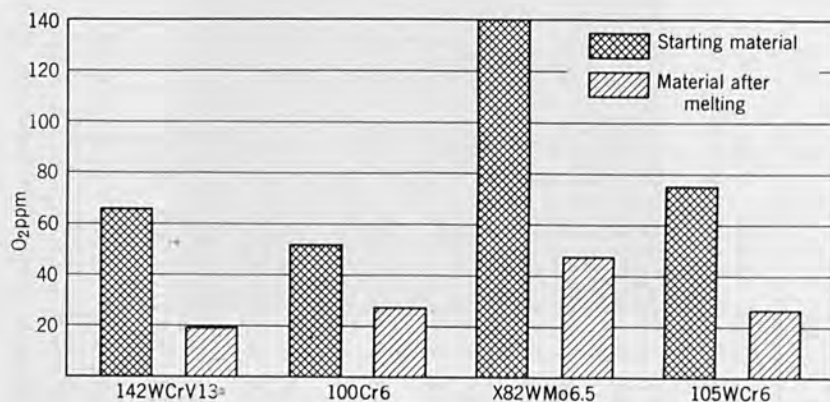


Fig. 7. Comparison of average oxygen content of steels from basic arc furnace and on the same material after remelting in 200-kw EMO.
(1) Initial material. (2) Remelting material.

Producers of high-precision measuring devices with steel marked X 20 Cr 13 (No. 8) suffered considerable material losses during process polishing and especially when the engraving was put on in the end. The engraving needles ran untrue due to non-homogeneities and slag. When vacuum steel was used, these difficulties were eliminated completely.

The extensive and necessary material working investigations for the determination of behavior under practical service condi-

tions in the case of vacuum steel--in comparison to open-air cast steels--have been only partially completed, but they indicate that a comprehensive description of the working material indexes only through the known processes of materials testing is incomplete and does not really tell us much. Proof of quality improvement must be supplemented by life testing. This is also necessary in order to justify the higher price on vacuum-processed steel as far as the steel customer is concerned. Investigations along these lines are now under way for steels 4, 5, and 6, but these studies have not yet been completed and therefore we cannot draw any conclusions at this time.

Stamping dies for the production of knife blades have been made of 40 NiCrMo 15. The first interim result was a reduction in the soft-annealing resistance of this brand of steel--which is very difficult to soft-anneal--down to 79 kG/sq mm. The hobbing capability and thus also the durability of the counterbores were improved.

Pressure molds made of 38 CrMoV 21.14 gave higher life times a consequence of the greater homogeneity and lesser liquation of the strands made in the EMO. In the case of artificial resin pressure molds made of 22 CrMo 4 (No. 6) and the previously mentioned pressure molds, it has been possible to obtain better machining properties, permitting higher cutting speeds for the vacuum steel, as compared to the basic electric steel. According to statements from mold makers, this fact alone justifies the use of vacuum steel for these purposes because the price of the working material is more than compensated for by the reduction in processing costs.

In conclusion we might say that the EMO can be used successfully as metallurgical aggregate for the remelting and refining of steels. On the basis of our results in the operation of the 1200 kW furnace at the Freital Noble Steel Plant, we can expect a decisive improvement in the economy of the method after the commissioning of the 1200-kW furnace which will enable us to make rolled strands for the blooming mill train and forged blocks with a weight of up to 15 tons for the 2000-ton forge press.

REFERENCES

1. A. M. Samarin, *Le Vide (Vacuum)* 97, 53-71 (1962).
2. K. Bunghard and H. Vollmer, *Stahl und Eisen (Steel and Iron)* 82, 401-419 (1962).

3. Neue Huette (New Mill), 7, 19 (1962).
4. V. Pasukhin and A. Fischer, "Vacuum Metallurgy," Moscow, 195 (1956).

ELECTRON-BEAM MELTING OF CERAMIC MATERIALS

INTRODUCTION

A large amount of effort has been devoted to the development of fabrication methods for nuclear fuels and especially ceramics such as UO_2 , UC, UC_2 , etc.

In such cases, the aim usually was a high-density product with physical and chemical characteristics (density, thermal conductivity, microstructure, and impurity content) controlled with high precision.

These efforts have led to a systematic evaluation of a number of different possible methods [1], [2], [3], [4].

One of them, using the electron-beam melting associated to continuous casting, has proved not only of interest for nuclear fuel fabrication, but also as a valuable tool for melting and casting of brittle materials.

The present paper describes: the reasons which have led to its selection, the design of the equipment used, the results obtained for uranium monocarbide and other refractory materials, and a discussion of the field of application of the method and of its possible cost level.

1 SELECTION OF A FABRICATION METHOD FOR URANIUM CARBIDE¹

This problem has been studied in detail in reference [5]. We shall only mention here the basic considerations. The sintering

*Commissariat de L'Energie Atomique, Saclay, France.

¹In this paper, uranium carbide or carbide is used for any uranium-carbon alloy with a composition close to that of the stoichiometric monocarbide.

methods did not appear as recommendable because of the low-production rates associated with hot pressing [6], or reaction hot pressing [7], or of the extremely high atmosphere purity required for a satisfactory cold pressing and sintering [3], [4]. If a melting method is to be successfully applied, two sorts of difficulties have to be overcome: (1) the contamination of the molten carbide, and (2) the casting flaws due to the brittleness of the carbide.

1.1 Contamination of Molten Carbide

The highly reducing properties of the carbide preclude any sort of hot oxide crucible for melting. The only possible containment is, therefore, a "skull" made of the material itself and kept unmelted by a suitable external cooling.

The reactivity of the carbide with oxygen, nitrogen, and moisture, requires melting under noble gas or vacuum. Vacuum is preferable since it allows a "decontamination" of the melt, oxygen and nitrogen being evolved by decomposition of the carboxide [8] or carbonitride [8] dissolved in it.

The high melting point and the necessity of a cooled crucible allow only the most powerful heat sources such as arc, induction, electron beam. Arc, although widely used, presents the drawbacks associated with a pollution of the melt by the electrode material (the consumable electrode fusion has not been successfully applied to carbide). Usually, a carbon electrode is employed which causes some carbon contamination. Induction has been successfully applied [2] to small samples, but the skin effect, which restricts the heat generation to a cortical zone of the heated body, causes this method to show little promise of producing the large quantities required for industrial applications. Electron beam is a "clean" heat source. The only difficulty involved in its use is due to the extreme thinness of the surface layer in which the heat is generated. This characteristic makes the heating of a large quantity of carbide impossible because of the surface over-heating which would be necessary and would provoke a large evaporation of the melt. We shall see later (section 2.1) how continuous casting makes possible the use of a thin, melted layer.

1.2 Casting Flaws

The cold plasticity of the uranium carbide is very low, therefore a casting technique for this material should avoid as completely as possible any thermal stress in the cast product.

A first step in this direction can be made by using continuous casting, the continuous extraction of the solid carbide at the bottom end of the mold establishing a steady-state temperature distribution in the crucible and immediately outside of it, which favors the obtainment of a stress free product.

This first step is nevertheless not sufficient, and the casting of uranium carbide has only been made possible by the following special procedure: the material being cast as a cylindrical rod, a "blanket" of unmolten material is built between the mold wall and the molten carbide. This "blanket" provides a good heat insulation which avoids any excessive temperature gradient in the slug during cooling. Meanwhile, this "blanket" acts as the skull mentioned in section 1.1, and avoids the possibility of a reaction of the solid slug with the mold during its cooling.

2 EQUIPMENT

The survey of the technical requirements for the selection of a fabrication method for uranium carbide, as sketched in section 1, leads to the conclusion that electron-beam melting and continuous casting constitute the "best fit" solution for this problem.

The principle of the apparatus designed for this purpose is illustrated in Fig. 1. An electron gun produces a vertical beam of electrons. This beam is directed to the top opening of water-cooled crucible made of copper. The bottom opening of the crucible is closed by a piston which can be moved vertically. An automatic feeding system supplies the raw material, made of small pre-reacted carbide particles, to the crucible where they are melted and welded to the "target," a refractory part fitted on the top of the piston. The piston is then moved progressively downward as more raw material is fed into the crucible. The velocity of the piston is adjusted so as to keep constant the level of the molten bath.

To build the blanket mentioned in section 1.2, the power and diameter of the electron beam are controlled in order to melt only a core in the particles present in the crucible.

In order to take full advantage of the continuous casting, it is necessary to provide a continuous removal system for the cast slugs. We shall term this section of the equipment "extraction."

Finally, the high flexibility of the electron-beam heating allows an automatic melting control, such a device will be described as "automatic control."

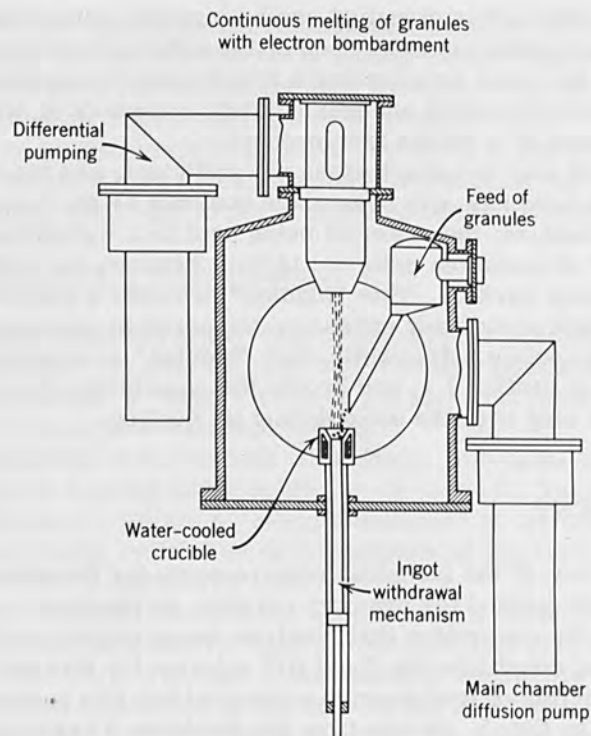


Fig. 1. Electron-beam melting furnace.

2.1 Melting and Casting (Fig. 2)

The electron gun made by ALCATEL has a maximum power of 20 kW. The aperture of the conical beam can be regulated by a magnetic diaphragm. The gun communicates with the melting chamber by a small hole located at the focus of the beam. This provides a high-vacuum impedance for a flow of gas and allows a separate pumping for the electron gun. Such a design enables one to keep a pressure as low as 10^{-6} mm Hg in the electron gun when the pressure in the melting chamber is as high as 10^{-3} mm Hg.

The position of the gun can be adjusted in order for the beam to be exactly centered on the opening of the crucible. This is achieved by two rectangular horizontal motions allowed by the flange connecting the gun to the melting chamber.

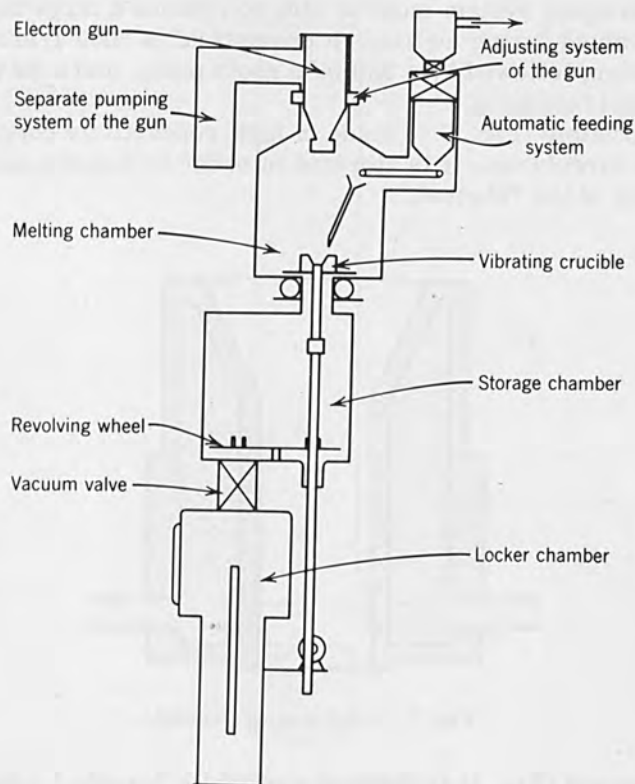


Fig. 2. Sketch of electron-beam melting furnace and continuous casting apparatus.

2.1 A The Melting Chamber. This part is a double-wall cylinder cooled by a water circulation between the two walls. This chamber is provided with the feeding system, the pumping system, and contains the crucible. It is connected with the cooling chamber located underneath.

The feeding system consists of a container loaded with the raw particles; these particles are carried from the container to the crucible by a belt conveyor followed by a funnel. The velocity of the belt can be controlled by regulating the voltage supplied to the electrical motor energizing it.

The pumping system must be able to remove a large amount of gas evolved during melting. It consists of: a 5000 l/sec oil diffusion pump, followed by a 350 m³/h roots pump, and a 60 m³/h mechanical forepump.

The crucible (Fig. 3) is made of high-conductivity copper cooled by water circulation. It is vibrated in order to achieve an even compacity of the "blanket."

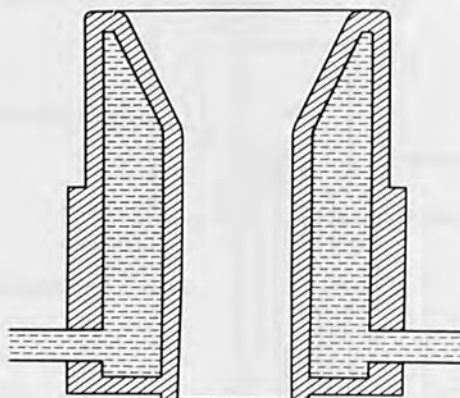


Fig. 3. Cold copper crucible.

The target (Fig. 4) is made of a graphite "candle," adjusted to the crucible diameter, screwed on a stainless steel block which is provided with a skirt to collect the blanket material after removal from the crucible. The piston can be locked to the target in order to bring the latter into the crucible by moving it upward, and to lower the target as the slug is built by feeding the raw particles into the crucible.

2.2 Extraction

When a slug with the desired length has been cast, the piston is moved completely downwards and the target comes at rest and is locked in a hole of a revolving wheel provided with twelve such positions. The wheel is moved in order to bring the next hole in front of the piston, which is provided with a new target to be brought in position for a next casting. The slug is then allowed to cool during the next six castings. It is then taken by a second piston which brings it to a locker chamber.



Fig. 4. Graphite
"candle" serv-
ing as target.

This chamber can be isolated from the cooling chamber by a vacuum valve which is then closed. The locker chamber is brought to atmospheric pressure and opened in order to remove the target and slug and supply the piston with a new target. A vacuum is established anew, and the next target is brought to the wheel where it is located at the same place as the one just removed. It is then ready to be taken by the piston No. 1 and brought to the crucible when the wheel will have moved $6/12$ turn.

2.3 Automatic Control [9]

For automatic operation of the melting and casting, rather than controlling the electron-beam heating, it proved to be easier to keep

steady the heating conditions (power and beam diameter) for a given crucible size and to pilot the feeding rate and the velocity of the piston 1 (casting rate).

As we shall see later (section 3.1), the metallurgical quality of the cast slug is primarily controlled by the residual pressure in the melting chamber. This is why we decided to have the feeding rate piloted by the residual pressure in this chamber, this pressure being kept at a constant preset value.

Because of fluctuations in the quality of the feeding stock, the rate of feeding will have to be varied, as will, in turn, the casting rate. To achieve good casting conditions, the level of the molten bath in the crucible should be kept constant. Because of the high temperature of the bath, the energy lost by radiation is important and constitutes useful information on the bath level in the crucible.

We decided not to use the optical part of this energy because the "dirt" lost by the carbide during melting would have covered the necessary optical systems and rendered them useless. Instead, we used a "thermal detector" made of two thermocouple junctions, one being in the radiation cone of the crucible when the liquid pool is too high, and outside when it is too low, the other thermocouple junction being always outside this cone and acting as a reference thermocouple.

The emf of the two thermocouples are opposed, and the resulting differential emf is then fed to an electronic system controlling the velocity of the motor moving the piston 1. This control system energizes the motor with a voltage adjusted so as to keep the position of the bath steady.

The principle of this regulation is sketched by Fig. 5. At the beginning of an operation, the piston motor 18 is energized by an estimated emf delivered by 96. The resulting thermocouple differential emf is opposed to a constant one in 75 (this constant emf has been determined experimentally as the thermocouple voltage corresponding to the correct position of the bath).

The resulting emf can be either positive or negative. It is amplified in 78 and 81, or 82. It is then fed into a three position switch system constituted of 83, 84, or 85. If the bath is too high, 83 is acted on and the motor 18 is fed with a high voltage. During this time, the motor 101 is excited with impulsions with such a polarity as to increase the voltage delivered by 96 (the frequency and length of impulsions can be adjusted in order to fit the inertia or the system). When the emf enters, 83 has reached a small preset value, 84 is energized, and 18 is fed with the new adjusted voltage through 96.

If the velocity of the piston is too high, the emf delivered by 78 is

[illegible]

If the characteristics of the impulsions acting on 101 are correctly chosen, with a small number of trial and error corrections, the emf delivered by 96 is the right one to keep the position of the bath steady as long as the feeding rate of the particles is kept constant. If the latter changes, a new sequence of corrections is triggered for a new adjustment of the piston velocity is thus practically maintained at the rate imposed by the feeding rate of the bath.

3 RESULTS

3.1 Uranium Carbide

The aim in this case was to cast slugs 20 to 30-cm long with a diameter of 20 to 25 mm. The slugs should be: crack free, of theoretical density (no porosity), with a fine equiaxial microstructure, with a carbon content controlled at least at $\pm 0.1W\%$ absolute,⁽²⁾ and with an oxygen and nitrogen content as low as possible.

We shall successively examine the operating conditions selected and the specifications obtained.

3.1 A Operating Conditions. We have used a crucible with a diameter of 30 mm and particles with a diameter ranging from 2 to 4 mm. This particle size has been imposed by two requirements: the particles should not be too small, in order to prevent them to flow between the crucible and the molten carbide. This would make impossible the formation of the necessary "blanket." On the other hand, they should not be too large, in such a case, they would not be completely digested by the molten bath and instead of a sound molten structure one would obtain porous inclusions in the cast slug.

The casting velocity achievable is dependant upon the residual pressure in the melting chamber. It is therefore governed by the amount of gas evolved during melting and the pumping power. With the equipment used, it has been possible to use casting velocities as high as 300/cm/h. The power necessary varies very little with the velocity and ranges from 3.75 kW for 60 cm/h to 4.1 kW for 300 cm/h. It should be noted that the power is very low because of heat insulation.

3.1 B Macrostructure of the Slugs. Figure 6 represents one of the first slugs obtained. It is roughly 35-cm long, with a mean diameter of 24 mm. It exemplifies the "orange peel" surface due to the "blanket casting."

3.1 C Microstructure of the Slugs. The microstructure of such a slug is shown by Fig. 7. It can be seen that the material is crack-free and contains no void.

² The theoretical carbon content for stoichiometric uranium monocarbide is 4.80%, the relative precision required was then $\frac{1}{48} \sim \pm 2\%$.

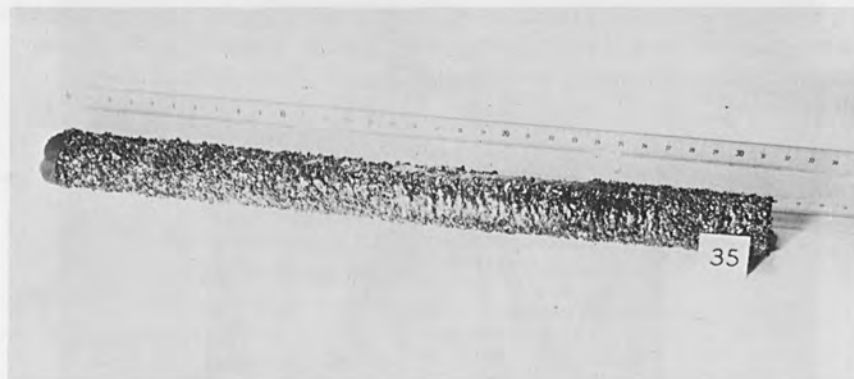


Fig. 6. Electron-beam cast UC slug.

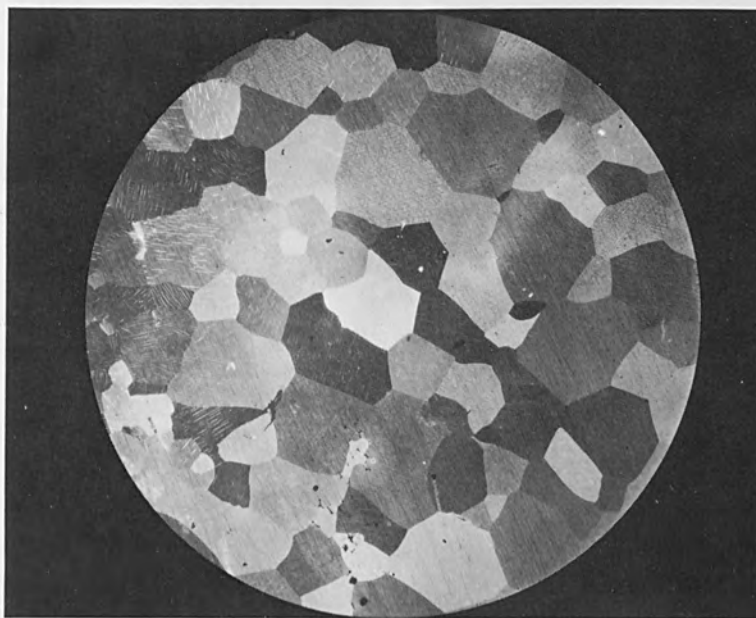


Fig. 7. Microstructure of electron-beam cast UC section parallel to the slug axis casting speed: 120 cm/hr.

We have found a critical casting rate under which the structure is a columnar one, as exemplified by Fig. 8, and above which the structure is equiaxial, as shown by Fig. 7. This rate is about 60 cm/h.



Fig. 8. Microstructures of electron-beam cast UC section parallel to the slug axis (columnar structure obtained for low casting rates).

It should also be noted that the grain size obtained for equiaxial structures is not very dependant on the casting rate, as shown by the comparison of Fig. 8, corresponding to a velocity of 120 cm/h, and of Fig. 9, corresponding to a velocity of 210 cm/h.

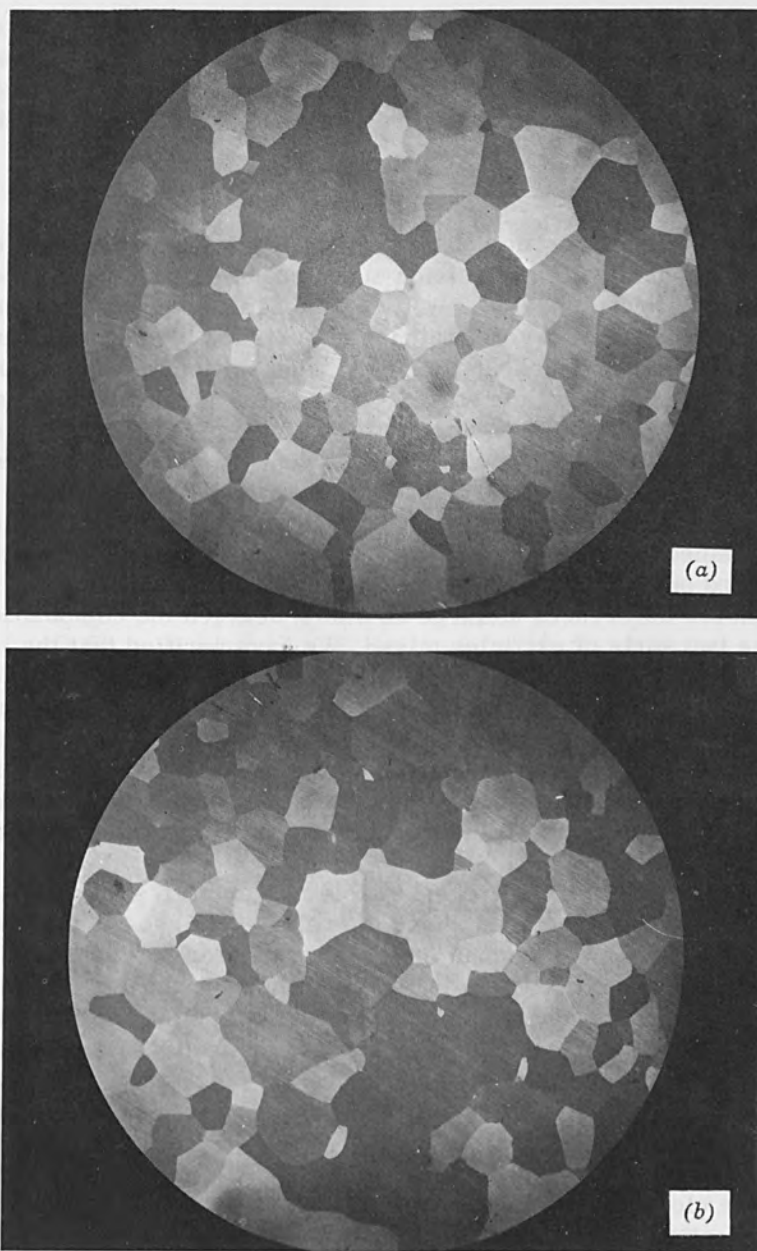
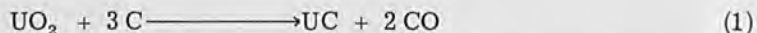


Fig. 9. Microstructure of electron-beam cast UC. (a) Section of the slug by a plan perpendicular to the slug axis. (b) Section parallel to the slug axis casting speed: 210 cm/hr.

3.1 D Carbon Content. The composition of the cast slug being specified, a method has to be devised to achieve a given composition with the required precision. The prereacted particles were prepared by solid-state carbon reduction of UO_2 according to the reaction



The exact composition of the melting stock thus obtained is difficult to control with a precision sufficient to respect the composition bracket required ($\pm 0.2 \text{ W} \% \text{ C}$). Therefore, we have chosen to use a feed made of several batches of particles blended after analysis in such proportions as to hit the composition aimed for. Owing to the pollution of the particles by oxygen during handling, some carbon was lost as CO during melting. For a given handling procedure, this loss was small and fairly reproducible. Experiment led us to accept it as 0.05% absolute.

Table I shows the precision with which a given composition can be obtained. The difference between the values aimed for and the obtained ones is of the order of magnitude of the analytical precision for differences as large as 0.65% between the compositions of the two sorts of particles mixed. We have verified that the blending of the particles did not introduce local fluctuations of composition, provided that the amounts of the two sorts of particles mixed are comparable and that the particles are not too large (2 to 4 mm is satisfactory for a crucible 30 mm in diameter). The precision with which a given composition can be obtained thus appears as ± 0.05 absolute ($\sim 1\%$ relative) and is better than the required one.

3.1 E Oxygen and Nitrogen. Table II exemplifies the usual oxygen and nitrogen contents. Since the values obtained for the lower casting rate are higher than the one obtained for the higher rate, we are led to think that these contents depend upon the gas content of the melting stock and the pumping power, but not on the casting rate (if the gas content was dependent on the casting rate, it should increase with the rate).

3.2 Refractory Oxides

The electron-beam melting and continuous casting, due to the combination of absence of contamination and stress-free cooling seemed particularly well suited for the casting of refractory oxides. The only obvious limitation of the method is the vacuum volatility of the oxide to be melted.

Table I

Carbon % of the Two Sorts of Particles Mixed	Carbon % of the Feeding Stock				Carbon % Obtained	$\Delta_1 C = C_0 - C_1$		$\Delta_2 C = C_4 - C_3$	
	C_0	C_1	C_2	C_3	C_4				
4.96	4.64	4.75	4.70	4.70 $\pm$ 0.05	0.32	0.00 $\pm$ 0.05			
4.94	4.71	4.80	4.75	4.70 $\pm$ 0.05	0.23	-0.05 $\pm$ 0.05			
5.28	4.63	4.85	4.80	4.84 $\pm$ 0.05	0.65	+0.04 $\pm$ 0.05			
5.28	4.63	4.90	4.85	4.80 $\pm$ 0.05	0.65	-0.05 $\pm$ 0.05			

Table II

Casting rate cm/h	Contents ppa	
	N_2	O_2
70	130 $\pm$ 30	400 $\pm$ 200
150	80 $\pm$ 30	150 $\pm$ 50

3.2 A Alumina. We applied this process to alumina. The melting stock was constituted of premelted particles. The power used was 4 kW, and the casting rate 80 cm/h. Figure 10 shows the first slug cast. X-ray analysis identified it as α alumina. Table III describes the composition before and after melting. Some purification took place (Fe, Na, Si, Ti), but the reasons why these elements, rather than others, were removed, are not clear.



Fig. 10. Electron-beam cast alumina slug.

3.2 B Zirconia. Owing to the action of electron bombardment, a white zirconia turned black. This transformation could probably be attributed to the formation of a nonstoichiometric oxide and a very strong piping prevented the making of a slug.

3.3 Refractory Carbides

The method being successful for uranium monocarbide, we tried to apply it to other refractory carbides.

3.3 A Uranium Dicarbide. The melting stock with a composition close to UC_2 was prepared by solid-state carbon reduction, and then melted and cast under conditions very similar to those for monocarbide. We obtained sound rods, the micrographic aspect of which are pictured by Figs. 11 and 12. It should be noted that a single-phase structure has been obtained.

Table III

Variation of the Impurity Content
during Melting and Casting of Alumina

Impurity	Before melting	After melting
B	20	20
Be	10	10
Ca	500	500
Cu	< 10	< 10
Fe	520	250
K	< 10	< 10
Li	< 0.5	< 0.5
Mg	185	160
Mu	20	20
Na	350	150
Ni	< 5	< 5
PG	5	5
Si	300	190
Ti	220	140
Zu	250	250

3.3 B Thorium Dicarbide. A melting stock with a composition close to ThC_2 was prepared by solid-state carbon reduction of ThO_2 . The microstructure of the rod obtained by melting and casting of such a material is shown in Fig. 13.

3.3 C Zirconium Monocarbide. Owing to a very strong volatilization of ZrC , it has not been possible to cast a sound rod of this material.

3.3 D Uranium-Thorium Mixed Monocarbide. In this case, it has been possible to cast rods, but none of them turned out to be crackfree, although the "blanket" thickness was increased up to 10 mm for a 30-mm crucible. The difficulty should be correlated to the extreme brittleness of the cast material.

3.4 Discussion

The variety of materials which have been successfully melted and cast, shows how powerful this method is.

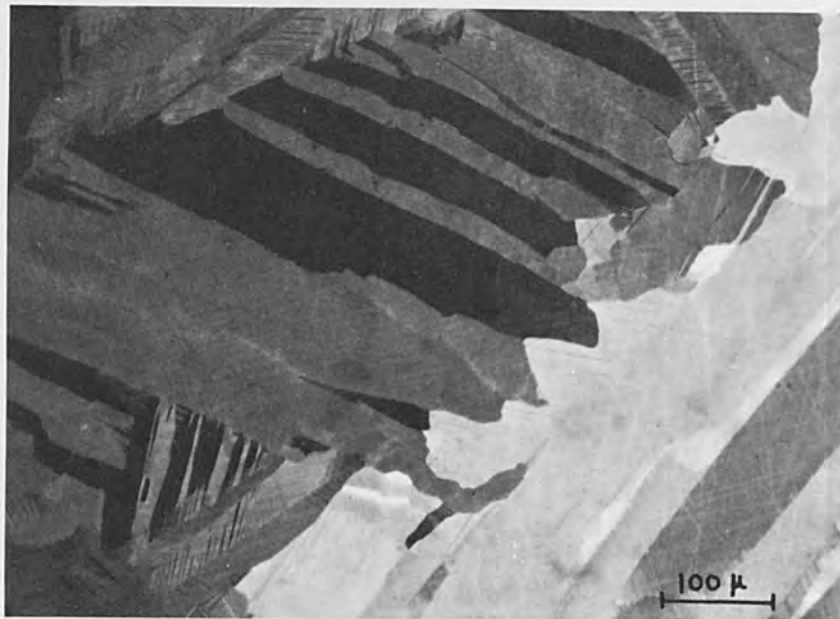


Fig. 11. Microstructure of electron-beam cast UC_2 .

The failure of the method for zirconia, zirconium carbide, and uranium-thorium mixed carbide shows its limitations to be the high volatility and high brittleness of the fabricated materials.

4 PRODUCTION RATE AND COST

From short production runs, it has been established that, for the type of slugs described above for uranium carbide, a production rate of 2 to 3 kG per hour is a very conservative one. If the equipment is used 2000 hours a year, the output is

$$3 \times 2000 = 6000 \text{ kG}$$

This production could probably be easily doubled. If the equipment is amortized in five years, since its cost is roughly 500000 F; the amortization cost per kG is

$$\frac{500000}{6000 \times 5} \approx 17 \text{ F}$$

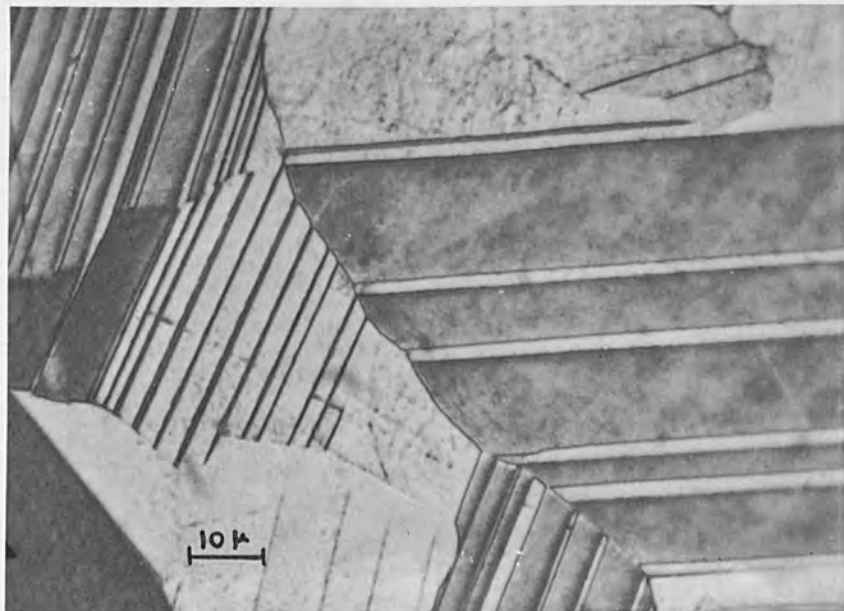


Fig. 12. Microstructure of electron-beam cast UC_2 .

If a yield of 90% is assumed (slugs with flaws, losses, etc.), the net amortization is

$$17 \times \frac{100}{90} \approx 19 \text{ F } (\sim \$ 4)$$

The cost of power is difficult to assess because most of it is consumed by auxiliaries (pumps), but it is certainly low. The necessary manpower can vary with the number of apparatus working in parallel. Because of the automatization, the only period of the making of a slug which requires the operator's intervention is the deposition of the very first layers of particles on the target. It seems reasonable to assume that the same operation could operate four to five furnaces.

5 CONCLUSION

Electron-beam melting associated with continuous casting allows making of sound slugs of brittle refractory materials such as pure

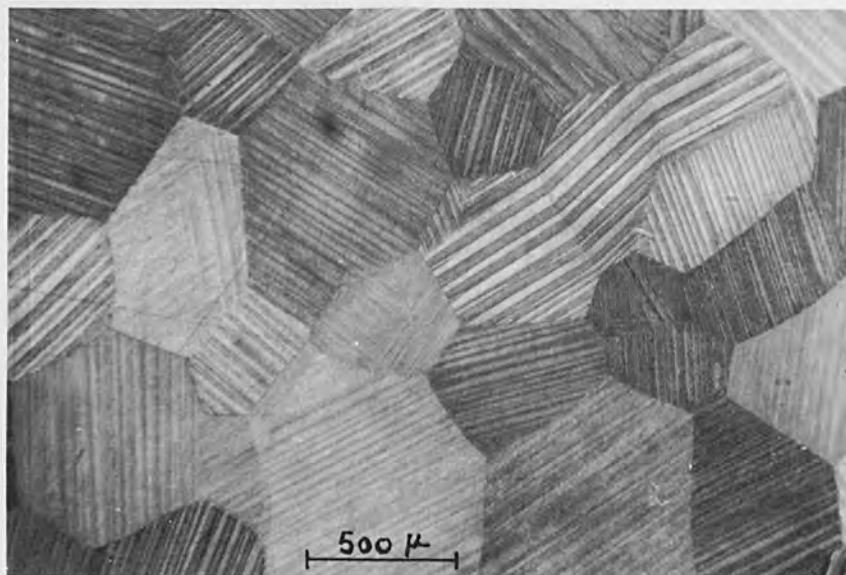


Fig. 13. Microstructure of electron-beam cast ThC_2 .

carbides or oxides. This result is made possible by the use of a special "blanket casting," which results in a slow uniform cooling of the cast material, and additionally provides a skull preventing the contamination of the melt by the crucible material.

Because of the vacuum melting and the "cleanliness" of the electron-beam heat source, a very high purity and good composition control can be achieved. Owing to continuous operation and automatization, the cost for production is low (at least compared to arc melting as used competitively for uranium carbide melting).

REFERENCES

1. A. Accary, "Préparation et mise en forme des alliages uranium-carboné," *J. Mat. Nucl.* 8 No. 3, 281-306 (1963).
2. B. Gayet, J. Holder, G. Kurka, J. Reboux, and J. Trouvé, Brevet CEA, No. P.V. 923.338.
3. P. E. Potter, "The effect of oxygen on the sintering of U - C and $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$," *Symp. on Carbides in Nuclear Energy*, Harwell, B 11, Nov. 5-7, 1963.

4. J. R. McLaren, M. C. Regan, and H. J. Hedger, "The Sintering Behaviour of Uranium Carbide," Symp. on Carbides in Nuclear Energy, B 12, Harwell, Nov. 5-7, 1963.
5. A. Accary and J. Trouvé, "Fusion par bombardement électronique et coulée continue - Méthode de mise en forme des combustibles nucléaires semi-métalliques," New Nuclear Materials Including Non-metallic Fuels Conference, IAEA, Prague (July 1963), vol. II, 375-398.
6. A. Accary and J. Trouvé, "Préparation par frittage d'alliages uranium-carbone," *Revue de Métallurgie*, L I X, No. 2, 105 (1962).
7. A. Accary and R. Caillat, "Study of mechanism of reaction hot-pressing," *J. Am. Ceram. Soc.* 45 No. 7, 347-351 (1962).
8. P. Magnier, J. Trouvé, and A. Accary, "The solubility of oxygen and nitrogen in uranium monocarbide," Symp. on Carbides in Nuclear Energy, Harwell, Nov. 5-7, 1963. A 9.
9. A. Accary, A. Treillou, and J. Trouvé, Brevet P.V. No. 956, 480.

THE PREPARATION AND PROPERTIES OF THE INTERMEDIATE PHASE BASED ON THE COMPOUND $\text{TiCo}^\dagger$

INTRODUCTION

In the course of binary phase diagram studies, many intermediate phases have been found which are based on compounds having the cesium chloride, or ordered body-centered cubic, structure. Not all these phases have been isolated and studied, but where they have, many scientifically interesting features have been revealed. Recent work, especially on the compound TiNi and alloys based on it, has shown that they may also have useful properties as engineering materials.

Because of this interest in TiNi , it was decided to carry out some exploratory work on the closely related compound TiCo . Work on the Ti-Co system, which has been summarized by Hansen and Anderko (1958), has revealed the existence of three compounds, Ti_2Co , TiCo , and TiCo_2 . Duwez and Taylor (1950) and other workers have shown that the structure of TiCo is body-centered cubic, but they did not obtain evidence for ordering. Recently, however, Pietrowsky and Youngkin (1960) have claimed to have observed super-lattice lines in X-ray powder patterns of the compound. The boundaries of the phase based on TiCo have not been established.

The purpose of the present investigation was to find a method of preparing the TiCo phase, preferably for compositions close to stoichiometry, and then to make a preliminary survey of some of its properties. First, therefore, the approximate positions of the phase boundaries were established. Then, a method of preparing

*Materials Research Corporation, Orangeburg, N.Y.

†This work was supported by ARPA under Contract AF 49(638)-1241.

the material in the form of rods was devised and, finally, property measurements were carried out on these rods.

STARTING MATERIALS

The starting materials were purchased from the United Mineral and Chemical Corporation. The titanium was in the form of a 0.375-in. diameter iodide crystal bar. This was electron-beam zone melted and then swaged into rods of 0.125-in. diameter. An analysis was carried out by the National Research Corporation using vacuum fusion techniques for oxygen and hydrogen, a conductometric method for carbon, the Kjeldahl technique for nitrogen, and semiquantitative spectrographic analysis for the other elements. The results are shown in Table I.

Table I

Impurity Content of the Electron-Beam Zone Refined
Titanium Used as a Starting Material

Impurity	Amount Present in ppm by Weight
Oxygen	189
Carbon	24
Nitrogen	15
Hydrogen	8.3
Iron	20
Silicon	20
Aluminium	< 10
Chromium	< 10
Nickel	< 10
Cobalt	< 10
Lead	< 10
Tin	< 10
Magnesium	< 5
Manganese	< 1
Vanadium	ND (< 50)
Molybdenum	ND (< 50)

The cobalt was obtained in the form of 0.120-in. diameter rods for which nominal analysis figures provided by the suppliers are shown in Table II.

Table II

Nominal Impurity Content of the Cobalt Used as
a Starting Material

Impurity	Amount Present in ppm by Weight
Silicon	5
Aluminum	2
Iron	2
Manganese	2
Nickel	1
Copper	1
Magnesium	<1
Calcium	<1
Silver	<1

PHASE BOUNDARY STUDIES

Preparation of the Alloys

In order to establish the approximate range of compositions over which the TiCo phase exists at room temperature, it was decided to prepare and homogenize a series of alloys having compositions ranging from about 46 to 55 at% cobalt and to examine them metallographically. This was carried out by two methods, crucible casting and arc casting.

The first series was made by melting the components together in a molybdenum-wound recrystallized alumina crucible contained in a dynamic vacuum system. In order to minimize the rate of heat evolution during the reaction the titanium, in the form of a rod, was slowly added to the molten cobalt by means of a manipulator controlled from outside the system. The pressure in the system was about 10^{-5} mm Hg.

Using this technique, ten ingots, varying in nominal composition from 46 to 55 at% cobalt, in steps of exactly 1%, and each weighing about 10 grams, were prepared. Because of the lack of stirring during the melting process, it was decided to give the cast specimens a homogenization treatment. This was carried out at 1000°C for 24 hours in a dynamic vacuum system operating at a pressure of 10^{-6} mm Hg. In order to reduce contamination, the specimens

were wrapped in tantalum foil which acted as a getter. The specimens were allowed to cool to room temperature over a period of several hours and then sectioned longitudinally. One-half of each were chemically analysed for titanium and cobalt,¹ and the other examined metallographically. Two of them, those containing nominally 49 and 55 at% cobalt, were found to be slightly inhomogeneous and were therefore rejected.

The second series, which were of the same nominal compositions as the first, were arc-melted in a purified argon atmosphere. Since adequate stirring of the melt occurs in this process it was considered unnecessary to carry out a homogenization anneal. After casting, the specimens were therefore sectioned, analysed, and examined metallographically. All were found to be homogeneous.

Analysis of the Alloys

Wet chemical methods, believed to be accurate to about $\pm 0.02\%$, were used for the titanium and cobalt analyses. The results are shown in Table III. It will be seen that the cobalt and titanium contents fail in every case to add up to 100%, there being an average deficiency of 0.23 wt% for the crucible-cast specimens and 0.18 wt% for the arc-cast specimens. In addition, the crucible-cast specimens lost an average of 0.63 wt% titanium during their preparation, while the arc-cast specimens lost an average of 0.29 wt% cobalt. Table III also shows the cobalt and titanium contents expressed in atomic per cent. These were calculated ignoring the deficit, i.e., by expressing each one as a percentage of the sum of the cobalt and titanium contents.

If it is accepted that the cobalt and titanium analyses are in fact accurate to $\pm 0.02\%$, the deficits encountered must be due principally to impurities introduced during the preparation of the alloys. Possible sources of contamination are: reaction with the alumina crucibles, the pickup of oxygen, nitrogen, and other gases existing as impurities in the argon used in the arc-melting furnace, and the pickup of tungsten and copper from the electrode and hearth of the furnace. To investigate these possibilities, vacuum-fusion analyses for oxygen, nitrogen, and hydrogen, and spectrographic analyses, specifically for tungsten and copper, were carried out on a specimen of both crucible and arc-melted alloys.

¹All of the analyses on the alloys were carried out by the Bridgeport Testing Laboratory, Bridgeport, Conn.

Table III
Analysis Results on Two Sets of Ti-Co Alloys

Nominal Co Content in atomic%	Crucible-Cast Specimens			Arc-Cast Specimens		
	wt% Co	at% Co	wt% Ti	wt% Co	at% Co	wt% Ti
46	52.62	47.59	47.13	51.02	45.94	49.80
47	52.04	47.02	47.70	51.81	46.74	48.01
48	53.20	48.13	46.41	52.68	47.57	47.14
49	...	...	...	53.88	48.78	45.97
50	55.21	50.17	44.56	54.91	49.86	44.88
51	56.16	51.13	43.64	55.27	50.29	44.54
52	56.71	51.67	43.12	57.09	52.06	42.74
53	59.07	54.14	40.70	56.99	51.98	42.81
54	60.48	55.57	39.30	59.58	54.59	40.26
55	...	...	...	60.13	55.20	39.67
						44.80

The results obtained on the arc-cast specimen (Table IV) showed that it contained a fairly high gas content (~ 0.06 wt%) but no detectable tungsten or copper. The results on the crucible cast specimen were, however, less satisfactory. Spectroscopic analysis showed that copper and tungsten were absent, but that the specimen contained an appreciable quantity of aluminum. A total of five gas analyses showed that large and very variable amounts of gas were present. It was therefore concluded that there had been considerable reaction between the molten alloy and the alumina crucible.

The particular arc-cast specimen to which the results of Table IV apply was also analysed for cobalt and titanium. The results obtained were 55.93 wt% for cobalt and 43.92 wt% for titanium. Adding up these two results, and the gas analysis results in Table IV, gives a total of 99.91 wt%, i.e., there is still a deficit of 0.09 wt%. It therefore seems that either there are some impurities not accounted for or the analysis techniques are slightly less reliable than is believed.

Table IV

Impurity Contents of Arc-Melted
Ti-Co Alloys

Impurity	Amount Present in wt%
Oxygen	0.04
Nitrogen	0.02
Hydrogen	0.0012
Tungsten	ND (<0.001)
Copper	ND (<0.0001)

These results leave no doubt that, in this case, arc-casting is the better of the two methods of alloy preparation. It does seem, however, that even this method leads to contamination of the specimens, presumably by impurities present in the argon atmosphere of the furnace. Having prepared the two sets of specimens, however, it was decided to equilibrate and metallographically examine them both.

Metallographic Studies

It has been reported in the literature that eutectics occur on both the titanium- and cobalt-rich sides of the phase diagram at temperatures of about 1020° and 1135°C, respectively (Hansen and Anderko, 1958). The temperature for equilibration of the specimens was therefore chosen to be 1000°C, i.e., just below the lower of the two eutectic temperatures. The crucible cast specimens were annealed at this temperature for 90 hours, which meant that, with the homogenization treatment they had already received, they were annealed at this temperature for a total of 114 hours. The arc-cast specimens were given a single anneal at 1000°C for 114 hours. The two sets of specimens therefore received essentially the same thermal treatments. The cooling rate was controlled so that the specimens cooled to 500°C in about 2 hours. The furnace was then switched off and the specimens were removed when they had reached room temperature.

On metallographic examination it was found, as expected, that some specimens contained essentially two phases and others only one. Of the crucible-cast specimens, those containing 51.13 and 51.67 at% cobalt were single phase, while of the arc-cast specimens, four were single phase, those containing 49.86, 50.29, 51.98, and 52.06 at% cobalt. Many of the specimens, especially those that were crucible-cast, also contained a small amount of a finely dispersed black phase which is believed to be an oxide.

According to our existing knowledge of the Ti-Co phase diagram, the TiCo phase is bounded on the titanium-rich side by a two-phase region in which TiCo is in equilibrium with Ti_2Co , and on the cobalt-rich side by a region in which TiCo and $TiCo_2$ are in equilibrium. It is therefore believed that the structures observed in our titanium-rich alloys consisted of Ti_2Co in a matrix of TiCo, and those in our cobalt-rich alloys of $TiCo_2$ in a matrix of TiCo. Examples of these structures are shown, together with an example of a single-phase TiCo structure, in Fig. 1.

Because of the higher eutectic temperature (1135°C) on the cobalt-rich side of the phase diagram, it is possible to further equilibrate the cobalt-rich alloy at a temperature higher than 1000°C. To take advantage of this, these alloys were given a further heat treatment of 24 hours at 1130°C and were again slowly cooled. This had the effect of converting two alloys which contained small amounts of $TiCo_2$ into single-phase structures. These were the crucible-cast alloy containing 54.14 at% cobalt and the arc-cast alloy containing 54.59 at% cobalt.

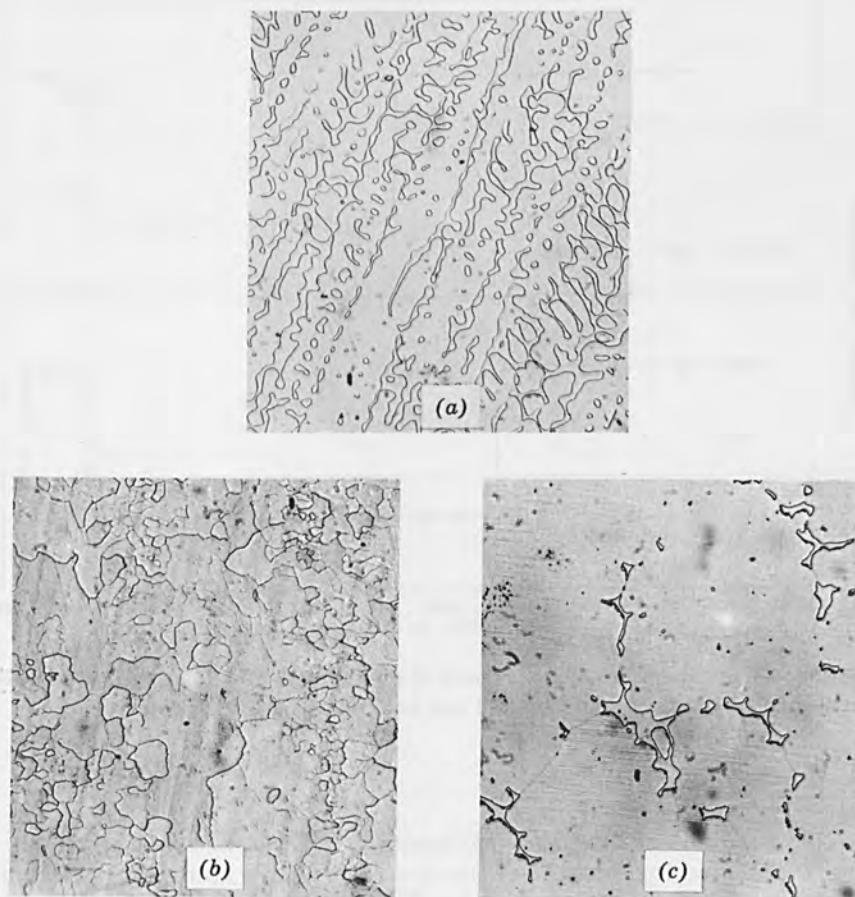


Fig. 1. The microstructures of three Ti-Co alloys showing
(a) Ti_2Co in a matrix of Ti Co,
(b) single-phase TiCo, and
(c) TiCo_2 in a matrix of TiCo (X 150).

The extent of the single-phase region is shown diagrammatically for both types of specimen in Fig. 2. It will be seen that for the crucible-cast specimens, it extends from about 50.7 to 54.8 at% cobalt, and for the arc-cast specimens from about 49.6 to 54.8 at% cobalt. There is an uncertainty in the position of each boundary of about ± 0.5 at%.

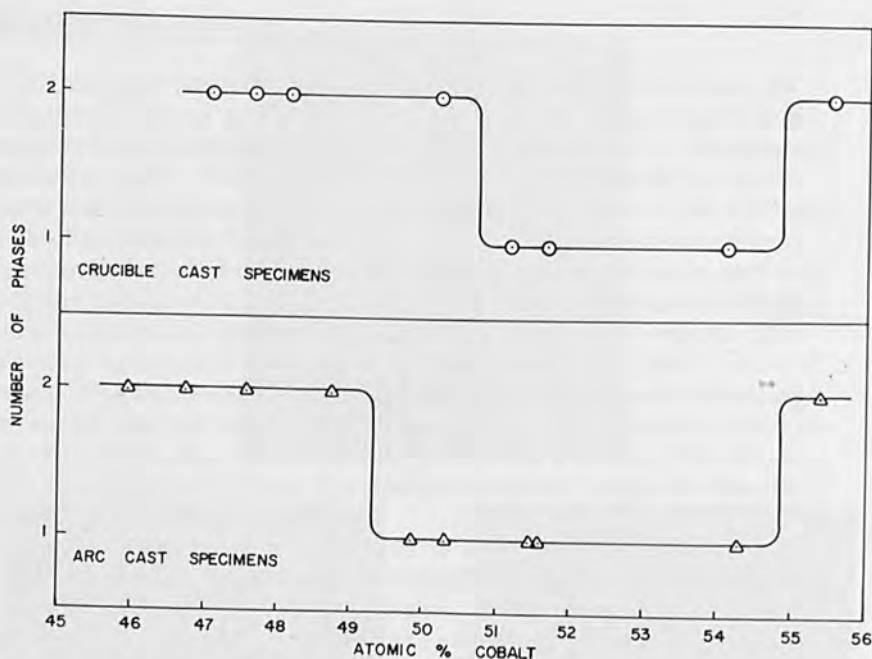


Fig. 2. The composition dependence of the number of phases in equilibrated and slowly cooled crucible melted and arc-melted Ti-Co specimens.

X-Ray Studies

To supplement the metallography, X-ray diffractometer traces were obtained for several of the arc-cast alloys using molybdenum radiation. Powder specimens were prepared for this purpose by crushing a part of each specimen and then annealing the powders obtained at 1000°C for 30 minutes.

The single-phase specimens showed peaks corresponding to a simple body-centered cubic lattices. No superlattice peaks were observed. Specimens which metallographically appeared to contain Ti_2Co as well as TiCo showed an additional set of peaks at angles agreeing well with those reported by Orrell and Fontana (1955) for Ti_2Co . The specimen containing 55.20 at% cobalt, which from the metallographic evidence appeared to be the only one to contain TiCo_2 , gave only TiCo peaks. Probably, however, the amount of TiCo_2 present was too small to be detected by this method. Traces for a specimen containing both TiCo and Ti_2Co and for one containing TiCo only are shown in Fig. 3.

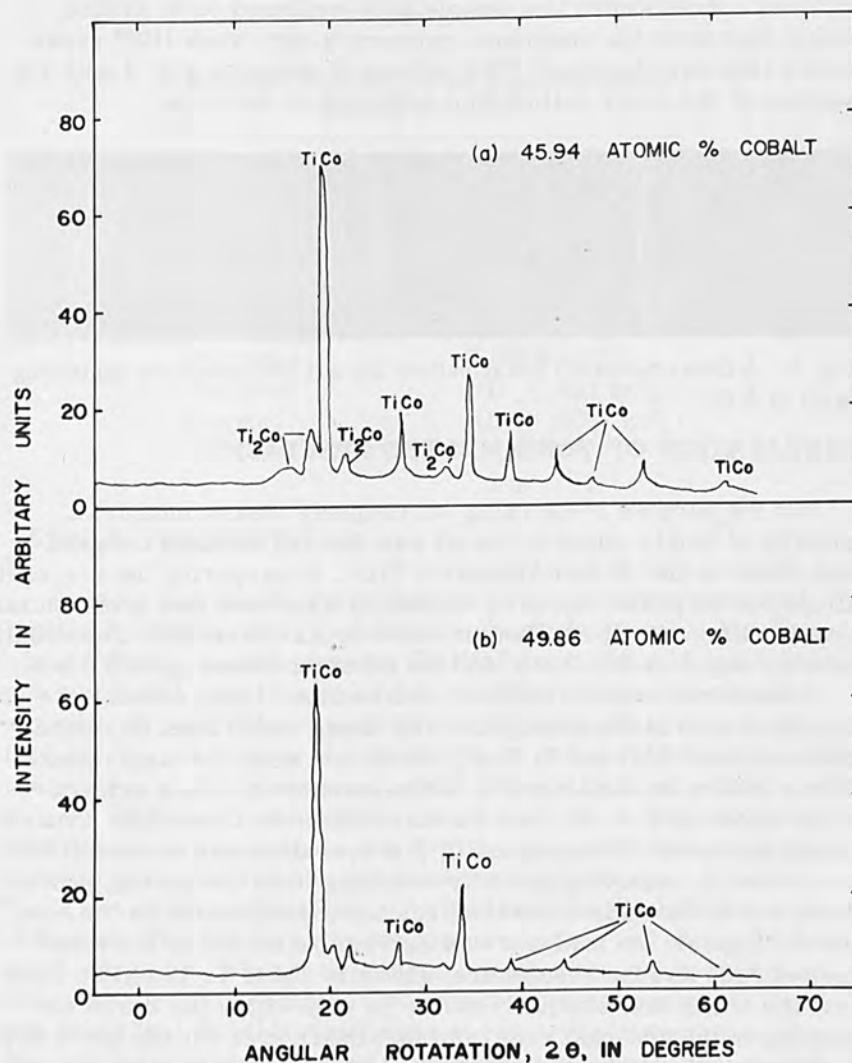


Fig. 3. Room temperature x-ray diffractometer traces for Ti-Co specimens containing (a) 45.94 at % Co and (b) 49.86 at % Co.

To obtain some further evidence as to the degree of order in the TiCo phase, several Debye-Scherrer patterns were obtained using powder prepared by crushing part of the crucible cast specimen containing 51.67 at% cobalt. Filtered cobalt radiation was used in a camera of nominal diameter 114.6 mm operating at 10kV and 10mA. Exposure times up to 30 hours were used. The patterns

obtained corresponded to a simple body-centered cubic lattice, except that after the maximum exposure a very weak (100) super-lattice line was observed. This pattern is shown in Fig. 4 with the position of the super-lattice line indicated by an arrow.



Fig. 4. A Debye-Scherrer x-ray pattern for a Ti-Co specimen containing 51.67 at % Co.

PREPARATION OF ZONE-MELTED SPECIMENS

For the purpose of carrying out property measurements, a quantity of single-phase material was desired having a composition close to that of stoichiometric TiCo. In preparing the arc-cast alloys for the phase boundary studies, it was found that preferential loss of either cobalt or titanium could occur (Table III). The cobalt losses ranged up to 1.0 at% and the titanium losses up to 0.6 at%.

A number of rods of nominal composition 51 at% cobalt were therefore cast in the expectation that these would actually contain between about 50.0 and 51.6 at% cobalt and would be single phase. These rods were then electron-beam zone melted once at a traverse speed of 4 in./hr in a standard MRC electron-beam zone refining apparatus. The purpose of this operation was to consolidate the material, removing porosity resulting from the casting procedure, and at the same time to effect some purification by the evolution of gases. An analysis was carried out on one of the zone-melted rods and the results are shown in Table V. It will be seen that the cobalt and titanium results lie well within the limits expected for the arc-cast rods, and that the figures for the gases are higher than those shown in Table IV for one of the earlier arc-cast specimens. The gas content of this specimen is substantially higher than that of the arc-melted specimen whose gas content is given in Table IV. Since the zone-melted rod was not prepared from this particular arc-melted specimen, it is not clear whether or not the gas content of the zone-melted specimen increased during the zone-melting operation. On the basis of the experience gained in zone-melting pure titanium (Table I) it seems unlikely that the zone-melting did in fact further contaminate the specimen.

Table V

Analysis Results on an Electron-Beam
Melted TiCo Rod

Impurity	Amount Present
Titanium	49.26 at%
Cobalt	50.74 at%
Oxygen	0.167 wt%
Nitrogen	0.028 wt%
Hydrogen	0.0015 wt%
Tungsten	ND (< .001 wt%)
Copper	ND (< .0001 at%)

TENSILE PROPERTIES

To study the tensile properties of the zone-melted material, tensile specimens were ground from the rods using an Alundum wheel. These specimens had a nominal gauge diameter of 0.075 in. and a nominal gauge length of 0.5 in. Before testing they were annealed at 1000°C for 1 hour to recrystallise their deformed surface layers. They were then tested in an autographically recording tensometer of the type described by Adams (1959) at a strain rate of $1.0 \times 10^{-4} \text{ sec}^{-1}$; the tests were carried out at temperatures between room temperature and 750°C using a Kanthal wound furnace through which argon was slowly circulated.

At temperatures up to 550°C completely brittle behavior was observed. The fractures were transcrystalline and showed "river line" markings. When a longitudinal section through a specimen tested at room temperature was examined metallographically, it was found that the cleavage facets were curved rather than planar. The fracture stresses tended to increase with increasing temperature but, as is usual with brittle materials, showed considerable scatter (Fig. 5).

Specimens tested at 600°C and 650°C showed very small amounts of plastic deformation (<0.1%). At 675°, 700°, and 750°C, however, much more deformation was observed and the load-elongation curves showed pronounced yield points. The specimens tested at 700° and 750°C necked down to a point and fractured after about 30% plastic strain.

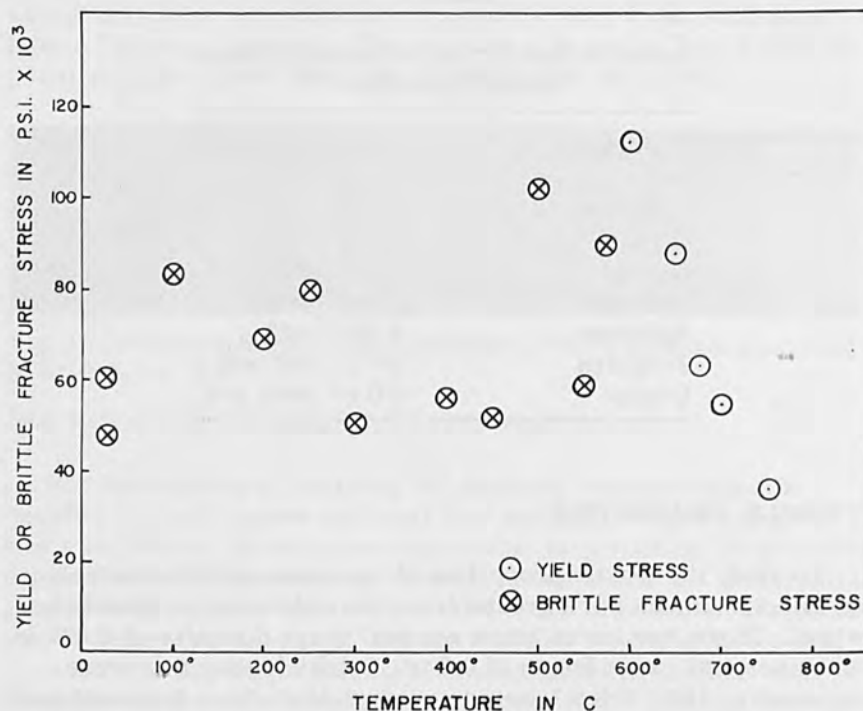


Fig. 5. The temperature dependence of the tensile properties of TiCo.

Metallographic examination of some of the fractured specimens revealed that their grain structure was somewhat inhomogeneous. The grain diameters were, however, of the order of 1 mm. All of the specimens tested at or above 350°C were slightly oxidized during testing.

ELECTRICAL RESISTIVITY MEASUREMENTS

Electrical resistivity measurements were carried out on one of the tensile specimens using the four-point probe method. Measurements were made at the temperatures of liquid helium, liquid nitrogen, and solid CO₂ + acetone, and at room temperatures and 86°C. The last of these temperature was achieved using a silicone oil bath. The results, which are shown in Fig. 6, show that the resis-

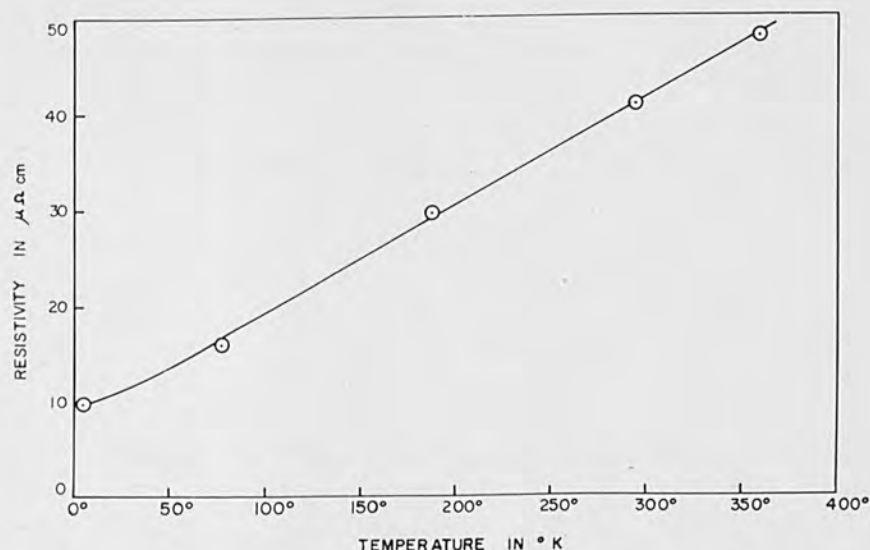


Fig. 6. The temperature dependence of the electrical resistivity of TiCo.

tivity varies from $47.9 \mu\Omega\text{cm}$ at 86°C to $9.6 \mu\Omega\text{cm}$ at -269°C . The room temperature value is $40.8 \mu\Omega\text{cm}$. It is estimated that the error in each measurement is about $\pm 2\%$.

OXIDATION AND CORROSION EXPERIMENTS

Oxidation in Air

To obtain some idea as to the oxidation rate of TiCo in air at high temperatures, a simple weight gain experiment was performed on a ground cylindrical arc-cast specimen approximately 0.25 in. in diameter and 0.5 in. in length. The cylinder was carefully weighed, placed upright in an alumina boat, and inserted into a platinum-wound furnace. It was then annealed for 1 hour at each of a series of successively higher temperatures, and weighed after each anneal.

For temperature up to and including 300°C there were no visible signs of oxidation and no measurable weight gains. From 400°C to 700°C , an oxide film was visible but no gain in weight was detected.

At 800° and 900°C weight gains were detected and after the 900°C anneal the oxide began to flake and the experiment therefore had to be stopped. Figure 7 shows the weight gain plotted against annealing temperature.

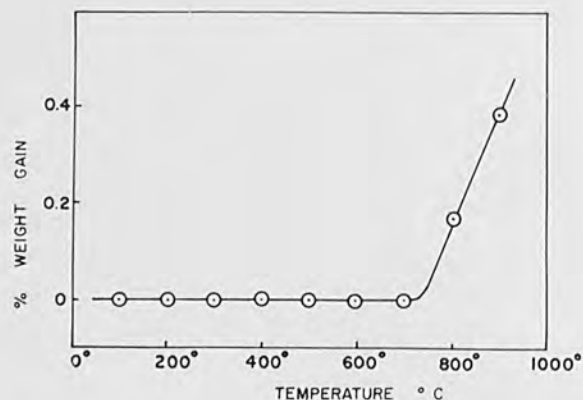


Fig. 7. Results illustrating the temperature dependence of the oxidation rate of TiCo in air.

A transverse section of the oxidized specimen was examined metallographically. The structure observed is shown in Fig. 8.

Corrosion in Liquid Environments

A second cylindrical specimen of approximately the same dimensions as the first was prepared for some experiments on the corrosion of TiCo in liquid environments. The liquids chosen were (a) tap water, (b) a 3 wt% solution of NaCl in water, (c) a 4 wt% solution of NaOH in water, (d) conc. HNO_3 , and (e) conc. HCl. All of the tests were carried out at 80°C.

The cylinder was weighed and then immersed in its environment for several successive periods of 1 hour. A weighing was carried out after each immersion. Tap water, NaCl solution, and NaOH solution were found to have no effect either on the appearance of the specimen or its weight. Concentrated HNO_3 dissolved it slowly, but concentrated HCl dissolved it rapidly, so that after an hour about 40% of it had dissolved. It was also observed that grain boundaries were strongly etched by the HCl.

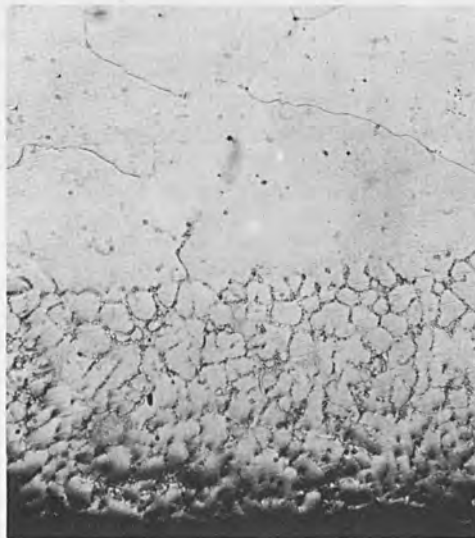


Fig. 8. A micrograph showing the structure of an oxidized rod (X150).

The results obtained with tap water, NaCl solution, NaOH solution, and conc. HNO_3 are shown in Fig. 9.

DISCUSSION

Phase Boundary Studies

The fact that the range of composition over which the TiCo phase could be obtained was different for crucible and arc-cast specimens presumably means that this range is sensitive to the impurity content of the alloys. If this is the case, the results obtained on the arc-cast specimens, the purer of the two sets, may still be in error to some extent since even these contained a relatively high impurity content. An objective of future work must therefore necessarily be the study of purer alloys. Perhaps the most obvious way of achieving this would be by paying more attention to the purity of the inert gas under which the arc-casting is carried out.

Although this work has established the approximate composition limits of the TiCo phase at room temperature, more careful

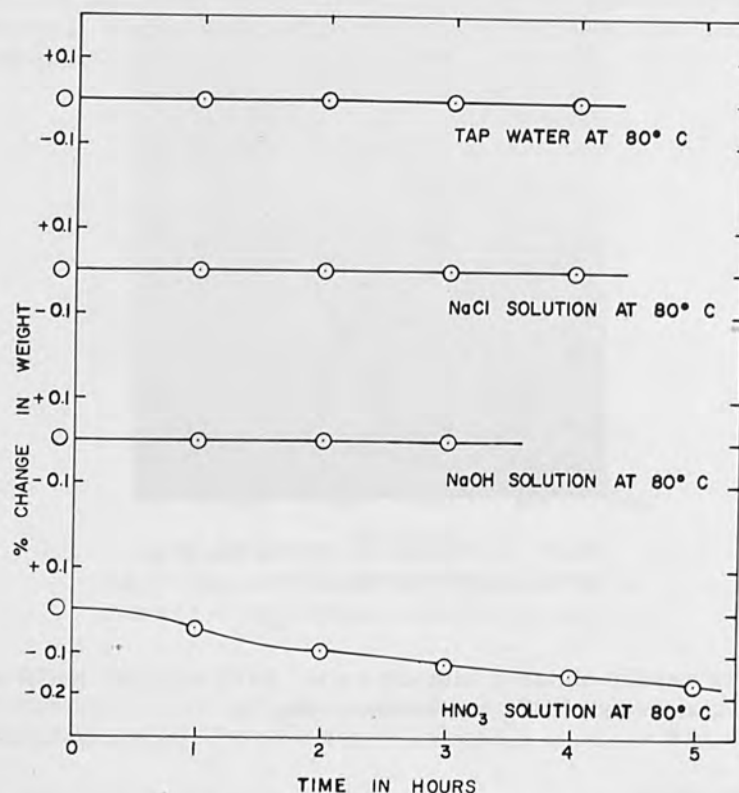


Fig. 9. Results illustrating the corrosion rate of TiCo in several liquid environments.

work, in which specimens are equilibrated at various temperatures and then quenched, will be required to find out how these limits change with temperature. It is also necessary to determine the variation of the solidus temperature with composition and to establish whether or not the stoichiometric compound melts congruently.

The presence of a weak superlattice line in the Debye-Scherrer pattern shown in Fig. 4 clearly indicates the presence of some ordering in TiCo. This is in agreement with the conclusion reached by Pietrowsky and Youngkin (1960). The absence of superlattice lines other than the (100) suggests, however, that the ordering might be rather weak. Some further work on this problem, perhaps using

neutron diffraction, therefore seems desirable. If the phase is ordered it must be a defect structure, at least for compositions other than stoichiometric. It should be possible, using a technique such as a combination of lattice parameter and density measurements, to find out the type of defect structure that is present. Work of this sort has already been carried out on some of the other cesium chloride type phases (e.g., Bradley and Taylor, 1937; Cooper, 1963).

Tensile Properties

The occurrence of a ductile-brittle transition in TiCo is of course in keeping with the behavior of most other body-centered cubic metals and compounds (e.g., Westbrook, 1960; Westbrook and Wood, 1963). The work of Westbrook and Wood in particular has shown that the ductile-brittle transition temperature of an intermetallic compound is extremely sensitive to composition and may change by several hundred degrees on going from one side of the stoichiometric composition to the other as the result of a grain boundary segregation effect. The present results must therefore be regarded as characteristic of one range of compositions only.

The tendency for the brittle fracture stress to increase with temperature is an unusual type of behavior but has been observed before in intermetallic compounds (e.g. Lowrie, 1952; Savitskii, 1960). It has usually been interpreted as due to the presence of small defects or cracks in the specimens. The stress concentrations arising from these defects or cracks can be relieved by plastic deformation more easily as the ductile-brittle transition temperature is approached, and the stress required to propagate one of them across a specimen therefore increases.

The development of oxide films on the specimens during the tests probably also influenced the results to some extent. It is well known that such a film can raise the yield stress of a metal and recently Hernaez, Smallman, and Adams (1963) have shown that it can also raise the ductile-brittle transition temperature quite substantially. In future work, more elaborate precautions should therefore be taken to avoid oxidation of the specimens.

Electrical Resistivity Measurements

The value obtained for the room temperature electrical resistivity of TiCo ($40.8 \mu\Omega \text{ cm}$) is characteristic of metallic conduction and is in fact close to the corresponding value for pure titanium.

The resistance ratio, however, i.e., $R_{r.t.}/R_4^{\circ K}$, has a value of about 4, which is lower than is usually encountered even in fairly impure metals.

The principal impurities in the TiCo, oxygen, and nitrogen, were present to the extent of 0.1 to 0.2 wt%. Nothing is known about the effect of impurities on the electrical resistivity of TiCo or any other cesium chloride type intermetallic compound. It is known, however, that in one body-centered cubic metal, niobium, the effect of oxygen and nitrogen is of the order of $50 \mu\Omega \text{ cm/wt}\%$ (Ang and Wert, 1953). If the same figure applies roughly to TiCo, the contribution of the oxygen and nitrogen to the resistivity will be 5 to $10 \mu\Omega \text{ cm}$. These two impurities alone could therefore lead to a value of the resistance ratio in the range 4 to 8, and could account for the low ratio. In addition to an effect due to impurities, there may also be contributions due to disorder, and to vacancies if the compound is not exactly stoichiometric.

Oxidation Experiments

The most interesting feature of the oxidation and corrosion behavior observed with TiCo is the metallographic structure shown in Fig. 8. This appears to show a band of internal oxidation advancing into the specimen.

The conditions under which internal oxidation can occur in an alloy have been studied extensively by Rhines (1940) and others and are quite well understood. One component of the alloy must react more readily with oxygen than the other, oxygen must be soluble in the alloy and be able to diffuse rapidly through it at the oxidation temperature, and the oxide formed must be relatively insoluble in the matrix. All of these conditions must therefore be satisfied by TiCo at 900°C . It is presumed that it is titanium oxide that is precipitated.

SUMMARY

It has been shown that in equilibrated and slowly cooled specimens, the intermediate phase based on TiCo exists over the composition range 49.5 ± 0.5 to 54.5 ± 0.5 at% cobalt.

The specimens used in determining this composition range were prepared by arc-melting and were of 99.8 to 99.9 wt% purity. The principal impurities were oxygen and nitrogen.

Arc-cast rods have been satisfactorily electron-beam zone

melted without appreciable preferential loss of either cobalt or titanium. The zone melting does not produce appreciable purification.

Some of the properties of TiCo specimens having compositions within about 1.5 at% of stoichiometry have been measured and are summarized in Table VI.

Table VI
A Summary of the Properties of the
Compound TiCo

Property	Value	Comments
Brittle fracture stress at r.t.	55,000 psi	Fractures are transcrystalline
Ductile-brittle transition temperature	$\sim 575^{\circ}\text{C}$	Unnotched tensile specimen ($\epsilon = 10^{-4} \text{ sec}^{-1}$)
Yield stress at 700°C	65,000 psi	Shows a yield point
Electrical resistivity at (a) -269°C	$9.6 \mu\Omega \text{ cm}$	-
(b) 25°C	$40.8 \mu\Omega \text{ cm}$	-
Temperature coefficient of resistivity ($0-100^{\circ}\text{C}$)	$0.0029/^{\circ}\text{C}$	-
Oxidation rate in air at (a) 300°C	-	No Oxide film formed and no weight gain
(b) 400°C	-	Oxide film formed but no weight gain
(c) 800°C	-	Oxide film formed and small weight gain
Corrosion rate at 80°C in (a) tap water	-	No effect
(b) NaCl soln.	-	No effect
(c) NaOH soln.	-	No effect
(d) conc. HNO_3	-	Slow dissolution
(e) conc. HCl	-	Rapid dissolution

ACKNOWLEDGMENTS

This work was supported by the Advanced Research Projects Agency under contract AF 49(638)-1241. The authors are indebted to many members of the MRC staff for their help and cooperation, but especially to J. Varon, R. Vigue, A. Iannucci, N. Sabisch, R. Schwabik, and N. DelFrate. It is also a pleasure to acknowledge valuable discussions with Dr. Benjamin Post who also took the Debye-Scherrer Photographs.

REFERENCES

- Adams, M.A., 1959, *J. Sci. Instr.* **36**, 444.
Ang, C., and C. Wert, 1953, *Trans. AIME* **197**, 1032.
Bradley, A.J., and A. Taylor, 1937, *Proc. Roy Soc. (London)*, **A**, 159, 56.
Cooper, M.J., 1963, *Phil Mag.*, **8**, 805; *Ibid* 811.
Duwez, P., and J.L. Taylor, 1950, *Trans. AIME* **188**, 1173.
Hansen, M., and K. Anderko, 1958, "Constitution of Binary Alloys," McGraw-Hill, New York, p. 511.
Hernaes, J., R.E. Smallman, and M.A. Adams, 1963, *J. Inst. Metals*, **91**, 279.
Lowrie, R., 1952, *Trans. AIME*, **194**, 1093.
Orrell, F.L., and M.G. Fontana, 1955, *Trans. Am. Soc. Metals*, **47**, 554.
Pietrowsky, P., and F.G. Youngkin, 1960, *J. Appl. Phys.*, **31**, 1763.
Rhines, F.N., 1940, *Trans. AIME*, **137**, 246.
Savitskii, E.M., 1960, "Mechanical Properties of Intermetallic Compounds," John Wiley and Sons, New York, J.H. Westbrook (Ed.), p. 87.
Westbrook, J.H., and D.L. Wood, 1963, *J. Inst. Metals*, **91**, 174.
Westbrook, J.H. (Ed.), 1960, "Mechanical Properties of Inter-metallic Compounds," John Wiley and Sons, New York.

EFFECT OF ELECTRON BOMBARDMENT HEATING ON SURFACE OF TUNGSTEN

INTRODUCTION

Recent developments in electron-beam technology have provided an efficient means of heating for annealing and mechanical property evaluation of refractory metals up to the very high temperatures near their melting points. However, it has been observed that at these high temperatures and for long periods of time, bombardment often produces changes in the surface of the specimen taking the form of a general roughening, Fig. 1, herein referred to as damage [1]. The deterioration of anodes in vacuum tubes has also been attributed to impulses of heat flow due to electron bombardment [2]. Electrons bombarding the surface of materials penetrate and dissipate their kinetic energy within a very shallow layer. It can be calculated [3] that a 100-kV beam bombarding tungsten dissipates its energy in a layer 8×10^{-3} mm in depth, while a 10 kV beam dissipates its energy in only a 10^{-4} mm layer. Heat generated in such a thin layer implies the possibility of rapid heating and the existence of a steep temperature gradient.

It was the purpose of this investigation to examine the morphology and causes of the damage to electron-bombarded surfaces, as well as to determine operating conditions under which it occurs. The effects of heating tungsten in the range of 1600 to 3000°C by thermal radiation and by electron bombardment with both stationary and oscillating beams were compared. Grain surfaces and grain boundaries in both coarse and fine grain sheet specimens as well as foils were separately examined to determine the effects of each mode of heating.

*U.S. Naval Laboratory, Washington, D.C.

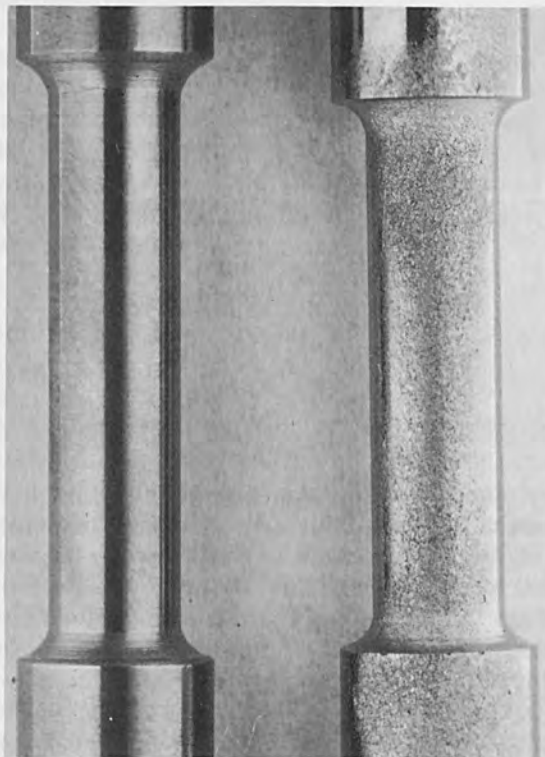


Fig. 1. Typical damage to surface of tungsten specimen by electron bombardment. (Left: unexposed; right: damaged at 60 cps and 2000° C). X2.7.

EXPERIMENTAL PROCEDURE

Equipment

In this study specimens to be examined for the effects of bombardment were heated by 10-kV electron beams generated in two 3-kW self-accelerating guns. The guns are positioned 180 degrees apart, as shown in Fig. 2, so that both sides of a specimen can be heated simultaneously. The beams, which may be focused to spot diameters of 0.5 to 2.0 cm, can either be held stationary or be caused to oscillate sinusoidally and sweep over the surface of the

specimen at frequencies from 20 to 3000 cps. It should be mentioned that the beams are not uniform in density and the accelerating high voltage contains a 4% ripple. Also, the filaments and other beam-control circuits are powered by alternating currents. The total power of the beams, and hence the temperature of the specimen, is controlled by the adjustment of emission current. The guns and the specimen are contained in a water-cooled chamber evacuated to an operating pressure of 8×10^{-6} torr.

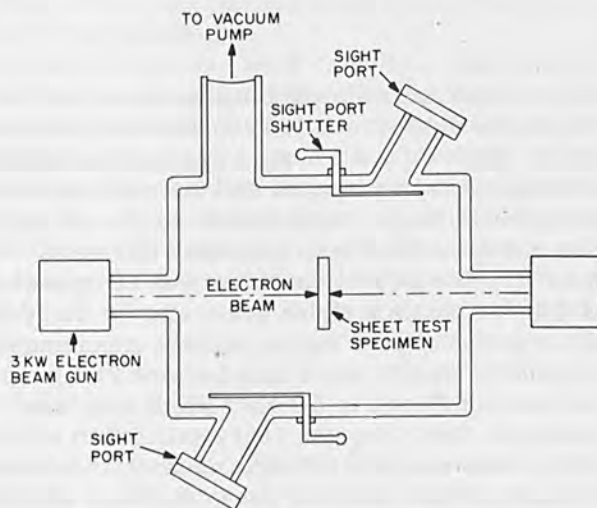


Fig. 2. Schematic plan view of electron-beam furnace.

The specimen could be heated not only by direct bombardment of the surface but also by thermal radiation using two susceptors adjacent to the specimen. In addition, when only one side of a specimen was bombarded, thermal radiation heating was simulated on the opposite side. It is not likely that electrons bombarding one surface penetrate the entire thickness of the specimen and dissipate energy in the opposite surface or otherwise disturb it. Even with the foil, such penetration is unlikely since its thickness of 0.13 mm is far greater than the calculated penetration depth of 10^{-4} mm. Since the surface of the side opposite the beam displays the same characteristics as surfaces heated to the same temperature between susceptors, these two means of heating will be considered equivalent.

The procedure for temperature measurement employs the standard technique of drilling a black-body hole (depth to radius ratio of 16) into a dummy specimen whose surface has the same geometry and finish as that of a test specimen. After correcting for sight glass attenuation, an empirical correlation between surface brightness and black-body temperature was established with a calibrated optical pyrometer [4]. To minimize the errors introduced from metallic vapor deposits, shutters covering the sight port are opened only when making measurements.

Materials

Three heats of nominally 99.95% tungsten were used in the study; two heats (WG and WH) were of 1/16-in. sheet and the other heat (WF) of 0.005-in. thick foil. All heats were manufactured by standard powder metallurgical techniques and their spectro-chemical analyses showed their major impurities to be Fe, Al, Mo, and Si. Specimens for examination of both sheet and foil were cut into tabs 2-1/8 in. by 3/8 in. Each sheet specimen was recrystallized for 90 minutes at 2300°C to obtain a stable grain size so that grain boundary migration would not occur during damage experiments. Only the foils were not so treated since they become brittle on recrystallization and more difficult to handle without breakage.

The two heats of sheet tungsten recrystallized to widely different grain sizes. One heat (WG) characteristically recrystallized to an unusually coarse grain size of 1 cm in diameter which facilitated the separate examination of damage to grain boundaries and grain surfaces. The other heat (WH), however, recrystallized to a fine grain size of 1 to 2 mm in diameter and permitted examination of damage in more commonly found polycrystalline materials.

A smooth surface was obtained by electropolishing all specimens in a 2% sodium hydroxide solution at 1 amp/cm² current density. Prior to electropolishing, however, the sheet specimens required mechanical polishing to 3/0 paper.

Experiments

In order to better understand the nature and causes of damage produced by electron bombardment, the following experiments were performed. First, damage in grain boundaries and on grain surfaces was systematically observed as it developed under stationary and oscillating beams at various temperatures. These effects were compared to changes manifested by thermal radiation heating.

Second, by comparing the depths of grain boundary grooves, the temperature difference between the side bombarded by a stationary beam and the side opposite was investigated in order to determine if any unusual temperatures were present in the surface.

Third, an attempt was made to determine if thermal cycling was related to the type of damage produced by oscillating beams. Since foils have a lower thermal inertia than do the sheet specimens, it was possible to induce a rapid cyclic change of temperature on the side of the foil opposite a beam sweeping at low frequency. Thus cyclic thermal radiation heating could be effected in the absence of bombardment.

A fourth experiment was performed to investigate the effect of temperature on the extent of damage by oscillating beams when the magnitude of each thermal pulse, as the beam passes a point on the specimen, is held constant. Also, the effect of the magnitude of thermal pulse at equal temperatures was examined.

An expression was developed to calculate this magnitude, ΔT , in terms of the sweep frequency, beam power, beam diameter, and sweep amplitude. By inspection of the diagram provided by Vibrans [3], it may be seen that for the range of dwell times used here the relationship

$$\Delta T = CP (\Delta t)^{1/2} \quad (1)$$

can be assumed applicable. The beam power density, P , is given in kW/cm² and the dwell time, Δt , is defined as the time, in seconds, taken by the beam to pass over a given point of the specimen. By selecting applicable values of ΔT and Δt for tungsten from the diagram, the constant, C , for a 1 kW/cm² beam power density was evaluated to be 447.2 degrees cm²/(sec)^{1/2} kW. Since the beam was oscillated sinusoidally according to $A \cos(2\pi \nu t)$, the relationship may be derived that

$$\Delta t = \frac{1}{2\pi\nu} \left| \cos^{-1} \left(\frac{x - L/2}{A} \right) - \cos^{-1} \left(\frac{x + L/2}{A} \right) \right| \quad (2)$$

where ν (cps) is the sweep frequency, x (cm) is the distance along the sweep path measured from its midpoint, A (cm) is the half amplitude of sweep as measured from beam centers, and L (cm) is the diameter of the beam. This relationship holds for

$$0 \leq |x| < \left| A - \frac{L}{2} \right|$$

where the surface is bombarded twice per cycle. Points at and beyond $(A - L/2)$ are exposed to the beam once per cycle during the movement of the beam to the end of the sweep and return so that for

$$\left| A - \frac{L}{2} \right| \leq |x| \leq \left| A + \frac{L}{2} \right| ,$$

$$\Delta t = \frac{1}{\pi \nu} \left| \cos^{-1} \left(\frac{x - L/2}{A} \right) \right| . \quad (3)$$

Substituting equation 2 into equation 1 yields an expression for the thermal pulse magnitude at any point of the sweep. Thus:

$$\Delta t = \frac{447.2}{\sqrt{2\pi\nu}} (P) \left| \cos^{-1} \left(\frac{x - L/2}{A} \right) - \cos^{-1} \left(\frac{x + L/2}{A} \right) \right|^{1/2} \quad (4)$$

$$\text{for } 0 \leq |x| < \left| A - \frac{L}{2} \right|$$

and by substituting equation 3 into equation 1 a similar expression for ΔT at the points near the ends of the sweep may be obtained.

A variety of combinations of beam power densities, 2-14 kW/cm², and sweep frequencies, 30-1470 cps, were so selected that the calculated amplitude of each thermal pulse was held constant even though the mean temperature varied from 1520 to 2540°C.

RESULTS AND DISCUSSION

Forms of Damage

The effect of bombardment on tungsten surfaces takes several forms. These include rippling and pitting of grain surfaces, grain tilting, uneven grain boundary grooving, and cracking. Figure 3 shows, in a general way, how these forms of damage progressed with increasing beam power and temperature. Each mode of heating produced characteristic changes in the surface. The surfaces of bombarded specimens were examined by light, interference, and electron microscopy as well as X-ray diffraction.

Heating by thermal radiation produced two different effects. At 1400°C for 150 minutes, grooving, of the type shown in the interference micrograph in Fig. 4, was apparent in both coarse grain and fine grain sheet material as well as foils. The grooving here should be noted for its evenness, a condition which persists to very high temperatures. At 3300°C, which is near the melting point of

tungsten, the surfaces of grains displayed pits, or facets, as shown in Figs. 5 and 6.

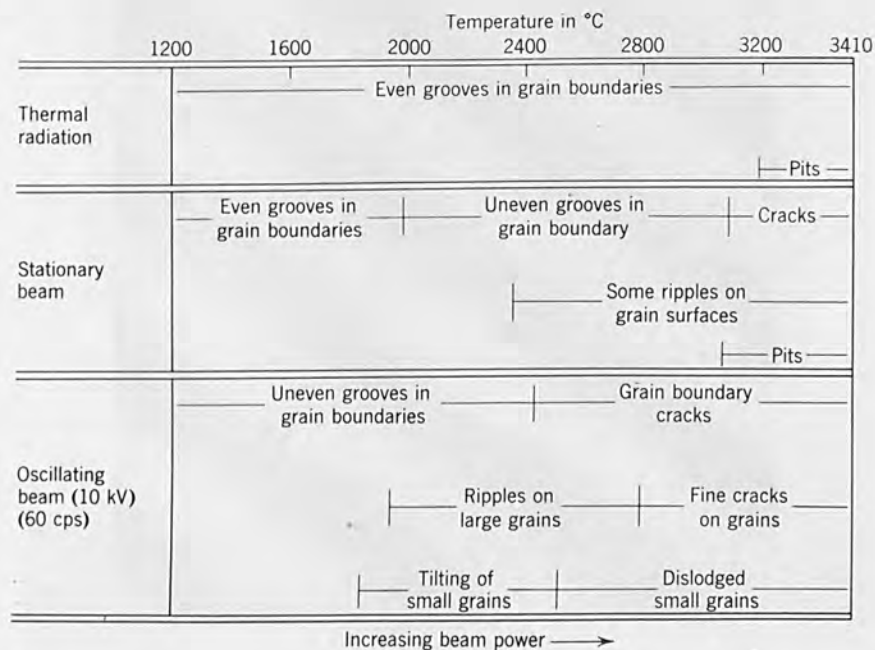


Fig. 3. Approximate temperature ranges in which various surface effects are induced in tungsten by different modes of heating for 30 minutes. (*Stationary beam has voltage and current fluctuations.)

Stationary beams produced changes in the surfaces of grains and grain boundaries similar in most respects to those produced by thermal radiation both at low and high temperature. At high temperatures near 3000°C, pitting on grain surfaces under the stationary beam had a form similar to the pitting produced by thermal radiation, but in addition the presence of ripples was observed. At temperatures below 1800°C, grain boundaries grooved the same as with thermal radiation. Grooving became uneven at higher beam powers and had the appearance of a series of connected pits as evidenced in the interference micrograph of Fig. 7.

Grain boundary grooving, which occurs in an even manner below 1800°C by both means of heating, is a well-known phenomenon

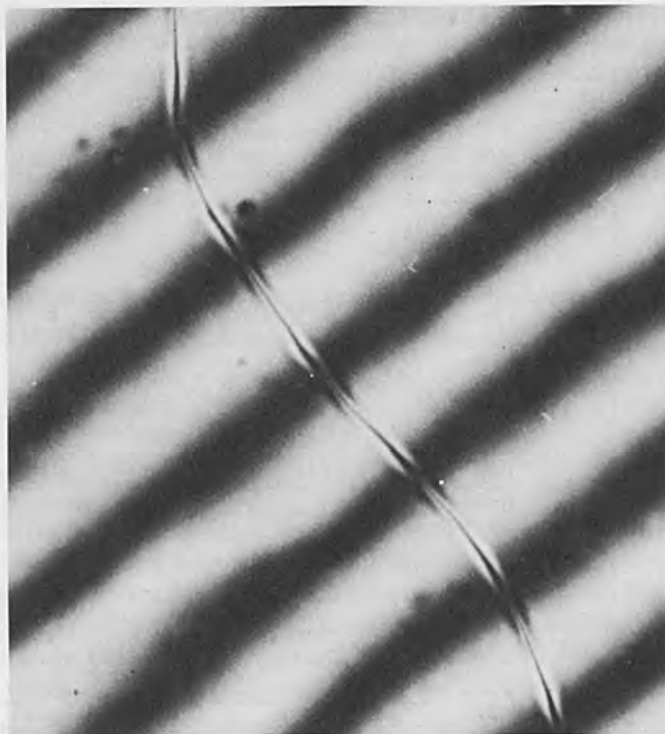


Fig. 4. Interference micrograph of an even grain boundary groove after heating to 2600°C for 90 minutes by thermal radiation. X650.

attributed to surface diffusion at high temperature [5]. The pits, developed at higher temperatures only, are probably due to the evaporation or expulsion of inclusions or impurities. But this pitting, in any event, is characteristic of the material at high temperature since it forms with thermal radiation. However, the appearance of some rippling and uneven grain boundary grooving will be seen to be characteristic of oscillating beams.

The most extensive damage to surfaces was caused by low-frequency oscillating beams, as shown in Fig. 8. Damage to grain boundaries developed in the following way. At low beam powers, and hence lower temperatures (1600°C), grooving was uneven in like manner to that shown in Fig. 7. These grooves at increased beam powers gradually developed into V-shaped cracks which became as deep as 0.010 in. This behavior was similar in grain

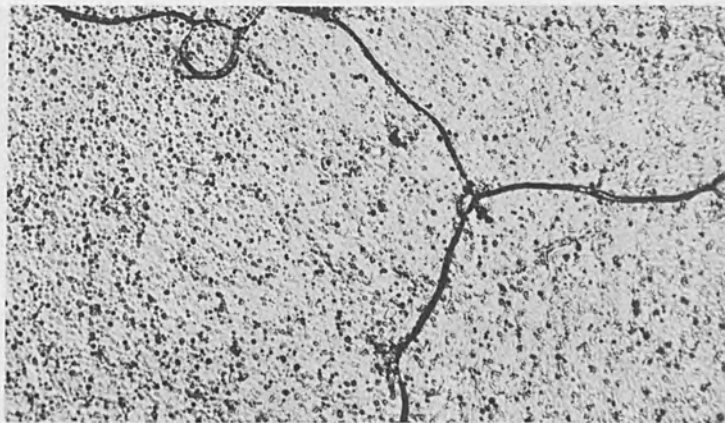


Fig. 5. Micrograph of pitted surface after heating to 3300°C by thermal radiation for 30 minutes. X100.

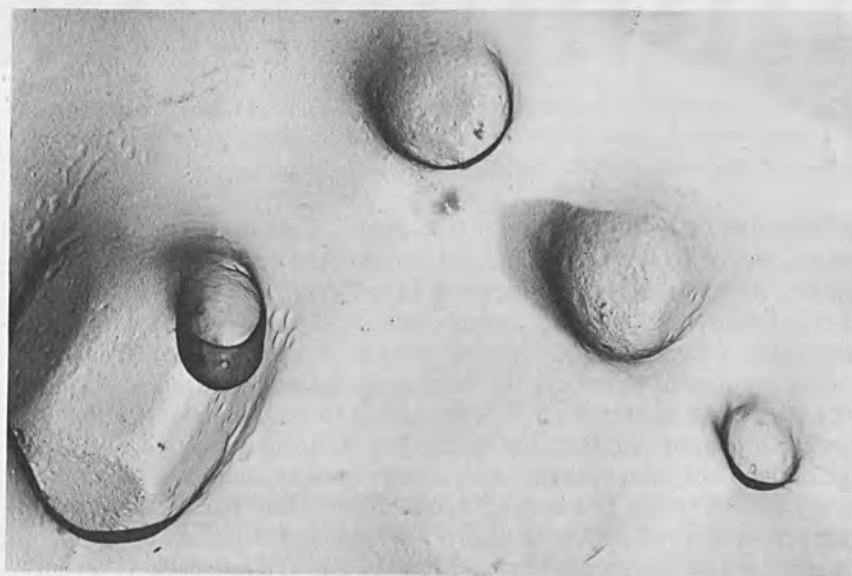


Fig. 6. Electron micrograph of pits formed on tungsten surface at 3300°C by thermal radiation for 30 minutes. X18,000.

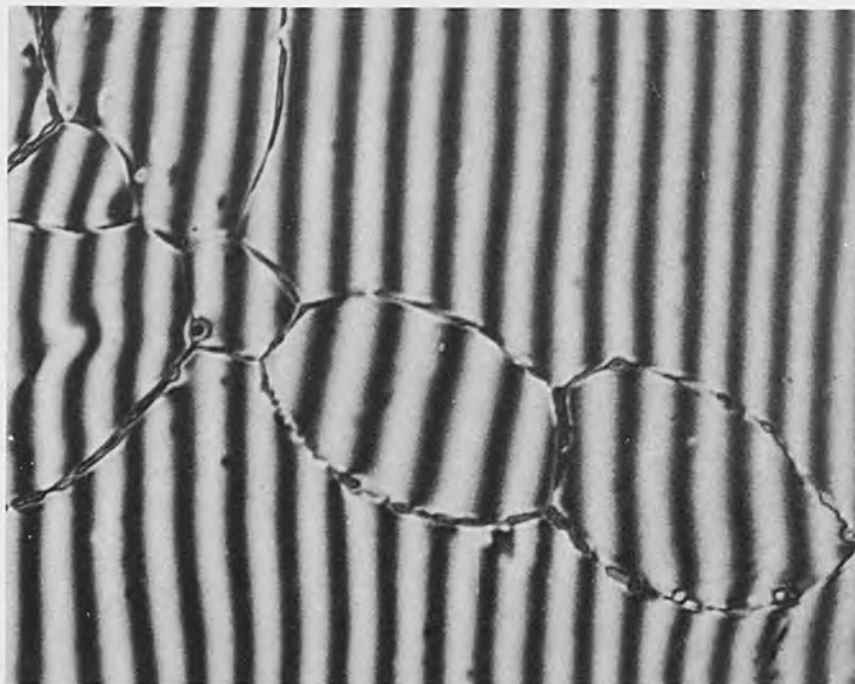


Fig. 7. Interference micrograph of tungsten surface heated by 60 cps oscillating beam at 2000°C for 30 minutes. Note uneven grain boundary grooves and tilted grains. X650.

boundaries of both coarse and fine grain sheet material. It was not as extensive in the grain boundaries of foils even at high temperatures. Damage to the surfaces of large grains in sheet specimens increased with increasing beam power and temperature at low frequencies. Oscillating low-power beams at low frequencies produced ripples to the surfaces of grains, as exemplified in Fig. 9. The direction of these ripples appeared to be dependent on the crystallographic orientation of the grain, although a precise correlation was not undertaken. Also, there was evidence of polygonization as seen in the breakup of spots in the Laue back-reflection pattern of Fig. 10. This breakup was induced when the rippling was caused at low-beam powers and low-sweep frequencies at a temperature of 1900°C.

The size of the grain itself showed an effect on the extent of damage that was observed on its surface. Shown in Fig. 9 is an example of a small grain that was unconsumed during recrystalli-

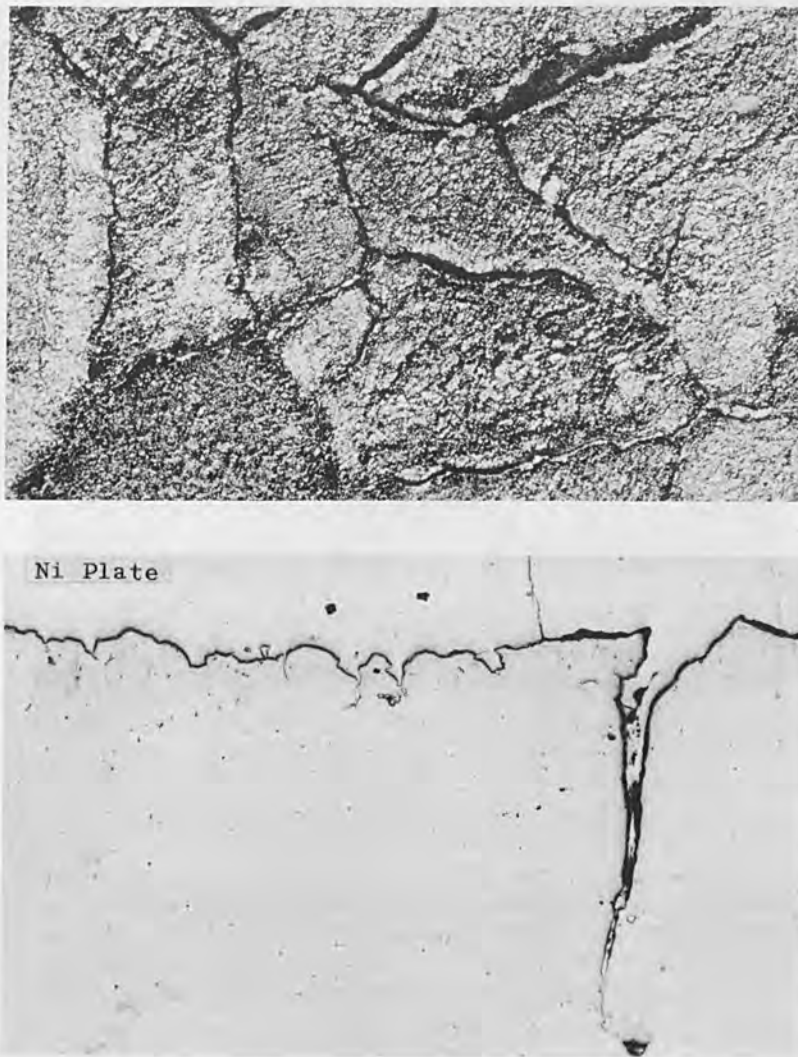


Fig. 8. Damage to tungsten heated to 3000°C for 30 minutes by an oscillating beam at 60 cps: (top) specimen surface (X16); and (bottom) tapered section, 2:1 (X100). Note deep "V" shaped cracks in grain boundary and shallow cracks in grain surfaces.

zation and subsequent grain growth in the coarse grain material (WG). A similar grain is shown in Fig. 11 taken with the interference microscope. In both figures it may be noted that the surface of the unconsumed grain is virtually undisturbed in comparison to

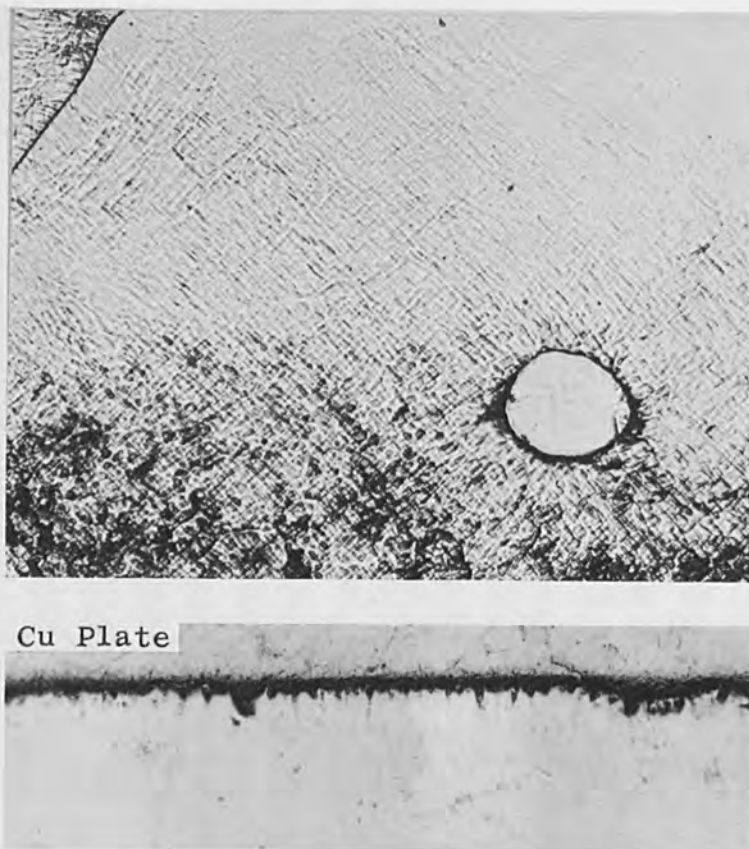


Fig. 9. Damage to tungsten heated to 2300°C for 60 minutes by a beam oscillating at 60 cps: (top) specimen surface (X200); and (bottom) tapered section, 10:1 (X200). Note in top micrograph the ripples on the large grain in contrast to the nearly undisturbed small grain.

the surface of the larger grain which surrounds it. Small grains also showed the tendency to be tilted. An example is shown by the interference micrograph in Fig. 6 where a grain is tilted $1/4$ degree from its original position.

It is likely that these forms of damage produced by oscillating beams are due to thermal cycling. Thermally induced cyclic stresses on surfaces may be accommodated in two ways. When the grain is large the stresses cause plastic deformation, as wit-

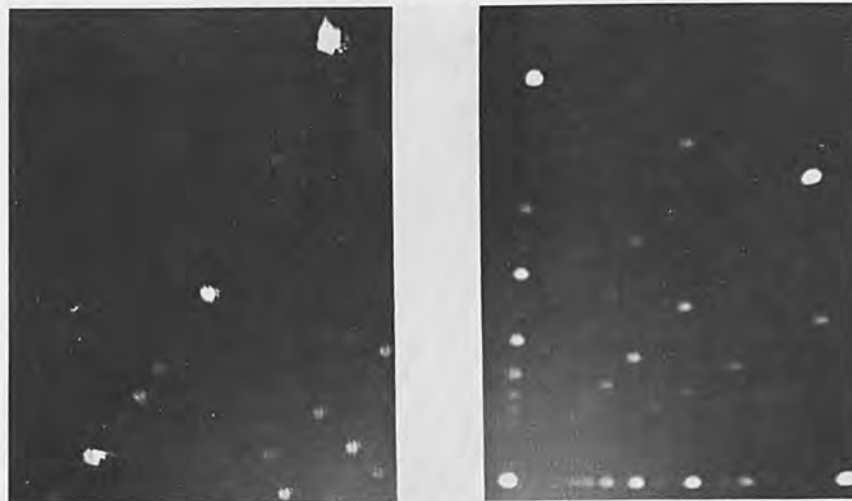


Fig. 10. Laue back reflection patterns of rippled single grain (left) and of as-recrystallized grain (right). Note breakup of spots due to polygonization produced at 1900°C by 20-cps oscillating beams in 3 minutes.

nessed by surface ripples showing X-ray evidence of polygonization. When the grain is small the grain boundary at sufficiently high temperature may accommodate the strain and cause the grain to tilt or even be dislodged without rippling. In such cases the grain boundaries do not provide the same constraint as material surrounding a point within a large grain, and ripples on the surface of the small grain are not so likely to occur. For similar reasons, heating foils by oscillating beams did not produce ripples on even its largest grain.

The formation of cracks in grain surfaces and boundaries, Fig. 8, also appears to be associated with the thermal cycling caused by oscillating beams. However, no means was available for producing thermal cycling on sheet specimens without direct electron bombardment.

Fine grain materials showed a combination of the effects produced in the coarse grain surfaces by oscillating beams, e.g., rippling, grain tilting, and uneven boundary grooving. In addition, small grains were seen to have been dislodged from the surface (Fig. 12). At temperatures very near the melting point, the metal appears to flow and produce mounds and valleys in the surface.

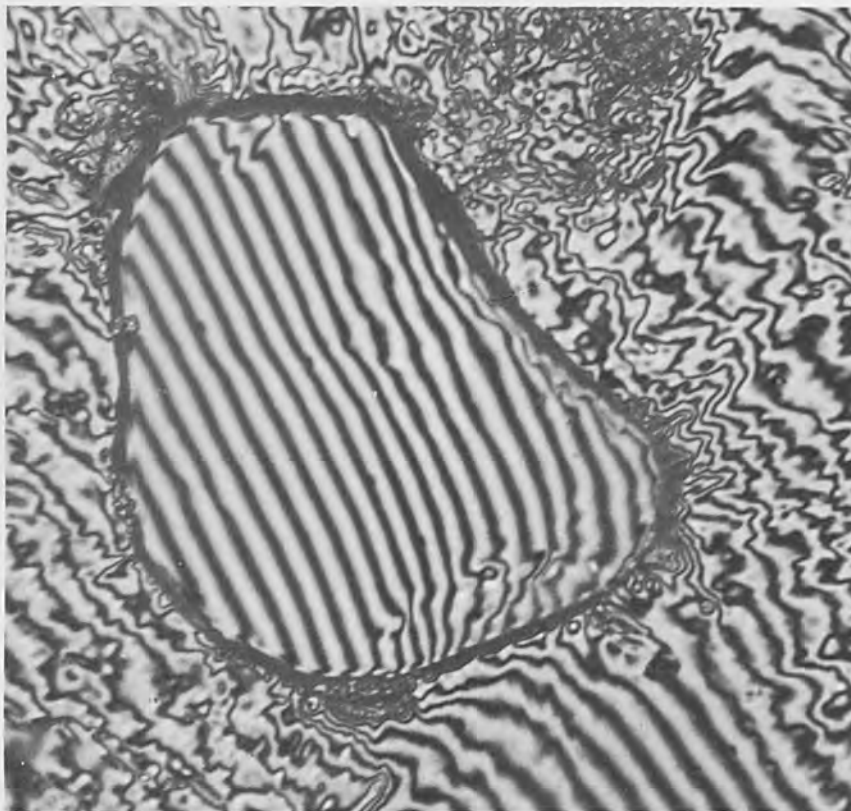


Fig. 11. Interference micrograph of an undisturbed small grain surface within a larger grain whose surface is rippled. X800.

Also at these temperatures some metal is extruded out of the surface at grain boundaries (Fig. 13).

In view of the effects produced by oscillating beams, the appearance of ripples and uneven grooves under stationary beams may be attributed to minor fluctuations in the beam itself. These fluctuations could be caused by the 4% ripple in the high voltage, as well as alternating currents in the filament and other beam-control circuits and may well induce thermal cycling similar to that induced by oscillating beams. With a steady beam it might be found that both thermal radiation and stationary beams would show very similar effects. These two means of heating however are not identical when the production of X-rays, secondary emission of electrons, etc., are considered.



Fig. 12. Surface of polycrystalline sample damaged by 60-cps oscillating beam at 2600°C for 30 minutes showing tilted and dislodged grains. X35.

Correlation of Surface Temperature and Damage

Measurements made with the interference microscope of grain boundary groove depths provided a means to indicate the temperature of surfaces bombarded by a stationary beam. A specimen heated to a mean temperature of 1800°C for 30 minutes showed groove depths of 700 to 900 Å in the area heated by the beam while those on the opposite side were less. To obtain comparable groove depths on the side opposite the beam a specimen had to be heated to a mean temperature of 2000°C . Thus a temperature difference of about 200°C was established between the two surfaces. The temperature difference indicated by the optical pyrometer was 100°C . The agreement although only approximate suggests that at least no unusually high temperature forms in the surface layer where the beam dissipates its energy. Above 1800°C in 30-minute groove depths could not be measured because of their unevenness.



Fig. 13. Severe damage to tungsten by 60-cps oscillating beam at 3000°C for 15 minutes showing metal extruded up adjacent to grain boundary. X20.

A similar, though less precise, relationship was observed on the bombarded area of a specimen heated to 3000°C. There pitting, as shown in Figs. 5 and 6, was comparable in extent and nature to that found in the area opposite the beam of a specimen heated to 3300°C. So that a difference in the order of 300°C was established between the bombarded side and the side opposite.

Cyclic Heating in the Absence of Direct Bombardment

The use of foils made it possible to determine the effect of thermal cycling in the absence of direct bombardment by observing the surface opposite the beam. When one side of a foil was bombarded with a beam oscillating at a low frequency (20 cps), it was possible to see a rapid cyclic change in brightness on the side opposite the

beam. Thus this side of the foil was being subjected to thermal cycling while not being bombarded. Grain boundaries on this side as well as the bombarded side were uneven. On heating, other foils to the same temperature between susceptors boundary grooving was even. This experiment clearly demonstrates the dependence of uneven grooving on thermal cycling. It is very likely that the ripples and cracks caused by oscillating beams are also due to thermal cycling.

Dependence of Damage on Temperature and the Magnitude of Thermal Pulse

It was found that the extent of damage is not only dependent on the magnitude of the thermal pulse, but on the temperature of the specimen as well. By varying the beam power density and sweep frequency (Table I), the thermal pulse, ΔT , according to the assumed relationship in equation 4, was held at nearly constant amplitude for the series of damage experiments at different temperatures shown in Fig. 14. The extent of roughening in specimen numbers 5 and 6 is seen to be quite similar although at the extreme temperatures the damage is reduced. The material is stronger at the lower temperature and can better accommodate the thermally induced stresses. At the highest frequency (specimen number 7) the extent of damage is also seen to be less. The relationship describing ΔT departs most from the more exactly calculated values [3] in the case of number 7 as well as numbers 1 and 2, so that the actual magnitude of thermal pulse may in fact be less than that calculated for this experiment.

The dependence of damage on the magnitude of thermal pulse is shown in Fig. 15. Here two specimens were heated to the same temperature, 2500°C, but the thermal pulse was different for both since they were damaged at sweep frequencies of 540 and 3000 cps. The difference in extent of the resulting damage yields a basis for establishing operating conditions where no damage of this type occurs. Decreasing either the beam-power density or the dwell time reduces the pulse magnitude and consequently the damage. Beam-power density may be decreased, still maintaining a given temperature, by broadening the beam-spot diameter or using several guns. The magnitude of the thermal pulse may also be reduced by increasing the sweep amplitude and frequency thus decreasing the dwell time. For a given temperature, the thicker the specimen the more power is required so that the thermal pulse is increased.

Damage associated with thermal cycling may be minimized, but

Table I
Operating Conditions for Heating with Constant Magnitude of Thermal Pulse

Specimen Number	Beam-† Power Density kW/cm ²	Sweep Frequency sec ⁻¹	Sweep Amplitude 2A cm	Calculated* Thermal Pulse at Midpoint °C	Calculated* Thermal Pulse at Ends °C	Mean Temperature °C
1	2	30	3.8	33	79	1520
2	4	120	3.8	33	79	1880
3	6	270	3.8	33	79	2040
4	8	480	3.8	33	79	2220
5	10	750	3.8	33	79	2360
6	12	1080	3.3	35	82	2450
7	14	1470	3.3	35	82	2540

*Calculations based on equations 1 through 4. Values at ends are calculated for the maximum dwell time at $x = A - L/2$.

†One beam was used with spot size of 0.5-cm diameter. All specimens of Fig. 14 exposed for 5×10^5 sweep cycles; thus 5×10^5 pulses at ends and 10^6 pulses at midpoint.

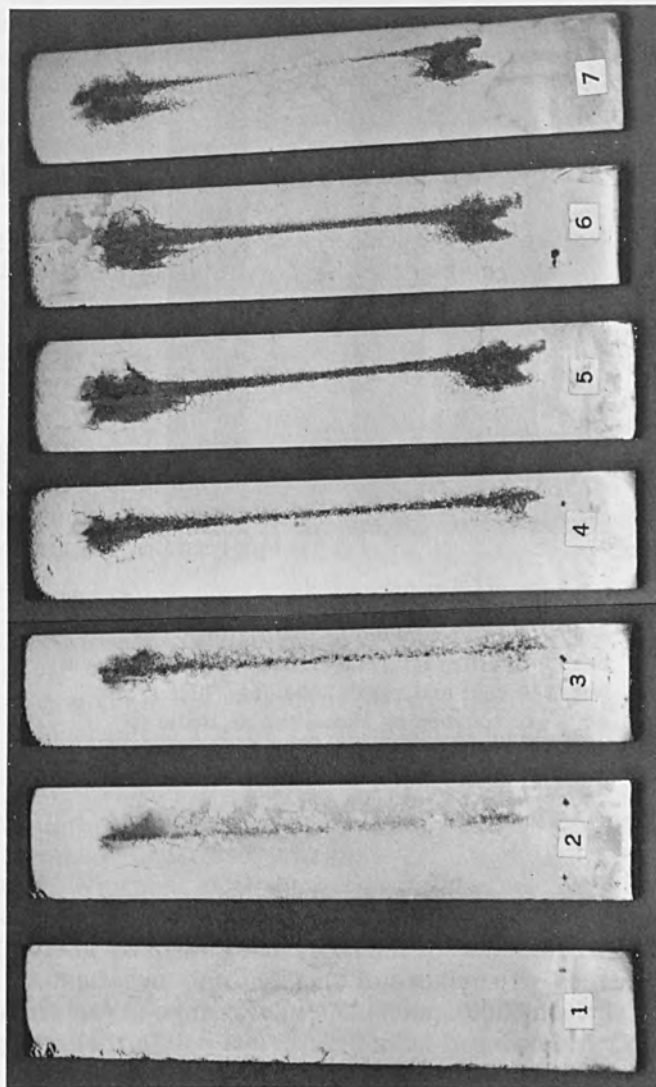


Fig. 14. Sheet specimens damaged at different temperatures but all subjected to the same thermal pulse. The extent of roughening indicates the dependence of damage on specimen temperature. The operating conditions which produced the damage are given in Table I. X1.5.

thermal grooving and pitting cannot be controlled since these effects are due to the mean temperature of the specimen alone.

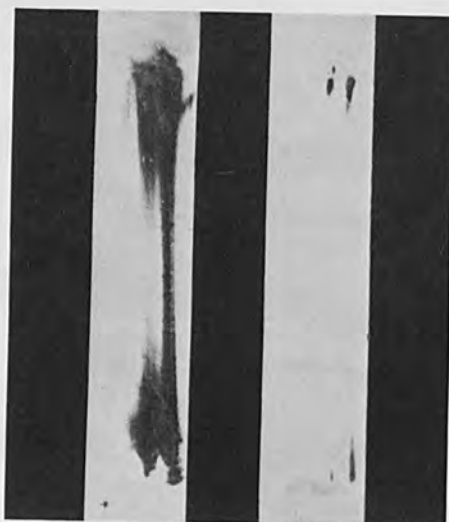


Fig. 15. Two specimens damaged at 2500° C for 30 minutes by oscillating beams at different sweep frequencies: left, 540 cps; and right, 3000 cps. The extent of roughening shows the dependence of damage on the magnitude of thermal pulse. X1.5.

SUMMARY

The nature of the effects (or damage) that a focused 10-kV electron beam produces on surfaces of tungsten was investigated. At temperatures between 1200° and 3400°C, the change in the surface was observed as it developed on both foils and polished sheet specimens. Electron bombardment produces damage to tungsten surfaces which appeared to be associated with the temperature to which the surface was subjected and with thermal cycling.

The types of damage produced by stationary and oscillating beams were found to be different. Stationary beams show charac-

teristic effects of grain boundary grooving and surface pitting similar to those manifested by thermal radiation heating.

On the other hand, oscillating beams produce damage of a different nature that has been observed to be associated with thermal cycling. As the oscillating beam sweeps past a given portion of the specimen, it induces a thermal pulse in the surface. The resulting damage includes ripples on grain surfaces, uneven grain boundary grooves, cracks, and tilted and dislodged grains. The extent of these effects is dependent on temperature as well as the magnitude of this pulse.

By increasing the frequency and amplitude of sweep and lowering the beam-power density, the magnitude of the thermal pulse and the damage associated with it can be minimized.

ACKNOWLEDGMENTS

The authors wish to express their thanks to Messrs. A. R. Donaldson, D. H. Price, and T. C. Lupton for assistance in the preparation of the photographs; to Mr. J. T. Atwell for invaluable aid with the experimental work; and to Dr. M. R. Achter for advice and review of the manuscript.

REFERENCES

1. "Development of an Electron Beam Heating Facility and its Use in Mechanical Testing of Tungsten to 6000°F," ASD-TDR-63-484, Lessells and Associates, Inc., May 1963.
2. P. W. Crapuchettes, "Cooling of Anodes Subjected to Long Pulses of High Peak Power," AIEE Conference Paper, Winter General Meeting, New York (Jan. 29 to Feb. 3, 1961).
3. G. E. Vibrans, "Calculation of the Surface Temperature of a Solid Under Electron Bombardment," Tech. Report No. 268, Lincoln Lab., Massachusetts Institute of Technology, Nov. 16, 1962.
4. H. Doering and P. Shahinian, "Brightness and Two-Color Pyrometry Applied to the Electron Beam Furnace," U. S. Naval Research Lab. Report No. 6062 (to be published).
5. B. Chalmers, R. King, and R. Shuttleworth, "The Thermal Etching of Silver," Proc. Roy. Soc. (London), A, 193, 465 (1948).

VARIABLE-GRADIENT, ELECTRON-BEAM HEATING METHODS FOR GROWING SINGLE CRYSTALS OF ZIRCONIUM

INTRODUCTION

Cylindrical single crystals of most refractory metals can easily be produced by floating zone-melting techniques. In contrast, only tedious and lengthy methods have been available for growing crystals of titanium, zirconium, and hafnium. These three metals are body-centered cubic in the high-temperature (β) phase (stable above 870°C in zirconium), and the low-temperature (α) phases are close-packed hexagonal with anisotropic physical and mechanical properties. All three metals are so chemically reactive that it is extremely difficult to prevent contamination at temperatures where crystal growth can be produced. As a result of these difficulties, only a few examples of the use of single crystals of these metals can be found in the literature.

The work described below was undertaken to provide single crystals of zirconium in a variety of forms (strip, foil, round bar) for an AEC-sponsored program of research. Using rather simple electron-beam heating methods, it was possible to grow crystals of more or less arbitrary length, at a lineal rate of about an inch per hour, by simply traversing a beta zone the length of a strip or bar, or by zone melting. Previous methods have required periods of hundreds of hours to grow crystals perhaps an inch long.

The success of the methods to be described apparently depends upon steep temperature gradients, which are easily produced by electron-beam heating; moreover, development of the methods was made possible by the unique ease with which electron beams

*Oak Ridge National Laboratory, Oak Ridge, Tenn. Operated for the U.S. Atomic Energy Commission by Union Carbide Corporation.

may be directed and controlled to optimize conditions for crystal growth in situ. Perhaps more important than the methods developed for growing zirconium crystals is the demonstration of the potentialities (presently unrealized) of electron-beam heating for producing and studying crystal growth in solid metals. The value of certain arbitrary temperature patterns has long been recognized for growing crystals in sheet and strip form, but the means for producing the desired patterns have been subject to many experimental limitations. With electron-beam heating it is possible to produce continuously variable temperature patterns and temperature gradients with simple apparatus and at high-power densities.

Rapperport [1] first produced large crystals of zirconium by simply annealing crystal bar (i.e., iodide, or van Arkel-de Boer process) zirconium just below the α - β transformation temperature for 100 hours. Langeron and Lehr [2] obtained crystals by an α - β cycling process. A cycle consists of heating into the beta range, cooling at a rate of 150°C/hr into the high alpha range, and holding in the alpha for 100 hours. The cycling is continued for weeks or months. Anderson [3] had earlier developed a similar technique for titanium. Disadvantages of these methods include the long time required; the uncertainty of results (yields of only a few per cent); the virtual impossibility of preventing contamination for long times at high temperatures; and, in the case of α - β cycling, the shape of the specimen may become badly distorted. Some of the difficulties are illustrated by Anderson's experience with titanium: the oxygen content increased by factors of 3 to 10 in sealed vacuum capsules; whereas, filling the capsules with 1/5 atmosphere of argon suppressed growth entirely!

Strain-anneal methods will work if deformation twinning can be controlled; otherwise, twin intersections act as growth nuclei. Churchman [4] used compressive straining (on the faces of a rolled strip) to grow crystals of titanium. Bokros [5] was able to produce limited exaggerated grain growth in sponge zirconium after tensile straining. The small grain size and the lower purity of the sponge zirconium probably minimized the influence of twinning.

Kneip and Betterton [6] and Langeron et al. [7] carried out floating-zone refining of zirconium. Rather surprisingly (in view of our experience here) the growth of large crystals was not reported. Induction heating was used, and the bars were irregular; therefore, large grains may have been produced but were obscured by the irregular surface.

APPARATUS AND MATERIALS

Experiments were conducted in a 4-in. Pyrex pipe attached to an unbaked, baffled, and liquid-nitrogen trapped, oil-diffusion pump system with a speed of 80 liters/sec at the pipe. The system blanked off below 1×10^{-8} torr, and experiments were not begun until the pressure dropped below 5×10^{-8} torr. Maximum pressures occasionally rose as high as 5×10^{-5} torr when a specimen was evolving hydrogen during initial heating. The system pressure during the first traverse of a specimen averaged 5×10^{-7} torr (again due to hydrogen). During subsequent passes, the pressure dropped by a factor of 10 to 20. Means were provided for traversing the specimens past the heat source at constant rates of travel.

Figure 1 shows the dimensions of a typical electron gun used for the solid-state experiments, and the relation of the gun to a strip specimen. The beam strikes the specimen on an area roughly 15-mm wide and 1-mm high. Masks could be fitted in front of the gun to change the beam shape; further adjustments could be made by biasing the body of the gun negative with respect to the cathode. Normally two identical guns were used, one facing each side of the strip. If variable heating patterns were desired, the guns, for example, could be masked so each heated only half the width of the strip. With equal power in the guns, line heating (as in Fig. 1) resulted. The lateral distribution of heat could then be shifted by changing the relative power in the two guns by control of the respective cathode heater currents. The power for these experiments was supplied by four or six 6-kV, 18-mA neon-sign transformers in series parallel. The unfiltered dc output, through a bridge rectifier, was 5 kV at 20 mA. The transformers are current-limited, and the regulation is of course poor. These characteristics were desirable to protect the apparatus and rectifiers in case of short circuits; and, under the conditions of the experiments, the power was self-regulating for small changes in cathode emission.

For zone-melting and refining the ring (also called annular) gun in Fig. 2 was used. Although designed for 6-mm bars, the inside diameter was 25 mm in order to accommodate kinking or misalignment of the specimens. The beam at the specimen was narrowed to less than 1 mm to impart maximum superheat. The same gun has since been used with specimens up to 15-mm diameter. A commercial, 10 kV, 100 mA power supply with 15% regulation was used. The beam current was held constant by a servo-controlled Variac supplying the cathode heating transformer. Automatic con-

trol of the beam current is a necessity when melting zirconium (even if the melting is superficial or localized), because the emission of the tungsten cathode increases when zirconium is being deposited on it.

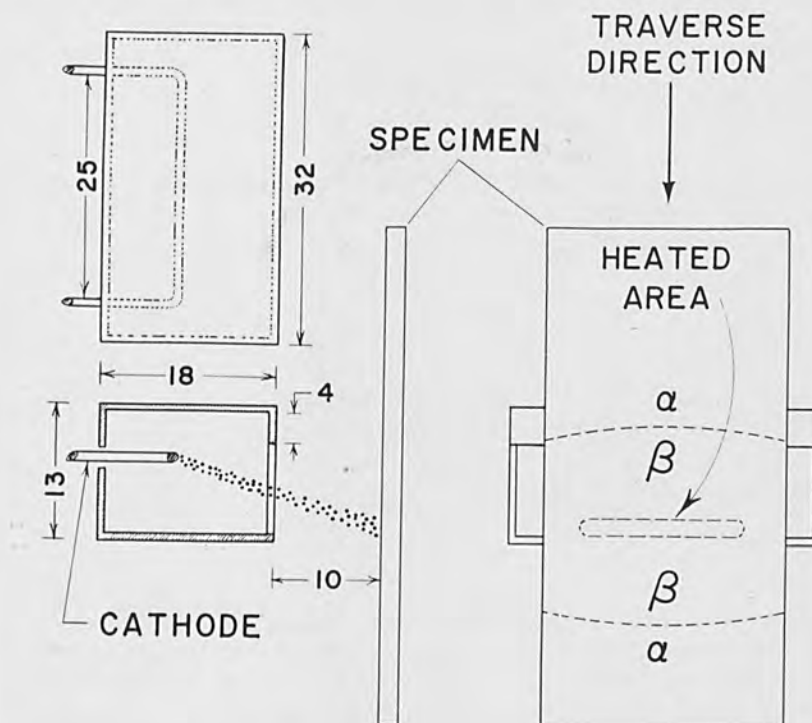


Fig. 1. Electron gun for producing line heating. The gun is shown in relation to a typical 1- by 25-mm strip. The shape and size of the beta zone for a maximum temperature of 1200°C are indicated on the strip. Dimensions in millimeters.

Crystal-bar zirconium meeting reactor grade I specifications was used for all experiments. Material was swaged and rolled directly from carefully conditioned crystal bar or arc-melted and rolled. Surface defects were removed by grinding and chemical polishing when necessary. Specimens were used in the cold-worked state (70 to 99% reduction in area).

Routine analyses of the six materials used are summarized as follows: B, Be, Cd, Pb, less than 2 ppm; Al, Cr, Mn, Sn, less than

20 ppm; and Ca, Co, Cu, Mg, Mo, Ni, Si, V, Zn, less than 100 ppm. Iron and hafnium ranged from 50 to 200 ppm each, and oxygen and carbon were between 5 and 100 ppm each. Hydrogen and nitrogen were below 1 ppm each after heating in vacuum.

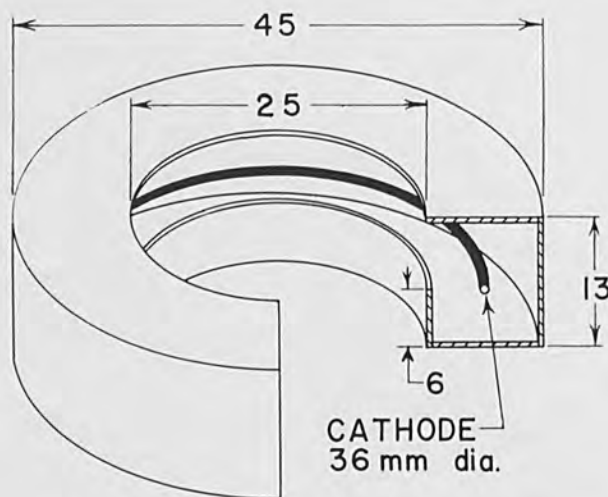


Fig. 2. Ring electron gun for zone melting. Dimensions in millimeters.

One crystal bar of hexagonal cross section (22 mm across flats) with exceptionally well-formed crystal faces was used for most of the solid-state growth experiments. Large crystals could be grown more easily in this material, designated bar A, at lower temperatures and at faster rates than any other stock. The second best material in which crystal growth could be produced was bar B, made from arc-melted crystal bar. The major differences in composition for bars A and B, respectively, are (in parts per million): 26 and 110 oxygen, 116 and 55 iron, 1 and 10 tungsten, 1.4 and 7 nickel, and 10 and 50 silicon.

SOLID-STATE GROWTH

In principle, large grains of zirconium are obtained by traversing a zone, heated into the beta range, along the length of a bar or strip. There are three arbitrary steps in the process: (1) production of a small number (preferably one) of beta grains across the

width of the strip; (2) traversing the zone to obtain lengthwise growth or elongation of the grains so formed by consuming the matrix of small alpha grains (produced by recrystallization ahead of the beta zone) at the advancing α - β interface; and (3) the transformation of each large beta grain into one alpha grain at the receding α - β interface behind the zone.

In operation, a static beta zone is first formed by heating to some temperature in the beta range and holding a few minutes to an hour to allow some grain growth to occur. The zone is then moved at a constant rate until near the end of the strip. The power is slowly reduced, and as the temperature falls the beta zone narrows until the zone is all in the alpha phase. Typical conditions for producing growth in a 1 by 25 mm strip are: maximum temperature 1200°C; beta zone height, 22 mm (see Fig. 1); power, 80 watts; beam area, on one side of strip only, 1/2 by 15 mm; traverse rate, 4 cm/hr after holding 15 minutes at temperature at the start.

Figure 3 shows results of runs on three different materials under the above conditions. In crystal bar A (Fig. 3a) large beta grains grew from the highest-temperature region down to the α - β interface in a few minutes. When the traverse began these same grains grew with the advancing α - β interface. After perhaps 5 cm of travel only about three grains occupied the width of the specimen, and the boundaries of these grains extended in the direction of travel from advancing to the receding α - β interfaces.

Nucleation of alpha grains started after the zone had moved far enough for the end of the strip to cool below 870°C. As the power was reduced at the end of the traverse, the alpha grains above the zone continued to grow as before when the beta zone contracted. Below the beta zone new alpha grains were nucleated, or grew from existing alphas, in the opposite direction.

Bar B (Fig. 3b) obviously behaved in a different manner under the same conditions. In the original (static) beta zone, equiaxed grains up to about 3 mm were produced. A few grains became elongated in the steeper gradient below the beam, and some became elongated during the traverse. However, the very long grains produced were mostly nucleated below the original beta zone. After 4 cm of traverse the situation in bar B was exactly the same as in bar A, except that the beta grains were much narrower. In the last position of the zone, it can be seen that the beta boundaries extended the full length of the zone.

The upper portion of bar C (Fig. 3c) is composed of roughly equiaxed alpha grains transformed from betas of the same size.

There was little elongation of these grains. At the point indicated, the travel was stopped, the power turned off and later raised to a higher temperature (1300°C, 100°C higher than before) and traversed at 1/8 the previous rate. Elongated beta grains were formed as in bars A and B.

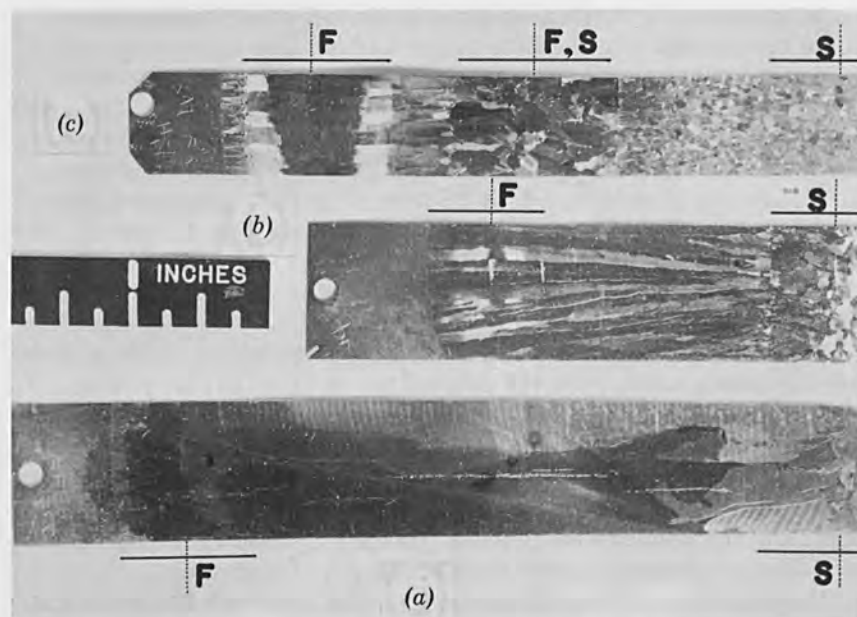


Fig. 3. Solid-state crystal growth in three lots of 1- by 25-mm zirconium strip (see text for conditions). On the photograph, *S* indicates extent of beta zone at start, *F* the extent of final beta zone, and the dotted line shows beam location in zones. Unetched. The long prominent white lines are prior beta boundaries made visible by offsetting (normal to strip face) of adjacent grains. Differences in shading are the result of thermal (or thermochemical) etching in the beta or transformation roughening, or both.

Thus, markedly different results were produced in three lots of zirconium of, by today's standards, respectably high purity. However, the last two steps in the process, the elongation of beta grains in the direction of growth to the full length of the beta zone, and the tendency for each beta grain to transform to a single alpha, are apparently the same.

The problem, then, is to produce a few wide grains as soon as possible. In bar A, grains of large size were produced in a few

minutes in a steep temperature gradient. In bar B the larger grains produced at the start did not grow; however there was apparently a competitive growth process that slowly reduced the number of grains across the width. In bar C grains of width equal to the beta grains were produced and these grew with little tendency to widen. In principle the growth process, after the first large growable grains are produced, is almost isothermal. The important processes occur at α - β transformation temperature at the boundaries of the beta zones. The high temperature at the center of the zone is needed only to get the large initial grains.

The examples in Fig. 3 were chosen to illustrate the differences in behavior in the materials, and they were not grown under optimum conditions. In bars B and C grains up to 4-cm long can be grown in a strip up to 1-1/2 by 13 mm. Higher temperatures and slower traverse rates improve the yield. Most of the following remarks will concern crystal bar A, which was most studied but which, unfortunately, does not appear to be representative.

In early attempts to improve the yield, some effort was made to secure larger grains (or perhaps more favorably oriented ones) by controlling the direction of the temperature gradients. When a beta zone is first formed there is a strong tendency for the boundaries of the large beta grains to be elongated in the direction of the local, maximum temperature gradient. Figure 4a shows three static beta zones. One was produced by a line beam as used for the specimens in Fig. 3. The other two were produced by off-center spot beams (approx. 2-mm square) at two different power levels. The tendency of the beta grains to elongate down the temperature gradient is easily seen.

By varying the lateral temperature gradients (while watching the results) it was possible to influence the direction of growth and grain size in the first formed beta zone, and to obtain larger grains for subsequent growth by traversing. The operation was tedious, but with low temperatures (limited by the original power supply) improved yields were obtained. It was also found that the path of the long beta boundaries could be altered when growth was well underway. Using the line beam and an off-center spot beam (of constant total power) on opposite sides of the specimen, the heating pattern was periodically and smoothly varied by adjusting the relative power of the two beams. The results are shown in Fig. 4b. Note particularly the boundaries (see arrows in Fig. 4b) between adjacent beta grains that have been "steered" to describe a sinuous path. In the light of later developments, the need for the steering was reduced, and attempts to utilize steering in other

materials (where the steering was more needed) showed very little effect.

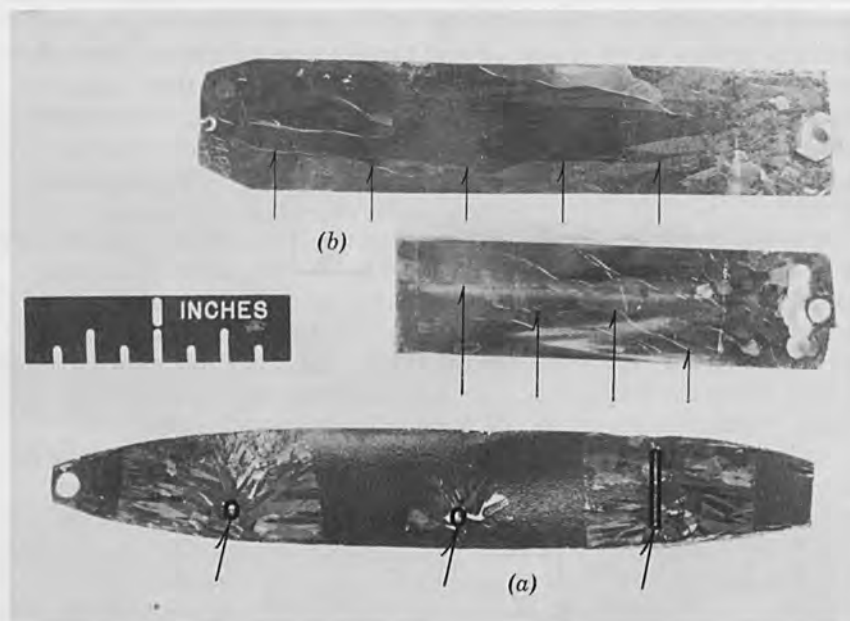


Fig. 4. (a) Effect of heating pattern and direction of temperature gradients in static beta zones. Shape and size of beam indicated by areas adjacent to arrows. Chemical polish—etch. (b) Arrows point to beta boundaries whose direction of growth was periodically changed by varying lateral temperature distribution with one line beam and one spot beam on opposite faces of specimen. Other boundaries also show some winding. Unetched.

The value of variable gradients was demonstrated in an attempt to grow crystals in thicker material. As a rule, reducing the surface to volume ratio of a specimen (which of course reduces the temperature gradients) made it more difficult to grow crystals, even in bar A. In 3 by 13 mm strip of bar A, poor yields resulted with line heating. Several inches of traverse were required for competitive growth to reduce the number of grains across the width. By varying the heating patterns it was found that heating only at opposite edges of a strip caused one, or at most two, beta grains to be formed immediately across the width of the strip, Fig. 5b and c. Although the first-formed beta grain never grew, subsequently formed grains were equally large so that longitudinal

growth began early. The yield of crystals in 15-cm-long strip was increased by a factor of four.

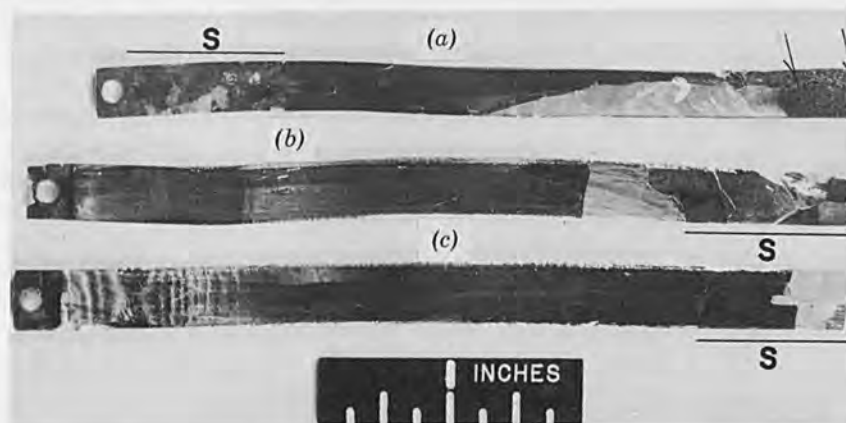


Fig. 5. (a) Large grains of zircaloy-2 in 2- by 12-mm strip. In last zone (arrows) the white line shows the location of the single former beta boundary in the zone. (b) and (c) zirconium strips, 3 by 12 mm, in which edge heating was used to produce transverse beta boundaries in starting zone (indicated by S). Distortion of strip, typical of thicker stock, is evident.

Figure 6 shows that improved yield in this (1 mm thick) strip could be obtained by starting the zone in a narrowed section. As the grains elongate, they widen and give a lateral component to the boundaries which causes a number of them to grow to the edges. This method was developed from observation of the effects of heating initially with a centered spot and then widening the spot during the traverse.

The value of nonuniform heating was further demonstrated when long crystals could not be produced in 6-mm-diameter rods of bar A heated uniformly around the periphery with a line beam. When the same amount of power was applied to a 2-mm-square spot, crystals up to 5-cm long were easily produced.

Large crystals of Zircaloy-2 can be grown in strips up to 2 by 13 mm, Fig. 5a, at rates up to 6 mm/hr if the starting material is water quenched from 1000°C. Although large beta grains are produced in steep temperature gradients, they do not appear to elongate in the stable manner observed in zirconium, and it is difficult to produce grains over 3-cm long. Since nucleation of undesirable grains occurs mainly at the edges, it is probable that improved

results would be obtained by changing the heating pattern from the line heating employed. The number of grains across the width can often be reduced by stopping the traverse, cooling for a few minutes and reheating to the original temperature. Often one of the grains will grow to the full width of the strip. Distortion of the strip is much greater than in zirconium. Precipitation, probably of the complex Zr-Sn-X phase, occurs on parallel planes (not yet indexed) making an angle of generally less than 20° with the growth direction. Some loss of alloying elements occurred; deposits formed by evaporation from the specimen analyzed 40% each of iron and chromium, 10% tin, and the remainder zirconium, nickel, manganese, and aluminum. The losses might be reduced by using a beam of lower power density.

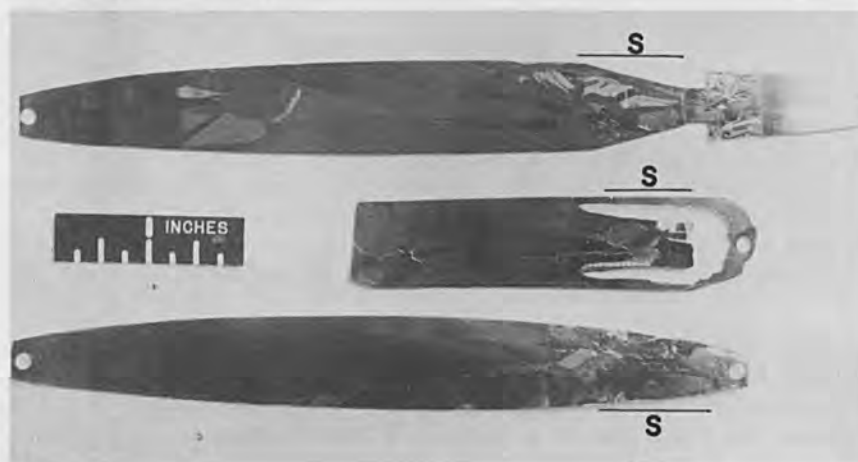


Fig. 6. Typical grain growth in 1- by 25-mm strip zirconium. Starting zone indicated by S is in narrowed portion of strip in order to hasten lateral growth of excess boundaries to edge of strip.

The zirconium and Zircaloy-2 crystals produced by the above methods generally show streaked or diffuse Laue spots. Alpha-annealing for periods of 30 hours usually results in sharp spots. In general, little change in boundary positions takes place during these anneals, but occasionally a small grain will grow larger. The maximum of boundary motion observed is over 1 mm/hr.

Columnar structures, such as that produced in Fig. 3b, are very stable in the alpha and low-beta temperature ranges. Introduction of nuclei by rapid heating (or cold work) has been observed

to cause grain growth at rates of the order of 1 mm/hr into this structure.

ZONE MELTING AND ZONE REFINING

Electron-beam, floating-zone refining has been used primarily for purification purposes, but large crystals of zirconium have been produced concurrently. Cylindrical bars from 4 to 14 mm in diameter have been zoned without difficulty using the gun shown in Fig. 2. The molten zone was stable and the large variations in specimen diameter experienced by others [6], [7] (using induction heating) did not occur with power levels controlled to only $\pm 5\%$. The zones were stable for either direction of travel: Langeron and Lehr [2] had reported that the zone was unstable when traversing upwards. The thermal conductivity of zirconium is low, and the beam must therefore heat the specimen evenly about its periphery to prevent off-center unfused regions. Because of the anisotropic thermal expansion of alpha zirconium, a kink may form at the trailing α - β interface where a few large grains with unfavorable orientation relationships are transforming. Kinks on the specimen axis as large as 5 degrees have been observed. In contrast to the solid-state growth experiments, the yield of crystals is about the same in all materials. Nucleation of the beta, and often the alpha, grains that grow the length of the bar frequently occurs in the unmelted starting section adjacent to the first zone.

The yield of single crystals from one or two passes on a round bar ranges from 20 to 100% at a 5 cm/hr travel rate. In the hot zone it is extremely difficult to see grain boundaries, but they often become visible at temperatures below 1000°C or in the alpha region. When it is apparent that several grains are growing, the number of grains may sometimes be reduced by rapidly traversing to the start, holding a few minutes, and restarting the traverse.

The degree of purification can be judged from the following data for a 7-mm-diameter bar, 20-cm long, that was given four passes at 3 cm/hr. Except for hafnium, which had decreased from 95 to 80 ppm, all 18 metals sought in analysis were below 10 ppm, or a detection limit of 20 ppm near the head of the bar. Carbon increased from 20 to 60 ppm with little variation from head to tail. Oxygen increased from an initial value of 5 ppm to 32 ppm in the head and 10 ppm in the tail. Nitrogen and hydrogen were below 1 ppm. Resistivity ratios ($\rho_{273^\circ\text{K}}/\rho_{4.2^\circ\text{K}}$) as high as 450 have been measured near the tail of a similar bar after two passes; the high-

est value previously reported for zirconium was 280 [8]. Apparently solutes that raise the melting point ($k > 1$) of zirconium are mainly responsible for the relatively low resistivity ratios measured in the past, because the highest ratios are measured at the tail. The carbon pickup probably comes from the pump oil or its pyrolytic decomposition products.

Alpha annealing, as with the crystals produced in the solid state (see above), is usually necessary to obtain sharp Laue spots. The basal plane of the crystals produced tends to be parallel to the rod axis.

The large gun has allowed as-deposited crystal bar to be melted and zone refined directly, or conditioned for mechanical working by zone melting part (or all) the way through the bar to get a smooth, defect-free surface. Even a well-formed crystal bar has voids or unbonded areas (between adjacent prior beta grains) that crack upon working. Elimination of the defects by machining is costly in material and time. Figure 7b shows a bar of titanium, with typical, spiny, as-deposited crystals. After two passes (left end of the bar) it became as smooth as if originally machined. Even the center section, which received only one pass, is respectably uniform in diameter. The corners of the originally square, as-deposited zirconium bar, Fig. 7a, were filed off to remove pits and the bar zoned directly. The bar, shown after ten passes, demonstrates the extent of misalignment that can be tolerated in the large ring gun. The offset, at the tail, occurred in the seventh pass.

DISCUSSION—SOLID-STATE GROWTH

The growth process has been divided into three, more or less, arbitrary steps. The last, and perhaps easiest to understand, is the transformation of each large beta grain to, in general, one alpha grain. The cooling rates used (at the receding α - β boundary) are so slow that supercooling of the beta is negligible; because of the steep temperature gradients the volume in which nucleation of alpha can occur is small. The rate of nucleation of new alpha grains should therefore be very low. The thermal-conductivity and thermal-expansion anisotropy of adjacent alpha grains of differing orientations could upset conditions at the α - β interface, but with a very large grain such disturbances are minimized. It is also possible that growth structure in the beta, or defects created by transformation stresses at the interface, affect the continuity or stability of the transformation.

That crystals long in one dimension can be produced is mainly

due to the tendency for large beta grains to grow unchanged into fine-grained alpha matrix. One cause of this unique phenomenon may be the effect of beta-stabilizing elements of low solubility in

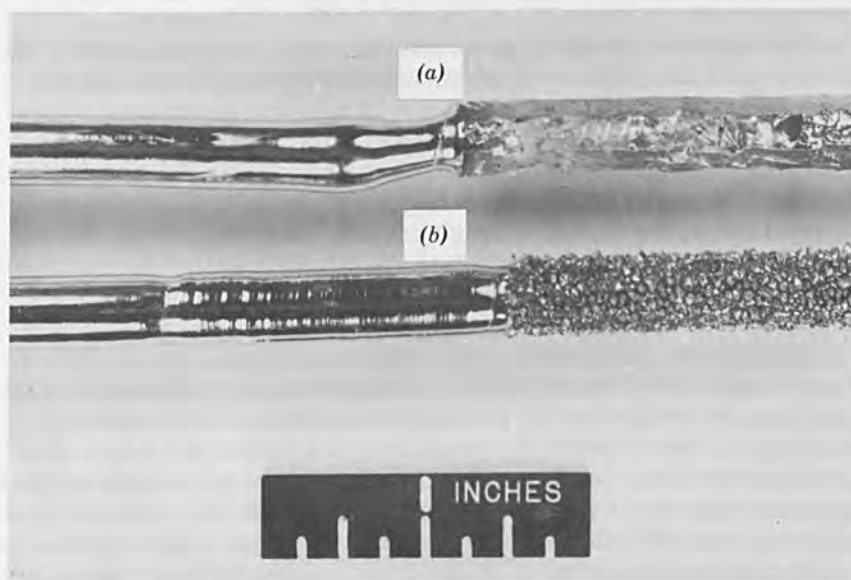


Fig. 7. Tail of as-deposited zirconium and titanium crystal bar after floating zone refining. (a) As-deposited bar of zirconium (square cross section) had corners filed off to reduce porosity. Bar shown after ten passes. Offset at tail (developed early in 7th pass) shows ability of large diameter gun to accommodate misalignment. (b) Titanium bar. Center of bar shows finish after single pass; section on left received two passes.

alpha zirconium (such as iron), on the growth interface. The alpha grains that recrystallize ahead of the beta region are small (0.1 to 0.5 mm) and stable for the periods and temperatures involved. The alpha plus beta field in the zirconium used is 10 to 30°C wide. Furthermore, iron, the major insoluble (in α) impurity, is commonly segregated at, or precipitated on, the grain boundaries. Therefore, the "alpha-beta interface" is not a plane but a volume several (alpha) grain diameters deep. Because of the segregation of the iron, the advancing beta front will not be flat since the grain boundary regions are iron rich. Because iron lowers the α - β transformation temperature, the beta front will extend farther into the alpha in the grain boundary regions. Therefore, each alpha

grain will, in effect, be isolated from its neighbors by a sheath of beta (which constitutes part of the advancing beta grain) before it reaches its own bulk transformation temperature. In the unlikely event that the grain transforms to a different orientation than the beta grain, its size and energy make survival unlikely.

The maximum speed at which crystals can be grown has not yet been determined; but, once very large beta grains are produced, they will grow at the rate of at least 15 cm/hr. Alpha grains may transform from beta at 150 cm/hr and still appear as single crystals under the microscope. Under these conditions the Laue patterns are unreadable, but annealing (in the alpha region) gives sharp Laue spots over the entire crystal. Macroscopic distortion is much increased at high rates of growth; this suggests that high strains are produced at the growth rates usually employed, but they have time to relax or anneal out.

At this time little can be said about the processes responsible for growth of the large, original grains, their dependence on composition, steering of these grains, or steering in the traversing beta phase. Because the temperature gradients utilized here are much steeper than have been employed in other solid-state crystal growing methods, it is possible for a number of less common phenomena, that depend on steep gradients, to affect or be responsible for the growth observed. For instance, solid-state (α - β) zone refining [9] of iron may affect the distribution of impurities, and temperature-gradient zone melting [9] (of iron, for instance, in both eutectic and eutectoid regions) may cause impurity transport or interface movement, or both. In the temperature gradients used, a driving force for boundary migration of the order of 200 dyn/cm² could be developed according to Luecke and Stuewe [10]. Although this driving force is probably at least 100 times less than forces from grain boundary or surface energies, it is high compared to temperature gradient forces in other crystal growing experiments. Further, impurity distribution is likely to be affected by volatilization in the beam, the concentration gradients produced thereby, as well as by grain boundary diffusion in the unusual configuration of grain and phase boundaries that develop during growth. Explaining the growth appears to be much more difficult than producing it.

Even in thin specimens we have noticed that the appearance is different on opposite sides of a specimen where only one side has been exposed to the beam. Numerous very shallow surface grains (much less than 1 mm in size) occasionally form on the back of very large crystals when only one face has been exposed to the beam. This is one reason that line heating gives better results

with heat on both faces. Medoff and Miller [11] observed increased growth rates in critically strained silicon iron where the grain growth was taking place in the beam. They suggest that the action of the beam is a disordering process akin to radiation damage, and that the energy for faster growth is from this source. We believe that the thermal stresses and some of the considerations discussed in the paragraphs above may be at least equally important. In any case, it appears that crystal growth in the electron beam may depend on more subtle factors than steep temperature gradients alone.

DISCUSSION—ZONE MELTING AND ZONE REFINING

The growth of single crystals by zone melting is not surprising in view of the solid-state growth. The difficult business of getting large grains is eliminated by melting the middle of the beta zone. It appears likely that seeding of crystals to obtain specific orientations is possible in spite of the phase transformations. The grains that grow (both alpha and beta) in the zone-melting experiments are frequently found to have originated several centimeters back in solid-starting material that is never melted. Since it is well known that a large alpha grain can be heated carefully into the beta and back into the alpha with the same orientation [12], it is not unreasonable to expect that a carefully prepared alpha seed of suitable length (so that one end is always alpha) could be welded to the start of the rod to be zoned in the same manner as metals without allotropic transformation.

From the limited zone-refining results it appears feasible to reduce all metallic impurities (except hafnium) below 10 ppm with little difficulty. Since oxygen can be moved by zoning (and may also be removed to an appreciable extent by volatilizations [13], it should be possible to reach values below 10 ppm with a baked vacuum system. Since carbon does not transport appreciably by zoning (it is possible that the contamination level obscured zoning) and is not likely to be removed by reaction with oxygen, great care in selection and vacuum techniques will be required to achieve low carbon levels.

DISCUSSION—ELECTRON-BEAM HEATING TECHNIQUES

The development of the solid-state, crystal-growing processes was made possible by the ease with which arbitrary heating pat-

terns could be produced with simple, if not downright crude, electron guns and limited power. All the solid-state experiments described here were done with less than 120 watts of power, the electrical components cost less than \$400, and the guns were made (or could be drastically modified) with hand tools in a few hours. Electron-beam heating on this scale at least, is efficient, economical, and immensely versatile for growing crystals and studying crystal growth.

For zone melting the advantages of using ring guns with inner diameters several times the specimen diameter should be considered. The specimen and molten zone are visible from all positions; specimen kinking, bulges from zone instability or overheating, and misaligned specimens cause no difficulties; less specimen material is evaporated onto the gun; and specimen stock too large for zone melting can be consolidated for mechanical working by surface zone melting. The same gun can be used for solid-state growth of strip. Four, 13-mm-wide, strips arranged in a hollow square can be accommodated in the gun in Fig. 2. Essentially line heating is produced on each strip. For edge heating the aperture of the gun can be masked to provide two, opposed, spot beams. The gun can also be tilted (with respect to the bar axis) to produce a canted liquid-solid interface, as described by Aust and Chalmers [14], to reduce or eliminate striation in melt-grown crystals.

The advantages of local superheat from using a narrow electron beam in zone refining cannot yet be proved. From the fact that iron and nickel analyses from the tail of our bars showed little increase over the initial concentration (and zone purification did take place in the rest of the bar), we deduce that much more of these elements was volatilized in our case than in similar experiments by others [6], [7] who used induction heating and showed large concentrations of these impurities at the tail.

The zone shape appears to be slightly different with a narrow beam; the centerline of the molten zone appears to lie below the beam. Increased convection stirring, at least at the upper liquid-solid interface, should result from the superheat in the narrow beam. Whatever the mechanism, vigorous stirring of the molten zone is observed. If the stirring results from the considerable force resulting from impact of the beam, then a narrow beam should provide more efficient stirring. The force from the beam is probably due to recoil from evaporating atoms [15]. The force has not been measured, but it is sufficient to impart rather brisk motion to freely suspended strips weighing several grams at power densities and temperatures much lower than used in zone melting.

CONCLUSIONS

Large single crystals of alpha zirconium have been produced in the solid state by traversing a beta-phase zone along a specimen at rates of the order of 1 cm/hr. Temperature gradients of at least several hundred degrees (C) per cm are required in, and adjacent to, the zone.

The initial step in the grain-growth process, the production of a few large beta grains to nucleate longitudinal growth during the traverse, is the most difficult and variable. The results depend on the material, temperature, and time, and sometimes on the direction of the temperature gradients.

The other two steps in the growth process do not depend upon the material and both take place at α - β interfaces (about 870°C). (a) Beta grains grow (in the direction of travel) by consuming alpha grains at the advancing α - β interface. (b) Generally only one alpha grain forms from each large beta grain at the receding α - β interface.

Crystals have been produced in a variety of forms including 6-mm-round bars, 1 by 25 and 3 by 13-mm strips, and in foils less than 0.3-mm thick.

Single crystals of Zircaloy-2 have been produced, for the first time, in up to 3 by 12 mm strips by the solid-state methods used for zirconium.

Crystals of zirconium have been grown by floating zone melting of 4 to 13 mm diameter bars at the rate of 3 cm/hr.

As-grown crystals usually require an extended alpha anneal to obtain sharp, Laue, diffraction spots.

Under certain conditions the existence of an allotropic transformation in a metal may aid, rather than obstruct, the preparation of single crystals at reasonable rates.

Electron-beam heating is a powerful tool for producing and studying crystal growth in metals because it is possible to produce continuously variable arbitrary, heating patterns, temperatures, and temperature gradients with a minimum of equipment and expense.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the many useful discussions with J. O. Betterton and D. S. Easton; the resistivity ratio measurements by the latter; X-ray orientation determinations by

O.B. Cavin; and chemical analyses by many persons in the Analytical Chemistry Division.

REFERENCES

1. E. J. Rapperport, *Acta Met.* 3, 208 (1955).
2. J. P. Langeron and P. Lehr, *Compt. Rend.* 243, 251 (1956), and *Mem. Sci., Revue de Met.* 55, 901 (1958).
3. E. A. Anderson et al., *Trans. AIME.* 197, 1191 (1953).
4. A. T. Churchman, *Proc. Royal Soc. (London)* 226, 216 (1954).
5. J. C. Bokros, *Trans. AIME.* 218, 351 (1960).
6. G. D. Kneip and J. O. Betterton, *J. Electrochem. Soc.* 103, 684 (1956).
7. J. P. Langeron et al., *Compt. Rend.* 248, 35 (1959).
8. *Ibid.*, *Compt. Rend.* 256, 5570 (1963).
9. W. G. Pfann, "Zone Melting," John Wiley and Sons, New York, 1958.
10. Kurt Luecke and H. P. Stuewe, "Recovery and Recrystallization of Metals," L. Himmel, Ed., p. 171, Interscience, New York, 1963.
11. J. I. Medoff and A. Miller, "Proc. 4th Symp. on Electron-Beam Technology," R. Bakish (Ed.) Boston, Mass., p. 204, 1962.
12. J. W. Glen and S. F. Pugh, *Acta Met.* 2, 520 (1954).
13. L. Renucci, P. Albert, and P. Lehr, *Mem. Sci., Revue de Met.* 60, 829 (1963).
14. K. T. Aust and Bruce Chalmers, *Can. J. Phys.* 36, 977 (1958).
15. O. C. Wells, *Proc. 4th Symp. on Electron-Beam Technology*, R. Bakish (Ed.), Boston, Mass., p. 106, 1962.

SUBSTRUCTURE AND IMPURITIES IN NIOBIUM SINGLE CRYSTALS GROWN BY THE ELECTRON-BEAM, FLOATING-ZONE TECHNIQUE

In conjunction with various investigations of the electrical, magnetic, and mechanical properties of niobium single crystals, a survey of the substructure, by etch-pit and electron-microscopic technique, and the resistivity ratios (300° and 4.2°K) was made as a function of operating conditions during crystal growth, particularly the system pressure and the crystal growth rate. For growth rates of the order of 4 mm/min, regular arrays of etch pits were observed; trace analysis, on the {111} and {112} surfaces, indicates the etching planes to be of the type {110} and the sub-boundaries to lie in {111} planes. However, analysis of etch-pit densities at apparent two- and three-boundary intersections by the method of Amelinckx did not corroborate the existence of any simple body centered cubic (bcc) sub-boundary. Further examination, by transmission electron microscopy, revealed a much greater dislocation density than that indicated by the etch-pit data, and predominantly irregular configurations. Similar investigation of crystals grown at 0.2 mm/min showed relatively few etch-pit arrays; electron micrographs revealed the dislocation density to be roughly two orders of magnitude lower than that of the rapidly grown crystals. Resistivity ratio measurements, taken along the length of the crystals, revealed greater dependence of this property on interstitial contamination, as a function of operating pressure, than on

*Metals Processing Laboratory, Massachusetts Institute of Technology, Cambridge, Mass.

substructure or possible zone-refining effects. Some evidence of precipitation was found, with morphology dependent on the rate of crystal growth. It was concluded that the high dislocation density (of the order of 10^9 lines/cm²) in the crystals grown at 2-4 mm/min precluded any unique relationship between etch pits and the actual substructure. In addition, a decoration mechanism appears to be active. Single crystals of niobium grown at the higher speeds, in vacua of the order of 10^{-5} - 10^{-6} torr, therefore are not, without subsequent annealing, of a perfection expected from the etch pit densities; growth rates below 0.5 mm/min must be used to attain dislocation densities below 10^7 lines/cm². Higher resistivity ratios may be attained by growing crystals in higher vacua; otherwise, the influence of interstitials on the electrical resistivity of the above material will predominate, and the influence of substructure, if any, will be concealed.

INTRODUCTION

The electron-beam, floating-zone technique has been a ready source of oriented single crystals of the refractory metals, with relatively high purity and perfection [1], [2], [3]. Investigations carried out at the Metals Processing Laboratory have been concerned with the mechanical [3], [4], [5], electrical, and magnetic properties [6], [7] of various Group V and Group VI materials, particularly at low temperatures; the electron-beam, floating-zone technique has been used throughout in the production of single crystals of W, Ta, Nb, V, Mo, and W-Ta mixed crystals. In the course of these studies, two patterns of deleterious effects have become increasingly apparent: excessive substructuring and high residual (4.2°K) resistivity. Both problems are particularly pronounced for the Group V elements. Where single crystals of tungsten and molybdenum may be grown with resistivity ratios ($R_{300}/R_{4.2^\circ \text{K}}$) above 100,000 and 50,000 respectively [8], [9], the situation is much more difficult for tantalum and niobium; single crystals of the latter grown in this laboratory commonly have resistivity ratios ranging between 75 and 100. The latter may be partially alleviated for tantalum and niobium by the annealing of thin wires in ultra-high vacuum, as near to the respective melting points as possible. Ratios of approximately 10,000 for tantalum [10] and 1900 for niobium [11] have been achieved in such a manner.

It is common practice to use relatively rapid zone rates (1-5 mm/min) with the electron-beam, floating-zone technique. The

formation of extensive substructure would then be not unexpected, and in fact would have several possible sources. Vacancy supersaturation in the wake of the molten zone may lead to extensive dislocation loop formation. Enhanced dislocation mobility, also due to high-vacancy concentration, combined with the stresses due to the shape of the liquid-solid interface, would be expected to result in extensive polygon structures. There is also the possibility that the substructure in the "seed" crystal may persist by growth into the new material. A good correlation between zone speed and dislocation density in molybdenum single crystals has been found by Adams and Iannucci [12]. By etch-pit techniques, the investigators found the dislocation density to increase from $10^5 - 10^6$ lines/cm² to $10^7 - 10^8$ lines/cm², when the zone speed was increased from 3-1/4 in./hr to 14 in./hr.

The measurement of the resistivity ratio is a particularly flexible and sensitive indication of the overall crystal quality, and is especially important where the intended use involves low-temperature electronic properties. Resistivity ratio measurements are consistently accurate at very low interstitial concentrations, where vacuum-fusion and other traditional methods of analysis break down [13]. Also, the method is nondestructive and affords a means of estimating variations in resistivity along the length of the crystal.

In the light of the above, extensive investigation of substructures and low-temperature electrical resistivity by various techniques was initiated, with special emphasis on the influence of operating conditions.

EXPERIMENTAL PROCEDURE

General Outline

The intention was to use systematic variations of growth rate and system pressure during growth to induce various levels of substructure and interstitial impurities. Subsequent investigation would then proceed by etch-pit techniques, transmission electron microscopy, and measurement of electrical resistivity at ambient and liquid helium temperatures.

Crystal-Growing Apparatus

The floating-zone apparatus used was of the type first described

by Calverley and co-workers [1]. Simple electrostatic focusing of the emission from an omega-shaped filament by a cylindrically symmetric gun, which itself was driven upwards by mechanical means, accomplished the zone traversal. Emission current was regulated by means of special grid-control rectifier circuits which modulate the filament heater power, according to feedback signals from the emission circuit. The voltage applied to the gun usually was set slightly under 2 kV, with emission current of 100 mA. The vacuum system consisted of a standard mechanical roughing pump of 13-cfm capacity, a 6-in. fractionating diffusion pump topped by a liquid nitrogen baffle, and a base-plate-collar combination topped by a pyrex bell jar. Elastomer gaskets were used throughout, and the limiting vacuum was approximately 10^{-6} torr. Operating vacua ranged down to 3×10^{-6} torr, and could be controlled by a variable leak.

Starting Materials

The initial material was electron-beam melted stock from the Kawecki Chemical Company; the manufacturer's analysis is presented in Table I.

Table I

Approximate Composition of Starting Material

Carbon	0.002% (by weight)
Oxygen	0.010%
Nitrogen	0.005%
Hydrogen	0.0001%
Tantalum	0.010% *
Iron	0.005%
Silicon	0.005%

*Neutron Activation Analysis performed on this material indicates a tantalum content of about 0.15%.

Crystal Growing Procedure

With the intention of separating the effects of system pressure and growth rate as operating variables, several different sets of crystals were grown, as indicated in Table II. Diameters were all 1/8 in., and a single pass was always used.

Table II

Pressures and Growth Rates

Set	Pressure, torr	Growth Rate, mm/min
A	About 1×10^{-4}	5
B	About 1×10^{-4}	0.2
C	5×10^{-4} *	5
	5×10^{-5}	
	5×10^{-6}	
D	5×10^{-6}	0.2
		2.0
		3.0
		4.0
		5.0

*The system pressure in Set C was maintained at pressures listed by the use of a variable leak.

Seeding techniques were used to control the axial orientations, which were held to within three degrees of $\langle 100 \rangle$, $\langle 110 \rangle$, $\langle 111 \rangle$, or $\langle 112 \rangle$. Chemical analyses of specimens from Set A revealed significant changes only in the interstitial content, as reported in Table III.

Table III

Interstitial Levels in Single Crystals, Set A

Carbon	0.0005% (by weight)
Oxygen	0.012%
Nitrogen	0.0007%
Hydrogen	Not detectable

Etch-Pit Specimen Preparation

Material from sets A, B, and C (see Table II) was sectioned and polished mechanically on various $\{100\}$, $\{110\}$, $\{111\}$, and $\{112\}$ planes. Final polishing was accomplished chemically by a mixture of 10 parts of HF(48%), 20 parts concentrated HNO_3 , and 25 parts lactic acid, all by volume; about 0.02 in. of depth was removed by the final polish, in order to insure an unperturbed

surface. Etching was done with the reagent used by Rexer [13] for niobium.

Specimen Preparation for Electron Microscopy

Sections were taken from material in sets A and B. Thinning for transmission electron microscopy was accomplished by the technique of Meieran and Thomas [14]. Observations were made on planes perpendicular to the growth direction.

Resistivity Ratio Measurements

The resistivity ratios were measured by four-point techniques, with a voltage sensitivity of approximately 2×10^{-9} V. Measurements at 4.2°K were made in a magnetic field to insure normal resistive behavior. Voltage and current contacts were made by controlled spot-welding of fine nickel wires to the surfaces of the crystals.

EXPERIMENTAL RESULTS

A. Etch Pits

Etch-Pit Density vs. Operating Vacuum. Figure 1 shows three typical pictures, at an apparent magnification of 2000X. As the operating pressure decreases from 5×10^{-4} torr to 5×10^{-6} torr, the pit density decreases, and regular arrays of pits form, with very low density between the rows. The highest density, at the highest pressure, is approximately 5×10^6 pits/cm². All three pictures are {111} planes of crystals with <111> axial orientations.

Etch-Pit Density vs. Zone Speed. The photomicrographs in Fig. 2 are typical of the {111} planes of rapidly grown crystals, set A. Figure 3 shows pitting of the {112} plane of the same material; the growth rate was 5 mm/min in all cases. The regular lines of pits in Fig. 2 are always in the <110> directions; in Fig. 3, the lines are approximately in the <123> directions. Pitting of the slowly grown crystals, i.e., set B, occurred in an entirely different manner, as shown in Fig. 4. The rate of pitting and pit growth was extremely low, and resolution was in many cases impossible, even after long immersion times in the etchant.

Pit Shape and Pitting Planes. No pitting of the $\{100\}$ or $\{110\}$ sections of the single crystals was observed, unless straining was induced. The pit edges, that is, the intersection of the pit planes and the plane observation, were always in the $\langle 110 \rangle$ directions, when the $\{111\}$ planes were etched; the pit shape was triangular, as shown in Fig. 2. Pitting of the $\{112\}$ planes resulted in the pennant-shaped pits of Fig. 3. The long axis of the pits, in the latter case, lies along the $\langle 111 \rangle$ directions, and the truncated end is parallel to the $\langle 110 \rangle$ direction perpendicular to the long axis.

B. Transmission Electron Microscopy

Growth Rate and Dislocation Density. Material from the rapidly grown crystals of set A showed an extremely high average dislocation density; the estimated average is well above 10^9 lines per cm^2 , on the basis of all the photographs taken. In addition, the structure tended to cellularize into regions of high and low density, as shown in Fig. 5. The slowly grown crystals, set B, were considerably better; Fig. 6 is typical. The dislocation densities are then of the order of 10^7 lines/ cm^2 .

Precipitates and Precipitate Morphology. Precipitates were sometimes observed, with morphology dependent on growth rate. Figure 7 is a photomicrograph of a section from set A, the rapidly grown set. The precipitates in set B all resembled Fig. 8.

C. Resistivity Ratio Measurement

Influence of Operating Pressure on Average Resistivity Ratio. The cumulative results of the resistivity ratio measurements on set C are presented in Fig. 9. The data for the highest ratios were taken from previous work by the authors [15].

Influence of Growth Rate on Resistivity Ratio. No correlation was observed between the growth rate of the single crystals in set D and their resistivity ratios.

Segregation Effects in Slowly Grown Crystals. The material grown at the slowest rate of set D exhibited a variation of resistivity ratio characteristic of zone-refining effects. Figure 10 plots resistivity ratio versus distance for the first two crystals in set D. The faster grown crystal showed no segregation effects; the crystals grown at 3, 4, and 5 mm/min acted similarly. The

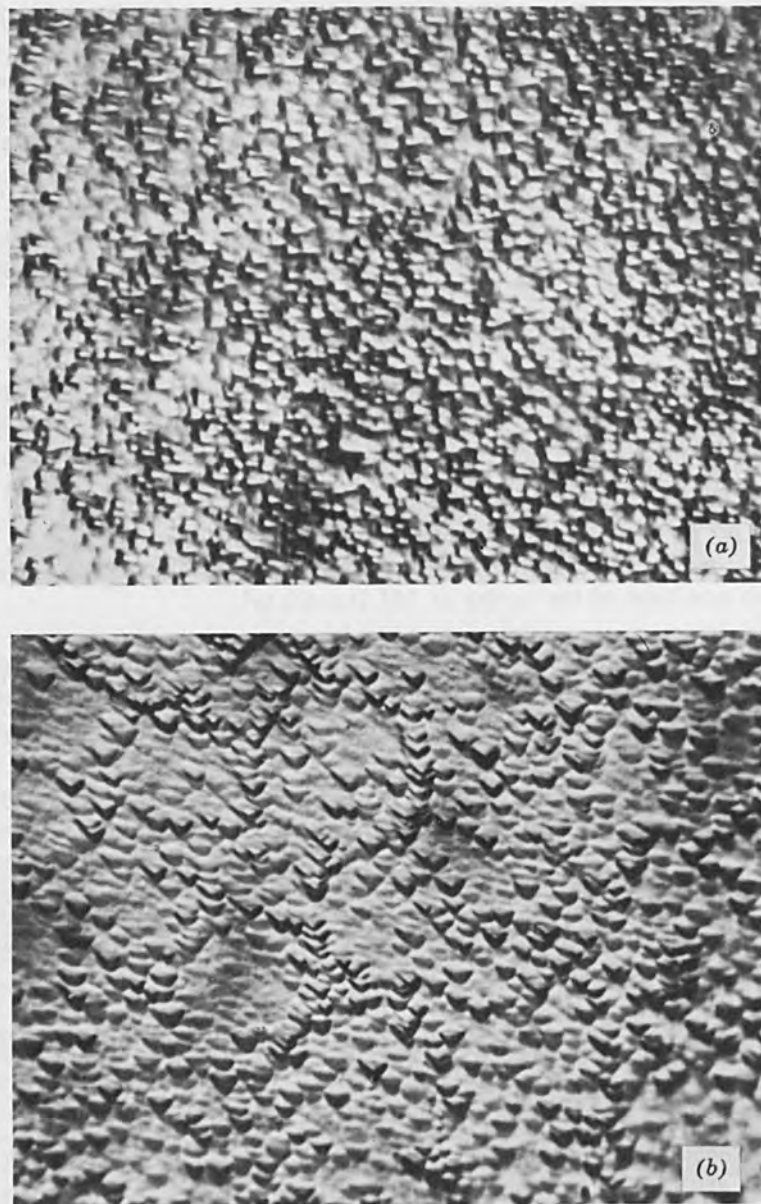


Fig. 1. Effect of system pressure on etch-pit density. Specimens from set C; growth rate was 5 mm/min. 2000X. (a) System pressure = 5×10^{-4} torr. (b) System pressure = 5×10^{-5} torr. (c) System pressure = 5×10^{-6} torr.



crystals were grown from the viewpoint of Fig. 10, from "left" to "right."

DISCUSSION

It is readily apparent, from the electron transmission micrographs, Fig. 5 and 6, that the relatively high growth rates, i.e., circa 5mm/min, result in very low levels of crystal perfection, in fact much lower than would be expected from the etch-pit results. In order to attain levels of perfection comparable to that which has been achieved in metals such as iron or copper, growth rates at least one order of magnitude lower appear to be desirable, and, in fact, as low as is compatible with the evaporation rate of the metal involved.

As the traces of the pitting planes are $\langle 110 \rangle$ and $\langle 111 \rangle$, the two possibilities for the etch-pit planes are $\{112\}$ and $\{110\}$. In view of the appearance of the pits, and the fact that the $\{112\}$ sections pit readily, and the $\{110\}$ surfaces do not pit at all, we are inclined to favor the $\{110\}$ as the etch-pit planes. The only positive statement possible about the relationship between the etch pits and

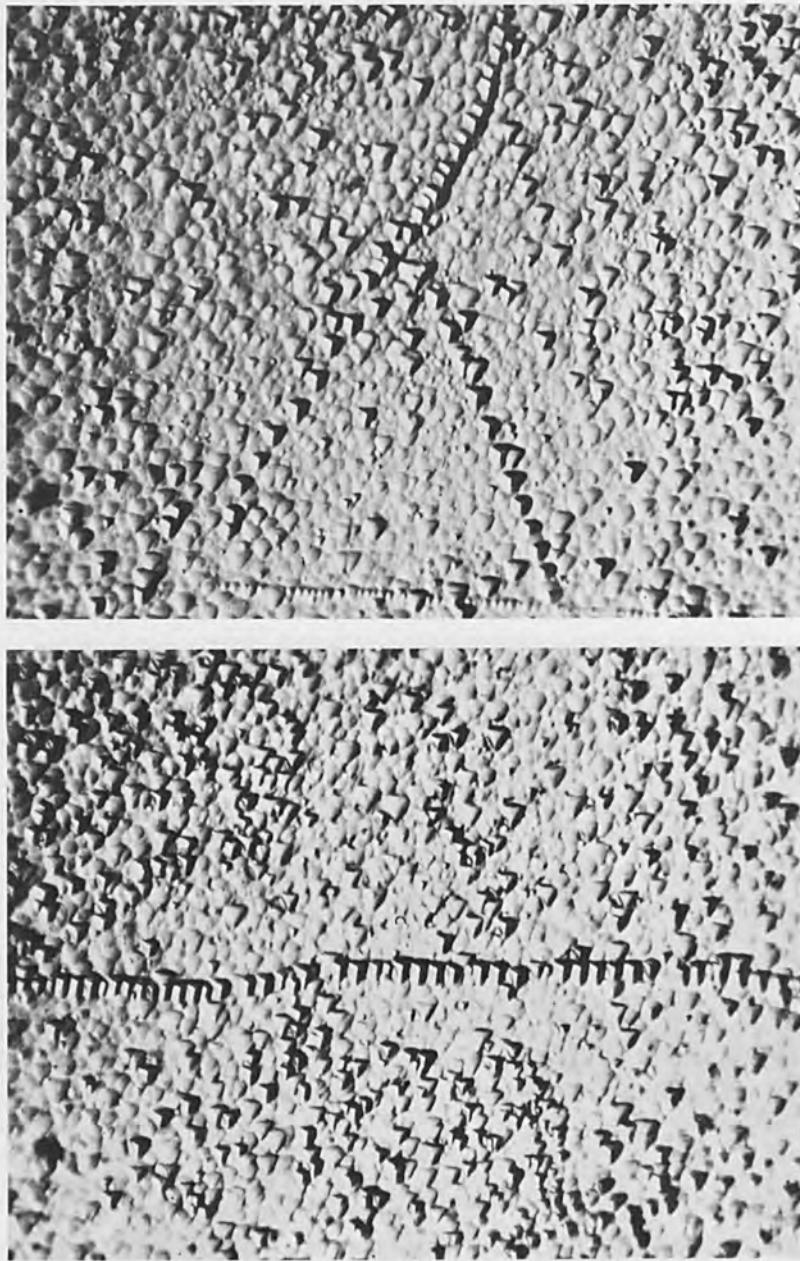


Fig. 2. Effect of rapid zone speed on etch-pit density. Specimens from set A; growth rate was 5 mm/min. Surfaces shown are $\{111\}$ planes or $\langle 111 \rangle$ axially oriented crystal. 2000X.

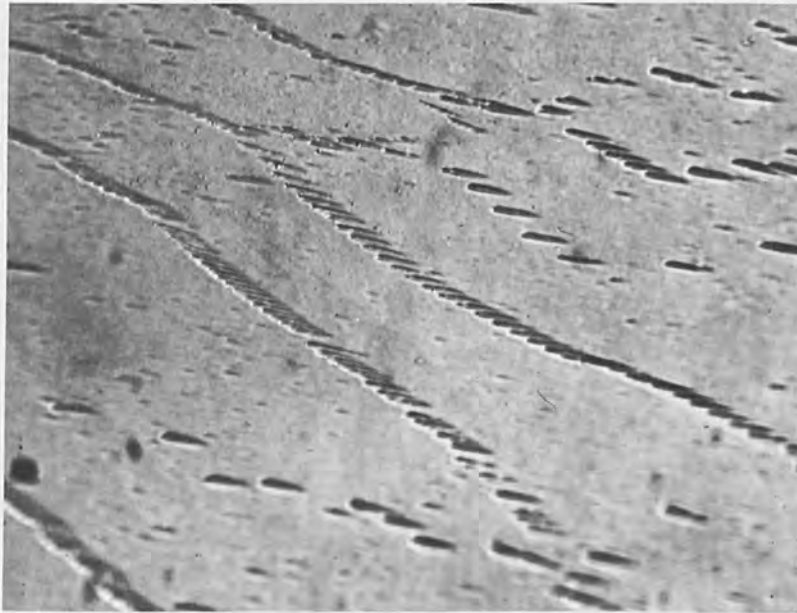


Fig. 3. Effect of rapid zone speed on etch-pit density. Specimen from set B; growth rate was 5 mm/min. Surface shown is a $\{112\}$ surface of a crystal of $\langle 111 \rangle$ axial orientation. 2000X.

the actual substructure of the single crystals, is that it is not direct. Besides the observation that the etch-pit densities obtained here are not indicative of the actual dislocation densities, it appears that the rows of etch pits in Figs. 2 and 3 do not completely identify the possible sub-boundaries they may represent. The implication of the rows of pits in Figs. 2 and 3 is of sub-boundaries lying in the $\{111\}$ planes, i.e., simple tilt boundaries which cut the $\{111\}$ planes in $\langle 110 \rangle$ directions, and the $\{112\}$ planes in $\langle 321 \rangle$ directions. However, the pit densities near the intersections of such boundaries must satisfy the relationships set down by Amelinck [16], based on Frank's Rule [17]. In short, any possible dislocation model must satisfy the relationship

$$\sum_{i=1}^N \left[\left(\sum_{j=1}^D \rho_{ij} \vec{b}_j \right) \cdot \vec{n} \right] \left[\vec{v}_i \cdot \vec{n} \right] = 0 \quad (1)$$

at any boundary intersection. $\vec{n}$ is the unit vector normal to the

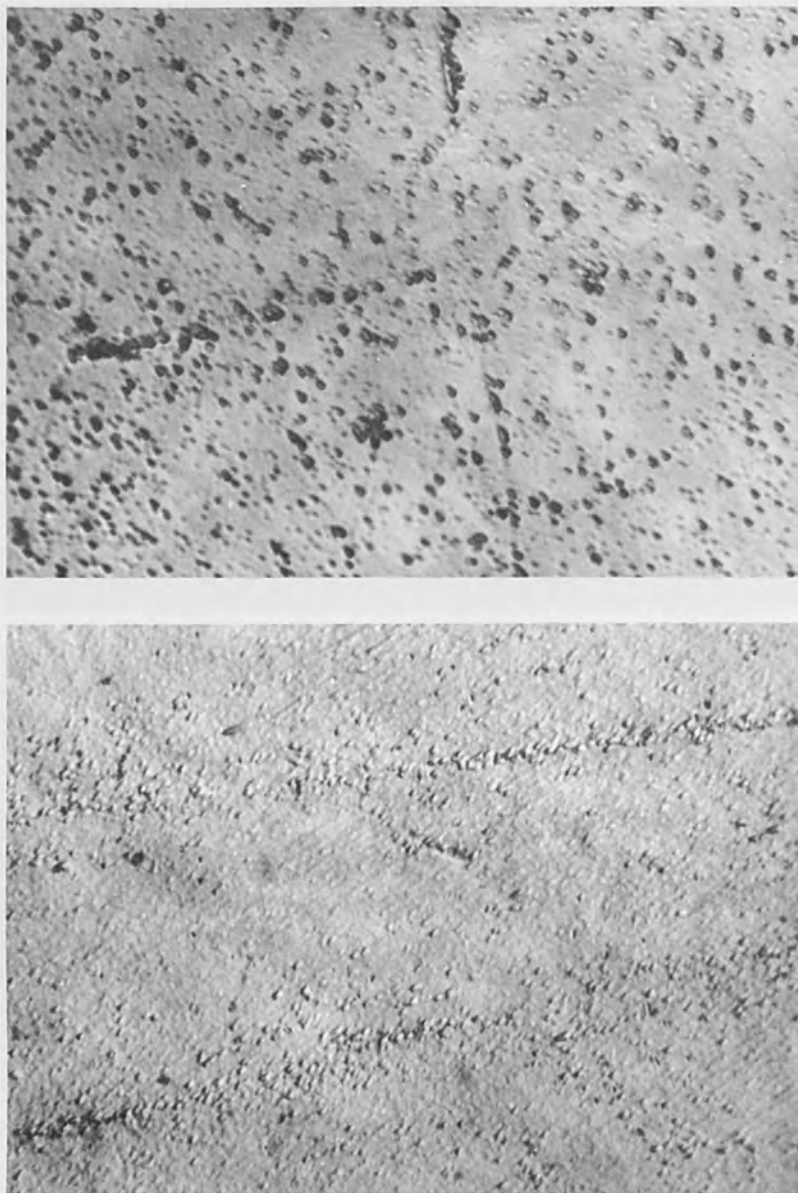


Fig. 4. Effect of slow zone speed on etch-pit density. Specimen from set B; growth rate was 0.2 mm/min. Surfaces shown are $\{111\}$ surfaces of crystals with $\langle 111 \rangle$ axial orientations. 2000X.

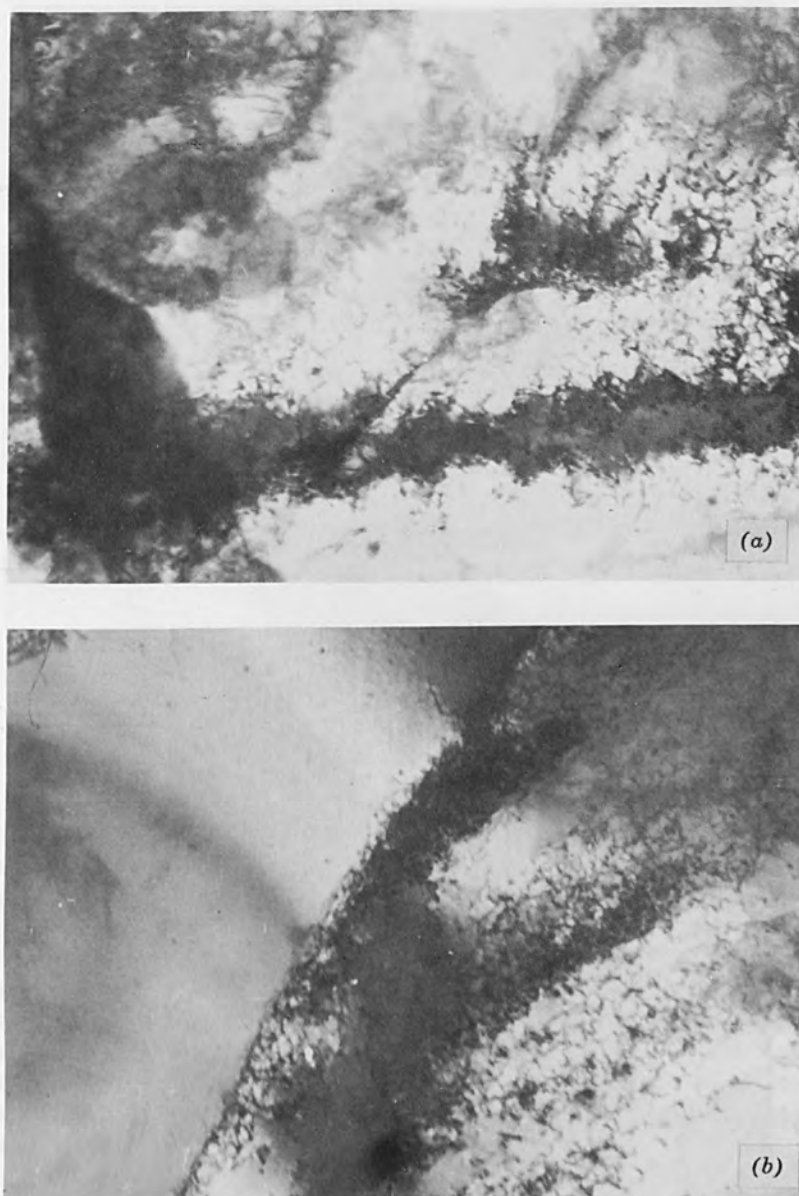


Fig. 5. Electron micrographs of rapidly grown crystals (set A).
(a) $\{111\}$ surface of a crystal of $\langle 111 \rangle$ axial orientation. 40,000X.
(b) $\{100\}$ surface of a crystal with $\langle 100 \rangle$ axial orientation. 40,000X.
Zone speed was 5 mm/min.

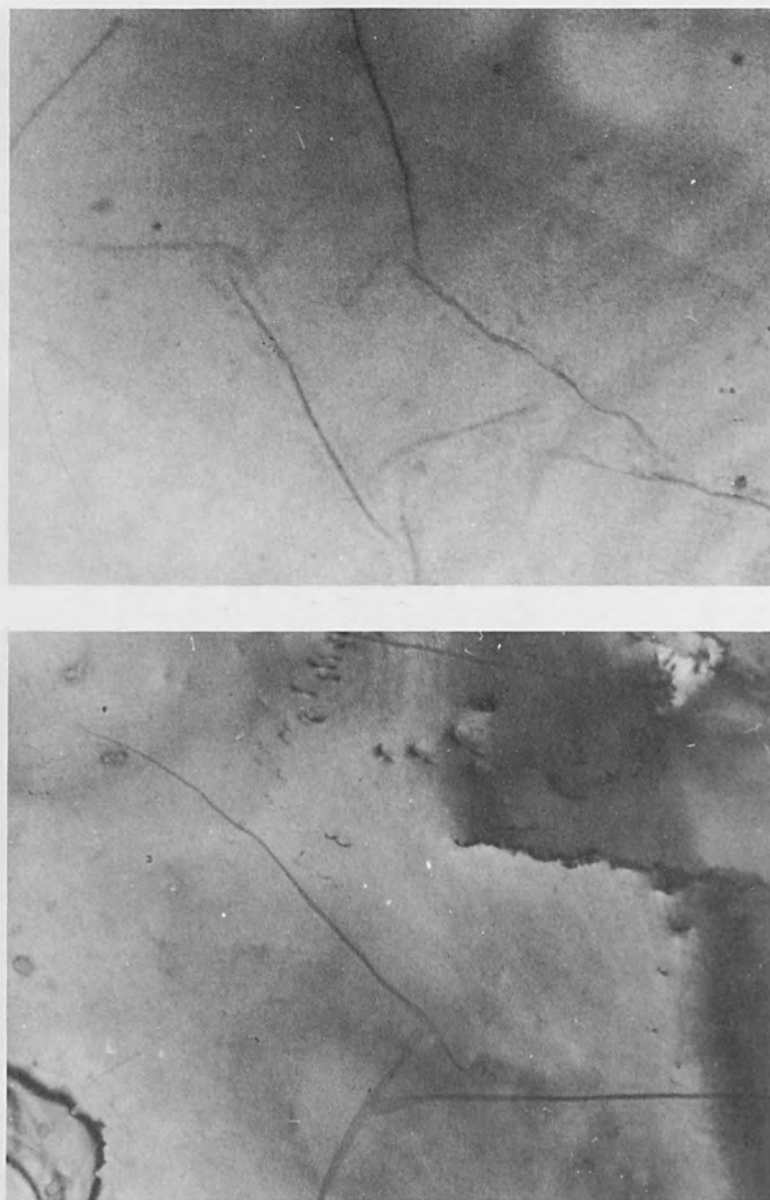


Fig. 6. Electron micrographs of slowly grown crystals (set B). Growth rate was 0.2 mm/min. Both micrographs are of $\{100\}$ surfaces in crystals with $\langle 100 \rangle$ axial orientations. 40,000X.



Fig. 7. Electron micrograph of precipitate in rapidly grown crystal. Specimen from set A; zone speed was 5 mm/min. Surface shown is a $\{111\}$ plane of a crystal of $\langle 111 \rangle$ axial orientation. 32,000X.

plane of observation; $\bar{n}_i$ is the unit vector normal to the plane of observation; $\bar{v}_i$ is the unit vector normal to the sub-boundary denoted by the subscript "i." " b_j " is the burgers vector of the parallel set of dislocations denoted by the subscript "j." ρ_{ij} is the linear density (along the line of intersection with the plane of observation) of dislocations of burgers vector b_j in the "i"th plane. Examination of a number of three-way intersections, such as those in Fig. 2, and two-way intersections, such as those in Fig. 3, together with consideration of a number of alternate bcc sub-boundary models, have led the authors to the following conclusion: that a linear correspondence between pit density and dislocation density may exist only if the dislocation reaction

$$\frac{1}{2} [1\bar{1}1] + \frac{1}{2} [11\bar{1}] \rightarrow [100] \quad (2)$$

prevails at all sub-boundary intersections. To date, no electron micrographic evidence for such extensive reaction has been found by the investigators. In view of the apparent dependence of pit



Fig. 8. Electron micrograph of precipitate in slowly grown crystal. Specimen from set B; zone speed was 0.2 mm/min. Surface shown is a $\{100\}$ plane of a crystal of $\langle 112 \rangle$ axial orientation. 100,000X.

density and configuration on the operating vacuum, a decoration or precipitation mechanism for etch-pitting is plausible. Oxide precipitation, for instance, would probably occur preferentially along the planes of high dislocation density shown in Fig. 5.

On the basis of Fig. 9, the large sensitivity of the residual resistivity to interstitial impurities, particularly oxygen and nitrogen, is established. However, the curves in Fig. 10 imply the zone-refining of an impurity which raises the melting point, as well as substantially lowering the resistivity ratio. Tantalum, tungsten, and molybdenum are the most likely possibilities. Because normal "wet" and spectrographic techniques have proven to be unreliable at these low levels, it is not possible at this time to know definitely the impurity element responsible for the behavior of Fig. 10. Neutron activation analysis may provide part of the answer [18].

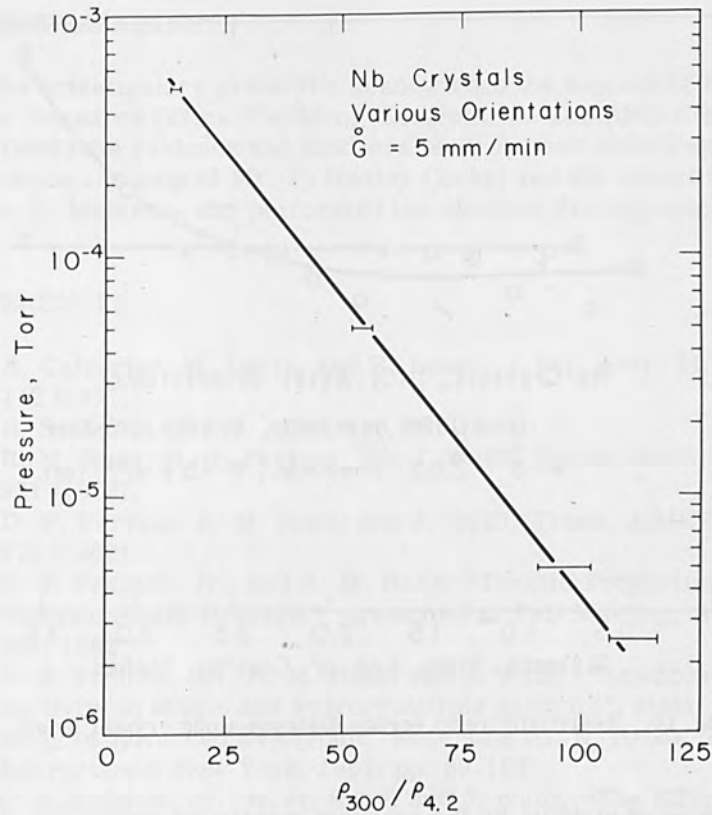


Fig. 9. Average resistivity ratio versus operating pressure of system.

CONCLUSIONS

Very high dislocation densities, above 10^9 lines/cm², exist in niobium single crystals grown at rates of the order of 5 mm/min by the electron-beam, floating-zone technique. A reduction in growth rate of at least an order of magnitude is necessary in order to achieve levels of perfection common in melt-grown single crystals of many nonrefractory materials.

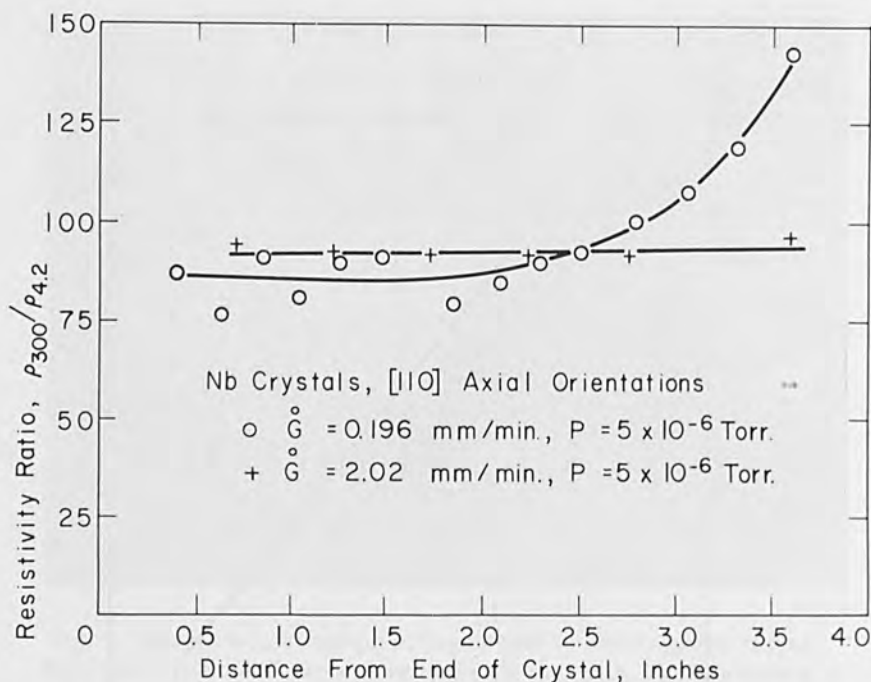


Fig. 10. Resistivity ratio versus distance along crystal length.

Etching of niobium by the reagents mentioned previously pits the $\{111\}$ and $\{112\}$ surfaces by revealing the $\{110\}$ planes. The $\{110\}$ and $\{100\}$ surfaces do not pit, unless strained material is present.

It is not possible at this time to set up a unique relationship between the etch pits observed on the surfaces of niobium and the dislocations and substructure present. In general, the actual dislocation densities may be far in excess of the observed etch-pit density.

The most important influence on the residual resistivity of niobium single crystals grown by the electron-beam, floating-zone method is the operating vacuum, to the extent that the influence of substructure and crystal perfection is concealed.

Zone-refining effects have been observed at growth rate of 0.2 mm/min, which may be due to molybdenum, tantalum, or tungsten.

ACKNOWLEDGMENTS

The investigators gratefully acknowledge the support of U.S. Army Research Office (Durham), Contract No. DA-ARO-(D)-31-124-G402; the guidance and kindness of Professor John Wulff; the advice and help of Mr. F. Hadley Cocks; and the expert help of Dr. E. Meieran, who performed the electron micrography.

REFERENCES

1. A. Calverley, M. Davis, and R. Lever, *J. Sci. Instr.* 34, 142 (1957).
2. H. Schadler, *Trans. AIME* 218, 649 (1960).
3. R. M. Rose, D. P. Ferriss, and J. Wulff, *Trans. AIME* 224, 981 (1962).
4. D. P. Ferriss, R. M. Rose, and J. Wulff, *Trans. AIME* 224, 975 (1962).
5. C. S. Tedmon, Jr., and R. M. Rose, "Tensile Properties of Niobium Single Crystals", presented at Fall Meeting, AIME, Oct. 1962.
6. C. S. Tedmon, Jr., R. M. Rose, and J. Wulff, "Superconductivity in Single and Polycrystalline Niobium", *Metalurgy of Advanced Electronic Materials* (G. E. Brock, Ed.), Interscience, New York, 1963, pp. 89-104.
7. C. S. Tedmon, Jr., R. M. Rose, and J. Wulff, "The Effect of Controlled Oxygen Additions on the Superconducting Properties of Niobium Single Crystals", presented at Annual Meeting, AIME, Feb. 18, 1964, New York. To be published.
8. I. Drangel, and G. T. Murray, "Parameters Pertinent to Purification by Electron-Beam Zone Refinement", presented at Annual Meeting, AIME, Feb. 17, 1964, New York.
9. E. Rexer, "On Some Properties of Electron Beam Melted Metals", *Proc. 4th Symp. on Electron Beam Technology*, R. Bakish (Ed.), 1962, Boston, Mass., pp. 245-275.
10. J. I. Budnick, *Phys. Rev.* 119, 1578 (1960).
11. T. F. Stromberg, and C. A. Swenson, *Phys. Rev. Letts.* 9, 370 (1962).
12. M. A. Adams, and A. Iannucci, WADC Tech. Rept. 59-441, Part II, Dec. 1960.
13. To be published.
14. D. A. Thomas, and E. Meieran, *Trans. AIME* 227, 284 (1963).

15. C. S. Tedmon, Jr., Sc.D. Thesis, Dept. of Metallurgy, M.I.T., June, 1964.
16. S. Amelinckx, *Physica* 23, 663 (1957).
17. F. C. Frank, Rept. Symp. on the Plastic Deformation of Crystalline Solids, Carnegie Inst. of Tech., Pittsburgh, 1950, p. 150.
18. F. H. Cocks of our laboratory is presently working on this problem.

ON THE DEFORMATION, RECRYSTALLIZATION, AND WELDING OF MOLYBDENUM SINGLE CRYSTALS

INTRODUCTION

Part of the research work done in the "Institut für angewandte Physik der Reinststoffe in Dresden" is the study of certain properties of pure VIa group transition metals, particularly those of molybdenum [1]-[7].

The behavior of electron-beam, zone-refined Mo-single crystals under plastic deformation has already been treated in several publications [8]-[11]. Repeatedly, for molybdenum single crystals, dependence of ductility on orientation has been mentioned [10]-[12]. Furthermore, the recrystallization temperatures of rolled Mo-crystals were found to depend on the orientation of rolling, i.e., on the crystallographic orientation of rolling plane and rolling direction [11].

The first part of this paper deals with these incompletely studied properties describing running experiments on the influence of different orientations on deformation and recrystallization behavior of Mo-single crystals.

PREPARATION AND PRETREATMENT OF SINGLE CRYSTALS

Mo-rods of 5-mm diameter produced by powder metallurgy¹ were processed into single crystals by triple electron bombardment zone refining. During melting the pressure was 5×10^{-6} mm Hg, the speed of zoning was 3 mm/min. The single crystals had a

*Institute for Physics of Pure Materials, Dresden, East Germany.

¹Supplied from VEB Berliner Glühlampenwerk.

mean diameter of 4.2 mm and a length of 80-110 mm. For zone melting, an arrangement was used which has been previously described [13].

The orientations of the crystal axes were predetermined by melting on suitable seeds. After fixing by X-ray back reflection photographs, the desired rolling planes were grinded on two opposite surfaces and electrolytically polished. In general, the errors arising from both the processes were kept below 5°.

PURITY OF AS-GROWN SINGLE CRYSTALS

From some single crystals the residual resistance ratio $R(4.2^\circ\text{K})/R(273^\circ\text{K})$ was determined to be 1.9×10^{-4} . Out of this, careful estimation [11] resulted in a total dissolved impurity content of only a few ppm by weight.

By means of vacuum hot extraction² and a carbon microanalysis [15] a content of 5 ppm by weight each of oxygen, nitrogen, and carbon could be determined. Owing to the measured residual resistance ratio, part of these impurities may be present not dissolved but in form of precipitation.

A first survey analysis carried out with the aid of a mass spectrometer for solids³ showed a content of 170-ppm tungsten and 2-ppm cadmium. All other impurities were below 1 ppm.

DEFORMATION BY ROLLING AT ROOM TEMPERATURE

Important factors for the deformability of single crystals are the orientations of the rod axis as well as that of the rolling plane [12], [16].

Variations of Hardness. Rolling was done at room temperature with a reduction per pass of about 0.003 mm down to the final thickness of 0.75 mm. Figure 1 shows schematically the variation of hardness due to rolling (scattering ± 5 VHN). Comparable to all hardness curves is the steep slope by 25-50 VHN up to a deformation of 10%. Further deformation caused no, or only weak, strain hardening in the case of the orientations combined in Fig. 1a, while those of Fig. 1b showed a further considerable increase.

²The vacuum fusion analyser GA 01 of Messrs. E. Leybold's Nachfolger, Köln, was used [14].

³Mass spectrometer MS 7, manufactured by the Associated Electrical Industries Ltd., Manchester (GB).

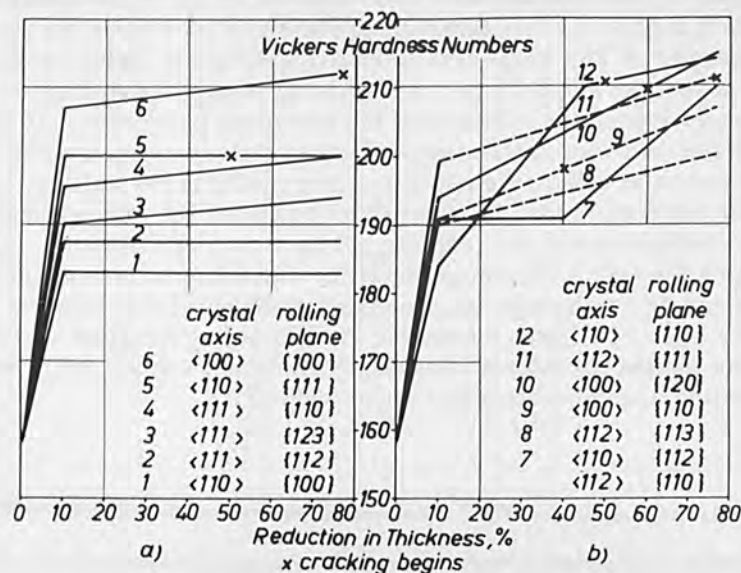


Fig. 1. Rolling of Mo-single crystals at room temperature.

The most favorable deformation properties were found at crystals with the orientation $\langle 110 \rangle \{100\}$ (Fig. 1, curve 1) known as mean rolling deformation texture. Such crystals also allowed rolling with a reduction per pass of 0.06 mm without causing any variation of the hardness curve or cracking.

For most of the other orientations, rolling with higher rates of reduction resulted in steeper hardness curves and earlier cracking.

Alterations of Shape. The crystallographic orientation influenced not only the strain hardening factor but also the alterations of crystal dimensions caused by rolling (see reference [17]). This effect is represented in Fig. 2 for three crystals of different orientations being rolled down to 0.75 mm as compared with a not deformed single crystal of 105-mm length. A paper to be published in "Zeitschrift für Metallkunde" will explain the different lengthening and widening of the crystals.

ANNEALING OF DEFORMED MO-SINGLE CRYSTALS

The samples obtained by rolling were suspended in a Mo-container and annealed for 60 minutes in a vacuum of 1×10^{-5} to 1×10^{-6} mm Hg by indirect inductive heating.

Bending Ductility and Recrystallization. In Fig. 3, the maximum bending angles at room temperature are plotted versus annealing temperature. The shape of the curves shown in Fig. 3a resulted from a first test period, the specimens having been cleaned only with acetone between the rolling and the annealing processes. At the beginning of recrystallization, the three different orientations had the maximum detectable ductility. Increasing recrystallization led to embrittlement, in general not reaching the highest degree at the temperatures of complete primary recrystallization. After running through a minimum, ductility was found to rise again. This can be compared with the observation published first by Perry et al. [18]-[20], that in the case of polycrystalline molybdenum the ductility after reaching a minimum can be restored by annealing at temperatures of about 2000°C.



Fig. 2. Comparison of Mo-single crystals rolled down to 0.75-mm thickness with an as-grown crystal, 1:2,5.

Furthermore, Fig. 3a indicates dependence of the recrystallization temperatures on orientation. From publications for Al [21] and Si-Fe [22] it is known that deformation with suitable orientations yields weak lattice distortion and inhibition of recrystallization. This fact was the starting point for a second test series with specimens which had been electrolytically polished after rolling⁴ in order to eliminate possible influences of the surface layer. Results are plotted in Fig. 3b. Crystals having the orientation $\langle 100 \rangle \{100\}$ showed incomplete recrystallization, but no embrittlement (curve 2). For the orientation $\langle 111 \rangle \{112\}$ the temperature of recrystallization and the minimum ductility shifted to higher values as compared with Fig. 3a (curve 3).

⁴For specimen polishing the electrolyte sulphuric acid-methyl alcohol (1:7) was always used [23].

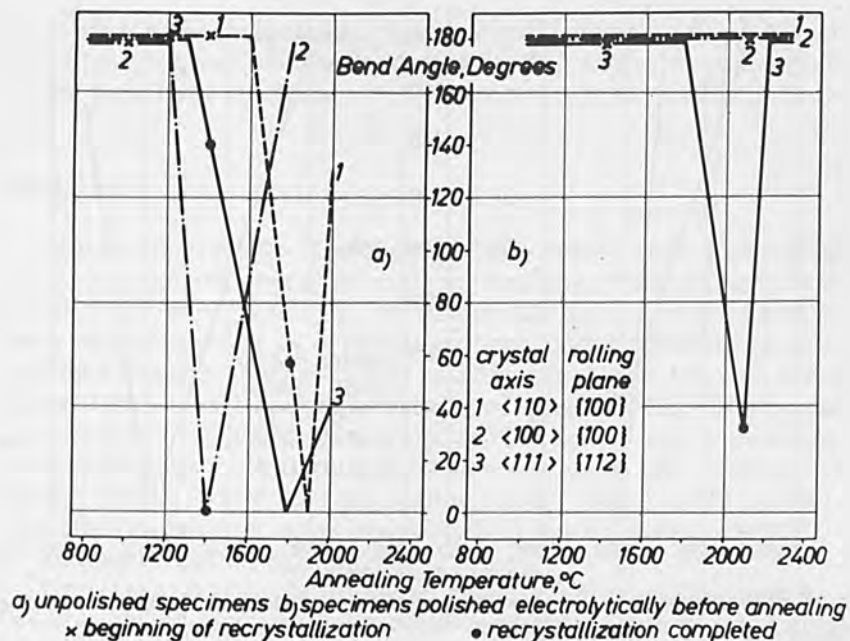


Fig. 3. Bend ductility of Mo-single crystals after rolling and annealing deformation: 70-80% annealing time: 60 minutes.

Figure 4 shows a similar comparison for three other orientations. Opposed to the behavior of those of Fig. 3, in the case of the unpolished specimens (Fig. 4a), the ductility could not be restored after it had reached the minimum. For polished specimens with the orientations $\langle 112 \rangle \{113\}$ and $\langle 112 \rangle \{110\}$ recrystallization without embrittlement could be found (Fig. 4b, curves 1,2). The orientation $\langle 110 \rangle \{110\}$ in the case of unpolished and polished specimens revealed similar recrystallization and ductility behavior.

ELECTRON-BEAM WELDING OF DEFORMED Mo-SINGLE CRYSTALS

A great number of papers [24]-[27] deal with problems arising from welding of molybdenum, particularly with the low ductility of the welding seams. For this reason electron-beam welding tests [28] have been made with rolled and electrolytically polished specimens having the six initial orientations shown in Figs. 3 and 4.

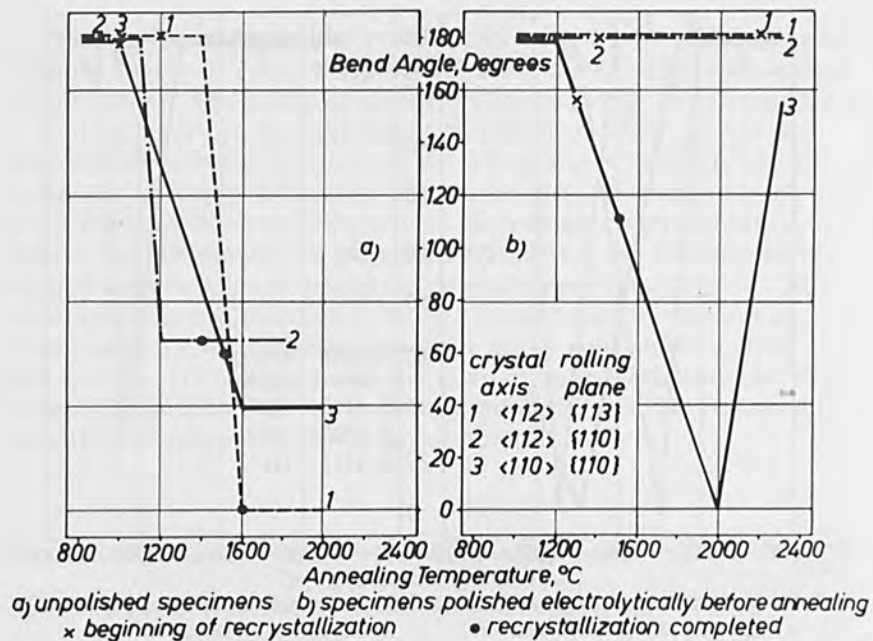


Fig. 4. Bend ductility of Mo-single crystals after rolling and annealing deformation: 70-80% annealing time: 60 minutes.

The specimens got a cross-welding seam, 1.5 mm in width. After having removed the edge penetration by grinding and electrolytical polishing, the welding seam was bent at room temperature. The following maximum bending angles could be observed:

Orientation		Maximum bending angle, degrees
$\langle 100 \rangle$	$\{ 100 \}$	180
$\langle 110 \rangle$	$\{ 100 \}$	180
$\langle 110 \rangle$	$\{ 110 \}$	45 - 130
$\langle 111 \rangle$	$\{ 112 \}$	180
$\langle 112 \rangle$	$\{ 110 \}$	180
$\langle 112 \rangle$	$\{ 113 \}$	180

That means that, in spite of the welding seams, very good ductility could be found. The only exception arose in the case of the orientation $\langle 110 \rangle \{ 110 \}$, the recrystallization behavior of which differed also from that of the other orientations (Fig. 4b, curve 3).

Comparative weldings at powdermetallurgically prepared technical molybdenum sheet resulted in bending angles of 40 degrees under equal test conditions.

SUMMARY

Mo-single crystals of predetermined orientations prepared by electron bombardment zone-melting could be rolled at room temperature by more than 80%. Strain hardening illustrated by hardness values depends on orientation. Subsequent annealing of unpolished specimens caused recrystallization connected with strong embrittlement. With electrolytically polished specimens of some orientations, recrystallization and embrittlement can be made to occur at higher annealing temperatures or not at all. Crystals having the axis $\langle 110 \rangle$ and the rolling plane $\{100\}$ showed lowest work hardening, and, after electrolytical polishing up to 2400°C, no recrystallization phenomena.

For a series of orientations no reduction of ductility could be observed after electron-beam welding of rolled and polished single crystals except of crystals of the orientation $\langle 110 \rangle \{110\}$.

ACKNOWLEDGMENTS

The author wishes to thank Messrs. K. H. Berthel, H. Engelhardt, K. Friedrich, H. Mai, D. Müller, W. Neumann, R. Petri, K. Schlaubitz, and E. Zedler, all of whom took part in this investigation.

REFERENCES

1. E. Rexer, *Exp. Techn. Phys.* **11**, 381 (1963).
2. E. M. Herbst, D. Schulze, and H. Wadewitz, *Exp. Techn. Phys.* **11**, 332 (1963).
3. H. Wadewitz, in "Reinststoffe in Wissenschaft und Technik," Akademie Verlag, Berlin, 1963, 301.
4. J. Prohaszka and H. Wadewitz, in "Reinststoffe in Wissenschaft und Technik," Akademie Verlag, Berlin, 1963, 313.
5. M. Schubert, *Monatsber. d. Dt. Akad. d. Wiss. zu Berlin* **5**, 518 (1963).

6. E. Zedler, P. Finke, K. Friedrich, D. Kraut, D. Müller, and K. Schlaubitz, in "Reinststoffe in Wissenschaft und Technik," Akademie Verlag, Berlin, 1963, 323.
7. E. Zedler, and U. Wiesner, Abh. d. Dt. Akad. d. Wiss. zu Berlin, Kl. f. Bergbau, Hüttenwesen u. Montangeologie 53 (1963).
8. H. S. Spacil, and J. Wulff, in "The Metal Molybdenum," ASM, Cleveland, Ohio, 262 (1958).
9. J. A. Belk, J. Less-Common Met. 1, 50 (1959).
10. A. Lawley, J. van den Syde, and R. Maddin, J. Inst. Metals 91, 23 (1962-1963).
11. E. M. Savitski, and Tao Tsu-Zun, Izvest. Akad. Nauk SSSR, Otd. Metallurg. i. Toplivo, 133 (1962).
12. R. Maddin, in "The Metal Molybdenum," ASM, Cleveland, Ohio, 214 (1958).
13. K.-H. Berthel, H.-E. Longo, and K. Schlaubitz, in "Reinststoffe in Wissenschaft und Technik," Akademie Verlag Berlin, 1963, 293.
14. G. Ehrlich and K. Friedrich, Neue Hütte 8, 27 (1963).
15. K. Friedrich, Microchimica Acta, 674 (1963).
16. K. T. Aust, R. Maddin, and H. K. Chen, Trans. AIME 5, 1477 (1953).
17. H. Gries and H. Esser, Arch. Eisenhüttenwesen 2, 749 (1929).
18. T. Perry, H. S. Spacil, and J. Wulff, Welding Res. Suppl. Sept. 442 (1954).
19. T. Perry, H. S. Spacil, and J. Wulff, Metal Progr. 65, 75 (1954).
20. L. Northcott, "Molybdenum," Butterworth Sci. Publ., London, 1956.
21. A. H. Lutts and P. A. Beck, J. Met., Trans. AIME 6, 257 (1954).
22. Hsun Hu, Trans. Met. Soc., AIME 224, 75 (1962).
23. W. J. Mc G. Tegart, "The Electrolytic and Chemical Polishing of Metals", Pergamon Press, New York, 1959.
24. N. A. Olshanski and A. V. Mordvinceva, Avtomat. Svarka 36 (1960).
25. S. M. Gurevic and G. K. Charcenko, Avtomat. Svarka 12 (1961).
26. W. N. Platte, in Met. Soc. Conf., Vol. 17, "Refractory Metals and Alloys II", Interscience Publishers, New York 1963, 307.

27. M. H. Scott and P. M. Knowlson, *J. Less Common Met.* 5, 205 (1963).
28. R. Göbel, E. Heinze, W. Neumann, E. Rexer, and K. Schlau-bitz, *Kernenergie* 5, 104 (1962).

FIELD-AIDED ELECTRON-BEAM ZONE REFINING OF TUNGSTEN

INTRODUCTION

Various methods have been devised to effect the separation of components of a solution at a solidifying interface, thereby purifying a primary phase. Fractional crystallization has long been employed as a method of purification, and more recently zone melting has resulted in the preparation of materials with purities hitherto unattainable. The use of electron-beam heating extended the application of zone refining to the refractory metals. The study of electrotransport of metallic ions in molten metal systems indicates that electrodiffusion can be used to control solute distribution. Pfann and Wagner [1] have shown in a theoretical analysis that electrotransport in a molten zone can be used to control the solute distribution coefficient over a wide range. Tougas and Hone [2] have succeeded in changing the effective distribution coefficient in a tin-2% lead alloy from 0.87 to 0.63 by establishing an electric field in the sample. The general expression "field freezing" is used to denote freezing methods in which an electric field is employed to alter the solute concentrations in solidifying liquids. The technique whereby an electric field is applied to a metal as it is zone refined is termed field-aided zone refining.

Control of the distribution coefficient in zone refining is of two-fold interest. A distribution coefficient of unity can be increased

*Department of Engineering Physics and Materials Science, Cornell University, Ithaca, N.Y. This work was performed in the High Temperature Laboratory of the Materials Science Center, Cornell University, and was supported by the Advanced Research Projects Agency.

or decreased, making possible the transfer of some previously inseparable solutes. Also, the general efficiency of zone refining can be increased. Since, in many of the refractory metal systems distribution coefficients of close to unity are quite common, it is expected that field freezing would be especially significant here. The effect of field freezing on the efficiency of zone refining was studied in a commercially pure, unalloyed tungsten. The purity level of the zone-melted tungsten was examined as a function of the polarity and intensity of the applied field using the relative resistivity ratio as a measure of purity. It is shown that significant increases in zone-refining efficiency can be achieved through field freezing and that higher levels of purity are produced.

The importance of zone-refining action in the purification of tungsten by electron-beam zone melting has generally been considered to be small compared with impurity removal by volatilization. The extent to which impurities are accumulated at the advancing solid-liquid interface and not removed by vaporization is made evident by the effect of electrodiffusion on the enhanced purification.

EXPERIMENTAL PROCEDURES

Electron-Beam Zone Refining of Tungsten

The floating zone method was used in zone refining the tungsten samples. The basic electron-beam unit, a modified Mark VIII melter-evaporator (Fig. 1), was designed and built by Alloyd Electronics Corporation. This equipment employs a hidden filament electron gun design to avoid contamination of the sample from the filament. The arrangement of filament and electrostatic focusing shields are shown in Fig. 2, as well as the path of electrons in the electrostatic focusing field. The nonthoriated tungsten filament (0.010 diameter wire $\times$ 1-in. loop) and its shield connected to the electrical leads are shown in Fig. 3 ($3/4$ in. $0.0 \times 3/4$ -in. long) and the entire electron gun assembly with the front cage members removed is illustrated in Fig. 4. The sample is traversed by the molten zone by moving the electron gun while holding the sample steady. Figure 5 shows the interior of the vacuum chamber, the electron gun, and a metal sample.

A vacuum system with an ultimate vacuum of about 3×10^{-7} mm Hg was supplied with the equipment. It consists of a National Research Corporation Type 200S rotary gas ballast type forepump

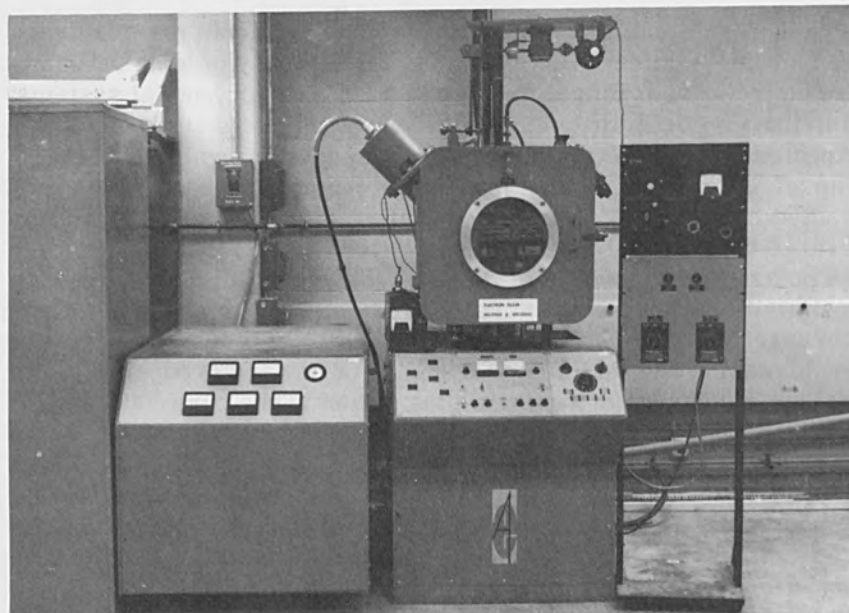


Fig. 1. Electron-beam vacuum chamber and power supply.

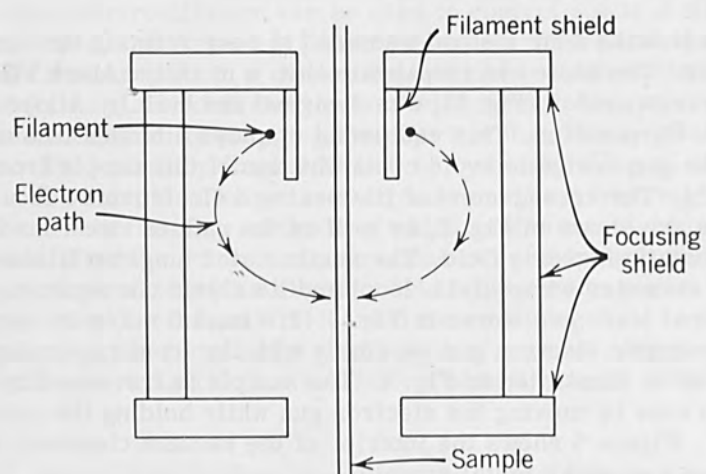


Fig. 2. Hidden filament gun design showing electron path in the electrostatic focusing field.

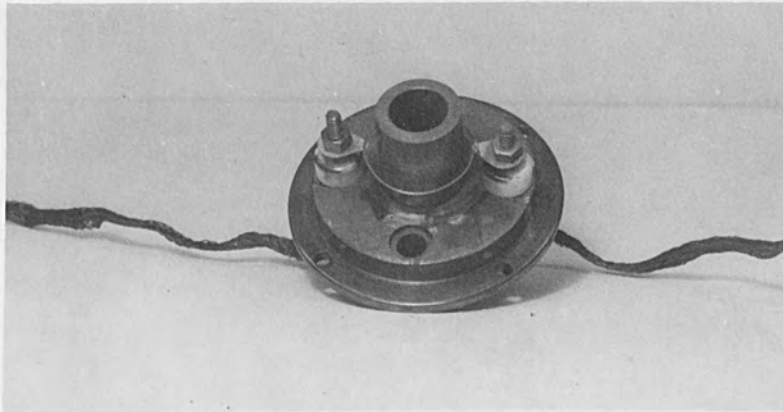


Fig. 3. Filament fixture and assembly showing the filament shield.

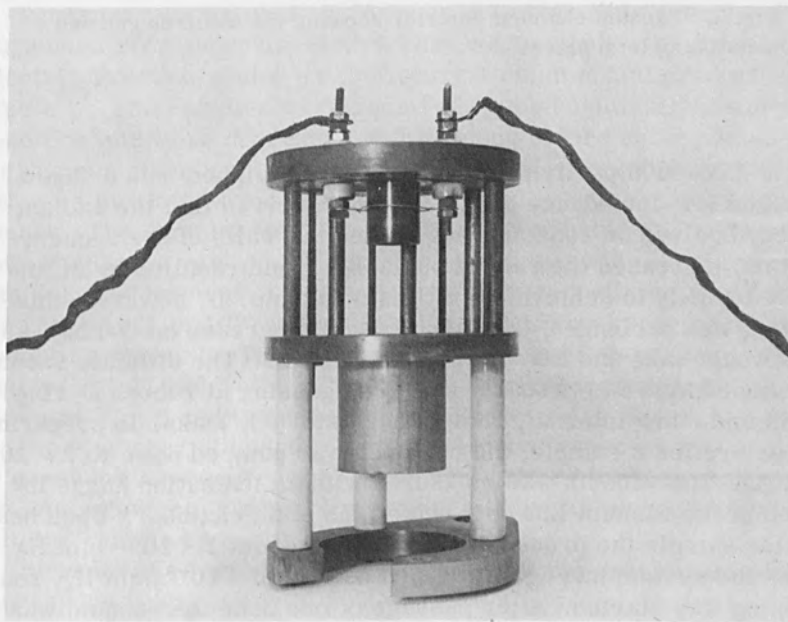


Fig. 4. Electron gun assembly.

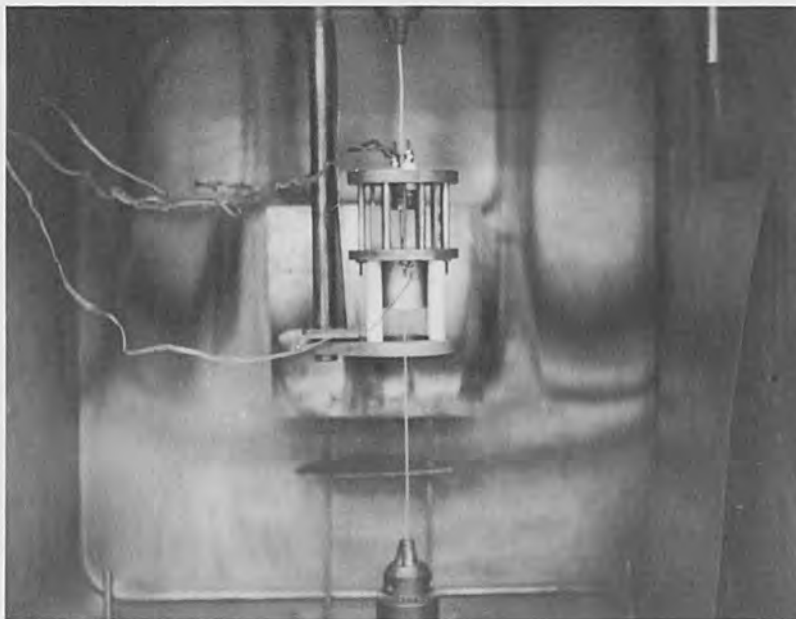


Fig. 5. Vacuum chamber interior showing the electron gun and a metal sample in place.

and a HS6-6500 purifying diffusion pump equipped with a liquid nitrogen low-impedance baffle. It was observed that the radiant energy evolved on zone refining heated the walls of the vacuum system, increased the rate of outgassing, and resulted in an improved ability to achieve the ultimate vacuum. If, however, this heating was prolonged, the vacuum-sealing grease on O-ring gaskets would bake and harden, producing leaks. The ultimate vacuum mentioned above is probably due to outgassing at rubber O-ring seals and other internal, rather than external, leaks. In preparing to zone refine a sample, the chamber was pumped down to 2×10^{-6} mm Hg. The vacuum was measured with an ionization gauge located in the vacuum line just behind the main chamber. Upon heating the sample the pressure increased to about 1×10^{-5} mm Hg. After the system had again pumped down to 6×10^{-6} mm Hg, zone refining was started. After passage of one zone the vacuum was approximately 4×10^{-6} mm Hg. It was necessary to let the chamber cool down between passes. Owing to the outgassing during the

effective "bake-out" while zone refining, the vacuum was 7×10^{-7} mm Hg on starting the second pass, and 3.5×10^{-7} mm Hg on beginning the third pass.

During several zone-refining trials it was noted that the power supply did not deliver a constant level of power. The fluctuations were such that a normally stable zone would suddenly either solidify or be superheated to the point where it collapsed. This is particularly a problem with the small diameter rod employed. It became necessary to monitor the emission current and control it by adjusting the filament temperature. This was accomplished by insulating the sample from its supporting chucks and passing the emission current through a known resistance. The voltage across this resistance was determined experimentally for a stable zone so that, with any fluctuations in the filament temperature, the impedance in the filament circuit was automatically altered to bring the emission current back to the setpoint. This was performed through a feedback arrangement, the details of which are described by Calverley et al. [3]. With a stable emission current the unit did not require constant attention during zone refining. This auxiliary unit is shown mounted on the rack to the right of the vacuum chamber in Fig. 1.

Tungsten welding rods, 1/16 in. in diameter and 14-in. long, were used throughout the study. They were supplied by Sylvania Electric Products and were their purest commercial product (Table I). The samples were received in the chemically cleaned condition and were degreased with acetone before being placed in the vacuum chamber. The ends of the sample were fitted with Transite insulators and then inserted into the water-cooled Jacob chucks. The samples were zone refined by passing the molten zone downwards at a rate of 2.5 mm/min. A travel rate of 2.5 mm/min was selected because it was known to produce resistivity ratios in the range 10^{+4} to 10^{+5} well within the sensitivity range of the resistance apparatus. To produce a molten zone 1/8-in. long required approximately 250 watts. The shape of typical molten zones is shown in Fig. 6 and typical samples illustrated in Fig. 7.

The electric field used to produce electrotransport was applied by passing a current from 12 V storage battery through the sample. An adjustable current limiting resistor was placed in series with the sample as shown in Fig. 8.

Currents of 10 and 20 amps were used in the experiment which correspond respectively to 500 and 1000 amps/cm² in the 1/16-in. diameter samples. The corresponding fields are 0.125 and 0.250 V/cm, which are in the range commonly used in electrotransport studies.

Table I
Lot Analyses of Tungsten Starting Material in
Parts per Million

Element	Samples	Samples	Samples
	1-11	12-40	21-30
Al	2	4	2
Ca	1	1	0.5
Cr	<1	3	1
Cu	<0.1	<0.1	0.3
Fe	3	4	25
Mg	<2	<2	<2
Mo	35	27	35
Ni	<1	4	1
Si	<1	8	5
C		<10	
H ₂	3.9*	<1	3.9*
O ₂	0.4*	7.5	0.4*
N ₂	1.0*	1.8	1.0*

*Typical analyses supplied by Sylvania Electric Products.

Current leads were run through a vacuum seal into the electron-beam chamber and attached to the bottom and top of the sample outside the range of traverse of the electron gun.

Resistivity Ratio Measurements in Liquid Helium

Adequate analytical techniques were not available for the low impurity levels in this study, although a mass spectrometer is now being calibrated. Hopefully, the analysis for both metallic and interstitial elements in the low ppm range will be of sufficient reliability that correlation with physical properties can be reported. In the meantime, purity levels were evaluated by the resistivity ratio method.

Resistivity ratios were determined from the ratio of resistance measurements made at room temperature (298°K) to those made at liquid helium temperature (4.2°K). Since ratios up to 10^5 were expected, the resistance apparatus had to resolve $5 \times 10^{-11} \Omega$ -cm. Voltages as small as 2.5×10^{-8} V over a 10-cm length in 1/16-in.

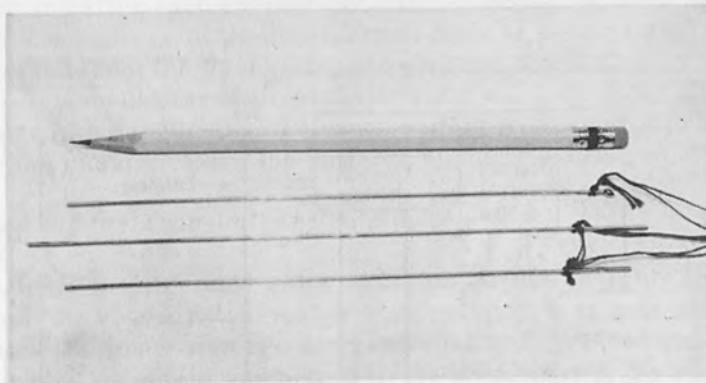


Fig. 6. Zone-refined tungsten samples.

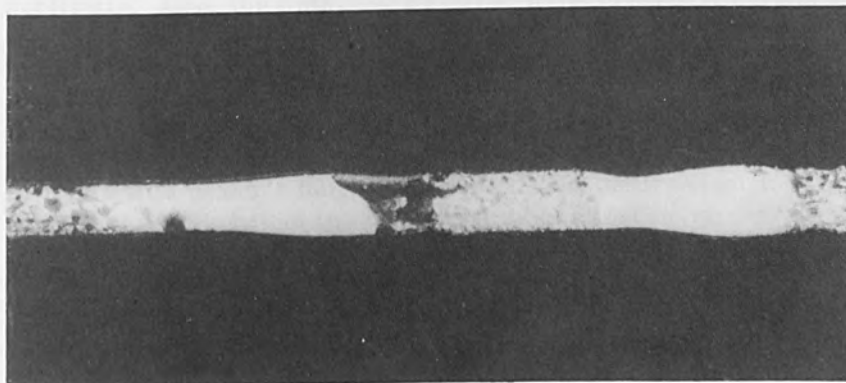


Fig. 7. Typical molten zones.

diameter samples would have to be measured at a current level of 10 amps. A Leeds and Northrup K-6 potentiometer, Cat. No. 7556, with a Keithley microvoltmeter, Model 149, as a null detector, was used for the potential measurement. It has a lower limit of resolution of 1×10^{-8} V. The current was supplied by two 12-V storage batteries in parallel and was measured by observing the

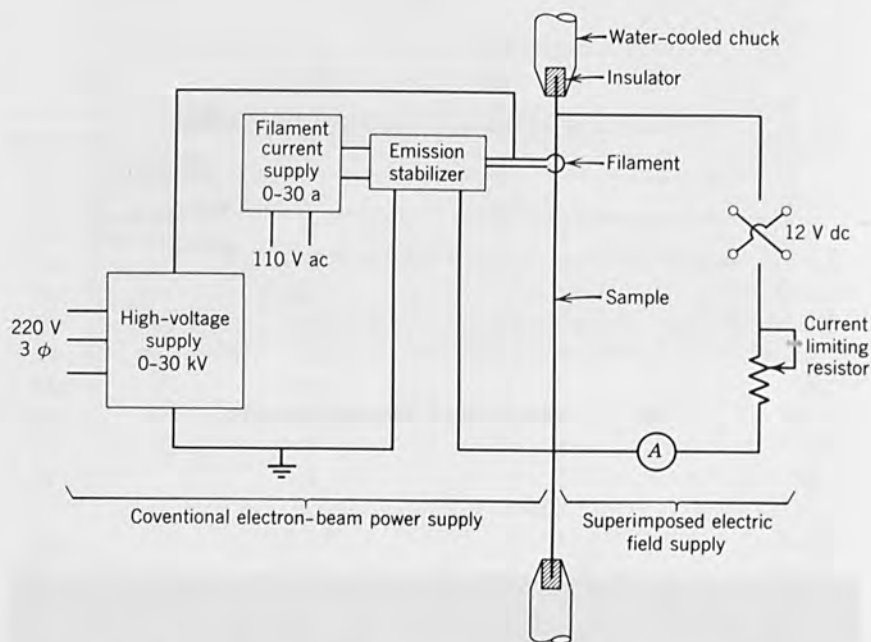


Fig. 8. Schematic diagram of electron-beam power supply and electron field superimposition circuit.

potential drop across a standard resistor in series with the sample.

Leads were connected to the specimens by soldering with high purity indium. Current leads were attached at each end of the zone-refined sample and potential leads 2.5 cm inside each end thus evaluating a 10-cm length. By avoiding the depleted section at the beginning and enriched portion at the end, it was expected that a more uniform and representative evaluation would be observed.

A resistance measurement was made at room temperature and, keeping the contacts in the same positions, the measurement was repeated with the sample immersed in liquid helium. At the low temperature, voltage readings consisted of two components, one a thermal emf and the other due to the sample resistance. To separate the two, a voltage reading was made with zero current

before and after each current-potential combination were measured. The average of the two thermal emfs at zero current was subtracted from the total voltage to give the potential drop due to the resistance. After the resistivity ratio was calculated from resistance, the samples were cleaned with hydrochloric acid to remove the indium leaving the tungsten virtually unchanged.

Analysis of the Zone-Refining Sublimate

In order to determine which of the impurities in Table I were removed by volatilization and by zone refining, a sample of the tungsten sublimate was prepared and analyzed. A clean stainless steel radiation shield was placed in the lower part of the electron gun to intercept the tungsten and impurity vapors. Sufficient material was deposited after three passes to be submitted for analysis. The film was nonadherent and could be removed by tapping the shield, thus minimizing contamination from the shield itself.

The sample was run on an emission spectrograph. A qualitative analysis revealed Al, Ca, Cu, Cr, Fe, Si, Mg, Ti, and Zn in the sublimate. Also, the following appeared to be present but could not be confirmed due to the complex nature of the tungsten spectrum: Pb, Mo, Mn, and Ag. If they are present, they are in such small quantities as to be just at the detection limit of the emission spectrograph. Ti, Zn, Pb, and Mn were not reported as present in the starting material and were probably below the spectrographic detection limits in these tungsten samples. The analyses show that a portion of a majority of the contaminants are lost by vaporization.

EXPERIMENTAL RESULTS

Effect of the Electric Field

Standard specimens were prepared without an electric field for comparison with those refined under the field-aided condition. A series of samples were given one, two, and three zone passes (samples 13, 1 and 2, and 16 respectively). The conditions under which all samples were zone refined are listed in Table II with their resistivity ratios.

To observe the effect of field polarity, samples 24 and 8, one and two zone passes, respectively, were prepared with a field of 0.125 V/cm parallel to the direction of traverse. Samples 22, one zone pass, and 9 and 10, two zone passes, were zone refined with

Table II
Operating Conditions and Resistivity Ratios of the Tungsten Samples

Sample Number	Number of Passes	Operating Vacuum 6×10^{-6} mm Hg	Current Amps	Speed mm/min	Resistivity Ratio
1	2		0	2.5	37,400
2	2	3	0	2.5	37,700
8	2	5	+10	2.5	29,000
9	2	5	-10	2.5	45,600
10	2	1	-10	2.5	45,300
13	1	1	0	2.5	24,900
16	3	4	0	2.5	40,000
18	2	2.5	-21	2.5	56,300
19	0	0	0	2.5	72
21	1	5	-20	2.5	41,260
22	1	5	-10	2.5	36,300
23	3	5	-10	2.5	49,700
24	1	5	+10	2.5	20,800
25	3	5	-20	2.5	60,900

the field antiparallel (-0.125 V/cm) to the travel direction. The resistivity ratios of these materials are listed in Table III. They show that a negative electric field increases the efficiency of zone refining.

The intensity of the electric field was increased to -0.250 V/cm in the antiparallel direction to observe the effect of electric field intensity on purity. Samples 21, 18, and 25 were prepared with one, two, and three zone passes, respectively. Their resistivity ratios are compared with those produced with zero field and -0.125 V/cm in Table IV. The effect of field intensity on zone-refining efficiency at three stages of zone refining is shown in Fig. 9. Increasing the magnitude of the electric field intensity, E , increases zone-refining efficiency.

The results are summarized in Fig. 10, in which the purity level (resistivity ratio) is shown as a function of the number of zone passes for two levels of electric field and for both polarities.

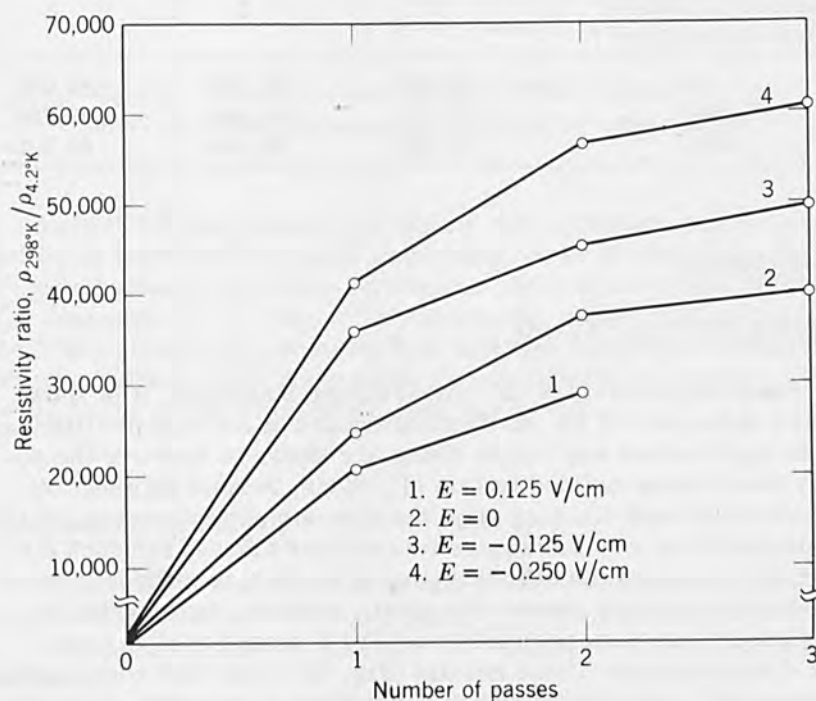


Fig. 9. Effect of electric field intensity on purity of three stages of zone refining.

Table III

Effect of Polarity of Electric Field on the Purity in
Terms of the Resistivity Ratio

Number of passes	1	2
Parallel field	20,800	29,000
No field	24,900	37,600
Antiparallel field	36,300	45,400

Table IV

Effect of Intensity of Electric Field on the Purity
in Terms of the Resistivity Ratio

Number of passes Current (amp/cm ²)	1	2	3
0	24,900	37,600	40,000
500	36,300	45,400	49,700
1000	41,300	56,900	60,900

DISCUSSION

Vacuum and Time Effects

From the analysis of the zone-refining sublimate, it is shown that a large part of the purification occurs by vacuum distillation. Both high vacuum and longer times are known to improve the purity level of zone-refined metals [4]. Since, in zone refining, diffusion is the rate limiting step, the ultimate purity level might be expected to vary as the square root of time allowed for such diffusion. Assuming the diffusion time to be directly proportional to the number of zone passes, the purity levels as represented by resistivity ratios were found to best fit a square root of time ($\sqrt{t}$) relationship. These results (Fig. 11) imply that zone refining does significantly contribute to purification of tungsten, since loss of impurities by volatilization would vary linearly with time in the presence of good stirring in the molten zone. Since tungsten has

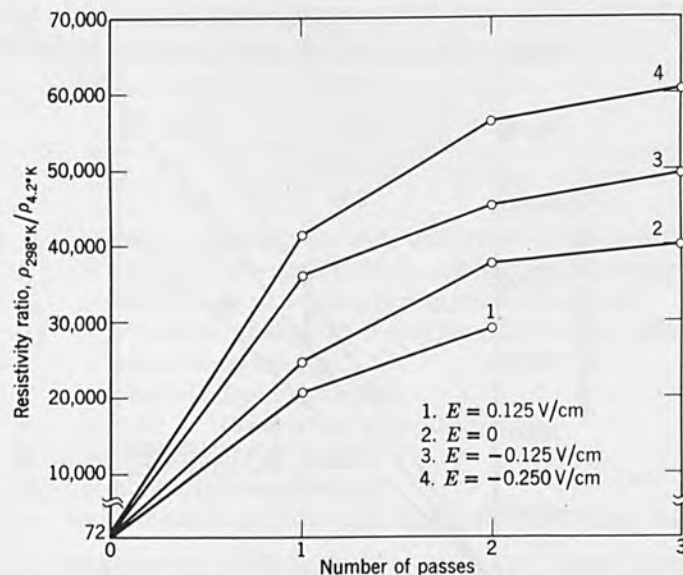


Fig. 10. Summary of experimental results showing the effect of electric field intensity and polarity on the purity.

the highest melting point of the metals and represents extreme conditions of superheat for most impurities, other investigators [5], [6] have assumed that zone-refining action is small or negligible. The dependence of purity on the square root of time and on the electric field both demonstrate that applying the principles of zone refining aided by field freezing to the purification of tungsten will result in the preparation of a higher purity metal.

Electrodifusion Effects

The details of electrodifusion and its effect on zone refining have been described by Hay and Scala [7], [8]. A summary is introduced here because of the good correlation between experiment and theory.

Under equilibrium conditions, the efficiency of zone refining with respect to a given impurity can be expressed by an equilibrium distribution coefficient

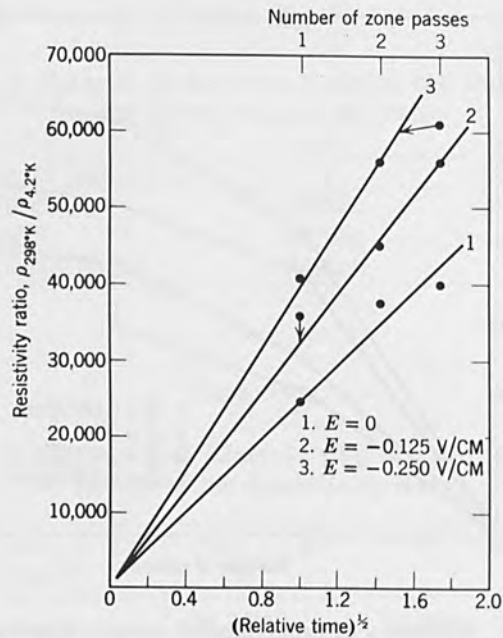


Fig. 11. Relation between purity level and the square root of the time in the molten state.

$$K_0 = \frac{C_s}{C_l}$$

where C_s is the equilibrium concentration of impurity in the solid, and C_l is the equilibrium concentration of impurity in the liquid at the solidifying interface. Under actual freezing conditions, however, complete equilibrium is not achieved and an effective distribution coefficient, K , is defined where $K_0 < K \leq 1$, for $K_0 \leq 1$. The limited data on tungsten alloy systems [9] show K_0 to be less than or equal to unity for its major contaminants. Of the impurities listed in Table I aluminum, chromium, iron, molybdenum, nickel, silicon, carbon, and nitrogen have distribution coefficients of less than or equal to unity. These solutes will tend to be collected in the molten zone and deposited in the tail end of the sample. The

effect of electric field intensity and polarity on the distribution coefficient of impurity is given by [7], [8]:

$$K_i = \frac{1 - \frac{D_i e E Z_i^0}{v k T}}{1 + \frac{1}{K_{oi}} \left(1 - \frac{D_i e E Z_i^0}{v k T} - 1 \right) \exp \left(\frac{t e E Z_i^0}{k T} - \frac{v t}{D_i} \right)}$$

where K_i = effective distribution coefficient of impurity
 K_{oi} = equilibrium distribution coefficient of impurity
 D_i = diffusivity of impurity i in molten solvent
 Z_i^0 = effective charge of impurity i in molten solvent
 e = electronic charge
 E = electric field intensity
 v = rate of transverse of molten zone
 k = Boltzmann's constant
 T = absolute temperature
 t = thickness of diffusion layer at solidifying interface

However, data such as diffusivities in the liquid state, distribution coefficients at very low concentrations, and effective valences of metals in liquid solution are not available for refractory metal systems. It was interpreted from the experimental results (Table III) that in tungsten transport of the majority of the impurity ions was anode-directed. These results are in agreement with those of Frantsevich et al. [10], [11] who observed in the solid state that migration of molybdenum and iron, the two major contaminants in the starting tungsten, was anode-directed. Thus, their effective valences are negative. When the intensity of an applied negative electric field is increased, the decrease in K_i from equation 1 becomes more pronounced and zone-refining efficiency is increased (Table IV). Thus, the results are in agreement with theoretical relationships (equation 1) and suggest the further investigation of (1) the nature of solute behavior, and (2) the potentially higher purities, achievable with (a) better starting materials, (b) more effective vacuum techniques, and (c) slower zone-refining rates.

CONCLUSIONS

The technique of field freezing was successfully applied to electron-beam zone melting and resulted in a significant increase in the efficiency of zone refining. Although detailed chemical analyses of the tungsten were beyond available techniques, based on

resistivity ratio results it has been shown that the method permits the removal of greater amounts of impurities than by zone refining alone for the same number of passes. A higher degree of control is obtained over the entire zone-refining process.

The experimental results also demonstrate that both the zone-refining action and the purification by vacuum distillation play important roles in high melting-point systems. With field freezing superimposed, a higher rate of purification may be obtained than with conventional zone refining alone with the potential for higher purities.

REFERENCES

1. W. G. Pfann and R. S. Wagner, *Trans. AIME*, **224**, 1139 (1963).
2. R. Tongas and A. Hone, *Can. Met. Quart.* **2** (3), 281 (1963).
3. A. Calverley, M. Davis, and R. F. Lever, *J. Sci. Instr.*, **34**, 142 (1957).
4. E. Rexer, *Proc. 4th Electron Beam Symp.* (R. Bakish, Ed.), Boston, Mass., 1962.
5. H. W. Schadler, *Proc. 1st Electron Beam Symp.* (R. Bakish, Ed.), Boston, Mass., 1959.
6. I. Dragel and G. T. Murray, *Proc. Symp. on High-Temp. Refractory Metals*, AIME, New York (1964).
7. D. R. Hay and E. Scala, "Refining Tungsten by Electrodiffusion," presented at AIME Annual Meeting, Feb. 17, 1964, New York, N.Y. To be published.
8. D. R. Hay, Masters thesis, Dept. Eng. Phys. and Mat'l. Sci., Cornell University, Ithaca, N.Y.
9. J. J. English, DMIC Report 152, April 28, 1961.
10. I. N. Frantsevich and I. I. Kovensky, *Dokladi Akad. Nauk Ukr. R. S. R.*, **9**, 1169 (1961).
11. M. D. Smolin and I. H. Frantsevich, *Fizika Tverd Tela*, **3**, 2115 (1961).

A MULTIPURPOSE, HIGH-TEMPERATURE TENSILE-TESTING MACHINE USING ELECTRON-BEAM HEATING

INTRODUCTION

Mechanical testing machines are required nowadays for multipurpose use in the temperature range between cryogenic temperatures and 3000°C. The design of any such machine will require the precision measurement of load, strain, and strain rate together with an accurate control of temperature.

A sensitive electronic weighing system, employing inertialess weighing for the detection and recording of applied load to the test sample, is best suited for a versatile mechanical testing machine. Such a weighing system is suitable for accurate autographic recording of load. The mechanical application of load is most accurately controlled through a screw mechanism driven by a positionally controlled servo mechanism. Machines of this design are currently available and will not be considered in detail.

The attainment of temperatures of 3000°C, and above, presents considerable design problems as illustrated by the variety of equipment used. Few solid materials appear suitable for use at temperatures above 2000°C—namely graphite, tungsten, and stabilized zirconia.

HEATING METHODS

Resistance heating has many disadvantages at temperatures around 2500°C. A constant current control must be used with oxide heaters (Keith, 1952) and the use of these heaters is not suitable for short heat treatment cycles (Davenport, 1950). Wire sag, sheet creep, and fabrication difficulties limit the usefulness of

*Department of Metallurgy, University of Cambridge, England.

tungsten at these temperatures (Alderman, 1950), (Randall, 1952).

The only resistance element suitable for temperatures up to 3000°C is that of graphite, but it has the disadvantages of long preheating times, danger of contamination of sample from volatiles, difficulties in operating at pressures above 1 atmos. to reduce carbon evaporation, and high stresses imposed on the element due to the fixed electrodes (Blackman et al., 1961). Despite these disadvantages, however, a number of units have been successfully adapted for use with mechanical testing machines (Green, 1960). These designs require large power input and long hot zones.

From a consideration of the radiation losses at temperatures above 2000°C, a sensible design would be one having a small hot zone. Heating with an RF induction coil or electron bombardment enables a large amount of energy to be concentrated into a small volume. Difficulties in gripping tensile specimens at high temperatures may be overcome by heating only the gauge length. Similar requirements for high-power density are to be found in welding and zone melting. The ease with which electron beams may be focused is a distinct advantage in obtaining selected area heat treatment. A consequence of this type of heating is its high heating rate thus eliminating long preheating programs.

For a versatile mechanical testing machine it is necessary that tensile tests, compression tests, bend tests, and creep tests be carried out with ease using the same furnace. With electron bombardment this is readily achieved as the heat is generated directly in the specimen from the same electron-beam source independent of the type of test. As a consequence of the lower power requirements the need for heavy water-cooled electrodes and busbars is eliminated. Using an eddy current concentrator to focus the energy of a magnetic field, similar power concentration to that of electron bombardment can be obtained (Johnson, 1963). However this method cannot seriously compete with the flexibility of electron beams.

An important question to ask is what materials require testing at these high temperatures? Basically it is the refractory metals, the carbides of these metals, and graphite. These materials react with oxygen even at small partial pressures, thus necessitating the use of high vacuum. Diefendorf (1960) has shown that some properties of graphite are dependent on the atmosphere used during testing. A vacuum of less than 10^{-3} torr is necessary for the effective operation of an electron beam and is thus ideally suited for the testing of these materials up to their melting points. The only restriction is that the sublimation temperature be not less than the melting point or the vapor pres-

sure greater than 10^{-3} torr. In the case of graphite the vapor pressure is about 6×10^{-3} at 2700°C (Pitzer and Clementi, 1959) which puts an upper limit on the testing temperature. By using separately pumped electron guns, higher furnace chamber pressures could be tolerated than 10^{-3} torr. Induction heating is suitable for high temperature testing in protective atmospheres at a pressure above 1 atm., or in a vacuum better than 10^{-3} torr, but difficulties involving sparking breakdown occur at intermediate pressures (Blackman et al., 1961).

The accurate control of temperature is always difficult at temperatures in excess of 2000°C , but it is a necessary requirement for accurate mechanical testing. Beirne (1961) claimed an absolute accuracy of $\pm 23^{\circ}\text{C}$ using a simple automatic temperature control in an electron-bombardment furnace. However, Johnson (1963), using an RF eddy current concentrator, maintained temperature manually within what is called "necessary limits." Stable automatic temperature control of RF heating can, however, only be achieved with great difficulty.

The apparatus to be described consists basically of a furnace mechanically attached to the moving crosshead of an Instron Tensile Testing Machine, an adequate vacuum system, and a power source capable of controlled electron bombardment.

Vacuum System

A vacuum system, with a pumping speed of 300 liters/sec at the baffle valve and a speed of 93 liters/sec in the vicinity of the electrodes, giving a pressure of 5×10^{-6} torr, is obtained using a 6-in. oil diffusion pump and a cold trap. This pumping speed is found to be sufficient even though it is below the 10 liters/sec per liter of pumped volume suggested by Holland (1950). An oil diffusion pump backed by a liquid nitrogen trap to prevent back streaming permits a higher pumping speed than would be possible with the same size mercury pump, the higher pumping speed being more important than a low ultimate vacuum. Provision is made for the inclusion of a molecular sieve pump to eliminate contamination from the oil if so required. The bombardment chamber is a flanged water-cooled 6-in. diameter $\times$ 10-in. copper cylinder with the provision of a silica window to view the electrode assembly. A second silica window is provided to eliminate evaporated deposits on the outer window. A door is provided to facilitate entry into furnace. Outgassing from copper is considered insignificant when testing small graphite specimens. A flap valve is incorporated which enables

the chamber to be let down to air and subsequently evacuated without switching off the vacuum pumps. Furnace flange seals are of indium and the remainder viton "O" rings. The pressure is measured by an ion gauge attached to the top furnace plate.

Electrode System (Fig. 1)

As with Calverly et al. (1957), it is found convenient to ground the specimen and run the cathode at a high negative potential. Also the whole electrode assembly is hung from the stainless steel plate (A). The cathode (B) is of 0.5-mm tungsten wire shaped in

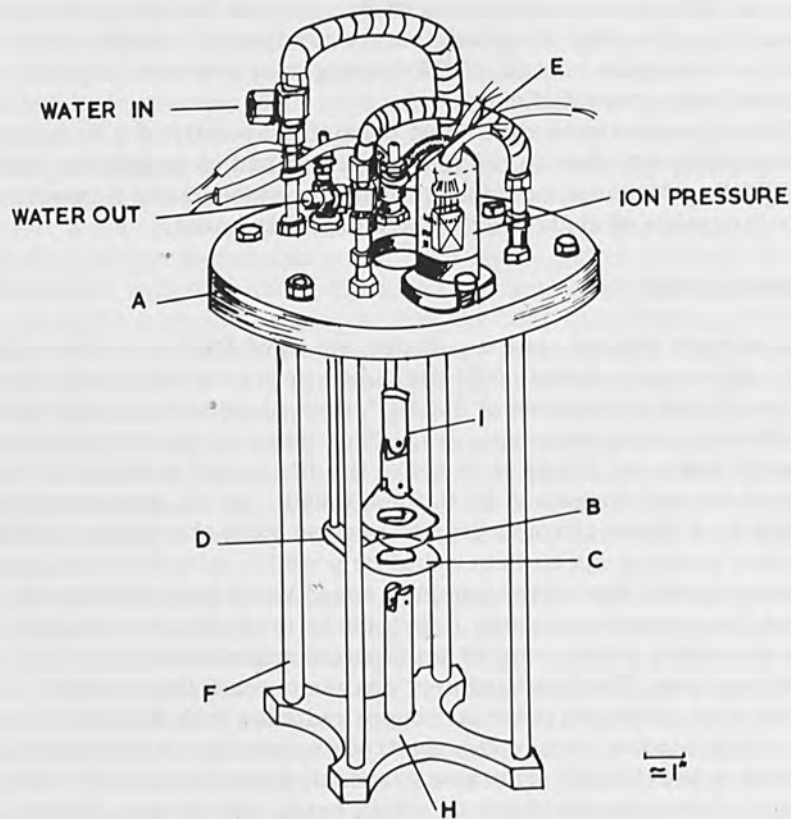


Fig. 1. Sketch of water-cooled tensile cage with filament cathode and focusing plates.

the form of a loop. Molybdenum focusing shields (C), at the same potential as the filament, are spaced 10 mm apart and connected to the filament terminal blocks (D). Vacuum-sealed electrodes (E) carry the cathode current to the filament. This arrangement gives a uniform temperature zone of $\pm 5^\circ\text{C}$ over 3 to 5 mm on small graphite single crystals. For specimens requiring gauge lengths of 1.5-cm uniform hot zone, a vertical tantalum tape may be used. Harris (1962) has shown (Fig. 2) that such a cathode gives a central hot zone of 2.5 cm with a variation of less than 50°C on Niobium

HIGH TENSION TERMINALS

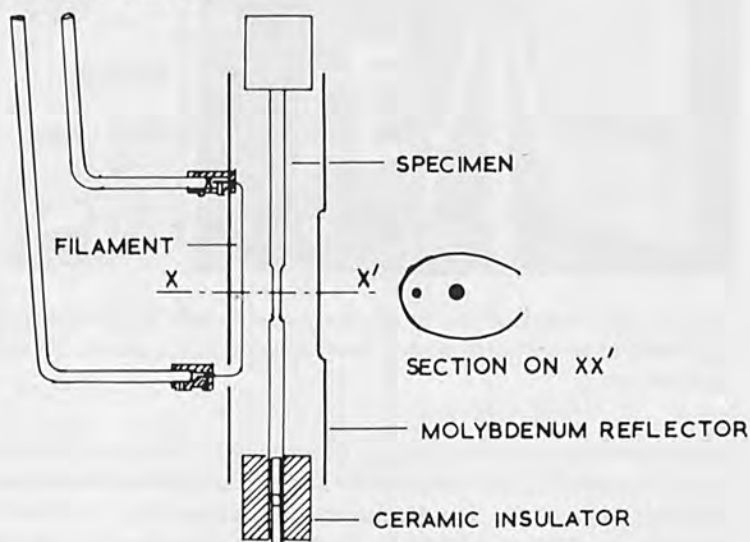


Fig. 2. Ribbon cathode and focusing shield (after Harris, 1962).

crystals 5 mm in diameter and by adding a small loop at each end of the cathode a more uniform central zone is obtained.

Tests on single crystal graphite, with the basal plane normal to the filament, show that a uniform hot zone of greater than 3 mm is obtainable. The size of the uniform hot zone will be limited not only by the cathode design but also by the gauge section and thermal conductivity of the material.

Test Arrangements

Figure 3 shows the general layout of the loading machine (A), furnace (B), vacuum system (C), power source (D), control panel (E)



Fig. 3. (A) General view of tensile machine, (B) electron-beam furnace, (C) vacuum system, (D) power source, (E) control panel, (F) and optical pyrometer.

and temperature measuring pyrometer (G). Figure 4 gives a diagrammatic layout of these components and in particular the method used to support the furnace. The stainless steel plate (Fig. 1) of the furnace chamber is attached rigidly to the moving crosshead (1) of the Instron tensile machine. The bottom flange of the chamber is attached to the vacuum system through the copper bellows (2). To compensate for the vacuum load thereby imposed on the moving crosshead and screw bearings of the machine, two copper bellows (3) of combined equivalent area to (2) are connected between the main frame (4) and the moving crosshead (1). Similar compensation is used for the bellows connecting the load cell (5) and loading arm (6) with the furnace chamber. By means of the magnetic valves (7) and (8), and the equalizing valve (9) simultaneous evacuation of the chamber and compensating bellows is obtained. Similarly by use of valve (10) air can be admitted. All testing rigs are attached to the top plate enabling the use of one furnace chamber.

Tension Rig (Fig. 1). This is basically a polanyi cage (F), water cooled to eliminate thermal expansion of the rods heated by radia-

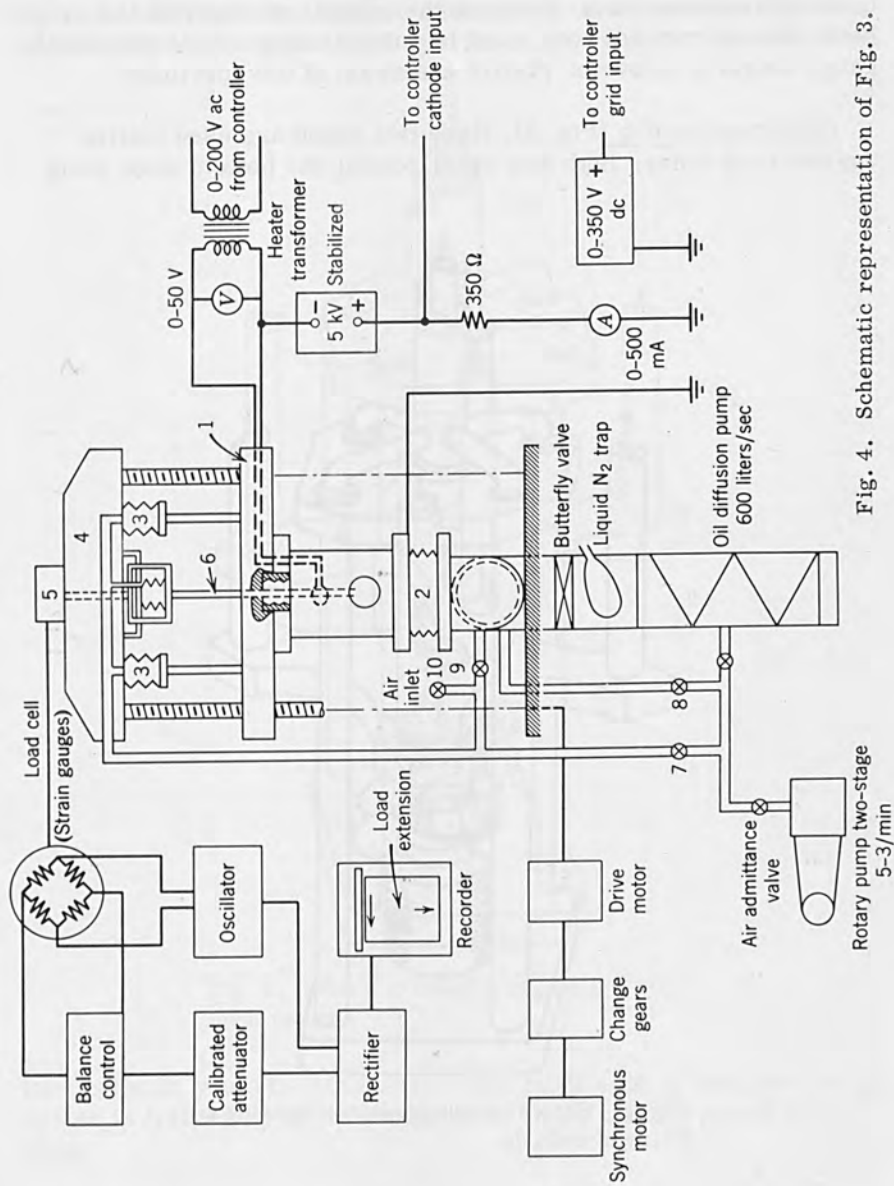


Fig. 4. Schematic representation of Fig. 3.

tion. One end of the tensile specimen is connected to the cage (H) and the other to the load measuring cell. Universal joints (I) are used in the connections. Because the cathode moves with the cross-head, the uniform hot zone must be initially longer than the tensile gauge length to allow for plastic extension of the specimen.

Compression Rig (Fig. 5). Here two cages are used pulling against each other. Both are water cooled, the bottom cage being

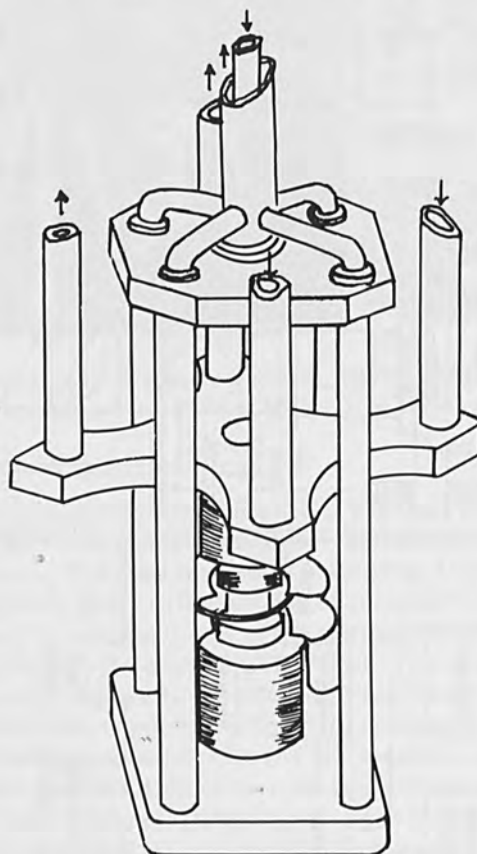


Fig. 5. Sketch of compression rig using a filament cathode.

connected to the load cell. The compression anvils could be made of cellulose carbon or graphite. Cathode arrangement is similar to the tensile rig.

Bending Rig (Fig. 6). This design is similar to the compression rig except that the anvils are replaced by either a 3- or 4-point graphite bend jig. The cathode filament could be focused either from above or below the bend specimen. For thicker specimens a

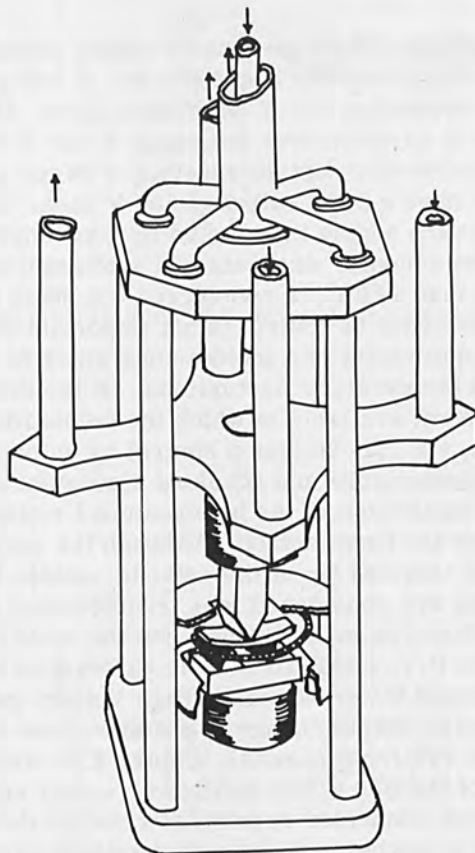


Fig. 6. Sketch of bending rig using a filament cathode.

tape cathode would be necessary. Maximum size of specimen to be heated is limited by its thermal conductivity and the power available.

Zone Refining. Calverly (1963) suggested the possibility of using this design as a zone refiner. This would be particularly useful in

preparing pure specimens for mechanical testing. The standard rates of crosshead movement available are 0.005 – 10 cm/min; however, vibration may present some problems.

Electrical System. The high-tension supply provides a fully stabilized dc voltage over the range 0-5 kV, at 500 mA, derived from 10 series connected 500-V stabilized units. The lowest supply in the chain is variable over the range 0-500 V in steps of 250 V with a fine vernier control covering a sweep of 250 V. The other nine units have a fixed output of 500 V each. The junction or tapping point of each supply is selected by a switch mounted on the front panel of the console. Accurate and automatic control of temperature better than $\pm 20^\circ\text{C}$, is considered essential for mechanical testing. This order of control is not important in zone refining as the major requirement is a molten zone which has a latent heat sink to even out temperature fluctuations. Of the two methods of temperature control available in which the bombardment current is kept constant, viz., (1) emission control by automatic adjustment of the cathode temperature and (2) using a constant current source with automatic adjustment of the bombardment voltage, it is found convenient to use the first method. Although the speed of response is limited by the thermal lag of the cathode, sudden fluctuations due to outgassing are considered less troublesome in a heat treatment zone than found in molten zones. As the other variable is fluctuation of the H.T. a stabilized H.T. supply was chosen. This supply unit designed by Brandenburg High Voltage gave a ripple of 10-mV peak, and an output change of not more than 0.1% against a mains change of 10% from nominal. Figure 7 shows the circuit diagram for one of the 500-V, 500-mA power supply units. The six EL 34-type valves connected in parallel regulate the current into the load in such a manner that the output voltage remains constant for varying values of load resistance. Control on the grid bias is obtained from a signal proportional to the difference between a sample of the output voltage and a reference voltage on the grids of the V 7 valve (type ECC 83). The variable supply is of necessity more complex in its arrangement.

The emission control circuit, Fig. 8, is identical with that used by Calverly et al. (1957). Basically, a voltage proportional to the emission current through a resistance is compared with a reference voltage from a stabilized supply. The amplified difference signal controls the filament power through two thyratrons.

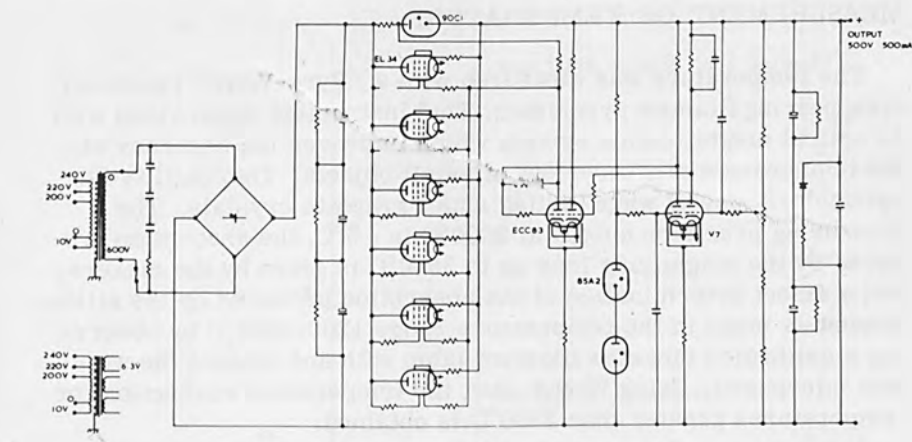


Fig. 7. Circuit diagram for 500 V, 500 mA stabilized power supply.

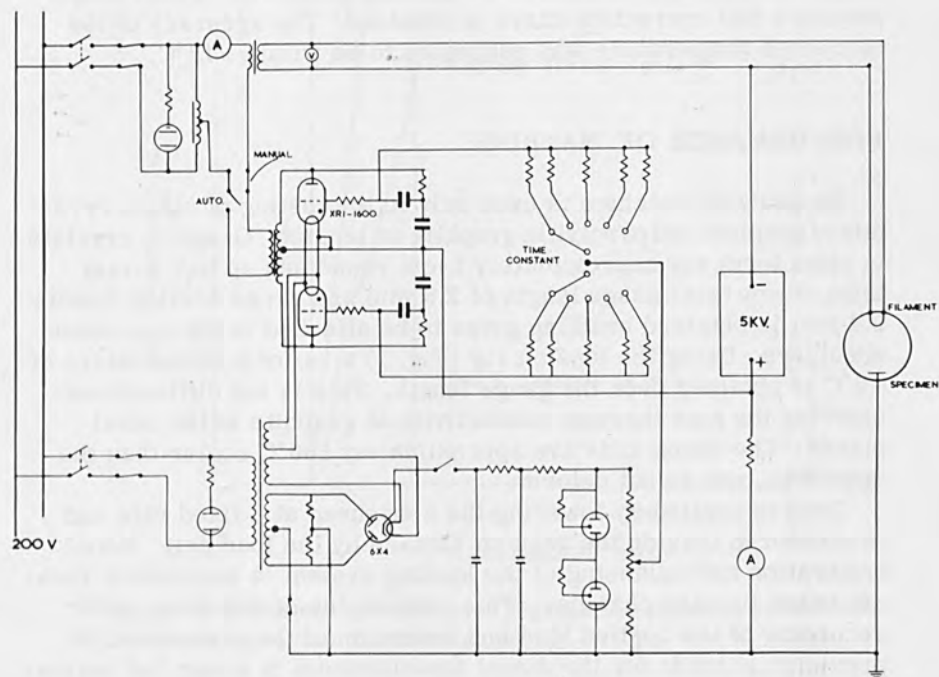


Fig. 8. Emission control circuit (after Calverly et al., 1957).

MEASUREMENT OF TEMPERATURE

The temperature was measured with a "Pyro-Werk" precision disappearing filament pyrometer. This instrument is provided with an optical magnification system which improves the accuracy of the temperature measurement of small objects. This facility is particularly useful when testing small graphite crystals. The measuring precision quoted at 2000°C is $\pm 5^\circ\text{C}$, the absorption produced by the magnifying lens up to 3000°C is given by the makers, and a direct determination of the absorption produced by the silica window is made in the temperature range 1200-2300°C by observing a calibrated tungsten filament lamp with and without the window interposed. Using Wiens' law, the temperature correction for temperatures greater than 2300°C is obtained.

Readings are also corrected for emissivity. In the case of commercial graphites the emissivity is well known for various surface conditions, but this is not the situation with graphite single crystals and a determination of the apparent surface temperature is compared with that of known commercial graphites. From these results a full correction curve is obtained. The accuracy of the corrected temperature was estimated to be within $\pm 12^\circ\text{C}$.

PERFORMANCE OF MACHINE

To date this machine is used only in the testing of single crystals of graphite and pyrolytic graphite in tension. Graphite crystals in plate form are approximately 1-cm round and up to 0.5-mm thick. From this a gauge length of 2.5 mm and cross section 1 mm $\times$ 0.4 mm is obtained enabling grips to be attached to the specimen shoulders. Using the tension rig (Fig. 1) a uniform temperature of $\pm 5^\circ\text{C}$ is obtained over the gauge length. This is not difficult considering the high thermal conductivity of graphite in the basal planes. The chuck ends are approximately 100°C cooler than the specimen, and do not deform.

Load is applied by lowering the crosshead at a fixed rate and is measured outside the vacuum furnace by the load cell. Initial calibration and balancing of the loading system is done under vacuum in the furnace chamber. This enables the direct autographic recording of the applied load and extension of the crosshead. No provision is made for the direct measurement of strain but various systems are available (Green, 1960).

Figure 9 shows a load versus crosshead extension curve for a

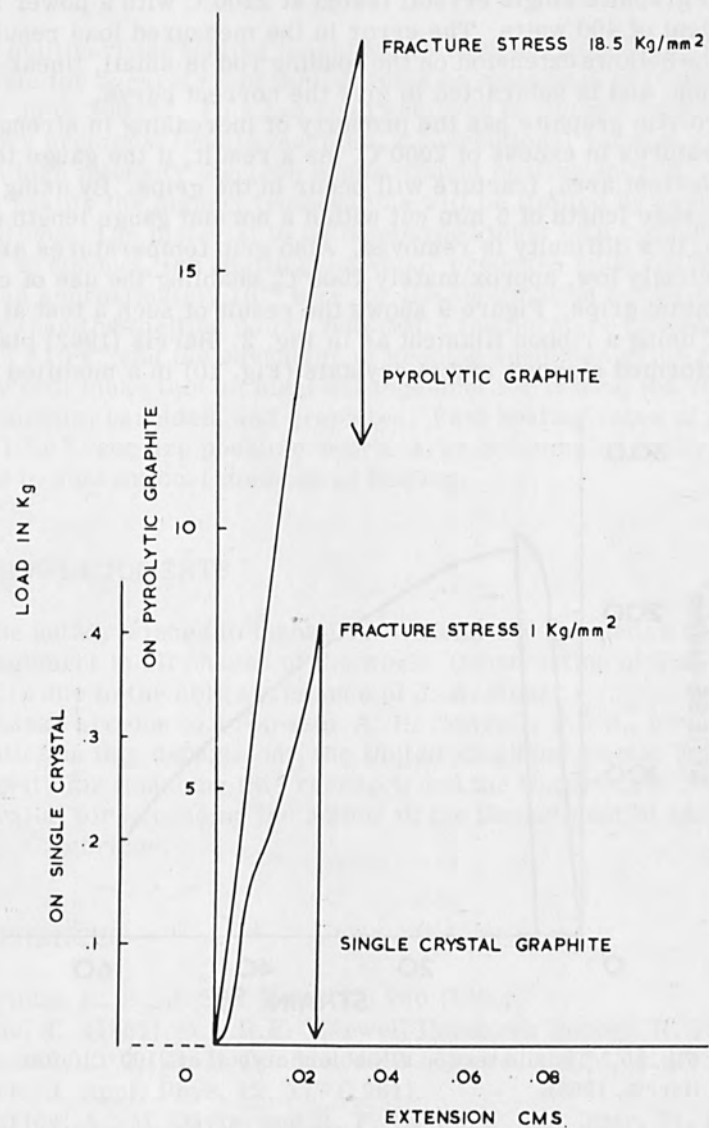


Fig. 9. Load-extension curves for a graphite single crystal at 2200°C and pyrolytic graphite at 2400°C.

natural graphite single crystal tested at 2200°C with a power requirement of 400 watts. The error in the measured load resulting from the bellows extension on the loading rod is small, linear with extension, and is subtracted to give the correct curve.

Pyrolytic graphite has the property of increasing in strength at temperatures in excess of 2000°C. As a result, if the gauge length is the hottest area, fracture will occur in the grips. By using a small gauge length of 5 mm cut within a normal gauge length of 25 mm, this difficulty is removed. Also grip temperatures are correspondingly low, approximately 1500°C, enabling the use of carbon or ceramic grips. Figure 9 shows the result of such a test at 2400°C using a ribbon filament as in Fig. 2. Harris (1962) plastically deformed niobium single crystals (Fig. 10) in a modified zone

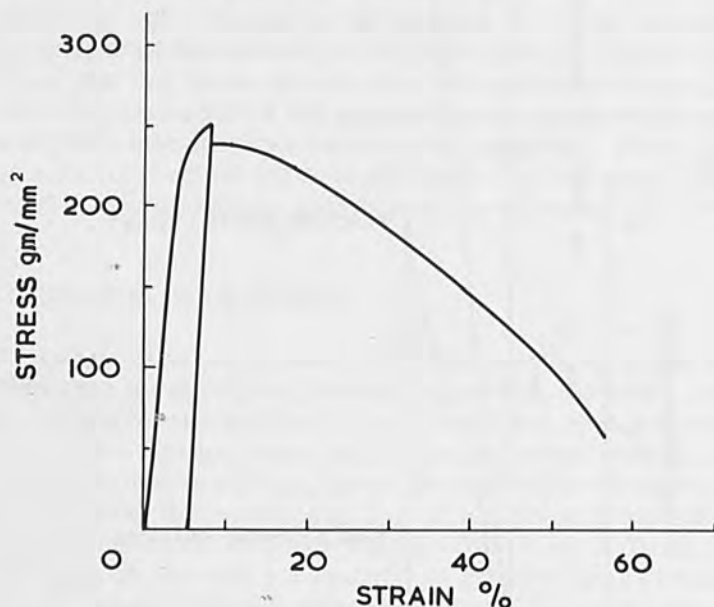


Fig. 10. Tensile test on a Niobium crystal at 2100°C (after Harris, 1962).

refiner at 2100°C. The initial pressure of the present system is 5×10^{-6} torr, but this pressure rose to 5×10^{-4} torr during operation. During a 1 hour annealing operation of single crystal graphite at 2200°C, a uniform temperature was maintained within $\pm 10^\circ\text{C}$.

CONCLUSIONS

A multipurpose testing machine has been developed which is suitable for accurate mechanical tests on materials at temperatures up to 3000°C. This has been achieved by using a typical laboratory precision tensile machine, a conventional vacuum system, and a high intensity electron bombardment heating source.

In reality the highest temperature will be limited by several conditions. The material to be bombarded must be a conductor or made to conduct, the equilibrium vapor pressure must not be greater than approximately 10^{-3} torr, or the sublimation temperature lower than the melting point. Automatic temperature control to within $\pm 10^\circ\text{C}$, and the advantage of keeping specimen grips relatively cool make this an ideal arrangement for testing the refractory metals, carbides, and graphites. Fast heating rates of greater than 1000°C/sec are possible with a large economy in power compared to conventional methods of heating.

ACKNOWLEDGMENTS

The author wishes to thank Dr. A. Kelly for his advice and encouragement in all phases of the work. Construction of this equipment is due to the able assistance of J. A. Stuart.

Thanks are due to Professor A. H. Cottrell, F.R.S., for use of facilities in this department, the United Kingdom Atomic Energy Authority for financing this research and the Department of Supply, Australia, for seconding the author to the Department of Metallurgy, Cambridge.

REFERENCES

- Alderman, K. B., *J. Sci. Instr.* **27**, 280 (1950).
- Beirne, T. (1961), A.E.R.E. Harwell Research Report, R. 3579.
- Blackman, L. C., P. H. Dundas, A. W. Moore, and M. A. Ubbelohde, *Brit. J. Appl. Phys.* **12**, 377 (1961).
- Calverley, A., M. Davis, and R. F. Lever, *J. Sci. Instr.* **34**, 142 (1957).
- Calverley, A. (1963), private communication.
- Davenport, W. H., S. S. Kistler, W. M. Wheildon, and O. J. Whitemore, *J. Am. Ceram. Soc.* **33**, 333 (1950).
- Diefendorf, R. J. (1960), *Proc. 4th Carbon Conference*, Buffalo, N.Y.

- Green, L., Jr., 1960, "Analytical Methods on the Study of Stress Strain Behaviour," Symp., Boston, Mass.
- Harris, B. (1962), Ph.D. dissertation, University of Cambridge.
- Holland, L. (1950), "Vacuum Deposition of Thin Films," Chapman and Hall, London, England.
- Johnson, R. W., J. Appl. Phys. 34, No. 2. 352 (1963).
- Keith, W. P., Bull. Am. Ceram. Soc., 31, 76 (1952).
- Pitzer, K. S., and K. Clementi, J. Am. Chem Soc., 81, 4477 (1959).
- Rendall, J. H., J. Sci. Instr., 29, 248 (1952).

ELECTRON BEAMS IN MATERIALS JOINING

The application of electron beams in welding dates back approximately ten years to Stohr of the French Atomic Commission, followed by Wyman in the United States. Siegerwald, at the same time in Germany, initiated microarc welding research with this new heat source.

Section V

ELECTRON BEAMS IN MATERIALS JOINING

In the past few years have seen a rapid growth in the use of electron beams for welding. This growth has been reflected by the transition from the research laboratory to the production shop. In the past few years we have seen a substantial increase of power levels in electron beam welding installations from 1 to 2 kW up to 10 to 40 kW, an almost twenty-fold increase. We have further seen the beginning of, and some growth in, automation of the process. Last but not least, a considerable amount of this technique in industry has taken place.

In 1962 we saw initiation of efforts to bring the electron beams for joining out of the vacuum. Today we have achieved reasonable success in welding with electron beams in inert atmospheres. We are working to develop electron guns capable of operating in outer space, where we need no vacuum pumps, and we hope in the near future to succeed in this task also.

Those interested in thoroughly surveying the work in the field to date can find a detailed list of most if not all known references in Mishler and Monroe (1962) and IAWRA (1962). For a digest of welding work completed by the end of 1962, see Bakish (1963), and for the most comprehensive and recent sources of information on electron-beam welding, see the handbook on the subject by Bakish and White (1964) and also Miller and Takemaki (1963).

The papers which follow show work in a number of areas of application. They show the progress effected to date and also give a feeling as to the types of tasks which can be successfully solved.

- Green, L., Jan. 1953, "Analytical Methods in the Study of Strontium Fluoride," *Symp. on the Study of Strontium Fluoride*, Boston, Mass.
- Harris, H. (1952), Ph.D. dissertation, University of Cambridge.
- Holmes, L. (1950), "Vacuum Deposition of Thin Films," Chapman and Hall, London, England.
- Johnson, R. W., *J. Appl. Phys.*, 34, No. 2, 353 (1963).
- Kelly, W. J., *Brit. Ass. Ceram. Soc.*, 31, 76 (1957).
- Piper, K. G., and K. Clemons, *J. Am. Chem. Soc.*, 81, 4477 (1959).
- Rosenthal, J. S., *J. Res. Natl.*, 70, 245 (1963).

Section V

ELECTRON BEAMS IN MATERIALS JOINING

ELECTRON BEAMS IN MATERIALS JOINING

The application of electron beams in welding dates back approximately ten years to Stohr of the French Atomic Commission, followed by Wyman in the United States. Steigerwald, at the same time in Germany, initiated micromachining research with this new heat source.

The past ten years have seen a most exciting growth in the application of electron beams for solution of joining problems. During this time electron beams in welding have successfully effected the transition from the research laboratory to the production stage. In this period of growth we have seen the complete obliteration of the high versus low voltage controversy so far as welding performance is concerned. We have seen substantial increase of power levels in electron beam welding installations from 1 to 2 kW up to 30 to 40 kW, an almost twenty-fold increase. We have further seen the beginning of, and some growth in, automation of the process. Last and not least, a considerable acceptance of this technique in industry has taken place.

In 1962 we saw initiation of effort to bring the electron beams for joining out of the vacuum. Today we have achieved reasonable success in welding with electron beams in inert atmospheres. We are working to develop electron guns capable of operating in outer space, where we need no vacuum pumps, and we hope in the near future to succeed in this task also.

Those interested in thoroughly surveying the work in the field to date can find a detailed list of most if not all known references in Mishler and Monroe (1962) and BWRA (1963). For a digest of welding work completed by the end of 1962, see Bakish (1962), and for the most comprehensive and recent sources of information on electron-beam welding, see the handbook on the subject by Bakish and White (1964) and also Miller and Takenaka (1964).

The papers which follow show work in a number of areas of application. They show the progress effected to date and also give a feeling as to the types of tasks which can be successfully solved

with electron beams. The joining with electron beams in inert atmosphere is also discussed. Although the Hamilton-Standard's and Heraeus representatives are extremely enthusiastic about its future growth, to me a somewhat more conservative estimate appears more appropriate.

Although considerable progress in electron-beam welding has been effected, much additional work has yet to be done. The emphasis here should be directed to development of complete specifications pertaining both to machine manufacture and performance, and to properties of electron-beam welded materials. I further believe that the fundamentals of the actual welding process and the exact nature of what happens in the material leading to the results obtained are still to be clarified. In conclusion I wish to state that electron beams in joining have achieved their place as an accepted tool, and that from now on the technique should enjoy continuous growth.

Editor

REFERENCES

- Bakish, R., 1962, "Introduction to Electron Beam Technology," John Wiley and Sons, New York.
- Bakish, R. and S. S. White, 1964, "Handbook of Electron Beam Welding," John Wiley and Sons, New York.
- BWRA, 1963, "Electron Beam Welding Bibliography," London.
- Miller, K. J. and T. T. Takenaka, 1964, "Electron Beam Welding in Aerospace Manufacturing," Welding Research Council, New York.
- Mischler, H. W. and R. E. Monroe, 1962, DMIC Rep. 174, Battelle Memorial Inst.
- Stohr, J., 1957, Fuel Elements Conf., Paris, TID 7546.

THE CURRENT STATUS OF ELECTRON-BEAM WELDING IN THE USSR

Electron-beam welding in a vacuum has been an industrial process in our country since the establishment in 1958 of the first laboratory electron-beam installation in the USSR.

Electron-beam welding finds application both for the joining of miniature parts in electronics and instrument making and for the joining of various articles with large overall dimensions having a length and diameter of several meters.

As is generally known, the high concentration of energy in the electron beam makes for small thermal distortion of articles during welding, and insignificant changes in the metal structure in the zone affected by the heat, ensures formation of a welded seam with a narrow fusion zone, a narrow heat-affected zone, decreases cracking susceptibility of the heat-affected zone, etc.

These properties determine the considerable interest in the study of "knife" welding (high depth-to-width ratio) in introducing this process into industry.

Electron-beam welding installations made in the USSR may be divided into three basic classes.

FIRST CLASS

The first class, a universal apparatus with multiposition change mechanisms and relatively small vacuum chambers, has a length of less than two meters and a diameter less than one meter. In-

*Moscow Power Institute and Bauman Technical Academy, Moscow, USSR.

†Paton Institute for Electro Welding, Kiev, USSR.

installations of this type have rather powerful vacuum systems, usually with a productive capacity of 1.0-2.5 thousand liters/sec at a vacuum of 1.10^{-4} - 1.10^{-5} mm Hg. The power of the electron-beam apparatus is up to 10-15 kW.

The universal character of installations of this type permits using them both in scientific research laboratories and in industrial enterprises.

A general view of an industrial installation of this type (U-3) is shown in Fig. 1.

The mechanisms of the installation permit welding of various flat and box-shaped articles and twelve articles with a cylindrical shape having a horizontal or vertical axis of rotation. The electron gun and power supply ensure formation of a beam with a power exceeding 10 kW (acceleration voltage 25 kV, beam current 500 mA).

The electron-beam welding installation ELU-1 (Fig. 2) is intended for simultaneous welding of ten cylindrical bodies with a length of up to 2000 mm. Automatic assembly of elements under vacuum is possible. When necessary, the drum for fastening of cylindrical articles may be replaced by a longitudinal table for welding flat articles. An electron gun type EP-25 with a maximum beam power of 2 kW at an acceleration voltage of 22 kV is used in this installation.

The industrial installation ELU-2 is of vertical construction with an automatic welding cycle. The installation has a rotating disc with collet clamps for fastening thirty articles. An EP-25 gun is also used on this installation.

ELU-4 and ELU-5 installations are also of the universal type. Some installations of this type have a mechanism for feeding an alloying wire under the electron beam for welding of ring-shaped and elongated seams; the welding of complicated curved seams in the horizontal plane is also made possible with the aid of a copying device.

For the needs of the radio electronics industry, a medium power electronic-beam welding installation is produced. This is also of the universal type with an acceleration voltage of up to 25 kV and beam current of up to 120 mA (type A306-05).

The chamber of the A306-05 installation (Fig. 3) is made in the form of a cube 500 mm on a side. An illuminator port with a diameter of 300 mm makes for good visibility and convenience in working. The chamber is designed for welding of ring-shaped seams in both the horizontal and the vertical plane as well as for welding of flat rectangular seams.

The main drawback of installations of the first type is their low

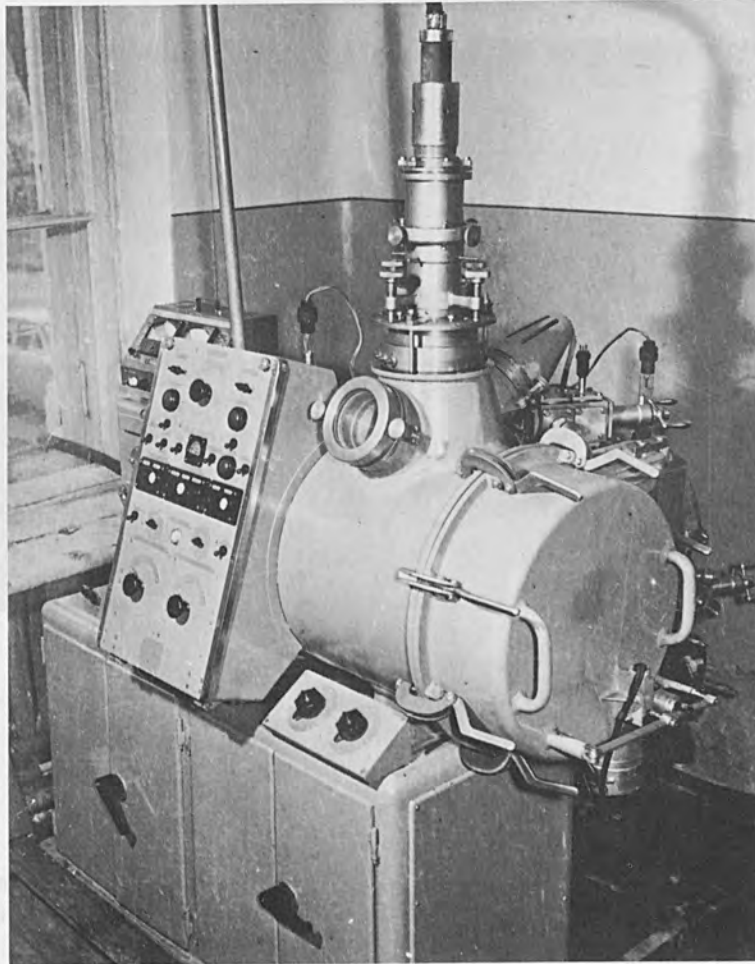


Fig. 1. Universal industrial installation type U-3.

productive capacity. In these installations only an insignificant portion of the time is taken up by the welding cycle. A considerable part of the time is spent in putting in the articles to be welded, exhausting the air from the chamber, and unloading the welded articles.

For this reason the greatest prospects for the welding of large numbers of small parts are offered by installations of the second class.



Fig. 2. Universal industrial installation type EDU-1.

SECOND CLASS

Installations of the second class are notable for the continuous process of loading, welding, and unloading of articles by the use of vacuum-loading hoppers or airlock chambers.

In some installations of this type, sectionalized vacuum chambers are used.

In Fig. 4 is shown the general view of an ELV-10 installation, which is intended for use in mass production of ring-shaped seams in the horizontal plane on articles of relatively small dimensions.

All mechanisms for loading and unloading of the articles are located outside the chamber. The fact that there is only one chamber and no auxiliary vacuum lead-ins makes the installation compact, simplifies its servicing, and increases the reliability of the work. Chambers of this type make it possible to carry out the welding with practically the same productive capacity and with as much ease as in welding in air.

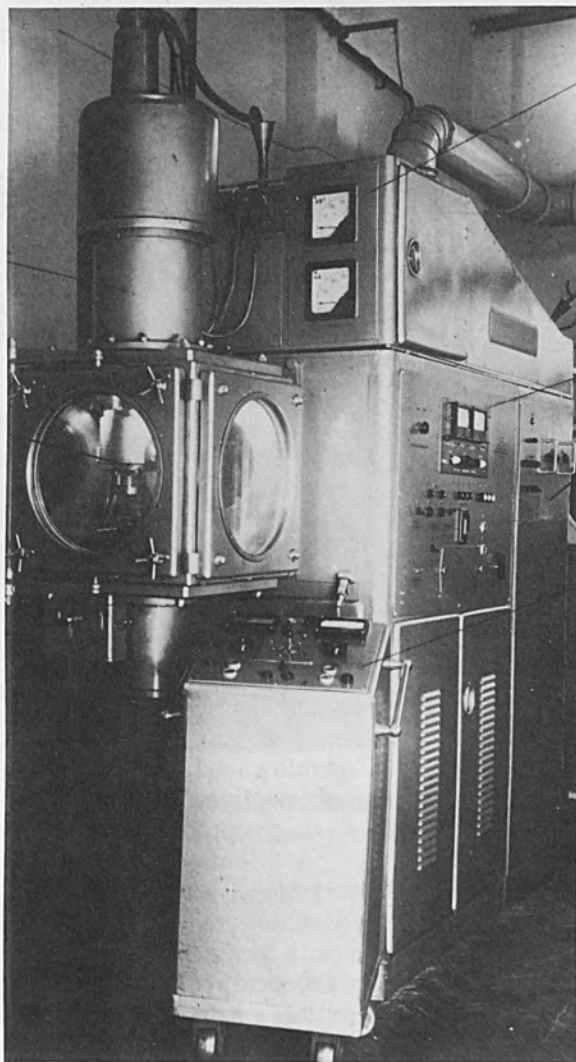


Fig. 3. Universal industrial installation type A306-05.

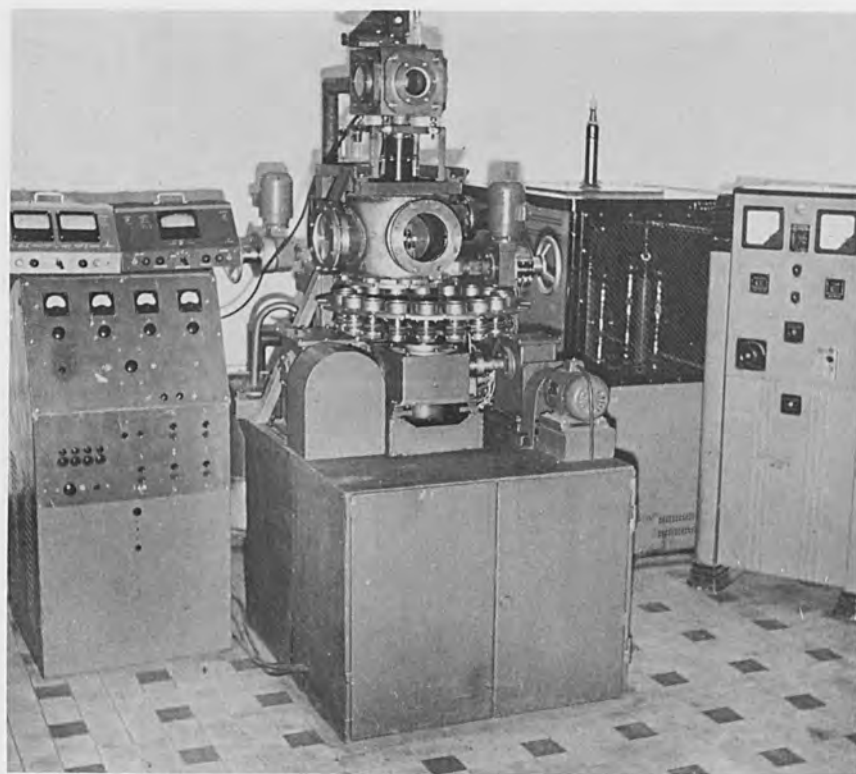


Fig. 4. High production installation ELV-10.

THIRD CLASS

To the third class of electron-beam welding installations belong installations for joining of articles with large overall dimensions.

Installations for electron-beam welding of articles with small and medium overall dimensions have been made, and have already been in use for quite a while in enterprises of the USSR. The creation of installations for electron-beam welding of articles of large overall dimensions required a new approach to designing of the installation as a whole as well as of its separate individual units. Mechanisms located in the chamber ensure precise and uniform rotation of articles being welded. The apparatus permits rapid and

convenient loading of the articles into the chamber. Since preparation of the installation for welding (unloading of the welded article, loading of the new article, exhausting the chamber to the working vacuum) takes up considerable time, several guns are mounted in the vacuum chamber, raising the productive capacity of the apparatus by making it possible to weld several ring seams in one loading and exhaustion cycle.

In Fig. 5 is shown an industrial installation of the Electric Welding Institute imeni Ye. O. Paton of the Academy of Sciences of the Ukrainian SSR for electric welding of articles with large overall dimensions (type U-86). Three welding guns are used in the installation. These are fed from one rectifier through a high voltage switch.

The maximum length of installations of this type which are already created reaches eleven meters; the diameter of the working chamber—three meters.

The basic types of welding guns used in industry in the Soviet Union are low voltage welding guns (E_{acc} up to 25 kV) and guns with intermediate acceleration voltage (E_{acc} up to 60 kV). The guns have a combined electrostatic-focusing and electromagnetic system. The basic characteristics of electron guns used in the USSR are given in Table I. The guns have metallic and boride-lanthanum cathodes and may work in continuous and in pulse conditions.

Scientific research being carried out in the Soviet Union in the field of electron-beam welding is directed toward the study of the properties of deep penetration welding. In the process of industrial adoption of electron-beam welding with the knife form of melting while using low voltage guns, a number of difficulties of a theoretical character were met.

It is generally known that in electron-optical systems with short focus electromagnetic lenses, insignificant changes in the current of the magnetic lens cause sharp changes in the focal length of the lens and, consequently, also the focal spot diameter on the article, which leads to instability in the welding process

$$\Delta f = \frac{2f_0}{I_M} \cdot \Delta I_M \quad (1)$$

In view of this drawback in short focus electron guns, welding guns were created with large focal distances which ensure formation of beams with a high-specific energy—on the order of 10 kW/mm². With the use of such guns, the diameter of the focal spot on the article is weakly dependent on the magnification of the magnetic

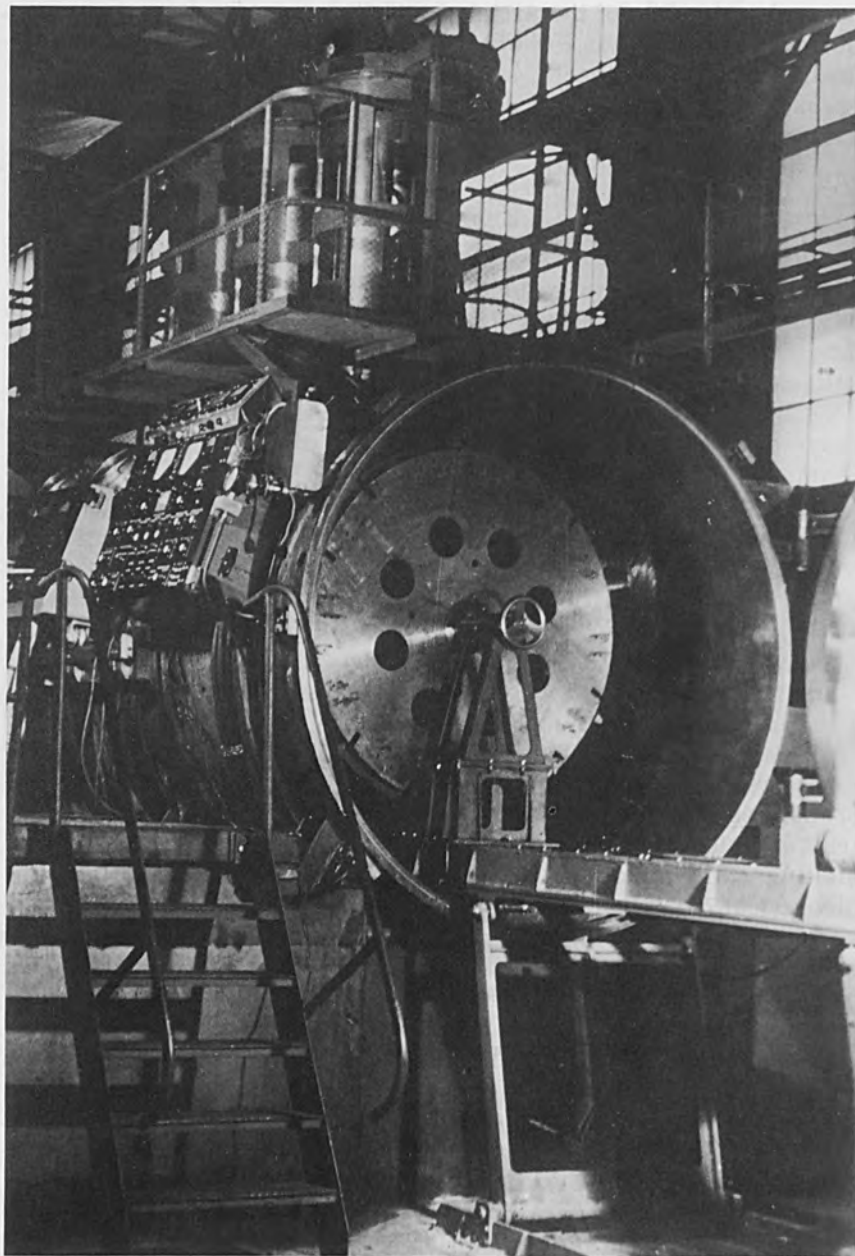


Fig. 5. Installation of U-86 for welding of articles with large dimensions.

Table I
 Characteristics of Industrial Electronic-Welding Guns
 of the USSR

Class of Gun	Acceleration Voltage E_{acc} kV	Beam Current I_b mA	Spot Diameter at Max. Power d mm	Specific Energy at the Focal Spot w/s kW/mm ²	Symbols
1. Low-voltage guns	20	200	1.0	5.0	A.306.05
	25	60	0.8-1.5	3.0	EP-25
	25	500	1.2-1.3	5-10	SP-30
2. Guns with intermediate voltage	60	35	0.6-0.7	5-7	EP-60
3. High-voltage guns	100	15	0.1-0.25	30-200	ELURO-1

lens $M = 1/d$, especially at values of M on the order of 1, which makes it possible to obtain stable parameters of the area of melting. At the same time, calculations of the minimum dimensions of the focal spot based on the paramount role of the negative space charge show a sharp change for the worse in the parameters of the knife melting with an increase in the distance between the magnetic lens and the article. Thus, calculation by the approximation equation of Hollway and according to Moss for a cylindrical beam of electrons ($0.05 < r_{f.s} < 2R$);

$$\frac{r_{f.s}}{R} = 10.5 \left(\frac{1}{R} \right)^{5/2} \frac{I_b^{5/4}}{E_{acc}^{3/2}} \quad (2)$$

for a beam with parameters $I_b = 200$ mA, $E_{acc} = 2 \cdot 10^4$ V, the radius of the beam at the output from the magnetic lens $R = 5$ mm— at $M = 1/a = 240/80 = 3$ — $r_{f.s}/R = 1.1$, whence $r_{f.s} = 5.5$ mm, i.e., the design radius of the focal spot should exceed by at least an

order the value of the radius of the beam at $M = 1$, when $r_{f.s} = 0.4$ mm. However in our experiments for cases of $M = 3$, the value obtained was $r_{f.s} = 0.6-0.8$ mm. Such a divergence of calculated and experimental data indicates the necessity for taking account of neutralization of the space charge of the beam by ions, since welding processes take place at a relatively low vacuum (on the order of 1.10^{-4} mm Hg) and with an intensive directed liberation of vapors and gases from the welding bath. Neutralization of the space charge facilitates the formation of long focus beams with a high-specific energy in the beam.

Until now there has been no clear presentation of the mechanism of formation of a welded joint during deep penetration electron-beam welding.

It is fully possible that an understanding of this mechanism would help in creation of an apparatus which would ensure better stability and reproducibility of the welding process.

It is noticed that during electron-beam welding that the surface of the welded seam is covered with small scales. A microscopic study of the seam microstructure has revealed the presence of solidification lines in the metal of the seam. The presence of scales and solidification lines in the metal of the seam indicates that solidification of the metal of the seam during electron-beam welding is periodic.

The presence of solidification lines in the welded seams, and the relatively small distance between them, testifies to the fact that with the use of a high-intensity electron beam, the metal at the forward end of the crater is melted in small portions and periodically fed to the crystallization area.

According to source material, the process of melting of the metal during electron-beam welding with deep melting takes place with the energy of the electron beam uniformly distributed along the surface of the cone of melting.

The action of the electron beam in such a presentation is analogous to the action of a stiff, red-hot rod (wire) which makes uniform contact with the surface of the crater.

Observations have shown that such a presentation of the process of electron-beam welding cannot explain many peculiarities of this process.

A more probable presentation of the thermal and mechanical action of an electron beam is to picture it as a jet of liquid or gas which washes over a dry, powdery substance. In a presentation of this type, the electron beam melts the metal at the front edge of the crater by degrees, layer after layer from the surface to the

depth, throwing small portions of liquid metal into the solidification area. Evidence in favor of such a presentation is Fig. 6, a photograph of the surface of the crater in which steps may be seen which are formed as a consequence of the uneven melting of the metal at the front wall of the crater.

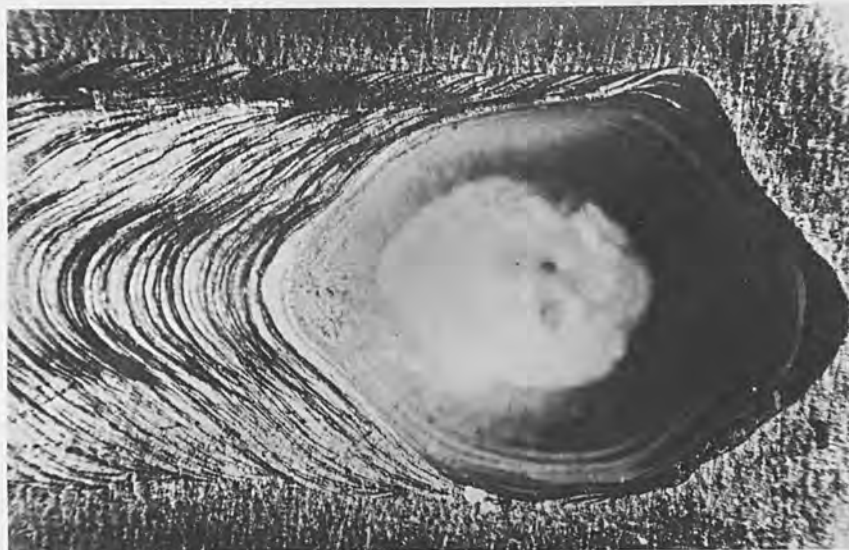


Fig. 6. View of the crater and the surface of the seam during electron-beam welding.

Besides this, the periodicity of the solidification and the formation of scales during electron-beam welding may be explained by interrupted motion of the stream of electrons. The stepwise motion of the electron beam could be explained by the action of ionized metal vapors over the welding bath. When the electron beam moves through the ionized vapors and gases, positive ions are concentrated at its center. The positive ions from a flexible potential tube from which electrons are unable to escape in a radial direction, as a consequence of which the beam of electrons may be held for some time within the boundary of the heating spot by the positive ions and a "step" of the electron beam to a new position is observed only when there is a definite deviation of the axis of the beam from the center of the welding bath as a result of motion of the article being welded. This stepwise motion of the beam and the unevenness of melting of the metal at the forward wall of

the crater are what causes the periodic solidification of the metal with the formation of scales on the surface.

The effect of the position of the focal spot with respect to the plane of the article being welded on the formation of a welded union with deep melting has been established experimentally.

When the focal spot is located below the surface of the metal (Fig. 7a), the melting zone has the form of a sharp wedge. In the

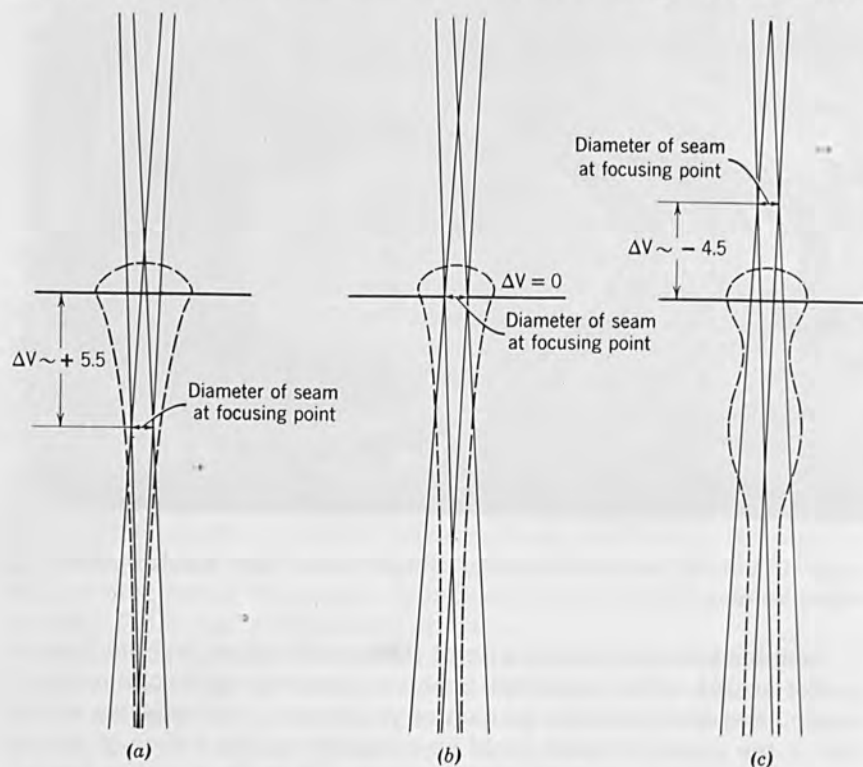


Fig. 7. Change in the form of the melting area depending on the location of the focal spot.

lower part of the seam, pores and discontinuities are observed. Pores of various sizes are also possible in the upper part of the melting area.

A favorable form of melting is obtained when the beam is focused on the surface of the metal (Fig. 7b).

When the focal spot is located above the surface of the metal (Fig. 7c), a narrow isthmus with large pores appears in the upper

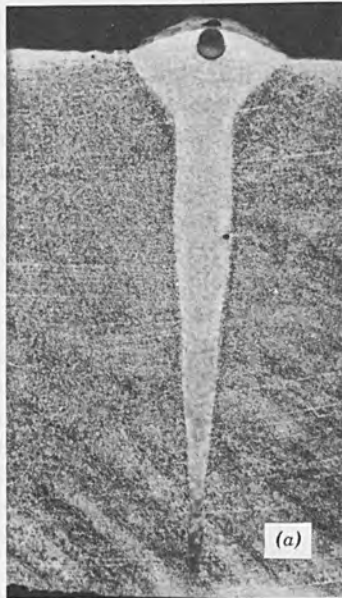


Figure 7a.

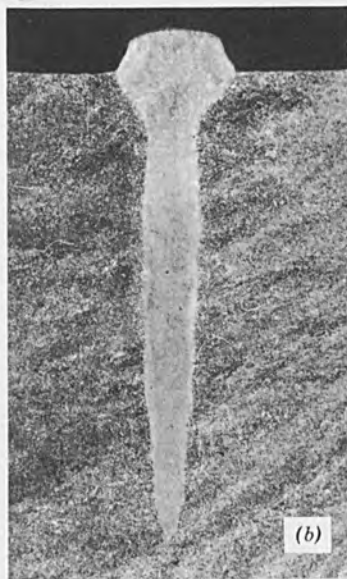


Figure 7b.



Figure 7c.

part of the seam at the place where the metal is locally superheated by the focused central part of the beam.

Welding practice shows that it is necessary to have uniform distribution of the current density throughout the area of the focal spot. During welding, with uneven distribution of the beam current in the central part of the seam where the most intensive part of the beam is in operation, a considerable narrowing of the seam is observed in its lower part. This phenomenon impedes the welding since small deviations of the beam from the line of the joint make even fusion impossible.

The complexity of bringing the beam, which has a narrow melting area, into line with the joint being welded is what determines the necessity for a more even distribution and sets the optimum for the specific energy in the focal spot.

In Fig. 8 is shown the experimental relationship of the dimen-

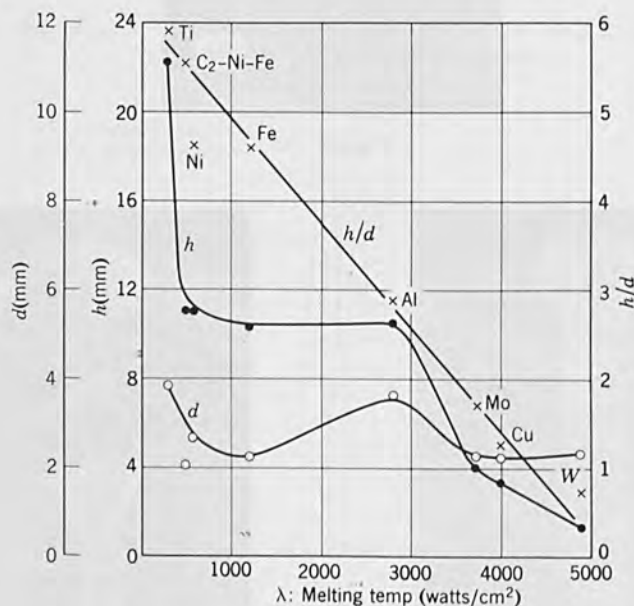


Fig. 8. Change in the dimensions of the melting zone depending on the thermo-physical properties of the metal.

sions of the fusion zone for materials with different values of λT_{fu} for a low-voltage gun—beam current 300 mA, acceleration voltage 21 kV, welding speed 30 meters/hr and $W_{spec} \cong 5 \text{ kW/mm}^2$.

It is evident from the graph that fusion parameters become worse with welding of more refractory materials. For welding of special steels and nonferrous metals with knife melting, it is efficient to use low-voltage welding guns with a specific power in the beam on the order of 10 kW/mm^2 and a general beam power of up to 10 to 15 kW. When welding refractory metals it is necessary to raise the specific energy in the focal spot of the beam by using high-voltage guns in order to ensure melting with a high ratio of depth to width.

Processes were cleared up which are connected with secondary emission of electrons from the welding bath and experiments were conducted on determination of the part of the energy which might be usefully exploited in electron-beam welding. In determination of the processes connected with secondary emission, the delayed potential method was used. The research was conducted with a type A-306 electron-optical system.

Losses of energy during passage of the electron beam from the cathode to the article may take place in the following sections:

- 1). At the accelerating electrode as a consequence of the fact that the projector elements are not coaxial and because of the intentional cutting off by a diaphragm of the peripheral part of the beam with a small energy density.
- 2). On the way from the anode to the article.
- 3). During bombardment of the metal of the article.

In the general case, the losses at the accelerating electrode may reach 5 to 10% of the general power of the beam.

Energy losses in the electron beam during passage through the drift space take place, in general, as a result of nonelastic collisions of electrons with molecules of residual gas. The magnitude of these losses depends on several factors; the magnitude of the current of the voltage acceleration beam, the degree of rarefaction, the distance between the cathode and the article.

It is established that at a pressure of $3 \cdot 10^{-4} \text{ mm Hg}$, energy losses of electrons in the beam through interaction with the plasma do not exceed 1% of the original energy of the beam. However, at a pressure of $4 \cdot 10^{-3} \text{ mm Hg}$, the energy losses already comprise 11% of the original energy of the beam. It may be assumed that losses by the electron beam in welding chambers on the way from the cathode to the anode at a working pressure in the chamber of $1 \cdot 10^{-4} \text{ mm Hg}$ and an acceleration voltage not less than 20 kV may be estimated at 1% of the total energy of the beam.

The most obscure part of the losses of energy of the electron beam during welding are losses with secondary electrons. According to source material, secondary electrons in X-ray tubes may carry off a considerable part (20-30%) of the total energy of the electron beam. There are no such data for the process of electron-beam welding.

Investigations were conducted with the aim of clearing up some of the aspects of phenomena caused by secondary emission in welding conditions.

In our experiments, a study was made of the dependence of the current of the part (I_3) and the current of the secondary electrons (I_2) on the total current of the beam (I_b), the voltage of the beam (E_{acc}) and the material of the article. For this purpose a series of experiments was set up whose system is given in Fig. 9.

As a result of the experiments conducted on electron-beam welding, it was established that the current of the secondary electrons (I_2) is altered to a considerable degree depending on the current and voltage of the beam and also on the properties of the metal of the part being welded.

The results of experiments on a molybdenum plate are given in Fig. 10. As may be seen, at a voltage of 15 kV, when the current in the beam is increased, the current in the part attains the value of the current of the beam and then diminishes to zero and changes sign. At the moment when a puddle of molten metal appears, a sharp decrease is observed in the current of the secondary electrons and the value of the current of the part exceeds in reverse polarity the current of the beam. However, the current of the part is always less than the secondary electrons by approximately the beam current.

The observed excess of electrons leaving the surface of the part indicates not only the reflection of primary electrons and dislodging of secondary electrons from the surface of the metal, but also the formation of a considerable number of thermal electrons.

It was established that the suppression part of the electron emission produces thermal electrons with energies of only a few electron volts in all. Thermal electrons comprise more than 90% of the total number of electrons emitted by the welding bath.

The total energy carried off by thermal electrons at a beam current of 60 mA and a voltage of 18 kV may reach 6 to 7 watts.

The secondary and reflected electrons have quite large energies and their distribution in the interval which we studied does not differ from the classical distribution. As shown by calculations for a beam with a current of 40 mA, the total power carried away

by the secondary electrons at a secondary emission current of 15 mA is from 30 to 50 watts. The total energy losses from secondary electrons which we calculated (for the following welding conditions: acceleration voltage $E_{acc} = 18$ kV; beam current $I_b = 40$ mA) were from 8 to 10%.

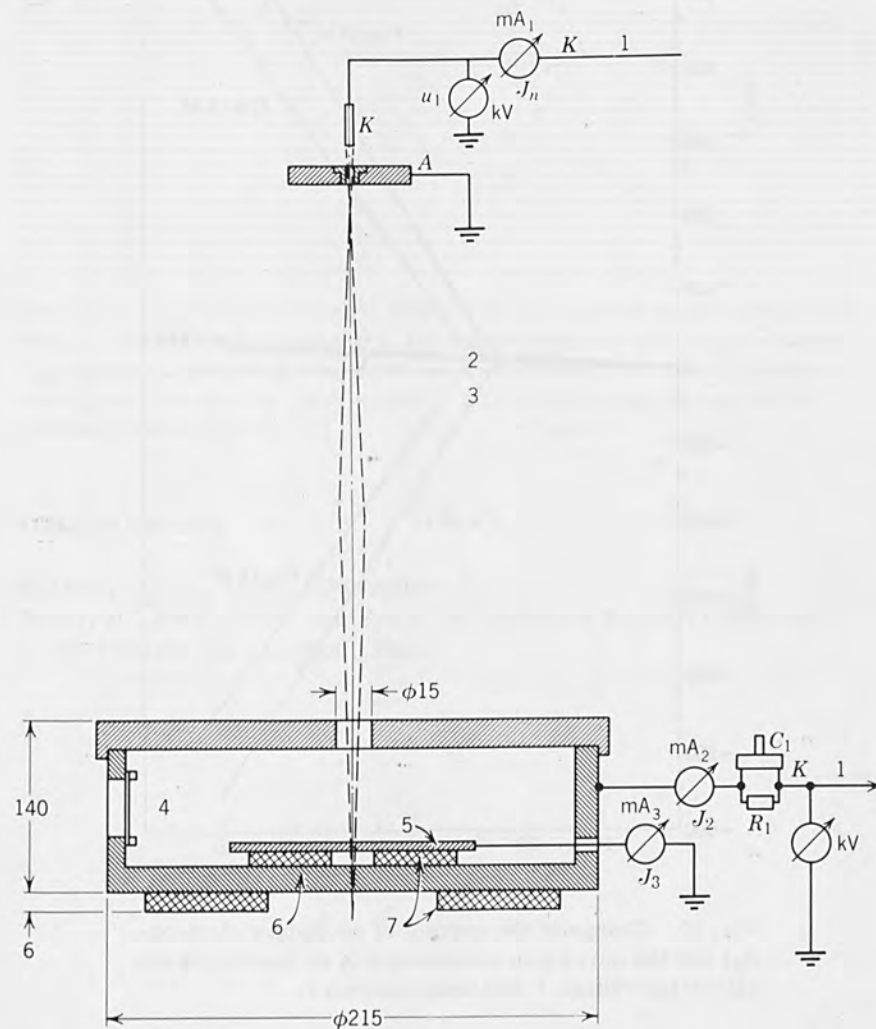


Fig. 9. System for measurement of the secondary emission current during welding. 1. Rectifier. 2. Current measuring circuit. 3. Secondary emission. 4. Sighting port. 5. Workpiece. 6. Screen. 7. Ceramic insulator.

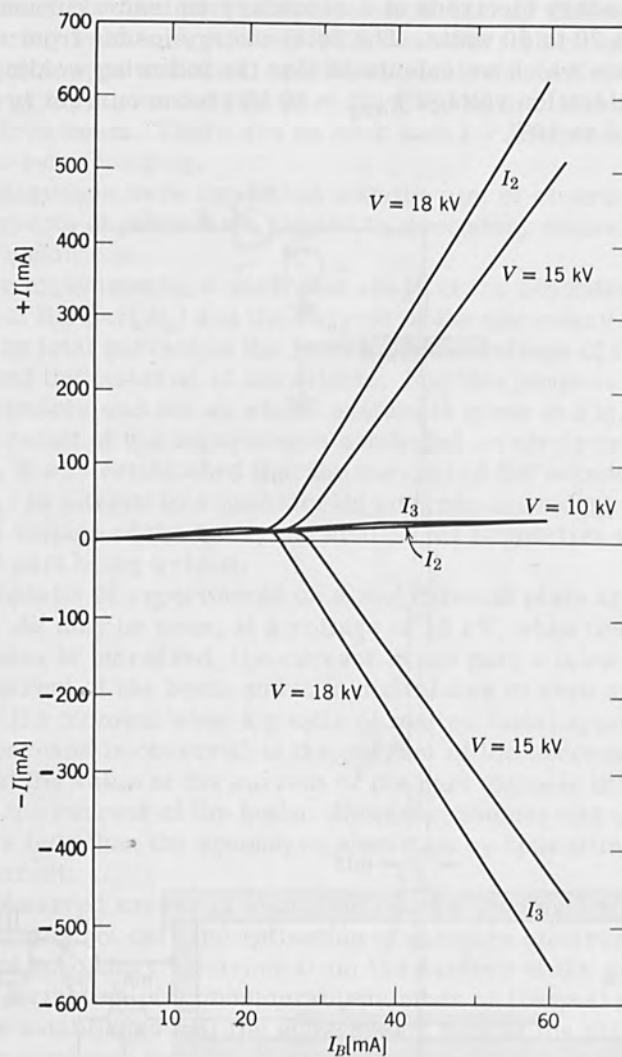


Fig. 10. Change in the current of secondary electrons (I_2) and the current in workpiece (I_3) as function of acceleration voltage V and beam current I .

In Fig. 11 the approximate balance of energy of the electron beam is given. As may be seen, the effective thermal power of the electron beam may be about to 88 to 93%.

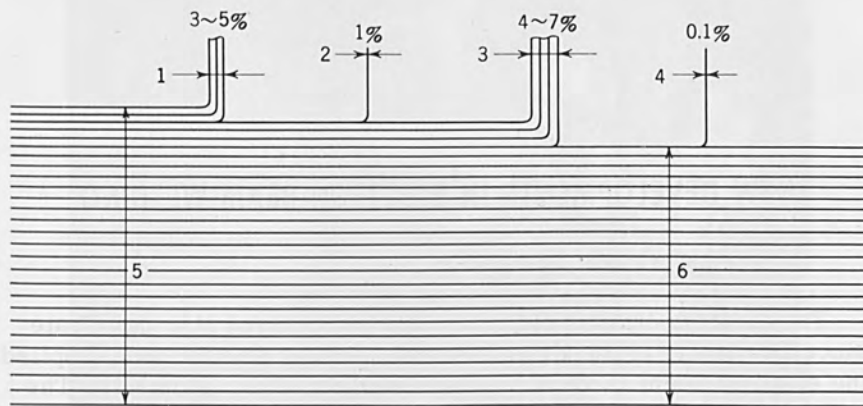


Fig. 11. Approximate thermal balance of the electron beam during welding. 1. Losses at first anode. 2. Losses on collision with gas molecules. 3. Losses of secondary electrons. 4. X-ray radiation. 5. Full thermal energy of the electron beam — 100%. Effective thermal energy of the electron beam 88-93%.

BIBLIOGRAPHY

- Hollway, D. L., 1962, *Electronics* 35, 7.
 Moss, H., 1962, *Proc. 4th Symp. on Electron Beam Technology*,
 R. Bakish, Ed., Boston, Mass.

NEW DEVELOPMENTS IN ELECTRON-BEAM WELDING

The electron-beam welding art has progressed at a rapid rate since Dr. Stohr, of the French Atomic Energy Commission, applied the electron beam in vacuum to the welding of fuel elements. The advantages inherent in this process have been enumerated in several of Dr. Stohr's papers as early as 1956. The first application of electron-beam welding was in the fastening of end closures for fuel element cases made of beryllium, zircaloy, and aluminum in thicknesses up to perhaps 0.050 in. The process has now been extended to many other fields and to a variety of materials and thicknesses.

Figure 1 illustrates the application of electron-beam welding to the joining of turbine wheels to shaft assemblies. The work was rotated about the horizontal axis. The gun was held fixed for each girth weld, but moved along a line parallel to the axis to weld four succeeding joints. The materials in the various components of the assembly are A286 stainless steel, 17-22A steel, SAE4130, and SAE4140. The shaft has an outside diameter of 5-1/2 in.

In stainless steel, welds have been made in thicknesses from 0.001 to 2 in. (Fig. 2), and aluminum plates up to 6 in (Fig. 3), have been welded on commercially available equipment capable of delivering 1 amp at 30 kV. Table I gives some indications of the parameters used in making sound welds in a variety of metals of different thicknesses. Tables II and III summarize the results of physical tests of some of the materials.

It is interesting to compare the energy required to weld by the electron-beam process against that used by other welding processes.

Figure 4 shows a weld made in 1/2 in. 304-stainless steel by the MIG welding process, which later had superimposed over the MIG weld a bead made by the electron-beam process. The MIG

*Sciaky Bros., Inc., Chicago, Ill.

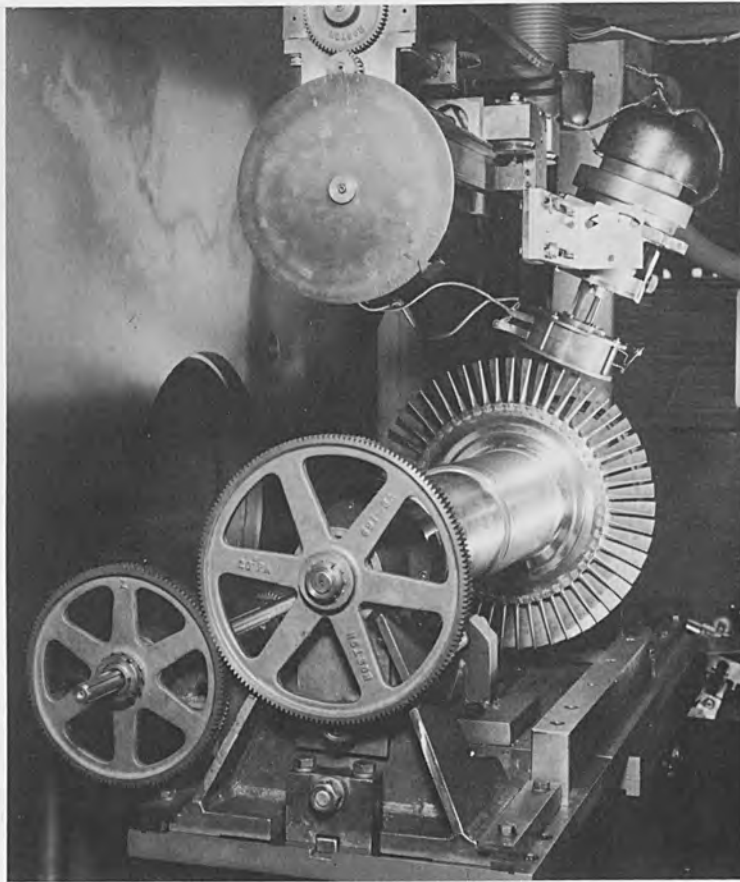


Fig. 1. Turbine wheel and shaft assembly.

weld was made at 6.5 in./min and required a current of 360 amp at 28.7 V, or a power of 10.3 kW. The energy input per inch of weld was 95.5 kJ. The electron-beam weld was made at 225 mA and 30 kV, at 37 in./min, a total power of 6.75 kW, and an energy input to the work of 9.1 kJ/in., less than 1/10 the energy per inch required when MIG welding this very same material. In the welding of 1/2 in. 2219 aluminum, the reduction in total energy input to the work, when using electron beam over TIG welding, is even more striking, since the electron-beam weld requires a beam current of 200 mA at 30 kV when welding this material at 95 in./

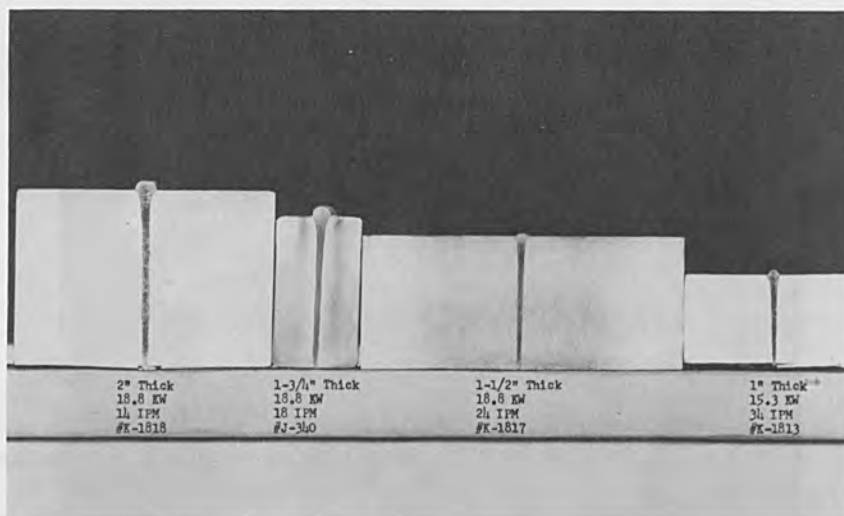


Fig. 2. Electron beam welds in 1- to 2-in. stainless steel welded at 15-in. gun-to-work distance.

min, or a total of 3.8 kJ/in., as against the two passes required in TIG welding this material at an average speed of 7 in./min with a total energy input to the work of 57.6 kJ/in. TIG welding this material requires fifteen times the energy input of electron-beam welding.

The wide range of power that may be delivered by a single electron-beam machine is also quite striking; for example, in welding 0.0008 in. titanium, a beam current of 1.25 mA at an accelerating potential of 7 kV was required to produce a full penetration weld at 45 in./min. This represents a power of 8.75 watts, as against the maximum power of 30 kW, or a ratio of 3420 to 1.

WELDING OF LARGE COMPONENTS

Inasmuch as at first thought it would appear that the piece parts to be welded by the electron-beam welding process must be smaller than the available vacuum chamber, considerable work has been done in an effort to overcome this limitation by attempting to develop equipment and methods whereby large structures could

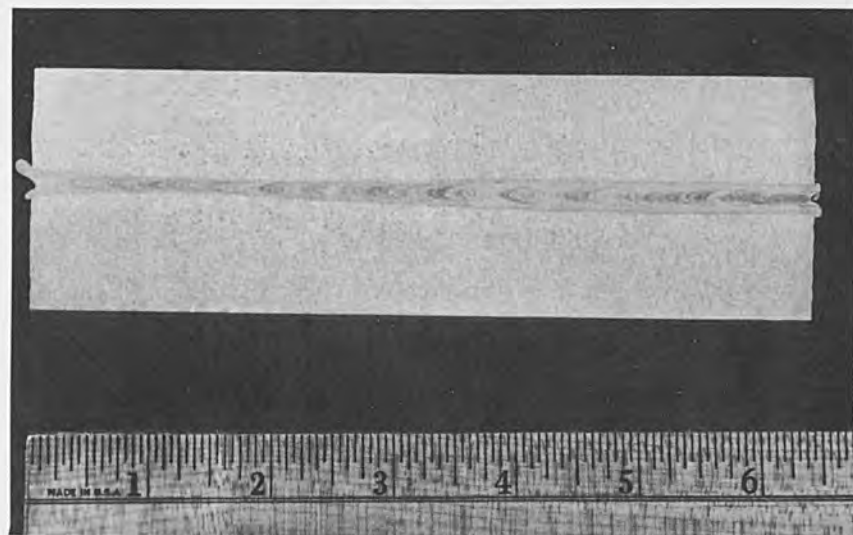


Fig. 3. Weld in 6-1/4-in. aluminum 5456-H343.

be welded, and at the same time retain the advantages inherent in the electron-beam welding process.

For the past several years, a great deal of effort has been expended in an attempt to produce an electron beam of high-power density, pass it through a series of orifices which are separated by differentials in pressure, and finally direct it to the work which is surrounded by an inert gas atmosphere at atmospheric pressure. The best results reported to date indicate that welds having a depth-to-width ratio close to 3:1 may be made in 1/4 in. stainless steel. However, in order to produce these results, it is necessary for the gun to be placed within 1/4 in. from the work. Welds in 1/8 in. material may be made with the gun at a distance of 1/2 in. from the work, at 150 kV, 10 mA, and a welding speed of 12 in./min. The energy input to the work in this case is 7.5 kJ/in.

If we compare this to the other welding processes, we find that 1/8 in. material may be welded by the TIG or MIG process at 20 in./min. with a total expenditure of 10 kJ/in. Welding by means of electron beam in a vacuum chamber requires the expenditure of 1.08 kJ/in. Inasmuch as welds produced out of vacuum cannot be as clean as those produced in vacuum, it appears as though electron-beam welding in air would have a very limited application.

Table I (a) - High-Temperature Alloys
Electron-Beam Weld Settings For Several Materials & Thicknesses

Material	Thick- ness Inch	Weld Type	Beam Voltage kV	Beam Current mA	Welding Speed In./Min	Welding Energy kJ/in.
Rene 41	0.015	SQB*	16.0	6	45	0.13
Haynes 25	0.015	"	19.0	8	64	0.14
Waspaloy	0.040	"	24.0	30	97	0.44
Hastelloy N	0.063	"	30.0	45	85	0.95
Inconel 718 †	0.080	"	30.0	26	39	1.20
Rene 41	0.093	"	23.0	54	48	1.55
Haynes 25	0.093	"	30.0	70	80	1.57
Haynes 25	0.375	"	30.0	180	51	6.35
Udimet	0.650	"	29.0	425	65	11.4
Rene 41	0.875	"	29.0	400	45	15.5
Inconel 718	0.875	"	26.0	435	50	13.6
Inconel 718	1.250	"	30.0	500	34	26.4

*SQB = square butt.

†Weld made vertically-up with gun horizontal

Table I (b) — Stainless & Maraging Steel
Electron-Beam Weld Settings for Several Materials & Thicknesses

Material	Thick- ness Inch	Weld Type	Beam Voltage kV	Beam Current mA	Welding Speed In./Min	Welding Energy kJ/in.
302	0.015	SQB*	31.4	32	56	1.08
302	0.250	"	30.0	100	60	3.00
304	0.500	"	29.0	330	63	9.10
309	0.750	"	29.0	470	56	14.6
321	1.00	"	30.0	500	40	22.5
316†	1.50	"	57.5	360	29	42.8
18% Ni	0.625	"	30.0	400	63	11.4
"	1.00	"	44.0	500	65	20.3
"	1.25	"	44.0	500	35	37.7

*SQB = square butt.

†Weld made vertically-up with gun horizontal.

Table I (c) — Aluminum Alloys
Electron-Beam Weld Settings for Several Materials & Thicknesses

Material	Thick- ness	Weld Type	Beam Voltage kV	Beam Current mA	Welding Speed In./Min	Welding Energy kJ/in.
6061-T6	0.050	SQB*	18.0	33	100	0.36
2024 T6	0.050	"	27.0	21	71	0.48
2014-T6	0.120	"	29.0	54	75	1.26
6061-T6	0.125	"	26.0	52	80	1.03
7075-T6	0.125	"	25.0	80	90	1.33
2219-T81	0.500	"	30.0	200	95	3.97
6061-T6	0.625	"	30.0	275	75	6.60
5086	2.00	"	30.0	500	36	25.0
2219-T81†	2.375	"	30.0	1000	43	41.8
5456-H343**	6.000	BOP†	30.0	1025	7	263

*SQB = square butt.

†BOP = bead on plate

‡Weld made vertically up with gun horizontal.

**Bead on plate made during investigation to determine maximum penetration of 30 kV, 30 kW electron-beam welding system.

Table I(d) — Titanium & Refractory Metal Alloys
Electron-Beam Weld Settings for Several Materials & Thicknesses

Material	Thickness Inch	Weld Type	Beam Voltage kV	Beam Current mA	Welding Speed In./Min	Welding Current kJ/in.
13V, 11Cr 3Al	0.080	SQB*	26.0	38	60	0.99
3Al, 1Mo, 1V	0.500	"	30.0	195	50	7.03
5Al, 2.5Sn	0.750	Lap	27.0	450	50	14.6
6Al, 4V	1.00	SQB*	27.0	450	41	17.8
Columbium 752	0.080	"	29.0	55	36	2.66
Columbium	0.500	"	29.0	340	46	12.8
TM-109 Tantalum	0.045	"	20.5	63	20	3.87
Nickel	0.018 to 0.025	Lap	30.0	19	100	0.342
Tungsten	0.250	SQB*	22.0	350	22	21.0

*SQB = square butt.

Table II
Per Cent Elongation and Per Cent Reduction in Area Data for Electron-Beam
Welded Square Butt Joints in 1/2 Inch Thick Aluminum Alloy 2219-T81

Weld No.	Specimen Geometry ($\times$, or Diam.)	Weld Width (at 0.5T) Inch	Elongation (For gauge length as indicated)				Reduction in Area %
			2.00 in.	1.00 in.	0.50 in.	0.25 in.	
	Inch		%	%	%	%	%
M-48	0.200 $\times$ 0.500	0.042	2.00	4.00	6.75	13.25	20
M-48	0.252	0.042	1.25	2.50	5.50	9.00	20
M-28	0.200 $\times$ 0.500	0.060	2.00	3.75	8.00	14.50	—
M-28	0.252	0.060	2.00	4.00	7.25	15.00	22
M-24	0.200 $\times$ 0.500	0.070	2.00	3.50	8.00	14.00	—
M-24	0.252	0.070	2.00	4.00	9.00	17.00	26

Table III

Joint Efficiency of Electron-Beam Welded Square
Butt Joints for Several Materials and Thicknesses

Material	Thickness inch	Yield Strength kips	Ultimate Strength kips	Joint Efficiency per cent
EZ-33A-T5 magnesium	5/8	...	17.5	97
5083-H322 aluminum	...	35.9	47.9	90
2219-T31 aluminum	1/2	42.3	50.7	77*
6A1-4V titanium	5/8	158	168	100
D6-AC steel	1/2	217	238	98
(tempered 900°F, 2 hours)				
D6-AC (Ausformed 50% steel	1/2	236	247	89
(tempered 900°F, 2 hours)				
Maraging steel - 18% Ni	1/2	249	254	...
(aged 900°F, 3 hours)				
Maraging steel - 18% Ni	1/2	264	268	...
(aged 875°F, 8 hours)				
AM 355 CRT Stainless steel	0.080	...	194	90

*Teat specimen was .252 in. diameter round bar. Full section (1/2 in.) specimens exhibited 82% joint efficiency.

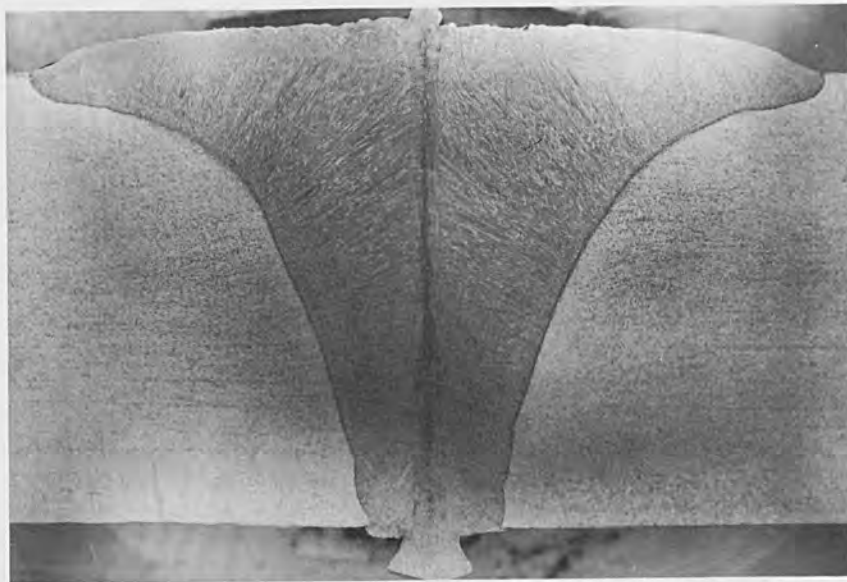


Fig. 4. Comparison of MIG to electron-beam weld in 1/2-in. thick 304 stainless steel.

even though the welds made by this process are acceptable. The influence of gun-to-work distance has a marked effect upon penetration of the weld because of the decreased current density caused by collision and scattering in the electron beam; for example, increasing the distance from 1/4 to 1 in., decreases the penetration by a factor of 7. Since the very narrow clean welds obtainable in a vacuum cannot be obtained by electron-beam welding in air, one wonders why TIG welding, with its much lower equipment cost, should not be used where the type of welding produced by electron beam in air are acceptable.

Another attempt to duplicate the quality of welds produced by the electron-beam vacuum has been through the use of the laser. In this process, the high-density monochromatic, coherent bundle of light emitted in a pulse having a duration of approximately 1 msec is directed through a lens and focused on the surface of the work, the high energy in the beam heating the material sufficiently to cause it to melt and vaporize. Although some joining has been attempted in several laboratories, the results reported to date by this method have not been generally satisfactory even in the thin-

nest materials, chiefly because the energy, although highly concentrated, is of short duration. What is required is a continuous laser beam of sufficient power, several kilowatts, which could be controlled in intensity.

Several very interesting methods by which parts larger than the vacuum chamber have been welded, and by which the highest quality electron-beam welds in vacuum are produced, have already been developed, manufactured, and put to use. One of the systems solved the problem of butt-welding long-diameter tubing. This was accomplished in a rather simple manner by inserting a plug in each of the tubes close to the end to be welded, supporting the tubes adequately and passing them into the vacuum chamber through plates which serve as rotating seals — the tubes being fixed and sealed to the plate by inflatable rubber seals — the sliding seal being made part of the plate which itself was well guided in relation to the vacuum chamber.

Another method was to add extensions to the chamber so that 12-ft-long parts could be accommodated as in Fig. 5.

Another interesting application is that illustrated in Figs. 6, 7, and 8. This installation was made for the purpose of producing long lengths of refractory tubing from the shorter lengths which are available from the manufacturers of this material. At the left of Fig. 6 can be seen a tube which is passed into the chamber through a fixed O-ring seal. The second tube to which this tube is to be welded may be seen at the rear of the machine. Figure 7 shows the method by which the weld is made. The gun is mounted on a rotating fixture within the chamber. The electron beam is directed to the joint and the electron gun is rotated in the fixture about the tube, thus making a continuous circular seam around the tube. The table upon which the gun is mounted is driven by a motor whose speed is controlled by a feedback-regulated motor control. Figure 8 shows the complete installation with control console and recording equipment used to monitor beam current, voltage, and motor speeds.

Another interesting application is that for welding segments to produce very large-diameter rings in heavy materials. An example of the type work that may be done with this equipment is the fabrication of the circumferential stiffening rings used in the construction of a submarine hull. Figure 9 illustrates how this was accomplished. Inasmuch as sections for this type joining are uniform, although not always rectangular, it was possible to produce a vacuum chamber which could be clamped locally at the area where the welding was to be done. The segments to be welded

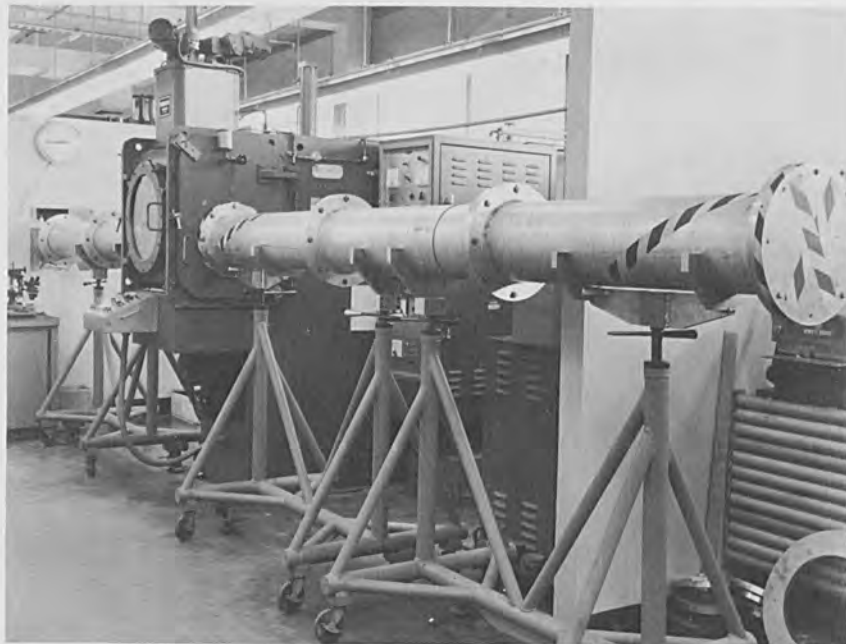


Fig. 5. Electron-beam welder type VX 32 x 20 x 30 with 12-ft extensions, installed at Solar Aircraft.

together are held in position with suitable fixtures. The vacuum chamber is then clamped around the parts, and by means of an electron-beam gun, which moves upward inside the chamber, the welding is accomplished.

An application which presented itself recently was the welding of segments to produce very large diameter rings in 2-3/8-in. thick aluminum. These rings were required on the S-1C stage of the Saturn-V launch vehicle. These rings, having a cross section resembling a "Y," are utilized to join cylindrical sections to end domes and skirts of the fuel, and oxidizer tanks for the S-1C stage of the Saturn V booster.

Figure 10 shows the placement of the four Y-rings on the vehicle.

Figure 11 shows a cross section of a Y-ring which is 33 ft. in diameter and weighs 3700 pounds.

The maximum thickness of the Y-ring was 2-3/8-in. and the cross section varied from 3/8 in. to this maximum.

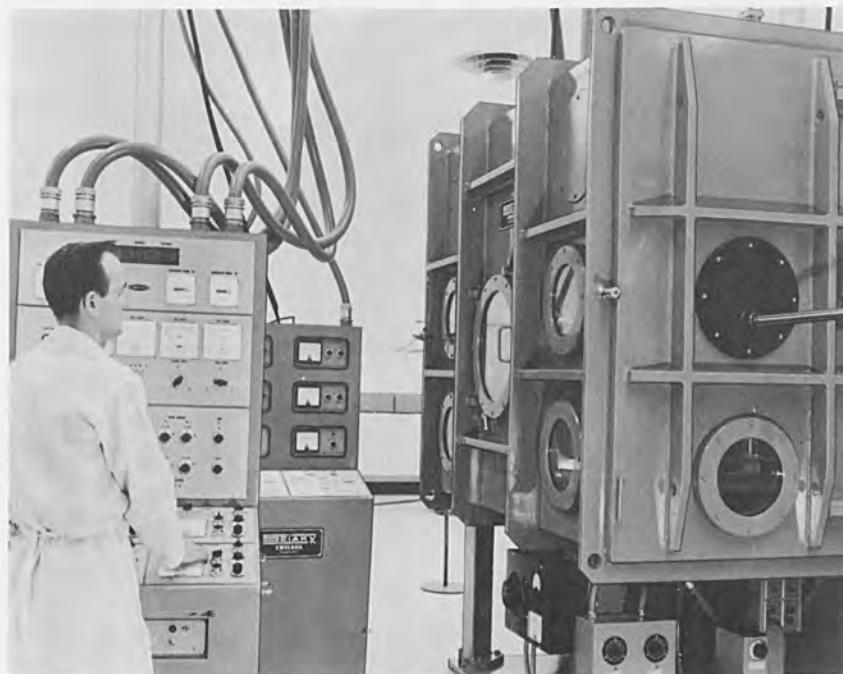


Fig. 6. Welding of continuous lengths of refractory metal tubing at NASA, Lewis Research Center, Cleveland, Ohio. Tubing passes through vacuum seals located on each chamber door.

Figure 12 is a proposal drawing, showing how the several sections of the Y-ring are handled and positioned into an electron-beam welding machine which clamps locally around the area where the butt weld is to be made. This vacuum chamber is only large enough to house the aligning fixture, the electron guns, and a small area of the Y-ring in the vicinity of the joint to be welded. In the development of this equipment, it was first necessary to determine the welding parameters required for the welding of 2-3/8-in. type 2219 T87 aluminum. By welding tests made on several thicknesses of 2219 material, which could be satisfactorily welded at the maximum capacity of the welding machines available at the start of the program, rated at 30 kV and 500 mA, it was determined, by extrapolation from the results, that the power required to weld the 2-3/8-in. aluminum would be 30 kW.

An electron gun, capable of delivering 1 amp at 30 kV, was then

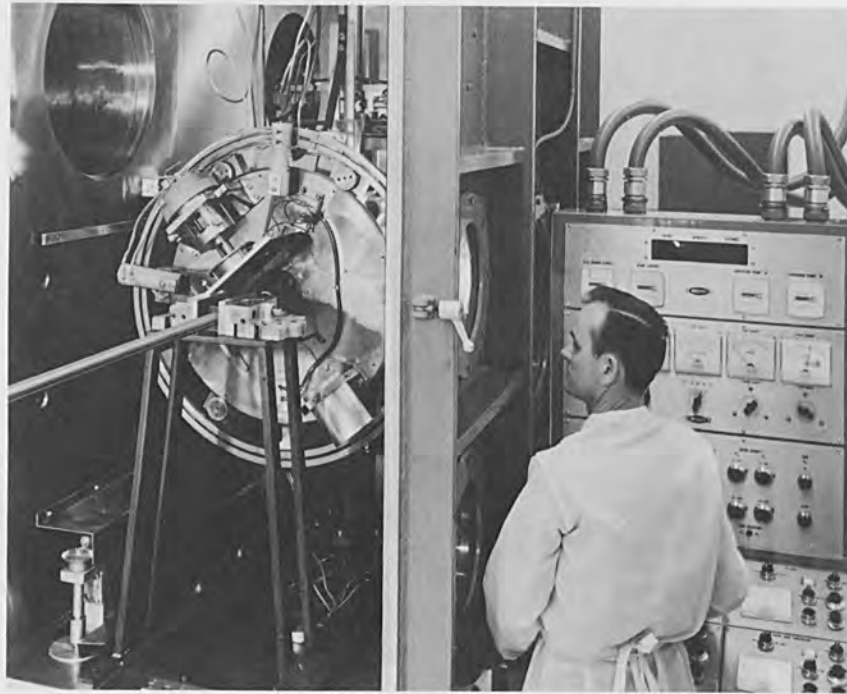


Fig. 7. Welding refractory tubing at NASA.

developed, and weld tests made with this 30-kW gun showed that satisfactory welds in 2-3/8-in. material could be made at a speed of 43 in./min. Inasmuch as the cross section of the Y-ring varied from 3/8 to 2-3/8 in., it was necessary to determine the beam parameters in order to program the current and voltage so that a satisfactory weld could be made on the Y-ring over the full section of the Y-ring. It was determined that the beam power required was directly proportional to the joint thickness for a constant welding speed and a constant weld width. Inasmuch as it is important that the width of the weld be kept constant, in order to avoid differential shrinkage and possible distortion of the assembly, it was decided that the welding would be done at a constant speed with the beam power programmed to be proportional to the thickness of the Y-ring at the point being welded; thus beam power must be programmed as a function of the gun travel along the joint. Since the electron-beam gun utilized was of the Pierce type, which



Fig. 8. NASA complete machine installation.

operates in the space-charge limited region in which current is proportional to the accelerating voltage to the $3/2$ power, the beam power therefore is proportional to the accelerating voltage to the $5/2$ power, or $P = KV^{5/2}$. Inasmuch as the electron-beam welding machine was provided with a regulated high-voltage power supply, whose output voltage could be controlled from a dc reference signal, programming the beam power, by means of varying the accelerating voltage, was easily accomplished.

The first equipment used a linear cam which was coupled to the reference potentiometer in such a way that a displacement of the linear cam, corresponding to a displacement of the gun in relation

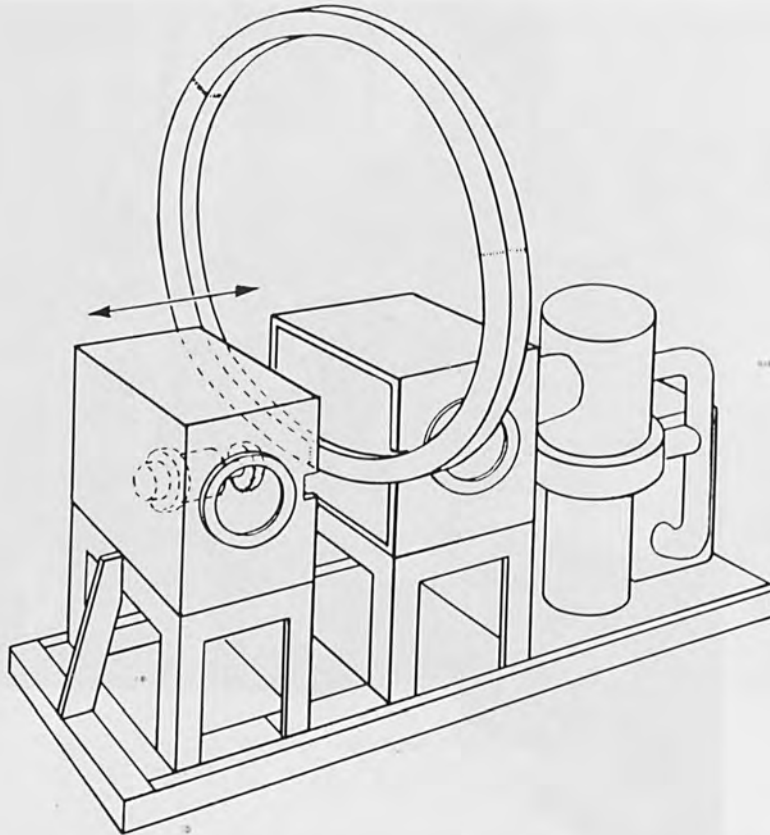


Fig. 9. Local clamp-on chamber concept.

to the work, produced the necessary reference signal to adjust the accelerating voltage to the required level for the beam power necessary for a satisfactory weld at that point along the tapered parts being welded.

Figure 13 shows the relationship of beam voltage, B_V , with Y-ring cross section.

The curve marked P_B , electron-beam power, is a representation of the cross-sectional variation of the part scaled to produce maximum power at the largest cross section. In the final realization of the equipment, in order to allow greater flexibility and ease in setting up the equipment, it was decided to utilize, in place of a

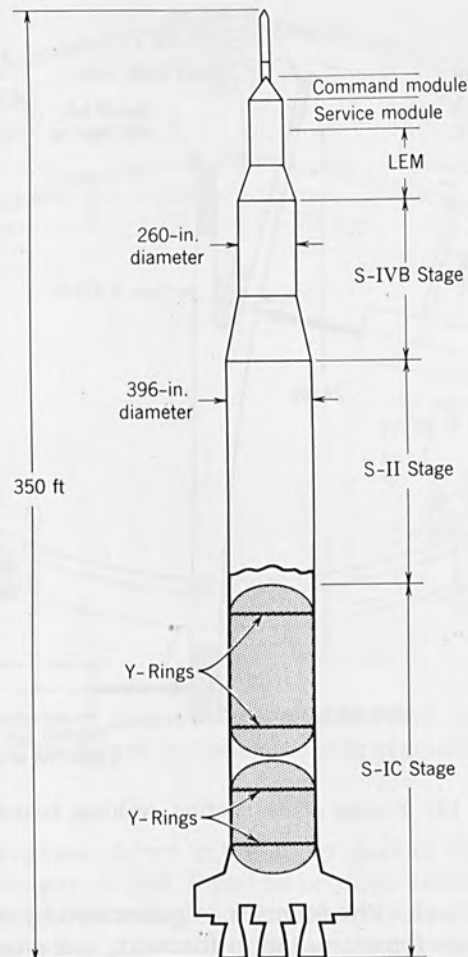


Fig. 10. Placement of the electron-beam welded, 33-ft diameter Y-rings in the S-1C stage.

machined cam, a device which could be easily adjusted so as to readily change the program of beam power versus displacement. The device utilized to generate the analog of the various parameters which have to be controlled was a Vernistat autotransformer. This device is essentially an autotransformer having 33 sets of tapoffs, each of which can easily be adjusted over the length of

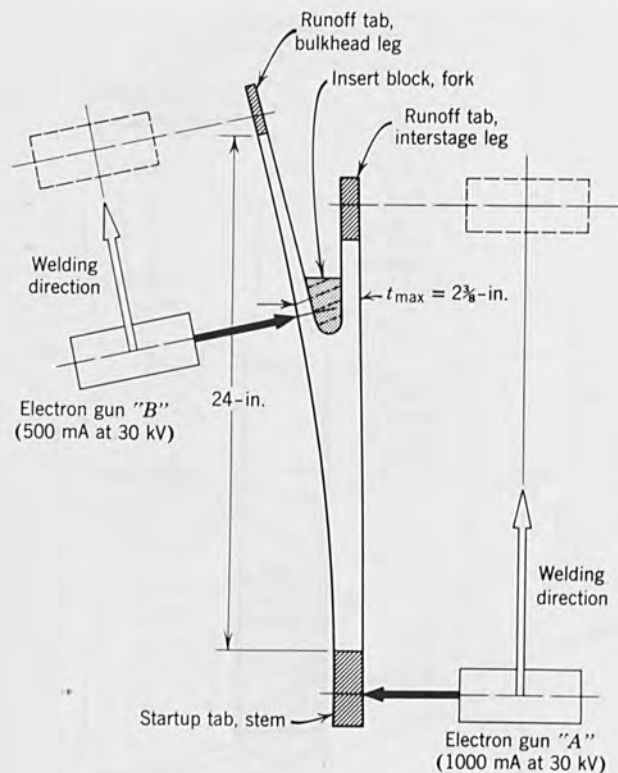


Fig. 11. Y-ring cross section, welding scheme.

the autotransformer. The function is generated by means of a motor which steps from one tap to the next, and samples the voltage at each tap and provides straight-line interpolation between taps. The output of the Vernistat, which varies in relation to the distance along the part to be welded, is fed to the input of the high-voltage regulator, thus developing the required program of power versus distance.

Figure 14 shows the arrangement of the programming control panel as used on the final equipment in which three parameters were controlled: welding speed, gun-to-work distance, and beam power. In welding the final part, two guns were used: Gun "A," having a maximum power of 30 kW, made the first pass in accordance with the program shown in the lower part of Fig. 13. Gun "B," having a maximum power of 15 kW, made the second pass

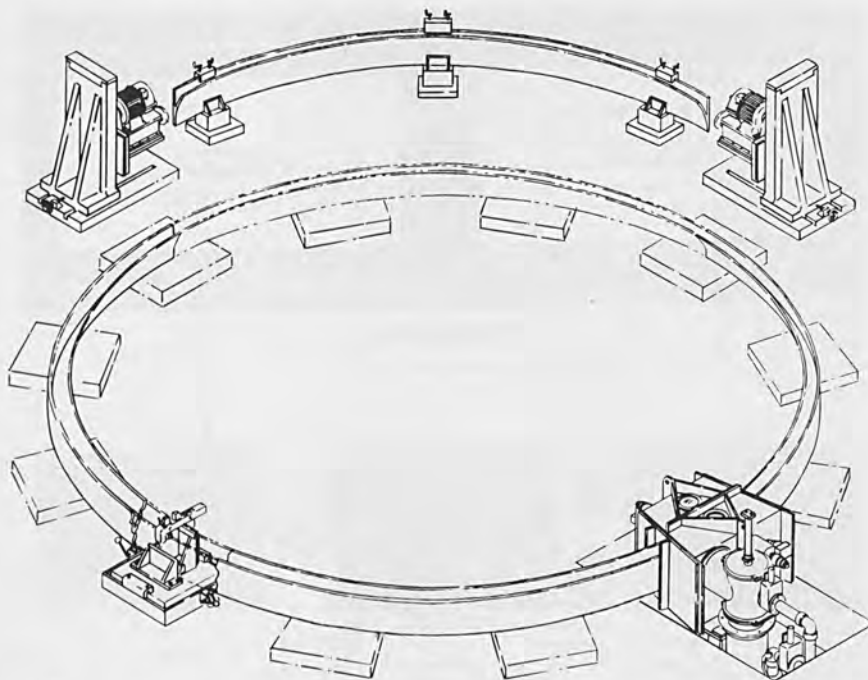


Fig. 12. Thirty-three-ft diameter, 3700#, 2219-T87, γ -ring for the S-1C for butt welding of the forged and machined ring segments.

following the program shown in the upper part of Fig. 13. The gun-to-work distance of Gun *B* had to be programmed in order to maintain a constant distance from gun to work.

When varying beam power by programming accelerating potential, the focus-coil current must also be programmed to maintain the beam in focus. Inasmuch as the focal length of the beam is related to focus-coil current by the equation $F = KV/(NI)^2$, the focus-coil current for a fixed focal length is proportional to the square root of the accelerating potential. In order to avoid the use of two separate analog programmers, one for varying the accelerating potential and the second to vary the focus-coil current, a device was developed which automatically adjusted the focus-coil current to the proper value. A voltage, proportional to the accelerating potential, was delivered to an electronic computing device which extracts the square root of the accelerating potential. This signal in turn provides the command signal for a regulated power

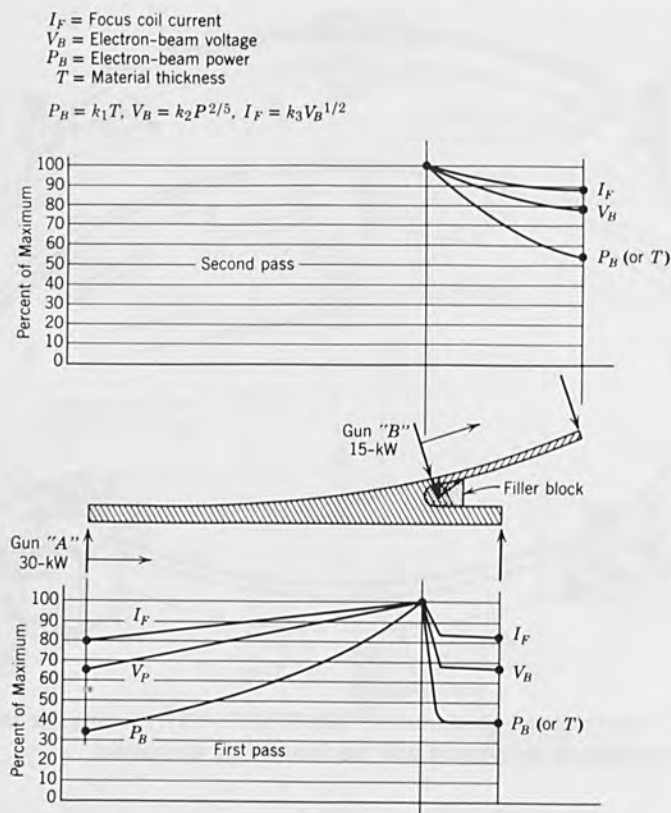


Fig. 13. Relationship of electron-beam welding parameters to Y-ring section thickness.

supply which delivered the focus-coil current in proportion to the signal it received from the square root extractor. In this way, the focus-coil current for any position of the gun relative to the work could be slaved to the accelerating potential; it was then necessary only to determine the proper focus-coil current for the maximum accelerating potential and gun-to-work distance to be used.

Figure 15 is a schematic representation of the method used to automatically program beam power and focus-coil current.

Figure 16 is a reproduction of a graph taken during the welding of a Y-ring section of the S-1C booster.

Trace No. 1 is a recording of the output of the analog program-

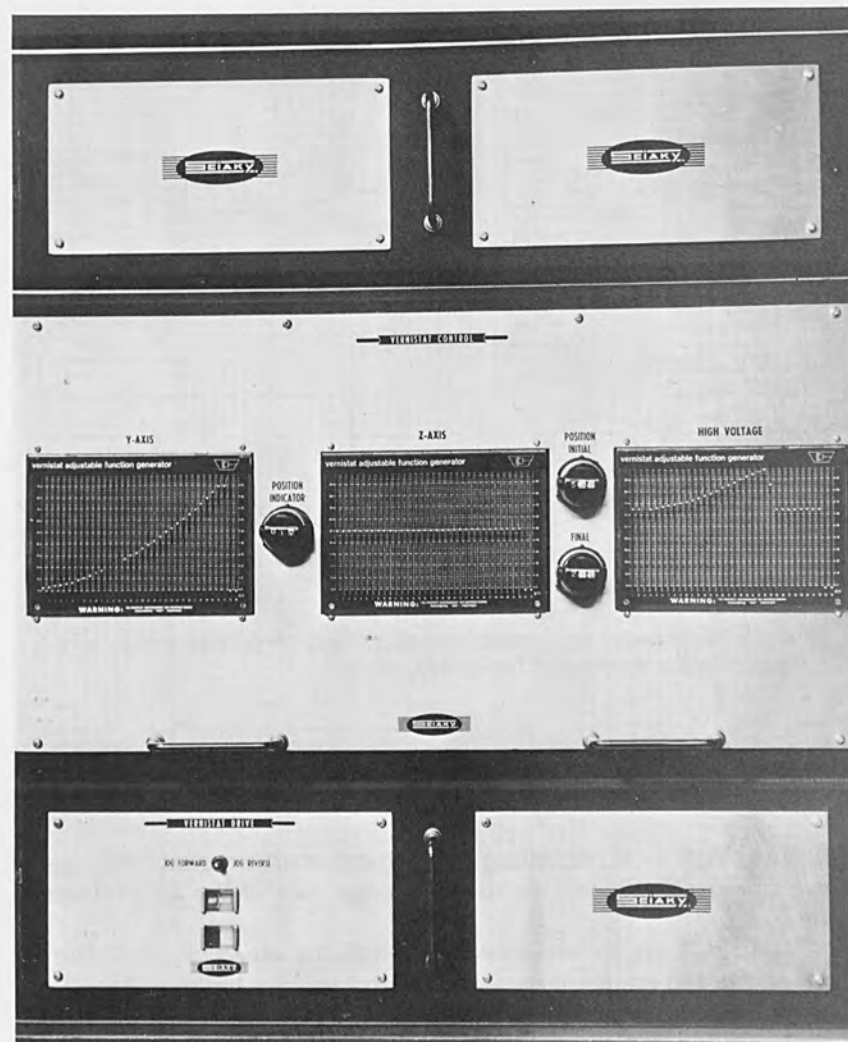


Fig. 14. Analog programmer for Y-ring electron-beam welder.

mer, showing the program of beam power. This output is a voltage following a preset law of variation which is an analog of the desired electron-beam power.

Trace No. 2 is a recording of the accelerating potential applied to the electron-beam gun.

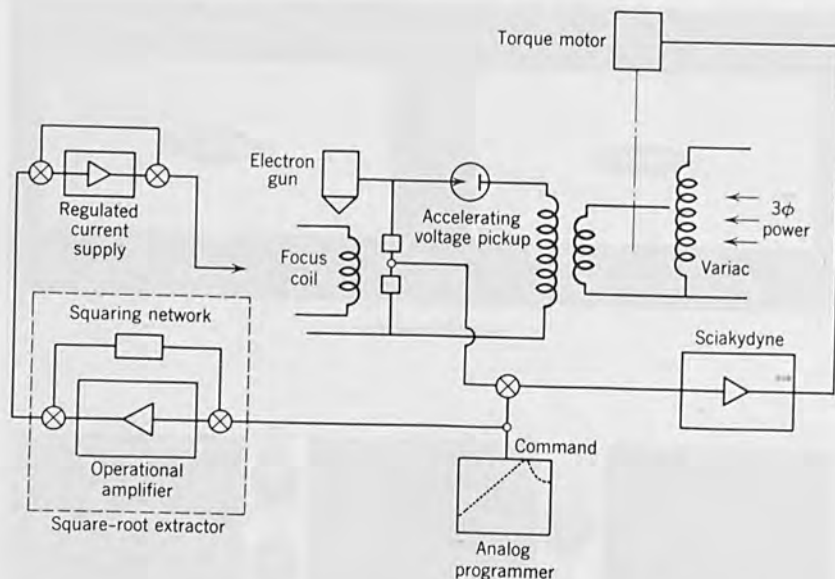


Fig. 15. Schematic representation of method used to automatically program beam power and focus coil current.

Trace No. 3 is a recording of the beam current resulting from the application of the accelerating voltage applied to the electron gun.

Trace No. 4 is a recording of the focus-coil current which is shown to vary in proportion to the square root of the accelerating voltage.

Trace No. 5 shows the travel speed of the electron gun. The trace shows the constant speed of the travel.

Trace No. 6 shows the output of a 40-turn potentiometer which is connected to the gun drive, and is, therefore, the gun position reference. The output voltage of this potentiometer is proportional to gun position. This output is used to drive the programmer so that the power varies as a function of electron gun position, and therefore of material thickness.

Figure 17 is a photograph of Y-ring sections being clamped into the fixture installed in the clamp-on electron-beam chamber. Prior to clamping, both ends of each Y-ring segment are machined simultaneously in a shell end mill to obtain parallelism within 0.0015 in. The segments are then placed on plates with air-bearing

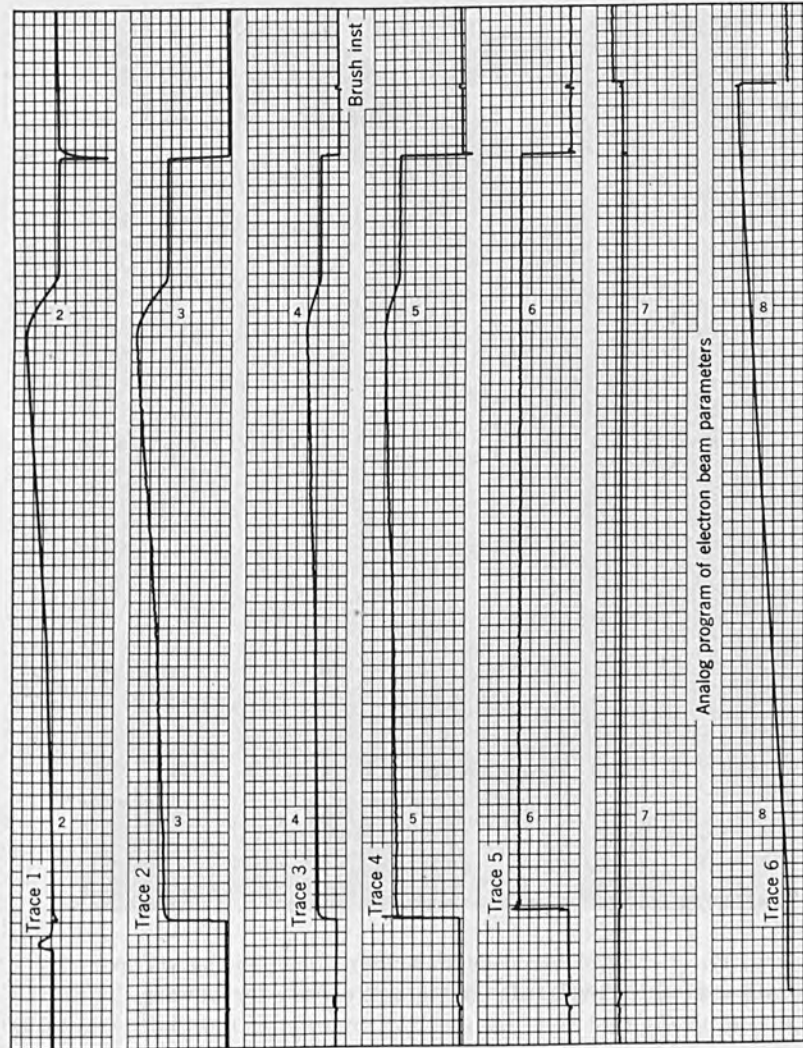


Fig. 16. Record of analog program values for Y-ring electron-beam welding.



Fig. 17. Y-ring electron-beam welder. Chamber shown open with curved Y-ring section being set in place for welding.

supports mounted on pedestals and moved into the chamber. Once in position, the parts are clamped in place and held level, and aligned, using air bearings. When the required fitup is achieved, the segments are located in position. The air bearings become inoperative and the movable half of the chamber is closed.¹

¹Electron-beam welding of large structures by Wm. A. Wilson, P. Gordon Parks, and Robert J. Schwinghamer, "Metal Progress," 1964.

Figure 18 shows sample sections of the Y-ring protruding from the closed electron beam chamber which has completely sealed itself around the Y-ring.

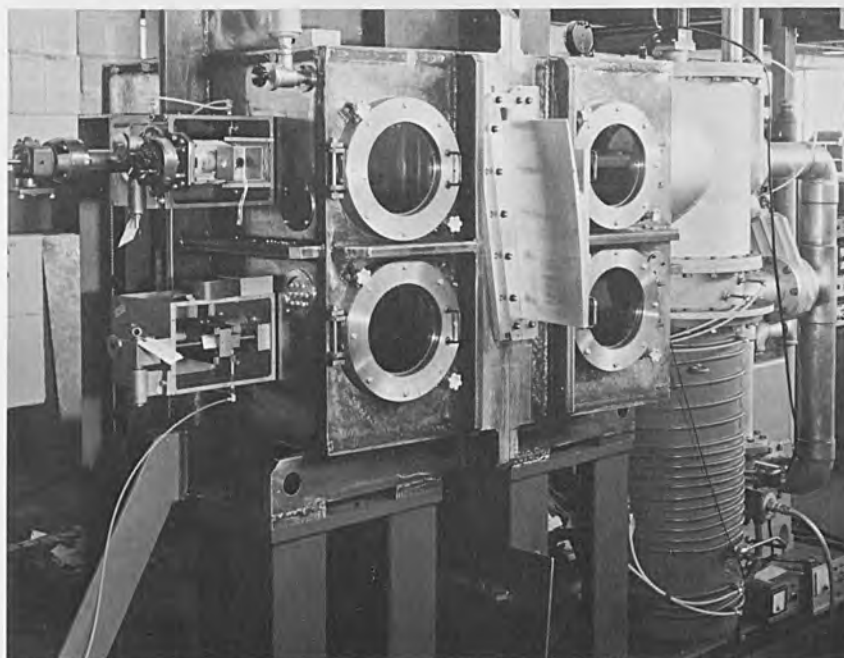


Fig. 18. Y-ring electron-beam welder, showing Y-ring sealed into vacuum chamber.

Figure 19 shows a cross section of the 33-ft diameter Y-ring made of rolled and machined 2219 aluminum, along with a 2-3/8-in. test plate of aluminum which has been butt-welded in the machine.

It is interesting to compare the welding of these Y-rings by the electron-beam process versus the inert-gas, metal-arc process. Table IV compares the two processes, and points out the great advantages of electron-beam welding process for structures of this type. The narrow weld and heat-affected zone resulting from the electron-beam process minimizes, to a great degree, shrinkage and distortion of the structure.

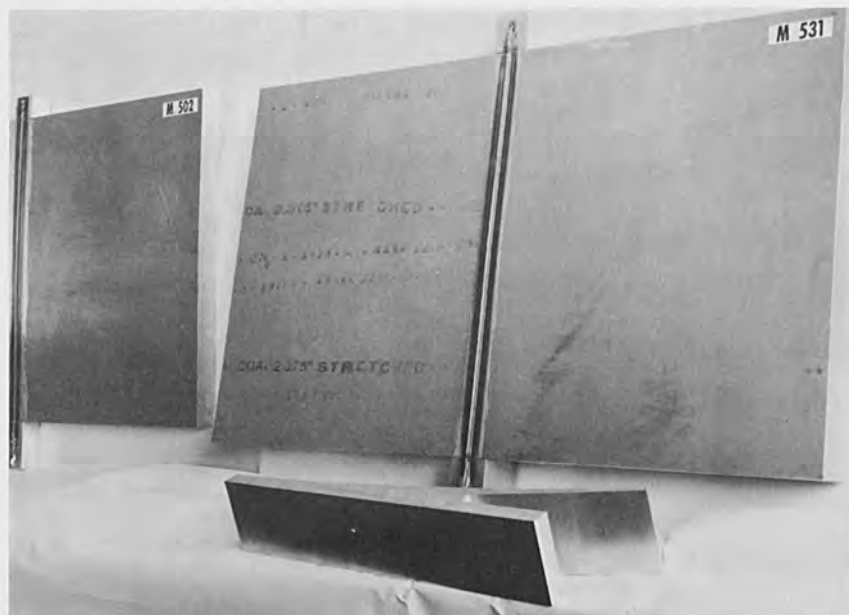


Fig. 19. 2.4-in., thick x 20-in. long 2219 aluminum electron-beam weld 30-kV, 1100-mA, 43 in./min in 4.4 A focus current, 1-1/2-in. gun-to-work, 7-1/2 degrees down gun tilt, fitup.

Electron-beam welding has become an important industrial tool. The advantages in the electron-beam system are due to the high-power density of the electron beam, the purity of the atmosphere in which the weld takes place, the stiffness of the electron beam which allows welding in all positions — downhand, vertical up, or overhead, which allows for the production of narrow deep penetrating welds of high joint efficiency and greatly reduced distortion and warpage. The electron beam has proven to be a very flexible tool by which welds in the very thinnest and thickest materials can be made. Our experience has shown that apparent limitations in the size of components which can be welded may be overcome by utilizing imagination in attacking the welding problem, so that the inherent advantages of electron-beam welding may be retained, although structures larger than the vacuum chamber are to be fabricated.

Table IV
Comparison of Welding Processes for Saturn S-1C Y-Rings

	Electron Beam	Inert-Gas Metal Arc
Passes	2	100
Setup and welding time	8 hr	80 hr
Filler material	none	300 cu in.
Weld quality	potentially high	moderate
Depth-to-width ratio	19 to 1	1 to 5
Joint efficiency	75%	50%
X-rays per joint	2	20
Welding speed	40 in./min.	4 in./min.

RECENT DEVELOPMENTS IN NONVACUUM ELECTRON-BEAM WELDING

INTRODUCTION

Electron-beam welding is becoming a more widely used and better known joining process. The outstanding capabilities of this process range from the assembly of all-welded microelectronic circuitry to the joining of heavy structural sections using a single pass at relatively high welding speeds. Intricate and complex shapes can be fabricated which are difficult and often impossible to construct by more conventional methods and, in addition, a wide variety of materials and material combinations, both metallic and nonmetallic, can be joined. Thus, electron-beam welding has proved to be a flexible, versatile, useful, and practical industrial process.

Basically, electron-beam welding involves the bombardment of the workpiece with a finely focused beam of high-speed electrons. Although the mass of an electron is very small, a stream of electrons at such speeds contains a significant amount of kinetic energy which is transformed into heat when this stream strikes the workpiece. It also is possible to focus and bend the electron beam with suitable electrostatic or electromagnetic fields. The process inherently lends itself to a high degree of controllability.

Because of their very small mass, electrons can easily be deflected upon impact with other bodies of greater mass. Therefore, an electron beam passing through a gas at atmospheric pressure is very quickly scattered and its effectiveness as a metalworking tool is lost. For this reason electron beams in most cases are

*Hamilton Standard, Division of United Aircraft Corporation, Windsor Locks, Conn.

generated, formed, and controlled in regions of low pressure, usually in a vacuum environment of about 10^{-4} mm Hg.

Today, most electron-beam welding machines contain the workpiece in the vacuum environment. There are at least two inherent advantages to be gained from this configuration. First, a vacuum environment is chemically very pure, making possible the welding of highly reactive materials without fear of chemical contamination. The second advantage is the capability of providing an electron-beam system having a long-working distance. Thus, it is possible to focus the electron beam at any point from 0.50 to over 30 in. from the electron optical structure, permitting welding in very inaccessible areas and, in general, adding great flexibility to the welding system. For these reasons plus technical problems associated with maintaining the workpiece at atmospheric pressure, practically all electron-beam welding to date has been conducted with the workpiece in a vacuum chamber.

However, enclosing the workpiece in a vacuum chamber imposes limitations upon the size of the workpiece and upon high-speed, high-production automation of the process unless large or complex chambers and vacuum systems are provided. The use of portable vacuum chambers also tends to be complex and cumbersome. Thus, the development of a practical system for electron-beam welding with the workpiece at atmospheric pressure would significantly increase the scope of application of this welding technique.

Two basic technical problems must be solved in order to produce a practical nonvacuum electron-beam welding system. First, a technique must be developed which permits the electron beam to escape from the vacuum environment in which it is formed and controlled into a region of gas at atmospheric pressure without significant loss of power. Second, the electron beam must be such as to provide a reasonable working distance external of the gun structure before scattering reduces its power density to levels too slow for metalworking.

The first problem is the more difficult to solve. Because of the high-power density necessary for electron-beam welding, thin low-density "windows" which would transmit electron beams of low-power density are not practical. A more practical system is a technique for staging the vacuum over a very short length using an orifice system through which the electron beam can pass. To be practical, this orifice system must be large enough to transmit the electron beam without significant loss of power, must be designed to minimize the pumping requirements for reasonable pres-

sure drop across each orifice, and must contain provisions to prevent clogging during extended welding operations.

The second problem can be solved quite readily by using a beam of relatively high-speed electrons, that is, a beam generated at relatively high voltage. It is well known that the lower the accelerating voltage, the more rapidly an electron beam becomes diffused when passing through a gas at atmospheric pressure. Thus, as the voltage is increased, it is possible to retain a significant power density at greater distances. At least 120,000 V are required for a practical working distance; a reasonable and practical range is 120,000 to 200,000 V. Today, a major portion of vacuum electron-beam welding equipment operates in the 120,000 to 150,000 range.

EQUIPMENT DEVELOPMENT

For the past two years, Hamilton Standard has carefully and extensively investigated the problems associated with nonvacuum electron-beam welding. Fortunately, production-type Hamilton Standard electron-beam welding equipment has a normal operating range up to 150,000 V, making this equipment admirably suited for conversion to nonvacuum welding. The electron gun and magnetic lens, together with the low-voltage supplies and controls, can be used without significant modification. Minor modification of the high-voltage supply permits operation up to 175,000 V. Thus, the problem of obtaining a practical working distance was relatively quickly and easily solved.

The more major problem, that of providing an escape passage for the electrons, required detailed analytical and development effort. Although we are not prepared at this time to disclose the details of the solution arrived at, it can be stated that a unique and practical solution has been developed. Transmission efficiencies in excess of 90% have been obtained, pumping requirements have been significantly minimized, and orifice clogging has been avoided.

In addition to the two major considerations previously discussed, it is necessary to provide inert gas protection of the workpiece during welding and shielding against X-rays which are emitted when the electron beam strikes the workpiece. Both of these are very straightforward problems which have been solved by conventional means.

Figure 1 is a photograph of a laboratory prototype nonvacuum electron beam welding machine. It is a modification of standard 6 kW equipment with no effort having been made to integrate and



Fig. 1. Laboratory prototype—nonvacuum electron-beam welder.

package the system for manufacture and use in production-type application. This laboratory unit operates very successfully at voltages up to 175,000 V and at powers up to 7 kW.

CAPABILITY EVALUATION

A significant amount of testing has been accomplished to determine the effects of nonvacuum electron-beam welding parameters and, also, to evaluate the current state-of-the-art welding capabilities.

For example, it has been shown that the deep-welding phenomenon associated with vacuum electron-beam welding also is encountered with the nonvacuum technique. Figure 2 is a cross-sectional view of a butt weld in 0.280-in. stainless steel. Although the depth-to-width ratio of the fusion zone is not as great as that normally experienced when welding in a vacuum, the deep-welding phenomenon certainly has been encountered. Depth-to-width ratio undoubtedly will increase as further work is accomplished.



Fig. 2. Nonvacuum electron-beam butt weld—AISI 302 stainless steel.

As expected, depth of penetration is strongly dependent upon accelerating voltage. Figure 3 illustrates this with a plot of penetration versus accelerating voltage for constant power input. This data confirms the requirement for high-voltage equipment.

Also, as expected, the depth of penetration decreases with an increase in working distance, that is, the distance between the orifice and the workpiece. Figure 4 shows this effect. Decrease in penetration results from lower power density caused by scattering of the electron beam during passage through gas at atmospheric pressure. However, as can be seen, working distances up to at least 1/2 in. yield practical welding results.

The type of gaseous atmosphere through which the electrons

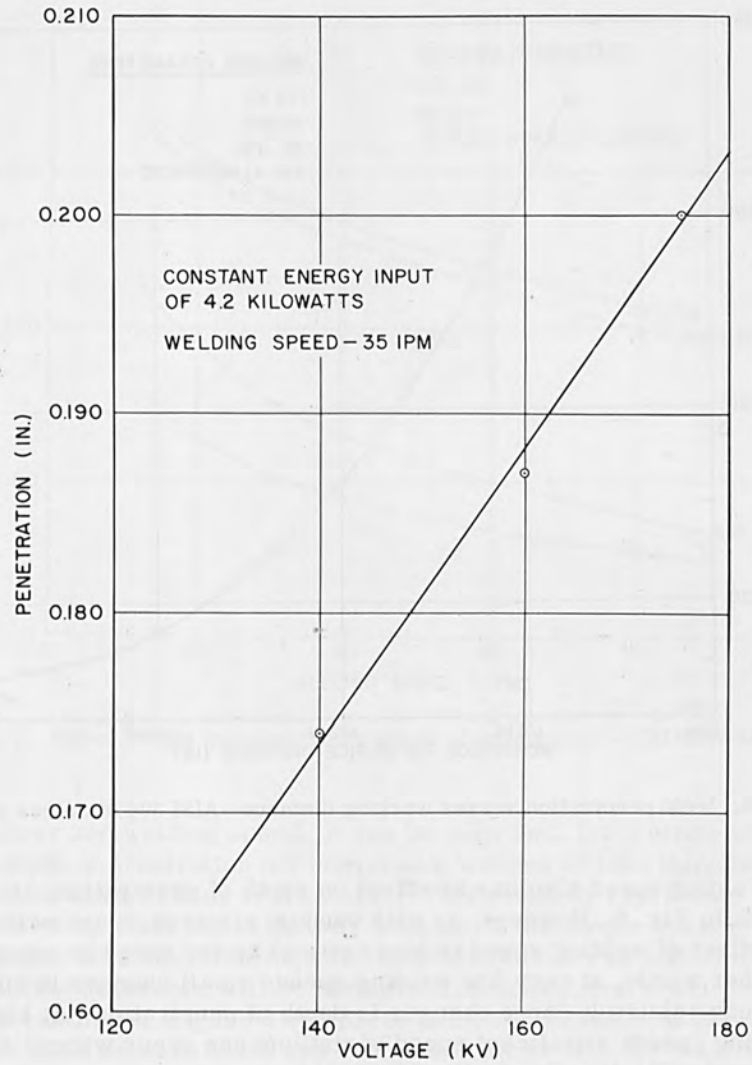


Fig. 3. Weld penetration versus accelerating voltage—AISI 304 stainless steel.

must pass also has a significant effect on scattering and, thus, on weld penetration. This is illustrated in Fig. 5. Again, as expected, the lower density of the protective gas used, the greater is the depth of penetration. Thus, helium gives significantly greater penetration than does argon.

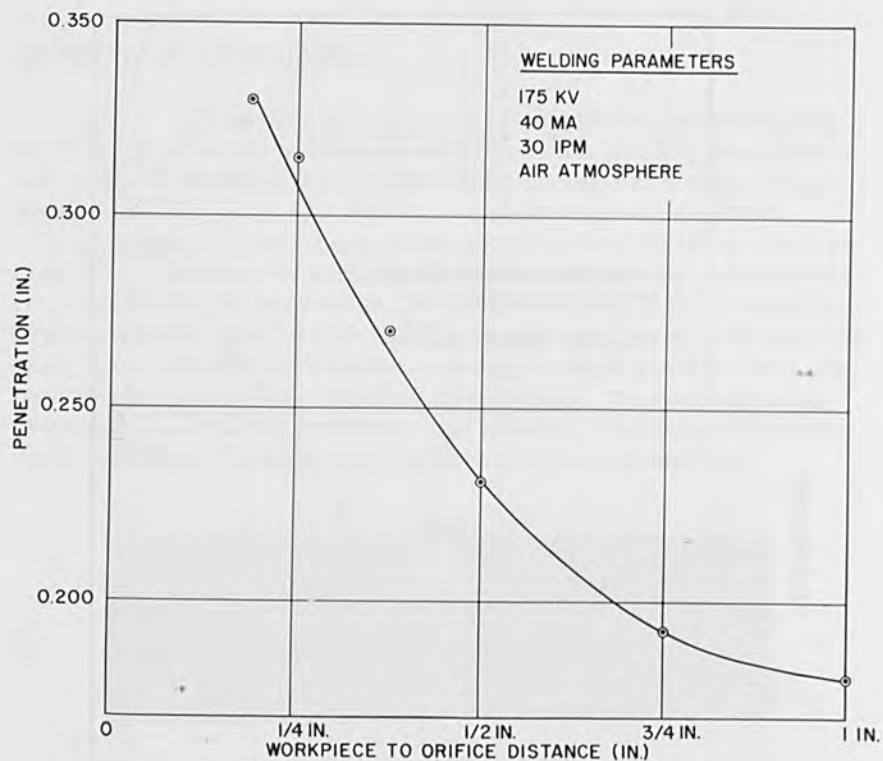


Fig. 4. Weld penetration versus working distance—AISI 304 stainless steel.

Welding speed also has an effect on depth of penetration, as shown in Fig. 6. However, as with vacuum electron-beam welding, the effect of welding speed is less critical as the speed increases. In other words, at very low welding speeds small changes in speed produce relatively large changes in depth of penetration. At higher welding speeds significant speed variations can occur without significantly affecting the depth of the weld.

Most of the nonvacuum welding work to date has been accomplished with four materials: 18-8 austenitic stainless steel, AISI 4340 CrNiMo low-alloy steel, Rene 41 nickel base high-temperature alloy, and TZM molybdenum alloy. Limited work has been done with other materials.

Figure 7 presents data on weld depth versus power in 18-8 austenitic stainless steel and compares these results with similar data obtained when welding in a vacuum under the same conditions

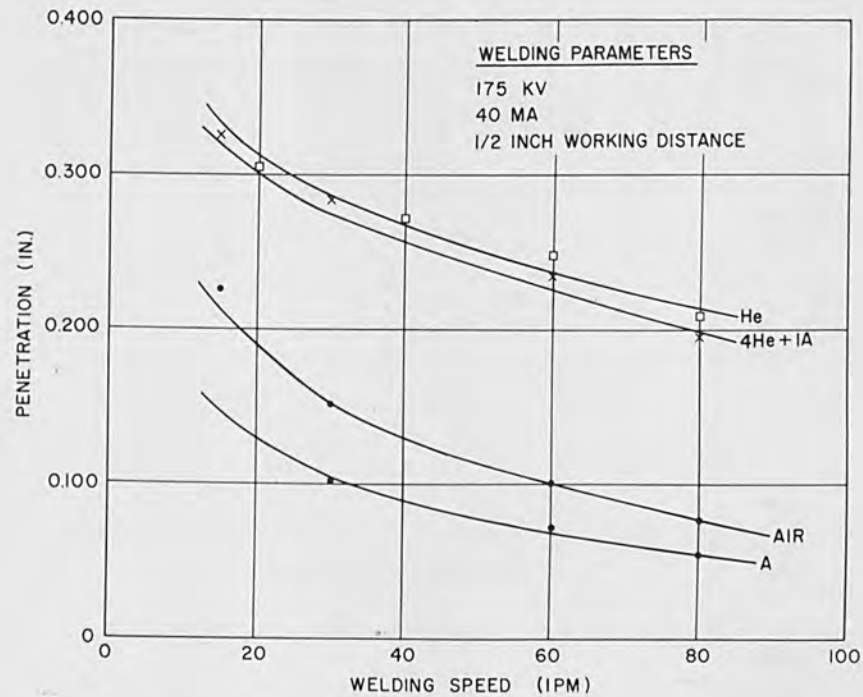


Fig. 5. Effect of gas atmosphere on depth of penetration—AISI 4340 steel.

of power and welding speed. It can be seen that, for a given power, the depth of penetration for nonvacuum welding is less than that obtained when welding in a vacuum. This probably represents a decrease in beam power density because of the larger beam cross-sectional area experienced with the nonvacuum technique. Undoubtedly, this difference will be minimized as nonvacuum development work continues. Figure 8 compares photomicrographs of the transition zone between fused and base material in AISI 304 stainless steel welded by both techniques. The characteristics are quite similar, although the structure in the fusion zone of the vacuum-welded material is somewhat finer than that of the nonvacuum material. This probably is the result of the relatively greater amount of fused material in the nonvacuum-welded sample with a resulting somewhat slower cooling rate.

Figure 9 presents tensile data on nonvacuum, electron-beam welded 0.250-in. thick annealed austenitic stainless steel with the weld surface machined slightly undersize to produce fracture in

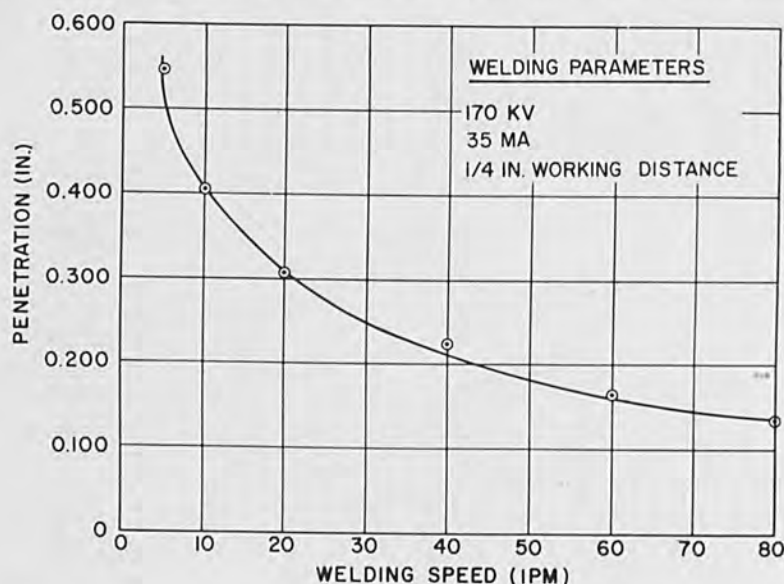


Fig. 6. Weld penetration versus welding speed—AISI 304 stainless steel.

that region. It can be seen that these data compare very favorably with handbook nominal and guaranteed tensile strength values. Figure 10 is a statistical plot of tensile strength versus probability of fracture for 0.062-in. annealed austenitic stainless steel sheet. In this case, tests were made on the as-welded specimen without surface preparation. Results are plotted statistically combining data on vacuum welds with data on nonvacuum welds. All data points fall on the same straight line indicating that statistically the nonvacuum welds have strengths equal to those of vacuum welds, and that both have strengths equal to the base metal.

Figure 11 presents photographs of nonvacuum welds in AISI 4340 steel in a thickness of 0.250 in. One weld was made in air at 35 in./min and the other in a mixture of three parts of helium and one part of argon at 60 in./min. Both welds are sound, but it can be seen that the weld made in the helium-argon mixture is somewhat narrower. Figure 12 compares photomicrographs of the transition zones in these two weld specimens. Figure 13 is a plot of penetration versus welding speed for AISI 4340 in a helium-argon atmosphere using working distances from 3/16 to 1/2 in. This shows the effects of both working distance and welding speed.

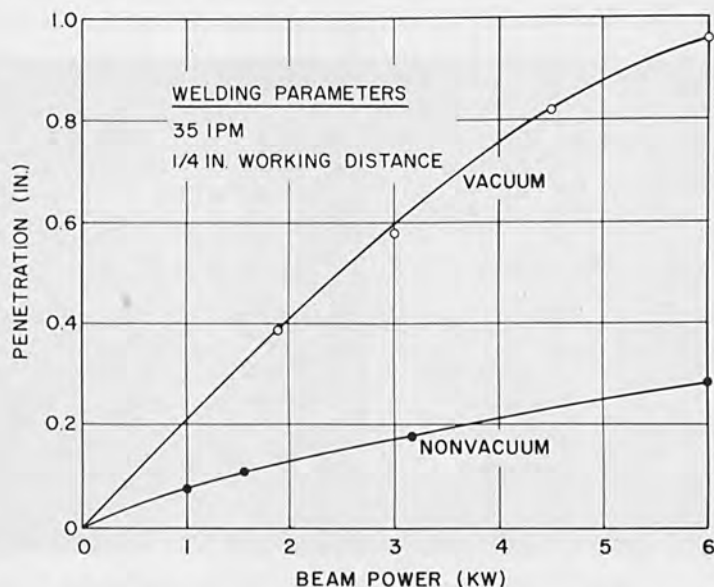


Fig. 7. Vacuum and nonvacuum weld penetration versus power—AISI 304 stainless steel.

Figure 14 is a photograph of a cross section through bead-on-plate passes made in a block of Rene 41. These were all run in an air atmosphere at 175 kV and 40 mA; three welding speeds, 60, 30, and 15 in./min, were used. Figure 15 is a plot of penetration versus welding speed for Rene 41 using a gas mixture of four parts of helium and one part of argon. Working distances from 3/16 to 1 in. are shown.

Figure 16 shows photographs of the cross section, face, and root of a non-vacuum electron-beam weld in TZM molybdenum made in a helium atmosphere at 7 kW, 15 in./min, and a 1/2-in. working distance. Figure 17 is a photomicrograph of the transition region of this weld. Radiographic inspection showed the weld to be very sound. Figure 18 presents curves of penetration versus welding speed in this alloy at working distances from 3/16 to 1 in. in a helium atmosphere.

Figure 19 is a cross-sectional view of an actual butt weld of 0.375-in., 2219 aluminum alloy plate. This was welded at 7 kW, 60 in./min, and 3/8-in. working distance in a helium atmosphere.

Figure 20 is a chart listing typical nonvacuum-welding para-

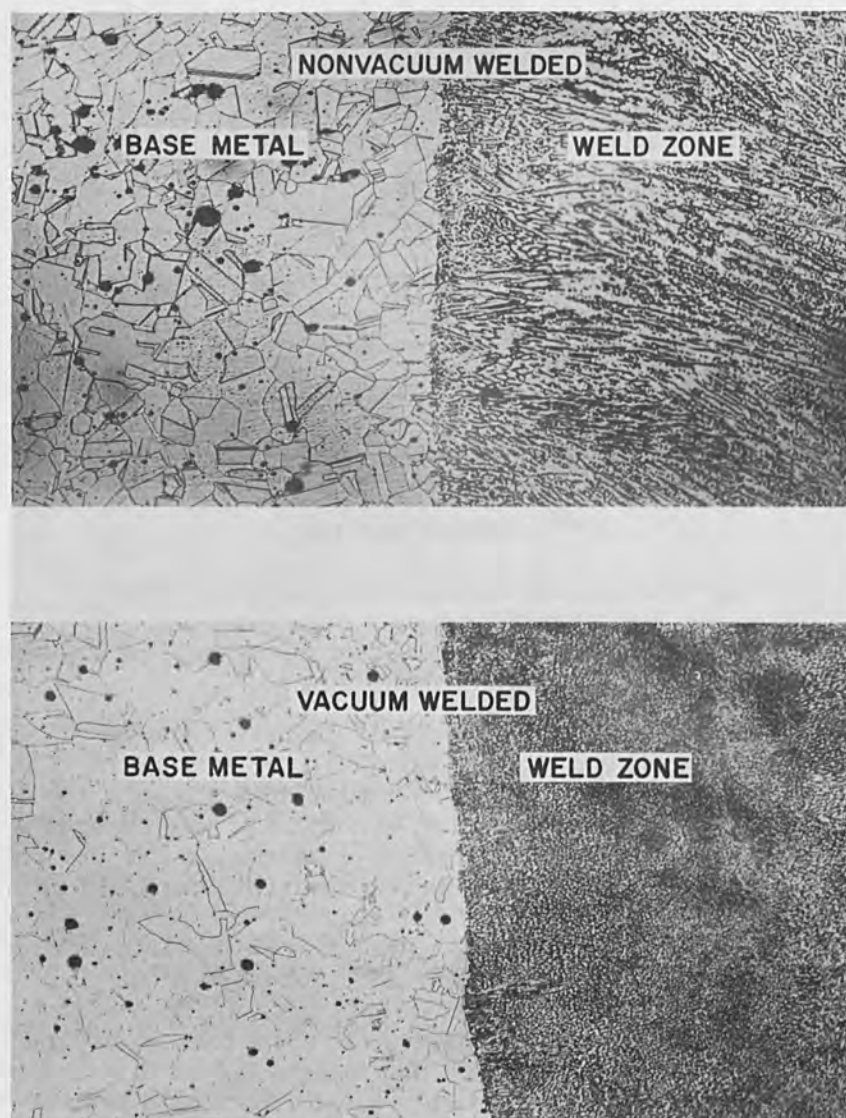


Fig. 8. Photomicrographs of electron-beam vacuum and nonvacuum butt welds—AISI 302 stainless steel (0.062-in. thick annealed sheet).

SPECIMEN	ULTIMATE TENSILE STRENGTH (PSI)	YIELD STRENGTH 0.2% OFFSET (PSI)	% ELONGATION IN 2 IN.	LOADING SPEED (IPM)
<u>WELDED SPECIMEN</u>				
1	82,600	35,300	57	0.05
2	81,900	36,200	53	0.05
3	84,000	36,400	63	0.05
4	84,100	37,100	49	0.05
<u>BASE METAL</u>				
NOMINAL ¹	85,000	35,000	55	—
GUARANTEED ² MINIMUM	80,000	30,000	50	—

WELD REINFORCEMENT ON THE TOP AND ROOT WAS
REMOVED PRIOR TO TESTING.

1 — METALS HANDBOOK, VOL. I PROPERTIES AND SELECTION
OF METALS 8TH, 1961, p 414.

2 — STAINLESS STEEL HANDBOOK, ALLEGHENY LUDLUM
STEEL CORP. 1956, p 2.

Fig. 9. Tensile test data—nonvacuum electron-beam welded AISI 304 stain-
less steel (0.250-in. thick annealed sheet).

meters for several different materials in different thicknesses.
This shows the wide range of capabilities of this process.

CONCLUSIONS

Experimental work conducted to date indicates that nonvacuum electron-beam welding is both a practical and useful welding technique. Results show that welds produced by this process are quite similar to those produced by vacuum electron-beam welding, and that these welds have excellent properties. This technique has the

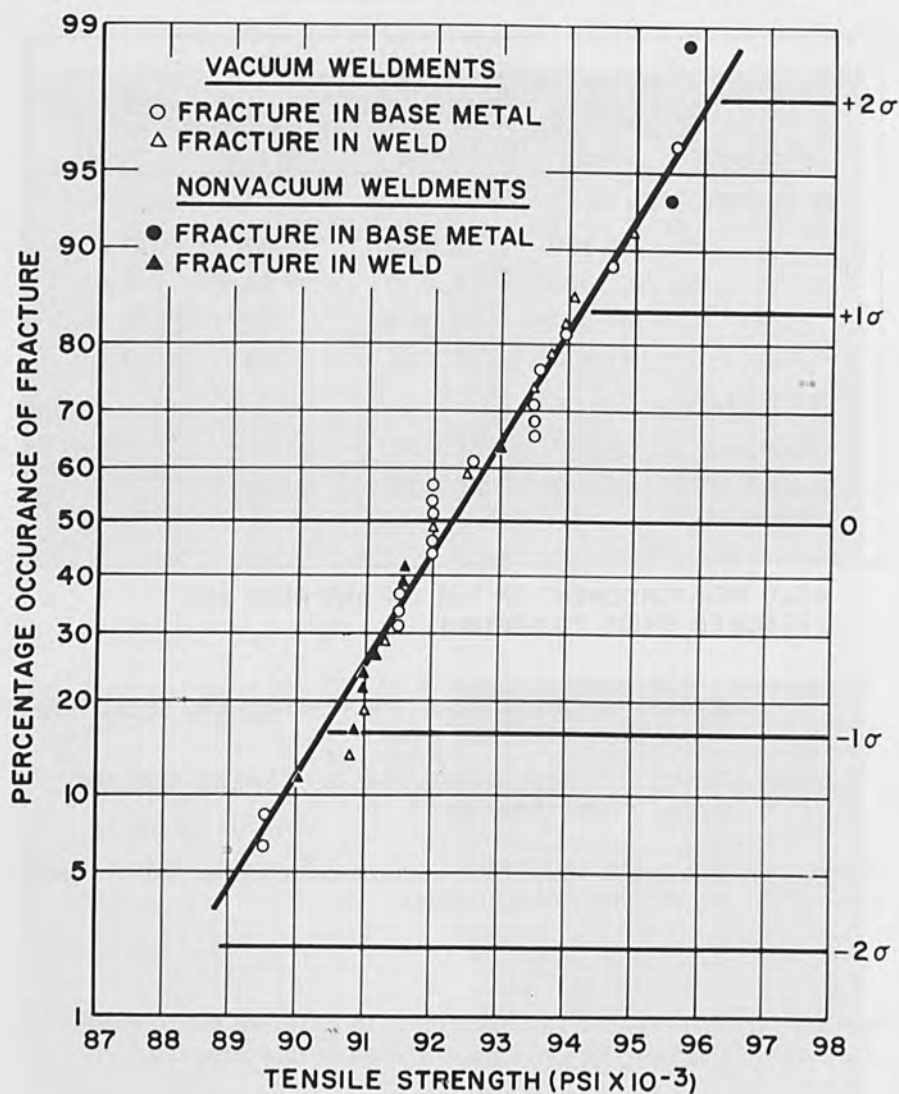


Fig. 10. Tensile strength of vacuum and nonvacuum electron-beam butt welds—AISI 302 stainless steel (0.062-in. thick annealed sheet).

advantages that no large or complex work chambers and vacuum systems are required and that high-speed, high-production processing is greatly simplified. It has the disadvantage that the working distance below the gun structure is somewhat limited, restrict-

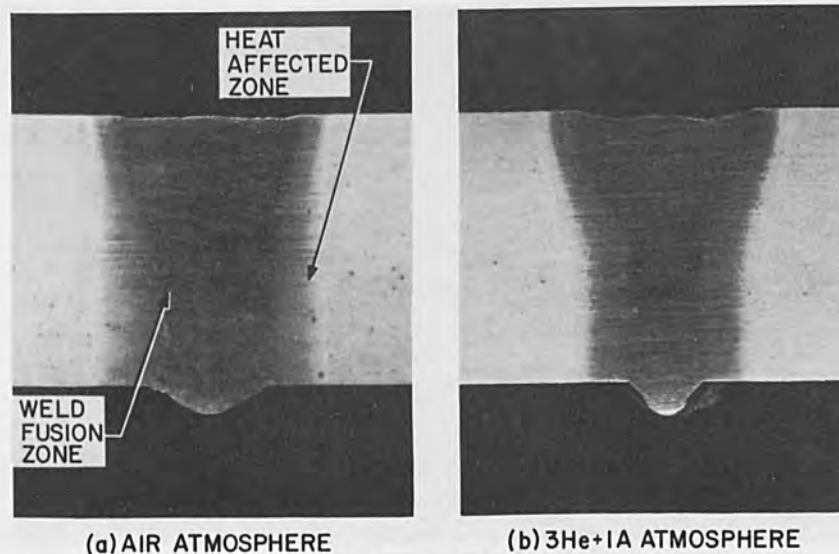
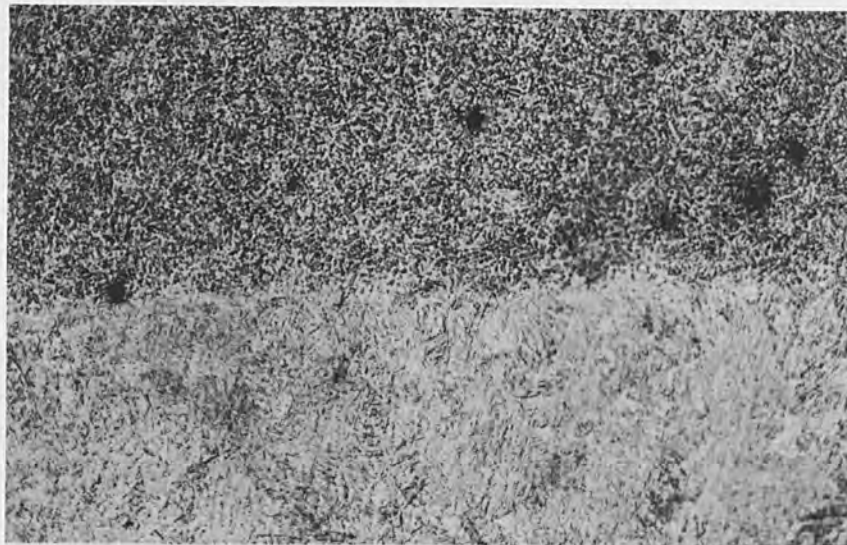


Fig. 11. Nonvacuum electron-beam butt welds—AISI 4340.

ing the shape of the workpiece being welded and also preventing welding in inaccessible areas. It is expected that this technique will supplement vacuum electron-beam welding, but it will not replace it. Widespread industrial interest in this technique already has been aroused. Potential areas of application of the process include the fabrication of thin and heavy-walled tubing, metal containers, missile cases, ship structures, and automotive parts. In general, any component requiring high-quality welding at high speed and having a reasonably smooth contour which permits access of the beam exit orifice close to the weld joint is a worthy candidate for this process. This development undoubtedly will significantly expand the roll of electron-beam welding in the metal joining industry.

ACKNOWLEDGMENTS

The author wishes to acknowledge sponsorship of some of the evaluation effort by the Fabrication Branch (Mr. F. R. Miller), Manufacturing Technology Division, Air Force Systems Command, Aeronautical Systems Division of the United States Air Force. He



(a) AIR ATMOSPHERE



(b) $3\text{He}+1\text{A}$ ATMOSPHERE

Fig. 12. Photomicrographs of nonvacuum electron-beam butt welds—AISI 4340.

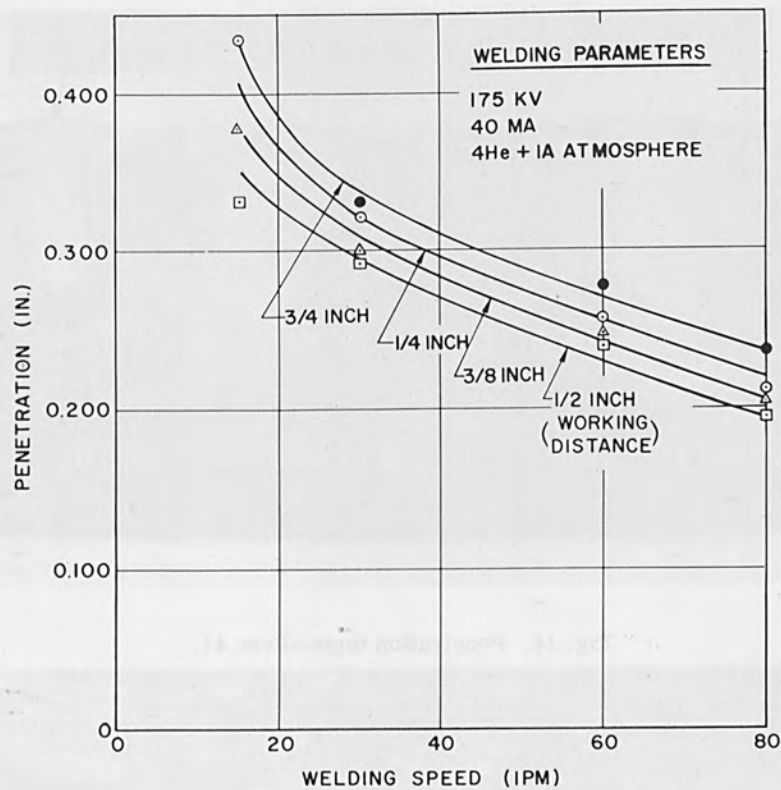


Fig. 13. Weld penetration versus welding speed—AISI 4340.

also wishes to acknowledge the efforts of Messrs. R. F. Donovan, R. F. Duhamel, R. M. Niedzielski, and F. R. Schollhammer in this program.

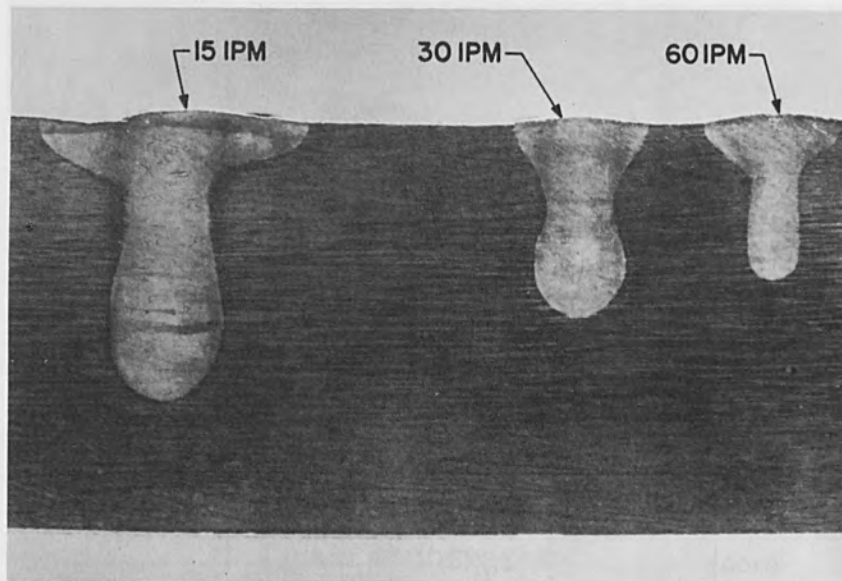


Fig. 14. Penetration tests—Rene 41.

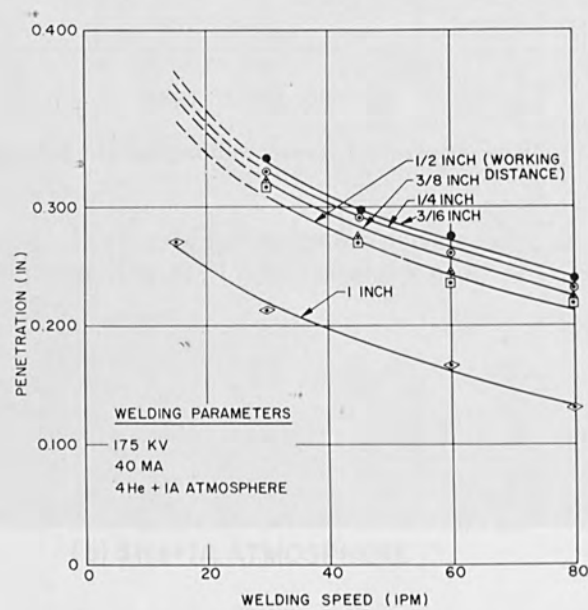


Fig. 15. Weld penetration versus welding speed—Rene 41.

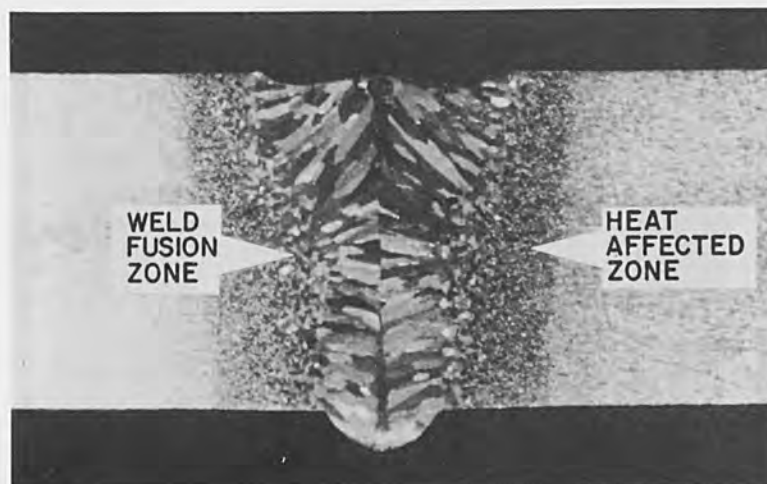


Fig. 16. Nonvacuum electron-beam butt weld—TZM molybdenum—helium atmosphere.

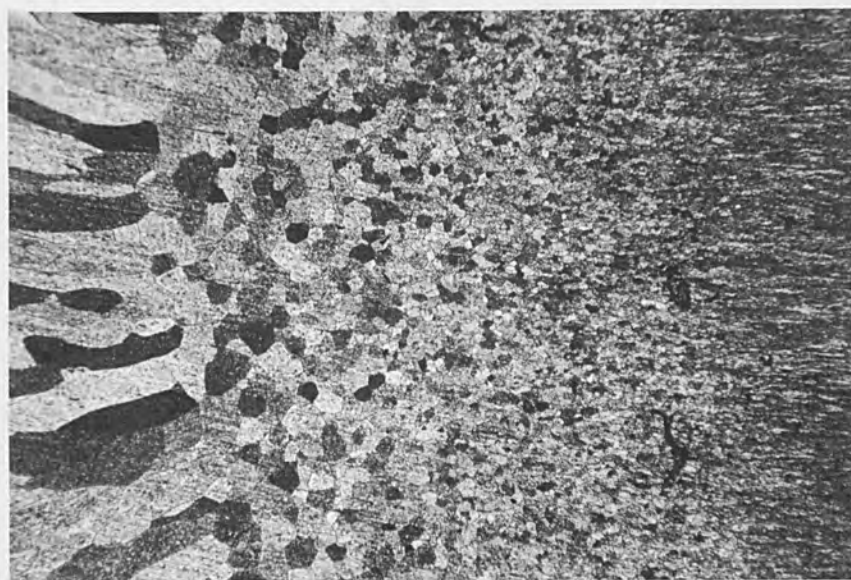


Fig. 17. Photomicrograph of nonvacuum electron-beam butt weld—TZM molybdenum—helium atmosphere.

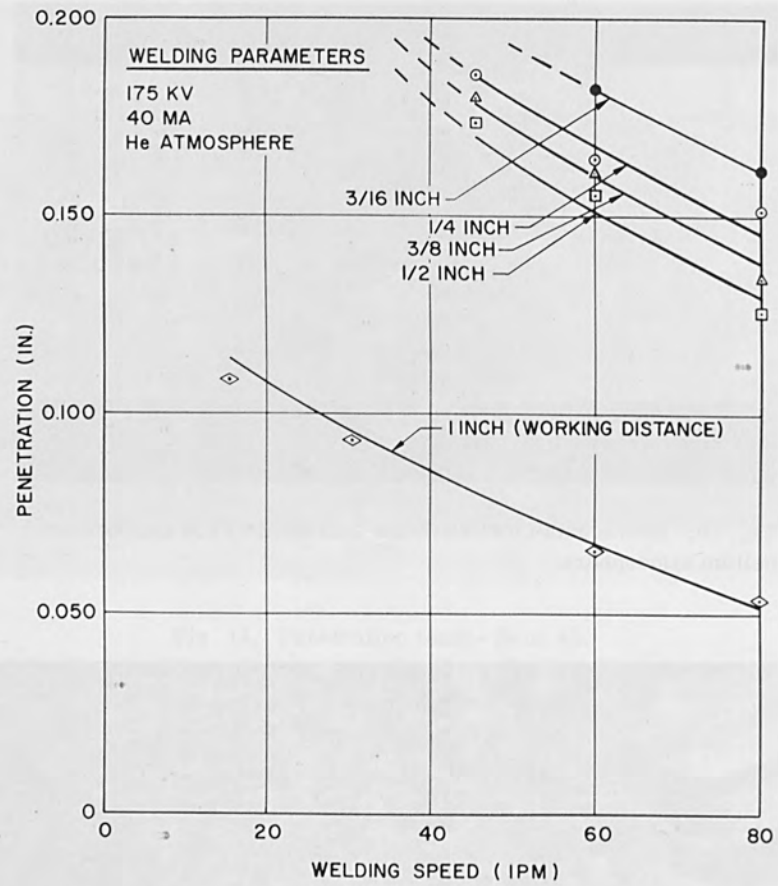


Fig. 18. Weld penetration versus welding speed—TZM molybdenum.

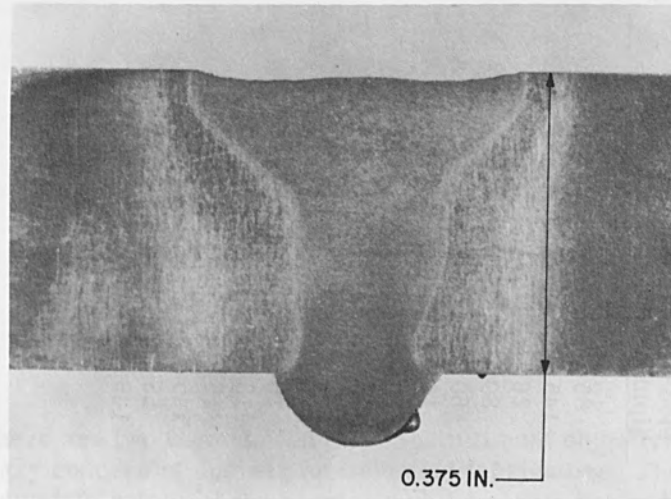


Fig. 19. Nonvacuum electron-beam butt weld—2219 aluminum.

Figure 20 Nonvacuum electron-beam welding parameters.

Material	Thickness (in.)	Voltage (kV)	Current (mA)	Welding Speed (ipm)	Working Distance (in.)	Atmosphere
Killed steel	0.125	175	30	85	1/4	air
SAE 1010 steel	0.010	150	2.5	145	1/4	air
SAE 4620 steel	0.400	175	40	26	3/16	air
AISI 4130 steel	0.025	175	4	80	1/4	4He-1A
AISI 4340 steel	0.250	175	40	60	1/2	3He-1A
AISI 4340 steel	0.430	175	40	15	3/16	4He-1A
AISI 321 stainless steel	0.060	175	20.5	130	1/4	air
AISI 304 stainless steel	0.280	175	29	36	1/4	air
AISI 304 stainless steel	0.400	170	35	10	1/4	air
Rene 41 high-temperature alloy	0.225	175	40	80	1/2	4He-1A
Rene 41 high-temperature alloy	0.275	175	40	15	1	4He-1A
Inconel X	0.125	150	25	18	3/16	air
TZM molybdenum	0.187	175	40	15	1/2	He
5052 aluminum alloy	0.0093	150	3	145	1/4	air
2219 aluminum alloy	0.375	175	40	55	3/8	He
Copper	0.008	150	8	145	1/4	air

ELECTRON-BEAM WELDING OF NUCLEAR FUEL CLADDING COMPONENTS

There are two factors which are the ultimate objectives and the primary concern of nuclear fuel element fabricators. These are the complete safety of the reactor and the ability to produce high heat generation rates. In the Advance Fuel Development Operation at Hanford Laboratories, we strive to stay three to five years ahead of the present-day fuel concepts. To do this, it is necessary to design, fabricate, and evaluate unique fuel elements.

Early in 1963, a program was initiated to demonstrate the flexibility of the electron-beam welding process and the feasibility of welding large, unique fuel element shapes that cannot be readily fabricated by other methods. Sections of each type of fabrication were thoroughly examined by destructive and nondestructive techniques to determine depth of penetration, extent of heat-affected zone, and the amount of porosity.

Aside from the ability of the electron-beam equipment to make unusual welds, we found that there are special considerations that must be taken into account which were not of serious importance when more conventional welding methods were employed. These are: the size of the weldment, fit-up, the fixturing, and the work carriages.

Size of Weldment. Size and complexity of the equipment are increased when the physical size of the weldment is increased. A longitudinal weld on a 1-ft-long section is relatively easy to make,

*Advance Fuel Development, Reactor and Fuels Laboratory, Hanford Laboratories, General Electric Co., Hanford, Wash. Work performed under Contract No. AT(45-1)-1350 between the Atomic Energy Commission and the General Electric Company.

but an 8-ft section can be considerably more difficult. When spacer ribs were welded to the outside of a 1-ft-long tube, distortion of the ribs was negligible. Little trouble from rib distortion or tube bow was encountered when this ribbed tube was inserted into a tube which had an inside diameter (I.D.) the same dimension as the outside diameter (O.D.) of the ribs, and "blind" burn-through welds were made through the outer tube into the ribs (Fig. 1). Distortion of ribs (and tube), when welded to an 8-ft-long tube, would not be negligible without elaborate fixturing to restrain the rib and tube. The burn-through weld is considerably more difficult to make on the 8-ft-long section than on the 1-ft-long section because the distorted rib cannot be seen through the outer tube. Also, since tube bow varies with length, good fitup and the burnthrough weld would be difficult to achieve.

Cost of weldment, then, increases with length. Fixturing is necessarily more elaborate and more care is required to assure quality work.

Fit-up. The fit-up of the pieces to be joined is important when conventional welding processes are used, but the importance is vastly increased when electron-beam welding. This is due to the extremely small diameter beam impingement area. Welding longitudinal ribs to tubes is an example. There is essentially a point contact between the rib and the tube. If there is a gap between the rib and tube of as little as 0.015 in. at one point, no weld may occur at that point or, at best, a thinning of the weld metal near the tube will take place.

Another example of necessity of good fit-up is vividly illustrated in Fig. 1. The bottom left photograph shows the result of a gap between the rib and the inside of the outer tube. The bottom right photograph indicates the results obtained from a good fitup of the same type of joint.

Fixturing. Fixturing plays an important part in obtaining quality welds on large, long, or intricately shaped pieces. For research and development work, effective fixturing can be kept simple; however, production of a large number of long pieces may change the fixturing picture. Drive mechanisms may be introduced to rotate the part into a new position after each weld, automatic clamping devices may be required that can release parts and reclamp after positioning the pieces, storage and feeding mechanisms may be needed to supply parts and all must be done in such a manner that the final product is a precise fabrication. Cost of elaborate fix-

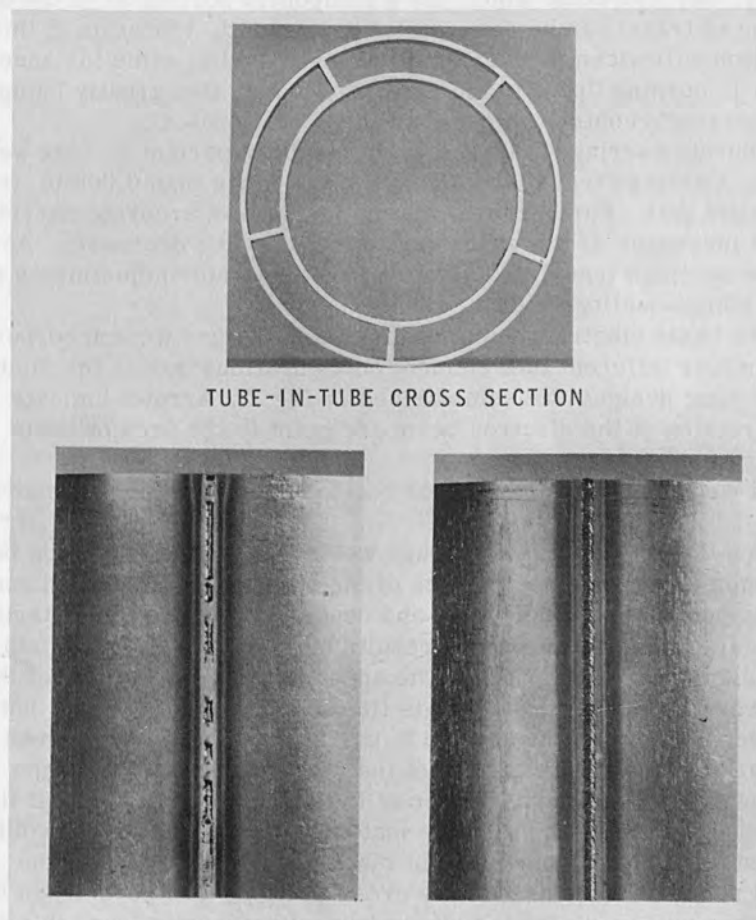


Fig. 1. Importance of fit-up illustrated by results of good and poor fit-up.

turing may be high and with each addition to the fixture, an accompanying increase in the size of the vacuum chamber must be considered.

Work Carriages. Smooth operation and essentially vibration-free movement are necessary features of work carriages. Vibration control is one key to quality work. Where pinpoint accuracy in placement of the small diameter electron beam on the joint is nec-

essary, any vibration which has a component horizontal to the direction of travel can be detrimental to the weld. Vibration in this direction will widen the weld unnecessarily and in some instances result in burning through thin cladding. It will also greatly hamper the operator's control over the welding.

Accurate carriage travel is particularly important on long weldments. Carriage travel should not deviate more than 0.005 in. from a straight path. Furthermore, the importance of accurate carriage travel increases as the wall thickness of the tube decreases. Accurate carriage travel will require fewer operator adjustments and assure high-quality welds.

Five basic electron-beam welding joint designs were incorporated in four different fuel element configurations during the study. These joint designs are summarized in Fig. 2. Arrows indicate the direction of the electron beam and point to the area of beam impingement.

All welding was performed on reactor grade Zircaloy-2 material.

Type-1 joints are burn-through welds in which the electron beam impinges radially on the surface of the outer tube. The beam completely penetrates the cladding and continues into the rib material.

Figure 3 clearly shows the results of poor fit-up between rib and tube in this type of joint. The appearance of the outside of the tube when this condition exists is illustrated in Fig. 1. Note that the area of the weld metal used to partially fill the void between the I.D. of the tube and the top of the rib is far greater than the area of the indentation at the top of the weld. The metal to fill the void did not come from the tube material directly above the void. It must have come from the joint material on either side of the metal shown in the plane of this cross section. The porosity in this joint type will be discussed later along with porosity found in all of the four remaining joints.

The bulge of metal shown in the top photograph of Fig. 4 is typical for this type of joint when the fit-up is good and the welding beam penetrates close to one side of the rib. There is two-point contact between the rib and the tube, i.e., since the tube is curved, only the two edges of the rib will touch it. As the beam penetrates the tube, the rib edge that is close to the beam heats, melts and the molten edge sags to form the bulge.

A good weld is illustrated in the bottom photograph of Fig. 4. There is good fit-up and the weld is only slightly off center.

A type-1 weld would be considered perfect if the welding beam penetrated the exact center of the rib and the weld was wide enough

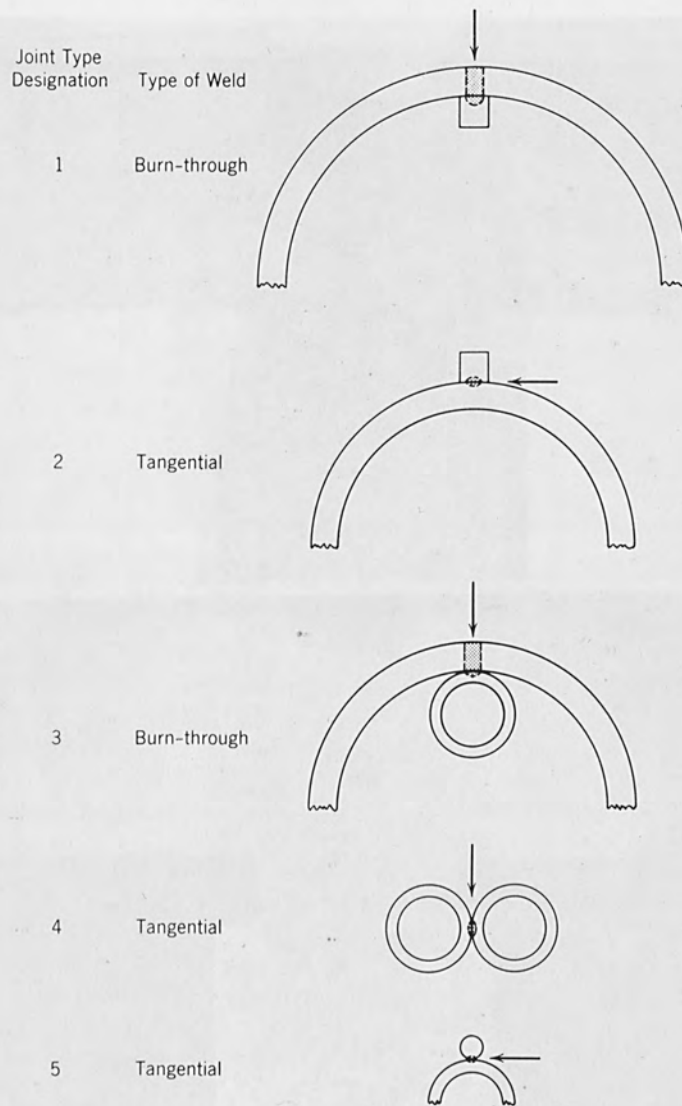


Fig. 2. Five basic electron-beam joint designs.

to eliminate the slight crevices between the rib and the tube on either side of the weld metal.

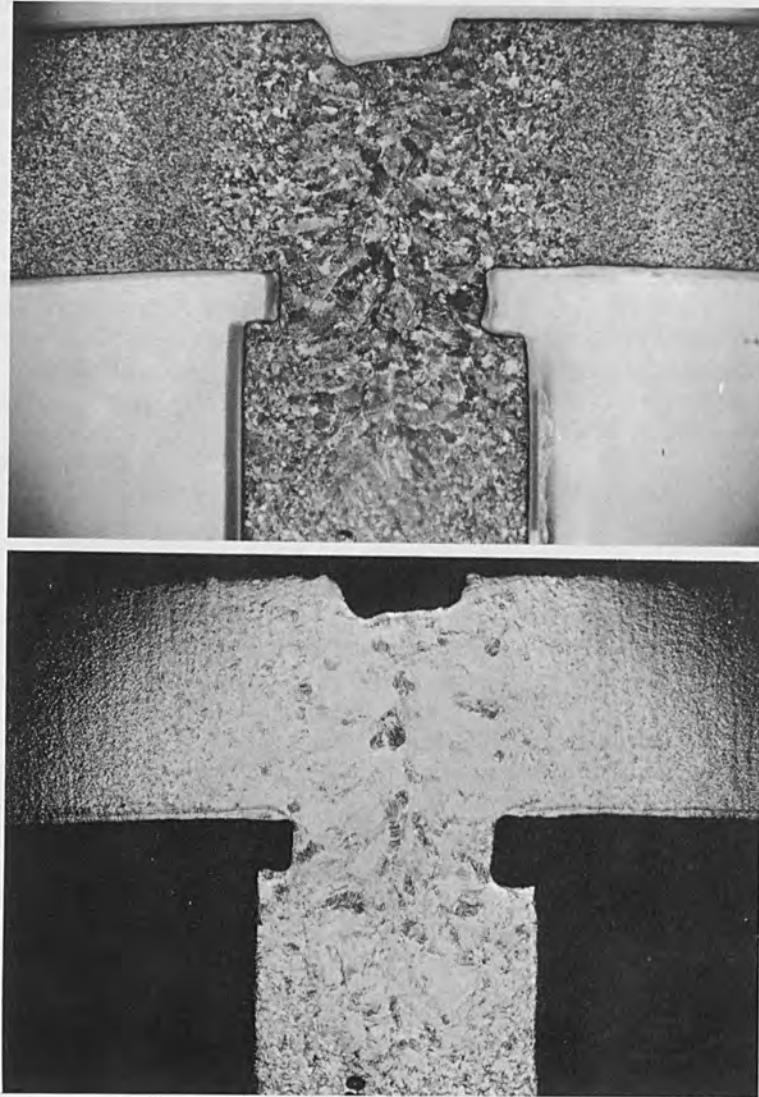


Fig. 3. Results of poor fit-up (cross section) on type-1 weld.

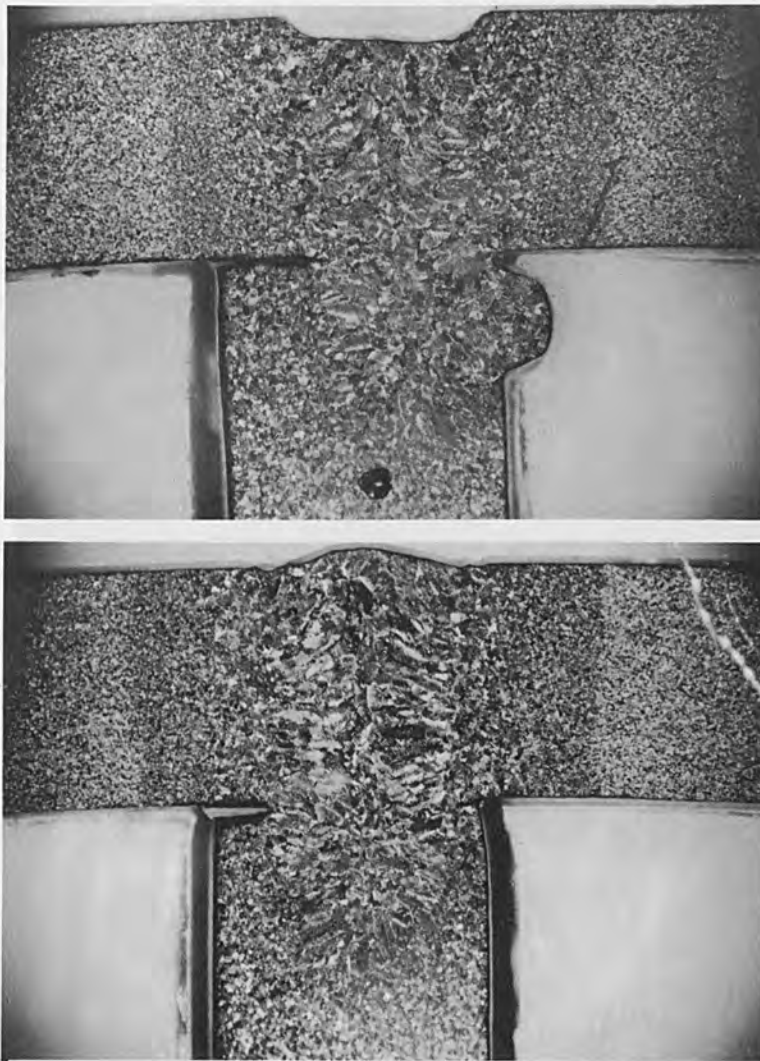


Fig. 4. One-sided (top) and good weld (bottom) on type-1 weld.

Type-2 joints are fabricated by aiming the beam tangentially to the cylindrical cladding and toward the point of contact between the rib and tube (Fig. 5). One weld pass on each side of the rib is made to assure complete penetration and to minimize crevicing between rib and tube.

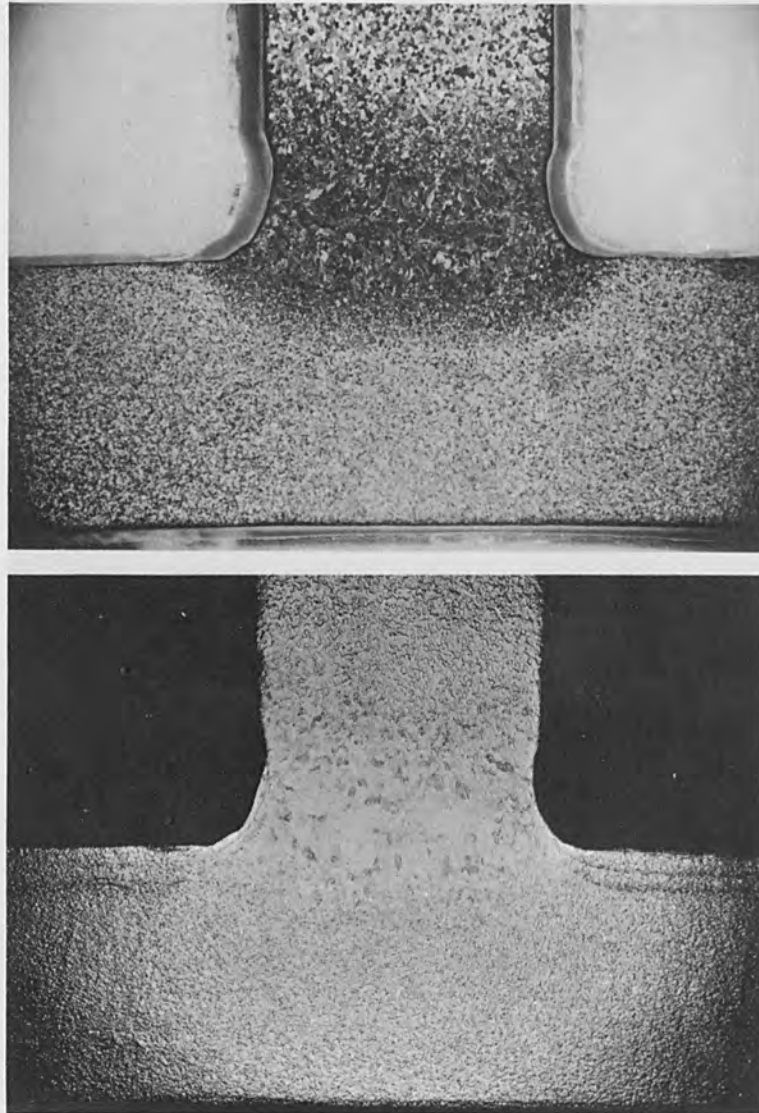


Fig. 5. Good type-2 weld.

Figure 5 illustrates the type of weld quality acceptable for nuclear fuel elements. The heat-affected zone is narrow and there is complete penetration. Undercutting of the rib is slight with a smooth contour. Also, there is no porosity in this section. Porosity is occasionally encountered with this joint, however.

A type-3 weldment comprises a burnthrough weld between two tubes with a small tube in contact with the I.D. of a large tube (Fig. 6). The electron beam initially impinges on the large tube and penetrates into the smaller tube to form the weld. When fit-up between the two tubes is good, a weld such as that shown in Fig. 6 results.

Figure 7 shows the results of excessive penetration and poor fit. Excessive penetration results in irregular protrusions on the inside of the small tube which are difficult to remove but do not affect the integrity of the weld.

A close fit between the two tubes may or may not be desirable, depending upon the ultimate use of the assembly. The photomicrograph shows a gap between tubes of approximately 0.01 in. A successful weld may be made with a much greater gap, but the optimum gap width is dependent upon the width of the weld and the size of the groove at the top of the weld that can be tolerated.

The interesting distribution of porosity apparent in Fig. 7 will be discussed later.

Two small tubes, laid side by side, make up a type-4 weldment (Fig. 8). The two tubes can be joined using either one pass or a pass on each side of the joint. Since the metal to fill the joint comes from the walls of the tubes, a close fit between the pieces is essential to prevent wall thinning. The greater the spacing, the more metal will have to be taken from the walls to fill the joint. This situation will result in a reduced tube wall thickness.

Figure 9 shows porosity in the two-pass and one-pass welds.

Very little mention of porosity has been made in the preceding paragraphs. This is because the porosity in all joints has much in common. The small porosity, with but few exceptions, occurred along the edges of the melts. The large holes which were found only in type-4 weldments always appeared near the point of maximum beam penetration.

Porosity is attributed to impurities in the materials used and not to the electron-beam welding process.

The round wire-to-tube joint (type 5) is extremely difficult to achieve. The point of beam impingement and the path of beam penetration have to be carefully selected in order to minimize melting of the wire and tube. Figure 10 is a cross section of a

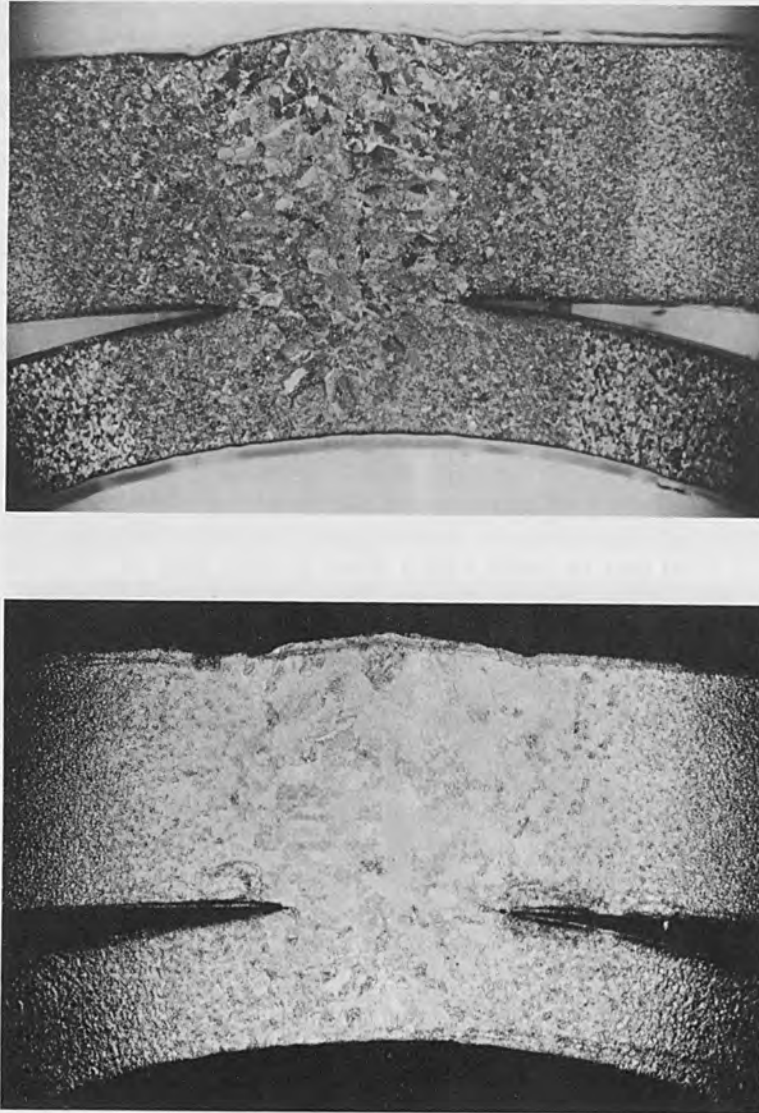


Fig. 6. Good type-3 weld.

type-5 weld. Because of point contact between the wire and tube, metal to fill the joint must be obtained from either the wire or the

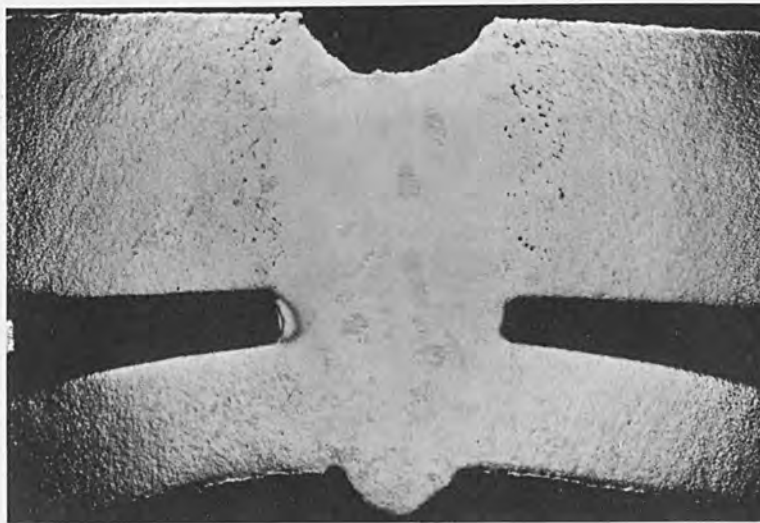
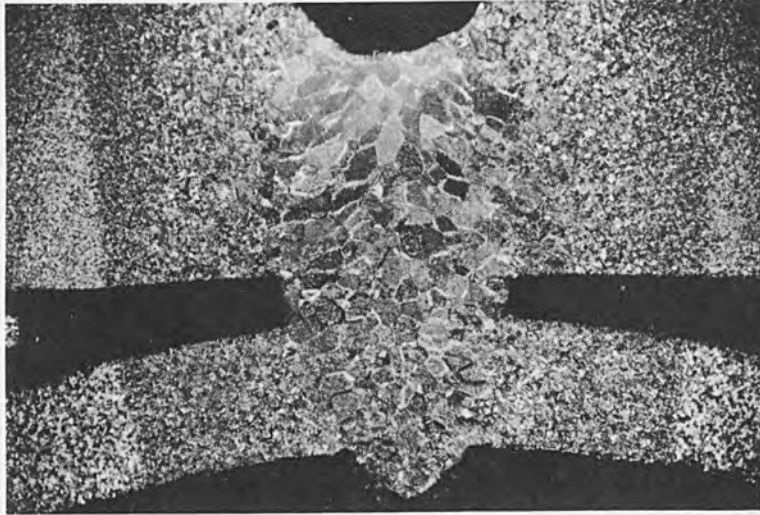


Fig. 7. Type-3 weld excessive penetration and poor fit-up.

tube. In most cases, the metal should be taken from the wire to eliminate tube wall thinning. If an assembly is to be made from a

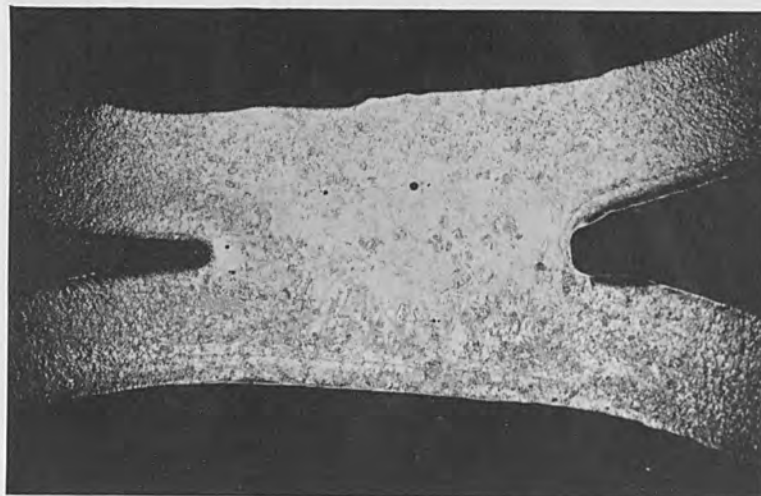
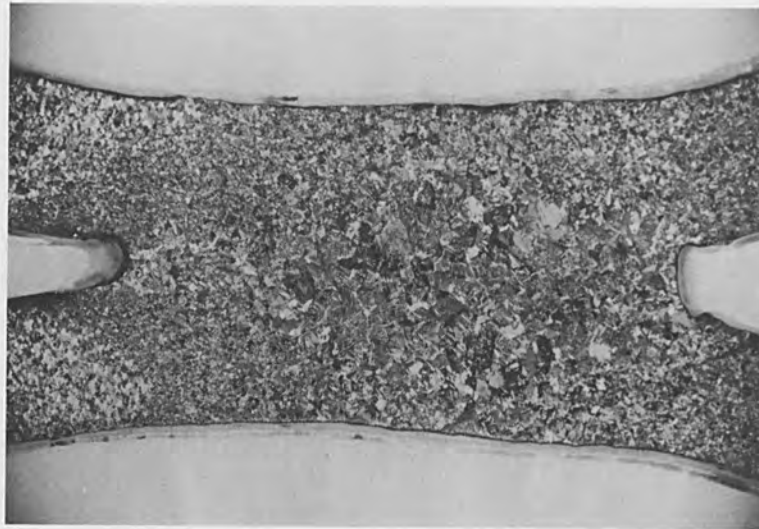


Fig. 8. A good two-pass type-4 weld (top). A good one-pass type-4 weld (bottom).

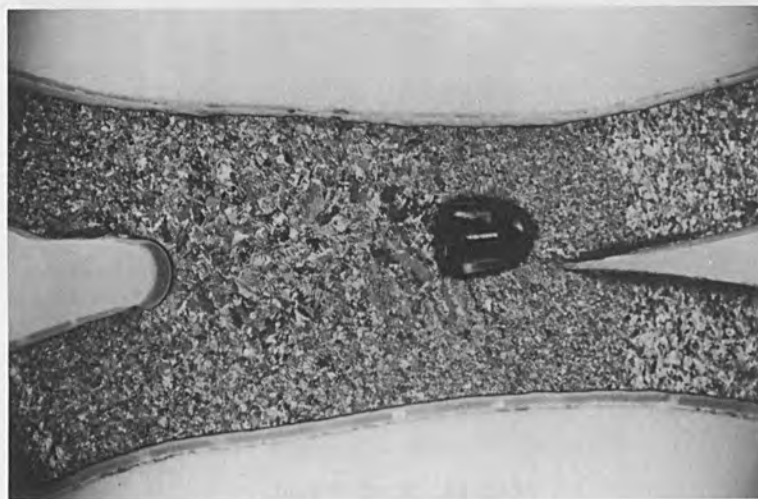
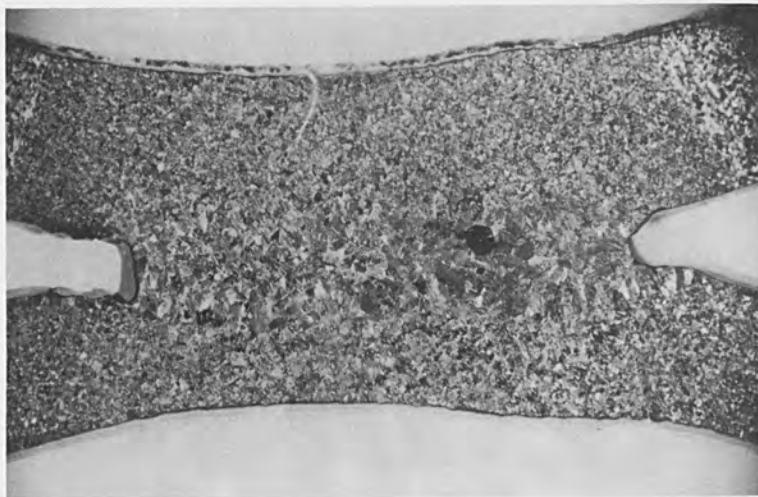


Fig. 9. Two-pass type-4 weld with porosity (top). One-pass type-4 weld with porosity (bottom).

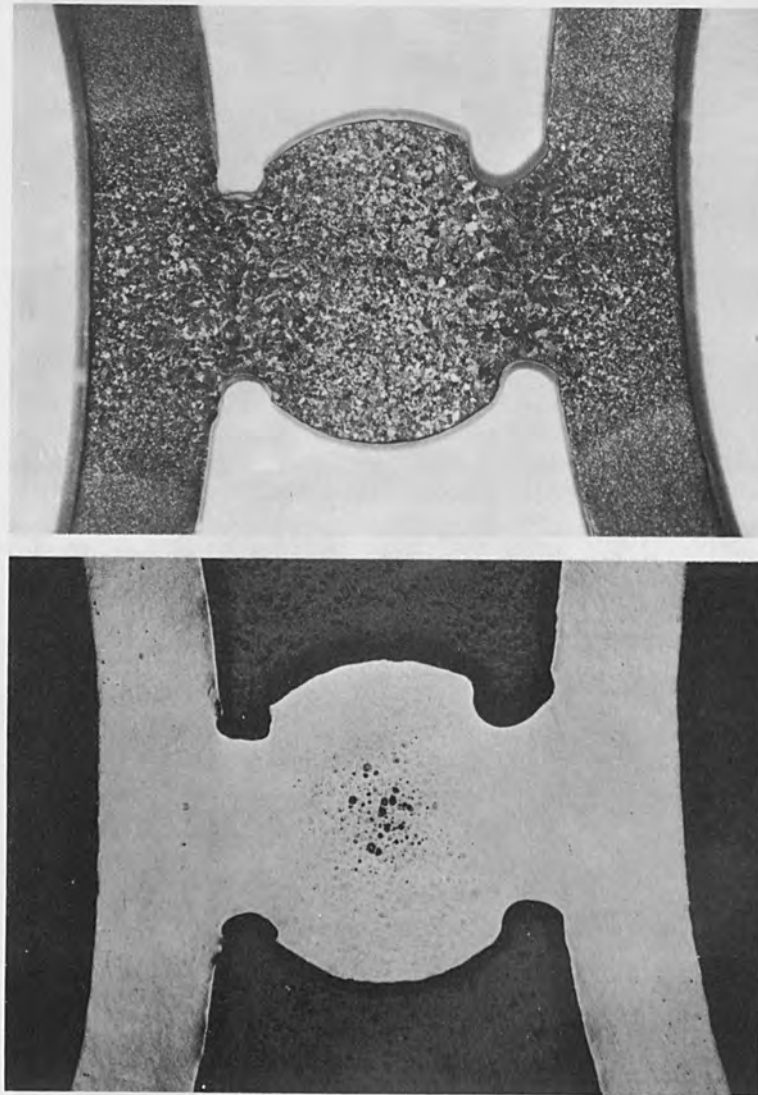


Fig. 10. Type-5 weld.

group containing joints of this type, dimensional changes should be anticipated.

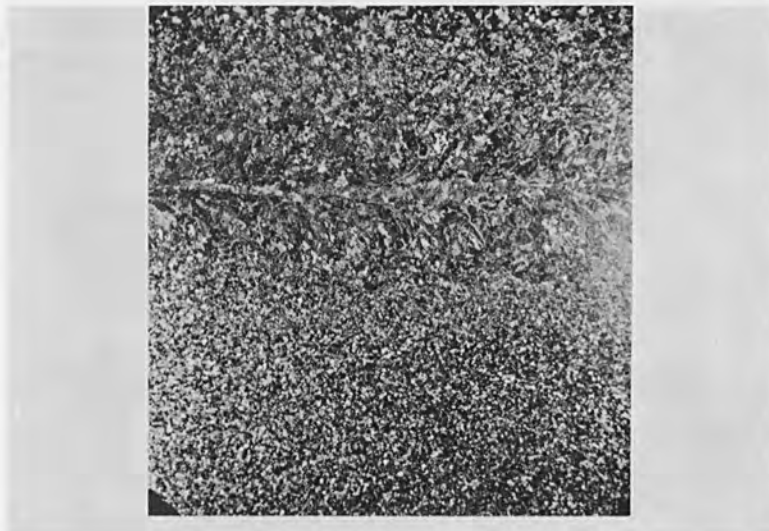
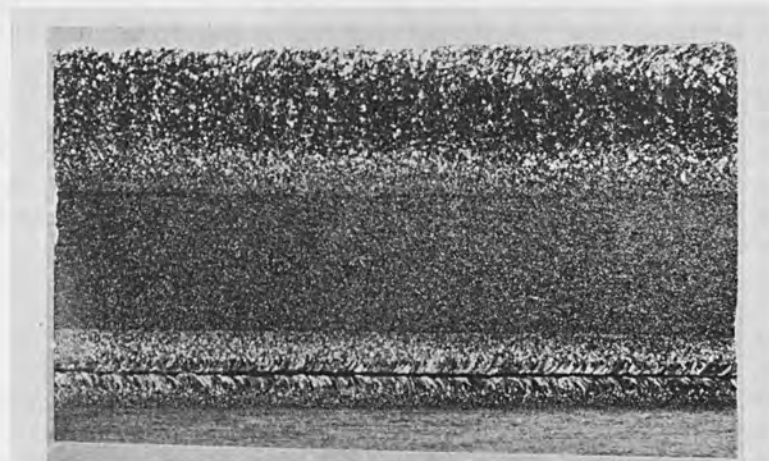


Fig. 11. Longitudinal sections of type-1 and type-2 joints.

Figure 11 is a photomicrograph of a longitudinal section of type 1 and 2 joints. Joint 1 is at the top of the top photograph and joint 2 at the bottom of the top photograph. Note the grain structure in the type-2 joint. The direction of travel of the welding beam is clearly discernible. The apparent line near the bottom of the top photo, in the type-2 joint, indicates the center of the melt with the electron beam coming from a direction perpendicular to the plane of the paper and traveling from right to left. The bottom photograph is an enlarged portion of this section.

CONCLUSIONS

The electron-beam welding process has proven highly successful for fabricating fuel element claddings having either standard or unusual configurations. The success of initial trials prompted the fabrication of actual pieces of cladding, using the full 9-ft long tubes. Photographs of these pieces may be seen in Fig. 12, which shows a preassembled 19-rod cluster and a tube-in-tube cladding assembly, and Fig. 13, which shows spacer ribs attached to the nested tubular cladding parts.

Figures 14 and 15 are additional experimental configurations that illustrate versatility of the electron-beam welding process.

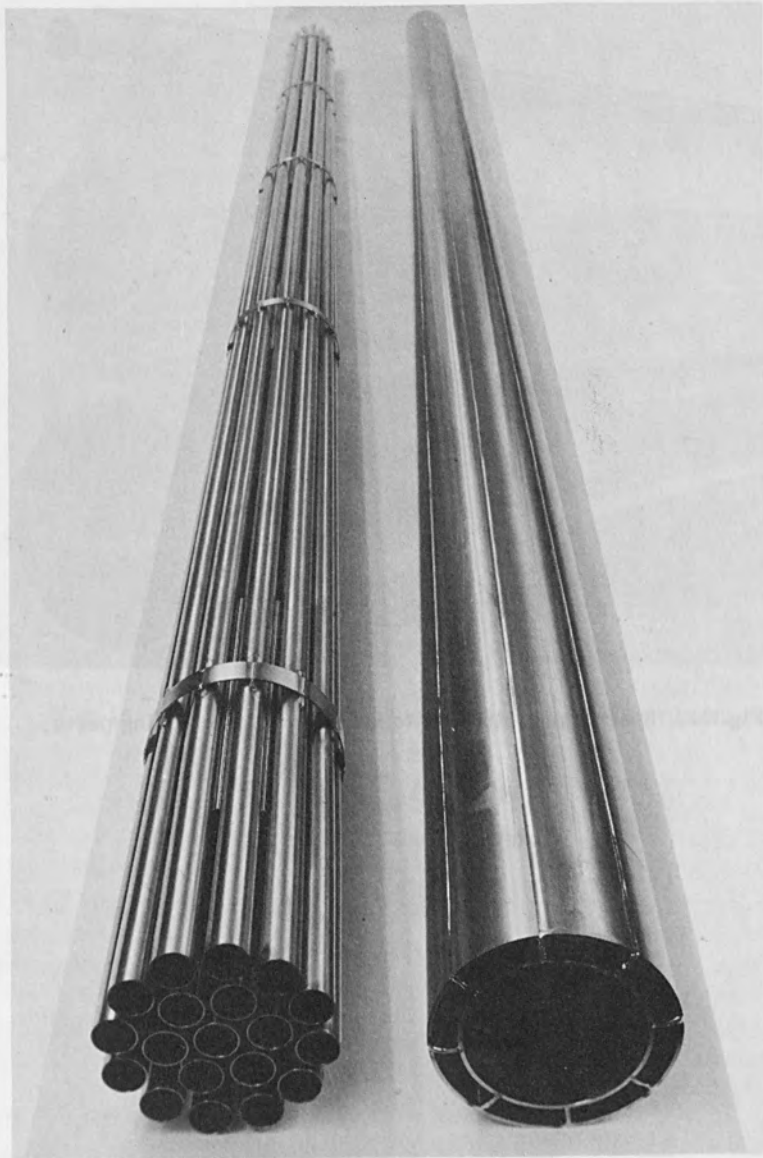


Fig. 12. Preassembled 19-rod cluster and tube in tube-cladding assembly.

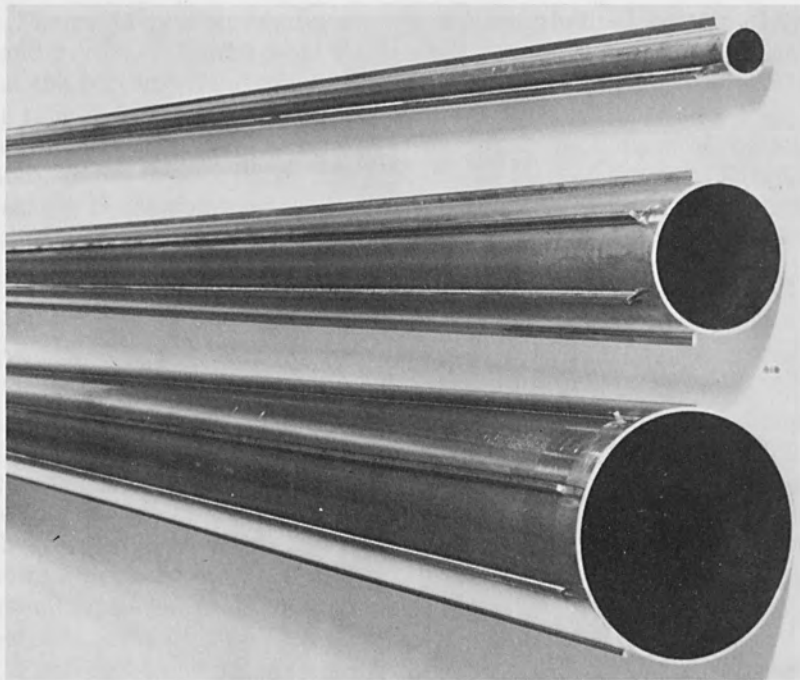


Fig. 13. Spacer ribs attached to nested tubular cladding parts.

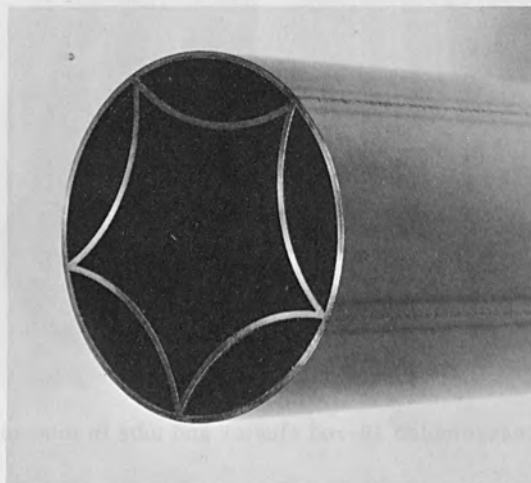


Fig. 14. Experimental fuel element cladding configuration.

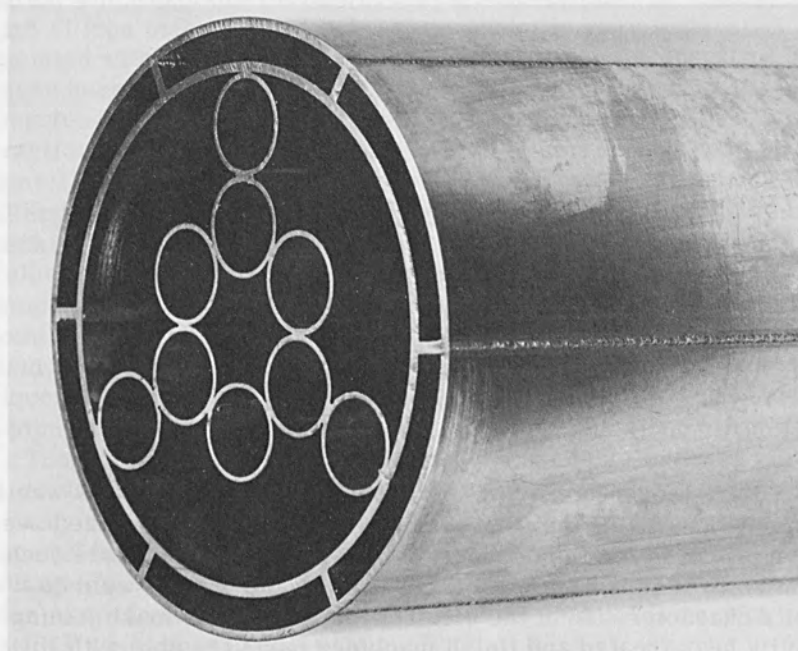


Fig. 15. Experimental fuel element cladding configuration.

PROPERTIES OF ELECTRON-BEAM WELDS IN ULTRA-HIGH-STRENGTH STEELS

INTRODUCTION

The electron-beam welding of ultra-high-strength steels was investigated to define the improvements which may be realized over other processes in the fabrication of aerospace components such as missile cases. The low total heat input and narrow weld deposits characteristic of the electron-beam process make joining of fully heat-treated and finish machined parts feasible with little or no distortion resulting from the welding. Added benefits are the high metallurgical quality of the vacuum-melted deposit and the extremely narrow heat-affected zone. The advantages to be gained by electron-beam welding large size components tend to counter balance the inconvenience of welding in a vacuum chamber.

The performance capabilities of aerospace and missile components have increased with the advent of the ultra-high-strength steels [1]. Strength levels in excess of 230,000 psi yield strength are readily obtained by conventional air-hardening heat treatment. Thermomechanical processing has produced even greater strengths in certain steels [2], [3]. Unfortunately, the ultra-high-strength steels with their higher alloy content have introduced problems in arc welding. They have been shown to be prone to weld cracking unless the sulfur and phosphorus contents of the base metal and filler wire are maintained at extremely low levels [4], [5] (in the order of 0.010%). Microsegregation of sulfur and phosphorus to the interdendritic regions of arc welds is a major factor in what has been termed hot short cracking [6]. Silicon promotes microsegre-

*Philco, A Subsidiary of Ford Motor Company, Newport Beach, Calif.

gation which explains the high cracking susceptibility of weld in the high silicon steels [6], [7], [8]. Large dendritic grains, possibly associated with one or more of the above mentioned segregation cases have been reported in conjunction with the low ductility of tungsten-arc welds in the ultra-high-strength steels [9]. A number of practices have been adopted as a means of correcting these problems. Weld ductility was improved by using lower carbon content filler wires [10] and by employing surface decarburization and cold working techniques [11]. In addition, the ductility of tungsten arc welds in H-11 steel was improved by preheating just below the M_s temperature (350° to 450°F) and immediately stress relieving. Joint efficiencies (welded and heat treated) greater than 95% and weld tensile strengths of 300 ksi were reported using these techniques [12]. Similar welds in D6-AC material averaged 220 ksi for a joint efficiency of 96% [13].

Tungsten-arc welding has been the predominate process in the fabrication of ultra-high-strength steels [6]. One investigator made a study of heat-affected zones in D6-AC steel simulating tungsten-arc-welding conditions in which he concluded that for given arc voltages and amperage, travel speeds must be above a certain minimum to ensure the desirable all-lower-bainite formation when a 600°F preheat is used [14].

Heat treatment after welding was standard procedure when welding by the tungsten-arc process. No data were available on the properties of tungsten-arc welds in material heat treated before welding, however, several investigators have applied the electron-beam process to welding of hardened steels. Tensile test results of electron-beam welded D6-AC steel heat treated to over 200,000 psi yield strength before welding were equivalent to those obtained for specimens welded in the annealed condition [15], and heat treated after welding. Ultimate tensile strengths were higher for high-voltage, electron-beam welds in H-11 steel heat treated to 45-50 Rc before welding than those obtained on specimens heat treated to a similar base metal hardness after welding [16]. Joint efficiencies of 90 to 95% and fracture toughness values ranging from 189.3 to 249.8 were reported for high-voltage electron-beam welds in 0.090 in. D6-AC steel heat treated prior to welding [17].

This paper discusses the results of an electron-beam weld evaluation program using five alloy steels. The effects of welding before conventional heat treatment, after conventional heat treatment, and after strengthening by thermomechanical processing are discussed. The weld evaluation tests include heat input determination, metallographic studies, microhardness traverses, tensile tests, and

fracture toughness tests. Techniques used in welding head closures and attachments to missile cases are described.

MATERIALS

The materials used in this investigation were selected to encompass the range of alloy steels employed in missile manufacture. The chemical composition of the five alloy types studied are listed in Table I. The steel with the lowest alloy content was air-melt 4130 steel obtained in the form of annealed 0.090-in.-thick rolled sheet. This steel was heat treated in the conventional manner before or after welding by oil quenching from 1550°F and tempering at 750°F.

A major part of the investigation work was concentrated on the welding of D6-AC steel, a consumable electrode vacuum-melted alloy developed by the Ladish Company. The chemical compositions for four different D6-AC heats are reported in Table I. The 0.090 and 0.100-in.-thick rolled sheet stock was used in welding studies on conventionally heat-treated material. The heat treatment consisted of oil quenching from 1650°F and double tempering in 2-hr cycles.

The 0.055-in.-thick D6-AC material was obtained by Ausform processing utilizing shear spinning as the deformation mode. Fourteen-in. diameter tube forgings with a 0.250-in. wall thickness were first austenitized at 1650°F, then cooled rapidly to 1025°F within the metastable austenitic region of the isothermal transformation diagram. The tube forgings were shear spun on an internally heated mandrel. After the desired deformation was accomplished, the part was quenched on the mandrel and then tempered. Ausform processing was also employed in the preparation of the 0.500-in.-thick material, except that the deformation was accomplished by open-die forging. Standard forging practices were varied only at approximately 1000°F during the quench from the austenitizing temperature.

The 0.100-in.-thick vacuum melted X-2 steel is a modified H-11 steel having a lower carbon content. This steel and the 0.100-in.-thick H-11 steel were Ausform strengthened by rolling.

The 250 series Maraging steel was included in this investigation since its precipitation hardening characteristics and high strength-toughness characteristics make it particularly suitable for missile manufacture. This steel was acquired in the form of 0.090-in.-thick solution annealed sheet.

Table I
Composition of Materials

Material	C	Mn	P	S	Si	Ni	Cr	Mo	V	Other
A. 0.090-in. 4130 rolled sheet	0.30	0.54	0.008	0.015	0.31	...	0.98	0.23	...	
B. 0.090-in. D6AC rolled sheet vac. melt	0.43	0.84	0.008	0.007	0.24	0.51	1.20	1.05	0.07	
C. 0.100-in. D6AC rolled sheet vac. melt	0.48	0.67	0.005	0.008	0.23	0.65	1.06	1.07	0.10	
D. 0.055-in. D6AC shear formed vac. melt	0.45	0.70	0.008	0.007	0.22	0.59	1.08	1.00	0.06	
E. 0.500-in. D6AC pancake forging vac. melt	0.47	0.77	0.010	0.007	0.27	0.56	1.13	1.02	0.07	
F. 0.100-in. X-2 sheet vac. melt	0.23	0.24	0.016	0.004	0.96	...	4.86	1.20	0.51	
G. 0.100-in. H-11 sheet vac. melt	0.38	0.29	0.007	0.008	0.90	...	5.24	1.32	0.54	
H. 0.090-in. 250 maraging steel	0.015	0.015	0.001	0.003	0.04	18.32	...	5.36	...	0.16Al 0.52Ti 9.16Co 0.003B

EQUIPMENT

Most of the welding during this investigation was done using a 30-kV Sciaky electron-beam welder as shown in Fig. 1. This welder is equipped with a 500 mA electron-beam gun which can be directed in any position within the vacuum chamber. In addition to the main 50-in. by 30-in. $\times$ 42-in. chamber, a 16-in. diameter chamber extension is available for welding elongated members such as missile cases as illustrated in Fig. 1. Special features added to this welder to facilitate production fabrication capabilities are: (1) focus-coil current decay mechanism which is synchronized with the beam current decay for smooth blending in of weld starts and stops; (2) a wire feeder with external motor drive which

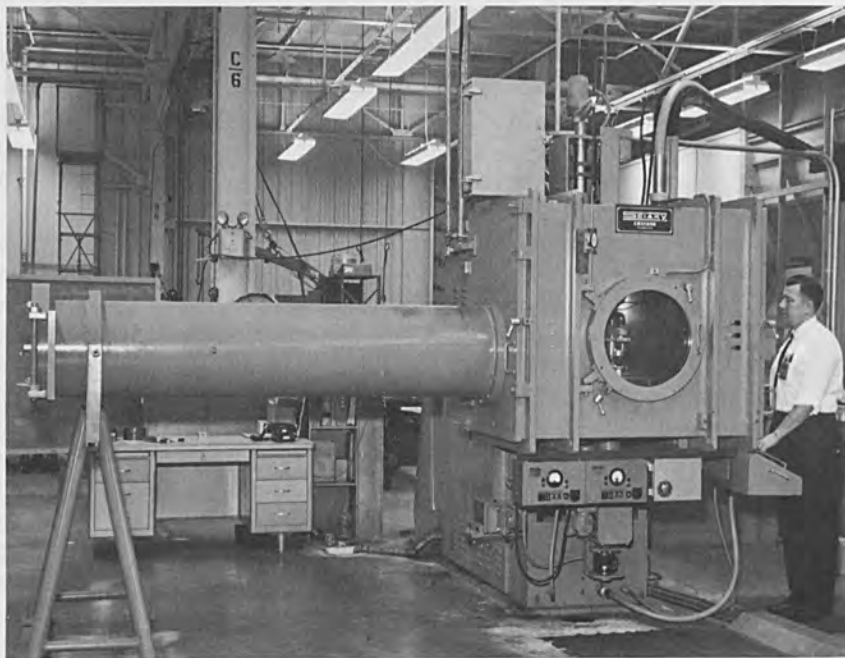


Fig. 1. Electron-beam welder with vacuum chamber extension.

houses a 14-in. diameter coil of wire within the vacuum system; and (3) an external micrometer dial gauge attached to the gun raising and lowering mechanism for accurate gun positioning.

WELDING PROCEDURES

As an expediency, welding schedules for a specified material thickness were developed by bead-on-plate welding without a joint. Welds made in this manner were comparable to those obtained using a square butt joint having a good fitup but not a joint with excessive gapping or misalignment. Joint preparations for electron-beam welding were specified on engineering drawings to have a maximum seam opening of 0.005 in., and a maximum joint misalignment of 0.005 in. The allowable edge mismatch was 10% of the metal thickness with a maximum of 0.010 in.

Cleaning

The most satisfactory procedure for cleaning of parts for electron-beam welding was to remove light oxidation from the parts by swabbing with a 20% dilute solution of hydrochloric acid and rinsing in water. Degreasing was accomplished with hot trichloroethylene and finally with acetone.

Weld Width

Procedures for electron-beam welding of the ultra-high-strength steels were based on several considerations. First, it was desirable to butt weld with as narrow a weld bead as practicable to minimize softening of the base metal adjacent to the weld in fully hardened materials. Fabrication of ausformed parts requires that welding be done when the material is in the hardened condition. Conversely, the second consideration dictated that the weld bead be wide enough to allow some slight variations in joint fitup and seam tracking within practical machining tolerances for large assemblies. The optimum weld width selected to best satisfy these criteria on the basis of preliminary tests was approximately 1/16-in. for material thicknesses ranging from 0.055 to 0.100 in. The welding conditions used on the various materials are given in Table II.

Table II

Welding Conditions

Material	Process	Preheat	Post-heat *	Pass Number	Voltage	Current	Travel Speed in./min	Filler Wire
0.090-in. 4130	Electron Beam	None	None	1	23 kV	90 mA	55	None
0.090 and 0.100-in. D6AC	Electron Beam	None	None	2	16 kV	52 mA	30	None
0.100-in. D6AC	Electron Beam	None	None	1	25 kV	90 mA	50	None
0.055-in. D6AC	Electron Beam	None	None	2	17 kV	54 mA	30	None
0.50-in. D6AC	Electron Beam	None	None	1	23 kV	38 mA	45	None
				2	17.5 kV	23 mA	35	None
		600°F	None	1	30 kV	300 mA	40	None
0.50-in. D6AC	Tungsten arc (manual)	600°F	800°F	1	15	110-140 amp	3	None
				Rem	15	145-155 amp	4	1/16 in. D6AC
0.100 X-2	Electron Beam	None	None	1	25 kV	90 mA	50	None
0.100 H-11	Electron Beam	None	None	2	17 kV	54 mA	30	None
				1	95 kV	7 mA	40	None
0.100 H-11	Tungsten arc	600°F	800°F	2	85 kV	3.5 mA	40	None
				1	15	75 amp	5	1/16 in. D6AC
0.090-in. 250 maraging	Electron Beam	None	None	1	23 kV	90 mA	55	None
				2	16 kV	52 mA	30	None

* All welds were stress relieved or retempered at the specified tempering temperature.

Cover Passes

The ultra-high-strength steels were particularly susceptible to weld undercut or starved areas on both sides of the weld bead as illustrated in Fig. 2a and 3a. When the welds were not to be machined, it was necessary to apply a wide cover pass which faired in smoothly with the parent metal for good appearance and for avoiding notch effects. Typical cover passes are illustrated in Fig. 2b, 3b, and 3c. As discussed in the Tensile Testing Section of this report, strength comparisons were made between single-pass welds with cover passes for 4130 and D6-AC steel heat treated before welding.

A recent innovation in electron-beam welding is the use of filler wire. Excellent results are obtained by adding filler wire either preplaced on the seam or fed by a wire feeder.

Preheat and Postheat

Neither preheating nor postheating were necessary when electron-beam welding any of the materials studied in thicknesses up to 1/8 in. No cracking was noted in any test welds or production welds in materials at this thickness level. The 0.50-in.-thick D6-AC material, however, showed extensive longitudinal cracking when attempts were made to weld without preheat. A 400°F preheat temperature eliminated all of the cracking except for extremely minute cracks in the base metal adjacent to the weld beam reinforcement. These were completely eliminated by using a 600°F preheat temperature.

Preheating was accomplished where required with quartz tube heaters inserted into longitudinal holes in support bars underneath the test plates. Heating with the defocused beams proved to be unsatisfactory for heavy plate welding. Temperatures were measured by means of Tempilaq indicator preplaced along side the seam of the parts to be welded.

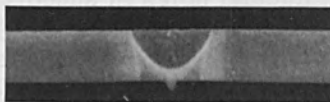
Postheating in the hold-down fixture was not deemed necessary, however, all test plates were furnace stress relieved or tempered immediately after welding at the specified tempering temperature.

Heat Input

Reputedly, the major advantage of electron-beam welding is the low total heat imparted to the material being welded. Calculations were made to determine the relative heat input for electron-beam and tungsten-arc welding in accordance with the formula,



A. SINGLE PASS WELD, HEAT TREATED
BEFORE WELDING



B. TWO PASS WELD, HEAT TREATED
BEFORE WELDING

P05479

3X MAGNIFICATION

ETCHANT: PICRAL + 2% HCl

Fig. 2. Macrographs of electron-beam welds in 0.090-in. thick 4130 steel.



A. SINGLE PASS WELD IN D6AC STEEL
HEAT TREATED BEFORE WELDING



B. TWO PASS WELD IN D6AC STEEL
HEAT TREATED BEFORE WELDING



C. TWO PASS WELD H-11 STEEL
AUSFORMED BEFORE WELDING

3X MAGNIFICATION

ETCHANT: PICRAL + HCl

Fig. 3. Macrographs of electron-beam welds in 0.100-in. thick steel.

$$\text{heat in kilojoules per inch} = \frac{\text{kV} \times \text{Ma} \times 60}{\text{ipm}}$$

The interpretation of heat input data is based on several considerations. For example, when multipass welding hardenable steels, the pass made with the highest power setting is the one which probably establishes the degree of heat effect on the parent material. The heat effect of subsequent passes is not additive. On the other hand, distortion of the welded parts is directly related to the number of weld passes being made. For this reason, the heat input data in Table III reflect both the heat input for a single pass and the total heat input for all passes of electron-beam and tungsten-arc welds in comparable material thicknesses. The calculations show that the electron-beam weld heat input per pass is less than 1/3 that of a tungsten-arc weld. Total heat inputs, in heavier section welds, may be in the order of 4% of those for TIG welds, reflecting the low distortion and thermal stress problems encountered.

Inspection and Repair Welding

All welded test plates and components were X-ray inspected after stress relieving. Porosity and cracking were practically non-existent in the electron-beam welds on ultra-high-strength steels. The X-ray defects were limited to occasional off-seam welds, lack of penetration, and weld undercut. A major feature of electron-beam welding is that these defects were repaired simply by rerunning the weld. No chipping out was necessary.

METALLOGRAPHIC STUDIES

Metallographic studies were made of electron-beam welds and heat-affected zones to establish a relationship between microstructure and the superior strength properties of electron-beam welds. Most of the studies were concentrated on the welds in 1/2-in. thick ausforged D6-AC steel. A macrograph of the entire weld and micrographs illustrating the microstructure of the weld, heat-affected zone and parent material are shown in Fig. 4. The weld has a typical cast structure but the columnar grains are extremely small. Likewise, the heat-affected zone exhibits a fine grain structure. Contrast this with the tungsten-arc-weld photomicrographs in Fig. 5 showing large overheated grains in the heat-affected zone. Colum-

Table III

Comparative Heat Input Developed
during Electron-Beam and Tungsten-Arc Welding of Steel

Electron-Beam Welding						
Steel Thick- ness	Voltage	Current	Travel Speed in./min	No. Pas- ses	Heat Input Kilojoules/Inch	
					Max. per Pass	Total All Passes
0.055	23 kV	38 mA	45	2	1.2	1.9
0.090	23 kV	90 mA	55	2	2.3	3.9
0.100	25 kV	90 mA	50	2	2.7	4.5
	95 kV*	7 mA	40	2	0.99	1.4
0.500	30 kV	300 mA	40	1	13.5	13.5
Tungsten-Arc Welding						
0.055	15	45 amp	5	1	8.1	8.1
0.090	15	60 amp	5	1	10.8	10.8
0.100	15	65 amp	5	1	11.7	11.7
0.500	15	125 amp	3	12	37.5	371.8

* Welded using high-voltage equipment.

nar grains in the weld are too large to be seen at the 500X magnification.

Some further studies were also conducted at higher magnifications to further illustrate the relative influence of microstructure on mechanical properties. The electron microscope, using negative replica methods, was used to study these structures at magnifications to 15000X. As might be expected, the fine structure of the cast-weld metals, was similar for both welding processes since they are both essentially martensitic (or tempered martensite) in nature. The differences noticeable in the dendritic patterns of the cast structure are not reflected in the morphology of the fine carbides. The fine structures in the heat-affected zones, Figs. 6

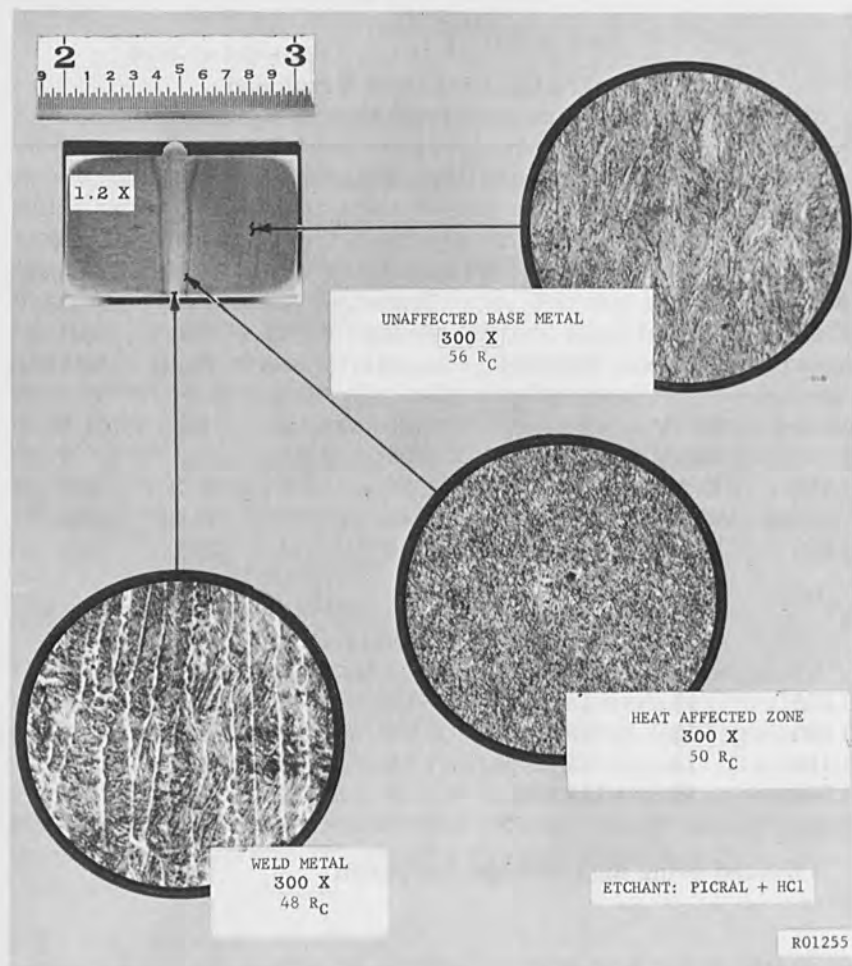


Fig. 4. Light macrograph and micrographs of electron-beam weld in 1/2-in. thick D6AC alloy ausformed 75% before welding.

and 7, are of a similar nature, although the size and density of the carbide precipitates are both slightly greater in that from the TIG-welded sample, Fig. 7. This provides a partial explanation for the higher joint efficiencies normally observed in electron-beam welds, in that the lesser extensive carbide precipitation and coalescence is indicative of a lesser degree of softening (overtempering). This

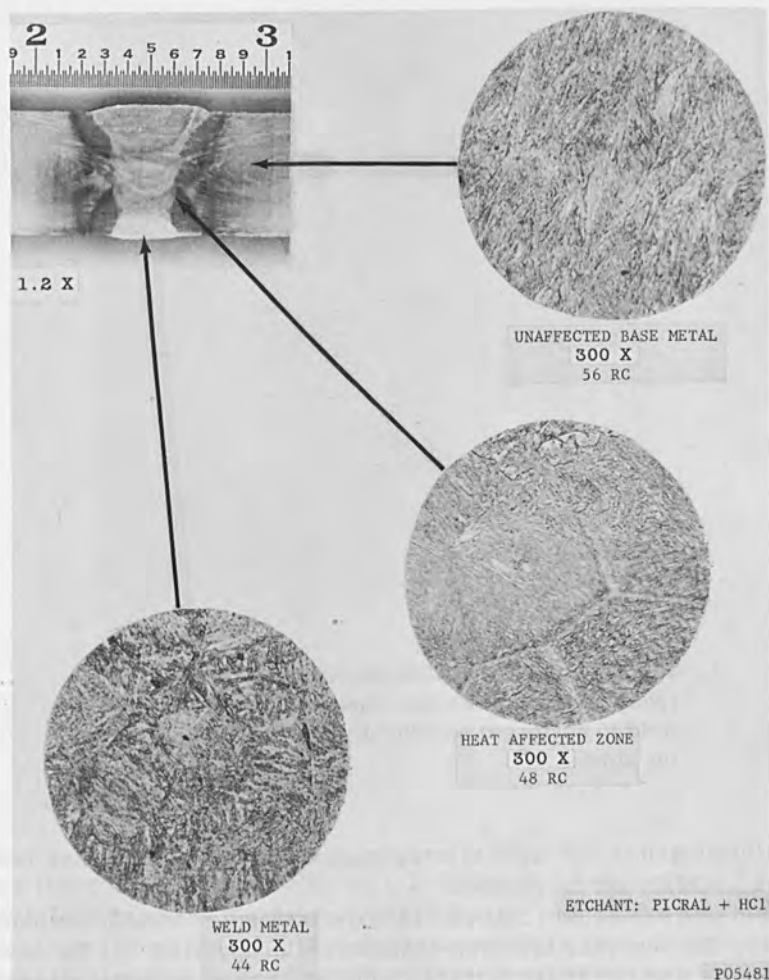


Fig. 5. Light macrograph and micrographs of tungsten-arc weld in 1/2-in. thick D6AC alloy steel ausformed 75% before welding.

is a result of the lower heat input values calculated for electron-beam welds so that there is less chance for gross alloy diffusion to occur.

However, this is only related to strength and hardness and not necessarily to the increased toughness also observed. The grain size and dendritic patterns observed on a more macroscopic scale

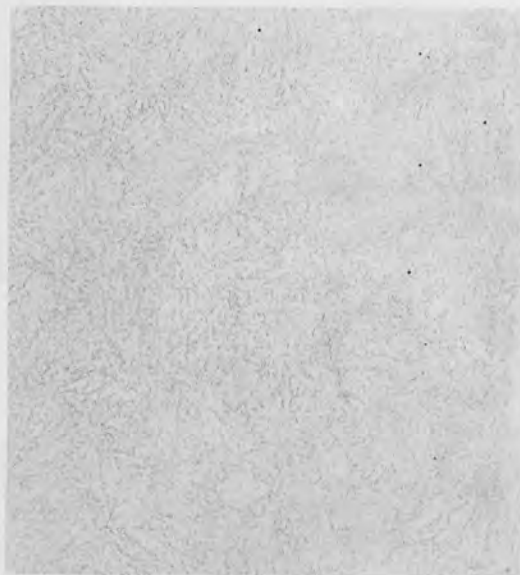


Fig. 6. Negative replica electron micrograph from heat-affected zones area of electron-beam weld in ausforged and 900° F temper D6AC alloy steel.

and illustrated in the light micrographs are factors that have been shown to influence toughness.

Therefore it seems that two factors both enter into the explanation for the superior electron-beam-weld properties: (1) the fine grain size and dendritic pattern in the weld zone contribute to the increased toughness and (2) the low heat input and subsequently lesser degree of softening in the heat-affected zones are responsible for the higher joint efficiencies.

MICROHARDNESS TRAVERSES

Microhardness traverses of the welds were made to establish the extent of the heat-affected areas. The traverses began at the center of the weld and continued across the heat-affected zone into the unaffected parent metal. Figure 8 graphically illustrates hard-

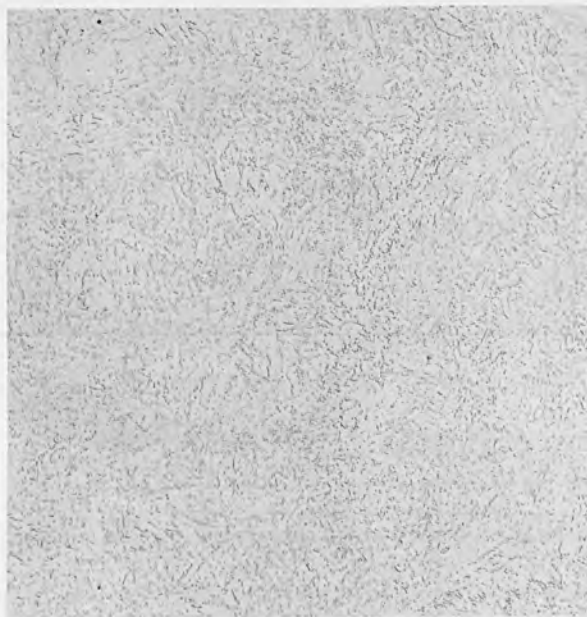
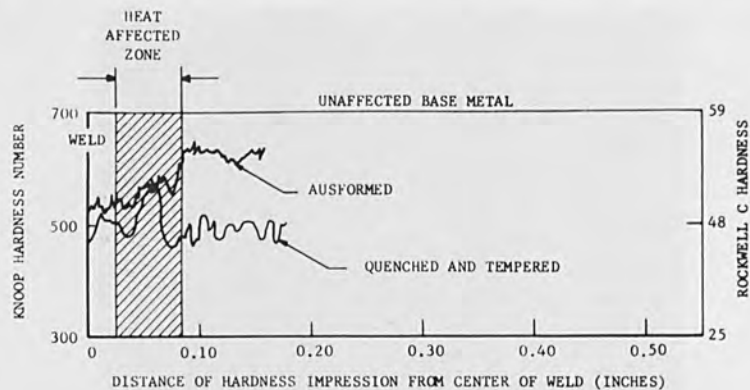


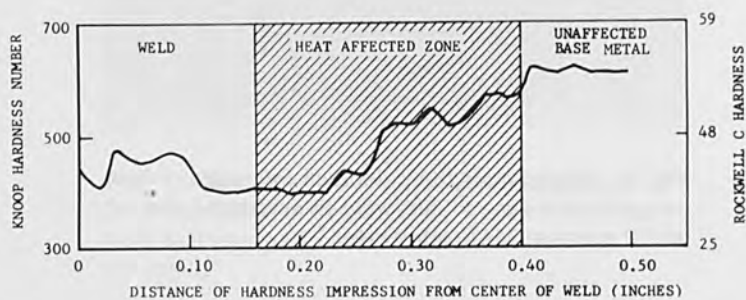
Fig. 7. Negative replica electron micrograph from heat-affected zone in TIG weld in ausformed and 900° F temper D6AC alloy steel.

ness traverses across welds in 1/2-in. thick D6-AC pancake forgings. Traverses of electron-beam welds in 75% ausformed material are compared with that in conventionally heat-treated material in Fig. 8a. It was significant that the weld in the ausformed material showed a fairly linear increase in hardness from the weld to the unaffected parent metal as contrasted to the conventionally heat-treated material which exhibited hard and soft peaks typical of heat-affected zones in hardenable materials. The traverse of the tungsten-arc weld in ausformed material in Fig. 8b, also showed the same linear increase in hardness even in the wide heat-affected zone.

In Fig. 9, it is seen that the electron-beam weld hardness traverse of ausformed X-2 steel tempered at 1100° F exhibits a linear increase in hardness to the unaffected parent material. Conversely, the ausformed H-11 steel weld traverse shows a peak hardness



a. ELECTRON BEAM WELD IN CONVENTIONALLY HEAT TREATED AND 75 PERCENT AUSFORMED D6AC STEEL



b. TUNGSTEN ARC INERT GAS (TIG) WELD IN D6AC STEEL AUSFORMED 75 PERCENT BEFORE WELDING

Fig. 8. Knoop hardness traverse of welds in 1/2-in. thick D6AC steel forgings, tempered at 900° F.

level in the heat-affected zone. This is a probably secondary hardening effect occurring at heat-affected zone temperatures higher than the 600° F tempering temperature.

TENSILE TESTS

Uniaxial tensile tests were made of both the welded and unwelded ultra-high-strength steels to determine the joint efficiencies to be expected in the fabrication of missile and other aerospace com-

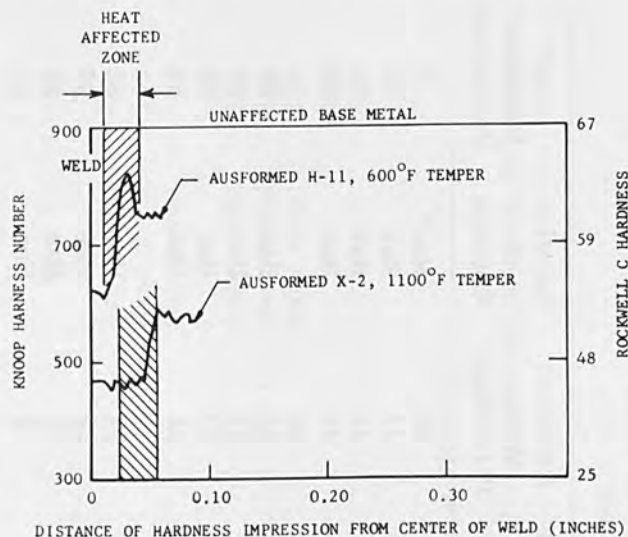


Fig. 9. Knoop hardness traverse of electron-beam weld in 0.10-in. thick H-11 and X-2 steel ausformed before welding.

ponents by electron-beam welding. Test plates of the sheet materials were cut into strips and prepared by grinding into flat transverse tension specimens. The tensile strength of welds in the 1/2-in. thick forged material were evaluated using 0.250-in. diameter round specimens. As is the custom for most investigators, data are reported for the yield strength and elongation obtained in the transverse weld tensile tests, however, it should be recognized that transverse weld specimens are inhomogeneous and these data may be misleading. Yielding in such specimens may occur simultaneously or separately at different locations. Elongation values were for a 2-in. gauge length and could be excessively low because all of the elongation was confined to a narrow, relatively ductile weld zone.

4130

The tensile properties of electron-beam welds in 0.090-in.-thick 4130 steel are summarized in Table IV. Results are reported for

Table IV, cont'd.

Heat treated after welding	wide cover pass	Weld not ground	187	192	3	9	Haz	94
			188	193	3	9	Haz	95
			187	193	3	9	Haz	95
			186	190	3	11	Haz	93
	Two-pass weld with	Weld	188	206	6	49	Base metal	100
		ground	188	204	7	48	Base metal	100
		flush	186	202	6	50	Base metal	99
	0.090-in. wide cover pass	Weld not ground	187	203	7	42	Base metal	100
			188	204	7	40	Base metal	100
			188	203	7	40	Base metal	100

¹ Heat treatment 1550°F oil quench, 750°F temper.² Test plates welded after heat treatment were retempered at 750°F.³ Haz = Heat-affected zone.⁴ Joint efficiency based upon average base metal ultimate strength; 204 ksi ultimate, 185 ksi yield, 7% elongation, and 38% reduction area.

both single-pass and two-pass welds in material heat treated before welding, and for two-pass welds in samples heat treated after welding. To evaluate the effect of weld bead geometry, specimens were tested with the weld ground flush and with the weld not ground.

As expected, the highest joint efficiencies (100%) were obtained with specimens heat treated after welding. The specimens heat treated before welding and retempered at 750°F after welding had an average joint efficiency of 95%. It was significant that the undercut in the single-pass welds as illustrated in Fig. 2a had no deleterious effect on the strength properties. Tensile strengths were only slightly higher with the single-pass welds than with the welds having a cover pass.

D6-AC

Table V lists the tensile properties of electron-beam welds in 0.090-in. thick D6-AC steel sheet conventionally heat treated after welding to 227-ksi base metal tensile strength. As typical with heat-treated electron-beam welds, the tensile fractures occurred in the base metal at a 100% joint efficiency.

Tensile strength results for single-pass and two-pass welds in D6-AC material heat treated before welding are shown in Table VI. These were obtained using 0.100-in.-thick D6-AC steel sheet. The slightly increased carbon content of this material as reported in Table I probably accounts for the higher 235-ksi base metal strength.

Photographs of the fractured tensile specimens of the single-pass and two-pass welds are shown in Figs. 10 and 11, respectively. It can be seen that the location of fracture was mostly in the base metal for all specimens except the single-pass welds not ground after welding. Interestingly, the joint efficiency of all samples approached 100% regardless of the test condition.

The electron-beam welding of ausformed D6-AC steel was investigated by welding together two 0.055-in. thick by 14-in. diameter cylinders which had been ausformed by shear spinning. The cylinders were girth welded using a penetration weld plus a cover pass. The cylinder was then cut into longitudinal strips for tensile testing. One set of specimens was tempered at 1000°F and another at 1075°F. Since the specimens were cut from a cylinder, they were curved across the width at a 7-in. radius, however, this appeared to have no effect on the test results as reported in Table VII. The joint efficiencies averaged 95%.

Table V
Tensile Properties of Electron-Beam
Welds in 0.090-In. Thick Ladish D6AC Steel
Heat Treated after Welding

Condition	0.2% Yield Strength, ksi	Ultimate Strength, ksi	Percentage Elongation, 2 in.	Reduction Area, percentage	Location of Fracture	Joint Efficiency, percentage
Unwelded base metal heat treated ¹	197	226	10	44		
	204	227	10.5	49		
Annealed, electron- beam welded and heat treated ²	197	225	9	46	Base metal	99
	205	227	9	44	Base metal	100
	202	227	9	45	Base metal	100

¹ Heat treatment 1650°F oil quench, 1000°F double temper.

² Welds were deposited using a penetration pass and a cover pass as described in Table II.

Table VI
Tensile Properties of Electron-Beam
Welds in 0.100-In. Thick D6AC Steel
Heat Treated before Welding¹

Condition	Yield Strength, ksi	Ultimate Strength, ksi	Percentage Elongation, 2 in.	Reduction Area, percentage	Location of Fracture	Joint Efficiency, Percentage
Single-pass weld, ³ ground flush	213	234	9	44	Base metal	100
	213	236	10	42	Base metal	100
	213	235	9	42	Base metal	100
Single-pass weld, not ground	212	234	7	13	Haz	99
	213	234	6	13	Weld	99
	213	234	6	15	Haz	99
Two-pass weld (0.090 in. wide cover pass), Ground flush	210	230	9	41	Base metal	98
Two-pass weld, not ground	211	230	8	41	Haz	98
	211	229	6	12	Base metal	97
	211	230	8	41	Base metal	98

¹ Heat treatment 1650°F oil quench, 1000°F double temper before welding plus 1000°F temper after welding.

² Joint efficiency based upon average base metal ultimate strength of 235 ksi.

³ Single pass welds were approximately 0.060-in. wide. Two two-pass welds had a cover pass approximately 0.090-in. wide. Refer to Figure.

⁴ Two samples broke prematurely owing to off-seam weld condition and lack of penetration.

Table VII
Tensile Properties of Electron-Beam Welds
in 0.055-In. Thick D6AC Steel
Ausformed before Welding¹

Condition	Yield Strength ksi	Ultimate Strength, ksi	Percentage Elongation, 2 in.	Reduction Area, percentage	Location of Fracture	Joint Efficiency, percentage
Unwelded base metal, 1000°F temper	261	265	5	28		
Ausformed	260	261	5	32		
electron-beam Welded	239	249	2	10	Haz ³	95
1000°F temper ²	239	242	2	10	Haz	92
Unwelded base metal,	250	256	2	10	Haz	97
1075°F temper	245	246	5	42		
Ausformed	249	251	5	36		
electron-beam welded,	226	234	5	27	Haz	94
1075°F temper ²	224	232	2	13	Haz	93
	232	239	2	6	Haz	96

¹ Samples were cut longitudinally from cylinders which were ausformed to 75% reduction and girth welded.

² Welds were made using a penetration pass and a cover pass. The welds were not ground flush with the base metal. Weld samples were retempered at specified temperature after welding.

³ Haz = Heat-affected zone.

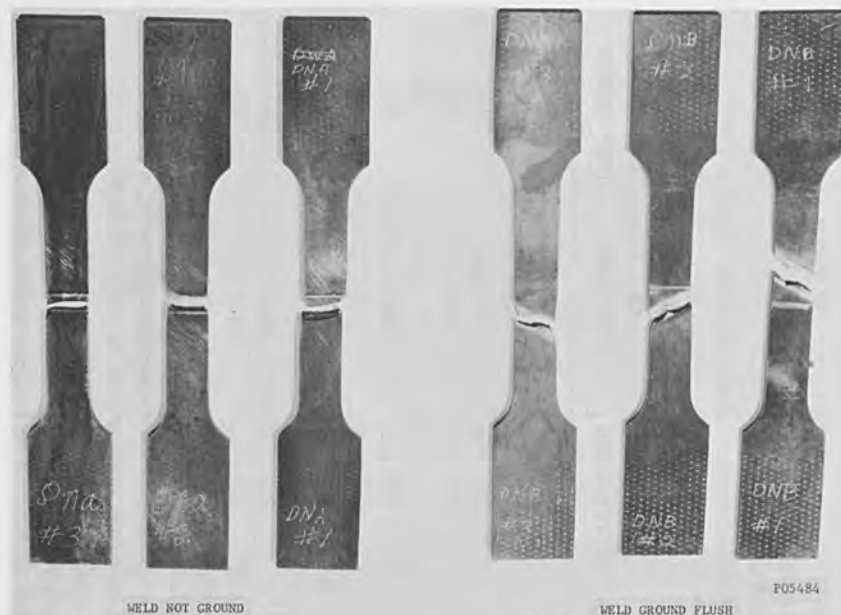


Fig. 10. Photograph of tensile test specimens of single-pass electron-beam welds in D6AC steel, heat-treated before welding.

The properties of electron-beam welds in 1/2-in.-thick D6-AC steel were determined using pancake forgings hardened to three strength levels before welding. Included were a conventionally heat-treated forging and two forgings which has been ausformed during forging to 50 and 75% reduction, respectively. For comparison, the 75% ausformed material was also tungsten-arc-welded.

As is seen in Table VIII the electron-beam weld joint efficiencies were about 97% in the conventional structures and 85 to 88% in the ausformed structures. Although the joint efficiencies in the ausformed steels were lower, higher residual strengths were observed. The joint efficiencies were exceptional for such thick material particularly when they are compared with the 65% joint efficiency obtained with the tungsten-arc welds. The retention of strength in the electron-beam welded material must be attributed to the rapid heating and cooling conditions which are inferred by the heat input calculations. As a result conditions necessary for softening through alloy diffusion are in existence for relatively short time periods.

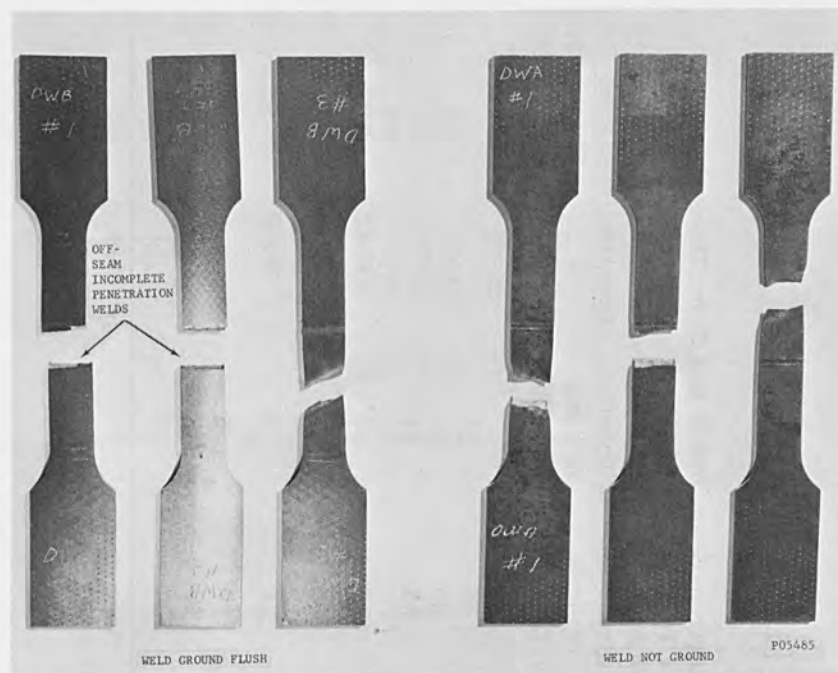


Fig. 11. Photograph of tensile test specimens of two-pass electron-beam welds in D6AC steel, heat-treated before welding.

X-2 Steel

The X-2 steel responds very well to Ausform strengthening and was included in the electron-beam welding investigation because of its potential as a missile steel. As shown in Table IX, welds in 0.100-in. X-2 ausformed by rolling to a strength level of 264 ksi had an average joint efficiency of 91%.

H-11 Steel

The welding of ausformed H-11 steel sheet demonstrated most significantly the potential of the electron-beam process in the fabrication of hardened steel. Welds in 0.100-in.-thick H-11 steel ausformed to 375 ksi before welding showed an average strength of 337 ksi for a joint efficiency of 90%. Strengths of this magnitude

Table VIII
Round-Bar Tensile Properties of Electron-Beam Welds in 1/2-In. Thick
D6AC Pancake Forgings Tempered at 900°F

Condition	0.2% Yield Stress, ksi	Ultimate Stress, ksi	Reduction of Area, percentage	Elongation in 1 in., percentage	Location of Fracture	Joint Efficiency, percentage
Conventional heat-treated base metal	222	244	32	11		
	221	243	35	11		
	220	243	35	11		
Conventional heat-treated electron-beam welded ¹	217	237	17	5	Weld	97
	215	237	18	6	Weld	97
	216	238	16	5	Weld	98
	220	238	21	7	Weld	98
Ausformed 50% base metal	261	278	17	6		
	264	281	19	6		
	261	277	20	6		

Table VIII, cont'd.

Ausformed 50% electron-beam welded	237	249	15	4	Weld	89
	236	245	12	3	Weld	88
	235	247	18	5	Weld	88
	236	248	12	3	Weld	89
Ausformed 75% base metal	281	293	26	8		
	282	296	22	8		
	277	293	15	7		
Ausformed 75% electron-beam welded	241	252	12	3	Weld	86
	243	249	15	3	Weld	84
	241	253	15	3	Weld	86
	241	242	15	3	Weld	82
Ausformed 75% tungsten-arc welded.	177	192	49	13	Weld	65
	179	193	53	12	Weld	65
	176	192	52	14	Weld	65

¹ Welded samples were retempered at 900°F after welding.

Table IX
Tensile Properties of Electron-Beam Welds
in 0.100 In. Thick X-2 Steel Ausformed before Welding¹

Yield Strength ksi	Ultimate Strength ksi	Elongation 2-In. percentage	Reduction Area percentage	Location of Fracture	Joint ² Efficiency percentage
(³)	241	3	8	Haz	91
(³)	239	3	9	Haz	90
226	243	3	5	Haz	92
223	242	3	9	Haz	92
218	238	3	5	Haz	90
222	242	3	11	Haz	92
227	243	3	4	Haz	92
(³)	242	3	7	Haz	92

¹ Material ausformed to 65% reduction and double tempered at 1100°F before welding, and re-tempered at 1100°F after welding. Welds were ground flush with parent material before testing.

² Joint efficiency based upon average base metal ultimate strength of 264 ksi.

³ Extensometer malfunction occurred.

Table X
Sheet Tensile Properties of Welds
in 0.100-In. thick H-11 Steel Ausformed before Welding¹

Type of Weld	Ultimate Strength, ksi	Percentage of Elongation, 2 in.	Location of Fracture	Joint ² Efficiency, percentage
Tungsten arc	200	3.5	Haz ³	53
	239	3.0	Haz	64
	239	3.0	Haz	64
	206	4.0	Haz	55
High-voltage electron beam	355	3.0	Haz	95
	334	3.0	Haz	89
	339	3.0	Haz	90
	342	2.5	Haz	91
	351	2.5	Haz	94
	315	2.0	Haz	84
Low-voltage electron beam	320	2.0	Haz	85

¹ Material ausformed to 70% reduction and double tempered at 600°F before welding, and tempered at 600°F after welding.

² Joint efficiency based upon average base metal ultimate strength of 375 ksi.

³ Haz = heat-affected zone.

across an as-welded joint are phenomenal. The results are compared in Table X with the 60% joint efficiency of tungsten-arc welds in the same material. Most of the electron-beam welding of the H-11 steel was done with high-voltage equipment, however, the result of one test with the low-voltage-type weld fell within the general strength range.

250 Series Maraging Steel

The tensile properties of electron-beam welds in 0.090-in.-thick 250 series Maraging steel are reported in Table XI. Welds were made in both solution treated and solution treated and aged material. Subsequent to welding, the test plates were aged at 900°F for 3 hours.

The joint efficiencies were comparable for both heat treat conditions, however, the welds made in solution treated and subsequently aged material showed better elongation properties.

FRACTURE TOUGHNESS

It is pertinent to question what effect a weld might have on the reliability of a fabricated structure. In order to define this, fracture toughness, K_{Ic} , evaluations were made of electron-beam weld and base metal samples [3]. Standard ASTM specimens [18], 3-in. wide by 12-in. long with 1 in. center slots were used. The slots were machined by the electrical discharge process to produce notch radii ≤ 0.001 in. Doublers were welded on both sides of the loading pin holes. Specimens were loaded at a load rate of approximately 5000 lb/min. To measure the slow crack propagation, the ink method was used.

Electron-beam weld specimens were prepared such that the weld was located at the center length of the specimen transverse to the direction of loading. Eloxed cracks were machined with the crack tips as nearly in the center of the weld as possible. In Table XII, it is seen that the fracture toughness level averaged 277 ksi $\sqrt{\text{in.}}$ for 0.090-in. D6-AC welded samples heat treated after welding and 305 ksi $\sqrt{\text{in.}}$ for base metal samples. Failures were 100% shear with the crack propagation occurring in the weld. The yield strengths of both base metal and electron-beam weld sections were in the order of 200 ksi.

Fracture toughness evaluations of welds in ausformed D6-AC steel were accomplished with longitudinal samples cut from the

Table XI
Tensile Properties of Electron-Beam Welds
in 0.090-In. Thick 250 Series Maraging Steel

Heat Treat Condition	Yield Strength, ksi	Ultimate Strength, ksi	Percentage of Elongation 2 in.	Reduction Area, percentage	Location Fracture	Joint Efficiency, percentage
Base metal solution treated and aged ¹	282 281	284 283	5.0 4.5	49 50		
Solution treated electron-beam welded ² and aged	237 248 278 269	266 273 280 270	4.0 4.0 3.0 3.5	38 38	Base metal Base metal Weld Weld	94 96 99 95
Solution treated aged	271 278	275 279	2.0 2.5	15 15	Weld Weld	97 98
electron-beam welded ² and aged	265 258	265 258	1.5 2.0		Weld Weld	93 91

¹ Aging temperature 900°F for 3 hours.

² Two-pass weld having cover pass approximately 0.090-in. wide.

Table XII
Tensile and Fracture Toughness Properties of 0.090 in. thick D6-AC Base Metal
and Electron-Beam Welds Conventionally Heat Treated
after Welding -1000°F Temper

Condition and Temper	0.2 Yield Stress, ksi	Ultimate Stress, ksi	Gross ¹ Stress, σ_g , ksi	Crack Length, in. 2 amp,	Fracture Toughness kc, ksi./in.	Net Section Stress σ net, ksi	Shear Lip, percentage
Heat-treated 1000°F	200.2	226.7	124.0 116.0 ²	1.50	305	245.0	100
Electron-beam welded, heat-treated, 1000°F	201.2	226.4	³ 123.0 96.0	1.57 1.9	308 266	255.0 257.0	100 100

¹ Gross stress of center-cracked 3-in. wide specimens.

² Did not fracture in notch.

³ Joint efficiencies were approximately 99.5%.

Table XIII
Fracture Toughness Properties of Electron-Beam Welded
and Base Metal Ladish D6-AC 0.055-In. Thick

Condition and Temper	0.2 Yield Stress, ksi	Ultimate Stress, ksi	Gross ¹ Stress, σ_0 , ksi	Crack Length, in. 2 amp	Fracture Toughness K_{IC} , ksi/in.	Net Section Stress σ net, ksi	Shear Lip, percentage
Base metal ausformed 1000°F	260.6	262.7	97.0 106.5	1.57 1.55	191 214	203 224	100 100
Ausformed electron-beam welded 1000°F	242.8	248.9 ³	92.5 104.0 107.5	1.27 1.46 1.32	155 194 187	173 196 191	100 100 100
Base metal ausformed 1075°F	247.0	248.6	121 >102 ²	1.54 ...	250 ...	239 ...	100 ...
Ausformed electron-beam welded 1075°F	227.1	234.7 ³	98.0 91.5 101.0	1.32 1.48 1.37	177 188 179	200 209 181	100 100 100

¹ Gross stress of center-cracked 2 1/2 - to 3-in.-wide specimens.

² Fractured in loading pin holes.

³ Joint efficiencies were approximately 94%.

0.055-in.-thick by 14-in. diameter electron-beam welded cylinder previously described. The test results are seen in Table XIII. Fracture toughness values of about 180 ksi $\sqrt{\text{in.}}$ were obtained for the welds for material tempered at 1000°F and 1075°F. In all cases, the crack propagation was entirely in the weld material and 100% shear failures were observed in this material which had yield strengths in the range of 240 to 270 ksi. Since the specimens were taken from large diameter shear spun material, fracture toughness of both base metal and welds should be representative of a fabricated cylinder. The toughness levels would be more than sufficient to avoid premature failure in a pressure vessel with girth welds.

PRODUCT APPLICATIONS

Electron-beam welding has been used for production fabrication of aircraft and missile components for 16 months at the Aeronutronic Division. A major production application has been the welding of 6-in. diameter missile cases which were previously welded by the tungsten-arc process with discouraging results. At that time, weld porosity was a continuous problem and fractures in hydrostatic burst testing generally initiated at the weld. In contrast, fractures in similar tests on electron-beam welded cases have never been associated with the weld.

A photograph of a typical missile case weld and a sketch of the slant butt joint design is presented in Fig. 12. The slant butt joint facilitates mating of the parts using simple end thrust tooling. The assembly was placed on a turntable and welded with the electron-beam gun in a horizontally placed position. Weld distortion was so minimal that machining was not necessary after welding.

Large size missile cases up to 14-in. in diameter and 10-ft. in length were welded using the chamber extension shown in Fig. 1. In some applications, conventionally heat-treated heads were welded to cases which had been ausformed by shear spinning. Only light tooling was needed to hold the abutting edges in alignment for welding.

An application involving electron-beam fillet welding of attachments is illustrated by the photograph in Fig. 13. The simulated fin support sample of 250 Maraging steel was welded to the curved section using preplaced filler wire. As indicated, no longitudinal distortion was observed. Also, the curved section was not altered in shape by the heat of welding. Tungsten-arc welding in such an

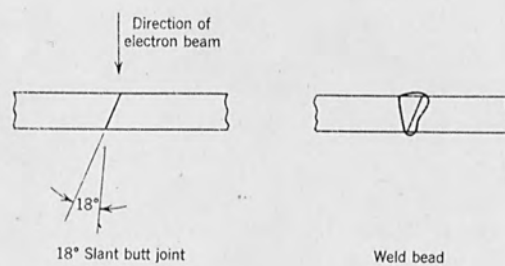
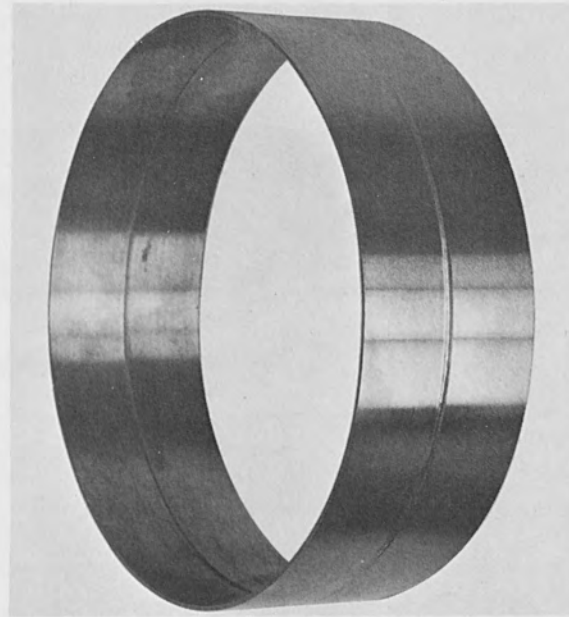


Fig. 12. Photograph illustrating electron-beam girth weld in 0.050-in. thick by 6-in. diameter D6AC steel cylinder.

application would have distorted the case beyond specified dimension tolerance.

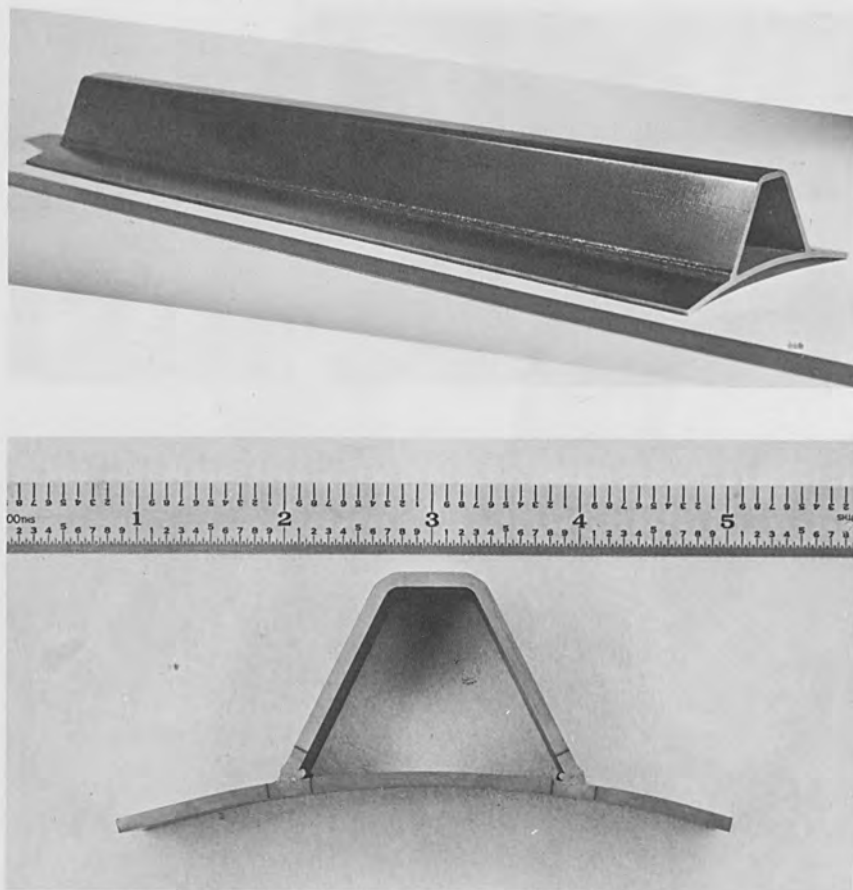


Fig. 13. Photograph illustrating electron-beam fillet welds in simulated fin channel of 250 series maraging steel.

CONCLUSIONS

The ultra-high-strength steels are readily welded both in the hardened and unhardened conditions by the electron-beam process without preheating or postheating in thicknesses up to 0.100 in.

Heat input developed by low-voltage-type electron-beam welding is less than one-third that developed by tungsten-arc welding in comparable material thicknesses.

Electron-beam welds and heat-affected zones show a fine grain metallographic structure which contributes to their superior properties over coarse grain arc welds.

Joint efficiencies approaching 100% of the parent material are obtained in uniaxial tensile tests of electron beam welds in 4130 and Ladish D6-AC steel when welded in the annealed condition and subsequently heat treated.

Electron-beam welding shows the greatest advantage in welding steels in the hardened condition. Joint efficiencies exceeding 95% are obtained in welding conventionally heat-treated steels. Welds in ausformed steels show joint efficiencies averaging better than 90% in sheet materials and 87% in 1/2-in.-thick D6-AC steel.

Tungsten-arc welds in materials hardened before welding show a maximum joint efficiency of 65%.

Fracture toughness values of electron-beam welds average 287 ksi $\sqrt{\text{in.}}$ in D6-AC heat treated after welding at a yield strength of 200 ksi and 180 ksi $\sqrt{\text{in.}}$ in D6-AC ausformed before welding at a yield strength of 250 ksi. All samples showed 100% shear lip fracture.

REFERENCES

1. H.J. Hucek, A.R. Elsea, and A.M. Hall, "Evolution of Ultra High Strength Hardenable Steels for Solid Propellant Rocket-Motor Cases," DMIC Report No. 154, Battelle Memorial Institute, 1961.
2. C.F. Martin, W. W. Gerberich, J. M. McCamont, and E. L. Harmon, "Research in the Mechanism of Strengthening in Ausformed Steel," ASD Technical Documentary Report No. ASD-TDR-62-692.
3. W. W. Gerberich, A. J. Williams, C. F. Martin, and R. E. Heise, "Ausform Fabrication and Properties of High Strength Alloy Steel," Presented AIME, Materials Congress, Cleveland, 1963, to be published in ASTM Bulletin.
4. R. P. Sopher, "Effects of Chemical Composition on Cracking Resistance of High-Strength Steel Weld Metals," Welding Journal Research Supplement, Nov. 1958, pp. 481s-492s.
5. H. W. Mischler, R. E. Monroe, and P. J. Rieppel, "Welding of High Strength Steels, for Aircraft and Missile Applications," DMIC Report 118, Battelle Memorial Institute, Oct. 12, 1959.
6. M. D. Randall, R. E. Monroe, and D. C. Martin, "New Developments in Welded Fabrication of Large Solid-Fuel

- Rocket Motor Cases," DMIC Report No. 173, Battelle Memorial Institute, Aug. 6, 1962.
7. J. T. Niemann and P. J. Rieppel, "Arc Welding of High-Strength Steels for Aircraft and Missile Structures," DMIC Memorandum No. 27, Battelle Memorial Institute, 1959.
 8. M. D. Randall and P. J. Rieppel, "Welded Fabrication of Steel Solid-Propellant Rocket Motor Cases," DMIC Memorandum No. 56, Battelle Memorial Institute, 1960.
 9. M. J. Albom and C. C. Titherington, "Investigation of Weldability of Ultra-High Strength Steels," Welding Journal Research Supplement, Sept. 1960, pp. 385s-391s.
 10. H. W. Mischler, R. E. Monroe, and P. J. Rieppel, "Welding of High Strength Steels for Aircraft and Missile Applications," DMIC Report No. 118, Battelle Memorial Institute, Oct. 12, 1959.
 11. J. F. Rudy and H. Swartzbart, "Welding of an Ultra-High Strength Steel for Missile Motor Cases," Welding Journal Research Supplement, June 1961, pp. 241s-252s.
 12. B. S. Blum and R. H. Witt, "Tungsten Arc Welding of H-11 Steel Sheet," Welding Journal Research Supplement, June 1962, pp. 274s-278s.
 13. J. R. Dyar and N. F. Bratkovich, "Reliable Weld Joint Design for High Strength Rocket Motor Cases," Welding Journal Research Supplement, March 1963, pp. 126s-133s.
 14. E. F. Nippes and E. W. Emmerich, "Heat Affected Zone Study of an Eutectoid Ultra-High Strength Steel," Welding Journal Research Supplement, Dec. 1963, pp. 547s-556s.
 15. R. E. Roth and N. F. Bratkovich, "Characteristics and Strength Data of Electron Beam Welds in Four Representative Materials," American Welding Society Presentation Fall Meeting, Sept. 1961.
 16. H. A. Hokanson and J. W. Meier, "Electron Beam Welding of Aircraft Materials and Components," Welding Journal, Nov. 1962, pp. 1000-1008.
 17. J. W. Meier, "Electron Beam Welding of Aerospace Materials," Technical Documentary Report No. ASD-TDR-63-132, Feb. 1963.
 18. ASTM "Fracture Testing of High-Strength Sheet Materials," Philadelphia, Jan. 1960.

ELECTRON-BEAM WELDING OF BRAZED REFRACTORY JOINTS

INTRODUCTION

The introduction of refractory metals into the aerospace program has brought with it many new problems in design, joining, and fabrication. One of the tools developed in recent years, which facilitates the welding of refractory metals, is the electron-beam welder (Fig. 1). This machine, by focusing electron flow into a



Fig. 1. Hamilton Standard electron-beam welder.

*The Martin Company, Baltimore, Md.

very narrow beam, generates high energy sufficient to produce 0.010-in. diameter welds in steel 1-in. thick. Since this high energy is generated over such a small area, the total energy involved in the process is less than that of conventional welding methods, resulting in narrower heat-affected zones and less distortion. Also, because the welding is accomplished in vacuum (10^{-4} mm Hg), contamination and porosity are minimized.

The Advanced Manufacturing Technology Laboratory of the Martin Company has conducted research into the electron-beam welding of brazed refractory joints. Power and weld speed settings were established for welding 0.008-in. D-36 columbium* and TZM molybdenum. These settings were then used to make welds necessary in the fabrication of a D-36 brazed honeycomb heat shield panel (Figs. 2 and 3) and a TZM brazed honeycomb structural panel (Figs. 4 and 5).

The effects of the honeycomb pattern, brazed onto the underside of the refractory sheet which was electron-beam welded, and



Fig. 2. D-36 brazed honeycomb heat shield panel.

*Other authors in this book prefer to refer to this refractory metal as niobium. Both names are accepted and used in industry (Ed.).

the effect of the braze alloy itself in the weld nugget, were studied. The results of this investigation were applied to the fabrication of brazed refractory honeycomb panels.

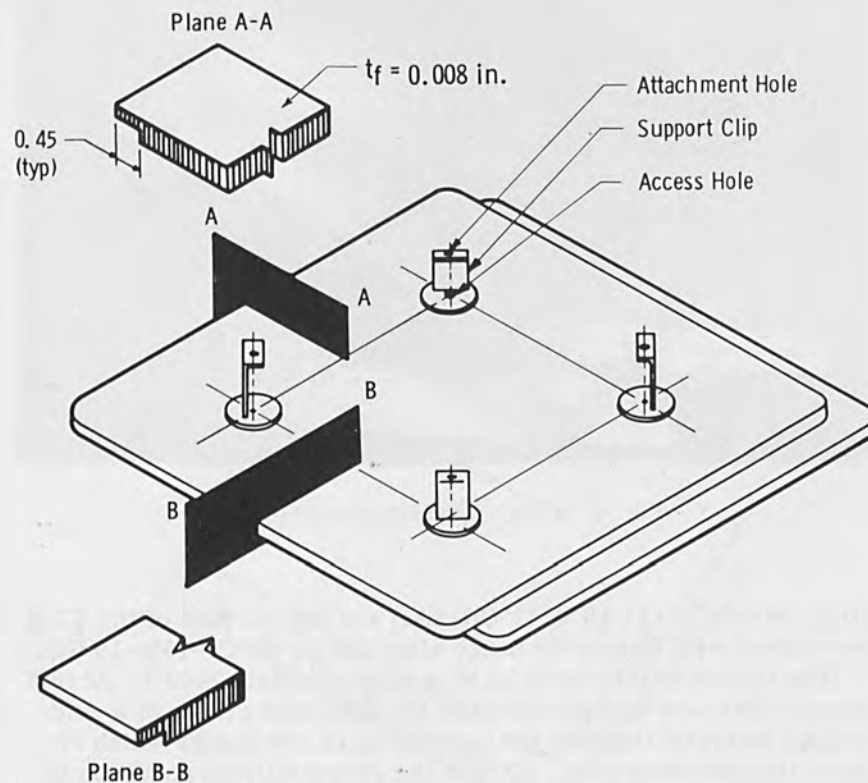


Fig. 3. Schematic of D-36 brazed honeycomb heat-shield panel.

EXPERIMENTAL PROCEDURE AND RESULTS

Materials

The refractory metals studied in this investigation were D-36 (Cb-10 Ti-5 Zr) columbium and TZM (Mo-0.5 Ti-0.08 Zr) molybdenum. One surface of the D-36 was coated with titanium braze



Fig. 4. TZM brazed honeycomb panel.

alloy, B120VCA (Ti-13 V-11 CR-3Al) and one surface of the TZM was coated with Haynes 25 braze alloy (55 Co-20 Cr-15W-10 Ni).

The recrystallization of TZM is approximately 2400°F. At this temperature, the energy stored in the deformed grains of a cold-worked material triggers the generation of new grains which replace the deformed ones. TZM in the recrystallized condition is extremely brittle. The ductile-to-brittle transition temperature of TZM is 250°F. Above this temperature, TZM is ductile.

Welding of D-36

Initial welds were run on flat pieces of D-36, 0.008-in. thick, to determine the settings that would result in a uniform weld nugget, with 100% penetration when no braze alloy was present. The D-36 used in this experiment was treated for five minutes at 3000°F, so that it would have the same metallographic structure as the material on the heat shield panel itself. D-36 in this condition has an extremely coarse-grained structure. All settings were recorded (Fig. 6). Three parameters were varied: voltage, current, and

table speed. The welds were inspected microscopically and by the Zeiglo method (fluorescent dye penetrant).

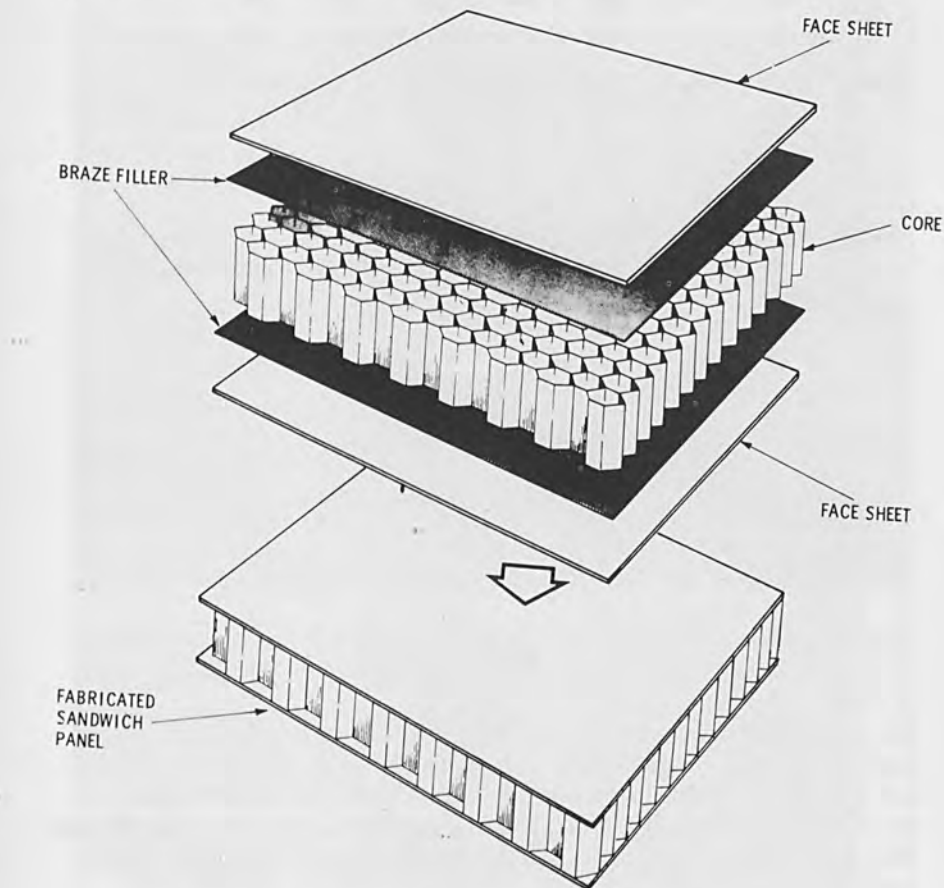


Fig. 5. Schematic of TZM brazed honeycomb panel.

Two settings were determined that would provide 100% penetration without cutting: (1) 50 kV, 1.75 mA, 20-ipm weld speed; and (2) 80 kV, 1.00 mA, 40-ipm weld speed.

The first setting is considered best. The high amperage-voltage ratio means that the work piece is bombarded by more electrons at slower speeds than in a low amperage-voltage ratio, and a wider beam is effected. Although the beam power in both settings is approximately the same, the power density, and also the energy density, are less in the first setting than in the second, and a lower

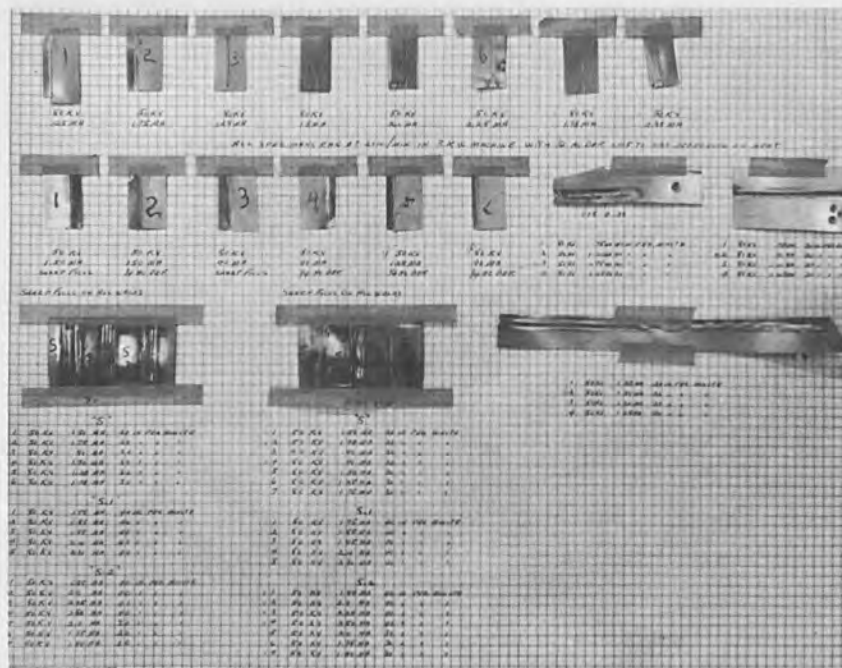


Fig. 6. Initial welds on D-36 using overlay and burn-through techniques. (Direct penetration welds on 0.008-in. sheet are also shown.)

weld speed is necessary to allow for sufficient energy per area to heat the part to its melting point. It is the author's opinion that, for thin-gauge refractory metals, this allows for a more uniform weld puddle and, as a result, a more homogeneous weld nugget, and that these advantages outweigh the disadvantages of a wider weld nugget and heat-affected zone.

B120VCA titanium alloy, 0.002-in. thick, was brazed to one side of the D-36 sheet. The welds run on the D-36 sheet were repeated, and the same settings were found to be best.

Parameters were also established for welding 0.008-in. D-36 to 0.008-in. D-36 by both the overlap and burnthrough methods (Fig. 7). These were: (1) 50 kV, 2.35 mA, 20-ipm weld speed (burnthrough); and (2) 50 kV, 1.65 mA, 20-ipm weld speed (overlap). In the overlap method, the beam was widened to 0.025 in. to enable flow to occur on both sides of the seam.

Sample straight line welds were made on a brazed D-36 honeycomb panel (Fig. 8) to determine settings that would penetrate the

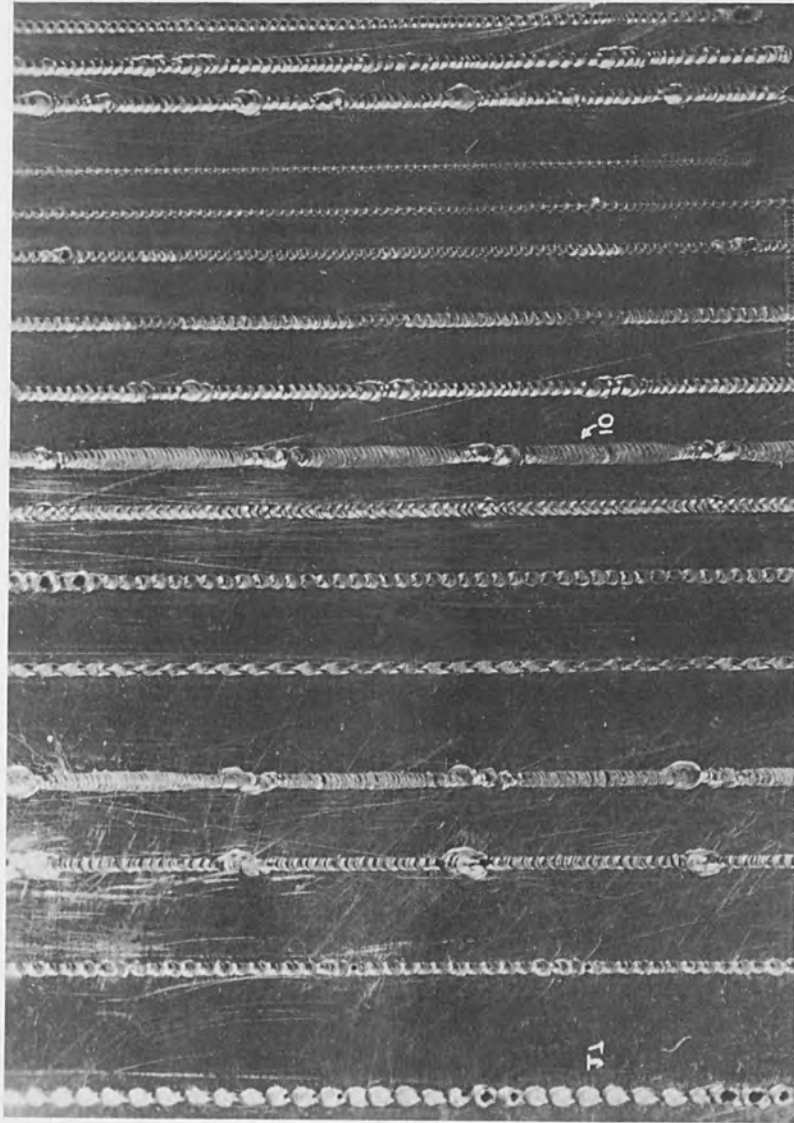


Fig. 7. Sample welds on D-36 heat shield (30X).

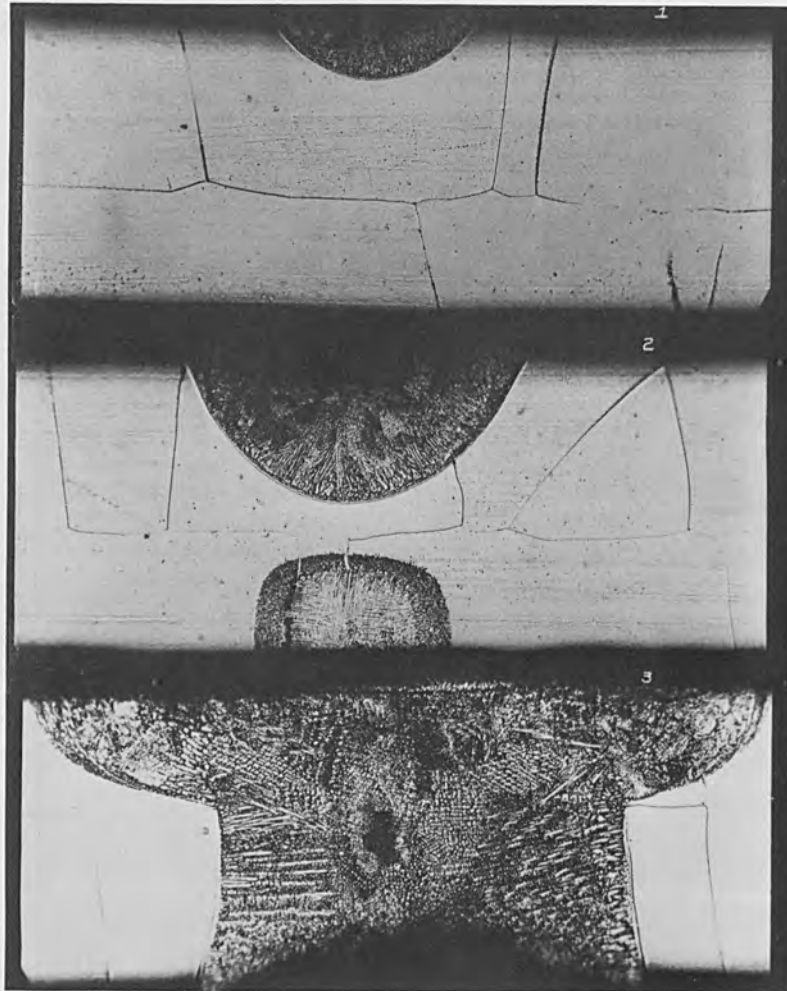


Fig. 8. Latitudinal cross section showing microstructure of welds on D-36 heat shield panels (etchant: mixed acid). 1. 30 kV, 0.50 mA, 40 ipm (300X). 2. 80 kV, 0.75 mA, 40 ipm (300X). 3. 80kV, 1.00 mA, 40 ipm (300X).

skin without leaving defects in the panel. These welds were examined micrographically (Fig. 9), determining the best setting as: 50 kV, 1.75 mA, 20-ipm table speed. This setting is the same one established for the penetration of a 0.008-in. D-36 skin. Examina-

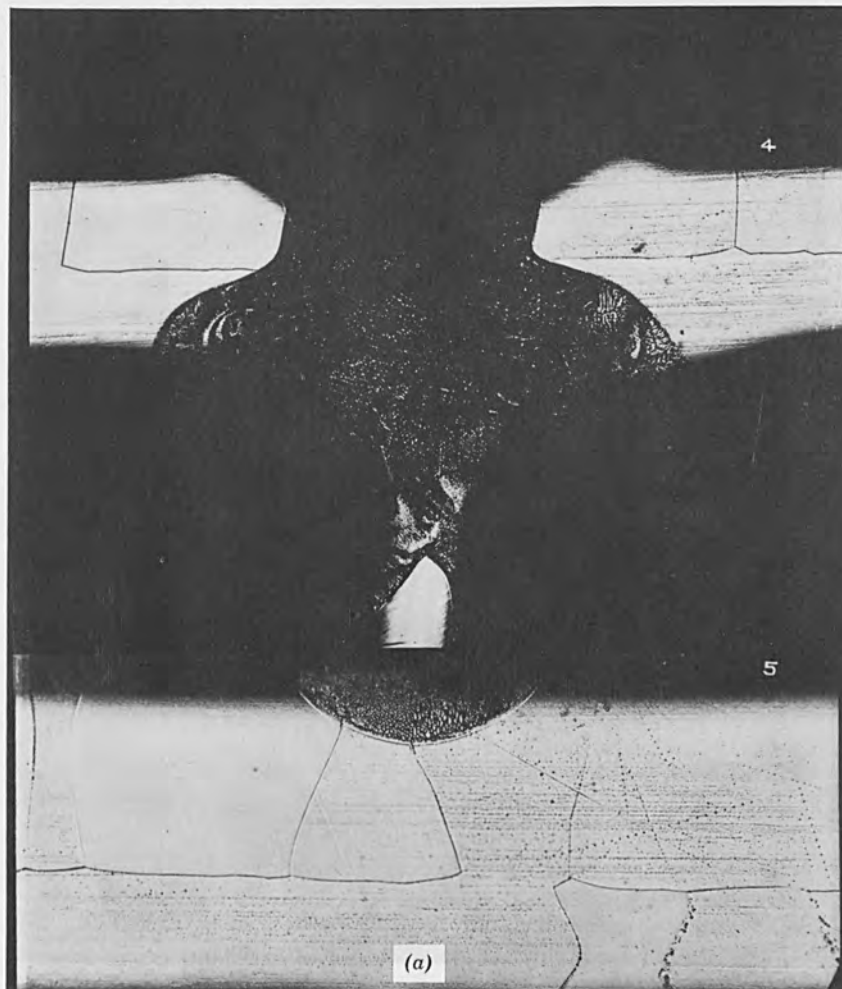


Fig. 8. 4. 80 kV, 1.25 mA, 40 ipm (200X). 5. 50 kV, 1.00 mA, 40 ipm (300X).

tion of Figs. 6 and 8 shows that the most common defect caused by too great a heat is pinholes in the weld, rather than the cutting which was apparent in a flat D-36 sheet. The pinhole defects occur at points where the honeycomb core is in contact with the D-36 skin (Fig. 10). The figure shows a photomicrograph of a weld area where a pinhole occurred, and that this defect is caused by a "pipe" effect similar to that seen in cast ingots. The honeycomb core,

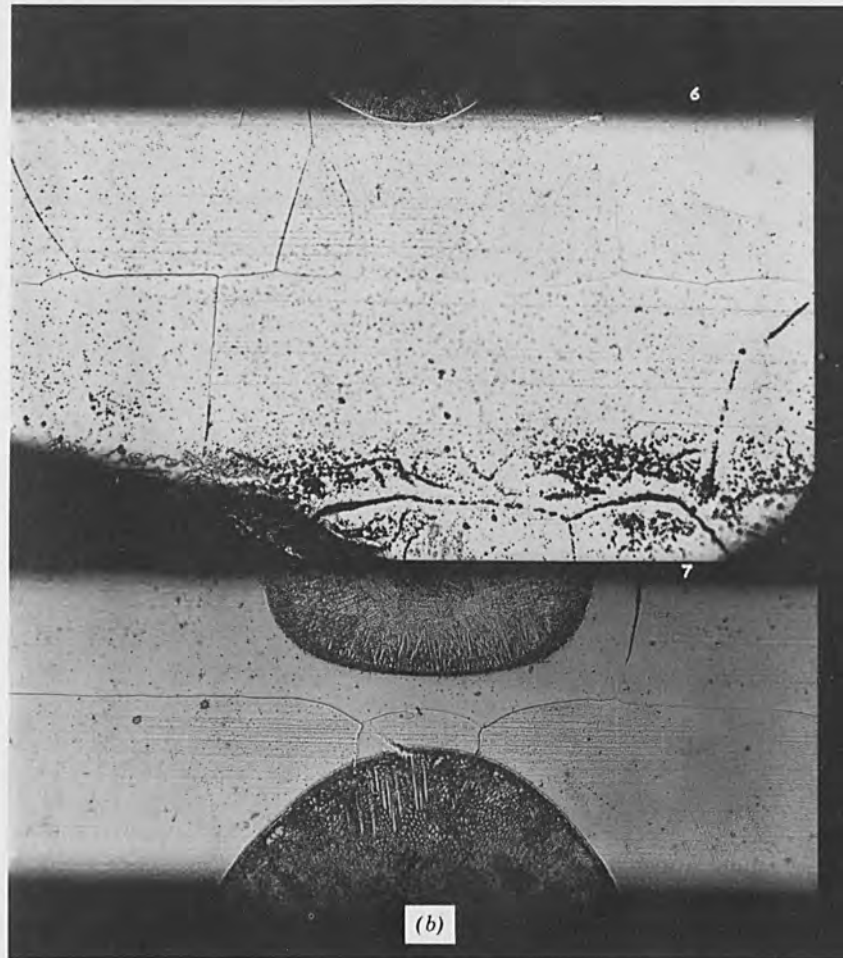


Fig. 8. 6. 50 kV, 0.75 mA, 40 ipm (400X). 7. 50 kV, 1.25 mA, 40 ipm (300X).

which is in contact at that point, acts as a chill on the molten nugget, causing solidification to occur unevenly. The resulting shrinkage in this area pulls the thin skin apart, and pinholes occur.

The final test on the D-36 alloy was the welding of a 0.008-in. thick D-36 patch onto a brazed honeycomb panel. The requirement for successful weld was the complete sealing of the area under the patch from the environment. Two shapes of patches were tested: a square 1-3/4 in. on a side, and a circle 1-1/4 in. in diam-



Fig. 8. 8. 50 kV, 1.50 mA, 40 ipm (200X). 9. 50 kV, 1.75 mA, 40 ipm (300X).

eter. Both burnthrough and overlap welds were made using the settings established above. Burnthrough welds were run at 50 kV, 2.35 mA, and a table speed of 20 ipm; overlap welds at 50 kV, 1.75 mA, and 20 ipm, with a 0.025-in. widening of the beam.

The burnthrough technique resulted in pinhole defects for both the circular and square patches. This is due to the "pipe" effect previously described. The overlap technique was successful on the square patches, but pinholes and intergranular cracks occurred in

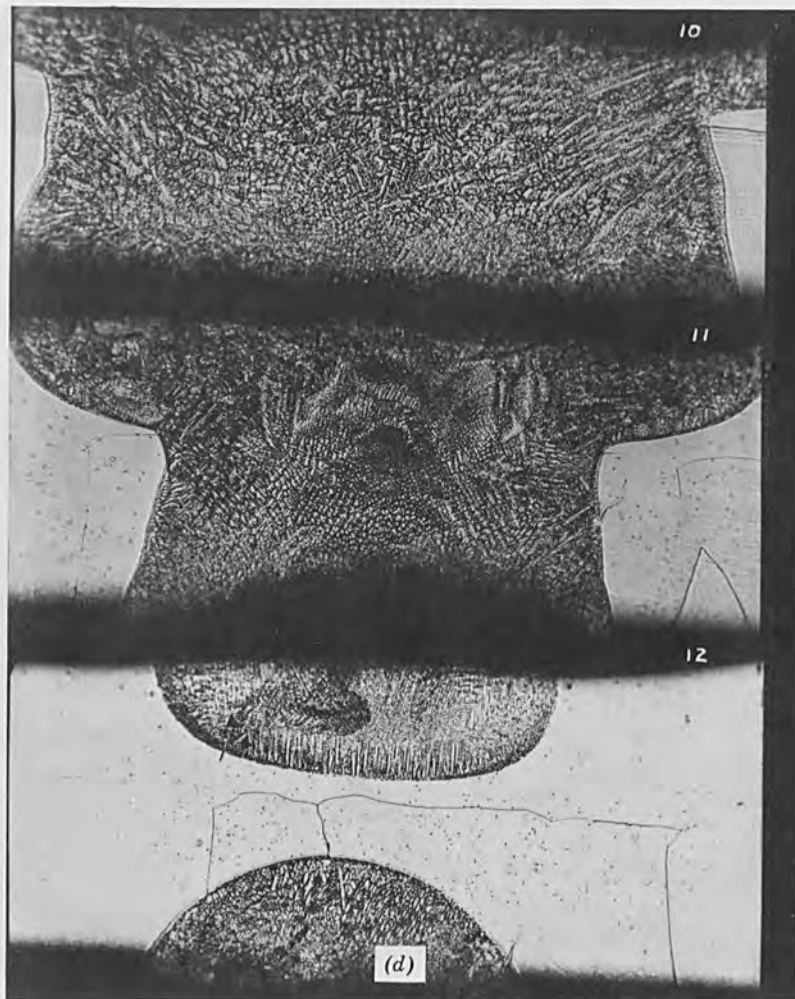


Fig. 8. 10. 50 kV, 1.75 mA, 20 ipm (300X). 11. 50 kV, 1.75 mA, 60 ipm (300X). 12. 50 kV, 1.75 mA, 80 ipm (300X).

the circular patch. Two reasons are hypothesized to explain the difficulty in welding the circular configuration. First, because the circle is not perfect, difficulty is encountered in focusing precisely on the seam, and the beam (less than 0.010 in. in diameter) cannot correct or absorb any differences. Second, the curvature of a circle means that all stresses on the weld nugget as it freezes are

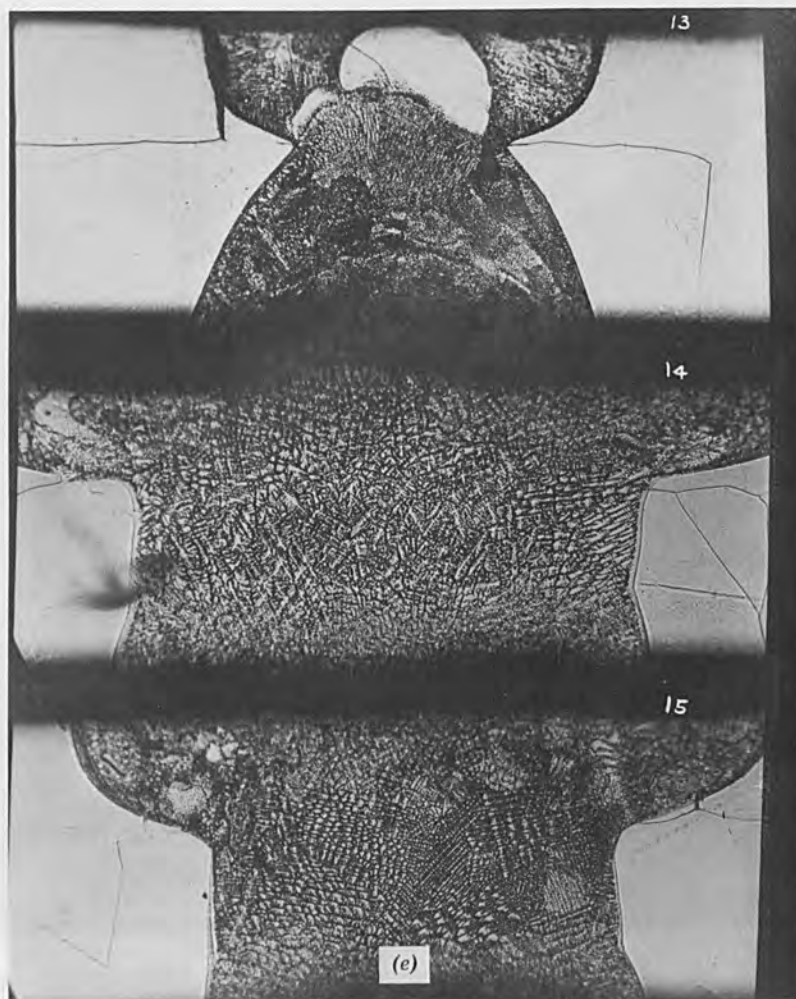


Fig. 8. 13. 80 kV, 1.25 mA, 80 ipm (300X). 14. 80 kV, 1.25 mA, 20 ipm (300X). 15. 80 kV, 1.25 mA, 60 ipm (300X).

more severe than in a straight line weld. In a square patch, areas of high stress are at the corners, but welding opposite sides of the square first, rather than adjacent sides, does much to alleviate this condition.

The advantage of an overlap weld is the ability of the liquid metal in the patch to flow into embryo pinholes or other defects

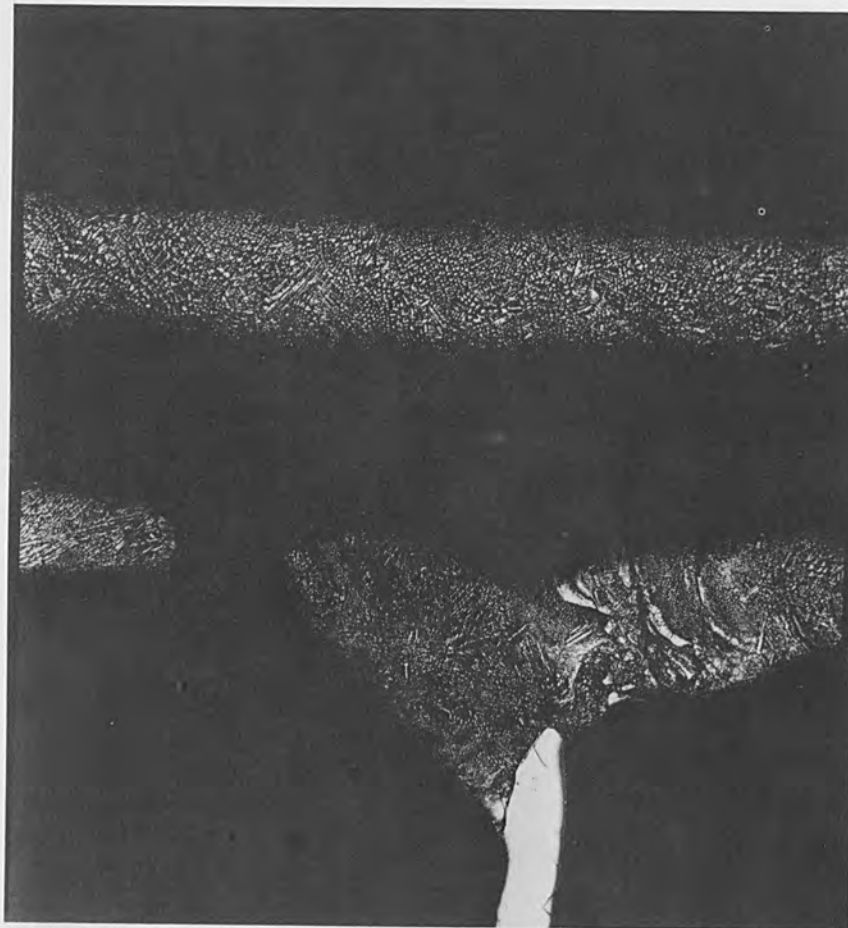


Fig. 9. Longitudinal cross section of microstructure of weld on D-36 heat shield—weld parameters: 50 kV, 1.75 ma, 20 ipm; Etchant: mixed acid (100X).

in the panel skin and make an homogeneous weld. Since the patch is not in contact with the honeycomb core, it is not subject to the chilling effect which causes pinholes.

Welding of TZM

The initial welding of TZM was similar to that done on D-36 (Fig. 11). TZM skin, 0.008-in. thick, was stress relieved for one

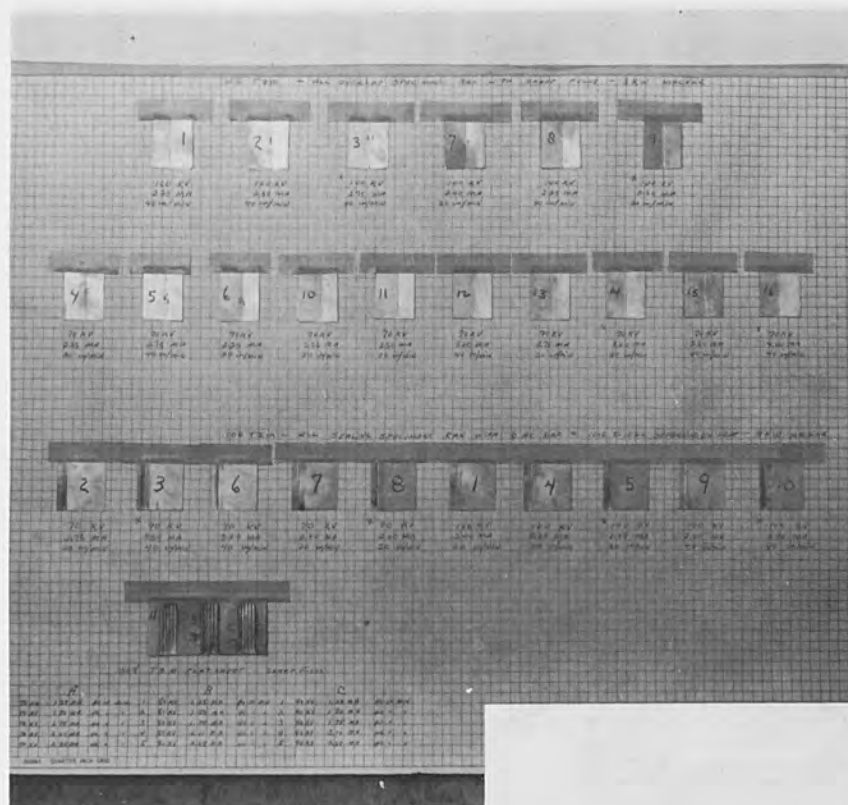


Fig. 10. Initial welds on TZM, using overlap and burn-through techniques (direct penetration welds on 0.008-in. sheet are also shown).

hour at 2100°F, as this is the condition of the honeycomb panels when the electron-beam welding is performed. Parameters were established, first for penetration of flat TZM sheet, 0.008-in. thick, and then for welding 0.008-in. sheet to 0.008-in. sheet by the overlap and burnthrough methods. The welds were inspected microscopically and by the Zeiglo method.

The best setting determined for penetration of TZM was 70 kV, 2.00 mA, 20-ipm table speed.

For welding 0.008-in. sheets together, the best settings were (1) 70 kV, 3.00 mA, 20-ipm weld speed (burnthrough), and (2) 70 kV, 2.00 mA, 20-ipm weld speed (overlap).

Here again, high amperage-voltage ratios were preferred.

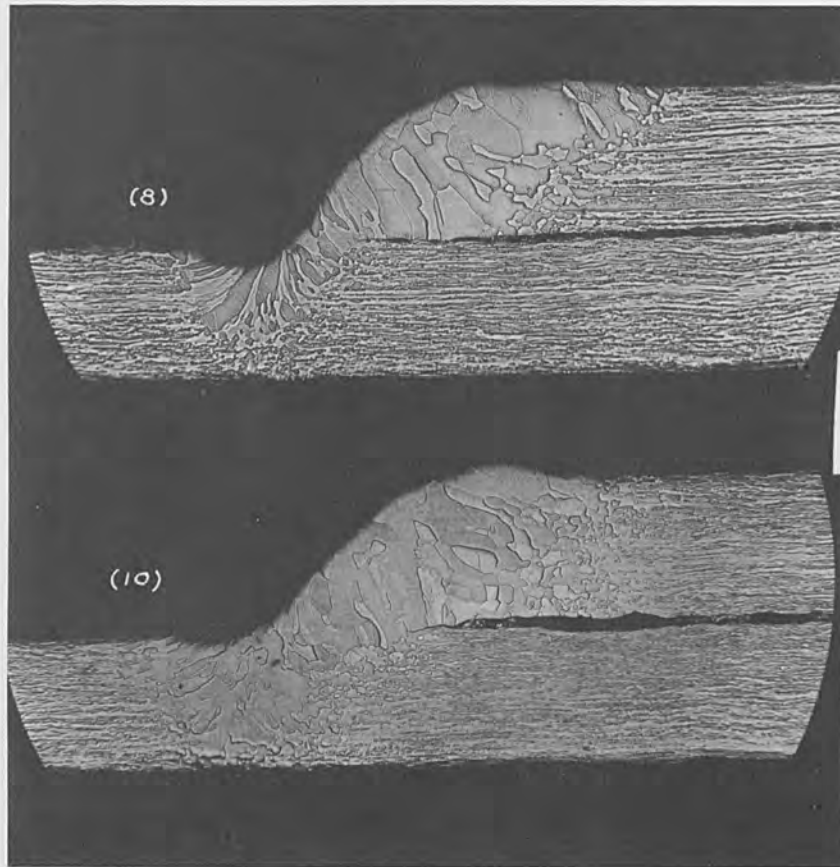


Fig. 11. Latitudinal section of TZM welds, welded by the overlap technique: 0.008-in. Sheet to 0.008-in. Sheet-etchant: No. 1 Mo. (8) 70 kV, 2.00 mA, 20 ipm (150X). (10) 100 kV, 1.75 mA, 40 ipm (150X).

The best welds were examined micrographically (Figs. 12 and 13). The weld nugget shows a coarse-grained structure, which results in an extremely brittle weld.

As in the case of D-36, the purpose of these tests was to weld a 0.008-in. thick patch to a brazed honeycomb panel. However, instead of welding the patch on a finished panel, the patch was welded onto a 0.008-in. facing prior to brazing. The braze alloy (Haynes 25) was sealed between the patch and the facing. This enabled the welding to be done without the effect of the honeycomb core underneath the flat skin. Because of the difficulties encountered in weld-

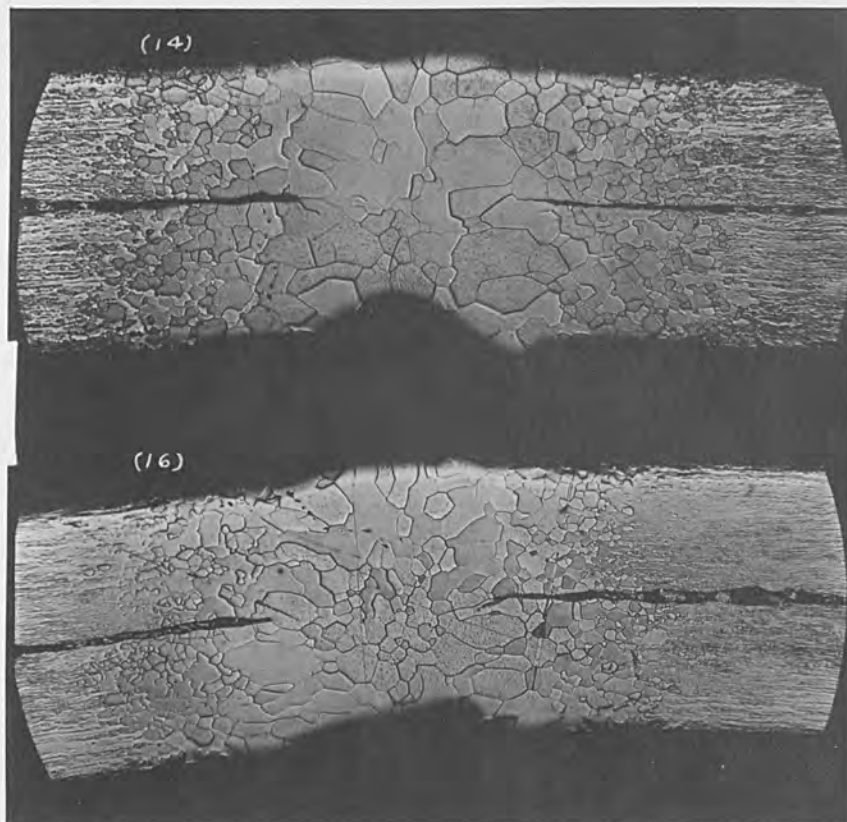


Fig. 12. Latitudinal cross section of TZM welds welded by the burn-through technique: 0.008-in. Sheet to 0.008-in. Sheet-etchant: No. 1 Mo. (14) 70 kV, 3.00 mA, 20 ipm (150X). (16) 70 kV, 4.00 mA, 40 ipm (150X).

ing D-36 circles, only square TZM patches were used. The patches were 1-3/4 in. on a side, the same as the D-36. Minimum warpage was one of the criteria for a successful weld, since a perfectly flat skin is necessary for successful brazing.

The welds were made by the overlap technique. All were extremely brittle; most were cracked. The large grained structure of the weld nugget, compounded with the fact that TZM at room temperature is below its ductile-to-brittle transition temperature, makes it almost impossible to make a successful weld in a restrained configuration.

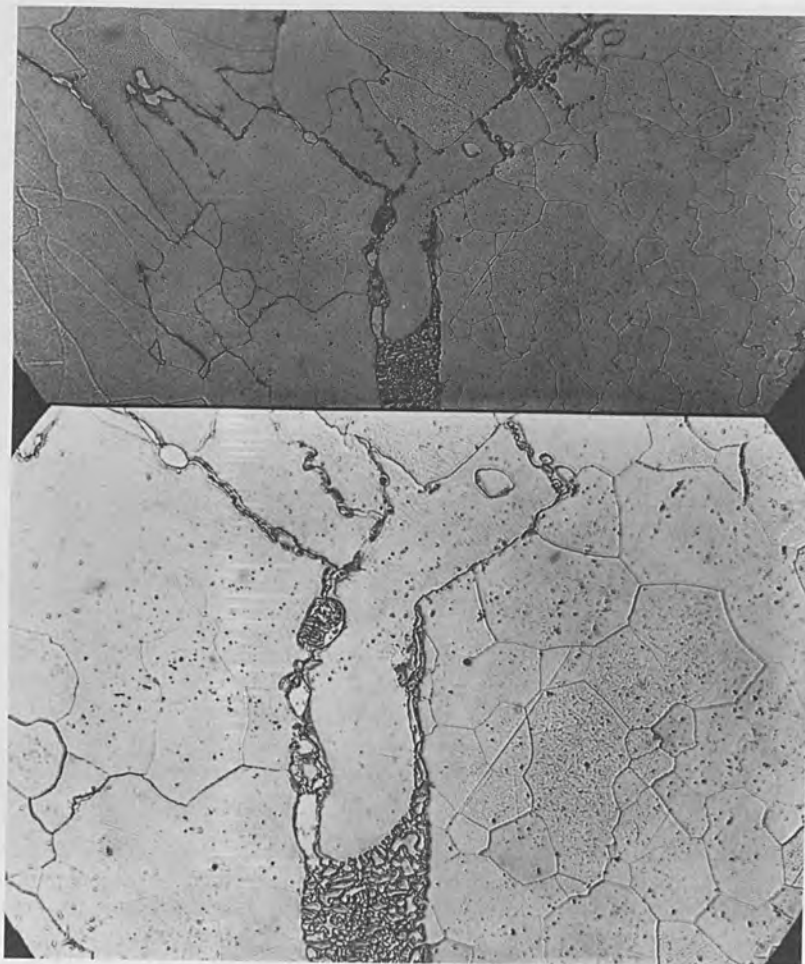


Fig. 13. Microstructure of TZM weld with secondphase present due to Haynes 25 braze alloy; etchant No. 1 Mo. Magnificant 350X (top), 700X (bottom).

A brazed honeycomb panel was sealed by electron-beam welding (Fig. 4). The braze alloy was sandwiched between a sheet, 0.012-in. thick, and a frame, 0.025-in. thick. After brazing, the weld was performed around the edge of the panel. Contamination of the weld with the Haynes 25 braze alloy could not be avoided. Because of the stresses resulting from the weld, the panel was

heated to 250°F (above the ductile to brittle transition temperature) before welding, and stress relieved at 2100°F for one hour after welding before it was allowed to cool to room temperature. Despite the extreme care taken to weld the panel under optimum conditions, the welding caused cracking of the panels.

Micrographic analysis of the cross section of the weld (Fig. 14)

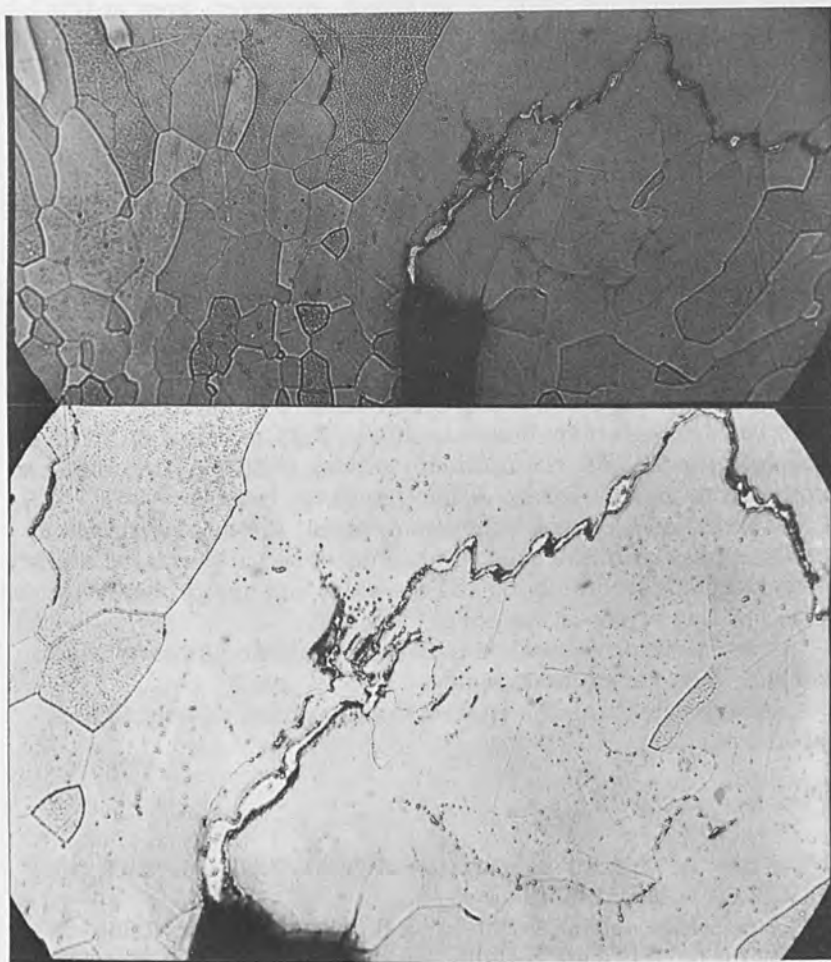


Fig. 14. Microstructure of TZM weld with second phase present due to Haynes 25 braze alloy after heat treating to 2100 °F and quench cooling: etchant: No. 1 Mo. Magnification 350X (top), 700X (bottom).

shows that the Haynes 25 braze alloy contaminated the weld nugget in two ways. It flowed into the grain boundaries of the TZM, and it alloyed with the TZM to form a precipitate. This precipitate could have caused a volume change, and thus may have introduced stresses into the weld.

A section of the weld was cut from the panel and heated to 2100°F in an inert gas atmosphere and quench-cooled to study whether precipitation could be retarded. However, even at this rapid rate of cooling, the precipitate was in evidence in the microstructure (Fig. 14). It is apparent that the welding of TZM in the presence of Haynes 25 introduces great stresses. Welds of high integrity in a constrained part could not be made.

CONCLUSIONS

D-36

The best setting for penetration of D-36, 0.008-in. thick, was 50 kV, 1.75 mA, and 20 ipm.

The best setting for welding 0.008-in. D-36 to 0.008-in. D-36 by the burnthrough technique was 50 kV, 2.35 mA, and 20 ipm. By the overlap technique, the optimum setting is 50 kV, 1.65 mA, and 20 ipm with a 0.025-inch ac deflection of the beam.

In welding on a brazed honeycomb panel, the most common defect was pinholes in the weld. These occurred at points of contact between the honeycomb core and the skin, and are probably caused by the chilling effect of the core.

The most effective method found to eliminate pinholes was to weld with the overlap technique.

A square patch was the easiest configuration to weld in a restrained part.

TZM

The best setting for penetration of TZM, 0.008-in. thick, was 70 kV, 2.00 mA, and 20 ipm.

The optimum setting for welding 0.008-in. TZM to 0.008-in. TZM by the burnthrough technique was 70 kV, 3.00 mA, and 20 ipm. By the overlap technique, the optimum setting is 70 kV, 2.00 mA, and 20 ipm, with a 0.025-inch ac widening of the beam.

TZM could not be welded in a restrained configuration without preheating to 250°F and stress relieving after welding.

TZM could not be welded in a restrained configuration when Haynes 25 was present in the weld zone, regardless of preheat or stress relieving.

E. B. Bas, G. Cremosnik,* H. Lerch,*
O. Winkler,† and O. Gmeiner†*

A 30-KV ELECTRON-BEAM WELDING MACHINE WITH INDIRECTLY HEATED TUNGSTEN CATHODE

INTRODUCTION

An electron-beam welding machine must fulfill various requirements, one of which is very important in practice: it must be greatly versatile in order to allow the effective study of the process of electron-beam welding, which is still very changeable, in various fields of technology. It is necessary, e.g., to obtain the smallest possible diameter in the welding spot, and that with low beam powers as well as with high ones. Two beam parameters are known to be important for the realization of high-power densities in the welding spot: accelerating voltage and emission current density of the cathode [1], [2]; the former having a stronger influence than the latter. However, various factors oppose too great an increase in accelerating voltage; among others, decrease in operation reliability and increase in X-ray output are serious disadvantages. In the equipment described in this paper, accelerating voltage has been limited to 30 kV, when special protection measures against X-ray radiation are not yet necessary. On the other hand, by increasing emission current density, the power density in the beam could be increased sufficiently to allow plates of stainless steel, several centimeters in thickness, to be welded through. Moreover, the equipment is characterized by ease of operation and versatility in application.

*Department for Industrial Research, Swiss Federal Institute of Technology, Zurich, Switzerland.

†Balzers A. G. für Hochvakuumtechnik, Balzers, Principality of Liechtenstein.

GENERAL CONSTRUCTIONAL DESIGN OF THE EQUIPMENT

Figure 1 shows the general structure of the welding machine. The welding chamber A, the electron-beam system B, and the high-vacuum pumping system C have been built as one compact unit held on a heavy pedestal D. The welding chamber is therefore easily accessible from all sides and is also adjustable in its vertical position by changing the holder column. It has been given large dimensions and has two side flanges, 25 cm in diameter, for the installation of additional chambers on both sides for long welds. There is also an opening at the bottom, 39 cm in diameter, as well as a front door of the same dimensions. The electron-beam system B had been inserted deeply into the welding chamber through an opening at the top, in order to get it as close as possible to the welding object. The pumping system consists of a 1500 liters/sec oil diffusion pump and a 170 m³/h or 90 m³/h mechanical forepump.

The electron-beam system consists mainly of an electron gun 1, a separating valve 2, which is operated pneumatically, an optical device 3 for viewing the welding object, a magnetic focusing lens 4, and a magnetic deflection element 5. The whole system, including the mechanism for adjusting the magnetic lens, is built very compactly, allowing flanging onto the welding chamber in any desired position. The lower part of the beam system, opposite the object, is water cooled. Besides, compressed air is introduced into the system at the level of the deflection element and flows vertically around various parts; it reaches the massive cathode head 7 through channels in the walls of the electron gun before leaving the system through the openings in the high-voltage screen 8 near the cable connection 9. This ensures the effective cooling of all elements in the beam system. The cathode chamber of the electron gun and the welding chamber communicate through a channel, the gas flow conductivity of which is about 0.3 liter/sec.

The cathode chamber is evacuated separately by means of an oil diffusion pump (60 liters/sec) through a valve 10. This makes it possible for the pressure in the welding chamber to be 100 times greater than in the electron gun when the pump speed at the opening of the chamber is about 30 liters/sec. It has been our experience that a good vacuum in the electron gun improves beam concentration and prolongs cathode life, as is reported in another paper. The separating valve 2 allows a continuous evacuation of the electron gun, thus improving both vacuum and high-voltage stability.

The optical device for viewing allows the observation of the welding process at an angle of 4 degrees to the electron beam.

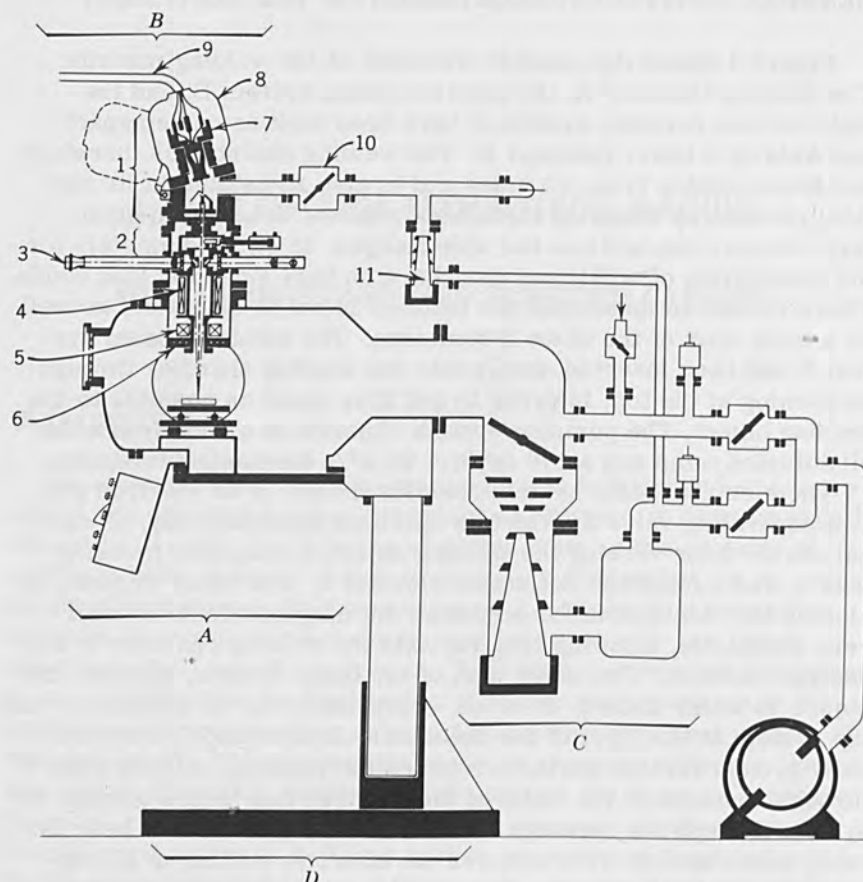


Fig. 1. Principal scheme of the new 30-kV electron-beam welding machine: *A* welding chamber, *B* electron-beam system, *C* high-vacuum pumping system, *D* holding pedestal. (1) Electron gun. (2) Separating valve. (3) Viewing ocular. (4) Magnetic focusing lens. (5) Magnetic deflection element. (6) Water-cooled bottom flange. (7) Cathode head. (8) High-voltage screen. (9) High-voltage cable. (10) High-vacuum valve. (11) Auxiliary diffusion pump.

It also permits the illumination of the object. The main problem about such devices, as is well known, is the evaporation onto the

viewing mirror; in order to solve it satisfactorily, we have built the whole system as an easily removable element, shown in Fig. 2. The whole system is mounted on a vacuum flange 1; the viewing mirror 2 can be adjusted during operation through a knob 3, with which the field of vision can be altered. There is a rotating glass plate 4 in front of the mirror as a protection against vapor condensation. It can be rotated by hand using the operating shaft 5, which can also be coupled to an electric motor for continuous rotation. In front of the glass plate, there is a fixed sector plate 6, with which one can regulate the width of the viewing window. Evaporation onto the protecting plate occurs behind that window only. A narrow opening is all that is needed in most cases. But it is recommended, if observation is not to be continuous, to adjust the plate in always the same position between observations, thus en-

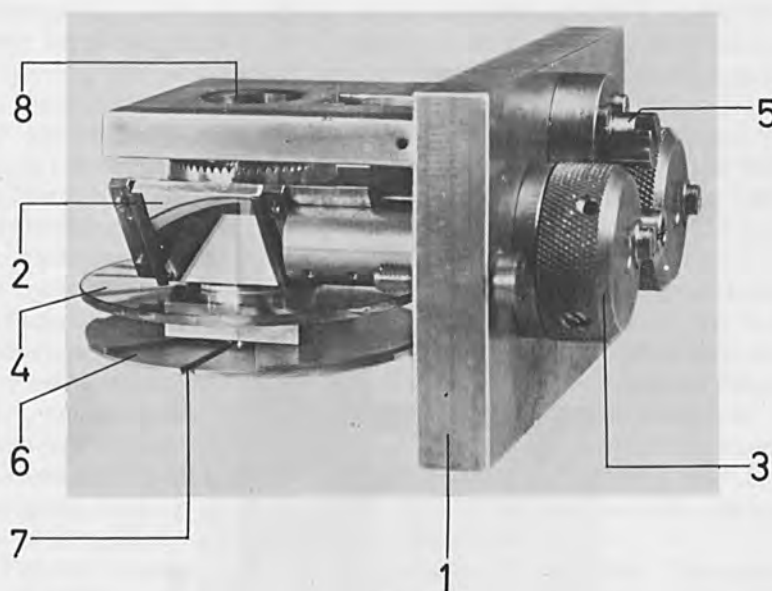


Fig. 2. Photograph of the viewing mirror device for the axial observation of the welding process. (1) Vacuum flange. (2) Mirror. (3) Knob for mirror adjustment. (4) Glass plate as screen against vapor condensation. (5) Drive for glass plate rotation. (6) Sector plate for the adjustment of the viewing window. (7) Viewing window. (8) Aperture diaphragm.

sureing that only a narrow sector of it will be coated by evaporation. If observation is to be continuous, then the plate must be rotated continuously also, at a speed which allows for a medium coating. Changing of the protecting plate is very simple. This optical device also carries the aperture diaphragm 8 which limits the aperture angle of the electron beam and which can be water cooled.

Figure 3 shows the general view of the electron-beam system. The magnetic lens, contained in the bottom container, can be centered during operation using the four screws 7. To achieve this, a relay circuit has been constructed to change over periodically the polarity of the lens excitation. Thus the centering can be controlled very quickly at any time; the electron beam itself can also be tilted during operation by displacing the anode, using the two screws 3 on the electron gun, and thus aligned onto the aperture diaphragm. The magnetic deflecting element is made of laminated iron foil, which allows an increase of deflection frequency up to

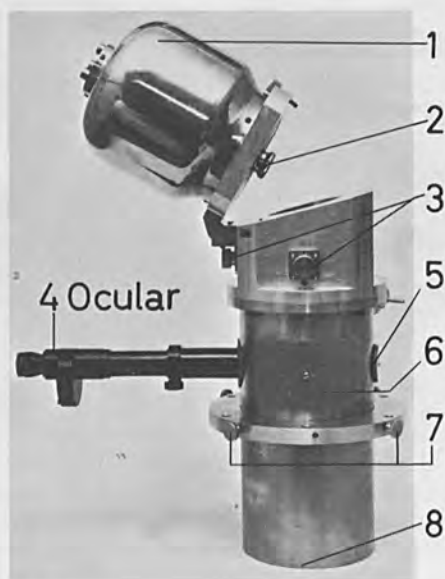


Fig. 3. Photograph of the whole electron-beam system. (1) High-voltage screen. (2) Cathode. (3) Anode adjustment. (5) Viewing ocular. (6) Place of the viewing mirror. (7) Screws for lens centering. (8) Water-cooled bottom flange.

100 C/sec. A further increase is limited by eddy currents in the metallic tube around the electron beam. This tube must be replaced by a tube of ceramics for higher deflection frequencies.

THE ELECTRON GUN

The electron gun is the most important part of the entire system described here. Its construction is characterized by two features: the ion separation in the accelerating chamber, and the special tungsten cathode heated by electron bombardment, which is called "bolt cathode" or *B*-cathode. Both these features have been described in previous publications [1], [3], [4], and will therefore be considered in no great detail here. Figure 4 shows a section through the electrode system of the electron gun. The anode makes a 60 degree angle with the axis of the electron-beam system, whereas the emitting surface of the cathode and the Wehnelt cylinder form an angle of 46.5 degrees with the anode. The electrons emitted by the cathode fly into the system axis on a curved path, and the positive ions generated on this electron path move on similar curved paths and cannot reach the cathode, but get to the Wehnelt cylinder. The emitting surface is protected against the bombardment by positive ions, and there is no ion crater in the cathode and therefore no disturbance in the focusing of the beam [1], [4]. The cathode consists of a massive tungsten rod, called bolt; the one front surface of which is plane polished and used as emitting surface, whereas the other end, which is narrower, is fixed in a holder. A tungsten spiral is wound around this tungsten rod, at a very small distance (0.6 to 0.25 mm, according to cathode diameter), and is heated by direct current; it then emits electrons which are accelerated with about 200 V onto the bolt. This electron bombardment allows the heating of the bolt to very high temperatures with optimal operating reliability, and permits realization of very high emission current densities.

Figure 5 shows the practical design of *B*-cathodes. The cathode cartridge with the conical-shaped Wehnelt diaphragm can be seen at the very top. It can be inserted easily into a socket of the electron gun. This simplifies cathode changing very much, because the cathode has been previously centered in the cartridge with the Wehnelt diaphragm and placed in the exact position in relation to the latter. The cathode structure as such can be seen in the center of Fig. 5; the holder pins for the tungsten spiral and the central holder for the bolt are soldered into a cylindrical plate made of

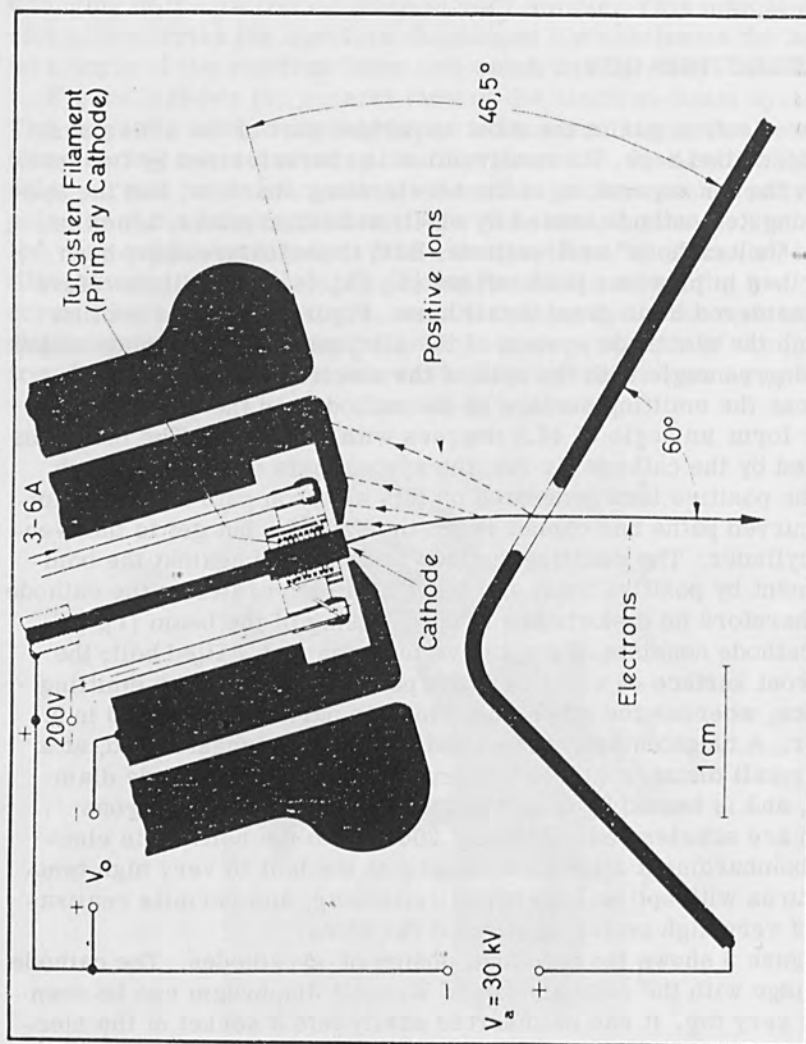


Fig. 4. Structure of the electron gun with ion separation.

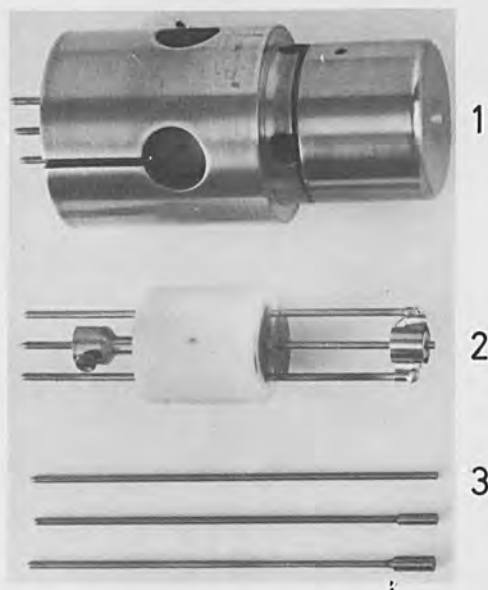


Fig. 5. Practical realization of the *B*-cathode. (1) Cathode cartridge. (2) Cathode system. (3) Cathodes of 1-, 1.5-, and 2-mm diameter.

ceramics. The changing of the cathode body (the bolt) is effected by loosening the screw at the rear end of the central holder. It is not necessary to take the cathode system out of the cartridge for this exchange.

At the bottom of Fig. 5, one can see the tungsten bolts with an emitting surface diameter of 1, 1.5, and 2 mm. For emitting surfaces with smaller diameters than 1 mm, a 1-mm bolt is used and ground at the front end to the desired diameter over a length of about 0.8 mm. Since it is very easy to change the cathode, it is possible to use in each case the emitting surface diameter corresponding to the desired electron-beam power, and thus to operate the cathode with maximum current density; in this way, the optimum focal point diameter can always be achieved.

In Fig. 6, the bombardment power N_B is shown in function of the emitting current I_e for cathode diameters of 1, 1.5, and 2 mm. This graph shows some characteristic properties. In the case of a 1-mm cathode, the heating power begins to increase steeply at a beam current of about 100 mA. This is the beginning of the

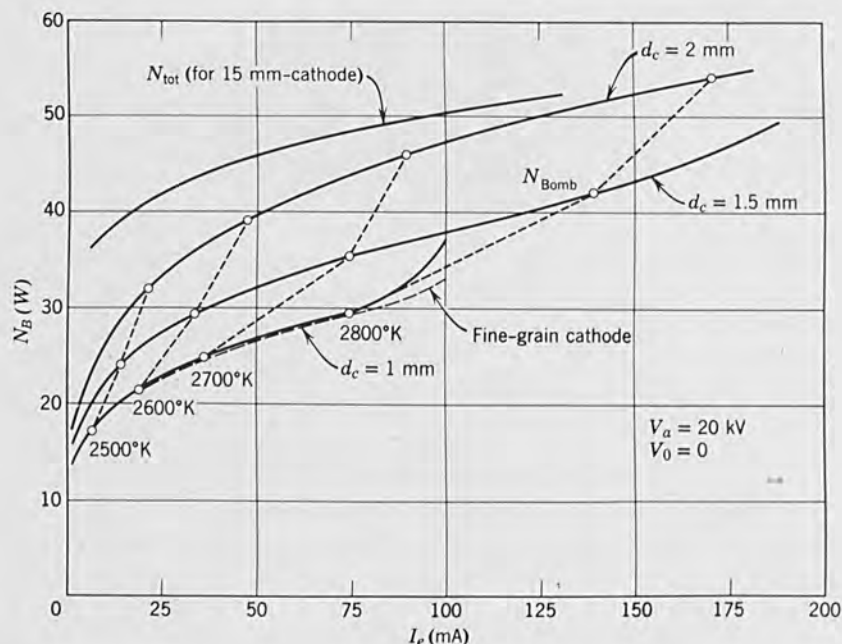


Fig. 6. Bombarding power N_B as a function of the emission current I_e at an acceleration voltage of 20 kV for B-cathodes of $d_c = 1, 1.5$, and 2 mm. For 1.5-mm cathode the total heating power N_{tot} is also given.

space-charge region; at the same time the aperture angle of the electron beam is greatly increased. The cathode can only be used up to that value of the beam current. In the case of a 1.5-mm cathode, space charge is first effective at about 200 mA. These data are valid for an accelerating voltage of 20 kV, since no other rectifier was available at the time the measurements were made. At higher voltages, the limit of the space charge moves towards higher current values.

Another factor seems to have an influence on this limit: it is the crystallographic structure of the cathode. The dotted line on Fig. 6 refers to a 1-mm cathode made of very finely crystallized tungsten. Whereas the curve coincides almost entirely with that for coarsely crystallized tungsten for low-current values (i.e., the emissivity is about the same on average), the space charge can first be noticed at higher beam currents in the case of finely crystallized tungsten. Cathode temperatures are also given for a few points on the diagram in Fig. 6. The beam currents of 1-mm and

1.5-mm cathodes for the same cathode temperatures are in a ratio of about 1:2, whereas the ratio of emitting surfaces is 1:2.25. Two reasons can be given for the variation in emitting efficiency: inhomogeneous electric field strength in front of the cathode, and variations in crystallographic structure. How strongly cathode emission can depend on crystallographic structure is shown very clearly in the heating characteristics for the 2-mm cathode, given in Fig. 6. This 2-mm cathode was chosen because of its extremely bad emission. One can see that beam currents of about the same values as in the case of the 1.5-mm cathode can be achieved for the same cathode temperatures, although the emitting surface is about 80% larger.

In Fig. 7, photographs of the current density distribution in the emitting surface, taken with an emission electron microscope, explain these great differences. In the case of the 2-mm cathode, a large grain with a bad crystallographic orientation (i.e., high work function) fills almost the entire emitting surface. The current densities measured in the emission microscope at lower temperatures (circa 2000°K) in the large grain and in a strongly emitting small grain are in a ratio of 1:2.5. At higher temperatures, this ratio is still more unfavorable. As we can see in Fig. 7, grain distribution of the 1.5-mm cathode is more favorable; the current densities in various grains vary at a ratio of 1:3. The most favorable case is that of a cathode made of finely crystallized tungsten, and the 1-mm cathode shown in Fig. 7 is an example of that. In this case too, variations of current densities occur at about 1:5. But grains of different crystallographic orientation are distributed much more evenly. The choice of tungsten quality also influences the emission distribution of the cathode during its operation, due to recrystallization.

The recrystallization process in normal operation is shown in Figs. 8 and 9, in the case of two 1-mm cathodes of different tungsten quality. The cathode shown in Fig. 8 was made of pure tungsten and was heated during operation to 2900°K, whereas the electron micrographs after 0 sec, 10 sec, 1 h, and 10 h, were made at a temperature of about 2000°K. An important alteration in the emission micrograph can be noticed. Contrary to that, the electron micrographs in Fig. 9 show, in the case of a 1-mm cathode made of doped tungsten, a much smaller alteration in the emission micrograph due to recrystallization. All these emission micrographs show clearly that electrons are only emitted from a certain number of emission patches, even in metallic cathodes. Only a fraction of the emitting surface will actually be contributing to the electron beam.

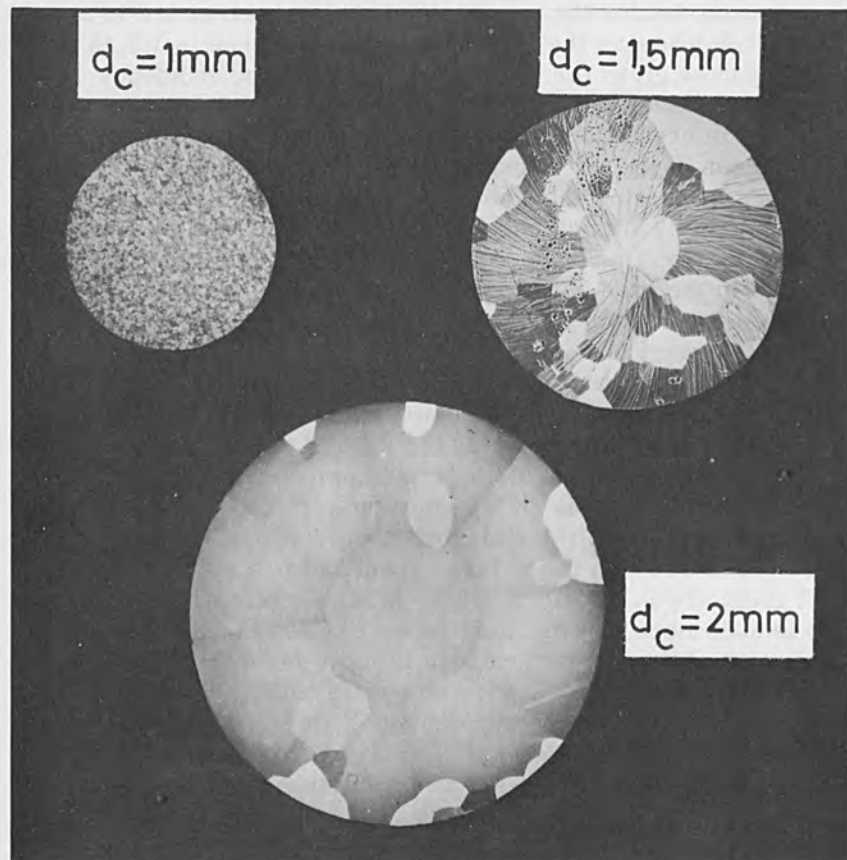


Fig. 7. Emission-electron micrographs of three *B*-cathodes corresponding to Fig. 6. (The micrographs of 1.5- and 2-mm cathodes are prepared from several single exposures because the viewing field in our emission microscope has a diameter of about 1 mm.)

This inhomogeneous emission decreases not only the yield of the cathode, it also influences the properties of the electron beam generated in the electron gun. It would be of great advantage to use single-crystal cathodes with crystallographic orientation which would be favorable for electron emission (e.g., 111-plane as emission surface), but there would be practical difficulties to overcome. Work is in progress in this direction.

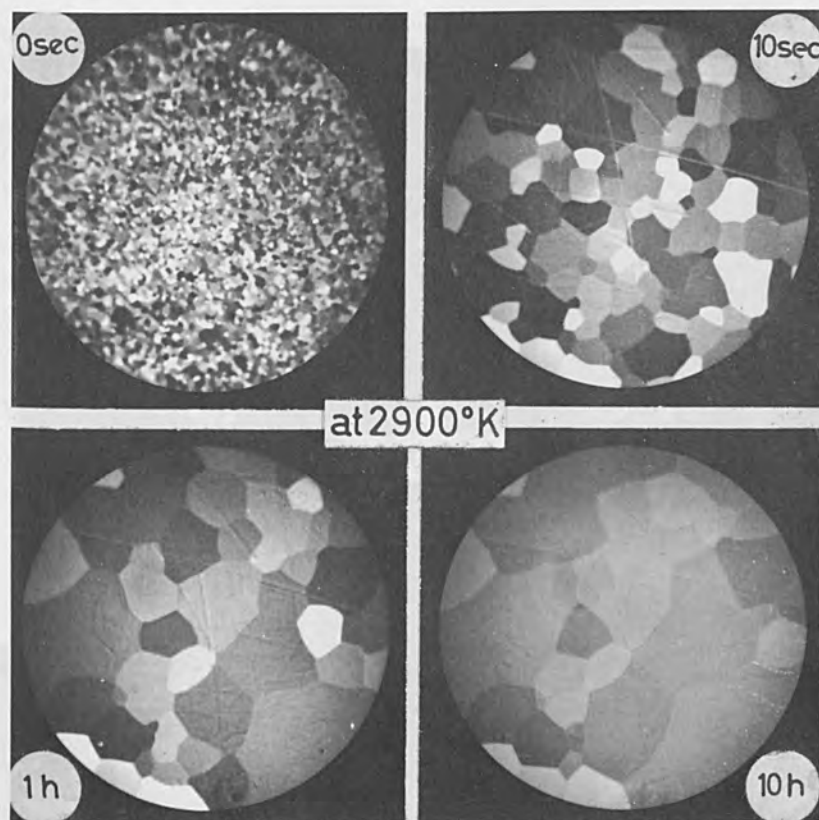


Fig. 8. Recrystallization of a 1-mm cathode made from pure tungsten. The cathode has been operated at 2900°K and the micrographs have been taken at about 2000°K after an operation time of 0 sec, 10 sec, 1 h and 10 h. The micrographs show the whole emission area of 1-mm diameter.

LIFETIME OF THE *B*-CATHODES

The lifetime of a pure metal cathode should depend solely on the rate of evaporation of the cathode material, i.e., on a material constant which would be influenced only by the temperature of operation. But experience has shown in various cases that this is

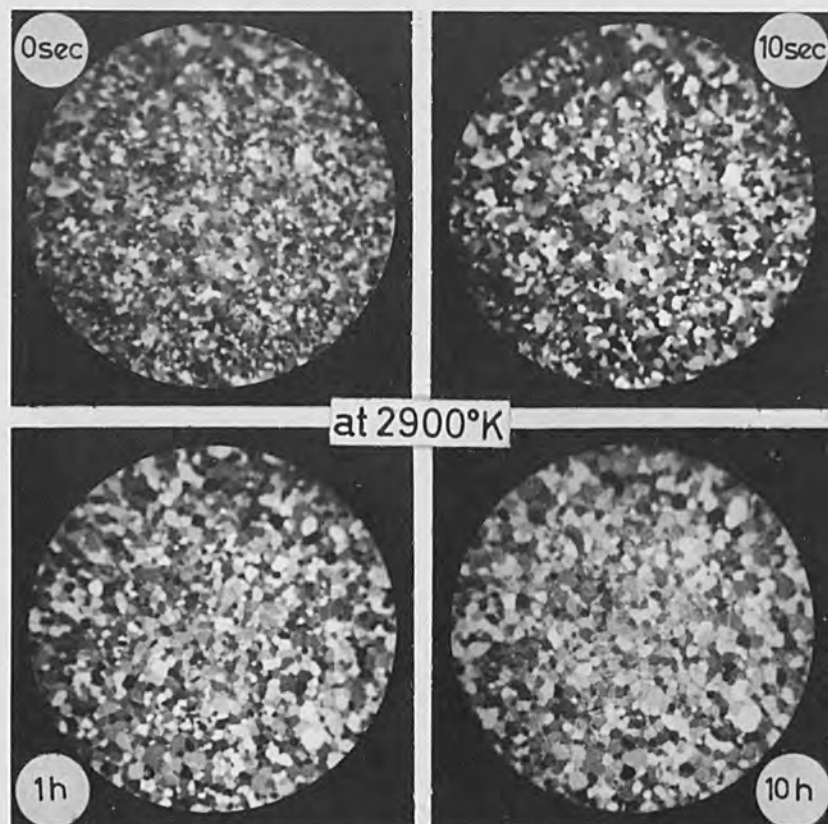


Fig. 9. Recrystallization of a 1-mm cathode made of doped tungsten to affect the grain growth. Data as in Fig. 8.

not the case for cathodes operated in dynamic vacuum systems [5]. The apparent rate of evaporation in such systems is much greater. As early as forty years ago, Langmuir [6] gave the reaction of tungsten with water vapor molecules remaining in the system as the reason for this accelerated material removal. Many mass-spectroscopy examinations have shown that in the dynamic, not bakeable vacuum systems the partial pressure of water vapor represents the main part of the remaining gas pressure [7].

Data about cathode lifetime experiments will always vary with the conditions of vacuum. The experiments reported here were made in recipients at a pressure of about 2×10^{-5} torr, as meas-

ured with an ionization gauge. The results should be regarded only as characteristic values. In the electron guns of the welding machine we have endeavored to obtain better vacua, and the separating valve in the beam path mentioned above was very useful for that purpose, since it avoids the necessity of flooding the electron gun when changing the welding object.

Two elements of a *B*-cathode play a part in its lifetime: the cathode as such, i.e., the "bolt," and the tungsten spiral. In the bolt, temperature reaches its highest values in the middle of the bombardment zone, and the cathode is destroyed there at the fastest rate. Figure 10 shows a 1.5-mm cathode after 40 hours operation at an emission area temperature of about 2900°K. The rate of material removal in the bombarding zone for 1-, 1.5-,

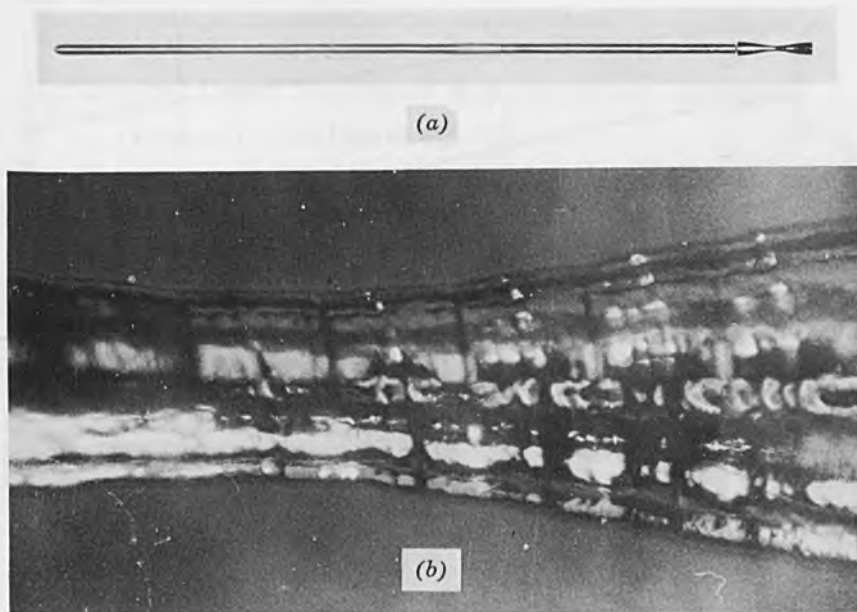


Fig. 10. (a) Photograph of an 1.5-mm cathode after an operation time of 40 hours at a temperature of the emission surface of 2900°K and a vacuum of about 2×10^{-5} torr. (b) Enlarged photograph of the central part of the bombarding zone showing projected traces of the filament.

and 2-mm cathodes can be seen in the graph in Fig. 11, where all experiments have been conducted at a temperature of the emission area of 2900°K , which represents extreme working conditions (cf. Fig. 6). The lifetime of a cathode is increased by a factor 5 if operation temperature is reduced to 2800°K . The material transport occurs more slowly in cathodes with larger diameter than in cathodes with smaller diameter, since the difference in temperature between the emitting surface and the bombardment zone is smaller. The decrease in emitting surface diameter at 2900°K is about 5×10^{-3} mm/h. But from the well-known evaporation data of tungsten [9], one calculates a diameter decrease of about 1.3×10^{-3} mm/h; that means the material removal in our experiments is about four times faster as would be expected from evaporation alone.

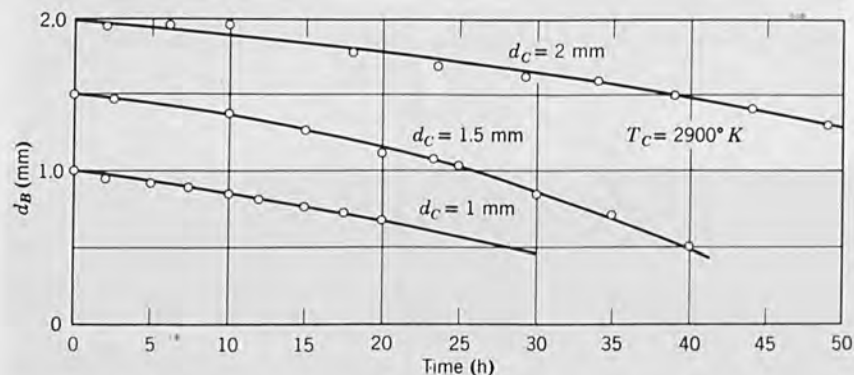


Fig. 11. Reduction of the cathode diameter in the bombarding zone as a function of the operation time for 1-, 1.5-, and 2-mm cathodes. The temperature of the emission surfaces has been held at 2900°K . The cathodes have been operated in a special vacuum chamber at about 2×10^{-5} torr.

The B -cathode works reliably even if the diameter in the bombarding zone has been reduced to less than 0.5 mm. For the 0.5-mm limit, we get from Fig. 11 the following lifetime at 2900°K : about 28 hrs for a 1-mm cathode, 40 hrs for a 1.5-mm cathode, and about 85 hrs for a 2-mm cathode. At higher vacuum we got much longer lifetimes.

The life of the heating spiral is not limited by material removal, but by material deposition. The tungsten atoms evaporating from the cathode condense on the spiral and increase thereby the cross

section of the spiral wire. Figure 12 shows the photograph of the cross section through the tungsten spiral wire after long operation. The wire grows towards the cathode. In Fig. 13, we see the change of spiral resistance R_H during the course of a lifetime experiment. The experiment was conducted with 1-mm cathodes, one of which was kept at an operating temperature of 2900°K during 10 hours before being replaced by a new one. The decrease in diameter in the bombarding zone of each bolt was about 0.15 mm per 10 hours operation, as shown in Fig. 13. After the tenth change, it was not possible to work reliably any longer, since the spiral wire had "grown" too much. During this experiment, the distance between the cathode and the spiral was 0.35 mm, but in later experiments that distance could be increased to 0.75 mm. At lower cathode temperatures the life of the spiral can amount to several hundred, even several thousand hours.

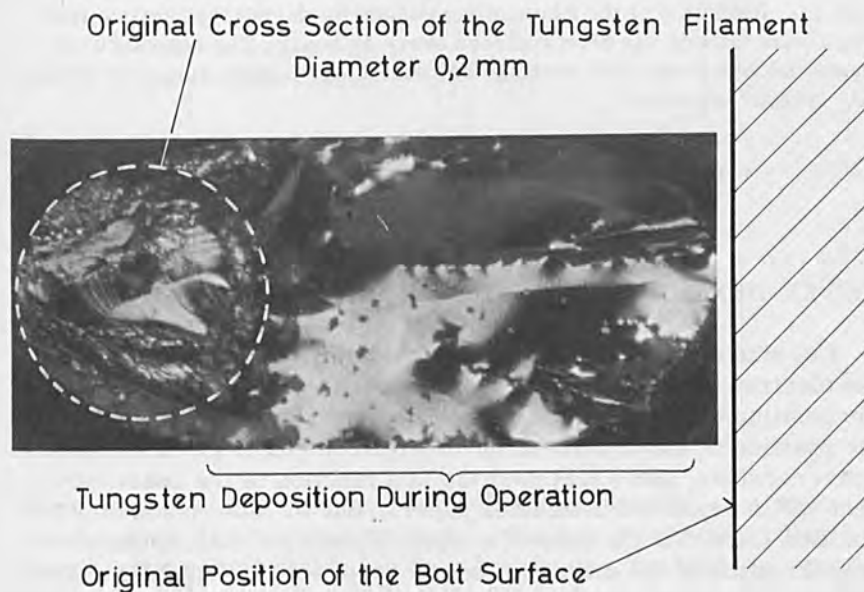


Fig. 12. Fracture cross section of filament wire (tungsten spiral) after a long operation. The dotted circle shows the cross section of the original 0.2-mm wire, the remaining part is the result of tungsten deposition during the operation of the cathode.

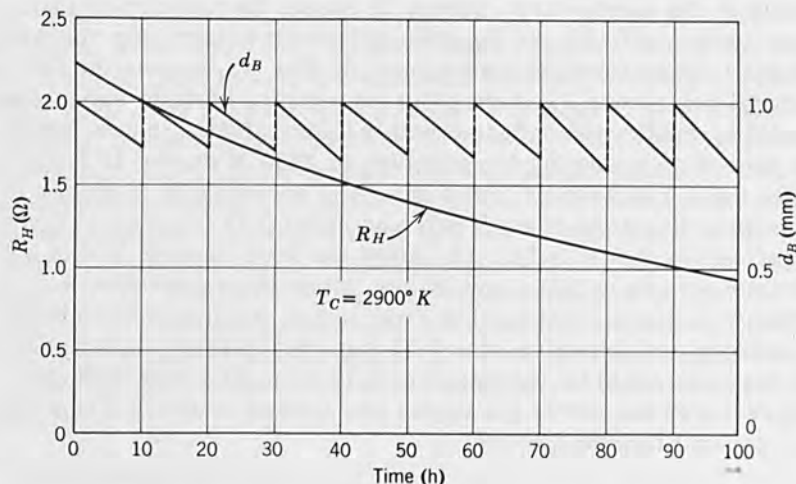


Fig. 13. Reduction of the filament resistance R_H during the lifetime test. The 1-mm cathode has been replaced every 10 hours. The second curve shows the reduction of the cathode diameter in the bombarding zone during this 10-hour operation.

POWER DENSITY IN THE WELDING SPOT

The size of the focal spot depends not only on the properties of the electron gun, but also on the dimensions of the beam path and the position of the magnetic lens. They are given in Fig. 14, where the position of the crossover of the electron gun is given for small beam currents, since that position is a function of the beam current (as is explained in another paper). But we shall consider this position fixed, for the sake of simplicity, and we shall speak of magnification of the magnetic lens in respect to this position. The minimum magnification is about 0.5 (which amounts to a demagnification), since in that case the welding spot comes to lie in the plane of the end-flange of the electron system. In the following, we shall refer to $M = 1$ as standard magnification. Current density distributions were measured at $M = 1.2$ and recalculated accordingly for $M = 1$. The welding spot diameter resulting from these measurements is shown in function of the beam current in Fig. 15,

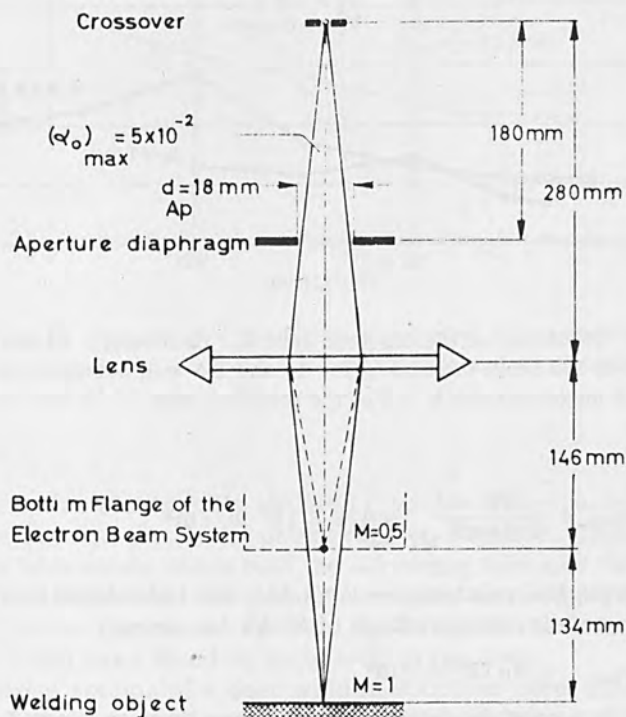


Fig. 14. Geometry of rays in the electron-beam system.

for 1- and 1.5-mm cathodes. We defined the welding spot diameter $d_{1/2}$ as the half-height width of the current density distribution. The values of Fig. 15 do not represent the smallest values obtainable; changing the position of the cathode inside the Wehnelt diaphragm (Parameter z_c) can bring about a smallest welding spot diameter of $d_{1/2} = 0.2$ mm for a 1-mm cathode at a beam current of 100 mA. Since these measurements were made at an acceleration voltage of 20 kV, we get for maximum power density in the welding focus:

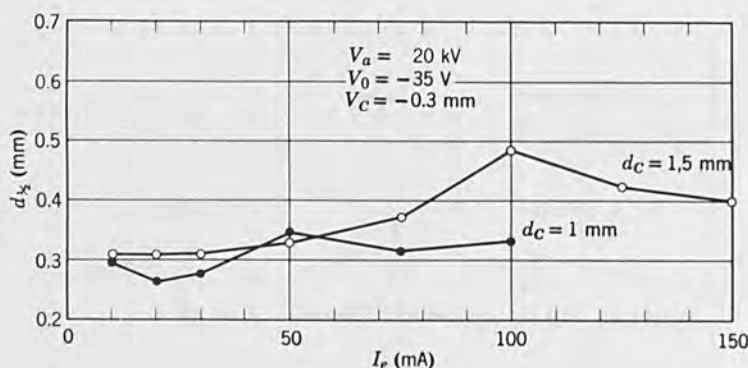


Fig. 15. Diameter of the electron spot $d_{1/2}$ (half-height width) as a function of the beam current I_e for 1- and 1.5-mm cathodes, at a standard magnification $M = 1$ of the focusing lens.

$$n_{\max} = \frac{VI}{1.44 \pi d_{1/2}^2} \approx 4.4 \times 10^6 \text{ w/cm}^2$$

If one accepts the relation $n \sim V^{7/4}$ [1], the calculated power density for an accelerating voltage of 30 kV becomes:

$$n_{\max} \approx 9 \cdot 10^6 \text{ w/cm}^2$$

But these data must be accepted with some caution since both measurements and calculations were based on a Gaussian current density distribution in the beam spot.

It has been mentioned earlier that the diameter of the emitting surface should preferably be decreased when operating with smaller beam currents. Definitely smaller focal spot dimensions can be achieved by increasing the negative Wehnelt voltage, as has been done, on Fig. 16, for an emission surface diameter of 0.6 mm and a beam current of 10 mA. The preparation of a wholly screened thermocouple, shown on Fig. 17, shall be an example of operation with small current and very small welding spot. In a Philips "Thermo-coax" thermocouple, 1 mm in outer diameter, the two thermocouple wires (diameter 0.1 mm) are welded together inside the stainless screening tube (Fig. 17a). The inner space around the thermocouple weld was then filled with MgO_2 and the end of the screening tube bent with a special tool. A cap of stainless steel was put on the tube end and welded by electron beam (Fig. 17b). In this way one can get the screening tube gas-tight

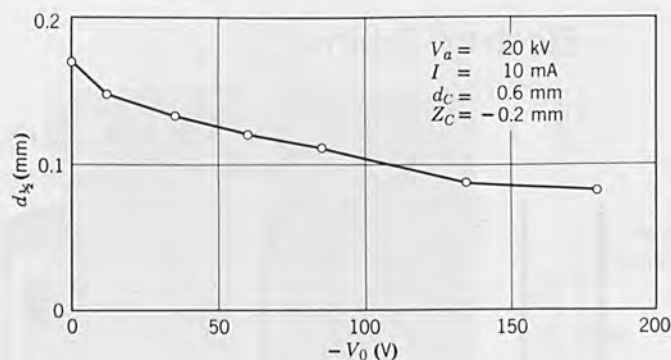


Fig. 16. Influence of the Wehnelt-potential on the electron beam diameter $d_{1/2}$ for a 0.6-mm cathode and an emission current of 10 mA.

sealed and also electrically isolated from the thermocouple wires, as can be seen on the X-ray photograph on Fig. 17c. There is no danger of corrosion, since both the screening tube and the cap are made of stainless steel. Such thermocouples were made in large numbers, about four years ago, for the European nuclear reactor "Dragon," and have stood up quite well to the test.

The cross section of a deep weld in stainless steel plates, shown in Fig. 18, is an example of operation with high beam power. These 30-mm thick plates (of stainless 304) were welded at a speed of 16 cm/min, using a 1.5-mm cathode and $M=1.2$. The beam current was about 220 mA and the accelerating voltage about 27 kV. Figure 19 shows another example, in which beam current density was so much higher that the cutting effect occurred, yielding a very sharp slot of 0.8-mm width, which is a measure for the beam spot diameter.

THE HIGH-VACUUM INSTALLATION AND THE ELECTRICAL CONTROL SYSTEM

The design of the high-vacuum installation has already been mentioned in connection with the general structure of the welding unit (Fig. 1). We have seen that it consists of two pumping systems, one for the electron gun and one for the welding chamber. Both systems must be operated simultaneously during some oper-

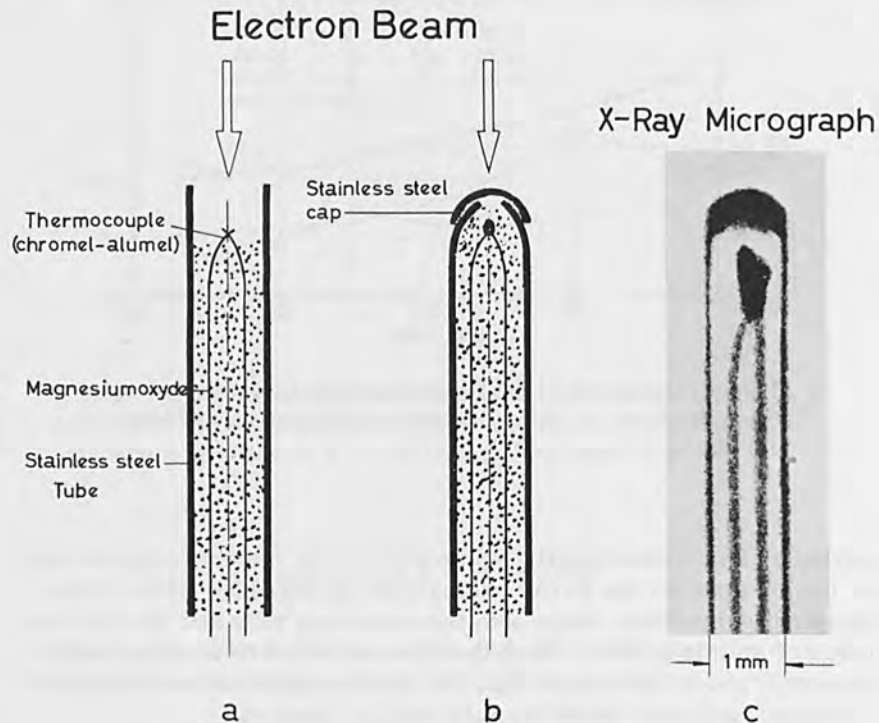


Fig. 17. Preparation of a screened thermocouple with a small electron-beam probe and low-beam current.

ations like the changing of the welding object or of the cathode. It is necessary to automatize the entire vacuum system in order to unburden the operator, and the latching between the high-voltage generator and the high-vacuum system can be achieved consequently. Automation, as applied to our equipment, allows the changing of the welding object or of the cathode by operating a single pushbutton and a two-position switch with which it can be decided whether the welding object or the cathode is to be changed. If the switch is in the position for changing the welding object, operating the pushbutton will execute automatically the following operations: the high voltage is switched off, the separating valve in the electron-beam system and the high-vacuum valve in the large diffusion pump are closed, the welding chamber is flooded. After changing the welding object, operating the pushbutton again will be followed

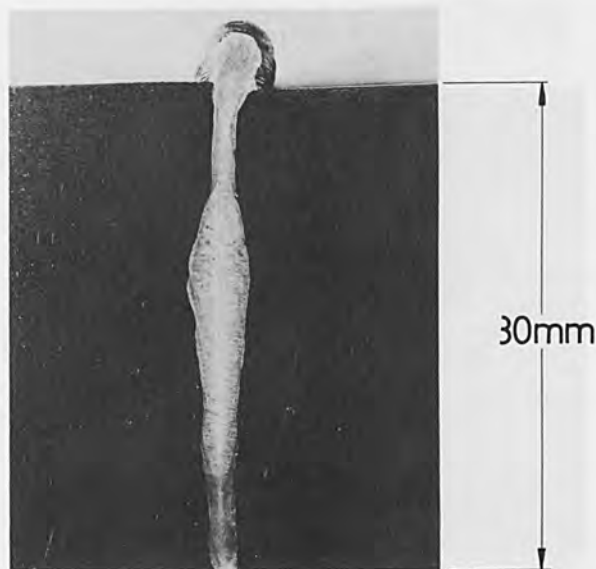


Fig. 18. Deep welding of stainless 304 plates with high-beam current: welding speed 16 cm/min, acceleration voltage 26.5 kV, beam current 220 mA.

by the following operations: the pressure is decreased, by means of the forepump through the bypass valve, until it reaches a value of 10^{-1} torr, the high-vacuum pump is then brought into operation, and the separating valve in the electron-beam system is opened. The switching on of the high voltage is only possible when vacuum has reached 8×10^{-5} torr in the electron gun. If the two-position switch is in the position for changing of the cathode, then operating the pushbutton will launch the same series of automatic steps as above, except that instead of the separating valve (which in this case remains open throughout the operation), the high-vacuum valve in the small diffusion pump is opened and closed (Fig. 1). Control of the equipment in this way guarantees reliable operation under all conditions. By the logic of this system, the operational steps are well defined, and facilitate the work of the operator, while eliminating unnecessary waiting time. It is thus possible, by operating a single pushbutton, to evacuate the welding chamber, which has a capacity of about 120 liters, in 47 seconds to a vacuum of 8×10^{-5} torr.

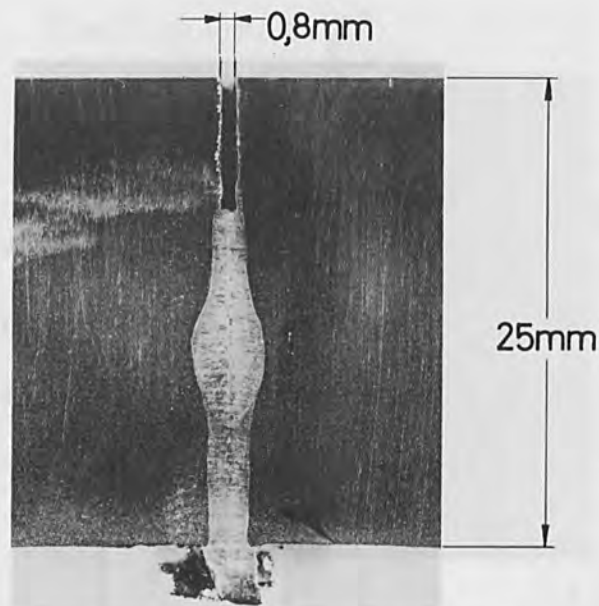


Fig. 19. Beginning of the cutting effect at high-power densities; speed 16 cm/min, acceleration voltage 27 kV, beam current 220 mA.

This high voltage is generated by a selenium rectifier which is protected against current overload by a current limiting transducer at the input. The high tension is well filtered and is kept constant by a servoregulating device. In order to eliminate the effects of the rapid voltage variations on the focusing of the electron beam, the lens current and the deflecting currents are electronically regulated proportionally to $\sqrt{V_a}$. A 10-turn potentiometer permits an exact adjustment of the focusing current in the magnetic lens; this adjustment can be made beforehand for any distance between welding object and electron-beam system, and for any needed beam current, using a calibration diagram.

The high-voltage rectifier and the circuitry for cathode heating are placed in a cupboard (see Fig. 20); the electronics for the lenses and deflecting currents, and for the pumping system and the control circuits for the welding object drive are in a second cupboard. The operation desk is separated and attached to the welding chamber. Figure 20 shows the complete equipment.

An important contribution to the realization of the equipment

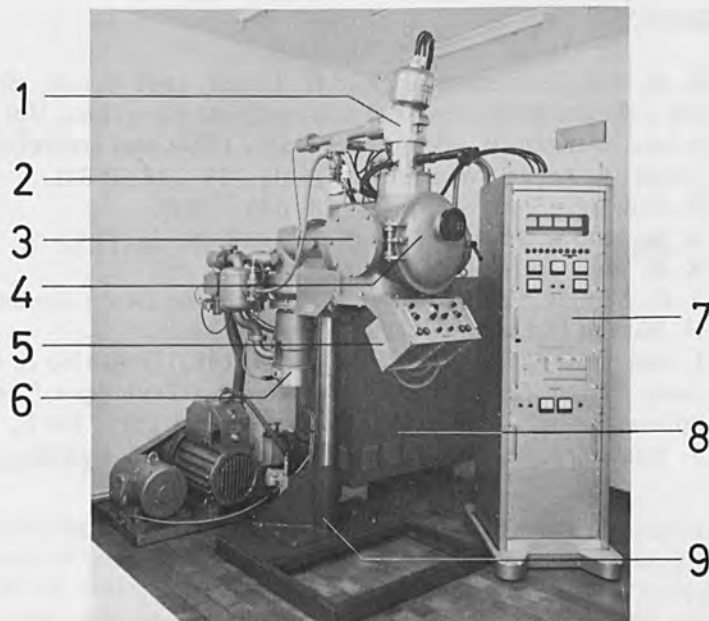


Fig. 20. Overall view of the welding equipment. (1) Electron-beam system. (2) Oil-diffusion pump for the electron gun chamber. (3) Welding chamber. (4) Large front door, with viewing window. (5) Control desk. (6) Main high-vacuum system. (7) Cupboard for electronics. (8) Cupboard for high voltage. (9) Pedestal with holder column.

described in this paper has been made by Mr. Bernhard Wihler, for the constructional design of the electron beam system, and Mr. Alois Meier, for the careful mounting and the manufacture of the cathodes, and we should like to thank them here. Our thanks also go to Professor E. Baumann, director of the Department for Industrial Research at the ETH, and to Professor M. Auwärter, Executive President of the firm "Balzers AG. für Hochvakuumtechnik und dünne Schichten," for their permission to publish this paper. We wish to express our thanks to Mr. J. B. Ansermoz for his assistance in the translation into English.

REFERENCES

1. E. B. Bas, G. Cremosnik, and H. Lerch, 1961 Trans. of the 8th Vacuum Symp. and 2nd International Congress, Vol. 2, p. 817, Pergamon Press, New York, 1962; and Schweiz. Archiv f. Angew. Wissensch. Techn. 28, 112 (1962).
2. H. Schwarz, Rev. Sci. Instr. 33, 688 (1962).
3. E. B. Bas, Z. Angew. Phys. 7, 337 (1955).
4. E. B. Bas, Optik 12, 377 (1955).
5. J. R. Morley, Proc. 3rd Symp. on Electron Beam Technology, R. Bakish (Ed.), p. 26 (1961). Boston, Mass.
6. I. Langmuir, Trans. AIEE 32, 1893 (1913); see also S. Dushman, "Scientific Foundations of Vacuum Technique," second edition, p. 647, John Wiley and Sons, New York, 1962.
7. J. Blears, J. Sci. Instr. Supplement No. 1, p. 36, 1951.

A SYSTEM TO VIEW ELECTRON-BEAM WELDING BY CLOSED-CIRCUIT TV

INTRODUCTION

In joining materials by electron-beam techniques, critical adjustment of beam power, focus, and position are necessary to produce welds with particular characteristics. At present these parameters can only be controlled by visual inspection of the welding zone. As systems become larger this becomes increasingly more difficult to do, because direct inspection of the weld is complicated by glass view ports and filters which separate the viewer from the weld by considerable distances.

One approach to viewing a weld in sufficient detail is the use of a telescope focused on the weld zone but located outside the chamber. This permits magnified inspection of the molten pool and has been used with great success for fixed electron-beam systems. This method becomes impractical, however, for a large system with an electron-beam gun driven in more than one axis.

Another technique which has become feasible only within the last few years is a system employing fiber optics for viewing the welding process. This can allow the target of a moving electron-beam gun to be continuously viewed outside the system. A complex fiber handling mechanism could be employed to follow the welding gun in as many as three axes of motion. The size of the welding system would be limited, since the length of the optical fiber would be several times longer than the length of the chamber. Furthermore, the high temperatures encountered in the welding process would require special treatment of the fiber to prevent

*Alloyd General Corporation, Cambridge, Mass.

it from overheating. This technique might also suffer from either a narrow field of view or a poor resolution.

A third approach is the use of a closed-circuit television system. This is the most versatile technique for remotely viewing the molten target of a moving electron-beam gun. Although costs and electronic complexity are generally greater for this scheme than with the other approaches, the results are far superior for large, complex welding applications. The television camera can be mounted on the electron-beam gun carriage to move with the target. A short optical system can be used to allow the camera to view the welding zone directly from above the workpiece. The television cable can be handled by the same mechanism which carries the high-voltage cable to the gun. The television monitor can be conveniently located near the welding controls. Although the dynamic range of a TV system is considerably less than that of the human eye, an acceptable picture can be presented on the TV monitor by illuminating the material with a light of narrow bandwidth and filtering the input to the camera with a narrow-band filter. This produces an extremely sharp visual display on the monitor from which fine adjustment and control of the electron-beam focus, power, and position can be made during the actual welding process.

SYSTEM DESCRIPTION

A closed-circuit television system has been employed in an automatic electron-beam welding system designed for making large rocket engine weldments. A block diagram of the television system is shown in Fig. 1. The electron-beam gun is mounted on a carriage which moves throughout the welding chamber with three degrees of freedom. The gun produces a high-density beam of high-energy electrons which is focused on the welding target by the focus coil. The beam passes through a hole in a mirror which is placed between the anode of the gun and focus coil at an angle of 45 degrees. The television camera is mounted above the electron gun and is shielded from the high-temperature radiation. The light emitted from the molten target is reflected to the television camera through a system of mirrors which optically align the axis of the camera with the axis of the electron beam. A light source producing a discrete spectrum is focused on the target material to illuminate the background. The light from both the welding zone and the base material returns to the camera through

the system of mirrors parallel to the incident electron beam. A narrow bandwidth filter passing one of the wavelengths emitted by the light source is placed between the lens and the vidicon tube in the camera permitting only a specific wavelength of light to be televised. The television cable is carried to the outside of the chamber with the same mechanism that carries the high-voltage cable to the electron-beam gun. The television monitor is located in the center of the welding console for convenient inspection and control of the welding process. The picture which is displayed on the monitor has the fine resolution, high magnification, and low contrast necessary to see the details of both the molten zone and base material. The field of view enables the gun position to be accurately and smoothly controlled during the welding cycle.

There were two major problems encountered in the design of the television viewing system. Since a 30 kV electron beam has an inherently short focal length for good weld penetration, it is necessary for the camera to view the target directly from above. This required the design of an optical system whose axis was parallel to the welding gun axis but would not interfere with the electron beam during the welding process. A second problem was the extremely high brilliance of the electron bombarded spot. The contrast in light between the welding zone and base material is typically several million at high temperatures. Since a television system has a maximum contrast range of two or three hundred, a sophisticated filtering system was required to simultaneously view both the welding zone and base material. The third problem was the deposition of the evaporating material on to the mirror between the anode of the gun and forms coil. These problems form the basis for the following discussion.

THEORETICAL DISCUSSION

The optical system projects the image of the electron-beam welding process into the television camera. It is presented symmetrically around the optical axis in Fig. 2. Any point of the target is described as a radius, R , about the axis.

The aperture in the focus coil has a radius R_2 of 1/2 in., and is limited by the optimum design of the focus coil for electron-beam welding. The aperture in the first mirror has a radius R_3 of 3/16 in., so that the mirror will not interfere with the electron beam as it passes through. The third mirror in front of the light source has a radius R_4 of 5/8 in., so that it will not severely re-

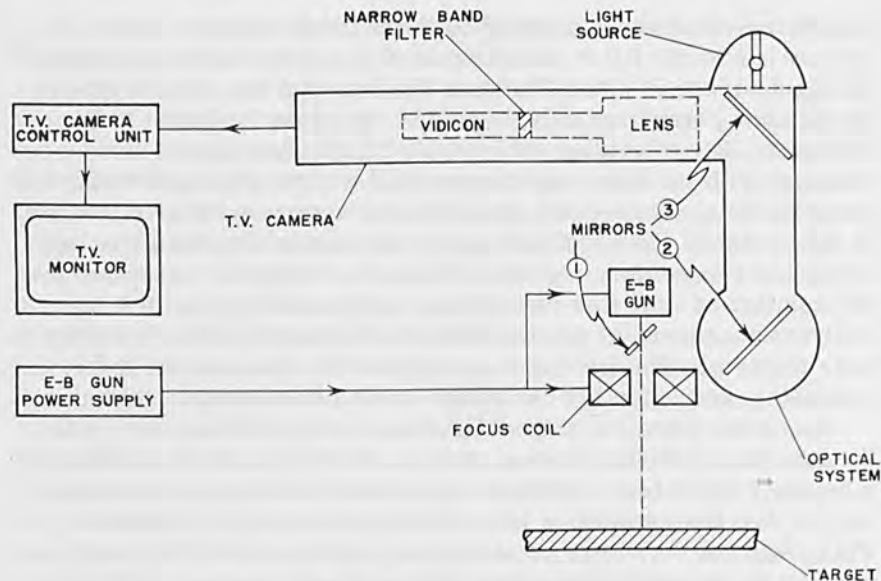


Fig. 1. System block diagram.

strict the light output from the lamp but will project all of the light from the target. A 6-in. focal length lens was used in the camera to provide an optimum magnification of the target. This lens has a radius R_5 of $3/4$ in. when opened to $f2.8$. The average radius of the vidicon image R_6 is approximately $1/4$ in.

The distance, L_1 , between the bottom edge of the focus coil and the top of the target is approximately 6 in. for good electron-beam weld penetration. The optimum distance between the top edge of the focus coil and the bottom of the electron gun is about 2 in. Since the focus coil used in the system was 1 in., the distance, L_2 , of the electron-beam aperture in the 45-degree mirror is 8 in. from the target. The 45-degree mirror in front of the light source is located a distance L_3 of 20 in. away from the target. The television camera in this system is mounted on the top of the electron gun so that the distance, L_4 , between the camera lens and target is approximately 24 in. The distance between the lens and vidicon tube in the camera is 8 in. in this case so that L_5 is about 32 in. With these parameters the magnification of the optical system is $1/3$; i.e., an object 1-in. wide at the target produces an image $1/3$ -in. wide in the vidicon. The magnification from the vidicon to

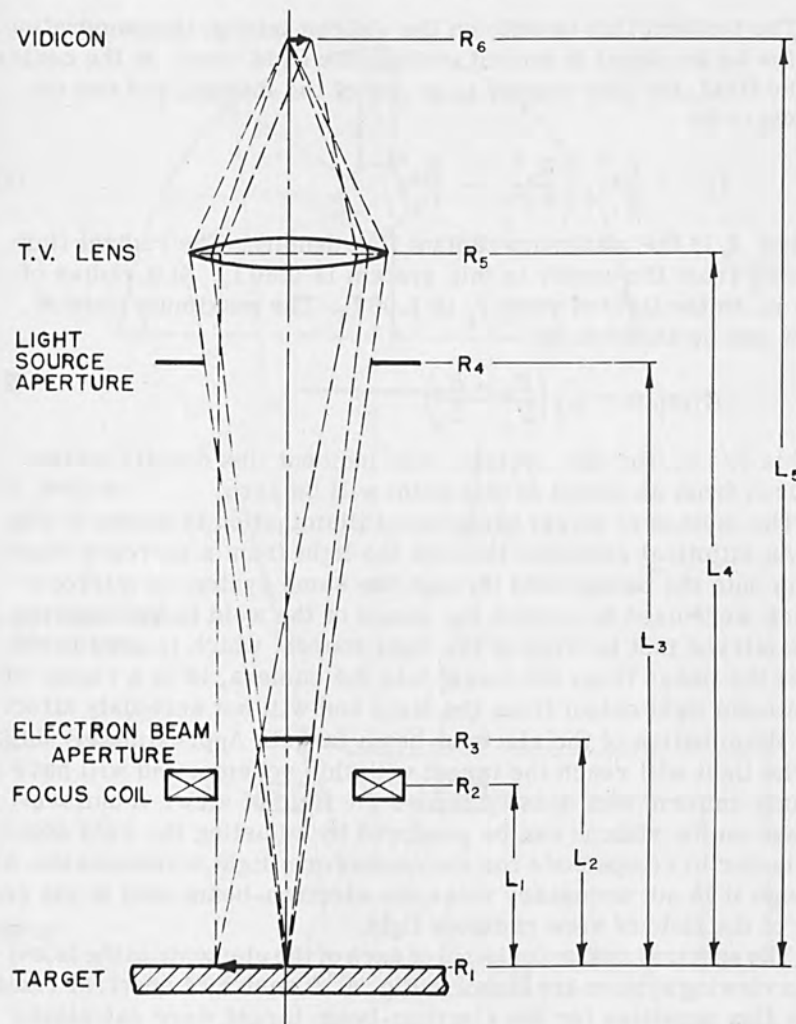


Fig. 2. Optical viewing system.

the monitor is 24. Therefore, a 1-in. object at the target will produce an 8-in. image on the monitor.

Ideally an optical system for television viewing should produce an image on the vidicon tube in the camera of constant flux density as an object on the target is moved through the field of view. However, the limitations around which this system was designed made this nearly impossible.

The incident flux density on the vidicon tube in the camera varies as an object is moved through the field view. At the center of the field, the flux density is at one of the minima and can be shown to be

$$I_i = \frac{L_4^2}{R_5^2} \left(\frac{R_4^2}{L_3^2} - \frac{R_3^2}{L_2^2} \right) I_0 \quad (1)$$

where I_0 is the maximum radiant flux density. The radiant flux density from the center in this system is $0.30 I_0$. At a radius of $1/2$ in. in the field of view, I_i is $1.00 I_0$. The maximum field of view can be shown to be

$$R_1(\text{max}) = L_4 \left(\frac{R_2 + R_5}{L_4 - L_1} \right) \quad (2)$$

and is $3/4$ in. for this system. The incident flux density on the vidicon from an object at this point will be zero.

The method of target background illumination is shown in Fig. 3. An elliptical reflector focuses the light from a mercury vapor lamp onto the background through the same system of mirrors which were used to project the image of the weld to the camera. The mirror just in front of the light source, which is used to reflect the image from the target into the camera, is in a region of minimum light output from the lamp and will not seriously affect the illumination of the electron-beam target. Approximately 50% of the light will reach the target with this scheme, and will have a nearly uniform flux density across the field of view. A uniform image on the vidicon can be produced by adjusting the light source reflector to compensate for the nonuniform light transmission, although it is not necessary since the electron-beam spot in the center of the field of view radiates light.

The spectral characteristics of each of the elements in the television viewing system are shown in Fig. 4. A series of spectral radiation flux densities for the electron-beam target were calculated for several temperatures and are given in watts per square centimeter per micron. This density, W_λ , was determined by assuming that the target was a Planckian radiator and using the equation:

$$W_\lambda = 3.697 \lambda^{-5} \left(e^{\frac{1.432 \times 10^4}{\lambda T}} - 1 \right)^{-1} \times 10^4 \quad (3)$$

where λ is the wavelength of the emitted light and T is the target temperature. At a color temperature of 5000°K , a tungsten target has a maximum output of $1.55 \times 10^3 \text{ W/cm}^2/\mu$ and peaks at a wavelength of about $800 \text{ m}\mu$. This corresponds to a true temperature

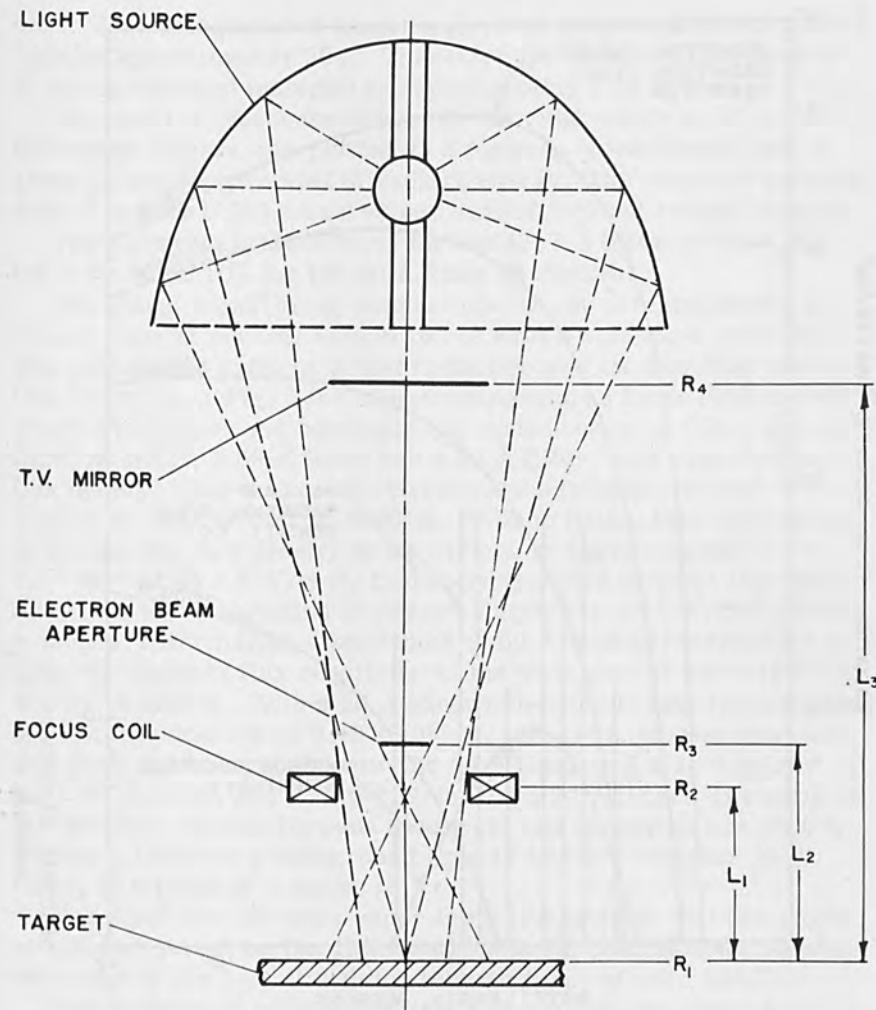


Fig. 3. Background illumination system.

of about 4600°K . At 2000°C , the output is only about $25 \text{ W/cm}^2/\mu$ and peaks around 1.5μ .

A mercury vapor lamp was selected to illuminate the background material because of its high efficiency in wavelengths below the target spectral radiation flux density peak. This lamp emits 50% of its output in three wavelengths 1850, 2537, and $4358 \text{ \AA}$. Since

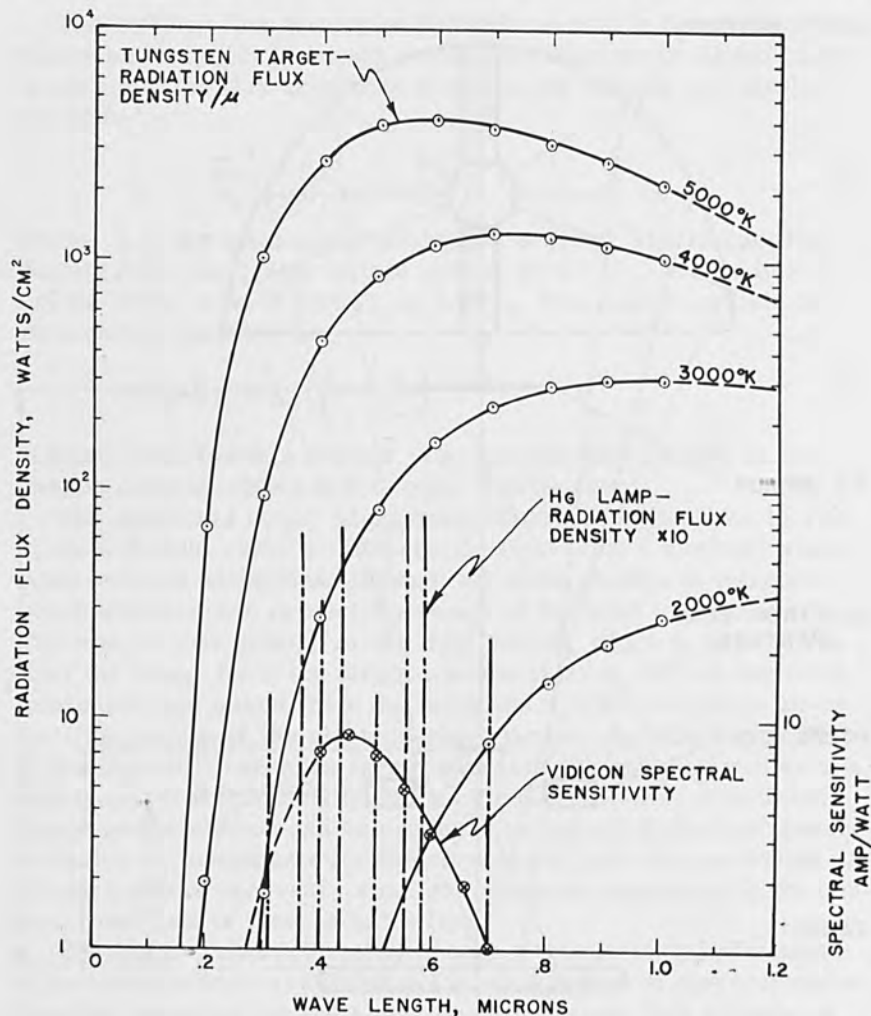


Fig. 4. Spectral characteristics.

optical glass has a cutoff typically around 3500 Å, the 4358 Å line was selected for the background illumination.

The radiation flux density of a 200-W mercury vapor lamp at this wavelength is approximately 5 W/cm². If it is assumed that 50% of this light reaches the background material through the optical system of mirrors, then the irradiation flux density is 2.5W/

cm^2 . The reflectance of tungsten at room temperatures for 4358 Å light is approximately 50%. Therefore the radiation flux density of the background material is approximately 1.25 W/cm^2 .

The spectral response curve for the vidicon tube used in the television camera was plotted as a function of wavelength and is given in ampere per watt of radiant energy. The response peaks at 4500 Å to give 0.085 amperes per watt of incident radiant energy.

The radiation transmission for the 4357 Å filter is about 20% for a 60 Å and 10% for the 20 Å filter bandwidth.

The output signal from vidicon tube can be determined for a molten zone at several temperatures with a cold base material. The contrast or ratio of target to background incident flux densities is shown in Fig. 5 for target temperatures from 1000 to 5000°K. The curves represent contrasts for conditions of no filter and no light, no filter, a 60 Å filter and a 20 Å filter. The total radiant flux density from the molten target varies from 5.7 W/cm^2 at 1000°K to 3549 W/cm^2 at 5000°K. With no filter, this corresponds to an incident flux density at the vidicon of approximately $5.7 \times 10^{-3} \text{ W/cm}^2$ to 3.6 W/cm^2 , respectively. This incident flux density from the background is approximately $6 \times 10^{-3} \text{ W/cm}^2$. With a 4358 Å filter having a bandwidth of 60 Å and a transmission of 20%, the incident flux density from the weld zone is $3.8 \times 10^{-3} \text{ W/cm}^2$ at 5000°K. With a 20 Å bandwidth filter having transmission at 10%, this reduces to $6.46 \times 10^{-4} \text{ W/cm}^2$. The incident flux density from the background with the 60 Å filter is $8 \times 10^{-5} \text{ W/cm}^2$ for a 60 Å filter, and $4 \times 10^{-5} \text{ W/in.}$ for a 20 Å filter. The ratio of incident flux density between the target and background at 5000°K without a filter or a background light is 5×10^4 . With the 20 Å filter, this reduces to about 15.

The filter and mercury vapor lamp combination will therefore produce an image on the vidicon which has a contrast that is within the range of the tube at welding temperatures of near 5000°K.

The problem of coating the first mirror with the vapor from the welding process can be reduced by employing a transparent tape passing in front of the mirror. By adjusting the speed of the moving tape, the heat from the molten target will have little effect on the properties of the tape. With the tape at a slight angle with respect to the perpendicular of the gun axis, a clear, reliable picture can be presented on the monitor for considerable lengths of time.

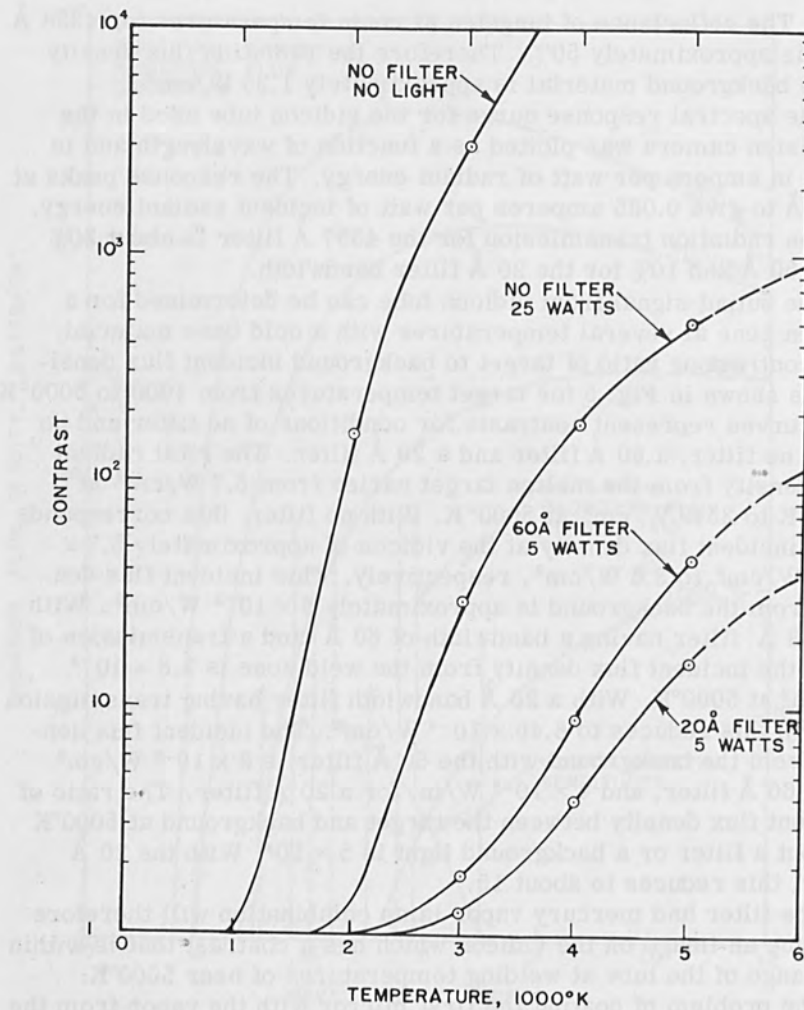


Fig. 5. Ratio of target to background incident flux vs. target temperature.

RESULTS

The television system was used to monitor the welding process

for two materials under several conditions. In all cases the television picture was adjusted for minimum background and maximum target intensities consistent with good definition of detail in both areas. The following three figures were viewed with the TV camera at an oblique angle with respect to the electron beam.

In Fig. 6 there was no filter between the lens and vidicon and no external light source illuminating the background. The target temperature was limited to about 1300°K for optimum detail in the target area. The detail of the background is just barely discernible at this level.



Fig. 6. 1000° C spot without light or filter.

In Fig. 7 the mercury vapor light source irradiated the background but no filter was used. The target temperature in this case was limited to about 1500°K for good definition in the highlights and on the background.

In Fig. 8 a 4358 Å filter with a 60 Å bandwidth was placed between



Fig. 7. 1200°C spot without filter.

the lens and vidicon. The target temperature could be increased to about 3000°K without losing detail in either the molten zone or base material.

In Fig. 9 a 20 Å filter was used, but the camera viewed the weld from directly above the workpiece through the set of mirrors described. In this case the electron gun is actually welding 1/2 in. stainless steel. Details of both the molten zone and base material are clearly definable and the contrast is within the limits of the vidicon tube in the TV camera.

CONCLUSION

The technique of using a light source of specific wavelength to illuminate the base material in an electron-beam welding process, and viewing the process through a highly selective filter, appears to be a very practical method. This technique has been success-



Fig. 8. 3000°C spot with filter and light.

fully employed in conjunction with a closed-circuit television system but could easily be used in direct inspection of the welding process with fixed gun applications in smaller electron-beam welding systems. It has also been shown that it is possible to view electron-beam welding by a relatively low-voltage electron gun directly from above. The apertured mirror placed between the focus coil and anode allowed the image of the weldment to be projected to a remotely located area but did not substantially change the beam characteristic.

ACKNOWLEDGMENT

The author wishes to thank Mr. Lawrence Leonard of the National Company for his assistance in designing the light source and making various calculations. Appreciation is also shown to D. Thomson, A. Shannon, and A. Sierko of Alloyd General for their

help and assistance during the design, construction and testing of the system. Thanks are also given to Mr. George Goldman for his handling of the figures.

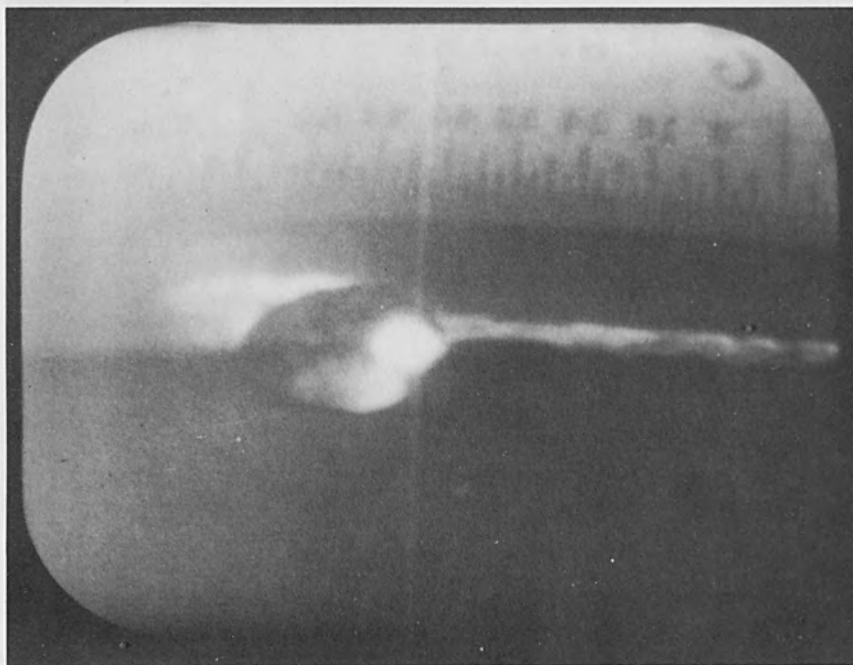


Fig. 9. Stainless steel weld at 2000° C.

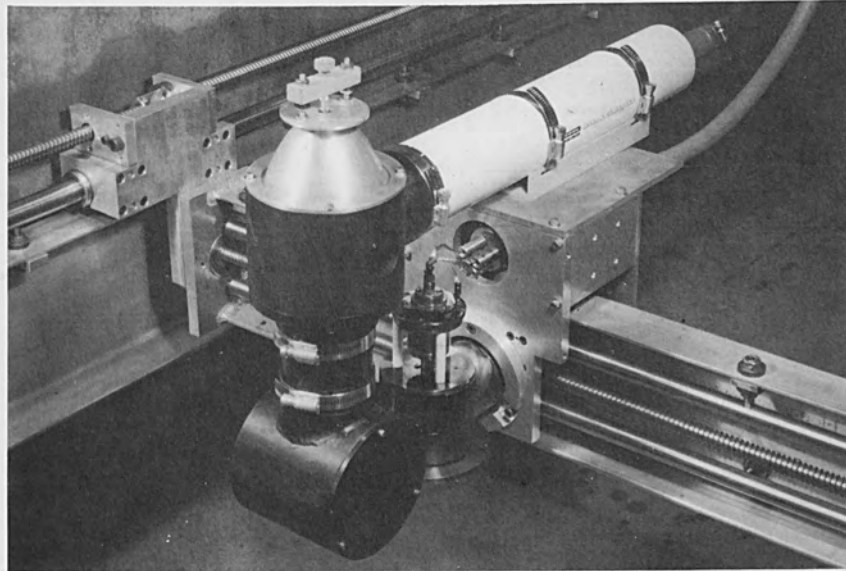
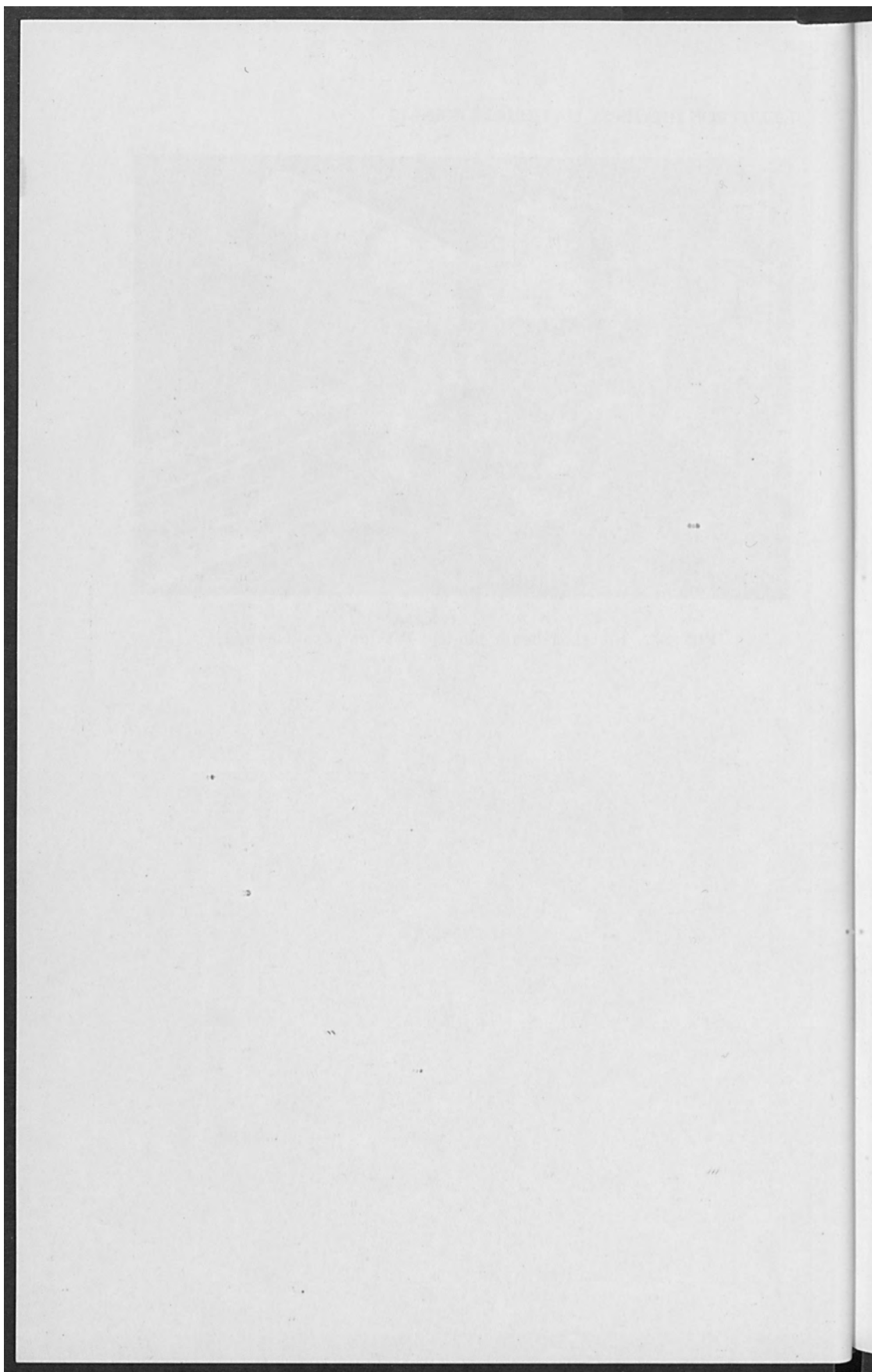


Fig. 10. Electron-beam gun and T.V. camera assembly.



Section VI

ELECTRON BEAMS IN MICROANALYSIS

For those who are not familiar with the technique, the subject is a new way of determining the composition of the unknown in vacuum with high energy electron beams and analyzing them with suitable means, the X-ray spectra emitted by the unknown. The technique is capable of both qualitative and quantitative analysis; the latter

Although all commercial products microprobe systems are capable of both qualitative and quantitative analysis, it is also possible to obtain the data either in air or under vacuum.

Section VI

ELECTRON BEAMS IN MICROANALYSIS

ELECTRON BEAMS IN MICROANALYSIS

Almost forty years elapsed from Moseley's discovery of the X-ray spectrum until Hillier (1947) and Castaing and Guinier (1949) developed the microprobe. The practical realization of microprobe analysis was an event of considerable importance to scientific progress, particularly to those engaged in metallurgy and solid-state work. We were no longer forced to infer the nature of local inhomogeneities from interpretation of average chemical analysis and phase diagrams. We were now able to perform analysis on areas as small as a micron in diameter. Furthermore, these analytical results were just as meaningful as those obtained by alternate reliable analytical methods from samples many times the size.

Much has been accomplished in this field since the discovery of this technique, and two recent volumes by Birks (1963) and Cosslett and Engstrom (1963) are strongly recommended to those wishing to become familiar with this new and powerful tool. An alternate way of obtaining information on the subject is through the courtesy of the producers of commercial microprobe equipment, although I suspect this information might be subject to some degree of bias.

For those who are not familiar with the technique, its essence consists of bombardment of the unknown in vacuum* with fine cross-sectional electron beams and analyzing them with suitable means, the X-ray spectrum emitted by the unknown. The technique is capable of both qualitative and quantitative analysis; the latter

*Although all commercially produced microprobe systems are vacuum instruments, it is also possible to obtain the data either in air or inert atmosphere.

is carried out after derivation of suitable calibration data.

Identification of unknown phases, diffusion studies, surface composition changes, composition inhomogeneities, gradient inclusions, and many related problems are readily handled by microprobe techniques. Although Professor Castaing will refer to this in his presentation, one should say here that today the practical limit of analysis is of the order of 100 to 500 ppm, based on statistical considerations. It is believed that the limit could be brought to the order of 30 ppm. This is considerably lower than the X-ray fluorescence techniques where we hopefully can resolve down to 1 ppm; yet when one considers the nature of these two techniques, i.e., local versus average, it can be seen that capabilities are virtually comparable.

As will be indicated later, one of the most important disadvantages of the microprobe today is the fact that it is under the same limitation relative to light element detection as the X-ray fluorescent analysis. In both cases we are not truly capable of detecting elements with atomic numbers much below 22. It is felt, however, that this is only a temporary limitation and when research to create large lattice spacing crystals produces the needed analyzer crystals this limitation will not hold any longer. For example, potassium acid phthalate crystal with $2d = 26\text{\AA}$, makes possible the detection of O_2 (oxygen) where $K\alpha = 23\text{\AA}$. Long-chain barium stearate crystals can further extend the light element detection range.

Regardless of its limitation, however, we are only a few years away from across-the-board acceptance of the microprobe as a routine analytical tool. This acceptance will be catalyzed by efforts to develop future instruments along two lines. The first line should create research instrumentation with all possible refinements for sensitivity resolution, stability with pricing being relatively unimportant as this type of instrumentation will be used only for advanced research tasks and have a limited market. The second line should create reasonably priced instruments, their price not exceeding 25,000 dollars. They need not have the ultimate in focusing, resolution, etc., but should be a most reliable rugged device, a workhorse for daily routine examinations.

The papers in this section present recent work in this field. After introducing the subject, Castaing and Slodzian discuss the new method of secondary ion micronanalysis. Quatert and Theisen give us their thoughts on quantitative microanalysis, while Theisen, in his second paper, discusses the detection limits of electron microprobe analysis. Moll then shares with us his experience on practical methods for experimental calibration of concen-

tration limitations, while Leroy presents results of a diffusion study, i.e., microprobe application research. Although these few papers do not cover the complete scope of electron-beam microanalysis's problems of today, they certainly give us a good appreciation of some of the important matters involved in this technique and the considerable benefits which can be derived from its application.

Editor

REFERENCES

- Birks, L. S., 1963, "Electron Probe Microanalysis," John Wiley and Sons, New York.
- Castaing, R., and A. Guinier, 1949, *Comp. Rend.*, **237**, 1220.
- Cosslett, V. E., and A. Engstrom, 1963, "X-Ray Optics and X-Ray Microanalysis," Academic Press, New York.
- Hillier, J., 1947, U.S. Patent No. 2,418,029.

ELECTRON AND ION BEAMS IN MICROANALYSIS†

Electron and ion optics have been closely associated in the course of the last fifteen years to the development of new analytical tools in the field of solid-state physics: electron probe microanalysis and secondary ion microanalysis.

The first of these techniques is well known; it is essentially a refinement of the very ancient method of X-ray emission analysis. The basic operation consists in bombarding with an electron probe the point at the surface of a massive sample which has been selected for analysis, then measuring by X-ray spectrometry the characteristic lines which are emitted by the various elements present in the irradiated volume. By comparing the intensities of these characteristic lines to the intensities of the corresponding lines emitted in the same conditions by pure standards, the local concentrations of the various elements, in a volume of the order of one micron cube, are obtained with an accuracy of about 1%.

The local character of the analysis is very important indeed, since it opens the way to an impressing number of applications in the fields of metallurgy, mineralogy, or even biology. The metallurgist, for instance, is now able to analyze practically all the precipitates, segregations, or inclusions that he is able to see under the metallographic microscope. But the main advance brought about by electron probe microanalysis results more from the new principle involved in the quantitative measurements than from the spot character of the analysis. The simple ratio of two measurements of the same characteristic line, when emitted successively by the point of the sample to be analyzed and by a reference standard of the pure element, may be considered as a fair approximation for the mass concentration of the element in the analyzed material. Such a linear behavior holds for any concen-

*Department of Solid-State Physics, University of Paris, Orsay, France.

†This is one of the four Plenary Session presentations.

tration; it is independent of the chemical bonds and it undoubtedly constitutes the most important advantage of X-ray emission analysis over other methods such as light spectrometry for instance.

Some problems require a high level of absolute accuracy; such is the case for instance when electron probe microanalysis is used for establishing the equilibrium diagram between two metallic elements; a diffusion couple is prepared and the concentration of both elements is plotted across the diffusion zone. Some difficulties may occur from the steep concentration gradients in the immediate vicinity of the phase boundaries; they may be overcome only by combining a good quantitative accuracy—1% or better—with the best resolving power, say 1μ . The same is true for the identification of precipitated phases in the alloys; an accurate quantitative analysis is necessary for distinguishing between neighboring tentative formulas.

Such an accuracy may be obtained by applying some corrections to the raw measurements. The corrections involve the self-absorption of the characteristic radiation in the sample itself and the secondary fluorescence emission of the sample excited by both characteristic and continuous spectrum; furthermore, the deceleration and the backscattering of the electrons are not the same in various targets, resulting in an atomic number correction. The important point is that, except for a few exceptional cases, the corrections are small and unaffected by the chemical bonds.

Other problems, for instance, segregation studies, do not require so high a level of absolute accuracy: the main interest lies more in the variation of the concentration of a given element across an extended region than in its true concentration at a given point. Such problems may obviously be solved by performing a series of point-by-point analyses and plotting the results on a concentration map; but this type of measurement is made much simpler and less tedious when using the scanning method of analysis which was developed by Cosslett and Duncumb. The electron beam is scanned across the specimen by two pairs of coils or electrostatic plates and the output of the spectrometer is used to control the grid of a cathode ray display tube which is scanned in synchronism with the main electron beam. In this way a picture is obtained of the specimen surface, showing the distribution of the element whose characteristic line has been chosen for analysis.

Scanning analysis is very attractive, and many experimenters have brought their attention on this type of semiquantitative analysis and disregarded a little the main quality of electron probe microanalysis, which lies in the possibility of absolute point-by-

point quantitative analyses. Then, in regard of this fundamental quality, X-ray emission analysis is subject to some limitations: the analysis of very light elements such as carbon, nitrogen, and oxygen is very difficult because of the extreme softness of their characteristic lines, although interesting results have been obtained recently in that respect by using diffraction gratings or organic layers for separating the characteristic lines.

Concerning the resolving power, its limitation to about 1μ , which arises from the electron diffuse penetration in the sample, is not too disturbing when considered in the surface direction; but a similar limitation occurs for the depth-resolving power, which prevents practically the analysis of thin superficial layers on massive substrates.

The difficulty may be overcome by using thin specimens; the continuous spectrum is much weakened and the faint characteristic lines of the superficial layer may be easily detected. For example, Fig. 1 shows the K spectrum of krypton registered on a thin sample of aluminum—400 Å units thick—which has been bombarded for 50 seconds with a beam of Kr^+ ions (4 keV, $1\mu\text{A}/\text{mm}^2$). The $K\alpha$ and $K\beta$ lines of Kr protrude strongly over the continuous spectrum, although the amount of krypton inserted in the sample was corresponding to less than three atomic layers.

Another advantage of this type of transmission analysis lies in the possibility of obtaining a much better resolving power. The resolution is no more limited by the electron diffuse penetration in the sample; if the current density may be kept high enough in very small probes by correcting the probe-forming lens for spherical aberration, it may be hoped that a resolving power better than 400 Å will be obtained on metallic specimens where the thermal effects are small.

Nevertheless, it may be concluded that, for the moment electron probe microanalysis, is not well adapted to very light elements; is limited to a resolving power of about 1μ when operating on massive samples; and is not convenient for analysing thin superficial layers on massive substrates.

Fortunately, another method of microanalysis is now available; its principle is entirely different, as it uses ion optics for producing "characteristic images" which give the distribution of the various constituent elements across the sample; its qualities and defects are in some way complementary of those of electron probe microanalysis.

When a solid surface is bombarded by a beam of fast particles such as ions, a sputtering process occurs. The sample is pro-

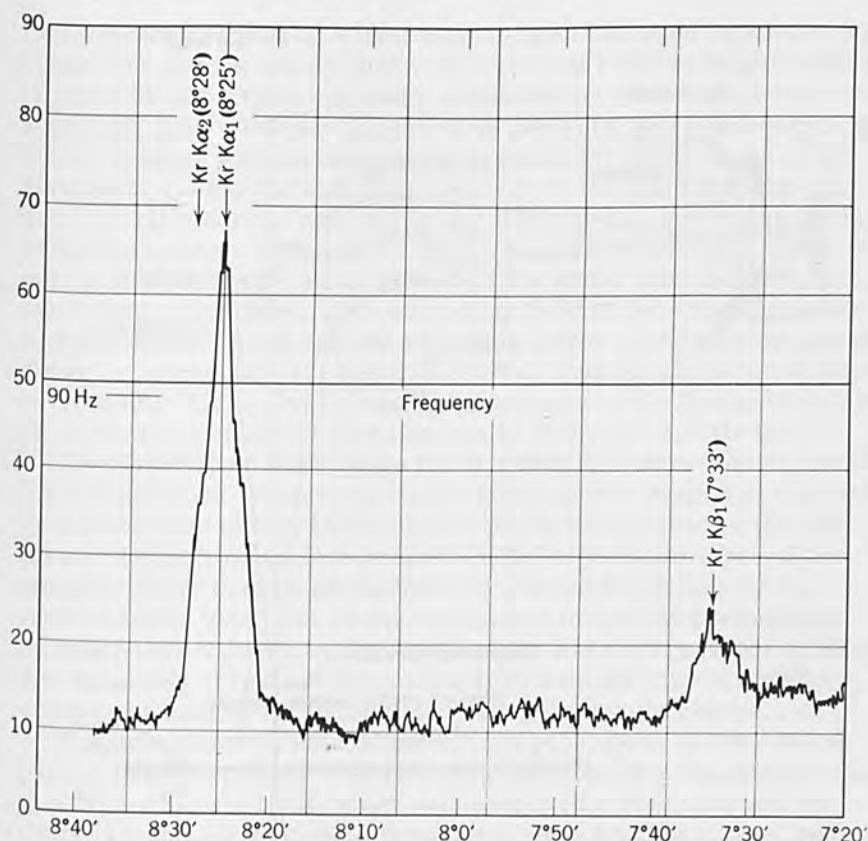


Fig. 1. *K* spectrum of krypton inserted in aluminum.

gressively etched down and a non-negligible part of the particles which are extracted from the surface by the bombardment are constituted by ions. These ions are formed from the atoms which are present in the first atomic layers; they leave the bombarded surface with low energies, of the order of a few electron-volts; it is thus possible, by focusing them with an emission lens, to obtain an image of the sample surface which is formed by the whole of the secondary ions. This "material image" can then be separated by mass spectrography into its component "characteristic images"; each of the characteristic images is carried by a given type of ion and it brings with it the map of distribution of the corresponding element—or better isotope—across the specimen surface. A schematic diagram of the instrument is represented on Fig. 2.

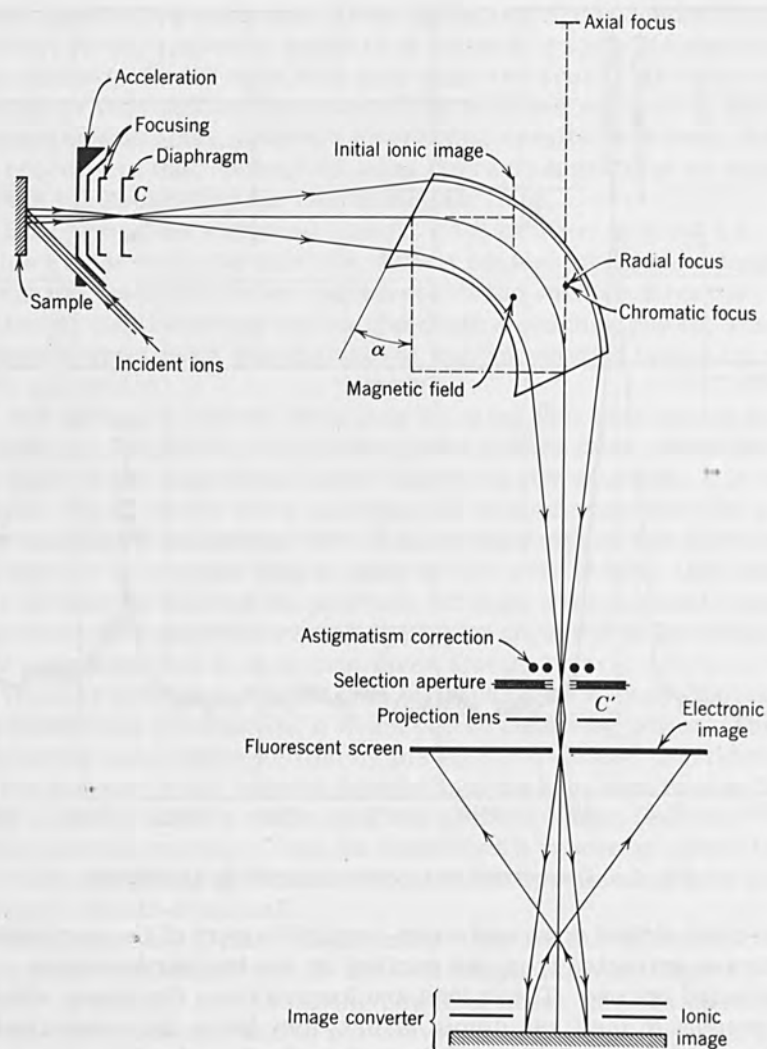


Fig. 2. Schematic diagram of the secondary ion microanalyzer.

The primary ion beam—argon ions or protons with an energy of some keV—is produced by a conventional high-frequency gun; it is focused onto the sample by an electrostatic condenser. The diameter of the irradiated part of the object surface is about 0.5 mm for a total ion beam intensity of about $20\mu\text{A}$. The secondary

ions are accelerated by a first electrode and focused by an electrostatic lens into an image of the object surface whose magnification is about 30. This first ion image is formed by the whole of the characteristic ions which are focalized in the same way whatever their mass; in some way, it reproduces in space the upper layer of the specimen. An aperture at the crossover of the emission lens is used for eliminating from the image those of the ions whose initial velocity is larger than a given limit. To give an example, a 0.4-mm aperture practically eliminates the ions whose initial energy is much larger than 1 eV. The extracting field at the object surface is about 10,000 V/cm and the resolving power of the first image is about 1μ . Obviously, the resolving power may be increased at will by reducing this aperture, but the brightness of the image decreases at the same time; we shall return to this point a little later.

This first composite image is formed by the superimposition of many component images; each of the component images is carried by a given type of ion, for instance one of the isotopes of the different elements which are present in the superficial layer of the sample. Some images are formed by ionized molecules or by multicharged ions; one of the component images is produced by primary ions diffused by the sample with low energy; these images are generally very faint when compared with the characteristic component images formed by ions bearing a single charge.

The separation of the component images occurs in the second part of the instrument; this part is constituted by a magnetic prism of a type (uniform field) which was proposed a long time ago by Cotte [1], and whose optical properties were studied by Hennequin [2] in our laboratory. The deflection of the ion beam is 90 degrees. For a convenient choice of the angle of incidence of the ion beam on the magnetic gap, the ion trajectories are focused in the same way in both transverse directions. The conical ion beam issued from the crossover of the emission lens which carries one of the component images converges after the magnetic prism onto a second crossover; at this level a selecting slit isolates the ions whose mass is corresponding to the excitation of the magnet. At the same time, the magnetic prism gives from the first image a second virtual image; unfortunately the prism is a non-Gaussian optical system and the only couple of stigmatic points is constituted by the two conjugated crossovers; as a result, this second virtual image is strongly astigmatic and each object point gives rise to two focal lines.

Furthermore, this second image is generally blurred by a strong chromatic defect. This defect arises from the fact that the

ion beam is not exactly monoenergetic, as the first aperture is limiting the lateral component only of the initial velocity of the secondary ions. Fortunately, it can be shown that a variation of the velocity of the ions, or a variation of the magnetic field, results in a rotation of the emerging trajectories around a "chromatic focal line" which is situated behind the exit pole face, at a distance which is theoretically equal to the two-thirds of the curvature radius of the ion trajectories. It is then possible, by properly adjusting the excitation of the first emission lens, to arrange things so that the second image is formed in the plane of this chromatic focal line, which constitutes an achromatic plane. In more precise words, the radial focal lines of the second image are formed on this achromatic plane. In such a way, the chromatic defect is reduced to the second order and a conventional stigmator is only necessary for obtaining a filtered image whose quality is the same as that of the first image.

This ion characteristic image is generally much too faint for a direct observation on a fluorescent screen whose sensitivity for the heavy ions is extremely low. For observing it, we use an image converter of the type which was proposed by Mollenstedt [3]. The virtual ion image is focused by a telescopic arrangement of electrostatic lenses onto the curved cathode of an electron emission lens; this cathode is made of aluminum and carefully polished, so that it is devoid of any visible structure by itself. The impingement of the ions onto this cathode gives rise to a secondary electron emission; the secondary electrons are accelerated backwards and produce a final image on a fluorescent screen. The efficiency of the image converter is large enough for giving, from an ion image which would be invisible if observed directly on a fluorescent screen, an electron image visible in daylight at a magnification of some hundredths.

This electron image might be deflected again for recording it conveniently on a photographic plate; that will be done in the near future. For the moment, we have obtained the pictures by photographing the fluorescent screen from the outside of the instrument; such a procedure results in a considerable loss of sensitivity, and consequently of resolving power as we shall see later, but it was convenient for the first trials.

A general view of the experimental instrument is represented on Fig. 3. The vacuum is obtained by differential pumping down; it is of the order of 10^{-7} mm Hg in the main body of the instrument, and better than 10^{-6} mm Hg in the region between the object surface and the first aperture of the emission lens.

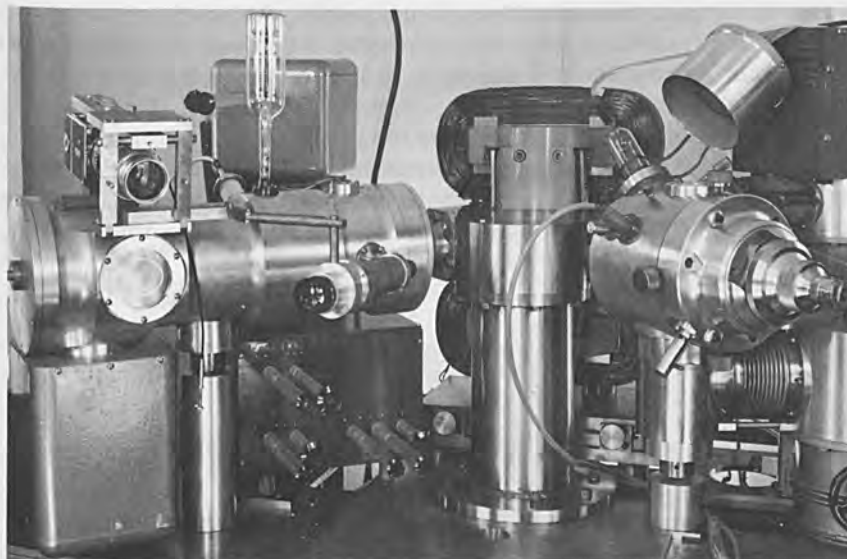


Fig. 3. General view of the secondary ion microanalyzer.

Let us consider now the possibilities of the method; the instrument is delivering images which represent the distribution maps of the various elements or isotopes across the specimen surface; one of the main characteristics of the method is the resolving power of those images.

The first limitation of the resolving power arises of course from the aberrations of the optical system, which comprises the first emission lens, the magnetic prism, the telescopic arrangement of electrostatic lenses, and the image converter. It should be noted that, if the magnification produced by the first emission lens is large enough, the aberrations introduced by the other parts of the optical system will be negligible because of the very small aperture of the image pencils. As a matter of fact, we have verified that the sharpness of the last image, observed on the fluorescent screen, is nearly identical to that of the initial image produced by the first emission lens. But some amount of distortion could be introduced by the whole of the optical system; apart from the second-order distortion produced by the magnetic prism, a pincushion distortion is produced by the first emission lens, the telescopic arrangement of electrostatic lenses and the electron

emission lens of the converter. Fortunately, the whole of those pincushion distortions may be compensated by the barrel distortion which arises from the acceleration of the ions by the electrostatic field of the converter lens; for a convenient choice of the potential applied to the cathode of that lens, the compensation is perfect and the last image is affected only by the second-order distortion introduced by the magnetic prism; that distortion is very low as may be seen in Fig. 4 (25- μ copper grid pressed on an aluminum block, Al^+ image).

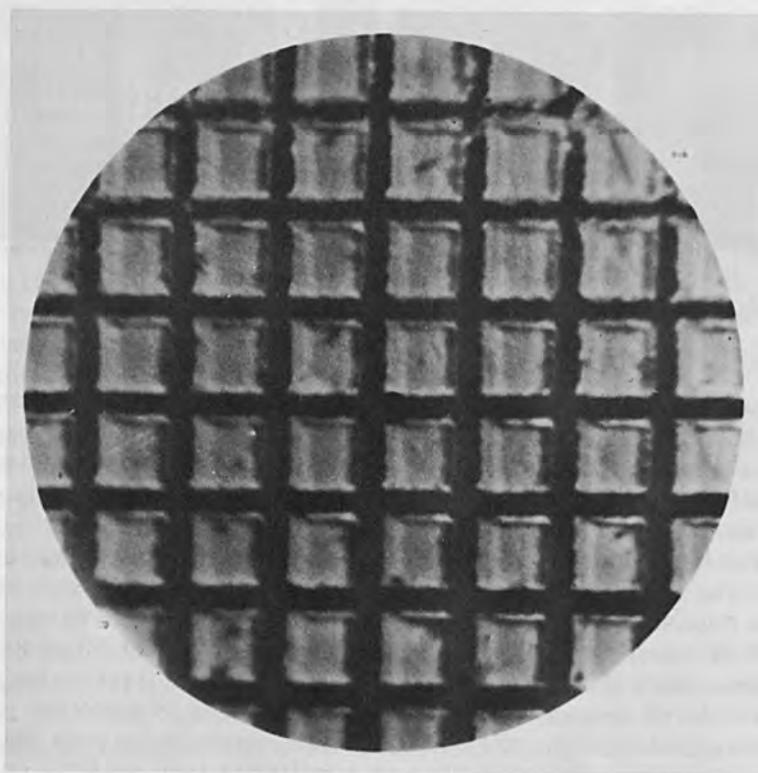


Fig. 4. Copper grid pressed on an aluminum block (Al^+ image).
The diameter of the imaged regions (Figs. 4 to 9) is about 0.2 mm.

We may thus consider that the resolving power of the analysis is identical to the resolving power of the ion image produced by

the first emission lens. Now, this resolving power may be increased at will by reducing the diameter of the aperture which is located at the crossover of that lens; for example, the image obtained with Al^+ ions on a sample of cast Al-Mg-Si alloy containing dendritic precipitates of Mg_2Si phase is represented in Fig. 5; the resolving power is about 1μ and could be considerably improved by reducing the diameter of the crossover aperture and using a direct recording of the electron image for compensating the loss of brightness. It seems at first sight that the resolving power

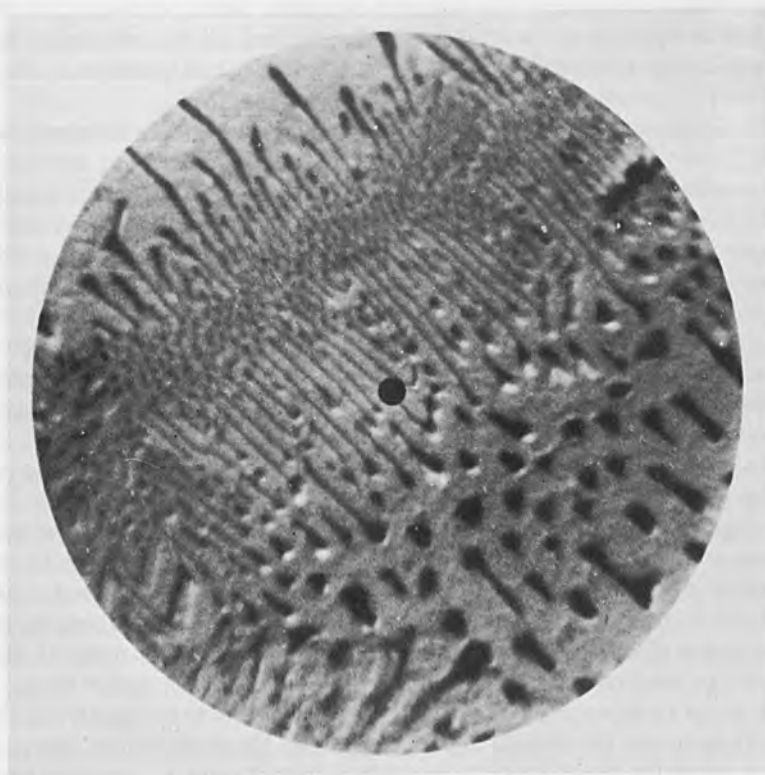


Fig. 5. Al-Mg-Si cast alloy (Al^+ image).

of the analysis is unlimited; as a matter of fact, no fundamental limitation is occurring here from the diffusion of the primary particles inside the sample, as it is the case in electron probe

microanalysis; the penetration of the incident ions inside the sample is of the order of some atomic layers only.

Nevertheless, there remains a limitation for the ultimate resolving power of the method, which arises from the destructive character of the analysis. The ions which contribute to the image formation are extracted from the sample itself; now the number n_i of the characteristic ions which are produced by a given bombardment is much less than the number n_0 of neutral atoms which are etched off during the same time; furthermore, the contrast aperture, if its diameter is small, eliminates from the image a large fraction of these characteristic ions. As a consequence, a minimum volume of material must be etched off for obtaining an image: hence a limitation in the ultimate resolving power of the analysis.

For estimating this limiting resolving power, we may proceed as follows: Let us designate by ϵ the limit of resolution and by p the accuracy which must be obtained for the quantitative analysis of the elementary volume ϵ^3 . In the most favorable case where we are provided with perfect calibration curves for deducing the concentration from the secondary ion emissions and where these secondary ion emissions are measured with a perfectly sensitive receiving unit — that is, a recorder which is able to detect the individual ions — the accuracy of the determination of the concentration will be limited by the statistical fluctuations which affect the number of characteristic ions emitted by the elementary volume. As a result, the elementary volume must be able to send in the receiving unit a number of characteristic ions equal to $1/p^2$ for a quantitative analysis of accuracy equal to p . We see immediately that the main factor for the limiting resolving power is the efficiency of the secondary ion production, that is the ratio n_i/n_0 between the number of characteristic ions which contribute to the image of a given part of the object and the number of neutral atoms which are etched from this part of the object at the same time. This ratio is proportional to the area of the contrast aperture; on the other hand, the resolving power of the first emission lens, which must be equal to ϵ , is roughly proportional to the square of the diameter of this aperture for a given extracting field at the object surface, say 10 kV/cm; as a result, the ratio n_i/n_0 is proportional to the desired resolution.

Let $(n_i/n_0)_{\text{exp}}$ be the value of the ratio n_i/n_0 which is obtained in a standard experiment where the contrast diameter is adjusted to the value which corresponds to a one micron resolving power (0.4 mm in our present experimental conditions). We may write,

expressing ϵ in microns:

$$n_i/n_0 = \epsilon (n_i/n_0)_{\text{exp}}$$

Combining with $n_0 = K\epsilon^3$ (K is a constant depending upon the lattice parameter) and $n_i = 1/p^2$ (from statistical considerations) we obtain:

$$\epsilon(\mu) = K^{-1/4} p^{-1/2} (n_i/n_0)_{\text{exp}}^{-1/4}$$

The value of K is about $6 \cdot 10^{10}$ atoms/ μ^3 . The efficiency coefficient $(n_i/n_0)_{\text{exp}}$ obviously depends upon the concentration of the imaged element; furthermore, for pure elements, it varies in very large limits from one element to another; for pure aluminum for instance, it is about 10^{-3} , for pure copper it is only 10^{-7} . As a result, for our present experimental conditions where the extracting field at the object surface is 10,000 V/cm, the limiting resolving power for a 10% accuracy would be about 350 Å in the case of aluminum, 0.35μ in the case of copper. Progress will certainly be possible in that respect, by increasing the value of the extracting field in the first emission lens and by improving the ionizing conditions by a proper choice of the nature and energy of the primary particles.

In the course of our estimation of the limiting resolving power, we have considered the "accuracy" p of the analysis; as a matter of fact, p was the accuracy which could be deduced from the measurements if we were provided with perfect emission-concentration relations. Unfortunately, those relations are far from being linear and, in that respect, secondary ion microanalysis is much inferior to electron probe microanalysis where the concentrations are related to the measured intensities by simple proportionality laws.

To give an idea of the complexity of the secondary ion emission process, let us consider first the case of a pure metal M . The main part of the emission is made of ions M^+ bearing a single charge; but apart from those single ions, the spectrum comprises molecular ions M_2^+ , M_3^+ , ... and multicharged ions M^{++} , or even in some cases M^{+++} , depending on the outer shell of the atom. When the metal is made of a mixture of isotopes, a lot of different molecular ions may thus be obtained.

In the alloys the phenomena are rather complex. When the alloy is made of neighboring elements such as Al, Mg, Si, or Cu, Ni, the ion emissions are roughly proportional to the concentrations as it was the case for isotope mixtures.

For example, the micrographs obtained on a sample of cast Al-Mg-Si alloy containing precipitates of pure Si and precipitates of Mg_2Si phase are represented on Fig. 6. The three images are

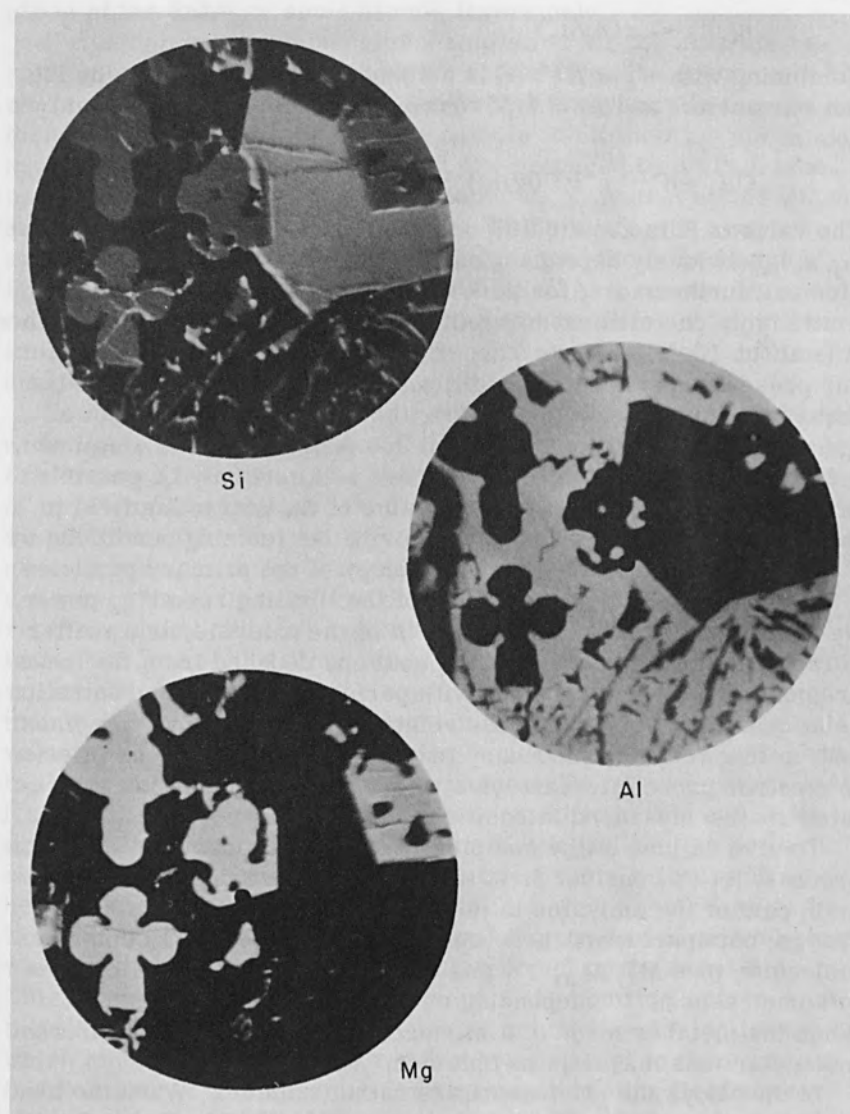


Fig. 6. Al-Mg-Si cast alloy.

corresponding to the distributions of ^{24}Mg , Al and ^{28}Si , respectively. The experimental procedure for obtaining such images is quite simple; the specimen is carefully polished; it is introduced in the instrument by using a vacuum lock which enables the high vacuum to be obtained after about half a minute pumping down. Then, when the ion primary beam is on, it is only necessary to modify slightly the excitation of the magnet to observe successively on the screen three identical magnesium images (masses 24, 25, and 26), the aluminum image (mass 27) and three identical silicon images (masses 28, 29, and 30), the two last being very faint. The successive images appear and disappear without any motion if the focusing is correctly adjusted in the achromatic plane.

The images obtained on the same sample by using scanning electron probe microanalysis would be quite similar, so that it may be said in this case, where the component metals have similar atomic numbers, that the ion emissions are roughly proportional to the concentrations. But the situation is quite different for instance in the case of Cu-Al or Cu-Be alloys; the $^{63}\text{Cu}^+$ emission of a Cu-Al alloy containing 5% Al is seven times higher than the emission of pure copper in the same conditions.

Figure 7 represents the image obtained with $^{63}\text{Cu}^+$ ions on a sample of copper containing nodules of Cu_2O . The copper oxyde appears in bright on the micrograph, whereas its copper content is certainly lower than that of pure copper! As a general rule, in the case of compounds, the emission is strongly dependent upon the chemical bond. Ionic compounds give rise to a very strong emission of the metallic ion, which may exceed that of the pure metal by two orders of magnitude.

As a conclusion, a better knowledge of the true mechanism involved in secondary ion emission will be necessary before we can assess the true possibilities of the method for quantitative measurements. For the moment, the situation is about the same as in light spectrometry for instance, where the empirical calibration with known standards is the general rule.

On the other hand, the dependence of the secondary ion emission on the nature of the chemical bonds may possibly be advantageous in some cases; we may hope that the combination of both types of analyses, X-ray emission and secondary ion emission, will give informations on the structure of the compounds that we could not obtain by using X-ray microanalysis only.

Furthermore, it could be feared that the emission of multi-charged ions and that of molecular ions will very much complicate the interpretation of the images by introducing the possibility of

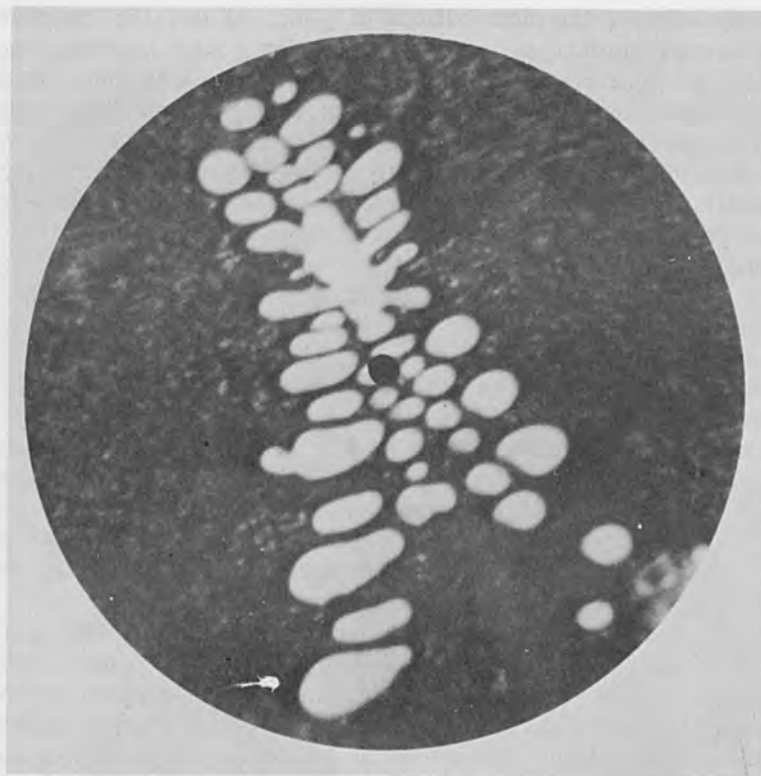


Fig. 7. Copper containing nodules of Cu_2O ($^{63}\text{Cu}^+$ image). The oxide appears in bright.

confusion between different ion combinations of the same mass. Fortunately, the difficulty may generally be overcome by using the isotopes. For example, an image corresponding to the mass 58 may be produced, either by $^{58}\text{Ni}^+$ ions or by $^{58}\text{Fe}^+$ ions; if the image is carried by nickel ions, an identical image will appear for the mass 60; this image will not appear in the case of iron ions.

These difficulties concerning the quantitative interpretation of the results are compensated by many advantages. First of all, the method seems particularly well adapted to the study of the light elements such as lithium and beryllium, which are practically out of the scope of the electron probe microanalyser. We have observed, for instance, the emission of oxygen positive ions by the oxides, of nitrogen ions by the nitrides, so that the identification of those

compounds will be easy by using the new method. For elements such as fluorine and chlorine, especially in the case of ionic compounds, no positive ion is emitted, but a new adjustment of the various potentials makes it possible to use the negative ions emitted by those elements for obtaining characteristic images.

The image obtained with $^{35}\text{Cl}^-$ ions on a sample prepared by evaporating a sodium chloride solution on a stainless steel substrate is represented on Fig. 8.

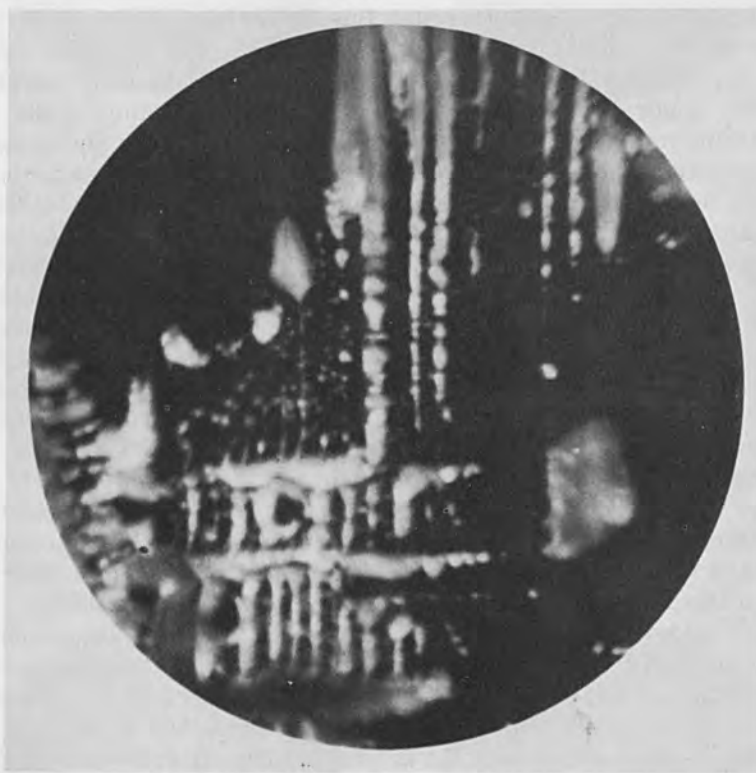


Fig. 8. Sodium chloride on stainless steel ($^{35}\text{Cl}^-$ image).

It should be noted that the method may be applied to insulating samples. The experimental procedure is more complicated than in the case of electron probe microanalysis, where it is only necessary to evaporate a thin layer of metal onto the insulator.

Such a procedure is obviously unapplicable to the case of secondary ion microanalysis where the emission is restricted to the first atomic layers. The difficulty may be overcome by evaporating aluminum onto the insulating sample through a mask formed by parallel metallic wires. The result is the deposition on the sample of a set of metallic wires whose distance is about 0.05 mm; the charging up effect is strongly reduced and the quality of the images is nearly as good as in the case of metallic samples.

The method has been extensively applied, in conjunction with Capitant and Troly, of the French Bureau de Recherches Géologiques et Minières, to the study of beryllium ores extracted in the Spor Mountains of the United States. It was known that the ore contains 1 to 2% of beryllium, but the exact mineralogical nature of the beryllium compound was undetermined. The study has shown that the beryllium was partly associated with silicon; as a result, it may be asserted that this ore contains the mineral known as "bertrandite." A similar work has been carried on jadeite, showing for the first time the substitution of sodium by aluminum in the vicinity of the grain boundaries. The same type of substitution was observed on different types of mica. As an example, Fig. 9 corresponds to an insulating sample of granite. In the observed region, particles of lepidolite (mica containing lithium) are included in a quartz matrix. The various images show, respectively, the distributions of lithium, sodium, aluminum, silicon, potassium, and calcium across the sample. The silicon content in the quartz is clearly higher than in the lepidolite (aluminosilicate); it is to be noted too that in the lepidolite, the lithium rich regions are poor in sodium and vice versa. It is observed by looking at the aluminum distribution image that the aluminum ions emitted by the insulating sample appear at the same time (that is for the same adjustment of the magnetic prism) that the ions are emitted by the aluminum wires; the conclusion is that the charging up effects in the sample have been maintained to a negligible level (less than 20 V).

As a conclusion, comparing the respective advantages of electron probe microanalysis and secondary ion microanalysis is difficult, and the two methods would better be considered as complementary from one another. Concerning the quantitative interpretation of the results, the ion method is for the moment, and will probably remain, much inferior. On the other hand, its resolving power is no more limited by diffusion considerations and reducing it below 0.1μ is only a matter of technology. Furthermore, the depth-resolving power is considerably better than that of electron probe microanalysis; the resolution that we have estimated above

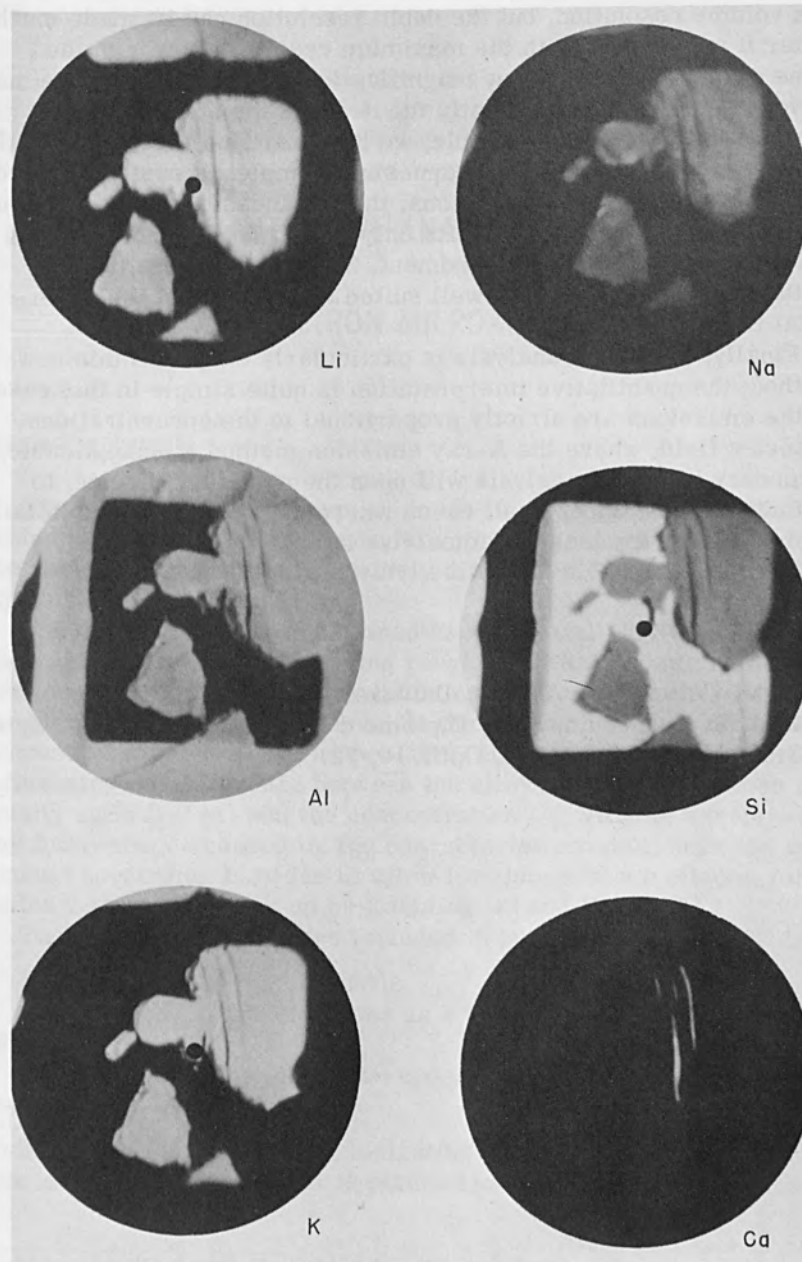


Fig. 9. Granite (lepidolite inclusions in quartz).

is a volume resolution, but the depth resolution can be made much better if we do away with the maximum resolving power in the plane of the surface. At low magnifications, the exposure time may be very short, and consequently the depth of the etched layer is very small. To take an example, we have verified that the natural oxide layer, which covers a magnesium sample, is easily detected through its oxygen secondary ions; the thickness of the oxide layer is some tenths of angström units only, and it is eliminated during the first second of the bombardment. We can thus hope that the method will be particularly well suited to the study of the superficial films.

Finally, isotopical analysis is particularly easy with this new method; the quantitative interpretation is quite simple in this case, as the emissions are strictly proportional to the concentrations. In such a field, where the X-ray emission method is unapplicable, secondary ion microanalysis will open the way, for instance, to self-diffusion studies, in all cases where the tracer method is failing because of the lack of radioactive isotopes.

BIBLIOGRAPHY

1. M. Cotte, 1938, Thesis, University of Paris.
2. J. F. Hennequin, 1960, Diplôme d'Etudes Supérieures, Paris.
3. G. Möllenstedt, 1953, *Optik* 10, 72.

QUANTITATIVE ANALYSIS USING AN ELECTRON MICROANALYZER

INTRODUCTION

The purpose of this paper is, on the basis of calculations published by Castaing in his thesis, to set up correction formulas for the analysis of alloys having widely different atomic numbers, e.g., uranium and aluminium.

The correction curves obtained experimentally by Castaing [1], and verified for elements having roughly similar atomic numbers, are insufficient to enable absorption correction to be carried out for elements as far as uranium. In the setting up of our formulas, account was taken, as far as possible, of all the physical factors influencing the difference between the alloy standard-specimen intensity ratio $I_A/I(A)$ and the concentration C_A , with the exception of any fluorescence caused by the characteristic radiation or the continuous spectrum. In order to allow for fluorescence effects, formulas were already set up by Castaing [1] and Henoc [2].

The following factors are included in our calculations:

- 1) Absorption of the electrons.
- 2) Diffusion of the electrons as a function of the penetration depth.
- 3) Variation of the ionization cross section as a function of the energy of the electrons.

In the present report, the ionization cross section K will be considered a constant, but it is planned to extend the numerical

*Euratom Research Center, Ispra, Varese, Italy.

†Institut für Angewandte Reaktorphysik, Kernforschungszentrum, Karlsruhe, West Germany.

calculations in order to take account of the variation of K as a function of the energy of the electrons. By this, we do not mean to imply that our correction formulas might possibly replace Castaing's experimental correction method. Any theoretical correction is by necessity based on absorption and diffusion laws governing electrons, such as Lenard's and Bothe's laws, which have hitherto proved to be at best an approximation. A further difficulty, which should not be underestimated, is the insufficient available knowledge concerning the absorption coefficients of soft X-rays and electrons.

We hope, however, that our contribution will increase the accuracy of microprobe analyses on such occasions when the preparation of standard samples is too difficult or complicated, especially since, in the preparation of homogeneous standard samples, insurmountable difficulties are frequently encountered.

Recapitulation of Castaing's Pinpoint Analysis Principle

A microprobe is aimed at a point on the surface of a massive sample the chemical composition of which is to be studied. Under the impact of this electron bombardment, the very small volume of matter irradiated by the electrons (in the order of a cubic micron) emits X-rays containing the characteristic lines of the various elements present at the point of impact of the probe. The respective concentrations of these various elements at the point of impact can then be obtained by spectrographic analysis.

Let us consider a series of alloys AB , in which $I_A/I(A)$ is the ratio between the intensities emitted in the $K_\alpha(A)$ line under the same excitation conditions by the alloy and the pure element A , respectively. Let us consider the curve representing the variations in $I_A/I(A)$ as a function of the mass concentration C_A of the element A in the alloy. It is obvious that this curve starts at the origin $C_A = 0$ to reach the point $I_A/I(A) = 1$ when $C_A = 1$ (pure element A).

The simplest hypothesis is to assimilate this curve into a straight line, i.e., to suppose that the intensity of the line $K_\alpha(A)$ emitted by the alloy is proportional to the concentration of A . It will be seen that this hypothesis amounts to considering that an electron beam of given characteristics irradiates a fixed mass of any given substance, encountering a mass of the element analyzed which is proportional to its mass concentration.

It will be seen that, despite its simplicity, this hypothesis is broadly speaking a good approximation. Moreover, it is possible to get even closer, for if, for instance, instead of assuming, as we

have just done, that equal masses of two elements A and B have the same absorption power for electrons of a given energy, we simply suppose that a mass of m_B of the element B can be found which is equivalent from the interaction viewpoint to the beam of a given mass m_A of the element A , we obtain for the ratio $I_A/I(A)$ an expression in the form:

$$\frac{I_A}{I(A)} = \frac{\alpha_A C_A}{\sum \alpha_i C_i}$$

when α_i are the constant coefficients attached to each element in the alloy.

It will therefore then be possible to obtain a general equation containing one or several parameters to be determined empirically with the aid of several standard alloys which can be prepared.

Castaing's microprobe can be used for the pinpoint quantitative analysis of elements with an atomic number higher than 11 (Na). The resolution power (volume affected lower than one cubic micron and concentration limit about 0, 1%) provides an indication of the performance of this apparatus, which is capable of analyzing less than $1 \mu^3$ of matter.

QUANTITATIVE ANALYSIS—GENERAL REMARKS

We have seen that approximate results can be obtained by assuming $I_A/I(A) = C_A$, where I_A is the X-ray intensity generated in the alloy and $I(A)$ is the X-ray intensity generated in a standard specimen of element A under the same conditions for high voltage, probe current, spectrometer setting, etc.

In order to obtain more accurate results the following corrections should also be carried out:

- 1) Absorption of characteristic X-rays in the anticathode (sample and standard).
- 2) Electron diffusion in the anticathode.
- 3) The dependence of the ionization cross section on the energy of the electrons.
- 4) In some cases, $\lambda < \lambda_k$ the absorption of an X-photon from either characteristic radiation or a part of the continuous spectrum in the anticathode and production of further ionization by fluorescence.

CORRELATION BETWEEN THE RATIO FOR THE CHARACTERISTIC LINES $I_A/I(A) = C$ AND THE CONCENTRATION C_A (CASE OF CONSTANT EFFICIENCY R)

Hypothesis: The efficiency of the X-ray emission remains constant regardless of the depth z in the anticathode. In thick anticathodes the electron intensity varies according to Lenard's law [6] as follows:

$$I_e(z) = I_0 e^{-\sigma \rho z}$$

where I_0 is the incident electron intensity, ρ the density and z the depth of the anticathode; σ is largely independent of the composition of the alloy and only depends on the accelerating voltage of the incident beam $\sigma = \text{const.}/U^2$.

Since at 30 kV: $\sigma_{30\text{kV}} = 1750$

$$\sigma = 1750 \left(\frac{30}{U} \right)^2 \quad \begin{cases} U : \text{in kV} \\ \sigma : \text{in cgs} \end{cases}$$

In order to simplify the calculations let us consider the case of a binary alloy

$$\rho = m_A + m_c$$

where m_A = is the mass of pilot element per unit of alloy volume

m_c = is the mass of the additional element per unit of alloy volume

A_A and Z_A = are, respectively, the atomic weight and atomic number of the element A

The intensity I of the characteristic line in the direction of the emergence θ of the X-rays can then be expressed, as follows

$$I_A = K A_A^{m_A} N_0 \int_0^\infty e^{-\sigma \rho z} e^{-\mu/\rho \csc \theta \rho z} dz \quad (1)$$

where K depends only on U/U_k (ratio of acceleration voltage and critical excitation voltage); $N m_A/A_A$ is the number of atoms T per unit of volume; μ/ρ the mass absorption coefficient of the characteristic line T in the alloy analyzed.

Let us assume that $\mu/\rho \csc \theta = \chi$, χ (alloy) being a linear combination of χ_T (standard) and χ_c (additional element).

$$\chi = C_A \chi_A + (1 - C_A) \chi_C \quad \left\{ \begin{array}{l} \chi_A = \frac{\mu}{\rho} \frac{\mu_{\text{line } A}}{\mu_{\text{in } A}} \text{ cosec } \theta \\ \chi_C = \frac{\mu}{\rho} \frac{\mu_{\text{line } A}}{\mu_{\text{in } C}} \text{ cosec } \theta \end{array} \right.$$

By substituting and integrating the second member of equation 1 we obtain:

$$I_A = \frac{KI_0 N}{A_A} \frac{1}{\sigma + \chi} \frac{m_A}{\rho}$$

It should be noted that m_A/ρ is the same as the concentration of the element A in the alloy, hence

$$I_A = \frac{KI_0 N}{A_A} \frac{C_A}{\sigma + \chi} \quad ; \quad I(A) = \frac{KI_0 N}{A_A} \frac{1}{\sigma + \chi_A}$$

and the ratio of the intensity of the X-rays in alloy and standard

becomes
$$\frac{I_A}{I(A)} = C = C_A \frac{\sigma + \chi_A}{\sigma + \chi}$$

i.e.,
$$\Delta C = C_A - C = C \left[\frac{\sigma + \chi}{\sigma + \chi_A} - 1 \right]$$

Allowing for the relation

$$\chi = C_A \chi_A + (1 - C_A) \chi_C$$

we obtain

$$\Delta C = C(1 - C_A) \frac{\chi_C - \chi_A}{\sigma + \chi_A}$$

By carrying out the approximation $C \cong C_A$ in this correction formula, we have:

$$\Delta C = C(1 - C) \frac{\chi_C - \chi_A}{\sigma + \chi_A} \quad (3)$$

CORRELATION BETWEEN THE RATIO OF THE INTENSITIES AND THE CONCENTRATION C_A (CASE OF VARIABLE EFFICIENCY R)

In contrast to the hypothesis put forward in the last section, account is taken here of the diffusion of the electrons as a function of the penetration depth, which directly causes the efficiency of a dz layer to increase as a function of the penetration depth. It is assumed, however, that the ratio U/U_K is sufficiently large to ensure that the ionization cross section K remains virtually constant.

Calculations of the Efficiency $R(\rho z)$ of a Layer as a Function of the Penetration Depth

As the electrons penetrate the metal they deviate from their original course. Let δ be the angle which they form with the normal on the test sample (it is assumed that the electrons enter the sample perpendicularly).

The ionization caused by an electron in a dz layer increases by a factor of $1/\cos \delta$, since it is assumed that the number of ionizations increase with the path length and that the path length ds is equal to $dz/\cos \delta$ in the dz layer. This relation does not apply, however, when $\delta \rightarrow \pi/2$, and it is indeed physically inconceivable that the path length should become infinite when $\delta = \pi/2$. The fact that the electrons deviate from their original course results in an increase in the efficiency $R(\rho z)$ of a dz layer as the penetration depth increases.

Let us calculate the efficiency $R(\rho z)$ as a function of the penetration depth z . Calculation of the efficiency amounts to calculation of the mean value of $1/\cos \delta$ for all electrons.

The distribution of the deviation angle as a function of the penetration depth is provided by Bothe's law:

$$F(\delta) = \frac{dN/N_0}{d\Omega} = \frac{1}{2\pi\Delta^2} e^{-\delta^2/2\Delta^2} \quad (4)$$

- where $F(\delta) = \frac{dN/N_0}{d\Omega}$ = the intensity distribution for a solid angle $d\Omega$
 $\frac{dN}{N_0}$ = the part dN of N_0 electrons scattered into the solid angle $d\Omega$
 δ = the scattering angle
 Δ = the most probable scattering angle

The most probable scattering angle is given by:

$$\Delta = \frac{800}{U} \frac{U + 511}{U + 1022} Z \sqrt{\frac{\rho z}{A}} \quad (5)$$

where

U = the energy of the incident electrons in keV

Z = the atomic number

ρ = the specific gravity (g/cm³)

A = the atomic weight

z = the thickness of the layer (cm)

or, if U is sufficiently low (0-30 keV)

$$\Delta = \frac{400}{U} Z \sqrt{\frac{\rho z}{A}} \quad (6)$$

Let $k = 400/U$ then

The mean value of $1/\cos \delta$ is given by:

$$R(\rho z) = \frac{1}{2\pi\Delta^2} \int_0^\infty e^{-(\delta^2/2\Delta^2)} \cdot \frac{1}{\cos \delta} \cdot 2\pi \sin \delta d\delta$$

and with

$$\operatorname{tg} \delta \cong \delta + \frac{\delta^3}{3} + \dots$$

we obtain

$$R(\rho z) = 1 + \frac{2k^2}{3} \cdot \frac{Z^2}{A} \rho z \quad (7)$$

Let us further assume that when $\Delta = \pi/4$ the electrons are completely diffuse. R remains constant above this value. The efficiency is thus expressed as follows:

$$\text{with } \rho z = \rho z_1 \quad \text{for } \Delta = \frac{\pi}{4}$$

$$\text{when } 0 < \rho z < \rho z_1 = \frac{\pi^2}{16} \cdot \frac{A}{Z^2} \frac{U^2}{400^2} \quad R(\rho z) = 1 + \frac{2k^2}{3} \cdot \frac{Z^2}{A} \cdot \rho z \quad (8)$$

$$\text{when } \rho z_1 < \rho z \quad R(\rho z) = 1 + \frac{\pi^2}{24} \quad (9)$$

Remarks

The expression for $R(\rho z)$ was obtained by the approximation $\operatorname{tg} \delta = \delta + \delta^3/3$, which is, strictly speaking, only applicable for low

values of δ . The exact integral diverges, however, when $\delta \rightarrow \pi/2$, because the efficiency $dz/\cos \delta$ diverges. The use of this approximation for larger angles sets a limit to the path length $ds = dz/\cos \delta$, but this limit remains arbitrary.

The selection of $\Delta = \pi/4$ is also arbitrary.

Bothe's law only applies to small angles.

From the three remarks above, it can be seen that $R(\rho z)$ can only be a rough approximation. According to Bothe $k^2 = 400^2/U^2$; Lenard's law gives $\sigma = 1750(30/U)^2$. By elimination of U between these relations we obtain:

$$\sigma = g, 85 k^2$$

$$\text{and} \quad \rho z_1 = 6, 14 \frac{A}{Z^2} \cdot \frac{1}{\sigma} = \frac{\xi}{\sigma}$$

$$\text{with} \quad \xi = 6, 14 \frac{A}{Z^2}$$

Calculation of the Concentration as a Function of $I_A/I(A)$

The contribution of the dz layer to the X-ray intensity is equal to

$$dI_A = K \cdot n \cdot R(\rho z) I_0 e^{-\sigma \rho z} e^{-\chi \rho z} dz \quad (10)$$

where

$$\begin{aligned} K &= \text{the ionization cross section} \\ n &= \text{the number of atoms } A \text{ per cm}^3 \\ R(\rho z) &= \text{the efficiency of the } dz \text{ layer} \\ I_0 e^{-\sigma \rho z} &= \text{the electron intensity} \\ \chi &= \mu/\rho \operatorname{cosec} \theta \end{aligned}$$

We again assume that for the incident electrons U/U_K is equal to $3 \sim 5$, so that K can be regarded as a constant. A similar relation applies in the case of the standard.

$$dI(A) = K n_T R_T(\rho z) I_0 e^{-\sigma_T \rho z} e^{-\chi_T \rho z} dz \quad (11)$$

(The index T refers to the standard.)

The number of atoms A per cm^3 for the alloy and the standard are, respectively:

$$n = \frac{\rho C_A N}{A} \quad n_T = \frac{\rho_T N}{A}$$

After integration we obtain:

for the alloy

$$I_A = \frac{K\rho C_A N_0}{A\rho} \cdot \int_0^\infty R(\rho z) e^{-(\sigma+\chi)\rho z} d\rho z = F(\chi, \xi) \quad (12)$$

and for the standard

$$I(A) = \frac{K\rho_T N_0}{A\rho_T} \int_0^\infty R_T(\rho z) e^{-(\sigma+\chi)\rho_T z} d\rho_T z = F(\chi_T, \xi_T) \quad (13)$$

and finally

$$\frac{I_A}{I(A)} = C_A \frac{F(\chi, \xi)}{F(\chi_T, \xi_T)} \quad (14)$$

In this correction formula $F(\chi, \xi) \neq F(\chi_T, \xi_T)$ since an atomic number correction has already been carried out in $R(\rho z)$. After substitution of $R(\rho z)$ in I_A and $R_T(\rho z)$ in $I(A)$ we obtain:

$$F(\chi, \xi) = \frac{1}{\sigma + \chi} \left[1 + \frac{\pi^2}{24} e^{-(\sigma + \chi)\xi/\sigma} \right] + \frac{\pi^2}{24} \cdot \frac{\sigma}{\xi(\sigma + \chi)^2} \left[1 - \frac{1}{[1 + (1 + \chi/\sigma)\xi]^{-\alpha + \chi/\sigma}} \right]$$

with

$$\xi_A = 6,15 \frac{A}{Z^2} \quad \xi = \sum C_i \xi_i$$

COMPARISON OF THE THEORETICAL RESULTS WITH CASTAING'S EXPERIMENTAL CURVES

By means of experiments on cylinders Castaing succeeded in recording the function $F(\chi, \xi)/F(0, \xi)$ as a function of χ ($\chi = \mu/\sigma \cos \theta$ varies because of the variation in the angle emergence θ). Figures 1 and 2 show Castaing's experimental curves together with our own results. Concurrence is good for high acceleration voltages, but insufficient for voltages below 15 kV.

CONCLUSIONS

1. The correction formulas can be used with a good degree of approximation as long as the acceleration voltage is higher than 15 kV.

$$C_A = \frac{I_A}{I(A)} \cdot \frac{F(\chi_T, \xi_T)}{F(\chi, \xi)}$$

where $F(\chi, \xi)$ is given by (15)

with $\chi = \sum C_i \chi_i$

and $\xi = \sum C_i \xi_i$

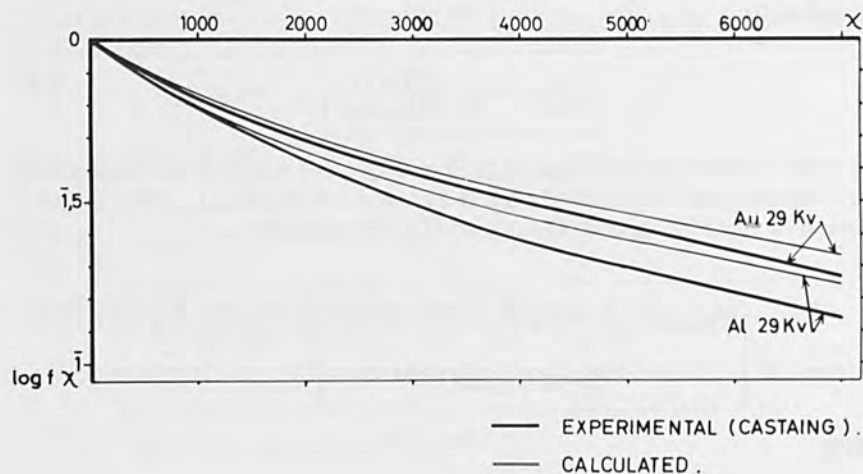


Figure 1.

2. Atomic Number Corrections.

(a) The fact that ξ is dependent on Z^2/A automatically causes and atomic number correction. This is a correction which is included because each element has a different ρZ_1 which is the depth at which diffusion is considered to be complete.

(b) In setting up our formulas we assumed that Lenard's coefficient σ is independent of the atomic number, but this is only an approximation. In other words we must carry out a second atomic number correction. After conversion of our formula in order to make proper allowance for ($\sigma_A \neq \sigma_B$), we have:

$$C_A = \frac{I_A}{I(A)} \frac{F(\chi_T, \xi_T)}{F(\chi, \xi)} = \frac{I_A}{I(A)} \frac{\frac{F(\chi_T, \xi_T)}{F(0, \xi_T)} F(0, \xi_T)}{\frac{F(\chi, \xi)}{F(0, \xi)} F(0, \xi)} = \frac{I_A}{I(A)} \frac{f(\chi_T, \xi_T)}{f(\chi, \xi)} \frac{F(0, \xi_T)}{F(0, \xi)} \quad (16)$$

$A \equiv T$ (the standard T is the pure element A).

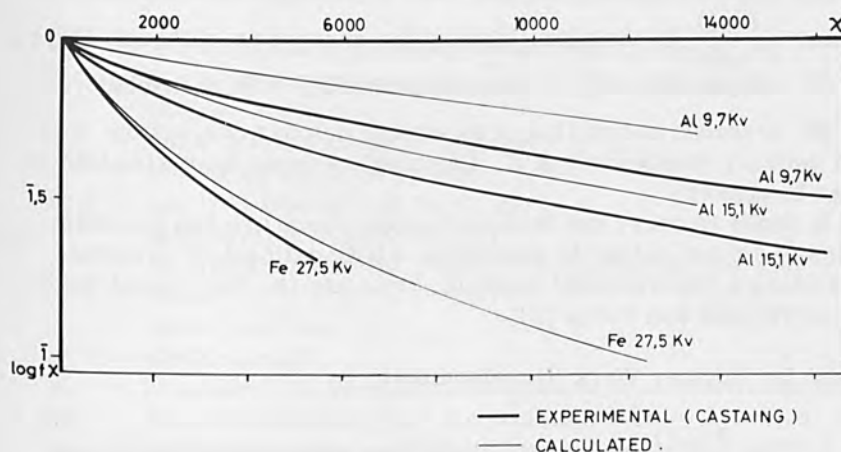


Figure 2.

where

$$F(0, \xi_A) = \frac{1}{\sigma_A} \left\{ 1 + \frac{\pi^2}{24} e^{-\xi_A} + \frac{1}{\xi_A} \cdot \frac{\pi^2}{24} [1 - (1 + \xi_A) e^{-\xi_A}] \right\} = \frac{1}{\sigma_A} \cdot \varphi(\xi_A)$$

and

$$F(0, \xi) = \frac{1}{\sum C_i \sigma_i} \varphi(\xi)$$

we calculate $\frac{f(\chi_i \xi)}{f(\chi_T \xi_T)}$ with a mean value of σ , but in calculating

$\frac{F(0, \xi_T)}{F(0, \xi)}$ we use the actual values of σ , obtaining thus:

$$\frac{I_A}{I(A)} = \frac{f(\chi_i \xi)}{f(\chi_T, \xi_T)} \cdot \frac{\varphi(\xi)}{\varphi(\xi_A)} \cdot \frac{\sigma_A C_A}{\sigma_A C_A + \sigma_B C_B} \cong \frac{F(\chi_i \xi)}{F(\chi_T, \xi_T)} \cdot \frac{\sigma_A C_A}{\sigma_A C_A + \sigma_B C_B} \quad (17)$$

The second factor of this formula has the same structure as the second approximation of Castaing and makes proper allowance for penetration effects.

If we want to include in this correction the backscatter effect also, we obtain:

$$\boxed{\frac{I_A}{I(A)} \cong \frac{F(\chi, \xi)}{F(\chi_A \xi_A)} \frac{\alpha_A C_A}{\sum \alpha_i C_i}} \quad (18)$$

α is in this case different from σ .

In this way two corrections are seen to emerge: (1) The first factor $\frac{F(\chi\xi)}{F(\chi_A\xi_A)}$ is the actual absorption correction, although $F(\chi, \xi,)$ is not independent of ξ , or in other words the atomic number.

(2) A second correction is an atomic number correction: σ is not entirely independent of Z . Castaing's second approximation is seen to appear.

In order to carry out this correction, there are two possible ways of action: either to determine α with calibration samples (Castaing's experimental method), or to use the theoretical method of Thomas and Poole [3].

Need for Further Work (Development)

1. The $\frac{F(\chi_i\xi)}{F(0,\xi)}$ concur well with those experimentally obtained by Castaing, as long as the acceleration voltage is in excess of 15 kV. Below this value the concurrence is insufficient, probably because our function $R(\rho z)$ takes too little account of the diffusion.

In order to check this we compared our ionization function with that obtained by Castaing and Descamps. Bothe's formula does in fact underestimate the increase in efficiency due to diffusion. These formulas might possibly be improved by adapting the $R(\rho z)$ curve to the experimental curves.

2. In our deductions it was assumed that the ionization cross section K remains constant with the penetration depth, which is, strictly speaking, not true, because the energy decreases with the penetration depth, and K is a function of the energy of the electrons.

It is our aim to examine the influence of this factor by means of numerical calculations. This influence will be especially important if U/U_K is less than 3 (major variation in the ionization cross section when $U/U_K < 3$).

3. However great the need for improving correction formulas, a satisfactory solution to the correction problem will only be possible when sufficient experimental results are made available concerning, e.g.: The accurate determination of absorption coefficients for soft X-rays, systematic determination of α -coefficients, etc.

LIST OF SYMBOLS

- I_A = X-ray intensity produced in an alloy (A line).
 $I(A)$ = X-ray intensity produced in a standard specimen (A line).
 C_A = mass concentration of element A in alloy.
 I_0 = impinging electron intensity ($z=0$)
 $I_e(z)$ = electron intensity at depth (z).
 U = acceleration voltage (in kV).
 σ = Lenard's coefficient (absorption coefficient for electrons).
 Z = atomic number.
 A = atomic weight.
 N = avogadro's number.
 m_A = mass of element A per unit volume.
 μ/ρ = mass absorption coefficient for X-rays.
 θ = angle of emergence of X-rays.
 K = ionization constant.
 δ = scattering angle of the electrons at depth z .
 Δ = most probable scattering angle at depth z .
 z = depth in specimen.
 z_1 = depth in specimen at which diffusion is complete.
 α = coefficient of Castaing which takes account of penetration effects and of backscattering effects.
 ξ = $6, 15 A/Z^2$.
 χ = $\mu/\rho \text{ cosec}$.

APPENDIX

In order to test our method we tried to correct some experimental results obtained by Ziebold and Ogilvie (see table below).

Measured and Corrected ($I/I_0/C$) for Binary Alloys

System	Line	True Comp.	Meas.	Absorption	Absorption + Electronic
Ni-Pt	Ni-K α	29.7%	1.027	1.222	1.057
		6.5%	1.031	1.300	0.998
Ag-Zn	Zn-K α	14.6%	0.877	1.203	1.103
Ag-Au	Ag-L α	40.6%	0.404	1.636	1.597
Ta-Os	Ta-L α	68.9%	1.067	1.004	1.003
W-Ir	W-L α	5.7%	1.105	0.988	1.034
UO ₂	U-L α	88.1%	0.938	1.008	0.985
UC	U-L α	95.2%	0.971	1.000	0.998

For atomic number corrections, the data of Poole and Thomas were used. Concurrence is good except for the system Ag-Au and in a lesser degree for the system Ag-Zn.

This discrepancy in the Ag-Au is probably due to the fact that the absorption coefficient is not known precisely for Ag $L\alpha$ radiation in gold.

REFERENCES

1. R. Castaing, Thèse de doctorat. Publication Onéra No. 55 (1952).
2. Henoc, "Contribution à la microanalyse par sonde électronique," Ministère des PTT, Etude No. 655, PCM.
3. D. M. Poole and P. M. Thomas, "Quantitative electron-probe microanalysis," Report AERE, R, 3815.
4. D. B. Wittry, J. of App. Phys. 29, 11 (1958).
5. R. Castaing and J. Descamps, "Recherches aéronautiques," No. 63 (1958).
6. M. von Ardenne, "Tabellen der Elektronenphysik, Ionenphysik und Uebermikroskopie—Band I," VEB. Deutscher Verlag der Wissenschaften, Berlin, 1956.
7. R. Castaing and J. Descamps, J. Phys. Radium 16, 304, 307 (1955).
8. R. Castaing, "Advances in Electronics and Electron Physics," Vol. XIII, 318-386 (1960).
9. P. Duncumb and Shields, Brit. J. Appl. Phys. 14, 10 (1963).
10. G. D. Archard and T. Mulvey, Brit. J. Appl. Phys. 14, 10 (1963).

DETECTION LIMITS OF ELECTRON MICROPROBE ANALYSIS—APPLICATION OF "INCREASED SENSITIVITY METHOD" IN METALLURGY

Since the development of electron microprobe analysis by Castaing [1] in 1951, the interest in metallurgy of local microanalysis of volumes of 10^{-12} ccm was at once appreciated and a considerable amount of experience has been obtained leading to a broadening of its field of application. For elementary, highly localized, microanalysis of elements with atomic numbers higher than 11, detection limits in the order of 0.5 wt% have been claimed, but the involved statistical principles have never been clearly defined. Based on theoretical considerations and experimental confirmations, the scope of this paper is to demonstrate that the method of X-ray emission microanalysis may be extended to trace analysis and therefore become a powerful evaluation method for a number of metallurgical problems (and in other domains as well) at present inaccessible, if appropriate experimental conditions are chosen.

THEORETICAL BASIS OF TRACE DETERMINATIONS

Quantitative microprobe analysis is based on the comparison of the emission I_A of the sample in the characteristic radiation A with the emission $I(A)$ in the same line and under the same experimental conditions of a reference target, consisting of either the pure

*Institut für Angewandte Reaktorphysik, Kernforschungszentrum, Karlsruhe, West Germany.

element *A* or, as in the proposed approach, a sample with low but known concentration of *A*. The mass concentration C_X of element *A* in the region analyzed is supplied by Castaing's [1] first approximation:

$$\frac{I_A}{I(A)} = c_X \cdot c_c \quad (1)$$

Considering that for the determination of low concentrations, X-ray absorption in the sample and secondary fluorescence emission effects are negligible, assuming that we measure the low concentration calibration C_c without error, that we determine the mean number of impulses by making infinite measurements, we obtain:

$$C_c \cdot C_X = \frac{\bar{N}_X - \bar{N}_{Bx}}{\bar{N}_c - \bar{N}_{Bc}} = N_X \frac{1}{\bar{N}_c - \bar{N}_{Bc}} - \frac{\bar{N}_{Bx}}{\bar{N}_c - \bar{N}_{Bc}} \quad (2)$$

For the range of low concentrations, but not for calibration with a pure element, N_{Bx} is not very different from N_{Bc} and we obtain the following linear relationship (Fig. 1):

$$C_c \cdot C_X = \alpha \bar{N}_X - \beta$$

$$\alpha = \frac{1}{\bar{N}_c - \bar{N}_B} \quad \text{and} \quad \beta = \frac{\bar{N}_B}{\bar{N}_c - \bar{N}_B} \quad (3)$$

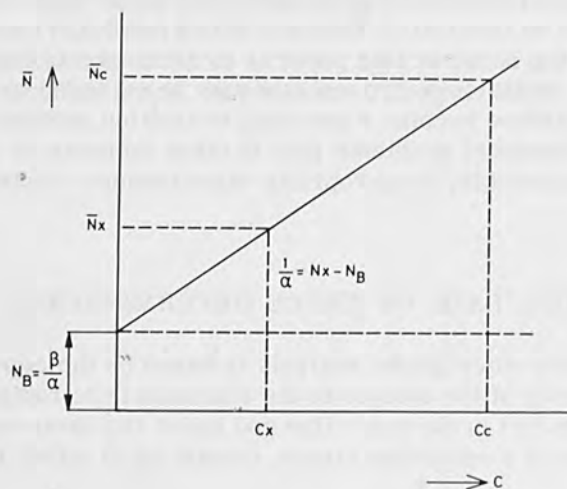


Fig. 1. Calibration curve for trace amounts determination.

- $\bar{N}_c$ = impulse counts for calibration concentration
 $\bar{N}_x$ = counts for trace concentration
 C_x = unknown concentration to be calculated
 $\bar{N}_{Bc}$ = counts for the background on calibration standard
 $\bar{N}_{Bx}$ = counts for the background specimen

The calculation of the concentration C_x from relation 2 if C_x tends to zero is done by means of the probability theory and laws of error[2].

$$\lim (\Delta C_x) = \sqrt{2} \cdot c_c \cdot (\bar{N}_c - \bar{N}_B)^{-1} (\Delta \bar{N}_B) \quad (4)$$

- if $C_x \rightarrow 0$
 $r^{-x} = \bar{N}_x / \Delta_t$ r^{-x} = mean counting rate for random sample
 Δ_t = time interval for one measurement
 $T = n \cdot \Delta_t$ T = time interval for the whole random sample
 n = number of measurements on the random sample

We get a detection threshold C_{DT} (concentration of an element in a matrix which still can be detected by the analytical method) of

$$C_{DT} = \frac{\sqrt{2} \cdot t_s \cdot \sqrt{r^{-x}_B} \cdot c_c}{\sqrt{T} (r^{-x}_c - r^{-x}_B)} \quad (5)$$

t_s = factor of "student" distribution depending only on number of measurements on the random sample (tabulated)

As detection limit C_{DL} , we define that concentration of an element, which still can be determined with a given accuracy, i.e., the relative error of the concentration C , shall not pass a value "a" in percentage.

$$\text{then } C_{DL} = \frac{100}{a} \cdot \frac{t_s \cdot \sqrt{2} \cdot \sqrt{r^{-x}_B} \cdot C_c}{\sqrt{T} (r^{-x}_c - r^{-x}_B)} \quad (6)$$

The fundamentally important parameter—the peak to background ratio of the analyzed characteristic radiation—is defined by the particular spectrometer setup.

For statistical reasons, the detection limit decreases with increasing total time interval "T" and increasing number of measurements within the same time interval T for a single point analysis.

EXPERIMENTAL PROCEDURE

The formation rate of the well-known contamination spot on the sample at the point of impact of the electron beam limits the counting intervals on one point in conventional microanalysis to about 50 to 100 sec per measurement.

As the detection limit decreases with longer time intervals, the formation of a contamination point was prevented by introducing through a high-precision valve and a capillary of nonmagnetic material a gaseous flow directed onto the electron-bombarded region, similar to experiences by Castaing [3] involving studies on low accelerated electron beams.

Even extremely small flow rates of air through the capillary (0.025 mg/sec) caused considerable reductions in lifetime of the electron emitting filament. By this fact, the reproducibility of the initial measurements was seriously restricted, as the change of the filament necessitates a complete focusing of the electron optical system. This drawback was eliminated by the use of an inert gas flow. The prevention of the contamination spot by the presence of a localized gaseous atmosphere at the point of impact of the electron beam is not due to an oxidation phenomenon, but almost certainly an ion-beam etching effect, as demonstrated by Quataert [4] using several inert gas atmospheres. The etching effect increased regularly with increasing atomic weight of the gas probe. Helium was rather ineffective when used to prevent contamination, whereas argon-bombarded metallic specimens exhibited perfect surfaces, even after an exposure of 10^4 sec. As the lifetime of filaments is not influenced by these small quantities of argon, this gaseous probe was exclusively adopted.

The upper time limit for the measurement of one concentration was fixed by economical considerations to 9000 sec. Then the gas valve was closed and the analyzed point localized by vacuum electron bombardment for about 100 sec. The dependence of the student distribution factor from the number of measurements shows that for more than 15 measurements, there is no more essential decrease for the detection threshold.

It was shown that the instrument stability is highly stable even for these long-time intervals, and that the fluctuations of a random sample are part of a Gaussian population. Random parasitic impulses coming from the electric mains were subtracted by an independent pulse height analyzer.

APPLICATIONS

Determination of Low Solid Solubility Limits in Alloys

The indirect metallic bonding of titanium-plated uranium to a magnox can has been studied in view of an improvement of fuel element fabrication of gas-cooled, graphite-moderated reactors.

For U-Ti interdiffusion, extensive studies have been reported [5], [6], and some complementary aspects will be outlined, but no information is available on the nature of the bonding between titanium and the magnox can.

After different annealings a perfect mechanical adherence of the two components was observed. However, the presence of the necessary diffusion bonding could not be observed by optical or electron microscopy, or by even conventional microprobe techniques.

By the described increased sensitivity method, it was possible to deduce that the limit of solid solubility of titanium in magnox L 80 is about 0.4 wt% at 550°C and about 0.5 wt% at 600°C. The corresponding concentration curves of titanium in magnox Al 80 and magnesium in titanium as a function of penetration are shown in Fig. 2.

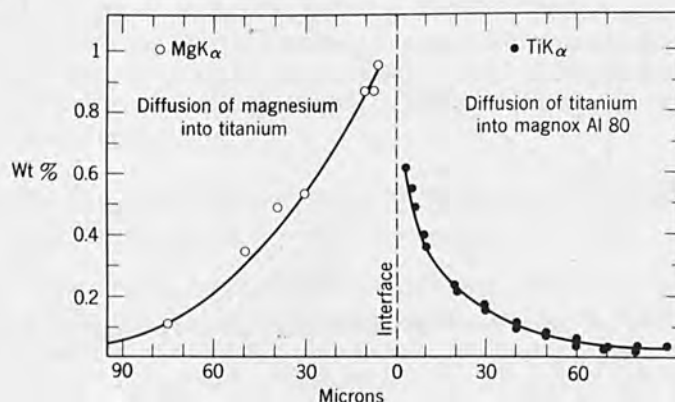


Fig. 2. Mg and Ti diffusion in Magnox Al 80 at 600°C.

The stability of the involved instruments has been checked for this specific application by statistical evaluation of the registered

impulse rates; numerical examples are described in [2] and demonstrate that the detection threshold is in the range of 50-100 ppm. The agreement of the experimental values with the basic linear relationship of equation 2 and Fig. 1 is excellent and is shown in Fig. 3 by the diagram of mean registered impulse rate on the specimen (N_x) versus measured concentration (c_x). It must be outlined however, that for these long time measurements, variations of experimental conditions may occur; for example, new electron optical alignment after filament exchange, fluctuations in the electrical mains, etc. The measured concentration c_x is essentially independent of these fluctuations, whereas the intensity rate of the radiation emitted by the sole specimen N_x may vary seriously. For this reason, the possible drifts were controlled before and after every series of determinations by intensity measurements of the characteristic radiation emitted by a pure element during 600 sec. If

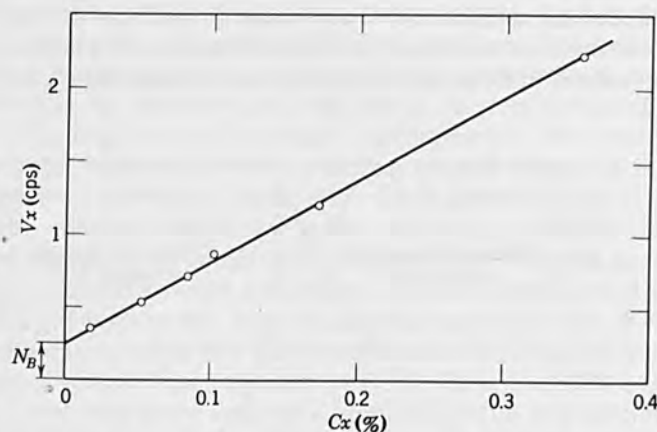


Fig. 3. Experimental calibration curve (Ti diffusion in Magnox).

necessary, a linear interpolation of the different N_x values is applied.

A similar study concerned the diffusion bonding of the different parts of a SAP (sintered aluminium product) fuel element can.

A silver film, 5μ thick, was evaporated onto the endcap before introduction into the tubular can. Then the assembly was vacuum annealed for one hour at 600°C . Microprobe analysis across the diffusion zone permitted us to plot the concentration versus penetration curve of Fig. 4, and demonstrate that an excellent metallic

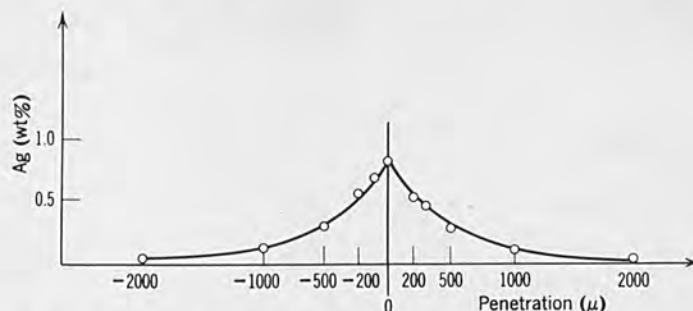


Fig. 4. Diffusion bonding of SAP by Ag diffusion S_μ Ag annealing one hour at 600°C .

bonding without brittle intermetallics is obtained. Maximum solid solubility of silver in SAP at 600°C is about 0.7 wt%.

It is interesting, furthermore, to notice the pronounced diffusivity into the endcap (positive penetration). This phenomenon corresponds to the more intimate initial contact of the silver layer, plated on the endcap.

Structural Microanalysis and Study of Intermetallic Diffusion Layer Formation

In uranium titanium diffusion couples, the diffusion mechanism is highly sensitive to the impurity level of the commercial purity uranium (carbon content 300-400 ppm). In the high-temperature range, owing to the relative diffusion rates of carbon and titanium in uranium, titanium carbide formation is localized exclusively at the original interface, since the quantity of carbon which migrates to the interface is sufficient to trap, in the form of thermodynamically very stable titanium carbide, the major part of titanium in uranium. Microprobe and microhardness results of uranium-titanium diffusion at 960°C are given in Fig. 5. The small zone of

carbides is seen on the micrograph and corresponds to the peak in the microhardness curve.

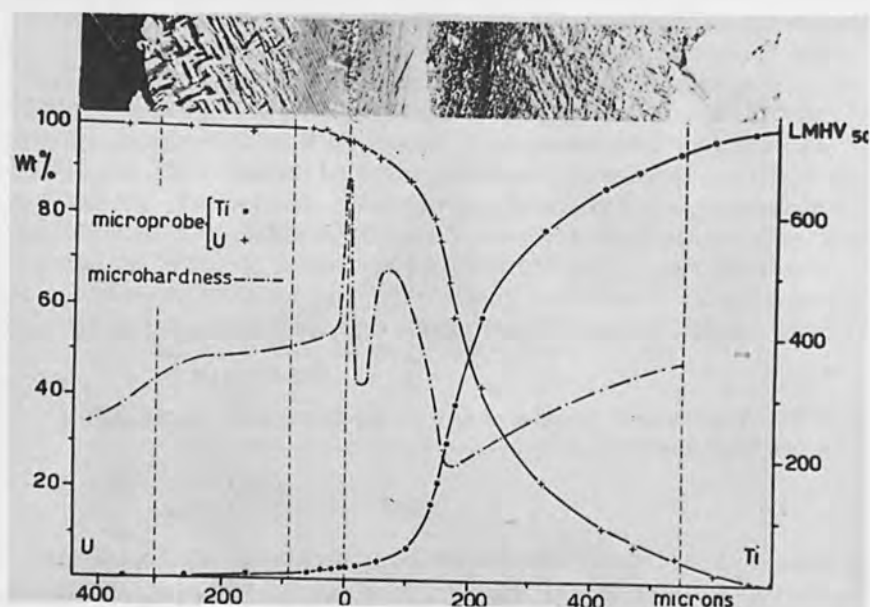


Fig. 5. U-Ti diffusion at 960° C.

At low-temperature ranges, the quantity of carbon arriving at the interface is insufficient to combine with all of the titanium and so the latter can diffuse freely into the uranium. The TiC, in this case, is distributed inside the zone of diffusion in uranium; their frequency decreases with distance from the interface. Uranium-titanium diffusion mechanism at 735°C is illustrated in Fig. 6.

Intergranular uranium diffusion into the titanium has not been observed in preceding studies. Its presence is, however, clearly revealed by the absorbed electron picture of Fig. 7 and the corresponding optical micrograph (Fig. 8) (polarized illumination, electrolytic etch). The decrease of uranium concentration in the grain boundaries was followed from 0.7 wt % to about 0.2 wt %. The cored structured, gray β retained titanium phase ("Christmas tree") revealed by the absorbed electron picture, corresponds to an

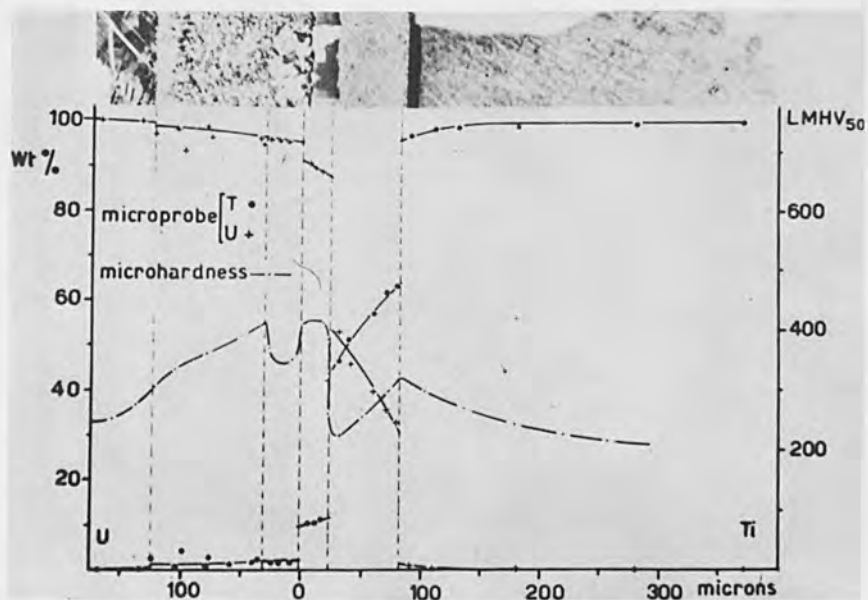


Fig. 6. U-Ti diffusion at 735° C.

impurity concentration of about 1200 ppm of iron.

Microanalysis of Radiation Damage in Irradiated, Pyrocarbon-Coated Uranium-Zirconium Monocarbides

The relatively high continuous background in microprobe analysis of irradiated materials causes a serious enhancement of the detection threshold. For quantitative determinations, an increase of the counting intervals is highly desirable.

In high-temperature, stable, fission product retaining, pyrocarbon-coated, uranium-zirconium monocarbide compacts, used as nuclear fuels for high-temperature gas-cooled reactors or thermionic converters, irradiation damage at high burnups, and at temperatures ranging from 1200°C to about 2000°C, shows a typical "spearhead" attack.

Depending on the structure of the first layer of the pyrocarbon coating, which can be either radially oriented columnar grains or loose carbon black, the uranium migration in the coating proceeds intergranularly or by volume diffusion, forming a $UC_{2+x} + C$ buffer layer.



Fig. 7. Uranium intergranular diffusion in Ti optical image.

A typical example is shown in Fig. 9, representing a pyrocarbon-coated, melted UC-ZrC particle, irradiated at about 1850°C to a burnup of 16% F.I.F.A.

The interface formed by the spearhead and the pyrocarbon contains about 0.1-0.2 wt % uranium and traces of barium. The geometrical form of the radiation damage can be logically explained by uranium intergranular diffusion and subsequent recoil action.

CONCLUSION

The detection threshold of quantitative microprobe analysis may be decreased to the range of 50 to 100 ppm, by the described method and statistical evaluation.

The preceding applications have been selected to illustrate both the reliability of quantitative trace analysis and the value of this new extension of microprobe technique to problems which are very difficult to resolve by conventional methods.

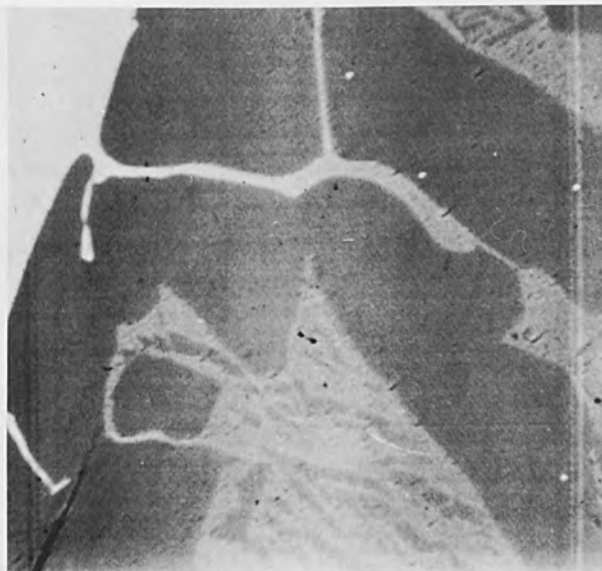


Fig. 8. Sample of Fig. 7, electron absorbed picture.

REFERENCES

1. R. Castaing, Thesis Paris, ONERA No. 55, 1951.
2. K. Täffner and R. Theisen, Proc. International Symp. on Electron Microprobe of Nuclear Materials, Ispra 1963, Euratom Report 1964.
3. R. Castaing, C. Rend. Acad. Sci. 238, 1506 (1954).
4. D. Quataert, private communication, 1963.
5. Y. Adda and J. Philibert, "Colloque sur la diffusion," North Holland Publishers, 1959.
6. M. Bastelli et al., Geneva Conference, 1964.

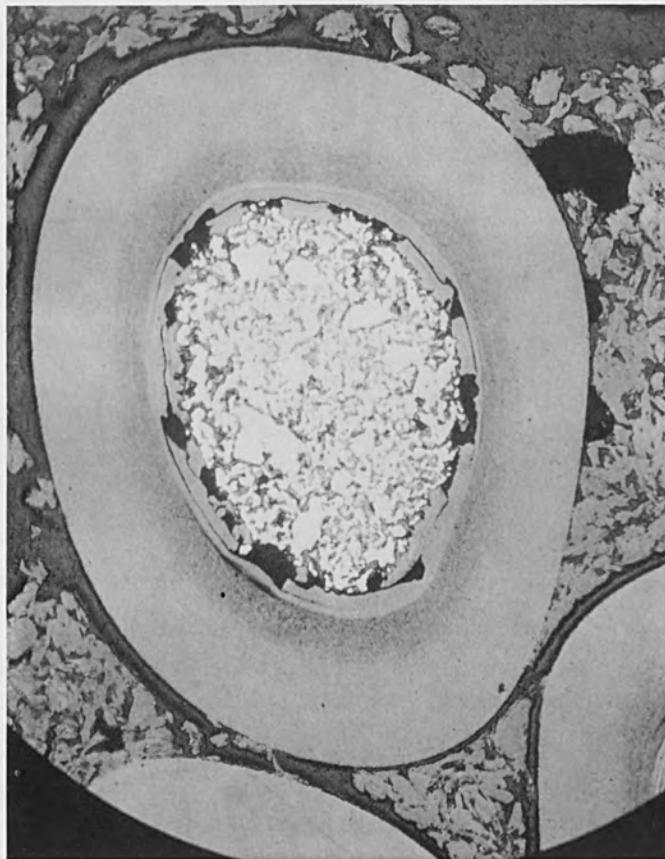


Fig. 9. Radiation damage in pyrocarbon-coated UC arc after 16% FIFA burn-up at 1850° C.

PRACTICAL METHODS FOR EXPERIMENTALLY CALIBRATED CONCENTRATION DETERMINATION IN THE ELECTRON-BEAM MICROANALYZER

A number of empirical or theoretical methods have been proposed to convert experimentally determined X-ray intensity ratios to concentration. A larger number of investigations have tested the accuracy of these methods and have indicated that only limited agreement between calculated and experimentally determined values can be obtained.

Unfortunately, it is now quite clear that for many studies for which highly accurate concentration determinations are required alloy standards must be employed. This would be the case for most diffusion or phase diagram studies, for instance.

The use of standard alloys to determine a working calibration curve is fairly straightforward. Exactly what constitutes an alloy standard is, however, somewhat less straightforward. It seems that the general consensus of opinion is that an alloy standard is required to be at least single phase and completely homogeneous over micron distances, and its chemistry should be precisely characterized.

It is the purpose of this paper to show that an alloy standard need not be either single phase or even extremely homogeneous. In essence, it is quite possible to employ even segregated multi-phase alloys as standards. It is only necessary, of course, that the average alloy chemistry be known.

This result, which has been proved experimentally, is of extreme importance. Very often the type of alloy standards which have been

*Advanced Metals Research Corporation, Burlington, Mass.

previously demanded are quite impossible to prepare, either practically or theoretically. Many alloy systems, whether binary, ternary, or quaternary, exhibit terminal solid solutions or intermediate phases of only limited solubility, and much of the possible range of chemistry is represented by multiphase alloys. In addition, the attainment of even reasonable homogeneity often requires alternate cold work and annealing treatments which are quite impossible in brittle compounds or alloys. It is, therefore, not only convenient, but in many cases absolutely necessary that less than ideal alloys be employed as standards.

There will be no attempt to justify these conclusions on any theoretical basis. Rather, it will be demonstrated that the conclusions have been justified experimentally. Both single-phase and two-phase alloys of known chemistry, from a number of binary alloy systems, have been employed to construct the usual calibration curves of intensity ratio as a function of concentration. It can then be shown that both the single-phase and two-phase alloys fit the calibration curve (which is drawn using the single-phase alloys only) with essentially the same precision.

In most available instruments, the diameter of the incident electron beam can be increased up to a few hundred microns with no change in incident electron current if the final lens is defocused. If a homogeneous alloy or pure standard is placed beneath the electron beam no change in measured X-ray intensity will be noted, as the beam is defocused as long as the focus of the X-ray spectrometer is not concurrently enlarged beyond the width of its final or receiving slit.

If a two-phase or segregated alloy standard is to be examined, an average X-ray intensity value is obtained utilizing a defocused electron beam. The specimen is translated systematically beneath the electron beam during some point counting interval. The choice of beam size and traverse path is chosen after it can be shown that consecutive runs will reproduce X-ray intensity values within the statistical deviation of the total number of counts collected (or its equivalent for those electronic systems which do not employ pulse counting scalars).

Figure 1 is a plot of normalized RuL_{α} X-ray intensity as a function of concentration in the Ru-Ta system (i.e., the usual calibration curve). It is observed that the average intensity values of the two-phase alloys fit the curve as well as the values from single-phase alloys. The fact that the two-phase alloy values fit calibration curves as well as single-phase alloy values may be shown in a more rigorous fashion, however.

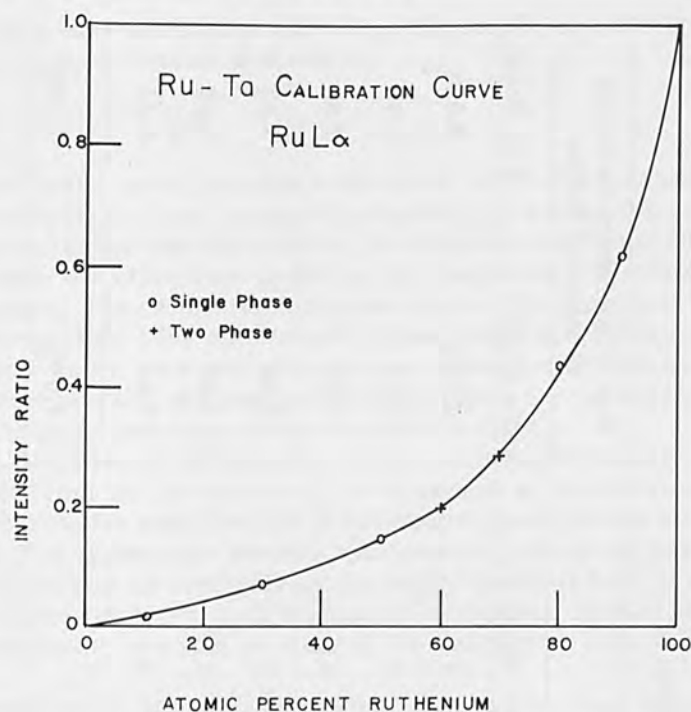


Figure 1.

Table I summarizes the results from six binary alloy systems in which both single-phase and two-phase alloys of known chemistry were available. The characteristic X-ray line from the first element listed for each binary system was employed to construct the calibration curve. Although a curve was actually obtained for each element, the curve which showed the most positive or negative deviation from linearity was chosen for presentation since it was felt that these would provide a more severe test of the applicability of the method. The number of single-phase and two-phase alloys studied for each system is also shown.

Once the intensity ratios were obtained for all of the alloys including the average values for the two-phase alloys, the calibration curves for each system were constructed utilizing the single-phase values only. For this purpose the best root mean square deviation fits to these values were obtained analytically by using the technique suggested by Zeibold and Ogilvie in *Analytical Chemistry*, Vol. 36, No. 2, page 322, 1964.

Table I

Binary System	Number of Alloys		Root Mean Square Deviation of Intensity Ratios	
	Single Phase	Two Phase	Single Phase	Two Phase
Ir-W	3	3	0.80%	0.52%
Rh-W	3	1	2.53	2.75
Pt-W	4	2	1.20	1.85
Ru-W	3	2	1.54	1.52
Pt-Ta	5	2	1.62	1.46
Ru-Ta	5	2	1.39	0.44
			1.07%	1.09%

Briefly each calibration curve is represented as a straight line by employing a function of the form

$$\frac{1-K}{K} = a \frac{1-C}{C} \quad (1)$$

where K is the usual intensity ratio and C is the concentration for each calibration alloy. A single parameter, a , defines the calibration curve and the deviation of each experimental intensity ratio from the value determined by the analytically calculated fit is obtained. The calibration data was found to be expressed very well by equation 1 for the concentrations investigated ranging between 5 and 95 per cent. The root mean square deviations of the experimentally determined intensity ratios for the single-phase alloys is given and these range from 0.8 to 2.5%.

The deviation of the experimentally determined intensity ratios obtained from the two-phase alloys as compared to the ratios calculated from the analytical fit of the single-phase data is also shown. It is quite clear that the two-phase values fit the calibration curves just as precisely as the single-phase values. It should also be noted that, for each system, no systematic shift of the two-phase values to one side or other of the calibration curve was noted.

In conclusion, it can be stated that multiphase alloys will serve as standard calibration alloys if an average X-ray intensity ratio is determined using the method prescribed.

COMPARISON OF THE DIFFUSION OF SEVERAL SUBSTITUTIONAL ELEMENTS IN NICKEL AND IN COBALT

INTRODUCTION

Atomic diffusion influences many solid-state reactions in physical metallurgy. This phenomenon is important, for example, in processes such as precipitation, recrystallization, creep, etc. Many radioactive isotopes now available for products of nuclear reactors have made it possible to study auto- as well as grain-boundary diffusion.

In the case of diffusion with an elevated gradient of chemical composition, the electron-probe microanalyzer has made it possible to easily and accurately determine the coefficients of volume diffusion. Many experiments have been undertaken in order to measure the coefficient of interdiffusion of two metals forming a substitutional solid solution.

Such a study in the case of nickel and cobalt base alloys presents, in addition to a considerable interest from a theoretical standpoint, a decided practical importance due to their extensive industrial application. The substitutional elements such as chromium, molybdenum, tungsten, and vanadium have been selected for study since they are most frequently encountered in these alloys.

The coefficients of interdiffusion have been measured by the use of couples in which one half was constituted by the pure element (nickel or cobalt) and the second half was composed of an al-

*Centre National de Recherches Metallurgiques, Paris, France.

†Research carried out at the CNRM under the joint sponsorship of the "Institut pour l'Encouragement de la Recherche Scientifique dans l'Industrie et l'Agriculture (IRSIA).

loy. The compositions and temperatures selected were such that diffusion took place in a face-centered-cubic lattice.

EXPERIMENTAL METHODS

The alloys were prepared from high purity metals in a vacuum induction furnace. The analysis of the base materials were the following: Co > 99.8%, Ni > 99.95%, Cr > 99.35%, W > 99%, V > 99.8% and Mo > 99.9%. Table I gives the composition of the alloys. After casting, the latter were subjected to an homogenization heat treatment of one hour at 1100°C in a protective atmosphere of argon. The results of the homogenization treatment were verified by both metallographic and electron-probe examination.

Table I

Chemical Composition of the Alloys in Weight Per Cent

Sample	Ni	Co	Cr	W	V	Mo
1		bal	15.2			
2		bal		14.66		
3		bal			14.88	
4		bal				15.16
5	bal		10.3			
6	bal			14.62		
7	bal				14.81	
8	bal					13.80

Preparation of Diffusion Couples. Each couple was composed of two cylindrical specimens 15 mm in diameter. One sectional face of each cylinder was lapped and then polished on wet emery-paper. The cylinders were then pressed and welded together under argon by joule effect. The quality of the welds thus produced was verified with an optical microscope. Studies by the electron probe showed that the initial conditions were not modified by this welding technique.

All diffusion heat treatments were performed in sealed quartz tubes containing an argon atmosphere. The pressure of the latter was determined by the temperature of this treatment: 48 H at 1300°C, 72 H at 1200°C, or 100 H at 1100°C. The surface layer of

each couple was removed after treatment in order to eliminate the material contaminated by surface diffusion.

Examination of the Couples. The curves concentration-penetration were determined by a Castaing [1] electron-probe microanalyzer from a plane surface normal to the welded interface and parallel to the direction of diffusion. The electron-probe employs a sharply focused electron beam permitting the irradiation of about one cubic micron of material. Quantitative analyses are obtained from the measurement of the intensity of the $K\alpha$ x -radiation characteristic of each element in the irradiated zone. The electrons were accelerated through a potential difference of between 20 and 30 kV. Since the couples were often composed of elements possessing widely differing atomic numbers, it was necessary to prepare calibration curves. The latter were obtained from solid solution binary alloys which were carefully homogenized before examination. Under these conditions, the precision of the diffusion measurement was approximately 1%.

Determination of Diffusion Coefficients

The calculation of the coefficients of diffusion were obtained following the method due to Grube. Under the present experimental conditions, Grube's solution of the second law of Fick could be employed. The solution proposed by Grube can be written as follows:

$$\frac{2(C - C_{\min})}{C_{\max} - C_{\min}} = 1 - \psi\left(\frac{x}{2\sqrt{Dt}}\right)$$

In this relationship $\psi\left(\frac{x}{2\sqrt{Dt}}\right)$ is the Gaussian integral, while t is the time of the diffusion heat treatment and D is the coefficient of diffusion.

The concentration-penetration curve for a Co-Co V Couple annealed at 1200°C for 72 hours is shown in Fig. 1.

The variation of $\frac{C - C_{\min}}{C_{\max} - C_{\min}}$ as a function of x is shown in Fig. 2a in a Gaussian diagram. If, in addition, one plots $\frac{x}{2\sqrt{Dt}}$ as a function of x , we obtain a linear relationship as shown in Fig. 2b. Such a behavior indicates that D is independent of the concentration.

The relationship between the coefficient of diffusion, D , and the concentration, C , is shown in Fig. 2c when it can be seen that

Table II
Results

Couples	T°C	Coefficient of Diffusion 10^{-12} cm ² /sec
Co/Co 15.2 Cr	1300	350
	1200	61.5
	1150	48.2
	1100	22
	1000	3.13
Co/Co 14.8 V	1300	780
	1200	202
	1150	142
	1100	44.5
	1000	11.7
Co/Co 15% Mo	1300	427
	1200	83.5
	1150	65
	1100	31
	1000	3.82
Co/Co 14.6% W	1300	91
	1200	26
	1150	18.25
	1100	5.7
	1000	1.3
Ni/Ni 10.3 Cr	1300	2100
	1200	385
	1150	244
	1100	73
	1000	20.6
Ni/Ni 14.8 V	1300	2150
	1200	340
	1100	112
	1000	21.4
Ni/Ni 13.8 Mo	1300	950
	1200	280
	1150	92.5
	1100	41.1
	1000	8.1
Ni/Ni 14.6 W	1300	161
	1200	20.4
	1150	17.35
	1100	4.9
	1000	0.71

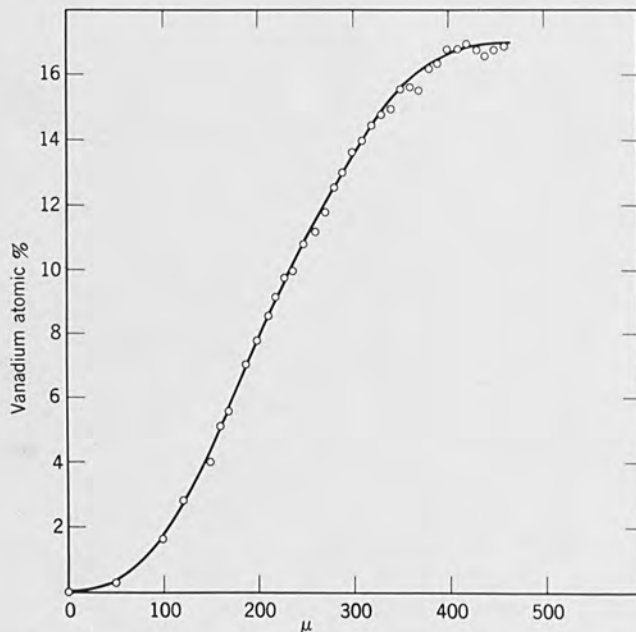


Fig. 1. Typical concentration-penetration curve, as determined for a Co-Co V couple after annealing at 1200° C for 72 hours.

D is independent of C . The linear relationship in Fig. 2b was employed to calculate the coefficients of diffusion which are shown in Table II, together with the temperature of diffusion and the composition of the couples.

If the logarithm of these experimental values are plotted as a function of the reciprocal of the absolute temperature, it is immediately evident that the coefficient of diffusion obeys a law of the type:

$$D = D_0 \exp \left(-\frac{H}{RT} \right) \quad (1)$$

Using the method of least squares, it is possible to determine the best linear fit of the experimental points. This can be employed to determine the frequency factor, D_0 , and the heat of activation, H .

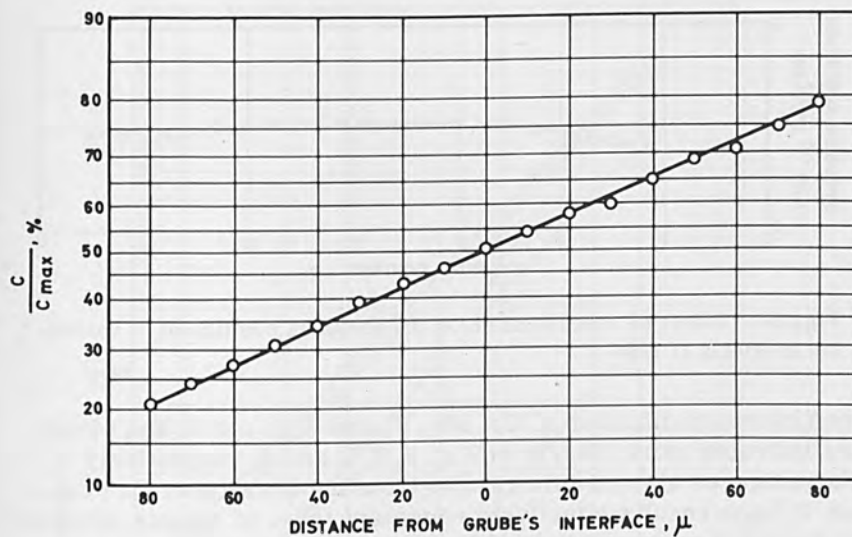


Fig. 2A. Typical $(x/c_{\max})$ probability plot as determined for a Co-Co V couple after annealing at 1200° C for 72 hours.

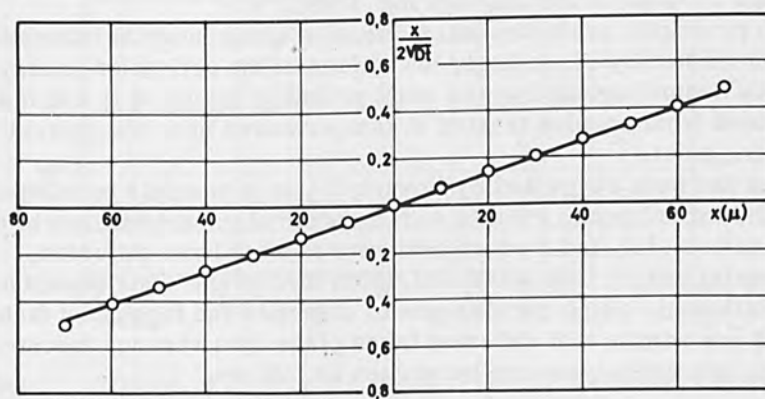


Fig. 2B. Typical $(x / \frac{x}{2\sqrt{Dt}})$ plot as determined for a Co-Co V couple after annealing at 1200° C for 72 hours.

RESULTS AND DISCUSSION

The experimental results, shown in Figs. 3 and 4 give the coefficients of diffusion of Cr, Mo, V, and W in Ni and in Co[2]. The

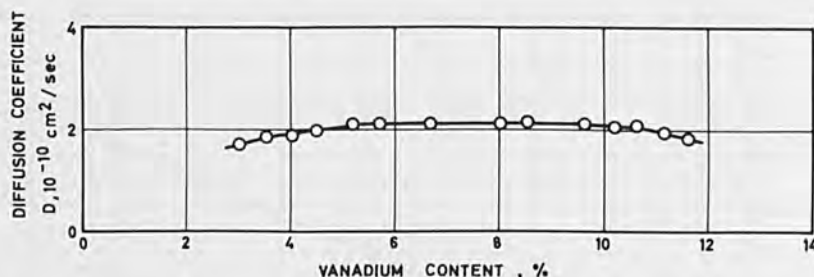


Fig. 2C. Effect of concentration on the diffusion coefficient of vanadium in cobalt at 1200° C.

coefficients of diffusion of Cr, Mo, V, and W in cobalt and nickel are indicated more clearly in Fig. 5, 6, 7, and 8, respectively. These figures also contain results from the literature. One can ask if these results effectively represent those of volume diffusion or if grain boundary diffusion has not also influenced the experimental points. Such parasitic effects are difficult to reveal if one does not have a sufficient amount of precise data obtained over a large range of temperatures in order to reveal the presence of a change in slope in the diagram $\log D$ vs T^{-1} .

In principle, grain-boundary diffusion along internal interfaces is manifested by exceedingly low values of the activation energy. It thus seems logical that the most probable values of D_0 and H are obtained from couples treated at temperatures near the solvent melting point.

As has been suggested by Nowick [3], it is possible by comparing the experimental results with theoretical considerations to obtain values of D_0 and H which best represent volume diffusion. Zener [4] has, in fact, advanced, from theoretical considerations, a relationship which permits one to calculate the frequency factor D_0 . If one admits that diffusion takes place by a vacancy mechanism, this expression can be written as follows:

$$D_0 = \gamma a^2 \nu \exp \left(\frac{\Delta S}{R} \right) \quad (2)$$

in which a is the lattice parameter, ν is the vibrational frequency and γ is a geometrical constant equal to unity for face-centered-cubic lattices, and ΔS is the entropy of activation of the mechanism. The latter can be written as the sum of two terms ($\Delta S = \Delta S_1 + \Delta S_2$) where ΔS_1 and ΔS_2 are, respectively, the entropy of formation and jumping of a vacancy.

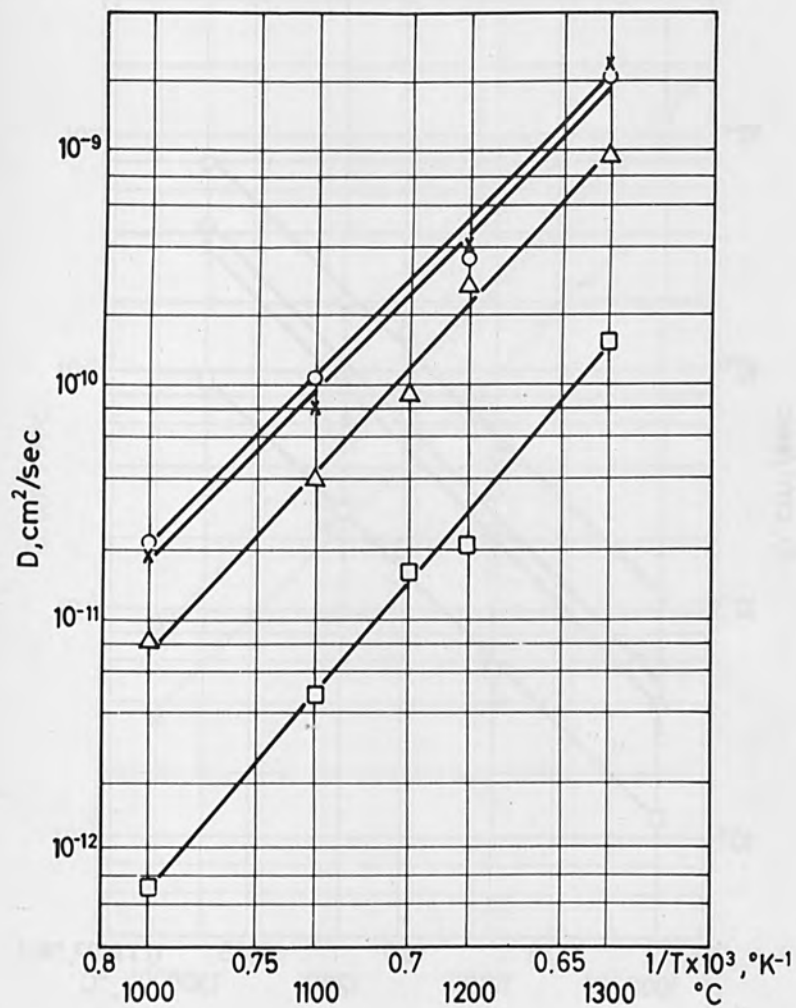


Fig. 3. Variation of $\log D$ as a function of $1/T$ for the diffusion of Cr, V, Mo, and W in nickel.

x Chromium. o Vanadium. Δ Molybdenum. □ Tungsten.

It can also be shown that

$$\Delta S = \frac{\lambda H}{\mu_0} \frac{d\mu}{dT} \quad (3)$$

where μ is the shear modulus, λ a constant equal to 0.57, and H

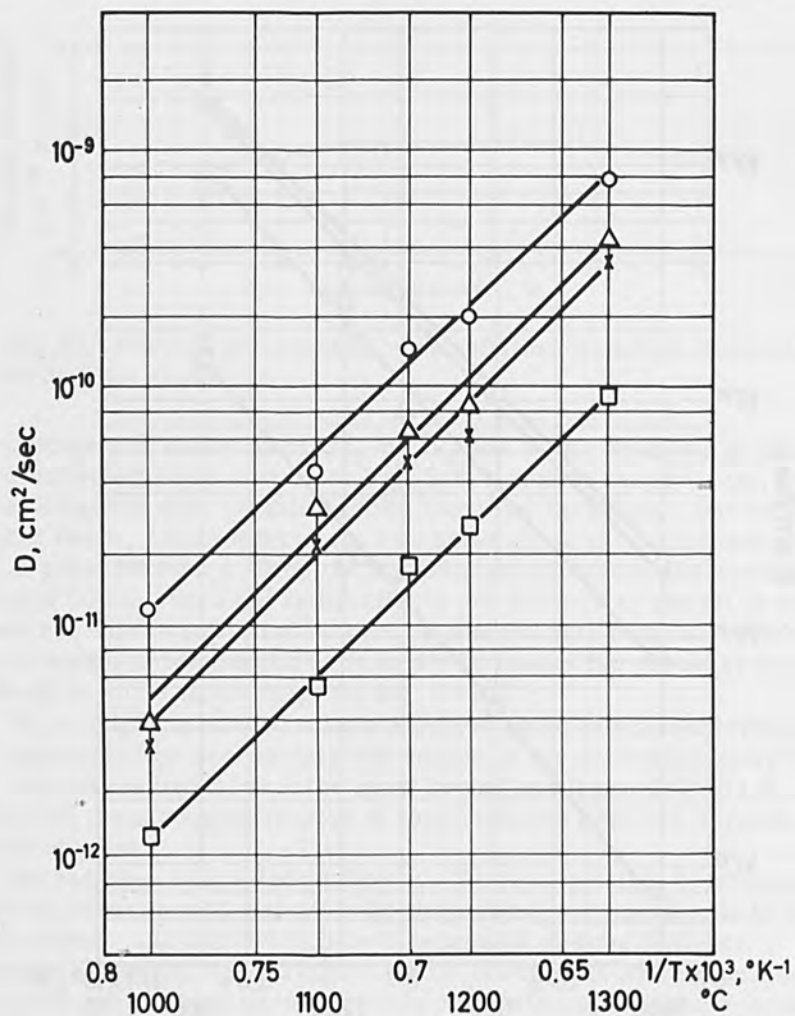


Fig. 4. Variation of $\log D$ as a function of $1/T$ for the diffusion of Cr, V, Mo, and W in cobalt.

x Chromium. o Vanadium. Δ Molybdenum. $\square$ Tungsten.

the enthalpy of activation. From the above relationships one can determine the coefficient of diffusion at any temperature if the enthalpy, H , is known.

Nowick has shown how one can determine the enthalpy from high-temperature measurements. If one denotes H' and $\Delta S'$ as the

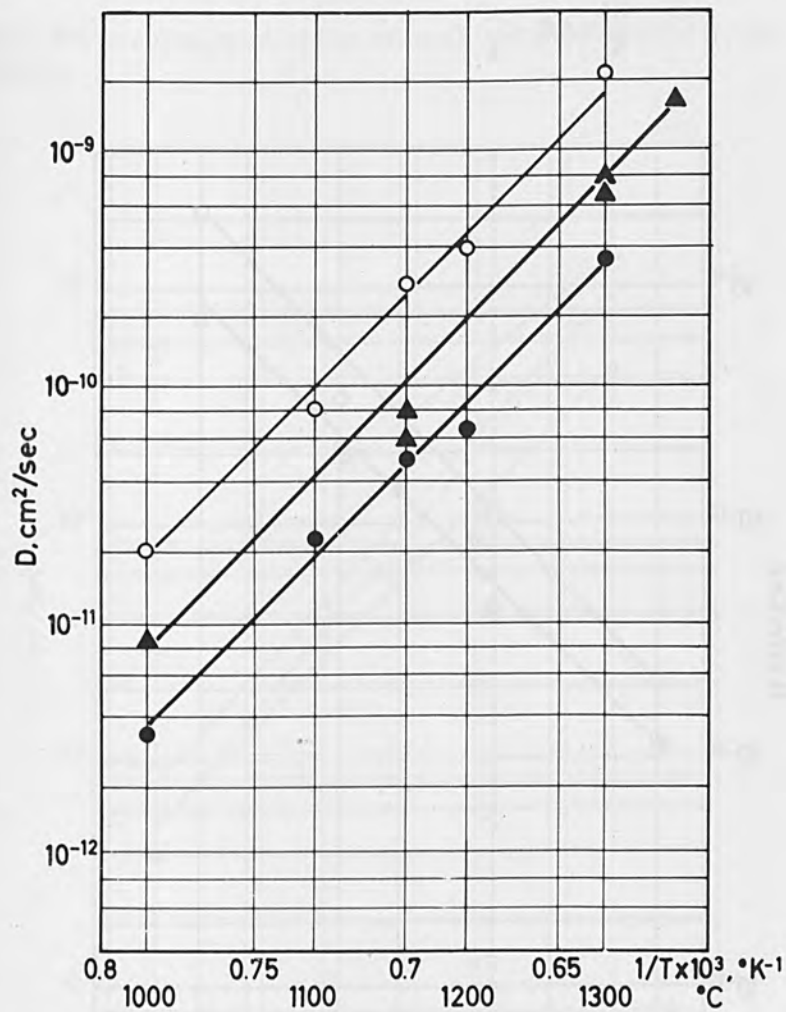


Fig. 5. Variation of $\log D$ as a function of $1/T$ for the diffusion of chromium in nickel and cobalt.

- Chromium in cobalt (present investigation).
- ▲ Chromium in cobalt (Weeton [13]).
- Chromium in nickel (present investigation).

experimental values, and H and ΔS as the true values of the enthalpy and entropy of activation, respectively, one can write at the temperature T_1 , near the melting point of the solvent that

$$\Delta S' - \frac{H'}{T_1} = \Delta S - \frac{H}{T_1}$$

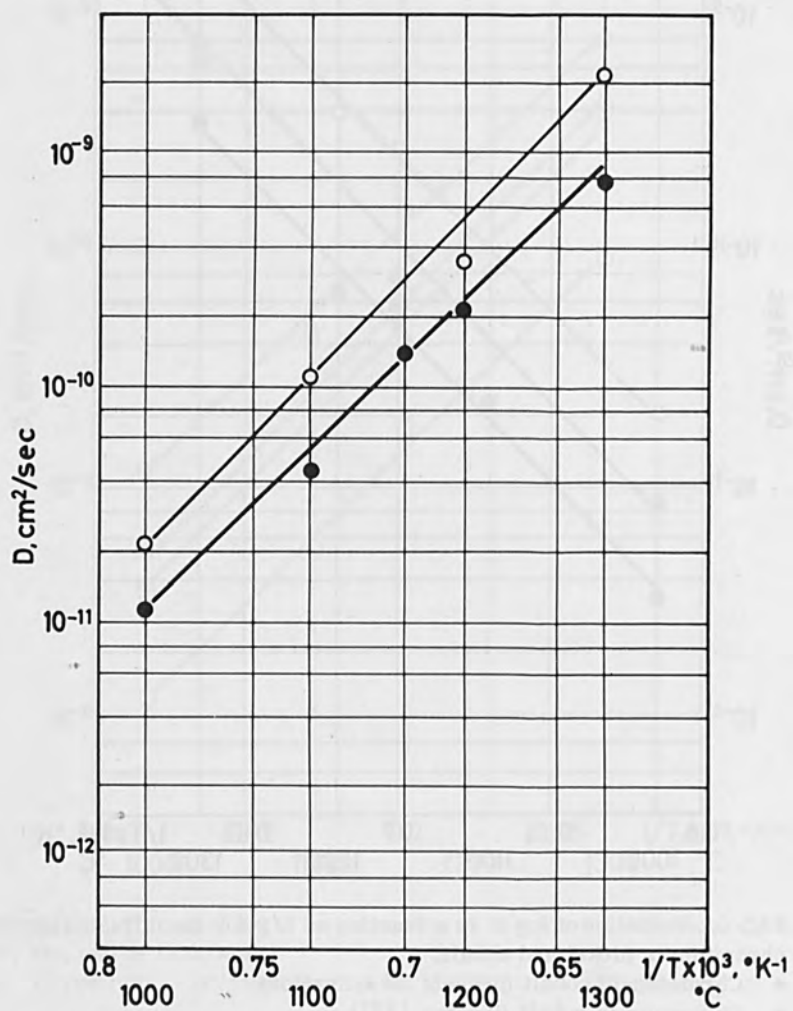


Fig. 6. Variation of $\log D$ as a function of $1/T$ for the diffusion of vanadium in nickel and in cobalt.

- Vanadium in cobalt (present investigation).
- Vanadium in nickel (present investigation).

When T_m (melting point of the solvent) is substituted for T_i , one obtains:

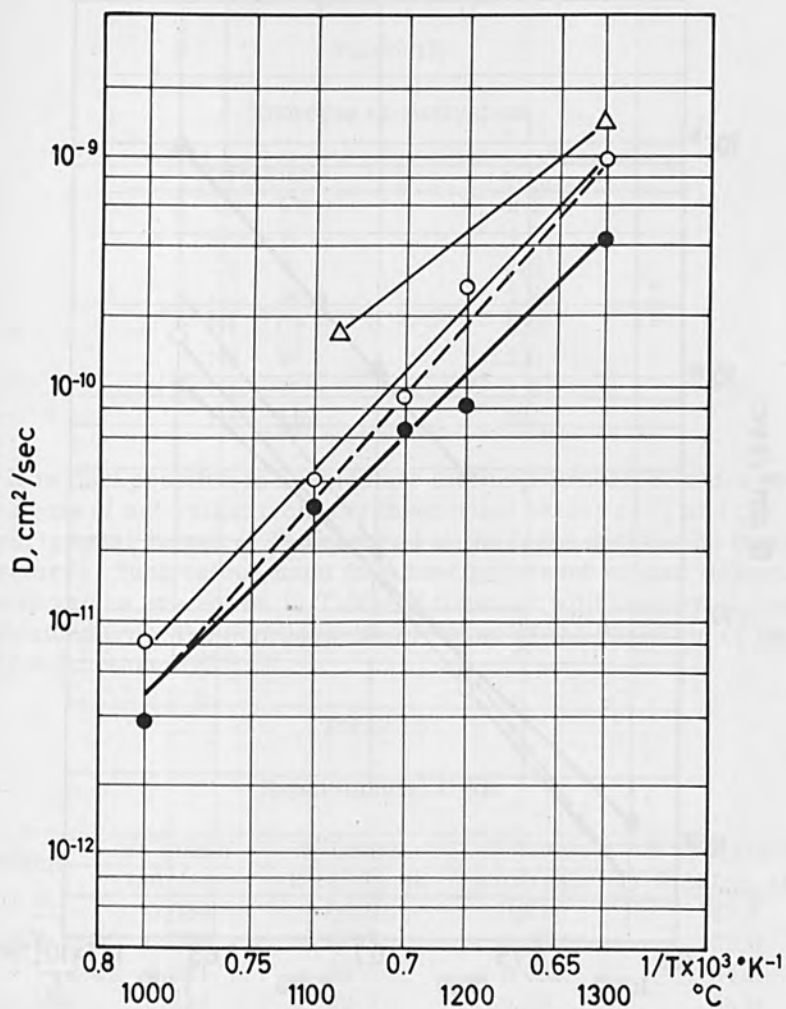


Fig. 7. Variation of $\log D$ as a function of $1/T$ for the diffusion of molybdenum in nickel and cobalt.

- Molybdenum in cobalt (present investigation).
- Molybdenum in nickel (present investigation).
- △ Molybdenum in nickel (Budde 14).
- Molybdenum in nickel (Swalin 15).

$$\Delta S' = \frac{H'}{T_m} + \Delta S - \frac{H}{T_m} \quad (4)$$

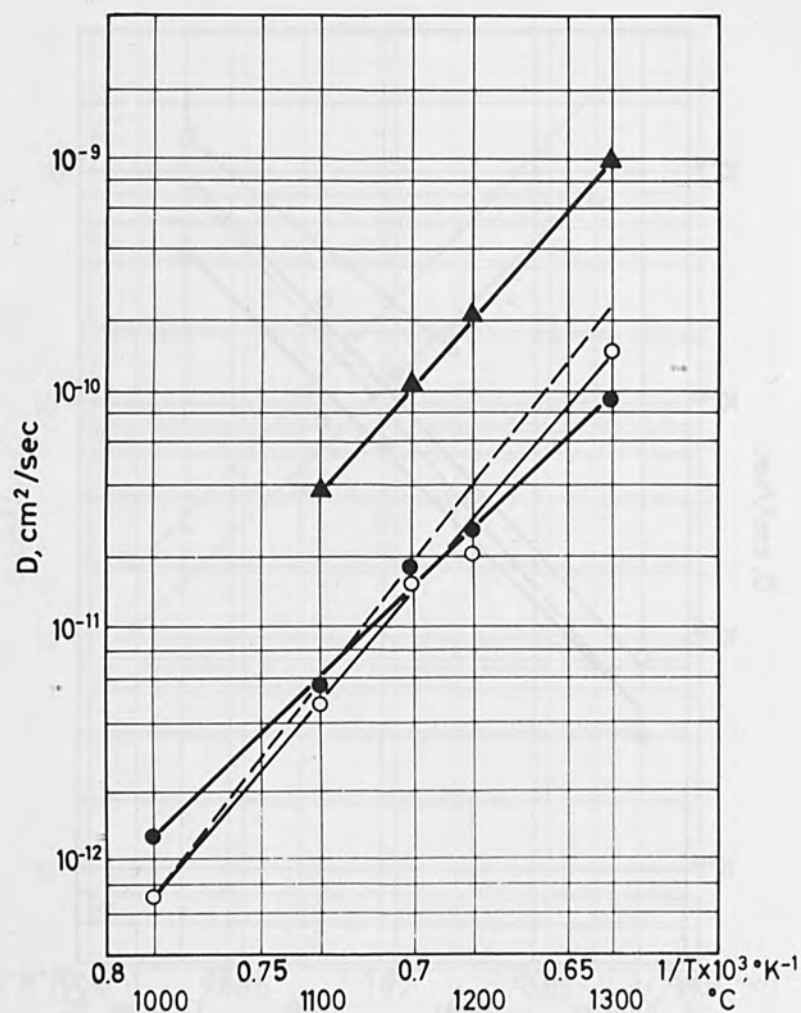


Fig. 8. Variation of $\log D$ as a function of $1/T$ for the diffusion of tungsten in nickel and cobalt.

- Tungsten in cobalt (present investigation).
- ▲ Tungsten in cobalt (Kovenskii [16]).
- Tungsten in nickel (present investigation).
- Tungsten in nickel (Swalin-Martin [17]).

The value of ΔS shall be calculated from the Zener equation, and H shall be taken as being equal to the heat of activation for self-diffusion. The values of $\Delta S'$, calculated from the above equations, are given in Table III.

Table III

Entropie of Activation

Specimens	E.U.
Co - Cr	6.1
Co - W	4.1
Co - V	2.0
Co - Mo	7.4
Ni - Cr	6.7
Ni - W	12.0
Ni - V	5.6
Ni - Mo	8.4

It is then possible to recalculate the most probable values of the heats of activation from the theoretical values of D_0 and the experimental values of D measured at the highest diffusion temperatures. Such values must thus best represent volume diffusion. These values are shown in Table IV together with those of D_0 and H obtained from the semilogarithmic plots of the variation of $\log D$ as a function of T^{-1} .

Table IV

Experimental Data

Systems	D_0 graph cm ² /sec	H graph k cal/g. at.	D_0 theor. cm ² /sec	H calculated k cal/g. at.
Co - Cr	0.084	60.6	0.21	63.5
Co - W	0.008	56.9	0.08	65.0
Co - V	0.021	53.0	0.023	54.2
Co - Mo	0.231	62.8	0.39	65.0
Ni - Cr	0.604	61.4	0.28	59.0
Ni - W	0.862	70.4	3.93	75.0
Ni - V	0.287	59.2	0.16	57.0
Ni - Mo	0.853	64.4	0.58	64.0

From these results one can see that except for the cases Co - W

and Ni - W, the heats of activation are only slightly modified by this new method of calculation. This leads one to think that, for the diffusion temperatures and other experimental conditions employed, the diffusion has been a volume phenomenon.

From the previous thermodynamical values, one can also calculate the free energy of the diffusion process using the relationship:

$$\Delta G = H - T\Delta S \quad (5)$$

The values of ΔG calculated from the 1200°C data are shown in Table V.

Table V
Thermodynamic Results

	ΔG k cal/g. at.
Co - Co*	51.6
Co - Cr	54.5
Co - W	58.9
Co - V	51.3
Co - Mo	54.1
Ni - Ni†	52.2
Ni - Cr	49.2
Ni - W	57.3
Ni - W	48.8
Ni - Mo	51.7
Ni - Mn‡	47.9
Ni - Al ‡	49.3
Ni - Tl ‡	48.1
Ni - W ‡	56.2

*Results given by Gruzin [6].

†Results given by Hoffman [7].

‡Results given by Swalin and Martin [5].

It must be pointed out that the values of the free energy are in better agreement than those of the heat of activation. This results from the fact that an error in the position of the straight line in the semilogarithmic relationship of D as a function of T^{-1} yields an error in ΔS and H which is in the same direction.

The results of Swalin and Martin [5] are also shown in Table V. It should be pointed out that these workers employed couples having a rather small solute-atom content. In addition, this table also

contains the values of the free energies of self-diffusion for nickel and cobalt calculated at 1200°C from the results of Gruzin [6] and Hoffman [7].

As has been previously mentioned, the coefficient of diffusion measured near the melting point of the solvent best represents volume diffusion. Table VI gives the values of D^i_m , the coefficient of diffusion at the melting point of the solvent, as obtained from a graphical extrapolation of $\log D$ vs T^{-1} . The values of D^2_m have also been calculated from the theoretical D_0 and the semi-empirical heats of activation shown in Table IV. These two groups of values are collected in Table VI.

Table VI

Coefficient of Diffusion at the Melting Point of the Solvant

System	D^2_m graph cm ² /sec	D^i_m calculated cm ² /sec
Co - Cr	2.5×10^{-9}	3.25×10^{-9}
Co - W	7.6×10^{-10}	8.10×10^{-10}
Co - V	6.1×10^{-9}	4.8×10^{-9}
Co - Mo	3.25×10^{-9}	3.9×10^{-9}
Ni - Cr	1.05×10^{-8}	0.97×10^{-8}
Ni - W	1.2×10^{-9}	1.39×10^{-9}
Ni - V	1.22×10^{-8}	1.03×10^{-8}
Ni - Mo	4.6×10^{-9}	5.2×10^{-9}

If one plots the values of D^i_m as a function of the square of the melting point of the solute element, it can be seen that the resulting relationship is linear. The result for several elements in the case of nickel are shown in Fig. 9a. The present results are shown on the linear relationship reported by Swalin, Martin and Olson [8]. Similar results of D^i_m in the case of cobalt are shown in Fig. 9b. Here, the agreement is somewhat less satisfactory in the case of cobalt.

The linear relationship as determined by the method of least squares has an equation:

$$D^i_m = \gamma_0 \exp(-\alpha T^2 m i)$$

In the case of cobalt the parameters α and γ_0 take on the values $2.3 \times 10^{-7} \text{ }^\circ\text{K}^{-2}$ and $1.45 \times 10^{-8} \text{ cm}^2 \text{ sec}^{-1}$, respectively, while the corresponding values for nickel are $2.2 \times 10^{-7} \text{ }^\circ\text{K}^{-2}$ and 3.6×10^{-8}

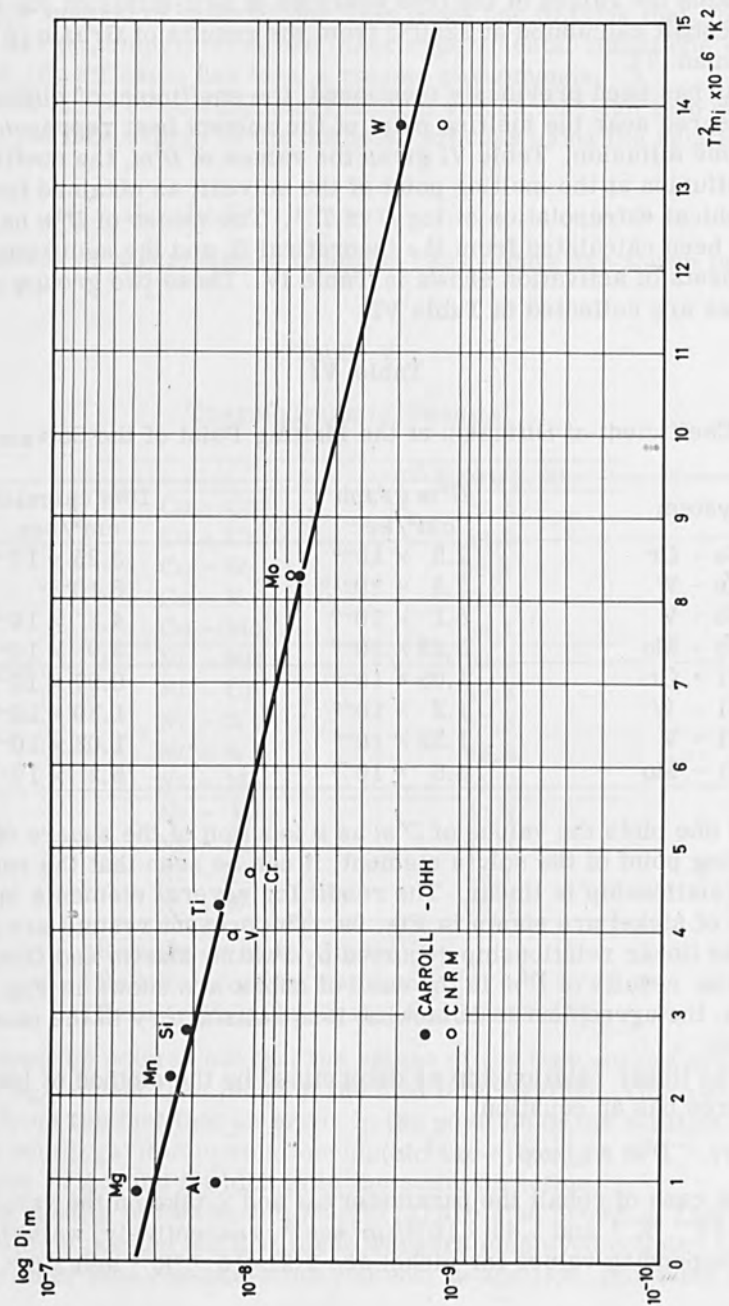


Fig. 9a. Variation of $\log D_{im}$ as a function of T_{mi}^2 in nickel.

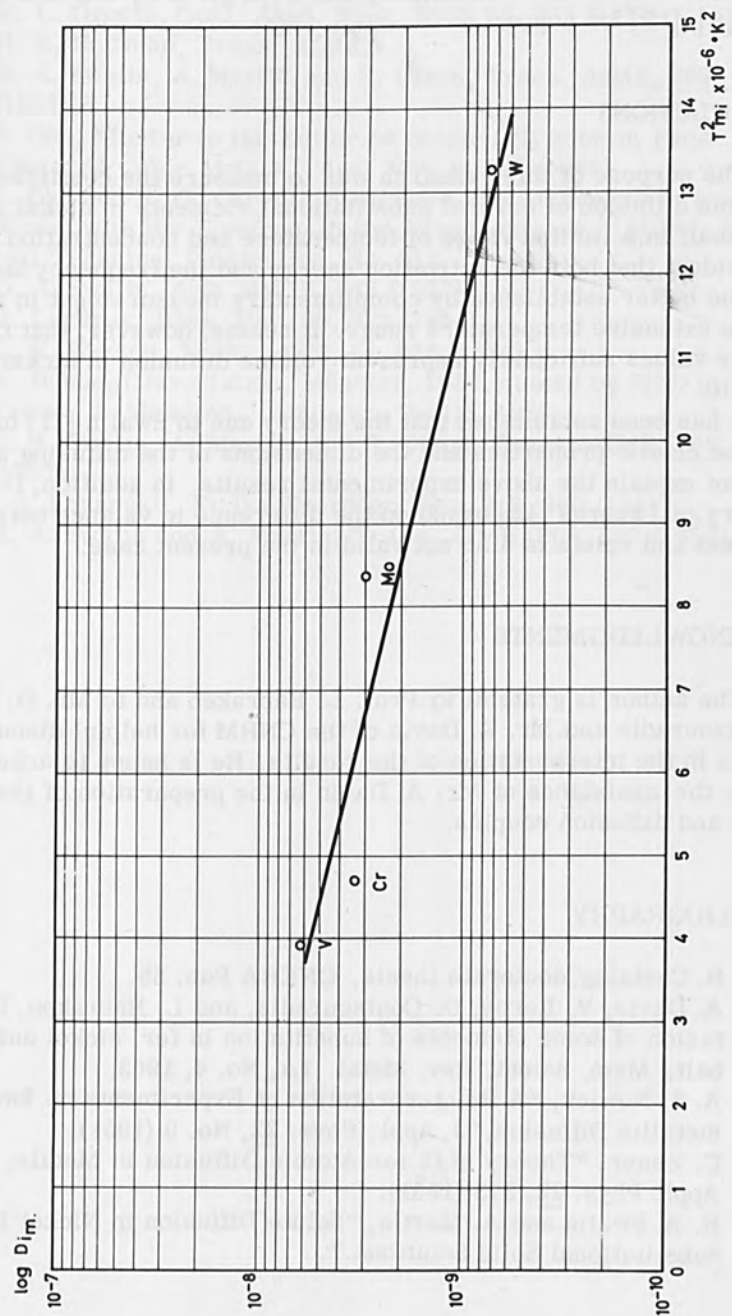


Fig. 9b. Variation of $\log D_m^i$ as a function of T_m^2 in cobalt.

$\text{cm}^2 \text{sec}^{-1}$. A similar relationship has been given by Ohh and Carroll [9], [10] for copper.

CONCLUSIONS

The purpose of this research was to measure the coefficient of volume diffusion of several substitutional elements in nickel and in cobalt in a limited range of temperature and concentration. It is evident that both the activation energy and the frequency factor can be better established by complimentary measurement in a more extensive temperature range. It seems, however, that the above values sufficiently represent volume diffusion in nickel and cobalt.

It has been established that the theory due to Swalin [11] based on the elastic properties and the dimensions of the diffusing atoms cannot explain the above experimental results. In addition, the theory of Lazarus [12] based on the difference in valance between solvent and solute is also not valid in the present case.

ACKNOWLEDGMENTS

The author is grateful to Prof. L. Habraken and to Mr. D. Coutsouradis and Mr. A. Davin of the CNRM for helpful discussions in the interpretation of the results. He is happy to acknowledge the assistance of Mr. A. Davin in the preparation of the alloys and diffusion couples.

BIBLIOGRAPHY

1. R. Castaing, doctorate thesis. ONERA Pub. 55.
2. A. Davin, V. Leroy, D. Coutsouradis, and L. Habraken, Diffusion of some elements of substitution in fer, nickel and cobalt. Mem. Scient. Rev. Metal. Lx, No. 4, 1963.
3. A. S. Nowick, "A Reinterpretation of Experiments on Inter-metallic Diffusion," J. Appl. Phys. 22, No. 9 (1951).
4. C. Zener, "Theory of D_0 for Atomic Diffusion in Metals, J. Appl. Phys. 22, 373 (1951).
5. R. A. Swalin and A. Martin, "Solute Diffusion in Nickel Base Substitutional Solid Solutions."

6. P. L. Gruzin, Dokl. Akad. Nauk. SSSR 86, 289 (1952).
7. R. E. Hoffman, Trans. AIME.
8. R. A. Swalin, A. Martin, and R. Olson, Trans. AIME, 209, 936 (1937).
9. S. Ohh, "Letter to the Editor on Solute Diffusion in Face-Centered Cubic Metals," Acta Met. 9, 387 (1961).
10. S. Ohh and R. G. Carroll, "Diffusion Measurements in Iron-Nickel with the Electron-Probe Microanalyzer," ASTM. Special Technical Publication, No. 317.
11. O. A. Swalin, Acta Met. 5, 443 (1957).
12. D. Lazarus, Phys. Rev. 95, 973 (1954).
13. J. W. Weeton, Trans. ASM, 44, 436 (1952).
14. K. Bubbe, Dissertation, Münster, 1954, quoted by Sifith in "Diffusion in Metallen." Springer, Berlin, 1955.
15. R. A. Swalin, A. Martin, and R. Olson, Trans. AIME, 209, 936 (1957).
16. I. I. Kovenskii, Fizika Tverdogo Tela, 3.2, 350 (1961).
17. R. A. Swalin and A. Martin, Trans. AIME, 206, 567 (1956).

1. J. L. Gervin, Dokl. Akad. Nauk SSSR 22, 266 (1963).
2. R. E. Hollman, Trans. AIME, 209, 936 (1957).
3. R. A. Swalin, A. Martin, and R. Olson, Trans. AIME, 209, 936 (1957).
4. S. Ohn, "Letter to the Editor on Solute Diffusion in Face-Centered Cubic Metals," Acta Met. 2, 382 (1954).
5. S. Ohn and H. G. Carroll, "Solute Diffusion in Face-Centered Cubic Metals," Acta Met. 2, 382 (1954).
6. R. A. Swalin, Acta Met. 2, 382 (1954).
7. D. Lazarus, Phys. Rev. 92, 913 (1954).
8. W. Westphal, Z. Naturf. 9a, 438 (1954).
9. K. Bopp, Dissertation, Münster, 1954, quoted by S. Ohn in Ref. 5.
10. R. A. Swalin, A. Martin, and R. Olson, Trans. AIME, 209, 936 (1957).
11. I. I. Korovin, Fizika Tverdogo Tela 3, 2 (1961).
12. R. A. Swalin and A. Martin, Trans. AIME, 209, 936 (1957).

ACKNOWLEDGMENTS

The author is grateful to Prof. L. Haberman and Dr. Mr. D. Choudhary and Mr. A. Davin of the CNRM for helpful discussions in the interpretation of the results. He is happy to acknowledge the assistance of Mr. A. Gervin in the preparation of the alloys and diffusion samples.

BIBLIOGRAPHY

1. H. Castaing, doctorate thesis, ONERA Pub. 55.
2. A. Davin, V. Leroy, D. Choudhary, and L. Haberman, Diffusion of some elements of substitution in fer, nickel and cobalt. Mem. Sci. Rev. Metal. La, No. 4, 1953.
3. A. S. Nowick, "A Reinterpretation of Experiments on Intermetallic Diffusion," J. Appl. Phys. 22, No. 9 (1951).
4. C. Zener, "Theory of Diffusion in Metals," J. Appl. Phys. 22, 373 (1951).
5. R. A. Swalin and A. Martin, "Solute Diffusion in Nickel Base Substitutional Solid Solutions."

THE SURFACE IONIZATION ION ENGINE

INTRODUCTION

As an introduction to the subject of ion propulsion, it is to review the reasons for investing in this type of propulsion system.

Section VII

for a rocket and its exhaust in free space. If the rocket is at rest, the following equation relating the exhaust velocity v_e to the mass of the rocket m and the mass of the rocket m_0 may be written:

ION PROPULSION

$$v_e = - \frac{1}{m} \ln \frac{m}{m_0}$$

Assuming that a given mission requires a certain velocity increment, this equation shows that a given velocity increment can be made by increasing the exhaust velocity. The physical reason for the improved performance is that for a given thrust level, exhaust velocity and propellant consumption rate are inversely related; as exhaust velocity increases, the corresponding decrease in the other.

For chemical rockets the maximum exhaust velocity is obtained by combining the highest chemical reaction energy with the lowest reaction product mass. The best combination is the hydrogen-oxygen reaction which gives a theoretical exhaust velocity of approximately 5×10^3 meter/sec.

If the exhaust is heated by a nuclear reaction, the exhaust velocity can be increased to approximately 10^4 meter/sec. using hydrogen as the propellant and heating it to 2500°K.

*Electro-Optical Systems, Inc., Pasadena, Calif.

This work supported by the U.S. Air Force Aero Propulsion Laboratory, Contract AF 33 (457)-10450.

Section VII

ION PRODUCTION

THE SURFACE IONIZATION ION ENGINE†

INTRODUCTION

As an introduction to the subject it is perhaps worthwhile to review the reasons for investing in the development of electric propulsion devices and systems.

By writing an equation expressing conservation of momentum for a rocket and its exhaust in free space and integrating once, the following equation relating the velocity increment imparted to the rocket, the rocket exhaust velocity and the initial and final masses of the rocket may be derived:

$$V_r = -v_e \ln \frac{m_i}{m_f} \quad (1)$$

Assuming that a given mission requires a specified velocity increment, this equation shows that significant gains in payload capability can be made by increasing the rocket exhaust velocity. The physical reason for the improved performance is of course that for a given thrust level, exhaust velocity and propellant consumption rate are inversely related; an increase in one implies a corresponding decrease in the other.

For chemical rockets the maximum exhaust velocity is obtained by combining the highest chemical reaction energy with the lowest reaction product mass. The best combination is the hydrogen-fluorine reaction which gives a theoretical maximum of approximately $5 \cdot 10^3$ meter/sec.

If the exhaust is heated by a nuclear reaction, the mean exhaust velocity can be increased to approximately $8 \cdot 10^3$ meter/sec by using hydrogen as the propellant and heating it to 2500°K.

*Electro-Optical Systems, Inc., Pasadena, Calif.

†This work supported by the U.S. Air Force Aero Propulsion Laboratory, Contract AF 33 (657)-10980.

Still higher velocities are desirable and even necessary for rocket flight to distant places. Thus in the last few years an experimental effort has been devoted to the development of ion rockets capable of operation at almost arbitrarily high exhaust velocities.

If equation 1, shown above, were the only one involved, the exhaust velocity should be made as high as possible and the photon rocket would be attractive. However, in an electric rocket system the power carried away by the exhaust must be generated in the vehicle. Since this power is proportional to exhaust velocity, high exhaust velocity implies a large and heavy electric power supply. Thus conflicting requirements are placed on the propulsion system and a compromise must be found. If the exhaust velocity is too low the propellant supply is unnecessarily heavy, and if the exhaust velocity is too high the electric power supply is unnecessarily heavy. As one might expect, the best situation is one where propellant supply and power supply weigh about the same. For missions of interest this corresponds to exhaust velocities in the range between $4 \cdot 10^4$ and $2 \cdot 10^5$ meter/sec.

If a useful system is to result, a charged particle generation and acceleration device should be made to satisfy the following requirements:

1. The device should be capable of producing exhaust velocities in the appropriate range. All particles should have approximately the same velocity and the angular spread in exhaust should be small.
2. Neutral particles should constitute only a small fraction of the total efflux.
3. The power required to run the ion source and accelerator should be very little larger than the power carried away by the exhaust.
4. The device must operate with high reliability for year-long periods.
5. The weight of the device should be small compared to the weight of the equipment used to generate power.

One of the most attractive ways of accelerating charged particles to the desired velocities is by the electrostatic acceleration of positive ions through an appropriate potential difference. The rest of this paper will concern itself with systems of this type.

CHOICE OF EXPELLANT

The choice of a specific expellant is closely bound to the choice of the specific method of producing the ions, but certain general considerations concerning the mass of the ion to be used can be made without regard to the nature of the ionization process.

The relation between the accelerating potential and mass required to yield exhaust velocities of interest is shown in Fig. 1.

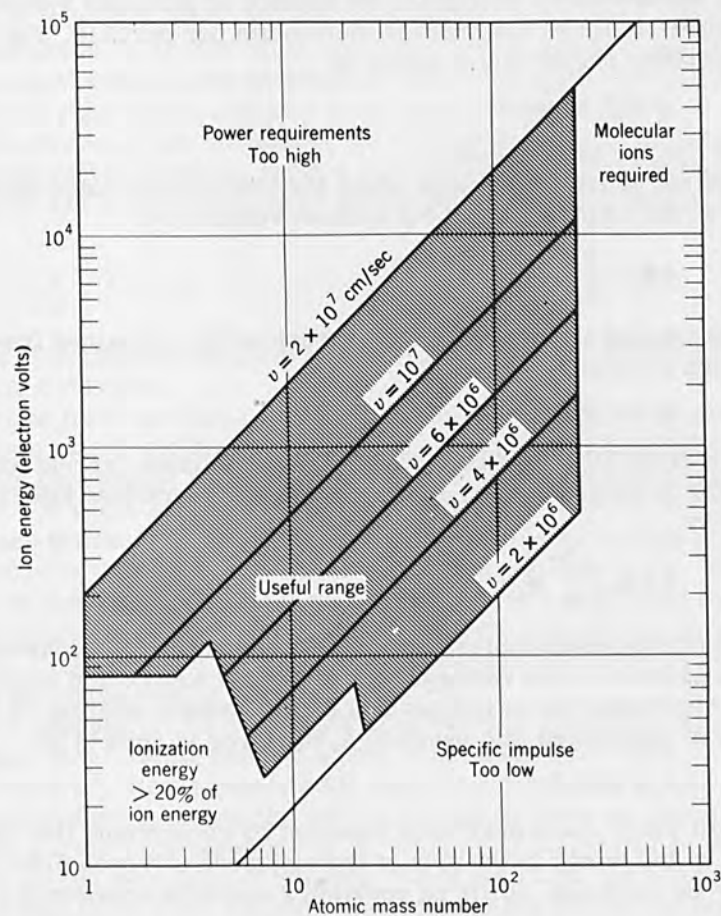


Fig. 1. Accelerating potential required to produce desired exhaust velocity as a function of ionic mass.

For low masses relatively low ion energies are desired and in some cases the energy which must be expended in producing the ions becomes too high. Because the energy expended in creating an electron-ion pair is usually several times greater than the ionization potential, the excluded region has been chosen as that for which the ionization potential exceeds 20% of the accelerating potential. The 20% figure may be somewhat conservative, but these considerations have the effect of eliminating light ions from the low velocity range.

The force exerted by a rocket engine is equal to the rate at which momentum is exhausted; the number of particles ejected per unit time times the average momentum per particle. For an electrostatic engine this is given by

$$F = (I/e) (\overline{mv}) \quad (2)$$

where I is the total current.

The net voltage Φ_0 through which the ion is accelerated is related to the ionic mass and the exhaust velocity by

$$e\Phi_0 = \frac{1}{2} mv^2 \quad (3)$$

Substituting in equation 2 the expression for v obtained from equation 3 yields

$$F = I\sqrt{2(m/e)\Phi_0} \quad (4)$$

In a given electrode configuration the maximum current which can flow is determined by space-charge considerations and is given by

$$I = K\sqrt{\frac{e}{m}} \Phi_0^{3/2} \quad (5)$$

where K is a dimensionless geometric constant, i.e., a constant which depends on the configuration of the ion source and accelerating electrodes but is independent of any uniform scaling. If equation 5 is substituted into equation 4, the force is seen to be

$$F = 2K\Phi_0^2 \quad (6)$$

a result which could have been foreseen by recognizing that the force on the motor is the sum of the electrode stresses over all electrode surfaces. Utilizing equation 2 converts equation 6 to

$$F = \frac{Kv^4}{2\sqrt{2}} \left(\frac{m}{e}\right)^2 \quad (7)$$

If we regard the exhaust velocity v as a constant defined by a mission we have a force from a given engine configuration proportional to $(m/e)^2$, with spectacular advantage indicated for small e/m values. Cesium, with a mass number of 133, is quite satisfactory in this respect; there is little to be gained by going to higher mass ions. This fact plus the fact that cesium is easily ionized strongly suggests the use of cesium as the expellant.

This analysis is based on the assumptions that there exist no limitation on current density from the ion source and no limitation on voltage which can be placed on the accelerating electrodes. In practice, however, ion source current densities are limited and there is a definite limit to electric field strengths which can be maintained at an accelerating electrode before sparking and sustained discharges occur between electrodes, a field strength which practical experience indicates is of the order of 10^5 V/cm.

The stress on the surface of the accelerating electrode is the force per unit area which, from a detailed point of view, is responsible for the force produced by an ion engine. This stress is given by

$$\frac{F}{A} = \frac{1}{2} \epsilon_0 E^2 \quad (8)$$

where E is the electric field intensity at the surface of the accelerating electrode.

In this form the force per unit area is seen to be independent of the ionic mass, and all that is required is to scale the geometry to yield the largest possible field strength E . For example, if $E_{\max} = 10^5$ V/cm and $\Phi_0 = 10^3$ V, accelerating gaps of the order of 10^{-2} are necessary. From this point of view the advantage in massive ions is that it permits relatively gross configurations. Dimensions of the order of 1/2 cm or larger are desirable, implying an accelerating voltage of 50 kV. Since the entire useful range shown in Fig. 1 lies below 5×10^4 V, the advantage to be gained by the use of higher mass ions is clear.

The considerations and relations presented above are based on a single accelerating gap and single accelerating potential. There are, however, strong reasons for departing from this simple scheme and using an accelerate-decelerate scheme as shown in the ion engine schematic of Fig. 2. Such an arrangement is suggested by the necessity of trapping electrons in the ion beam and preventing them from entering the accelerating region and bombarding the ion source with a consequent serious loss of power. A further advantage is that the voltage across the accelerating gap

$(\Phi_0 + \Phi_1)$ can be made much larger than Φ_0 , thus increasing the space-charge limited current or making possible the use of larger dimensions.

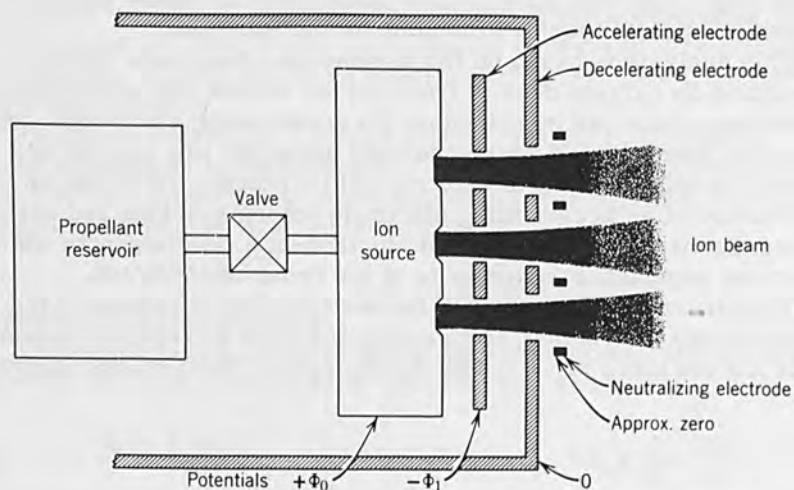


Fig. 2. Schematic representation of ion engine.

ION SOURCE

The general requirements for electric propulsion devices listed earlier place a number of requirements on the ion source. Quite obviously the source must be reliable and long-lived. In addition, there are some more subtle requirements imposed by the necessity for long operating times.

Clearly, from power considerations alone, one demands low interception of the ion beam by the accelerating electrode. A much more pressing requirement, however, for low interception arises from the erosion of accelerating electrodes produced by the sputtering by high-energy ions. For example, a 10 kV cesium ion striking a copper electrode will eject about 10 copper atoms, and unless the intercepted beam fraction remains very small, below about 1 ion in 10^4 , erosion becomes excessive. The technique of producing such low intercepted beam fractions is largely a prob-

lem of accelerating structure design and fabrication, but certain aspects of the ion source are also involved.

It is essential that the ions leave the source with randomly directed velocities extremely small compared to their velocity as they pass the accelerating electrode if the desired low interception values are to be obtained. Furthermore, the ions have to start from a surface which is stationary and satisfies criteria required for a good optical system.

Any material which leaves the source as thermal velocity neutral particles contributes negligibly to the thrust while increasing the propellant consumption rate. To avoid this propellant waste it is essential that the propellant efflux be close to 100% ionized.

The neutral efflux has a secondary effect related to lifetime and sputtering which may impose an upper limit on its allowed value much more stringent than that imposed by propellant utilization considerations. This is the effect of resonant charge exchange in the accelerating gap. When an ion passes through a neutral vapor it may capture an electron from an atom, proceeding as a high-velocity neutral atom and leaving behind a slow ion. In an accelerate-decelerate configuration, the slow ion has a high probability of striking the accelerating electrode, producing sputtering effects as discussed in the preceding section.

When the ion and atom are of the same species, so that no internal energy change is involved in the process, the cross section for the reaction can be quite large; values of the order of 10^{-14} cm^2 have been measured. Suppose we wish to avoid having more than one ion in 10^4 undergo charge exchange and that the ion path length in the vicinity of the accelerating electrode is 1 cm. Then, for a cross section of 10^{-14} cm^2 , it is necessary that the neutral atom density be less than 10^{10} cm^{-3} . If the mean neutral atom velocity away from the ion source is 10^4 cm/sec , this implies a neutral efflux from the source of $10^{14} \text{ atoms/cm}^2\text{-sec}$, which for singly charged ions is the mass flow corresponding to $1.6 \times 10^{-5} \text{ amp/cm}^2$. Since ion current densities of approximately $5 \times 10^{-3} \text{ amp/cm}^2$ are required for appreciable thrust densities, the requisite ionization percentage is approximately 99.7%. This is a difficult requirement to meet.

The limitation to the acceleration which can be obtained with electric rockets lies in the weight of equipment required to power the rockets. It is therefore of primary importance that the power required to generate the ions be as small as possible. Since ionization potentials are of the order of 10 V, and since heavy ions re-

quire accelerations of the order of 10 V, and since heavy ions require accelerations of the order of a kilovolt or more, the goal of nearly 100% efficient engines is, in principle, achievable. At present, however, sources require many times the ionization energy per ion produced and the power requirement is one of the most important parameters for ion sources.

One particularly simple mechanism for ion generation is that of surface ionization. The propulsion devices discussed in this paper make use of this mechanism. The process is illustrated in Fig. 3

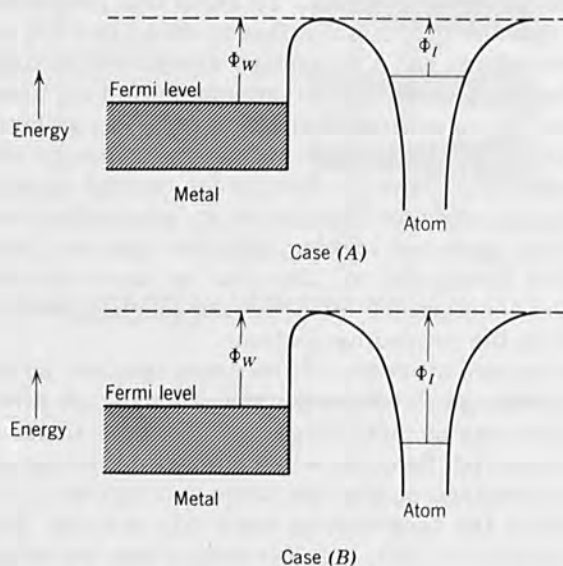


Fig. 3. Energy level diagram of atom near metal surface.

showing an energy level diagram of an atom in the vicinity of a metal. The metal is represented by a potential well filled with electrons up to the Fermi level.

For the case shown in Fig. 3(a), the ionization potential Φ_i is less than the work function Φ_w , and the energy of the system is higher than it would be if the outer electron were removed from the atom and placed in the metal at the Fermi level. Accordingly when an atom comes into thermal equilibrium with the surface before evaporation, the outer electron will usually remain in the metal and the evaporated particle will be an ion. If $\Phi_i > \Phi_w$ as in

Fig. 3(b), the lower energy state is that in which the atom is neutral and surface evaporation will thus consist mainly of neutral particles. Quantitatively the ratio of neutrals to ions can be expressed by the Saha-Langmuir equation

$$R_A = \frac{g_a}{g_i} \exp [- (\Phi_w - \Phi_i) e/kT] \quad (9)$$

where the g 's are statistical weighting factors for the atomic and ionic states ($g = 2s + 1$), where s is the atomic spin, k is the Boltzmann constant, and T is the absolute temperature.

The alkali metal atoms, lithium, sodium, potassium, rubidium, and cesium all have sufficiently low ionization potentials that surface ionization is easily accomplished using available metal surfaces. In the case of the alkali metals $g_a/g_i = 2$, and equation 9 becomes

$$R_A = 2 \exp \left[\frac{-11600 (\Phi_w - \Phi_i)}{T} \right] \quad (10)$$

where the potentials are in volts and the temperature in degrees Kelvin. Of the alkali metals, cesium is the heaviest and has the lowest ionization potential, so that it is clearly the optimum alkali for propulsion; we shall confine the remainder of our discussion to the surface ionization of cesium. Its use has been objected to on the basis of its cost and relative scarcity, but as the demand has increased the cost has very rapidly decreased and this appears no longer to be a serious consideration.

For cesium $\Phi_i = 3.8$ V and for tungsten, which has most extensively been used in surface ionization experiments, $\Phi_w = 4.52$ V. The substitution of these numbers in equation 10 shows that this combination is very favorable even at fairly high temperatures. This equation indicates that the neutral efflux ratio is lowest at low temperatures. However, a much more important consideration, the adsorption of cesium on the ionizing surface, rules out low temperature operation.

For any given rate of delivery of cesium to a surface there is, under equilibrium conditions, a concentration of cesium on the surface due to the finite time spent on the surface by an ion or atom before evaporation takes place. This concentration is expressed in terms of θ , the fraction of a monolayer coverage, where a monolayer of cesium on tungsten corresponds to about 4.5×10^{14} atoms/cm². The work function Φ_w which belongs in equation 10 is therefore that of the composite surface and falls very rapidly as θ increase, a fact which shows up in a quite remarkable fashion

in the electron emission data shown in Fig. 4. Examine, for example, the curve corresponding to a delivery rate of 10^{17} atoms/cm²-sec. At a temperature of 1800°K the emission current is 2×10^{14} electrons/cm²-sec; if the work function remained constant

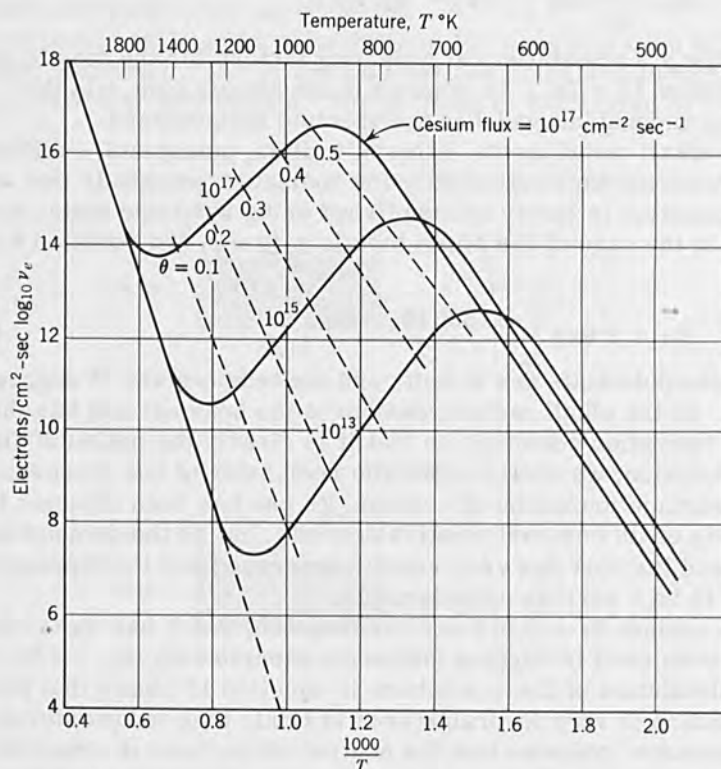


Fig. 4. Electron emission from cesium-tungsten surface.

the emission would follow the straight line and a decrease in temperature to 1000°K would cause the emission to drop to about 10^4 electrons/cm²-sec, a decrease by a factor of 2×10^{10} . Instead the emission is seen to rise to 10^{16} electrons/cm²-sec, an increase of a factor of 50. The large discrepancy is accounted for by the rapid decrease in work function as the surface concentration of cesium increases. At a given cesium supply rate, the increase in surface coverage with decreasing temperature shown in Fig. 4 is due to the increased lifetime before evaporation of cesium on the tungsten surface.

The implications of this work function variation with cesium coverage upon surface ionization are shown graphically in Fig. 5, in which the flux of ions and atoms evaporating from the surface is shown as a function of surface coverage for a particular tungsten

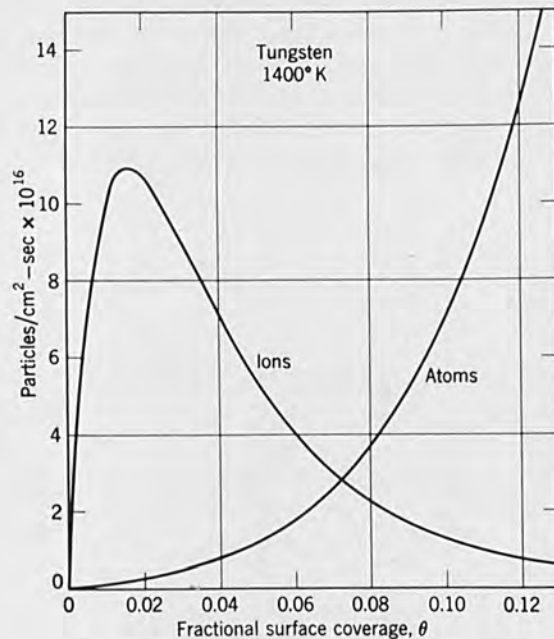


Fig. 5. Cesium ion and atom emission from tungsten surface.

temperature. For very small values of θ the work function at the surface is essentially that of tungsten, the ion emission is proportional to θ , and the atom emission is low. As the surface coverage increases, the work function of the surface decreases and the ion-to-atom ratio decreases as indicated by equation 9. The ion emission passes through a peak and decreases again; by the time θ has reached approximately 0.10, atom emission is strongly predominant.

The data shown in Fig. 5 can be combined with the corresponding data for other temperatures to produce a family of curves quite useful in discussing ionizer performance; these curves are shown in Fig. 6, in which ion current density is plotted against tungsten surface temperature. The curve labeled "maximum current density" is the envelope of the family of curves and gives the

maximum current density which may be obtained at a given temperature. The point at 1400° corresponds to the maximum ion emission in Fig. 5; other points are derived from similar curves. The individual curves of Fig. 6 indicate the maximum ion current which can be produced at a given temperature, with the added restriction that the ratio of atom emission to ion emission (defined as R_A) not exceed the value with which the individual curve is labeled. Thus it is seen that high current densities may be achieved only if corresponding atom emission densities are acceptable.

Let us try to understand physically why Fig. 6 looks the way it

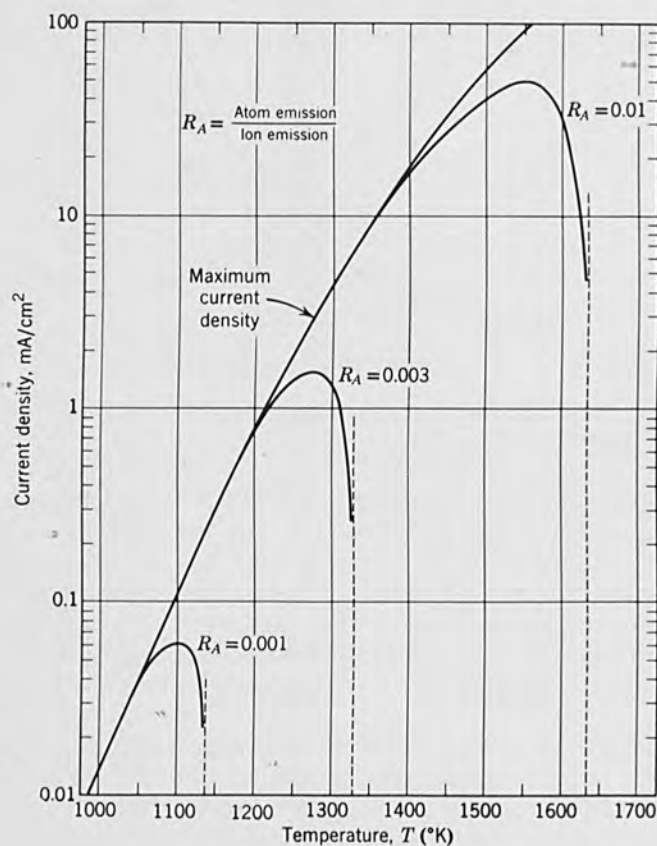


Fig. 6. Cesium ion current density obtainable from tungsten surface.

does. Cesium atoms or ions are bound to the tungsten surface by an attractive force characterized by an "energy of adsorption." Thus, in order to evaporate from the surface, the cesium must possess enough thermal energy to overcome the energy of adsorption; its probability per unit time of evaporation is proportional to the probability that its thermal energy exceeds the energy of adsorption. The distribution of thermal energy depends exponentially on energy and temperature as given by

$$N(E) = \exp \frac{-E}{kT} \quad (11)$$

As the temperature is increased, then, the probability per unit time of evaporation increases very rapidly. The rate of emission from the surface is proportional to the surface coverage θ and to the probability per unit time of evaporation. Figure 5 shows that maximum ion emission takes place at about $\theta = 0.015$. This value of θ will not change much with temperature; it must always be kept low to avoid decreasing the surface work function if efficient ion production is desired. Thus we see that the maximum ion emission is dominated by the probability per unit time of evaporation and increases very rapidly with temperature as shown in Fig. 6. The fact that there is a maximum ion current density for each value of R_A can be seen from an examination of equation 10, which for constant R_A can be written:

$$T = \text{const } x (\Phi_w - \Phi_i)$$

As the temperature is increased, Φ_w must also increase, which means that θ must decrease slightly as shown by Fig. 4. Finally Φ_w reaches the pure tungsten value and θ vanishes. At this limiting temperature (indicated by the vertical dashed lines in Fig. 6), the emission (which is proportional to θ) drops to zero.

As indicated earlier, the power required to operate an ion source is a very important consideration. In the case of the surface ionization source, the principal loss of power is through thermal radiation from the hot tungsten surface. For purposes of calculation we will assume here that the total heat loss is that from the front surface of the tungsten. The power loss per unit area is then $\epsilon \sigma T^4$ where ϵ is the total emissivity of the tungsten surface, σ is the Stefan-Boltzmann radiation constant, and T is the temperature in degrees Kelvin. The power put into the ion beam per unit area is $j\Phi_0$, where j is the ion current density, and Φ_0 is the net voltage through which the ions are accelerated. The ratio of radiated power-to-beam power is given by

$$R_B = \frac{\epsilon \sigma T^4}{j \Phi_0} \quad (12)$$

It is desirable to have R_B as small as possible. For a given temperature the maximum obtainable value of j may be obtained from Fig. 6. If the temperature dependence of ϵ is put in and the numerator is expressed in terms of j through the curve of Fig. 6, it is found empirically that equation 12 may be rewritten in a relation due to Forrester and Speiser

$$j = \frac{2.3}{(R_B \Phi_0)^{1.4}} \quad (13)$$

where j is in mA/cm² and Φ_0 is in kV. Thus, increased efficiency is to be gained either by increasing j or Φ_0 . The latter is usually limited by mission considerations. In order to increase j , electric accelerating fields must be increased and accelerating structure dimensions decreased, a procedure which cannot be carried on indefinitely. In addition, large current density is accompanied by large neutral efflux. Thus in any given situation the design of an ion source will be influenced strongly by compromises between competing considerations.

The most obvious approach to the problem of delivering cesium is to evaporate or spray cesium vapor directly onto the tungsten ionizing surface. In order to maintain an adequate flow of cesium the concentration of cesium atoms in the vicinity of the tungsten would have to be of the order of 10^{13} cm⁻³. There are at least two disadvantages to the scheme: the presence of the cesium vapor would increase the difficulty of maintaining strong electric fields in the accelerating region; and charge exchange interactions between outgoing ions and incoming atoms would produce unacceptably large numbers of slow ions which would subsequently be drawn to the accelerating electrode.

A scheme which overcomes the above objections and appears to work quite satisfactorily utilizes ionizers made of porous tungsten. There are a large number of interconnected pores in the material so that cesium vapor supplied to the rear of the tungsten under a pressure of a few millimeters of mercury travels to the front surface of the tungsten where it evaporates mainly as ions. It is necessary for satisfactory operation that the pores in the tungsten be small (a few microns in diameter) and close together (average spacings of 5 to 10 μ). The small pore size is necessary so that the predominant cesium transfer mechanism is surface diffusion which delivers cesium to the front surface of the tungsten,

rather than vapor flow which allows cesium vapor to pass out the end of the pore without being ionized. Cesium ions are emitted from an annular region surrounding each operative pore. This region is typically a few microns across; its size is determined by a competition between the rate of diffusion of cesium along the surface and the rate at which ions evaporate from the surface. In order that there not be large fractions of the surface which do not emit ions, then, the distance between pores must be comparable to the size of individual emitting regions.

The curves presented in Figs. 4, 5, and 6 apply to tungsten surfaces with uniform surface coverage and uniform emission and are thus not directly applicable to rear-fed porous tungsten on which emission and surface coverage have significantly different values at different points on the surface. Figure 7 shows some data taken with porous tungsten fabricated at EOS; it is plotted in the same way as Fig. 6. Comparing the two curves we see that the

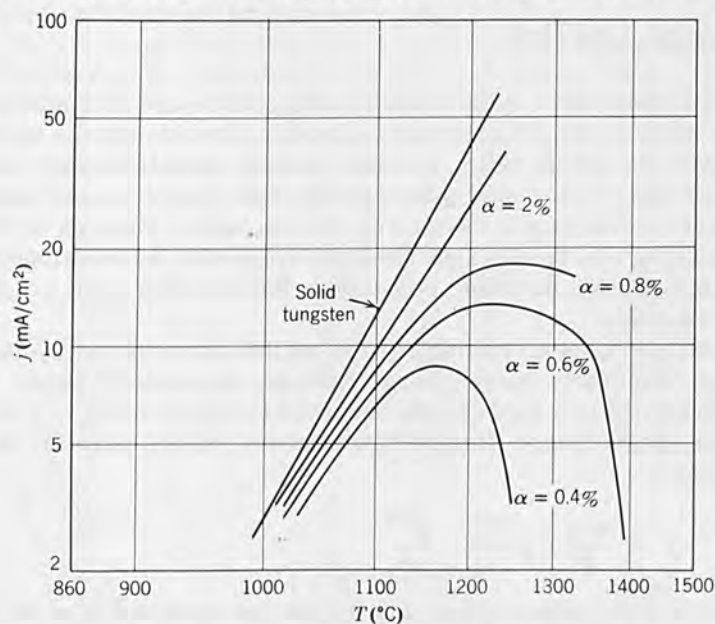


Fig. 7. Cesium ion current density obtained from porous tungsten.

porous tungsten is capable of producing almost the same maximum current at a given temperature, but that at a given current the neutral efflux from the porous tungsten is noticeably higher. This can be traced to two sources: vapor flow of neutrals from the pores,

and the fact that in order to achieve a given average current density, local current densities must be several times the average and local surface coverages must be several times the average.

In summation, the surface ionization process appears to provide a very promising ion source for ion motors. Ions of a single species are formed at a well-defined surface with very low energies, providing perfect conditions for the formation of a uniform velocity collimated ion beam. The neutral efflux is of the order of 1%, a negligible quantity as far as propellant utilization is concerned. Whether this value is low enough so that electrode erosion arising from charge exchange interactions is not prohibitive has not been completely resolved.

The power required to generate ions is only the power radiated from the ionizer structures. By operating at high ion current densities very attractive power efficiencies may be obtained.

ION ACCELERATION

The acceleration system and the ion source are intimately related. Many of the requirements placed on the ion source apply equally to the acceleration system; it must operate reliably for long periods of time, and must operate with power losses small compared to the power carried by the ion beam. Erosion of the accelerating electrode is particularly important as mentioned earlier; this erosion probably represents the ultimate limit to ion engine lifetime.

Although in actual operation, ion engine acceleration systems are not completely space-charge limited, questions of space charge and perveance are necessarily involved in engine design. The equation for space charge limited flow between infinite parallel planes is given by

$$i = \frac{4\epsilon_0}{9} \sqrt{\frac{2e}{m}} \frac{\Phi^{3/2}}{d^2}$$

From this, the perveance of a unit cube (an aperture of area d^2 at separation d) can be found to be

$$\rho = \frac{4\epsilon_0}{9} \sqrt{\frac{2e}{m}}$$

For cesium ions, this is equal to $4.7 \cdot 10^{-9}$ amp/ $V^{3/2}$, and furnishes an estimate of the perveance achievable from a circular source (or from a length of slit source equal to the source width).

Thus with a potential of 2000V, corresponding to an exhaust velocity of $5.5 \cdot 10^4$ meter/sec, a current of approximately $420 \mu\text{A}$ may be obtained. Since for useful propulsion devices, beam currents of tens of amperes will be required, space charge considerations thus imply that thousands of individual beams will be required. For purposes of discussion an array of circular sources will be assumed although an array of slits could also be used.

The effective perveance of an accelerating geometry can be increased significantly by using an accelerate-decelerate arrangement. Figure 8 shows a typical geometry. The final electrode is operated at zero potential, representing approximately the potential of the spacecraft being propelled. The ionizer is operated at positive potential; this potential determines the final velocity of the ions and is thus determined by mission considerations. The accelerating electrode is operated at a negative potential which may be chosen for best performance. Figure 9 shows some experimental data taken with an accelerating structure of the type shown. As the accelerate decelerate ratio $(\Phi_0 + \Phi_1)/\Phi_0$ is increased the beam perveance increases and the gun perveance decreases

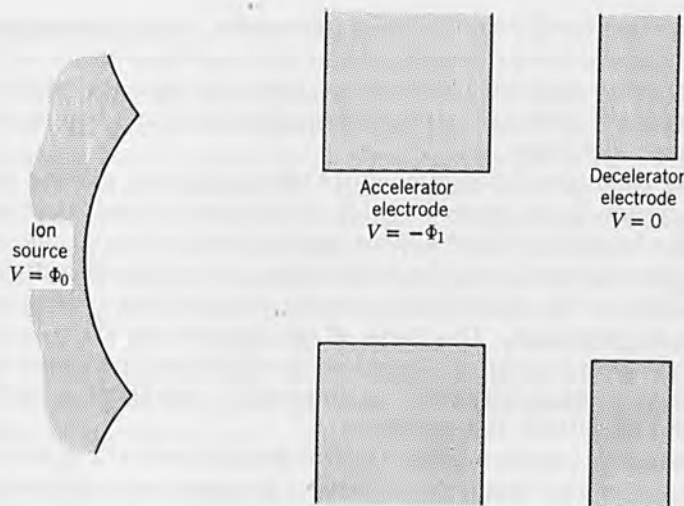


Fig. 8. Typical electrode configuration.

slightly. In the limit of small accelerator aperture diameter, the current depends only on the potential difference across the accelerating gap and the gun perveance is constant. In practice, however, the fields are influenced by the potentials of all three electrodes and the perveance decreases slightly as the accelerate-decelerate ratio is increased.

Φ_0	Φ_1	R	I	p_{beam}	p_{gun}
1.50	12.0	9.0	2.52	27.0	1.62
1.75	11.3	7.5	2.56	20.4	1.72
2.00	11.5	6.7	2.70	17.5	1.72
3.00	10.5	4.5	2.70	9.5	1.72
4.00	8.0	3.0	2.70	5.2	2.08

- Φ_0 Ionizer potential, kilovolts.
 $-\Phi_1$ Accelerator potential, kilovolts.
 R $(\Phi_0 + \Phi_1) / \Phi_0$.
 I Unit source ion current, milliamperes.
 p_{gun} $I [\Phi_0 + \Phi_1]^{-3/2} (10^{-9} \text{ amp/V}^{3/2})$.
 p_{beam} $I [\Phi_0]^{-3/2} (10^{-9} \text{ amp/V}^{3/2})$.

Fig. 9. Experimental accelerator performance.

In addition to increasing beam perveance, thus allowing operation at higher current density, the accelerate-decelerate arrangement prevents electrons, introduced into the beam for neutralization purposes, from traveling back up the beam and bombarding the ionizer structure.

It has been found experimentally that a physical ground potential electrode is not necessary for accelerate-decelerate operation. The boundary of the approximately zero potential plasma forms downstream from the accelerating electrode at a distance comparable to the ionizer-accelerator spacing and acts as a decelerating electrode. The shape of the plasma boundary is not exactly the same as that of a physical electrode so the shape of the beam is somewhat different, but in general operation is very similar with and without the electrode.

In order to improve engine performance several aspects of ion optic designs are being investigated. The goal is to achieve a design which has high perveance, uniform emission over the ionizer surface, and low interception by the accelerator of ions, either directly from the ionizer or produced by charge-exchange inter-

actions. High perveance allows operation at lower accelerating potentials, which in turn implies both reduced accelerator erosion by sputtering and reduced high-voltage breakdown. Uniform space-charge limited current density over the ionizing surface implies increased ionization efficiency and reduced accelerating voltage. Low interception is, of course, important from the standpoint of operating lifetime.

The approach being taken to the ion optics problems makes use of machine computation for the analysis of possible engine configurations. The procedure is an iterative one in which Poisson's equation is solved (initially with zero-charge density). From the calculated potential, distribution particle trajectories are then computed. From the trajectories the first approximation to charge density in the region is computed. This charge density is used in the second solution of the Poisson equation; new trajectories and space charge are computed and the procedure is repeated until the desired accuracy is obtained. On the basis of the final potential distribution the trajectories of ions formed at various places can also be determined. Figure 10 shows a section of a typical axisymmetric geometry and a space-charge limited solution. Figure 11 shows the trajectories of some charge-exchange ions. Although this computer technique is still being developed, it promises to be a very useful one.

NEUTRALIZATION

Since the topic of beam neutralization is covered in detail elsewhere, it will only be mentioned here. For the purposes of engine development, space operation is simulated by allowing the interior surface of the test chamber to "float," thus insuring that ion and electron currents are closely matched. The principal effort in this area is devoted to the development of efficient electron sources with characteristics compatible with engine operation. The two main energy losses, those associated with thermal radiation from the cathode and with the potential difference between the cathode and the beam, must be kept to a minimum. At the same time the source must be kept far enough away from the beam so that it is not damaged by stray ions.

ENGINE CONSTRUCTION AND OPERATION

The problem in ion engine design is one of combining the elements discussed above into a workable unit. The methods to be

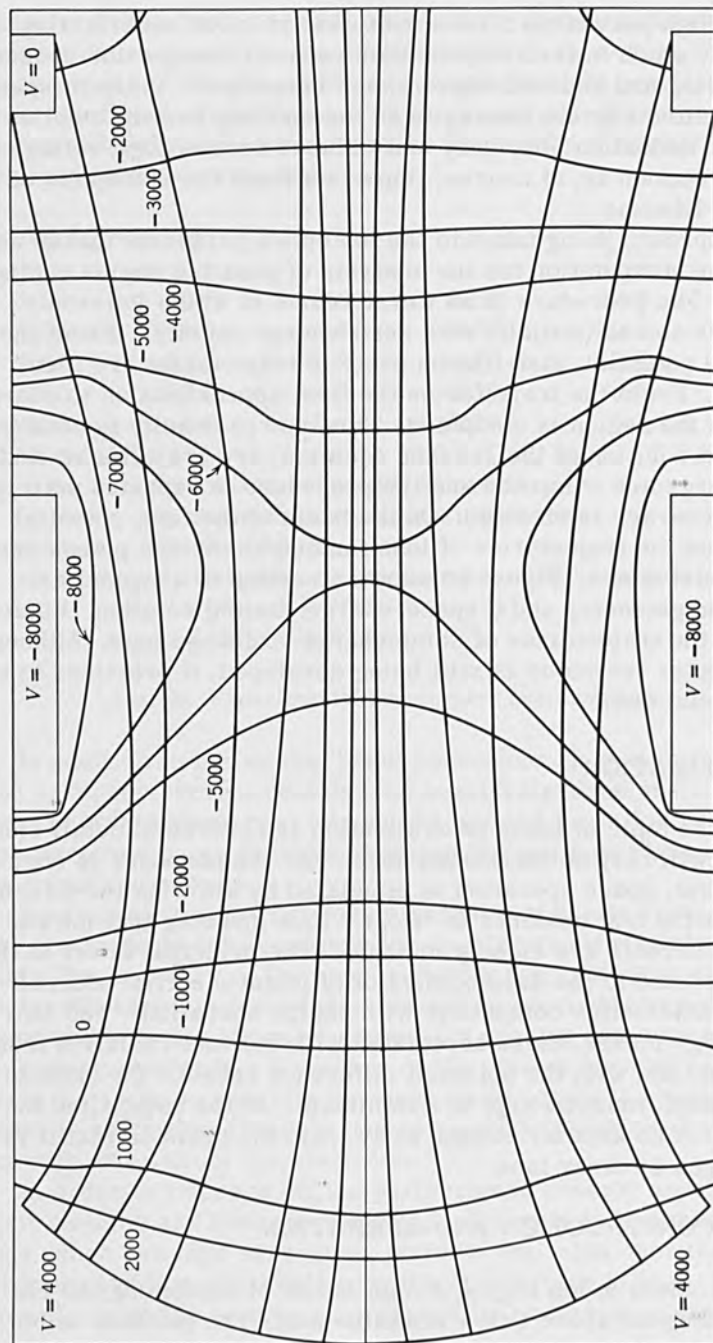


Fig. 10. Equipotentials and trajectories.

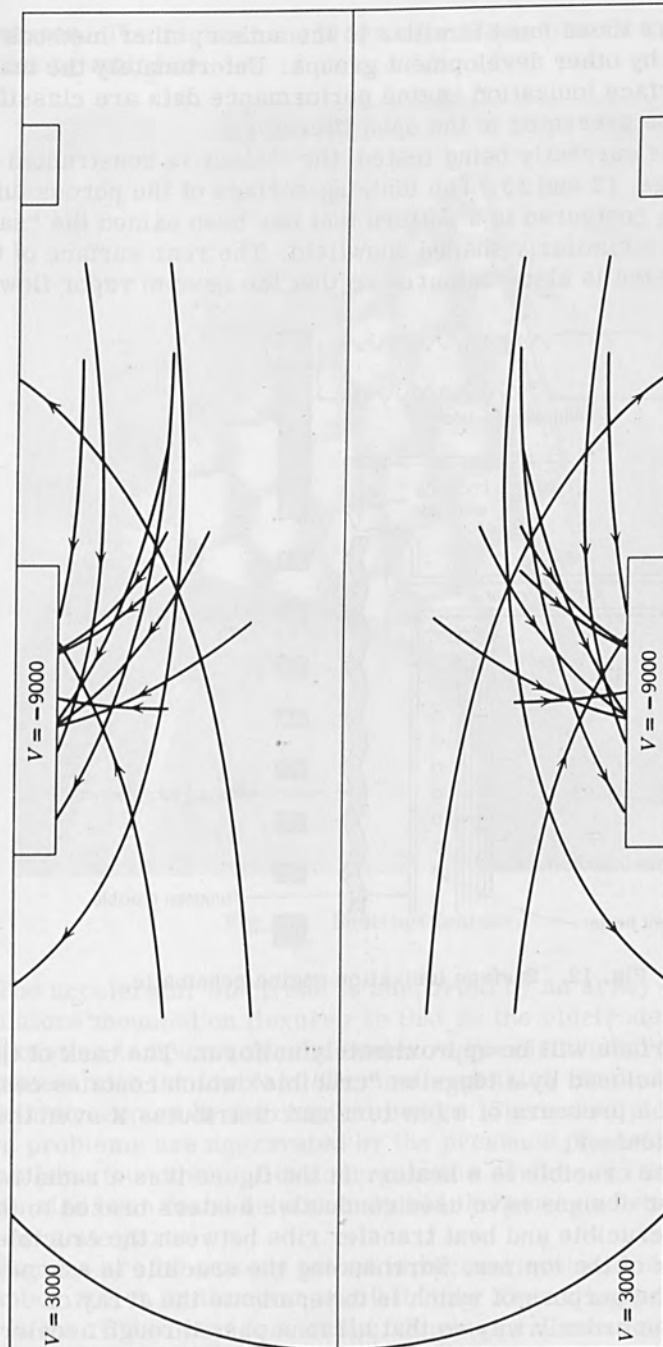


Fig. 11. Charge exchange trajectories.

described are those most familiar to the author; other methods are favored by other development groups. Unfortunately the majority of surface ionization engine performance data are classified and cannot be presented in the open literature.

In engines currently being tested, the ionizer is constructed as shown in Figs. 12 and 13. The ionizing surface of the porous tungsten plate is contoured in a pattern that has been named the "sas-trugi" after a similarly shaped snowfield. The rear surface of the porous tungsten is also contoured so that the cesium vapor flow to

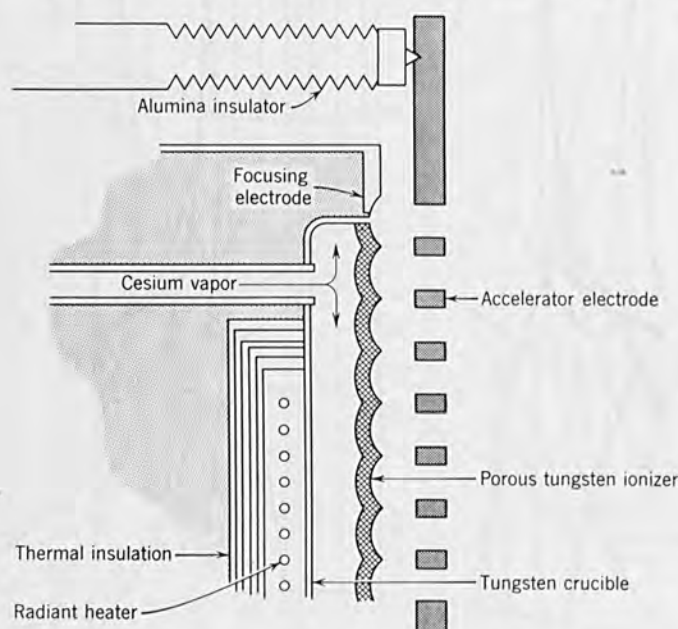


Fig. 12. Surface ionization engine schematic.

the front surface will be approximately uniform. The back of the ionizer is enclosed by a tungsten "crucible" which contains cesium vapor at a pressure of a few torr and distributes it over the back of the ionizer.

Behind the crucible is a heater. In the figure it is a radiative heater; other designs have used conductive heaters brazed to the back of the crucible and heat transfer ribs between the crucible and the back of the ionizer. Surrounding the crucible is a focusing electrode, the purpose of which is to terminate the array of sources in an orderly way so that all ions pass through accelerator

apertures. The region around the crucible is filled with thermal insulating material.

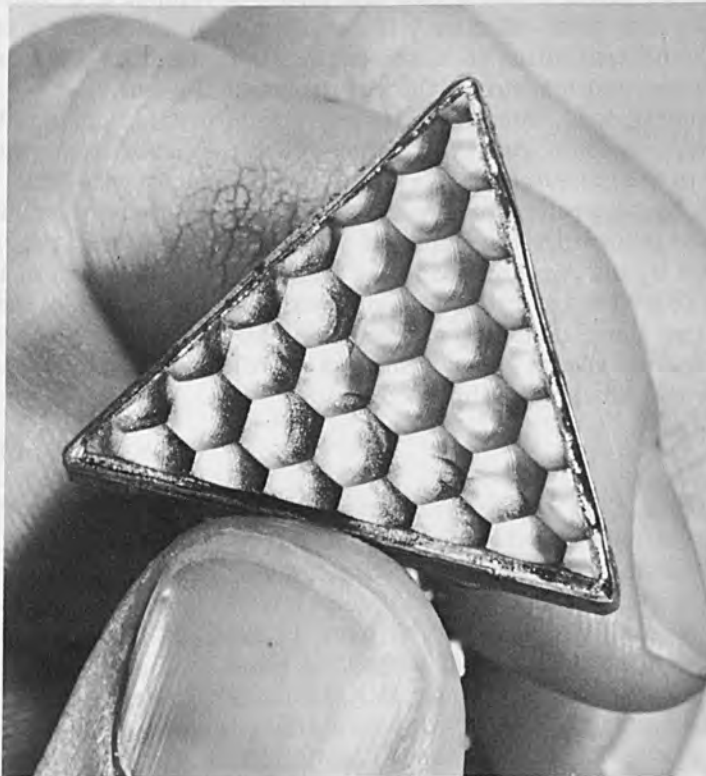


Fig. 13. Sastrugi ionizer.

The accelerator electrode is supported by an array of alumina insulators mounted on flexures so that as the electrode heats up to operating temperature, the center of the electrode remains in alignment with the ionizer. Thermal expansion must of course be taken into account in the original design. Since electrical breakdown problems are aggravated by the presence of cesium vapor, it has been found helpful to corrugate the surface of the insulators. It has also been found helpful to shield the junction between metal and ceramic parts.

Engines are laboratory tested in vacuum systems; a typical chamber is 3 ft in diameter and 9-ft long. Operating pressures are in the 10^{-5} to 10^{-7} torr range. Engine performance is meas-

ured electrically; currents and voltages delivered to electrodes and heaters are monitored and from these data, performance figures such as power consumption and thrust developed are calculated. The ionization efficiency is determined by collecting a fraction of the neutral efflux from the engine, ionizing it on a hot tungsten surface, and measuring the resulting ion current.

If properly done, laboratory testing should provide conclusive information on ion engine performance in all respects except those relating to beam neutralization which is influenced by the presence of a finite testing chamber. For this reason there exist, in addition to laboratory development and testing programs, programs sponsored by the Air Force and the NASA to test operation of ion engines aboard space probes fired on ballistic trajectories. Assuming that these tests are as successful as anticipated, the surface ionization engine will continue as a promising candidate for space propulsion applications.

A CESIUM PLASMA CAPILLARY EMITTER AS A SOURCE OF CESIUM IONS OR A SOURCE OF ELECTRONS OF HIGH-CURRENT DENSITY†

INTRODUCTION

The problem of the hollow cathode (cavity or cylindrical tube) as a source of electrons has been an extensive field of investigation for a long time. As a general conclusion it was stated that the electron current density, which can be extracted directly through the orifice, is much smaller than that which would be obtained if the electron emission of the whole inside walls of the hollow cathode would contribute to the electron efflux.

In the case where the hollow cathode is an infinite cylindrical tube, Weyman [1] has shown that the mean electron number density $\bar{n}$ in the cross section is the geometrical mean of the electron number density n_s at the tube wall, and the value n_0 , which is the electron number density on the tube axis. Since $n_0 < n_s$, the electron current density which could be extracted from the cross section of this tube is always smaller than the emission from an equal area of the pure surface of the wall metal.

When the hollow cathode is a cavity, a similar conclusion was given already by Jain and Krishnan [2]. Later, Brodie and

*CSF—Compagnie generale de telegraphie Sans Fil Centre de Physique Electronique et Corpusculaire Domaine de Corbeville, Orsay, S. & O., France.

†This paper is the result of work partially supported under Contracts (41/62 and 63-34205-00-280-75-01) with the Direction de Recherches et Moyens d'Essais of the French Ministere des Armees.

Niewold [3] stated that an effusion hole of a thermionic hollow cathode behaves like a conventional thermionic cathode with the same thermionic constant as the interior walls of the cavity, provided the diameter of the effusion hole is smaller than a certain value given by the space-charge density at the inside wall. With increasing diameters, a hollow beam is obtained, which means that the mean electron current density becomes lower and lower.

Since it was recognized that the limiting parameter is the electron space charge inside the two hollow cathode structures, the purpose of this investigation was to study the influence of the introduction of positive ions into a hollow structure of a highly refractory metal heated to high temperature. The positive ions are produced by thermal ionization of cesium vapor fed into the structure. Alternatively it was interesting to investigate the possibility of using such a structure as a cesium ion source.

Two structures were investigated: a small cylindrical tube (called "capillary emitter"), and a cylindrical cavity with an effusion hole on the top side.

EXPERIMENTAL STUDY OF CAPILLARY EMITTERS

Several devices have been examined, each provided with a small cylindrical tube of tantalum or molybdenum, with an inner diameter of 0.4 mm and a length of 10 mm. As already shown [4], some of these devices were provided only with an anode-probe at about 1 mm of distance to the outlet of the tube (Fig. 1a), others were gun structures, Fig. 1b. Typical I - V curves obtained with a probe structure are shown on Fig. 2a and with a gun structure on Fig. 2b.

The saturated electron and ion current densities measured with these structures are summarized in Figs. 3 and 4.

These results show that: (1) anode-probe structures and gun structures are giving the same saturation levels for the electron current density; (2) the electron current densities do not depend on whether the tube is made of tantalum or of molybdenum; and (3) the electron current densities cannot be confused with the values which correspond to the electron emission of tantalum or molybdenum if immersed in cesium vapor at a vapor density corresponding to the cesium pressure at the inlet.¹ (If the cesium vapor pressure at the outlet—which is unknown—would be chosen, the

¹The emission properties were taken from the article by Nottingham [5].

current density curves for tantalum and molybdenum would be shifted to lower values of the current densities.)

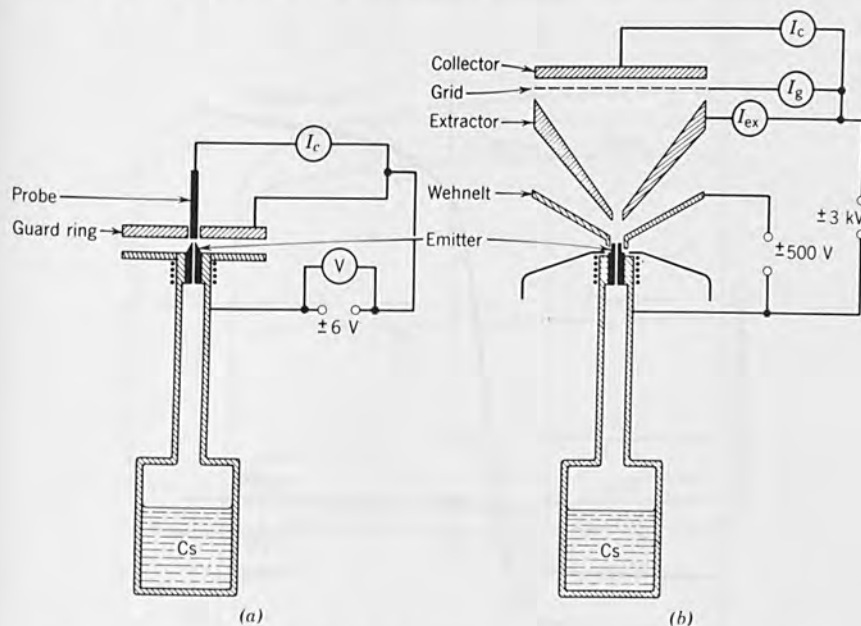


Fig. 1. Configuration of the test devices. (a) Capillary emitter with an anode-probe structure. (b) Capillary emitter with a gun structure.

The interpretation of these results, though qualitatively easy, becomes difficult when quantitative agreements with theoretical models is asked. The reason is the unknown pressure at the outlet of the tube.

Several hypotheses exist, especially for the ion emission of a cylindrical tube: (1) the cesium atoms migrate on the inside wall and evaporate as ions from the edge of the tube, as proposed for the case of a porous structure where the pores can be compared with cylindrical tubes of very small diameter [6], [7]; (2) the cesium ions are directly emitted from the inside wall through the open end of the tube; and (3) inside the tube a plasma is formed of which electrons or ions are extracted [4].

The fact that the tube operates under flow conditions does not make easy the verification of these hypotheses. Therefore a quasi-

static operation condition was investigated by replacing the cylindrical tube by a cavity provided with a small hole and put into communication with a cesium reservoir.

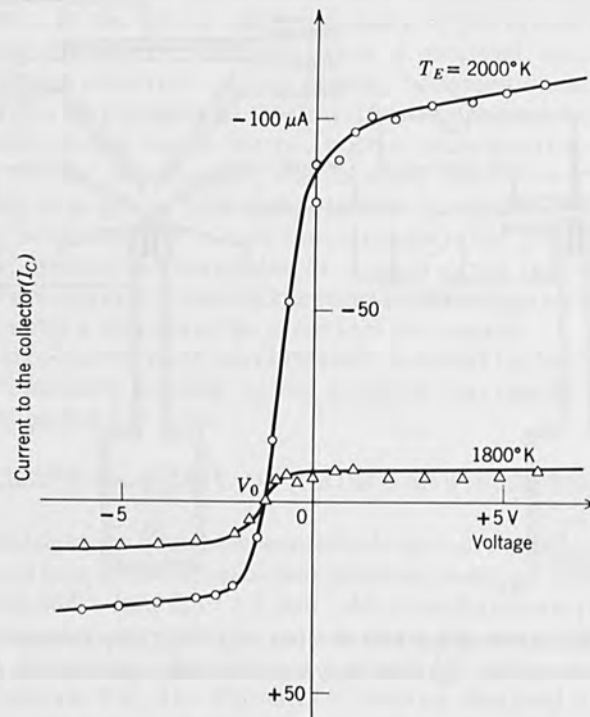


Fig. 2. (a) $I - V$ curves obtained with a capillary emitter probe structure for $T_{cs} = 453^\circ\text{K}$ and $d_e/C = 0.9\text{ mm}$.

STUDY OF A CAVITY EMITTER

The effusion hole of the cavity had the same diameter as the open section of the capillary emitter (0.4 mm). The cavity itself was a relatively great cylinder of an inner diameter of 10 mm and of a length of 15 mm. So, the ratio of the inside wall area to the hole area was 5000; and the effusion currents through the hole were supposed not to disturb the equilibrium pressure inside the cavity.

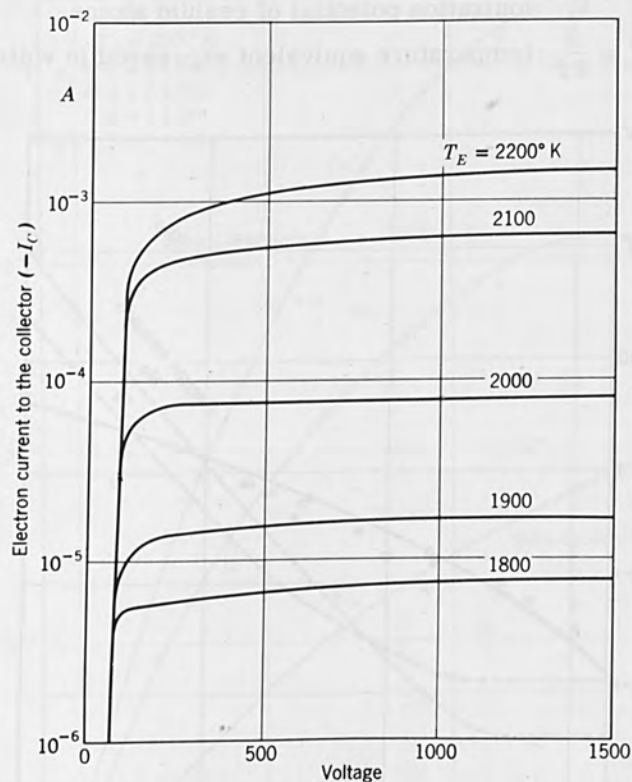


Fig. 2. (b) $I - V$ curves obtained with a capillary emitter gun structure for $T_{cs} = 400^\circ \text{K}$.

Theoretical Model

Under these conditions the cavity may be considered as an isothermal enclosure. It is known that under this condition a cesium plasma is formed whose density $\pm n_p$ of electrons and ions is given by the Saha equation:

$$\frac{\pm n_p^2}{n_0} = \left(\frac{2\pi - mkT}{h^2} \right)^{3/2} e^{-V_i/\bar{V}} \quad (1)$$

n_0 density of the cesium atoms under equilibrium conditions

$$\bar{V} \equiv \frac{V_i}{kT} \quad \begin{array}{l} V_i \text{ ionization potential of cesium atoms} \\ \text{temperature equivalent expressed in volts.}^2 \end{array}$$

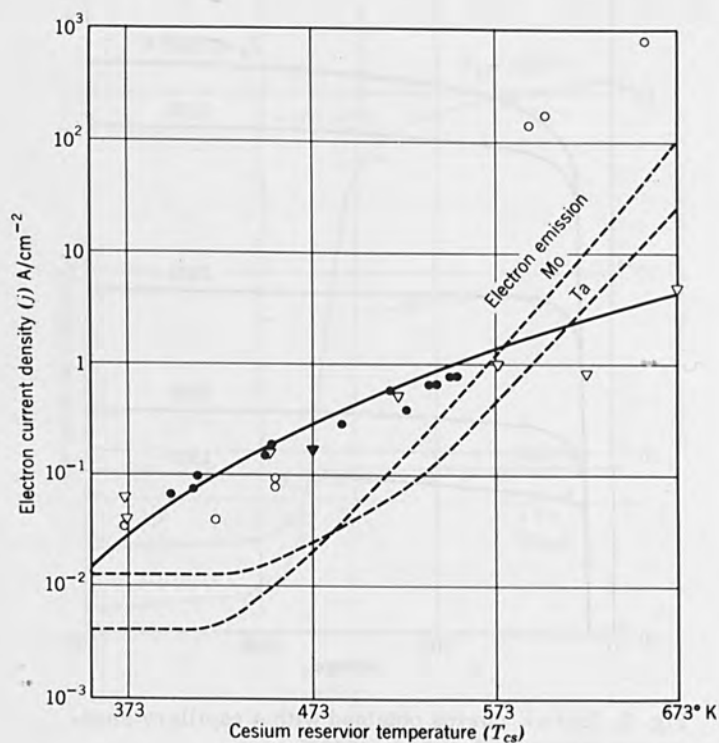


Fig. 3. Electron current densities measured with capillary emitter structures at -3 V polarization above the floating potential for the anode probe structure and at -1.5 kV for the gun structure. Comparison with theory.

²Alternatively, this equation may be written in the form:

$$\frac{\pm n_p^2}{(1-f)n_g} = \left(\frac{2\pi mkT}{h^2} \right)^{3/2} e^{-V_i/\bar{V}} \quad (1a)$$

n_g cesium atom density before dissociation, $f = \frac{\pm n_p}{n_g}$ fractional ionization. When f is very small, the gas density before dissociation may replace n_g .

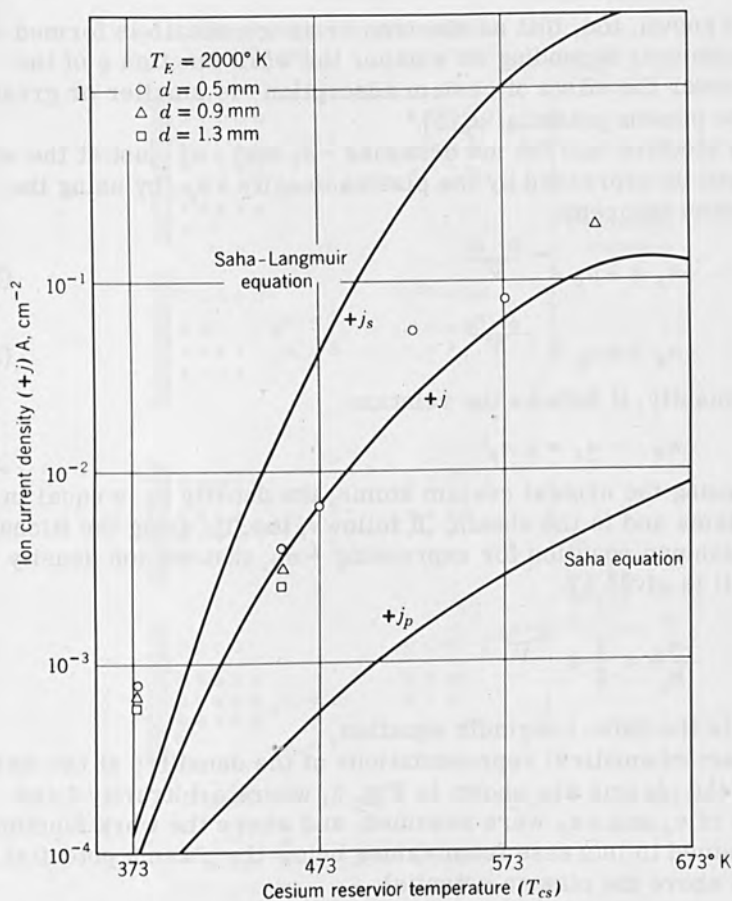


Fig. 4. Ion current densities measured with the molybdenum capillary anode-probe structure at -3 V polarization below the floating potential. Comparison with theory.

Inside the volume occupied by the plasma, the random currents V_p of electrons and ions are given by:

$$-V_p = \pm n_p \frac{-\bar{v}}{4} = \pm n_p \sqrt{\frac{kT}{2\pi - m}} \quad (2a)$$

$$+V_p = \pm n_p \frac{+\bar{v}}{4} = \pm n_p \sqrt{\frac{kT}{2\pi + m}} \quad (2b)$$

where $-\bar{v}$ and $+\bar{v}$ as well as $-m$ and $+m$ are respectively the mean velocities and the masses of electrons and of ions.

It is known, too, that an electron or an ion sheath is formed at the inside wall depending on whether the work function ϕ of the wall—under the effect of cesium adsorption—is smaller or greater than the plasma potential μ_p [5].³

The electron and the ion densities $-n_s$ and $+n_s$ just at the wall may then be expressed by the plasma density $\pm n_p$, by using the Boltzmann theorem:

$$-n_s = \pm n_p e^{-\frac{\phi - \mu_p}{V}}, \quad (3a)$$

$$+n_s = \pm n_p e^{\frac{\phi - \mu_p}{V}}. \quad (3b)$$

Consequently, it follows the relation:

$$+n_s \cdot -n_s = \pm n_p^2 \quad (4)$$

Concerning the neutral cesium atoms, the density n_0 is equal in the plasma and in the sheath. It follows, too, by using the Richardson-Dushman equation for expressing $-n_s$, that the ion density at the wall is given by:

$$\frac{+n_s}{n_0} = \frac{1}{2} e^{\frac{\phi - \mu_p}{V}} \quad (5)$$

which is the Saha-Langmuir equation.

Some schematical representations of the densities at the wall and in the plasma are shown in Fig. 5, where arbitrarily fixed values of n_0 and $\pm n_p$ were assumed, and where the work function is assumed to increase from values below the plasma potential to values above the plasma potential.

Experimental Study

A cavity was built of tantalum, put into a gun structure similar to that shown in Fig. 1b, and heated by electron bombardment. The following information was obtained:

Electrons Currents. As in the case of the capillary emitter, the electron current density extracted of the effusion hole does not correspond to the electron current density emitted by the inside

³ It may be emphasized that the plasma potential is dependant only on the ionization potential of the gas, of its temperature and of the initial (before dissociation) gas density, but not on the work function of the inside wall.

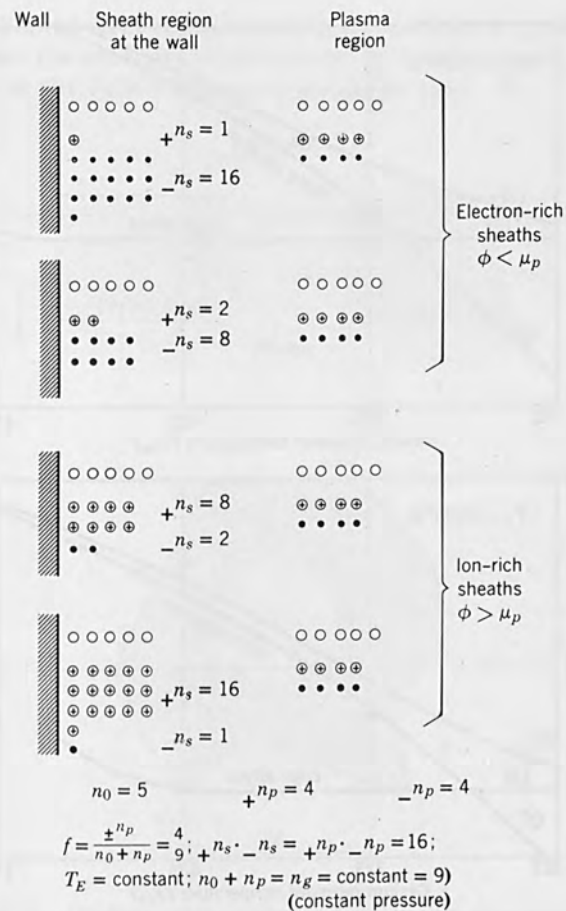


Fig. 5. Schematic representations of the neutral ions and electron number densities at the wall and in the plasma.

wall, but to the random current efflux through the hole of the cavity, hence given by the Saha equation as indicated in Fig. 6. Curve (1) gives the electron efflux through the hole after the Saha equation for temperatures of 2100°K and 2000°K, and the cesium vapor densities corresponding to the cesium reservoir temperature indicated on the abscissa:

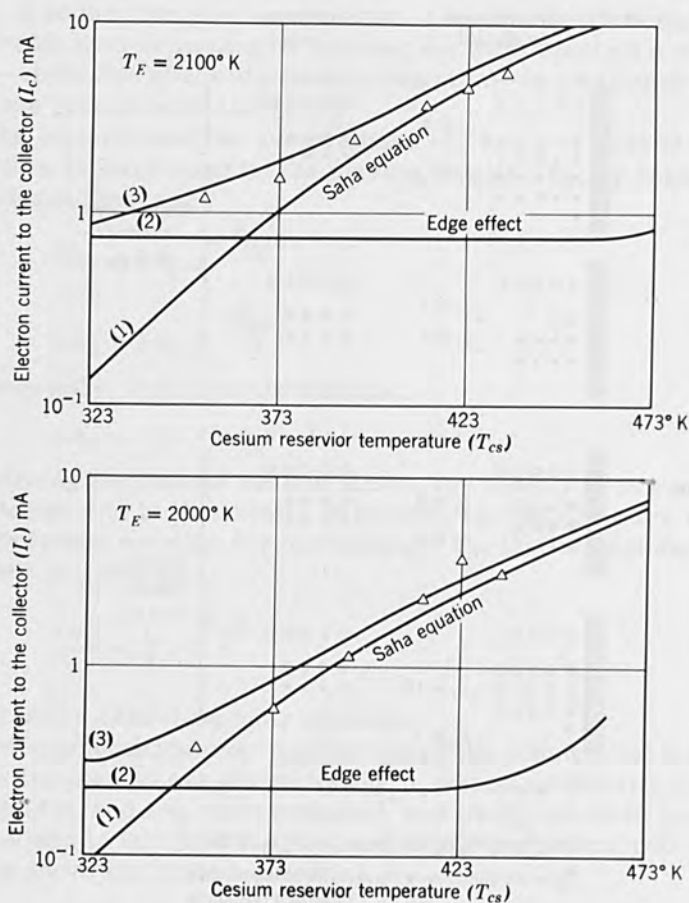


Fig. 6. Comparison between theory and experiment for the tantalum cavity emitter. Electron currents.

$$I_p = \frac{\pm n_p - \bar{v}}{4} \cdot \pi R^2$$

If the Richardson-Dushman electron emission at the edge of the hole is taken into account by measuring (curve 2), the agreement between the theoretical model and the measurement seems quite good.

Ion Currents. If there is a good agreement for the electron extraction, it is not so the case for the ion extraction which is about

ten times higher than indicated by the ion random current in the plasma. But the interpretation seems to become clear if the real conditions at the hole edge are considered (Fig. 7):

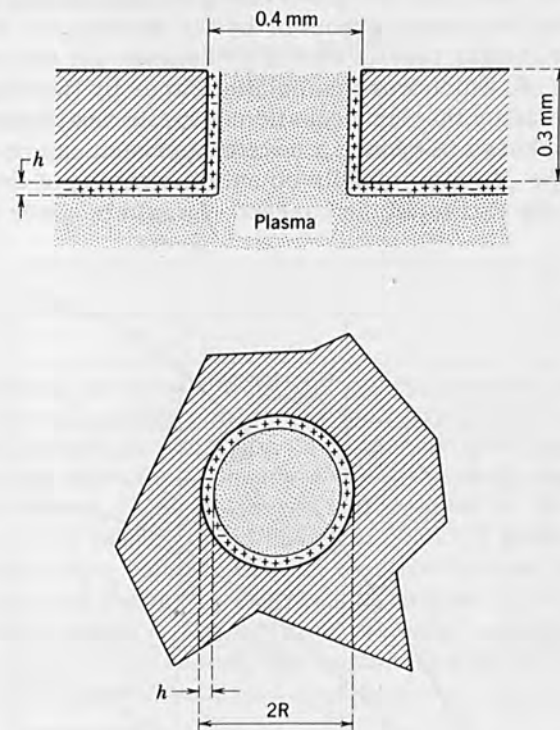


Fig. 7. Configuration of the real cavity effusion hole, showing the plasma and the sheath region.

1. The real hole is not an orifice of infinitely small thickness; in reality it is a small tube.
2. Under the interesting operating conditions, an ion sheath of small thickness is formed at the inside wall up to the open end of the hole.

When electrons are extracted, the presence of the ion sheath and the thickness of the cavity orifice make no great difference with the effusion flux from a hole of infinitely small thickness, for the ion sheath is low in electrons and has small thickness. However, when ions are extracted, it has to be kept in mind that the ion

sheath is much richer in ions than the plasma. So the extracted ions come out, as well of the plasma, as of the sheath. If $+I_s$ and $+I_p$ are the ion currents extracted from the hole when the ion density is uniform, and they are given either by the density at the wall $+n_s$ (giving an excess error) or by the density $+n_p$ in the plasma, the experimental results should be between the two curves indicated in Fig. 8, which were calculated either by the Saha-Langmuir equation or by the Saha equation. This is what happens. As the sheath thickness is known, a nearer approach can be made assuming that the density $+n_s$ fills up only the region of the sheath. In this case, the extracted ion current $+I$ would be given by:

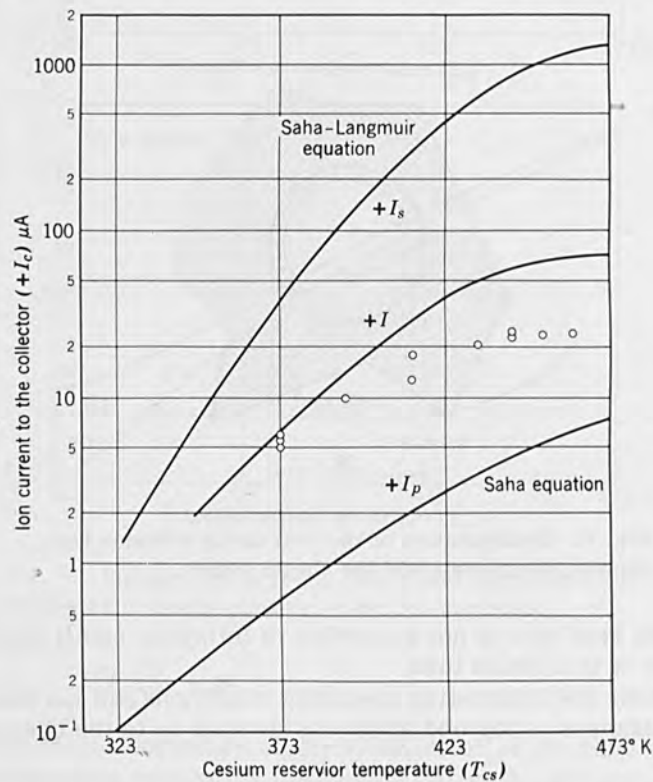


Fig. 8. Comparison between theory and experiment for the tantalum cavity emitter. Ion currents.

$$+I \approx \left(1 - \frac{2h}{R}\right) + I_p + \frac{2h}{R} + I_s$$

The sheath thickness h was calculated by the solution of the Boltzmann-Poisson equation [5]:

$$T_{\text{cavity}} = 1900^\circ\text{K} \\ (\text{tantalum})$$

T_{cs}	h
350°K	22.5 μ
400°K	10 μ
450°K	5.7 μ

Conclusion⁴

Summarizing the situation for the cavity, it can be stated that the experimental results concerning the extraction of electrons and of ions through an effusion hole in the cavity fit quite well with the predictions from the following model: the cavity contains an isothermal plasma which is limited with respect to the inside wall of the cavity by a sheath. This sheath is ion rich under the most chosen temperature conditions. The electron current which can be extracted is given mainly by the random current of the plasma, since the contribution of the ion-rich sheath is negligible. However, when ions are extracted, the contribution of the ion-rich sheath becomes predominant.

APPLICATION OF THE CAVITY EMITTER CONCLUSION TO THE CAPILLARY EMITTER

Trying to apply the cavity model to the results of the capillary emitter means to interpret the extracted electron and ion currents by the random currents in the cavity model.

Electron Currents

Figure 3 shows that the experimental points for $T_E = 2000^\circ\text{K}$ have the same behavior as the electron random current of the

⁴ The curve is indicated in Fig. 8. Considering that this calculation corresponds to an excess error, assuming constant density $+n_s$ through the whole sheath, the agreement between this modified model and the experimental results looks satisfying.

plasma in a cavity. When it is tried to fit the experimental points by an electron random current curve, it appears that the plasma density $\pm n_p(L)$ at the outlet of the capillary should be $1/20 \pm n_p(0)$ at the inlet.⁵

Furthermore, the same ratio is fitting the electron current for other capillary temperature, as is seen in Fig. 9.⁶

Hence, the experimental result seems to be satisfactorily fitted by assuming at the capillary outlet a plasma whose density is given by the equation:

$$\pm n_p(L) = \frac{1}{20} \pm n_p(0)$$

The problem is to understand this plasma fraction. Indeed, if we would have only a flow of neutral cesium atoms under Knudsen flow condition, then the neutral flow would be given by:

$$\Gamma_k = \frac{8R^3}{3L} \left(\frac{\pi}{2 \cdot m k T} \right)^{1/2} \Delta P$$

where $\Delta P = P(0) - P(L) = kT[n_g(0) - n_g(L)]$. The neutral flux density would be:

$$V_k = \frac{8R}{3L} \left(\frac{kT}{2\pi \cdot m} \right)^{1/2} [n_g(0) - n_g(L)]$$

Alternatively, this flux density would be given by the effusing neutral flux:⁷

$$V_k = \frac{n_g(L) \cdot \bar{v}}{4} = n_g(L) \left(\frac{kT}{2\pi \cdot m} \right)^{1/2}$$

So, it follows that

$$n_g(L) = \frac{n_g(0)}{1 + 3L/8R}$$

⁵In some older measurements, extraneously high electron current densities of more than 100 amp cm⁻² have been found in a capillary emitter diode with an anode probe at voltages of about 3 V (see Fig. 3). This may be due to an effect similar to the arc mode in a thermionic converter [8], [9].

⁶The fact that at relatively low cesium reservoir temperatures the experimental points are higher than the random currents may have the same reason as that for the cavity (edge emission).

⁷This is certainly not quite correct, since the velocity distribution is not isotropic.

which gives:

$$n_g(L) \simeq \frac{1}{20} n_g(0)$$

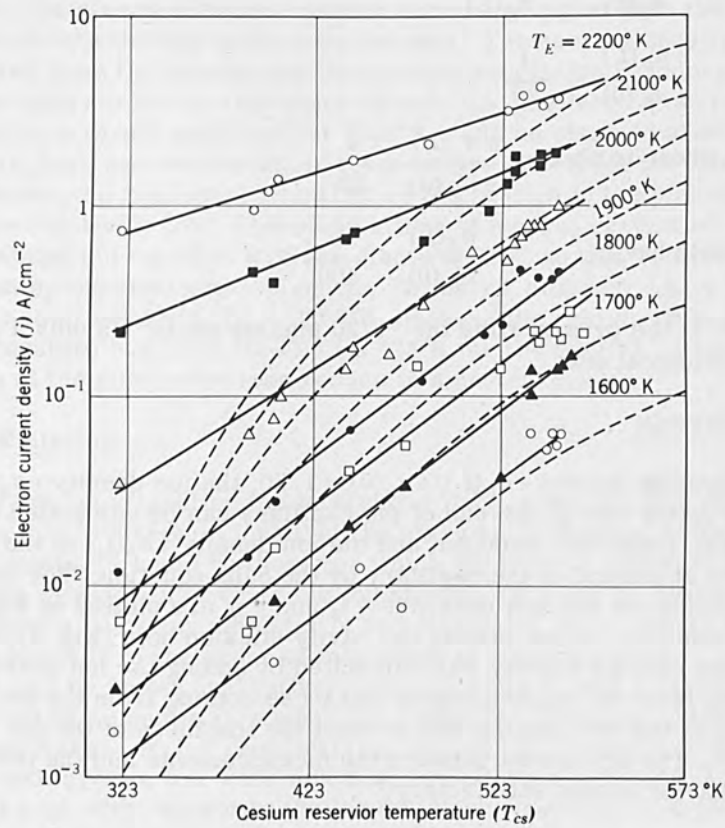


Fig. 9. Comparison between theory and experiment for the capillary emitter. Electron gun.

for the experimental capillary with $L = 10$ mm and $R = 0.2$ mm. But, if the Saha equation can be applied in the outlet of the capillary, this is not compatible with

$$\pm n_p(L) = \frac{1}{20} \pm n_p(0)$$

Since the fractional ionization will be relatively small throughout the capillary, the Saha equation would give:

$$\frac{\pm n_p^2(0)}{n_g(0)} \simeq \frac{\pm n_p^2(L)}{n_g(L)}$$

$$\text{or: } \frac{\pm n_p(L)}{\pm n_p(0)} \simeq \frac{1}{\sqrt{20}}$$

$$\text{and in order to obtain } \frac{\pm n_p(L)}{\pm n_p(0)} = \frac{1}{20}$$

$$\text{we should have: } \frac{n_g(L)}{n_g(0)} = \frac{1}{400}$$

Hence, at the present time the "1/20 plasma model" is only a phenomenological one.

Ion Currents

Supposing always $\pm n_p(L)/\pm n_p(0) = 1/20$, the ion density $\pm n_s(L)$ at the inside wall at the end of the capillary can be calculated by the Saha-Langmuir equation, and the ion density $\pm n_p(L)$ in the plasma at the end of the capillary by the Saha equation. The corresponding ion current densities $\pm j_s$ and $\pm j_p$ are plotted in Fig. 4. As it was done in the case of the cavity effusion hole (Fig. 8), the real ion current density was calculated by adding the ion currents coming from the plasma region and those coming from the ion sheath at the wall, taking into account the thickness of the ion sheath. The agreement between the measurements and the predicted behavior seems satisfactory.⁸

GENERAL CONCLUSIONS

Capillary emitters, as well as cavity emitters with a small hole may be used either as an electron source or as an ion source (cesium ions). For both emitters the favorable temperature conditions are those for which the plasma is separated from the inside wall by an ion-rich sheath.

⁸ However it has to be noted that with one of the tantalum capillary ion guns, an ion density of 480 mA/cm² under 9 kV has been measured at 1800° K.

Electron Emission

In the electron extraction the interesting part is the cesium plasma, since this is richer in electrons than the ion-rich sheath.⁹ Thus, the plasma electron random current is higher than the electron current density emitted by the wall. The current density extracted from the plasma can be equal to the plasma random current if the wall surface is important enough. So, an electron current density up to 100 amp/cm^2 at 2200°K could be obtained at sufficiently high cesium reservoir temperature (600°K for the cavity). However, the fractional ionization of the cesium in the plasma remains relatively low. Therefore a flow of neutral cesium is streaming out together with the electron flow, so that volume ionization by the electrons can occur. So, only 8 amp/cm^2 have been obtained presently at $T_E = 2200^\circ\text{K}$, because the cesium reservoir temperature had to be limited (to 423°K for the cavity). A special form of the electrodes may reduce this neutral flux.

Ion Emission

In the ion extraction the interesting part is the ion-rich sheath, since this is richer in ions than the plasma. So the best geometrical conditions would be those for which the plasma region would be largely reduced in favor of the ion sheath. This can be realized by choosing a capillary diameter of the dimension of the sheath thickness which is of the order of magnitude of some microns.

It may be shown qualitatively what happens if such a capillary is crossed by a cesium flow when it is heated to higher and higher temperatures. At low temperature, the cesium coverage at the inside wall gives a low work function; the electron emission is relatively high with respect to the ion emission (electron-rich sheath). Since the capillary has a diameter of the order of magnitude of the electron sheath, the volume inside the capillary is filled up by an atmosphere with electrons as majority carriers. Ion emission in this case occurs by a surface migration flow of neutral cesium atoms to the edge of the capillary and by evaporating there in form of ions (Fig. 10a).

When the temperature becomes sufficiently high, the coverage of cesium will diminish, and consequently the work function will

⁹ The inverse case of an electron-rich sheath is not interesting for electron extraction, since the area of the hole section could be replaced advantageously by an area of the wall material.

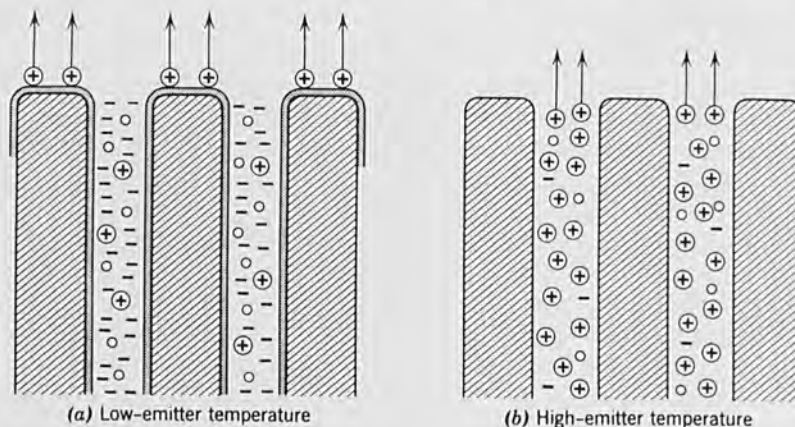


Fig. 10. Schematic view of the ion emission. (a) At the edge of the capillary. (b) From the open section of the capillary.

increase. So the ion emission inside the capillary will become predominant and an atmosphere with ions as majority carriers is formed. For example, for a capillary temperature of 1700°K and an equivalent cesium reservoir temperature of 400°K at the outlet, an ion-rich sheath of a largeness of $18\ \mu$ is formed with an ion density at the capillary wall of about 10^{14} ions per cm^3 . Therefore, the ions extracted from the capillary in this case come predominantly from the open section of the capillary and less from the edge area (Fig. 10b). This is what has been observed in the very instructive experiences made by Forrester, Kuskevics, and Marchant [10] with an ion-image microscope.

When ions are extracted from an orifice with a diameter of the dimension of the sheath thickness, a high ion current density can be obtained; for example, for tantalum an ion current density up to $3\ \text{amp}/\text{cm}^2$ (of the hole) at $T_E = 2000^{\circ}\text{K}$ could be obtained at adequate cesium temperature (500°K for the cavity). With a molybdenum capillary emitter of 0.4-mm diameter, an ion current density of $0.20\ \text{amp}/\text{cm}^2$ has been obtained only because we have a plasma in the capillary outlet.

A certain number of applications can be foreseen using the capillary emitter: (1) as an electron source for evaporation, fusion or welding in an electron gun structure; (2) as an ion source for machining or surface treatment by sputtering.

LIST OF SYMBOLS

d_E/C	= emitter-collector spacing.
f	= fractional ionization.
h	= Planck's constant.
h	= sheath thickness.
$+I$	= ion current.
$+I_p$	= random ion currents.
$+I_s$	= random ion currents.
$+j_p$	= random ion current density in the plasma.
$-j_p$	= random electron current density in the plasma.
$+j_s$	= random ion current density in the plasma.
$-j_s$	= random electron current density in the plasma.
k	= Boltzmann constant.
${}_om$	= atomic mass for cesium
$+m$	= ionic mass for cesium.
$-m$	= electron mass.
n_g	= number density of atoms before dissociation.
n_0	= number density of cesium neutral atoms in the plasma or the sheath.
$\pm n_p$	= number density of either electrons or ions in the plasma.
$+n_s$	= number density of ions in the sheath near the wall.
$-n_s$	= number density of electrons in the sheath near the wall.
$P(0)$	= cesium pressure at the inlet of the capillary.
$P(L)$	= cesium pressure at the outlet of the capillary.
q	= proton charge ($1602 \cdot 10^{-19} \text{ Cb}$).
R	= radius of the capillary and of the hole of the cavity.
T_{cs}	= cesium condensation temperature.
T_E	= emitter temperature.
$+\bar{v}$	= mean velocity of the ions.
$-\bar{v}$	= mean velocity of the electrons.
$\bar{V}$	= temperature equivalent expressed in volts.
Γ_k	= Knudsen flow.
μ_p	= plasma potential.
V_k	= neutral flow density.
$+V_p$	= random flow of ions in the plasma.
$-V_p$	= random flow of electrons in the plasma.
ϕ	= emitter work function.

REFERENCES

1. H. D. Weymann, "Electron density distribution in a cylinder," University of Maryland, Technical note BN 214, July 1960.
2. S. C. Jain and K. S. Krishnan, "The thermionic constants of metals and semi-conductors," *Proc. Roy. Soc. (London) A*, 213, 143-157 (1952); *A*, 215, 431-437 (1952); *A*, 217, 451-461 (1952).
3. I. Brodie and A. Niewold, "Zero field emission from thermionic hollow cathodes," *J. Appl. Phys.* 33, 3328-3334 (Nov. 1962).
4. H. Huber and R. Le Bihan, "Emetteur capillaire d'ions de césium: application à des convertisseurs thermioniques," Combustion and Propulsion Panel, AGARD-NATO, Athens, Greece (July 1963).
5. W. B. Nottingham, "Sheath and plasma theory of an isothermal diode," Topical Rept. for the Thermionic Emitter Materials Res. Program, Contract NONR 3563(00), Thermo Electron Eng. Co. (1962).
6. G. M. Nazarian and H. Shelton, "Theory of ion emission from porous media," *Electrostatic Propulsion*, edited by Langmuir, Stuhlinger and Sellen (Academic Press, New York, 1961), pp. 91-106.
7. D. Zuccaro, R. C. Speiser, and J. M. Teem, "Characteristics of porous surface ionizers," *Electrostatic Propulsion*, edited by Langmuir, Stuhlinger and Sellen (Academic Press, New York, 1961), pp. 107-139.
8. D. L. Dresser, W. G. Laurita, J. D. Dunlop, H. Huber, and R. Le Bihan, "Theory and experiment of a capillary-type emitter," *A.I.A.A. Journal* 2, 52-58 (Jan. 1964).
9. R. Breitwieser, "Cesium diode operation on three modes," Rept. on 23rd Annual Conference Physical Electronics, Mass. Inst. Tech., Cambridge, Mass., pp. 267-280 (1963).
10. A. T. Forrester, G. Kuskevics, and B. Marchant, "Ionization of cesium on porous tungsten," XIVth International Astronautical Congress, Paris, Sept. 25-Oct. 1, 1963.

A HIGH-INTENSITY MERCURY ION SOURCE OF DUOPLASMATRON TYPE

DESCRIPTION

Principle of the Source

The duoplasmatron consists of a low-voltage arc between a cathode F and an anode A , the discharge being strongly constrained by the electric field of a third electrode, the intermediate electrode IN , and by a very strong, inhomogeneous magnetic field parallel to the discharge in the region between IN and the anode A (Fig. 1). The resultant high-density plasma emerges from an aperture in the anode and diffuses within the expansion cup C . Ions are removed from the plasma and accelerated by the extraction electrode E , a few millimeters from A , operating at a negative potential of 20 to 30 kV.

Construction Details

To reduce the defocussing effect of the space charge of increasing importance with heavy ions ($M = 200$ for mercury), the rotational symmetry has been abandoned in favor of rectangular apertures.

Figure 2 shows the arrangement of the source: the tantalum filament forming the cathode F is situated within the central cavity of the intermediate electrode IN ; the latter, of mild steel, terminates in a truncated cone pierced by a slit F , $4 \text{ mm} \times 25 \text{ mm}$. The anode A , also of mild steel, is filled with a molybdenum insert containing an aperture of width e ($0.1 < e < 0.5 \text{ mm}$) and length $l =$

*Institut d'Electronique, Orsay, S & O, France.

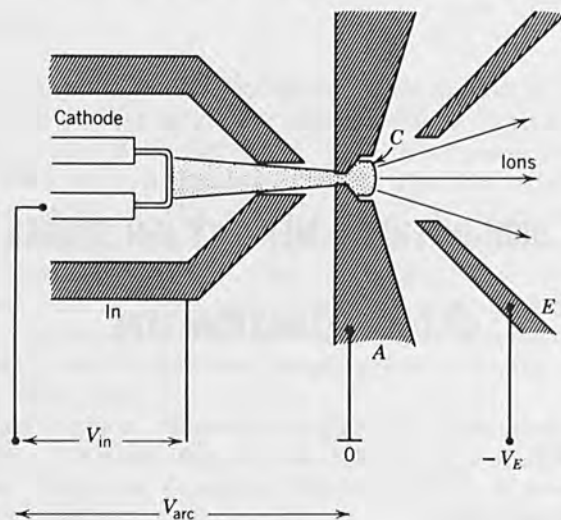


Figure 1.

20 mm. IN and A are electrically insulated by a quartz ring AI, 2-mm thick, which provides a leaktight closure of the discharge chamber between the high pressure region of the source and the high vacuum region of the extraction space.

The anode A is a mill steel rectangular parallelepiped, pierced by several channels, and free to move in a plane parallel to the front surface of IN. This permits alignment of the apertures without shutting down the source. The anode is grounded while the extraction electrode E is maintained at high negative potential V_E . E, in nickel-plated iron, is supported by porcelain insulators; it is cylindrically symmetric, and has a slit F_3 , 12-mm wide by 50-mm long. Permanent magnets produce a 200 gauss transverse field across the slit, preventing secondary electrons formed between E and the collector from returning to A.

Just after E, and carrying the same potential, V_E , is the collector C_o . A negatively polarized ring ($V \cong -5$ kV), between the two, stops secondary electrons emitted by C_o . A grounded pump circulates kerosene coolant through C_o by means of teflon tubing. The stainless steel vacuum chamber is evacuated to an ultimate vacuum of 5×10^{-6} torr using a liquid nitrogen baffled oil diffusion pump of 2000 l/sec speed.

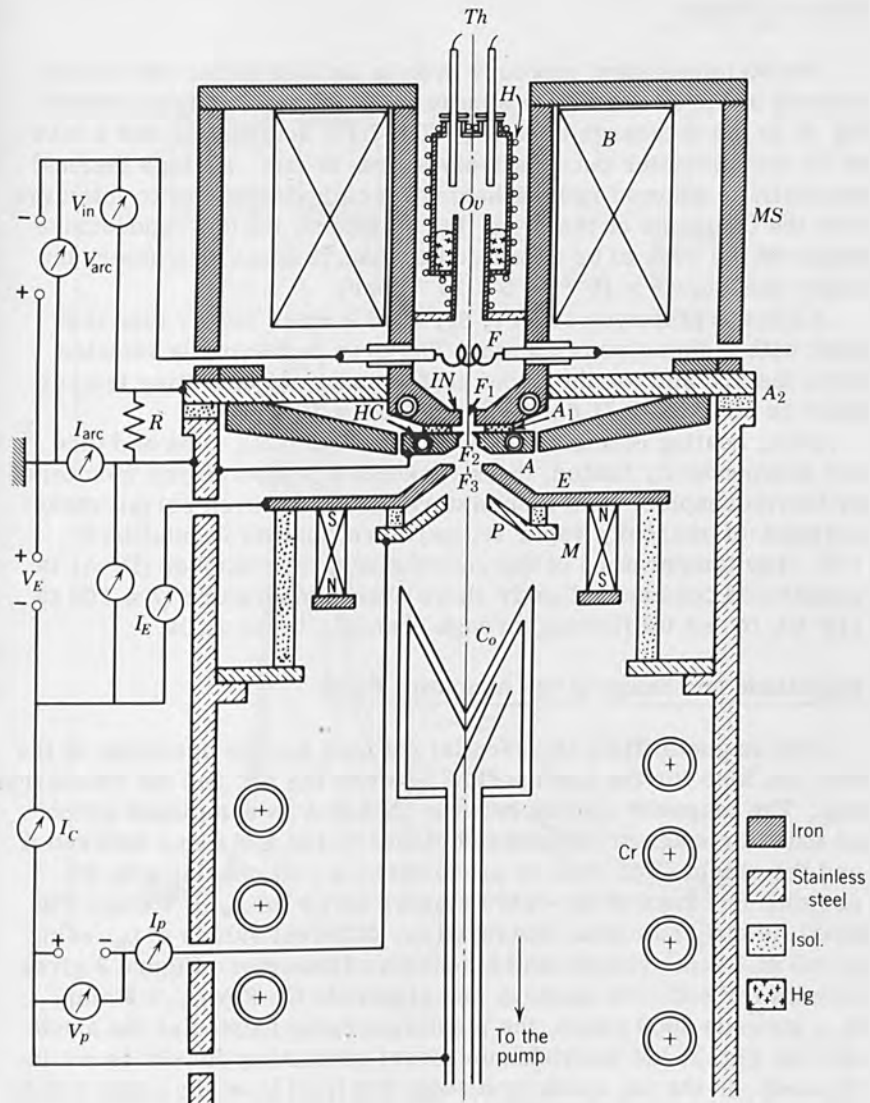


Figure 2.

Mercury Supply

The stainless steel mercury oven is located within the central channel of the intermediate electrode, a thin-walled tube connecting it to the discharge chamber. The first source, S_1 , had a tube of 10-mm diameter but relatively short: 40 mm. Its high thermal conductivity allowed radiant heat from cathode and arc to interfere with the operation of the oven. Furthermore, its low conductance required the oven to be operated between 75 and 115°C (mercury vapor pressure 6×10^{-2} to 5×10^{-1} torr).

A second prototype source, S_2 , used a much longer tube (150 mm) with a diameter of 20 mm. The oven is thermally isolated from the arc and for the same performance its operating temperature is 60 to 90°C ($1.6 \times 10^{-2} < p < 1.5 \times 10^{-1}$ torr).

Using heating elements soldered on their wall, oven and tube are independently heated, their respective temperatures monitored by thermocouples. Two thermistors inside the oven permit measurement of mercury vapor temperature, and its regulation to 1°C. The temperature of the entire discharge chamber (IN-A) is maintained constant, slightly above oven temperature ($T \cong 100$ to 110°C), by hot oil flowing through channels in its walls.

Magnitude and Shape of the Magnetic Field

The magnetic field is essential not only for the operation of the arc, but also for the plasma flow between the arc and the extraction cup. The magnetic circuit between IN and A is completed within an exterior magnetic shield and closes in the air space between A and the shield. The field is produced by a coil coaxial with IN, permitting a field of $nI = 2400$ ampere turns for $I_m = 4$ amp. The axial field B_z has been measured for different values of I_m using a coil of 1 mm³ volume and a sensitive fluxmeter. Figure 3 gives results for soft iron anode A and electrode E. For $I_m = 3$ amp, $B_{\max}$ exceeds 3000 gauss, the maximum being located at the anode slit (in Fig. 3, the molybdenum insert containing the slit is not indicated). In the ion extraction zone, the field is of the order 100 to 800 gauss. For a stainless steel anode, the $B(z)$ curves are broader, the maximum, which never exceeds 2600 gauss, is shifted slightly behind the anode. The field in the extraction region may exceed 2000 gauss. After improving the magnetic circuit, a field of 3500 gauss can be obtained for $I_m = 3A$.

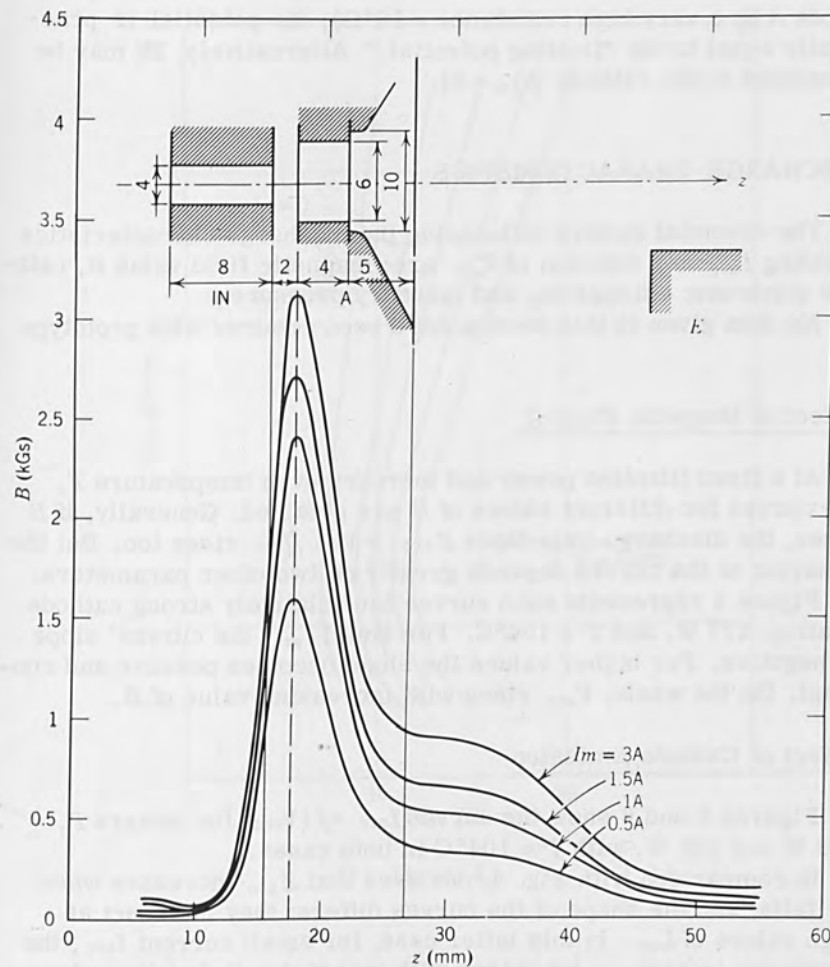


Figure 3.

Power Supply

The arc supply is adjustable between 0 and 200 V, with a maximum current of 12 amp. To reduce the number of parameters affecting the arc characteristics and to obtain readily reproducible results, the intermediate electrode is self-biased (IN connected to

anode A by a very high resistance $= 10^4 \Omega$), its potential is practically equal to the "floating potential." Alternatively, IN may be connected to the cathode ($V_{IN} = 0$).

DISCHARGE CHARACTERISTICS

The essential factors influencing the discharge characteristics yielding I_{arc} as a function of V_{arc} are: magnetic field value B , cathode electronic emission, and mercury pressure p .

All data given in this section have been obtained with prototype S_T .

Effect of Magnetic Field B

At a fixed filament power and mercury oven temperature T , the curves for different values of B are obtained. Generally, if B rises, the discharge impedance $Z_{arc} = V_{arc} / I_{arc}$ rises too. But the behavior of the curves depends greatly on two other parameters.

Figure 4 represents such curves for relatively strong cathode heating: 177 W, and $T = 104^\circ\text{C}$. For small I_{arc} , the curves' slope is negative. For higher values the slope becomes positive and constant. On the whole, V_{arc} rises with increasing value of B .

Effect of Cathode Emission

Figures 5 and 6 show the curves $I_{arc} = f(V_{arc})$ for powers $P_c = 153$ W and 129 W, with $T = 104^\circ\text{C}$ in both cases.

In comparison with Fig. 4, one sees that Z_{arc} increases when P_c falls, and the shape of the curves differs: they intersect at high values of I_{arc} . In this latter case, for small current I_{arc} , the discharge impedance increases with increasing B , but for values above the point of intersection, increasing B causes a decrease in Z_{arc} .

Effect of Pressure Variation

The real pressure inside the source is not precisely known. It is determined only by the mercury temperature T in the oven. A pressure rise has the same effect on Z_{arc} as an increase of electronic emission from the cathode.

Figure 7 shows results obtained at $T = 77^\circ\text{C}$ for $P_c = 153$ W.

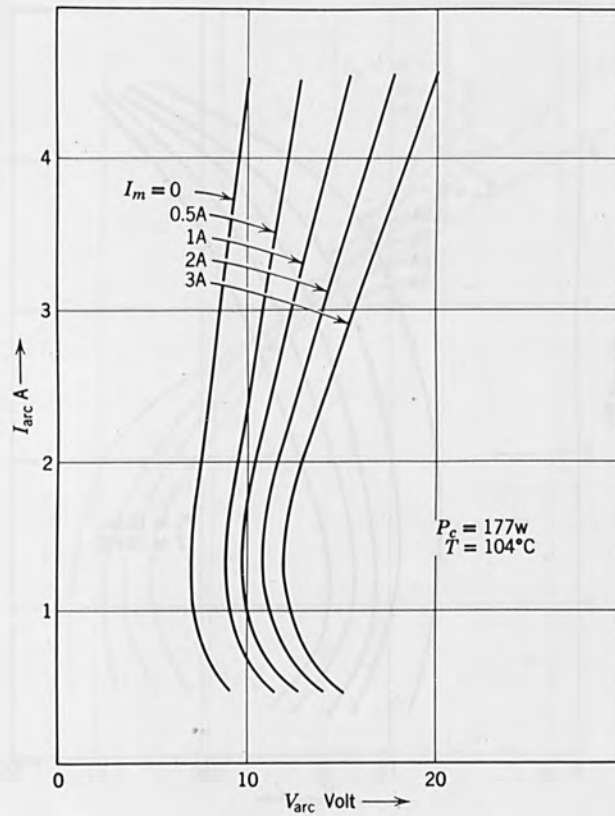


Figure 4.

On the same figure are the limiting curves corresponding to the value $T = 104^\circ C$, the heating power remaining fixed at $P_c = 153$ W.

MAXIMUM USEFUL ION CURRENT

The ion current extracted from the source plasma by the voltage on electrode E , V_E , and received by the collector depends essentially on the following factors: accelerating voltage V_E , arc current I_{arc} , magnetic field B , and potential difference between cathode and intermediate electrode V_N .

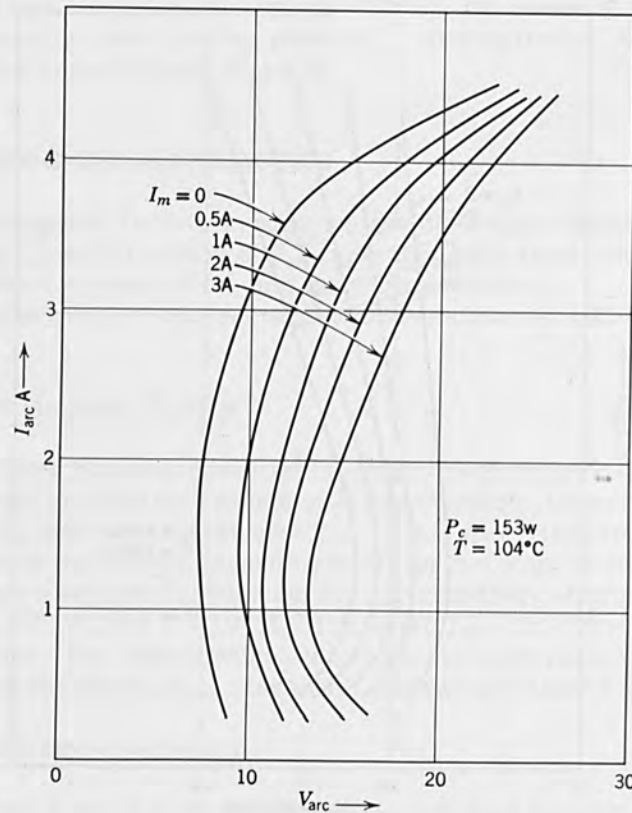


Figure 5.

The results presented below were obtained with prototype S_2 , the anode slit width being 0.3 mm.

Effect of Accelerating Voltage V_E

For a given set of operating conditions, the ion beam from the source plasma has an angular aperture α .

For low values of V_E , the boundary of the plasma is convex, and the beam highly divergent; part of the ion current strikes electrode E (extraction current I_E) and the remainder, the useful current, reaches the collector (current I_c).

If V_E increases, the plasma boundary recedes and tends to flatten; the aperture α decreases and I_E drops rapidly.

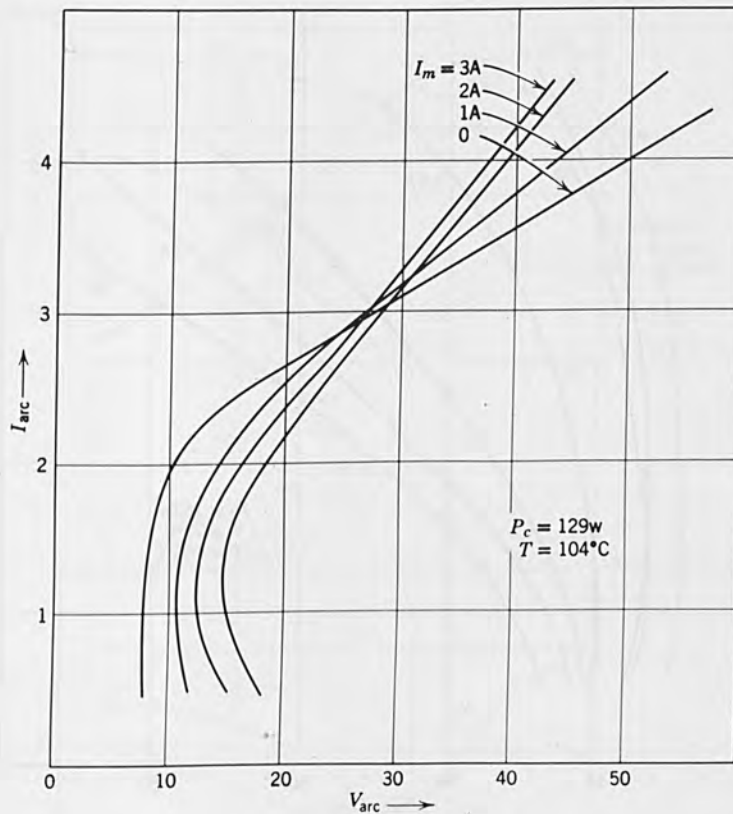


Figure 6.

For $V_E > 25\text{kV}$, all the extracted current reaches the collector, and its values remain constant with further increase in V_E .

Figure 8a shows the variation with V_E of collector current I_C and extractor current I_E . The polarized ring R , at potential $V_p \cong -5\text{ kV}$ with respect to the collector, stops all secondary electrons coming from the collector; I_C is therefore a true ion current.

However, I_E is the sum of the ion current to the electrode E , and the secondary emission electrons. The secondary emission coefficient γ of nickel (covering E) is not well known but is understood to be negligible for $V \leq 2\text{ kV}$, and is about 1 between 10 and 15 kV. To a good approximation the corrected ion current to E is $I'_E = I_E / (1 + \gamma)$. We therefore have: $I_{\text{total}} = I'_E + I_C$

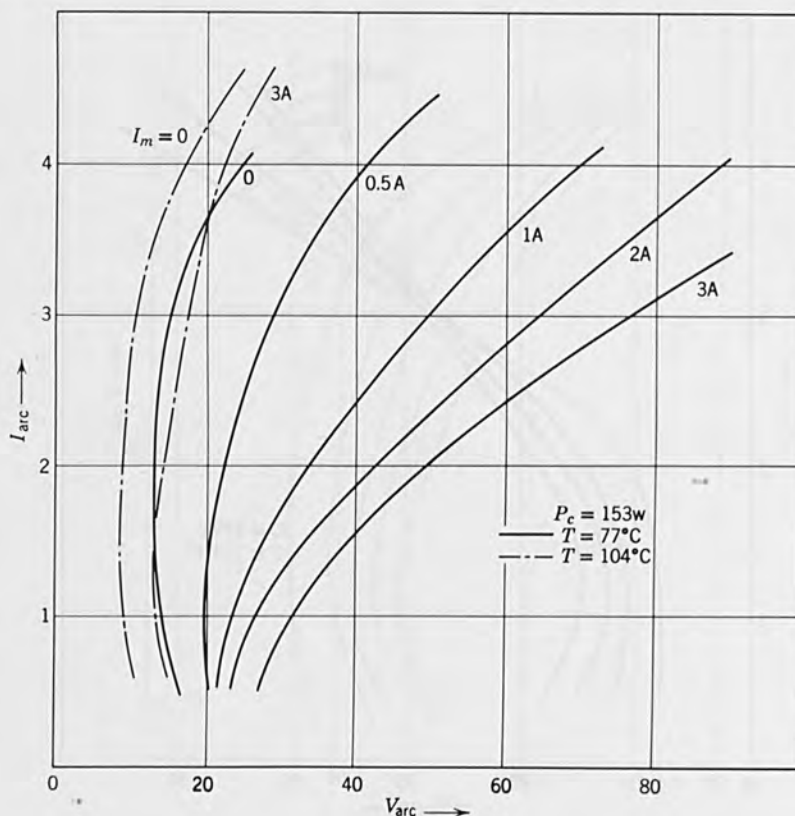


Figure 7.

One can see that the total extracted current remains constant, whatever the voltage V_E may be.

These results confirm those obtained with hydrogen [1] and argon [2], [3]. Duoplasmatrons: the duoplasmatron is a source of constant current. The extracted current depends only on arc characteristics, pressure p , and magnetic field B .

Still, it is recommended that V_E be increased in order to reduce the beam divergence and avoid interception of beam ions by the accelerating electrode.

With a slit 0.3-mm wide and an arc current I_{arc} of 10 amp, available ion current reaches 12 mA (Fig. 8b); beam divergence remains less than $\pm 5^\circ$. This result corresponds, at the slit plane, to a current density of more than 200 mA/cm².

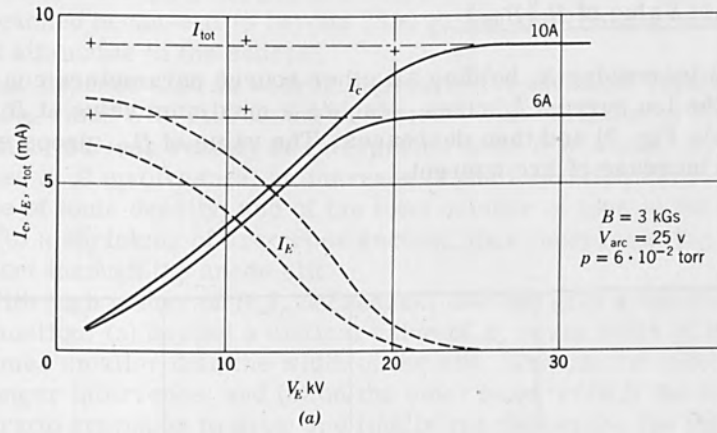


Figure 8a.

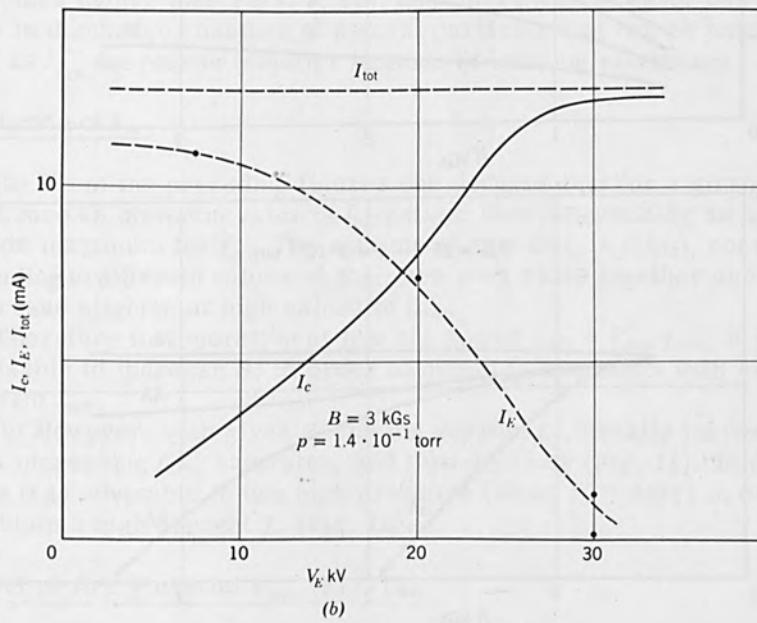


Figure 8b.

In the study below of the other factors influencing I_C , the accelerating voltage V_E is fixed between 25 and 30 kV.

Optimum Value of B (B_{opt})

With increasing B , holding all other source parameters constant, the ion current I_c rises, reaches a maximum value at $B = B_{\text{opt}}$ (see Fig. 9) and then decreases. The value of B_{opt} drops slowly with increase of arc current.

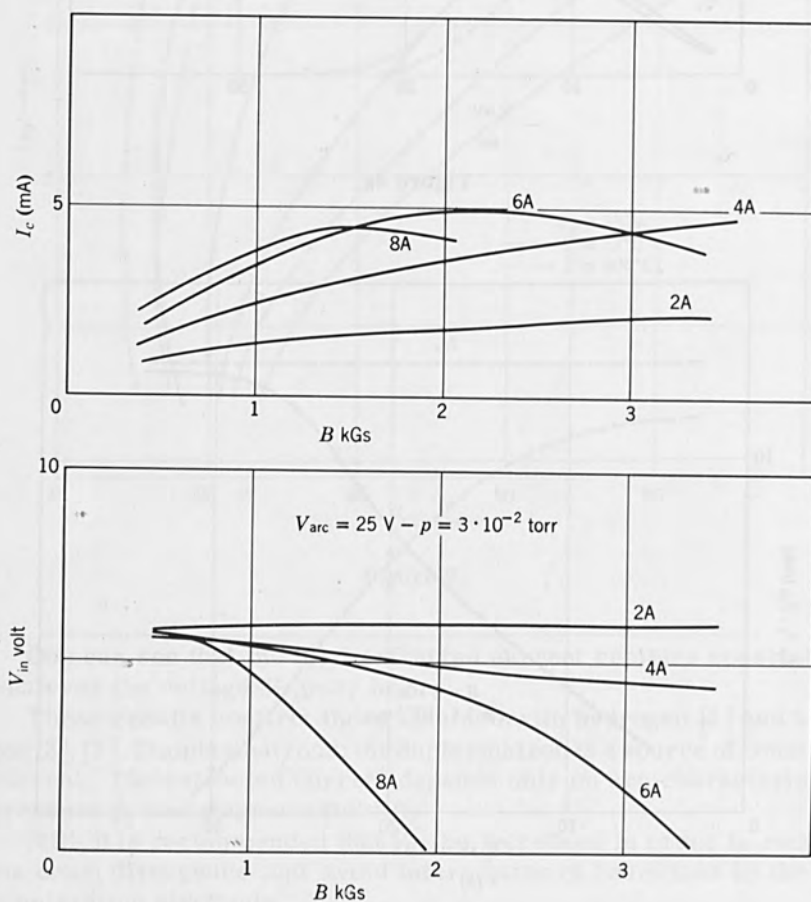


Figure 9.

For low intensity of I_{arc} , the corresponding value of B_{opt} cannot be reached because it is beyond 3500 gauss, the limiting value of field attainable in the source.

B_{opt} depends also on source pressure: for identical values of I_{arc} , B_{opt} rises with increasing pressure. This variation of I_c as a function of B can readily be interpreted when B is small; an increase of B involves: (a) an increase of the ionization ratio, and hence of ionic density, and of the total number of ions in the arc; and (b) a shrinking of arc cross section, thus increasing the ion current through the anode slit.

With high values of B , I_c decreases; one can give a tentative explanation: (a) beyond a critical value of B , beam width of the arc becomes smaller than the width of the slit. The constriction factor no longer intervenes; and (b) on the other hand, even if the ionization ratio continues to grow and finally reaches unity, the total number of ionized atoms passing through the anode slit will decrease.

This critical level of B , where the I_c curve changes direction, becomes higher (see Figs. 9, 10): as p increases (greater density in discharge chamber of neutral particles that can be ionized), and as I_{arc} decreases (smaller number of ionizing electrons).

Influence of I_{arc}

(a) From the preceding figures one deduces that for a given pressure an optimum value of I_{arc} exists, thus determining an absolute maximum for I_c . The maxima of curves $I_c = f(I_{\text{arc}})$, corresponding to different values of B_{opt} , are very close together and decrease slightly at high values of I_{arc} .

Therefore for operation at low arc power $p_{\text{arc}} = V_{\text{arc}} I_{\text{arc}}$, it is advisable to increase B , in order to obtain I_c maximum with low current I_{arc} .

(b) However, with given B , the ion current I_c initially increases with increasing I_{arc} , saturates, and then declines (Fig. 11). In this case it is advisable to use high pressure (about 10^{-1} torr) in order to obtain a high current I_c (Fig. 12).

Effect of Arc Potential V_{arc} (Fig. 13)

At fixed B and p , one varies V_{arc} while keeping I_{arc} constant:

(a) If IN is at floating potential, the current I_c remains constant, even with V_{arc} varying from 15 to 100 V.

The number of ionized particles drops slowly because at $V_{\text{arc}} >$

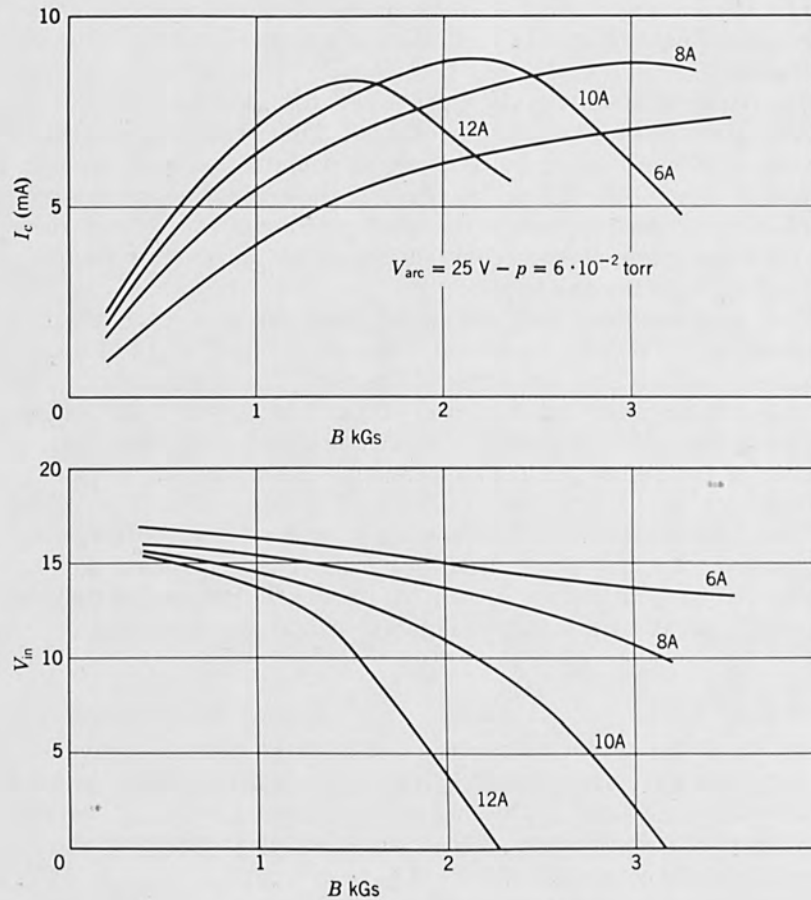


Figure 10.

40 V the probability of obtaining particles that are doubly or triply ionized, increases.

A q/M analysis of the ion beam should show a shift to multiple ionization with increasing V_{arc} .

(b) If $V_{IN} = 0$ (IN at cathode potential) I_c grows very slowly with increasing V_{arc} .

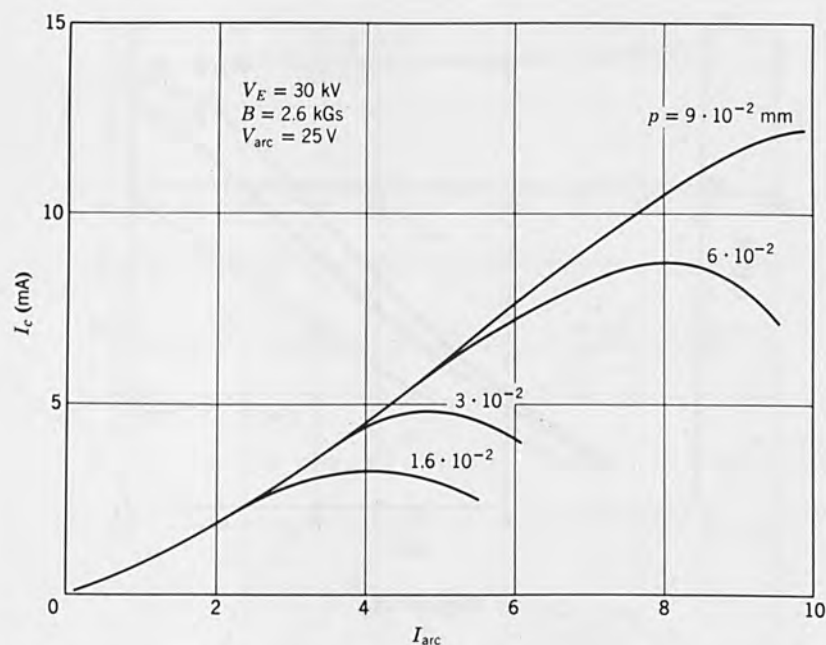


Figure 11.

Effect of Intermediate Electrode Potential V_{IN}

V_{IN} is the potential difference between intermediate electrode and cathode.

(a) in most cases IN is self-biased and at a potential near the floating potential. The data given above were obtained under these conditions. The curves traced in Figs. 9 and 10 represent the potential variation of IN as a function of B .

V_{IN} is practically constant for B small with respect to B_{opt} , but decreases rapidly as B approaches and exceeds B_{opt} . This variation of V_{IN} shows that the current in the bias resistor $R = 10^4 \Omega$ (connected to the anode) rises, and therefore that the difference between electronic and ionic currents reaching IN increases, the electronic component remaining larger.

(b) In some experiments IN was shorted to the cathode ($V_{IN} = 0$) and the current I_{IN} reaching IN was measured as a function of B . It was observed to be positive (ionic) and very intense at low B , becoming negative (electronic) for B greater than B_{opt} (Fig. 14).

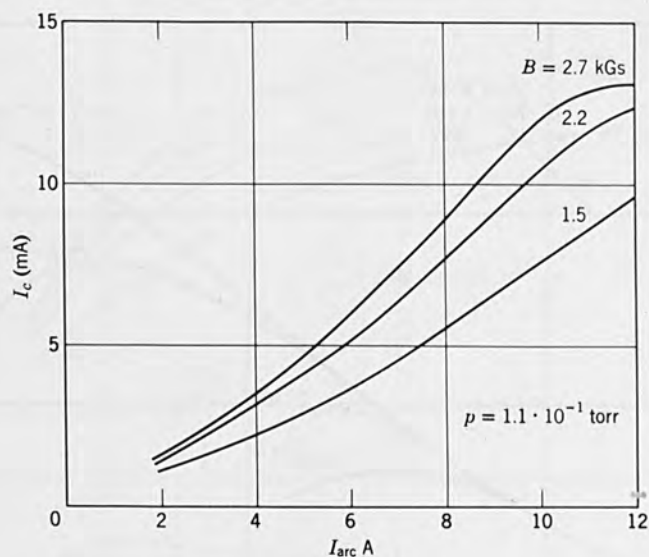


Figure 12.

But in both cases the curves of $I_c = f(B)$ show the same behavior with maxima of I_c practically the same.

(c) If V_{arc} is varied the difference $(V_{arc} - V_E)$ remains constant, provided that all other parameters are fixed; this law could be verified in all cases. In Fig. 13, the variation of V_{IN} is shown.

Effect of Pressure Variation

An increase in p is accompanied by an increase of the maximum value of ion current I_c (see Fig. 11). For a given arc current an increase of p allows operation at lower arc voltage V_{arc} and smaller cathode heating power.

CONCLUSION

We have constructed a mercury ion source, producing a useful mercury ion (Hg^+) current of 12 mA under the following conditions: anode slit 0.3×20 mm, arc current 10 amp, magnetic field strength 3000 gauss, and pressure 90μ .

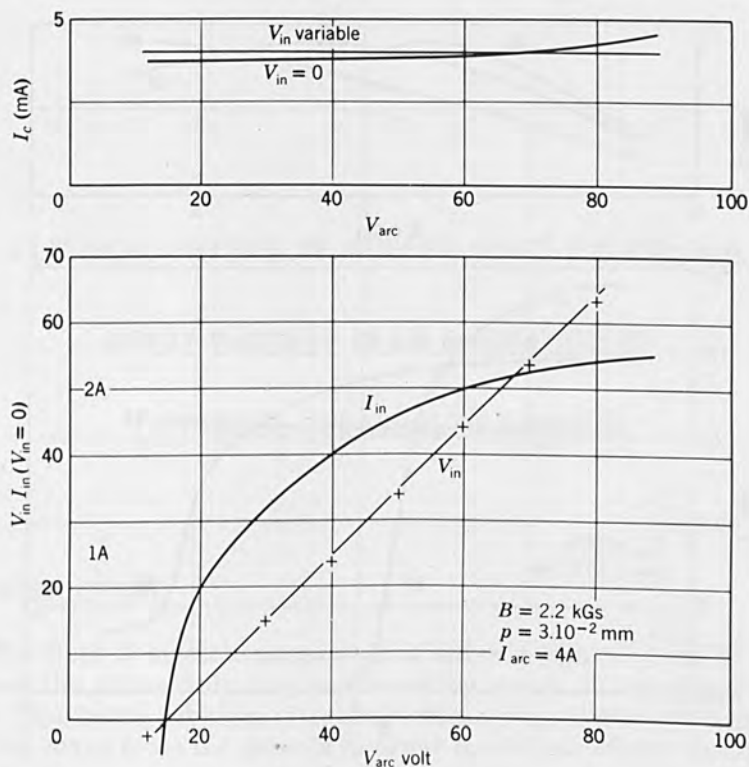


Figure 13.

Increasing the slit width to 0.45 mm results in a useful current of $I_c = 17 \text{ mA}$.

In all cases, for $V_E \geq 30 \text{ kV}$, the beam divergence is always less than $\pm 5^\circ$.

We have observed several interesting source characteristics:

- (1) the duoplasmatron is a current generator; the total ion current extracted from the plasma is independent of extraction voltage V_E .
- (2) There is an optimum magnetic field value B_{opt} , at which we have maximum I_c ; the maximum is very flat and almost independent of I_{arc} , in a large domain it depends only on the pressure.
- (3) The extracted current I_c is almost independent of V_{arc} .
- (4) With the intermediate electrode at floating potential, $(V_{arc} - V_{IN})$ is practically independent of V_{arc} , other parameters remaining fixed.

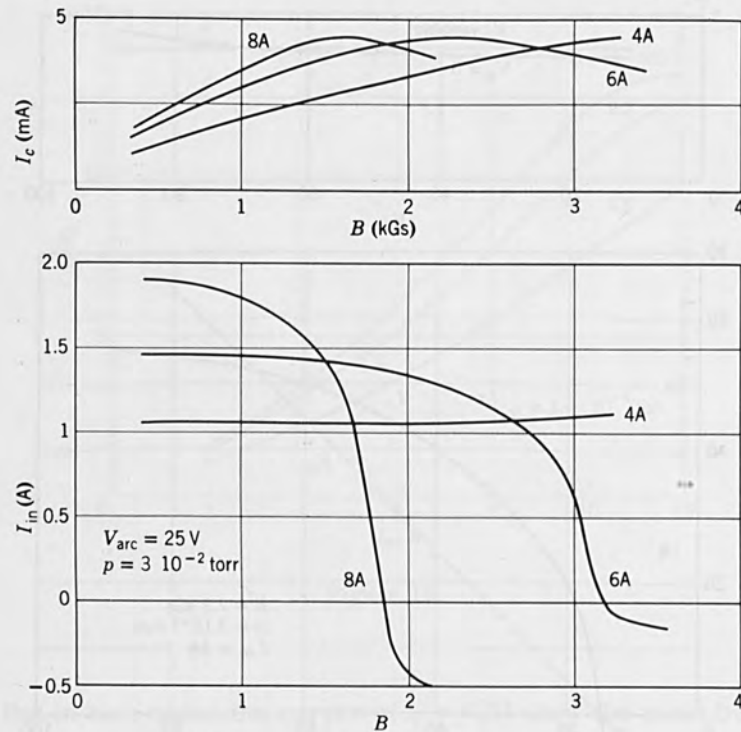


Figure 14.

ACKNOWLEDGMENTS

We would like to acknowledge the continued support of these studies by the Applied Physics Department of the French Nuclear Center of Saclay.

REFERENCES

1. R. Becherer, G. Gautherin, and A. Septier, *J. Phys. Rad.* **23**, June 1962, p. 121 A.
2. M. M. Gabovich and E. T. Kucharenko, *Soviet Phys.-JETP* **26**, 5, 1956, p. 996.
3. W. C. Beggs, et al., *NAECON Nat. Conf. Proc.*, 1961, p. 554.

CLASSICAL PHYSICS IN MODERN SPACE PROPULSION:
HEAVY PARTICLE BEAM FORMATION BY
ELECTRICAL SPRAYING OF LIQUIDS†

INTRODUCTION

The field of electric propulsion of space vehicles is one in which almost the entire field may be treated by means of classical physics. The basic problem of space-vehicle propulsion is that of expelling mass from the vehicle in order to use the effects described by Newton's formulations of the laws of motion. Although we can envision many complications, the most simple form of the problem may be resolved into the equation

$$F = \frac{d(mv)}{dt}$$

where F is the thrust on the vehicle produced by expelling mass m from the vehicle with a velocity v relative to the vehicle. If each element of mass dm is expelled with the same velocity v , the thrust is given by

$$F = -v\dot{m}$$

where $\dot{m}$ is the time rate at which mass is expelled from the vehicle. If we assume that sophistication may complicate the problem but may not be likely to change the basic situation, the problem re-

*Electrical Engineering Department, University of Illinois, Urbana, Ill.

†The research reported in this paper was supported by the U.S. Air Force Office of Scientific Research by grant AFOSR-107-63.

mains that of expelling mass particles at a rate of $\dot{m}$ kilograms per second at a velocity of v meters per second.

Electric propulsion usually refers to the art and science of propelling space vehicles through the use of electric and/or magnetic fields to obtain the velocity v of the expellent mass. The acceleration of mass particles may be accomplished by several means which provide also the major divisions in the field of electric propulsion. These are: (1) electrothermal (in which a gas is heated, expanded to a low temperature and exhausted from the vehicle); (2) magnetohydrodynamic (into which category we will place all systems utilizing $\vec{J} \times \vec{B}$ forces in an ionized gas or plasma); and (3) electrostatic (in which a beam of ions or heavy charged particles is formed and accelerated by static electric fields).

The problems in category (3) are those toward which the research reported in this paper is directed. The general problems of this category will be examined and some specific problems related to particular methods of particle and beam formation will be discussed.

ELECTROSTATIC PROPULSION

Let us assume a system in which thrust is to be produced by expelling one mass particle m with a velocity v . In an electrostatic system this can be accomplished by placing a charge q on the particle and accelerating it through a potential V_{acc} . The acceleration occurs inside the vehicle and to avoid charge accumulation on the vehicle and resultant "beam turn around" we will assume the particle charge to be neutralized immediately after acceleration and ejection from the vehicle. This allows us to eject a neutral particle with the desired velocity. (It should be noted at this point that neutralization may pose serious problems in some cases. These problems have been discussed in more detail by Sellen [1].)

The exit velocity v will be given by

$$v = \sqrt{2 \frac{q}{m} V_{acc}}$$

and the momentum given to the vehicle will be

$$mv = m \sqrt{2 \frac{q}{m} V_{acc}}$$

If n such particles are ejected per unit time, the thrust is given by

$$F = nmv = \dot{M}v$$

where $\dot{M}$ is the rate at which mass is ejected in the form of particles of mass m .

If the specific charge q/m for all particles is not the same, the particles are not all exhausted at the same velocity. In this case the thrust is given by

$$F = \dot{M} \sum_i g_i v_i$$

where g_i is the fraction of particles having velocity v_i and $\dot{M}$ is the total mass-flow rate. In the case of a continuous distribution of velocities, the sum reduces to the integral

$$F = \dot{M} \int v g(v) dv$$

Hunter [2] defines a beam power efficiency (beam power per unit thrust for a beam of particles having a velocity distribution divided by beam power per unit thrust for an ideal, monoenergetic beam with the same specific impulse) as

$$\eta_B = \frac{[\int v g(v) dv]^2}{\int v^2 g(v) dv}$$

The beam power efficiency is a measure of the degradation of the energy transfer from the accelerating potential power supply to the beam kinetic energy (and hence, vehicular energy). For a Maxwellian velocity distribution the beam power efficiency is approximately 85%.

In an electric propulsion system using only electric fields for particle acceleration, the particle velocity produced by a fixed accelerating potential is proportional to the square root of the particle specific charge (q/m). This means that the beam power efficiency can be related to the specific charge distribution. Thus the beam power efficiency becomes

$$\eta_B = \frac{\langle (q/m)^{1/2} \rangle^2}{\langle q/m \rangle}$$

where the angular brackets indicate the average of the contained quantity.

The foregoing discussion is significant because it provides a partial basis for comparing conventional ion-source techniques with electrical spraying of liquids as methods of producing charged particle beams for thrust.

A second point that must be considered in electric propulsion

discussions is the existence of an optimum expellent velocity. Introducing parameters such as the masses of the power supply and payload, transit time, etc., into a thruster problem results in the determination of an optimum expellent velocity for a specified mission. For reasonable parameters, the optimum velocity is usually found to be in the range 20 km/sec to 50 km/sec. This corresponds to a range of specific impulse¹, I_s , of approximately 2×10^3 sec to 5×10^3 sec. Figure 1 is a plot of specific charge

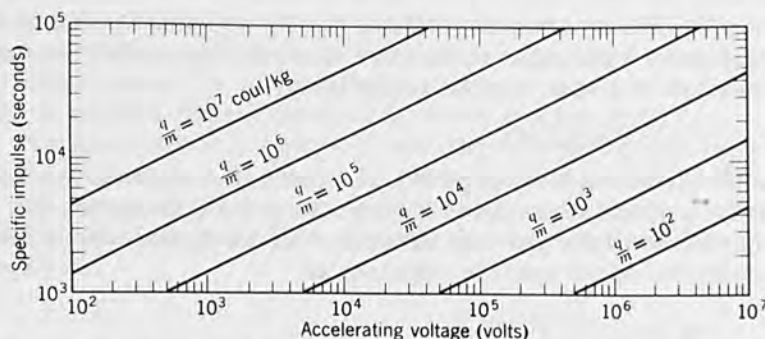


Fig. 1. Relationship between specific charge, accelerating potential, and specific impulse.

necessary to produce a given specific impulse as a function of accelerating potential in volts. To obtain a specific impulse of 2000 seconds with 10^4 V accelerating potential, a specific charge of about 2×10^4 coulombs/kilogram is required. It should be noted that an atomic hydrogen ion has a specific charge of about 10^8 C/kg. and a cesium ion has a specific charge of about 7×10^5 C/kg.

If a cesium ion beam is produced and used for thrust purposes, the accelerating potential necessary to achieve a specific impulse of 2000 sec is about 290 V. The energy imparted to the beam is then 290 eV per ion. The energy necessary to produce an ion from neutral cesium in a thruster is about 1000 eV [3]. Therefore the

¹If all particles in the expellent beam have the same velocity, specific impulse is given by

$$I_s = v/g$$

where v is the expellent velocity relative to the vehicle and g is the gravitational acceleration constant at the earth's surface.

maximum energy per ion imparted to the beam is a small fraction of the total energy expended in producing and accelerating the ions. Thus, the overall efficiency of energy transfer from the power supply to the beam is low.

Consider, on the other hand, a heavy particle composed of one ion and many neutral atoms. Let the particles in such a system have a specific charge of 10^4 C/kg. To obtain a specific impulse of 2000 sec with such particles requires an accelerating potential of 2×10^4 V. Assuming that 1200 eV is required² to produce the heavy charged particle, we see that the beam kinetic energy per particle is 2×10^4 eV. Since this energy is large compared with the 1200 eV used to produce the particle, if high accelerating potentials are available, it is more efficient to use heavy charged particles than to use monatomic ions.

LIQUID SPRAYING PRODUCED BY ELECTRIC FIELDS

During the latter half of the nineteenth century Lord Rayleigh studied the stability of electrified liquid surfaces [4], [5]. In particular, he was interested in the stability of an electrified liquid drop and in the stability of liquid jets, both neutral and electrified. From the time of the work by Rayleigh until about 1914 there was little, if any, research done in this field. In 1914 John Zeleny duplicated much of Rayleigh's work and added some of his own contributions to the literature [6], [7].

To more fully explain the instability of electrified liquid surfaces, first consider an isolated spherical liquid drop of radius r . Due to surface tension the pressure inside the drop is greater than the pressure of the surrounding medium. The excess pressure inside the drop is

$$p = \frac{2\gamma}{r}$$

where γ is the surface tension and r is the drop radius.

Let the drop carry a uniform surface charge density σ . The surface charge reduces the internal pressure by $\sigma^2/2\epsilon$, where ϵ is permittivity of the medium. Thus

$$p = \frac{2\gamma}{r} - \frac{\sigma^2}{2\epsilon}$$

²This includes 1000 eV to produce the ion and 200 eV as surface energy for a 0.01μ radius particle which has a specific charge of 10^4 C/kg.

If p is greater than zero the sphere is stable with respect to small perturbations of the surface. If p is zero or negative for some value of surface charge the drop is unstable, i.e., any small disturbance will grow and the drop will fragment.

Many years after Zeleny's work was completed, it was suggested that electrical instability could be utilized to produce beams of heavy charged particles for propulsion purposes. Research projects for the study of the spraying phenomena were initiated at several industrial laboratories, notably Ramo-Wooldridge Research Laboratory and Aerojet-General Corporation. These research projects utilized a modified form of the instability of droplets described above. A liquid was made to flow through a capillary or similar channel. When the liquid emerged from the channel, it was in a region of high electric field so that high surface charges were produced on the liquid. These surface charges caused the liquid surface to become unstable and a spray of highly charged particles resulted.

Basic investigations were instigated to determine the mechanism of charged droplet formation. Moreover, experimental determinations of the specific charge produced by a given liquid spray were made. This work was aimed at determining the properties of an "ideal" liquid for use as a propellant. Since the initial work was begun (about 1959) other groups have become interested in the problems of electrical spraying.

PRESENT RESEARCH

A schematic diagram of a typical system used for the study of the electrical spraying phenomena is shown in Fig. 2. Basically the apparatus consists of a liquid reservoir, a small diameter (50 to 400 μ) metal capillary tube, electrodes for production of electric fields, associated power supplies and electronic equipment, vacuum system, optical systems for illumination and photomicrography of the spraying process, and apparatus for studying the specific charge distribution of the sprayed liquid drops.

As the reservoir pressure is increased from zero, liquid flows from the capillary tube into the vacuum system at a rate determined by the pressure, the viscosity of the liquid, and the radius and length of the capillary. Upon the application of a sufficiently high electrostatic potential between the accelerating electrode and the capillary, the liquid surface acquires an induced surface charge high enough to cause instability of the surface. This instability re-

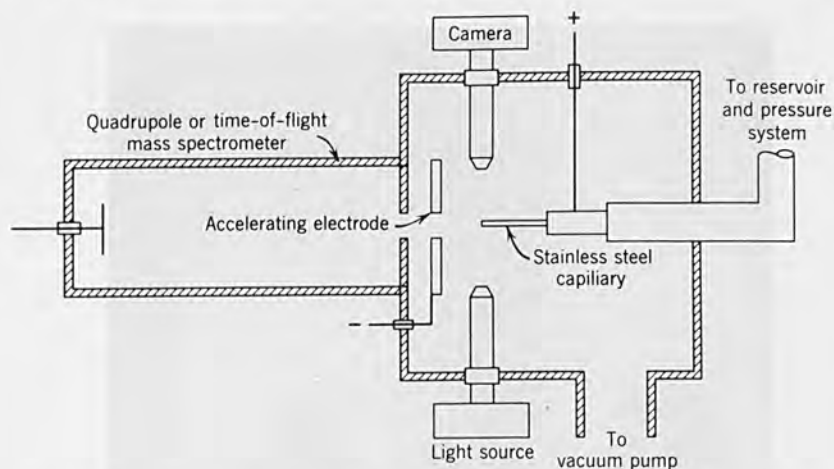


Fig. 2. Typical system used for the study of electrical spraying of liquids.

sults in the emission of charged drops from the surface. Possible configurations of the spraying surface are shown in the photographs in Fig. 3.

In general, the spraying produced for voltages near a certain minimum value is naturally pulsating in form and is characterized by the liquid necking down into a slender axial filament from the end of which the spraying of charged particles actually occurs. The liquid filament is longer for high-viscosity liquids. At voltages appreciably higher than the minimum spraying potential the spray originates at several points around the periphery of the capillary tip. At such high voltages the spray is usually cone shaped as determined by the direction of the electric field lines.

The specific charges of the sprayed drops have been studied through the use of both quadrupole and time-of-flight mass spectrometers and by using techniques for measuring the charges of individual drops. Figure 4 shows a typical specific charge distribution obtained by the use of a quadrupole mass spectrometer. In this case the electric field causing the spray gave rise to drops with rather low specific charge. The liquid in this particular case was glycerol. Other liquids, either pure or with additives, which have been studied include water, molten Wood's metal, and various organic liquids such as Octoil, dibutyl phthalate, and silicon oils. No liquid has been found which could not be sprayed electrically from the metal capillary.

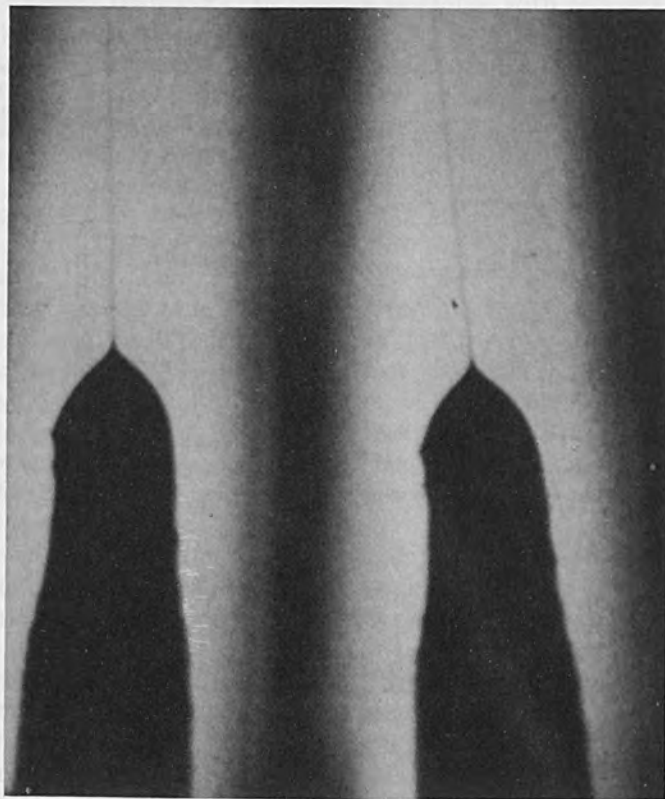


Fig. 3. Configurations of a liquid (glycerol) being sprayed electrically from a metal capillary.

It was found when dibutyl phthalate was sprayed in a certain high field situation that particles were produced which had a specific charge distribution peaking at about 10^5 C/kg. By assuming that the width of the distribution is due to the spectrometer resolution (which may or may not be true), we conclude that the particles are made up of two molecules of dibutyl phthalate with one electron removed, i.e., the particles are diamers. Present knowledge of this phenomenon is insufficient to state the propensity of other liquids to produce diamers, or other multimolecular particles, rather than larger drops.

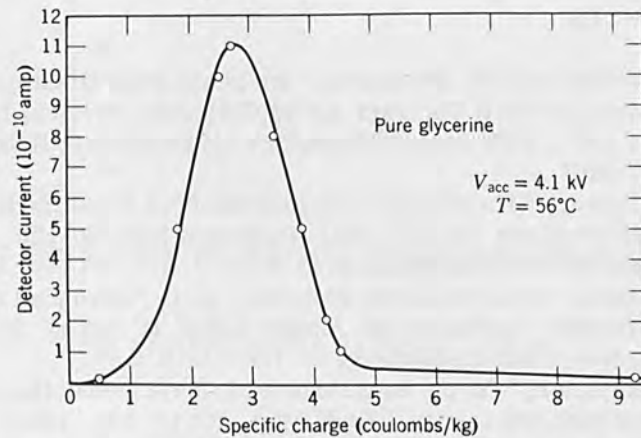


Fig. 4. Specific charge distribution obtained by the use of a quadrupole mass spectrometer. The liquid being sprayed is glycerol at room temperature.

RESEARCH GOALS AND CONCLUSIONS

At times it is well to review the goals toward which research is directed. In particular, in our research as applied to propulsion, the goals might be: (1) a beam of particles with a specific charge distribution centered at about 10^4 C/kg, and whose width at half-height would be less than 5% of the specific charge at the peak; and (2) a particle current of 0.1 to 1.0 mA per capillary. This basic research will extend the state-of-the-art in the generation and acceleration of heavy charged particles.

It is often satisfying to note that classical physics (mechanics, fluid dynamics, electrostatics, etc.) has a place in many modern research areas such as exotic-propulsion-systems studies. By applying the fundamental principles of physics to the problem of heavy particle beam formation by electrical spraying we should be able to produce thrusters for use in our modern space program.

The author acknowledges with thanks the assistance of Dr. Ralph S. Carson, Dr. John M. Schneider, and Miss Ruth Pembroke in the preparation of this paper.

REFERENCES

1. J. M. Sellen and W. Bernstein, "Ion beam neutralization," Final Report, AFOSR Contract AF 49(638)-886, Physical Electronics Lab., TRW Space Technology Laboratories, Redondo Beach, Calif.
2. R. E. Hunter, "Theoretical considerations of nonuniformly charged expellent beams," ARL Technical Note 60-138, Aeronautical Research Laboratory.
3. S. Naiditch, "Ion propulsion systems," p. 1, "Advanced Propulsion Systems," edited by M. Alpern and G. P. Sutton, Pergamon Press, New York (1959).
4. Lord Rayleigh, "On the equilibrium of liquid conducting masses charged with electricity," *Phil. Mag.*, (5) 14, 184 (1882).
5. Lord Rayleigh, "On the instability of jets," *Proc. Lond. Math. Soc.* X, 4 (1878).
6. John Zeleny, "On the conditions of instability of electrified drops, with applications to the electrical discharge from liquid points," *Proc. Cambridge Phil. Soc.* 18, 71 (1915).
7. John Zeleny, "Instability of Electrified Surfaces," *Phys. Rev.* 10, 1-6 (July 1917).

THE OPERATION OF AN ELECTRON BOMBARDMENT ION SOURCE WITH VARIOUS GASES

INTRODUCTION

Several areas of work on the electron-bombardment thruster appear to be reaching completion. The ion-chamber configuration [1], magnetic field design [2], and the accelerator structure [3] all fall into this category. A possible exception might be an attempt to produce a more uniform current density, which would permit considerably greater average power densities for the same accelerator system lifetime.

The cathode is probably the most important component research area remaining for these thrusters. The cesium autocathode, with no apparent lifetime limit, is one promising solution. The use of cesium, however, places more severe limits on the thrust-to-area ratio at low specific impulse than does mercury. For this reason, continued research is warranted on cathodes employing alkaline-earth carbonates. Very substantial lifetimes (more than 1600 hr) have already been obtained with cathodes of this type in component tests.

The present efficiency of the electron-bombardment thruster ranges from about 60 per cent at 4000-sec specific impulse to about 80 per cent at 10,000 sec [4]. These efficiencies are equivalent to power to thrust ratios of about 150 kW per pound at 4000 sec. and almost 300 kW per pound at 10,000 sec. This performance is adequate for most proposed electric propulsion missions.

The simplicity and ruggedness of the electron-bombardment thruster also makes it interesting as an ion source for a variety of experiments. Plain tantalum or tungsten cathodes may be ade-

*Lewis Research Center, NASA, Cleveland, Ohio.

quate for this application. Such cathodes have been evaluated for electric propulsion [5], and were found to be much less efficient and shorter-lived than the autocathode or oxide-matrix types, but the demonstrated lifetime of over 1000 hours should be sufficient for ion source applications. Also the ease of fabrication and operation make such cathodes a natural selection for this purpose.

Some plasma experiments in which the ion source is used simply to produce a beam or plasma column can use cesium or mercury, the elements of interest for propulsion. Other experiments, though, require the ionization of a number of elements. The performance of the ion-chamber and accelerator system were, therefore, evaluated with a number of gases - such as krypton, argon, nitrogen, helium, and hydrogen. This study was made with an existing 10-cm thruster that had been thoroughly investigated with mercury as the propellant. The data from these tests indicate the types of ions that this source can effectively produce.

APPARATUS AND PROCEDURE

A cutaway sketch of the 10-cm-diameter ion source used in these tests is shown in Fig. 1. The gas flow to the source is controlled by a calibrated lead. The material to be ionized passes through a flow distributor into the ionization chamber, which contains the cylindrical anode and an axially mounted cathode. An axial magnetic field prevents electrons, which are emitted from the cathode, from rapidly escaping to the anode. Escape of electrons to either end of the chamber is prevented by operating these ends at the same potential as the cathode. Some of the neutrals are ionized by the bombarding electrons, and some of these ions reach the perforated grid system at the downstream end of the chamber. This grid system (ion extraction system) focuses and accelerates the ions that reach the plane of the first grid into an ion exhaust beam. For thruster application, a neutralizer (not shown in Fig. 1) then current and charge neutralizes the ion beam.

The current density range at which ion chamber losses are minimized and accelerator durability is at an acceptable level occurs (for a 10-cm-diameter mercury source) for neutral flow rates equivalent to about 0.15 to 0.2 amperes of singly charged ions. The product of the equivalent neutral current, ionization cross section, and the square root of molecular weight (which determines residence time for a fixed neutral temperature) was used (with mercury as the standard) as a scaling parameter to estimate

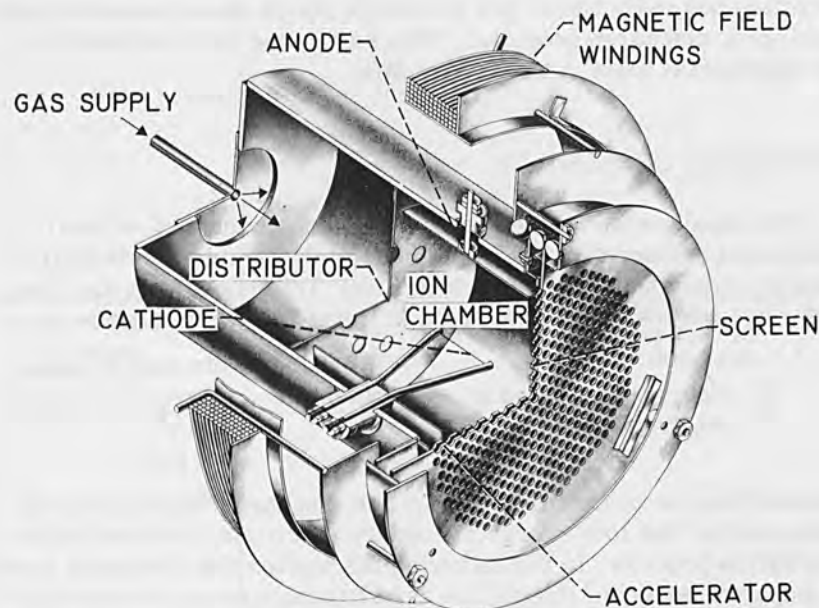


Fig. 1. Ten-cm-diameter electron-bombardment ion source.

a scaled flow rate for each gas tested. A constant value of this scaling relation implies a constant probability of ionization per unit path length of an ionizing electron. Flow rates above and below the values estimated from this simple scaling relation were explored to account for uncertainties in the relative values of ionization cross section and other discharge effects not included in the flow estimate.

Previous investigations have indicated that (with an optimized ion chamber geometry) the mechanism controlling the maximum propellant utilization efficiency that can be obtained is probably the rapid escape of neutral atoms from the ionization chamber. It was anticipated, therefore, that the lighter gases, in a chamber optimized for mercury, would yield propellant utilization efficiencies lower than mercury.

The use of a 10-cm-diameter mercury ion thruster results in another limit that must be mentioned. The refractory metal cathode configuration used in this investigation burns out very rapid-

ly at emission currents much above 10 amperes. This current restriction naturally limits the discharge power that can be obtained at a given discharge potential. This particular limit is noted in the discussion when it affects the data.

DISCUSSION OF RESULTS

The usual method of comparing the performance of several electron-bombardment ion sources is to determine the discharge energy required to produce a beam ion. This energy, in electron volts per ion (eV/ion), is arrived at in the following manner:

$$\text{Energy dissipated in discharge per beam ion} = \frac{\text{Discharge potential} \times \text{discharge current} - \text{beam current}}{\text{Beam current}}$$

Subtracting the beam current from the discharge (anode) current accounts for the low-energy secondary electrons liberated in the ionization process. In the following discussion the discharge power dissipated per beam ion will be used to compare the ionization efficiencies obtained for each gas with that of mercury. All of the tests were performed with the same source except for certain modifications made for light gases - which will be described with the appropriate experimental data.

The curves of Fig. 2 are data obtained with the 10-cm-diameter ion source and presented in reference [6]. The curves are intended to illustrate general trends rather than the absolute performance of the source.

Figure 2 (a) shows the variation of energy dissipated in the discharge per beam ion with increasing mass utilization efficiency. As the utilization efficiency is increased, the energy required to ionize an additional amount of material increases and is asymptotic to 100% efficiency. The mass utilization efficiency is determined for mercury by dividing the beam current by the equivalent current of neutrals and operating the source at discharge potentials that minimize the percentage of multiply-charged ions. The values for the other source parameters, which were held constant except where noted, are presented on the figure.

The effect of increasing magnetic field on discharge losses is displayed in Fig. 2(b). As the containment of high-velocity ionizing electrons increases with increasing magnetic field, the losses at first drop rapidly, but then approach a constant value at a magnetic

field strength that yields an electron cyclotron radius of about one-fifth the source radius.

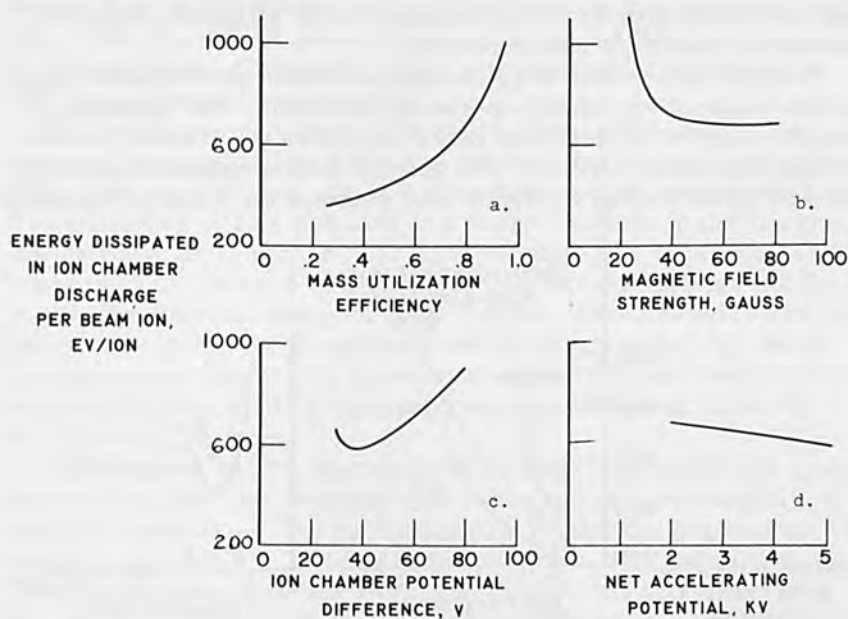


Fig. 2. Discharge performance of 10-cm-diameter electron-bombardment ion source using mercury.

Parameters when not specified: mass utilization efficiency, 0.8; magnetic field strength, 40 gauss; ion chamber potential diffusion, 50 V; net acceleration potential, 2.5 kV; neutral flow rate, 0.16 amp; accelerator potential, -2000 V.

Figure 2 (c) shows the effect of discharge potential on ion chamber losses. At low potentials the ionization cross section decreases rapidly and leads to large discharge currents to maintain a constant mass utilization efficiency. As the ion chamber potential difference (discharge potential) is increased, the losses go through a minimum. Above a discharge potential of about 50 V, the percentage of multiply charged mercury ions becomes significant [7] and the initial assumption of singly ionized exhaust beam particles less accurate. The range of discharge potentials anticipated for propulsion applications are at the minimum discharge loss point or below because of cathode durability considerations [5].

The final curve (Fig. 2(d)) indicates the effect of net accelerating potential on the energy dissipated in the ion chamber per beam ion. As the extraction field increases with a given geometry, the ions are withdrawn from the discharge more efficiently thus reducing the discharge losses per ion.

The first gas tested, krypton, gave excellent performance over a wide range of ion source operating conditions. The variation of source output with electrical parameters was qualitatively similar to operation with mercury. The general trends obtained with krypton are shown in Fig. 3. Figure 3(a) displays the trend of discharge

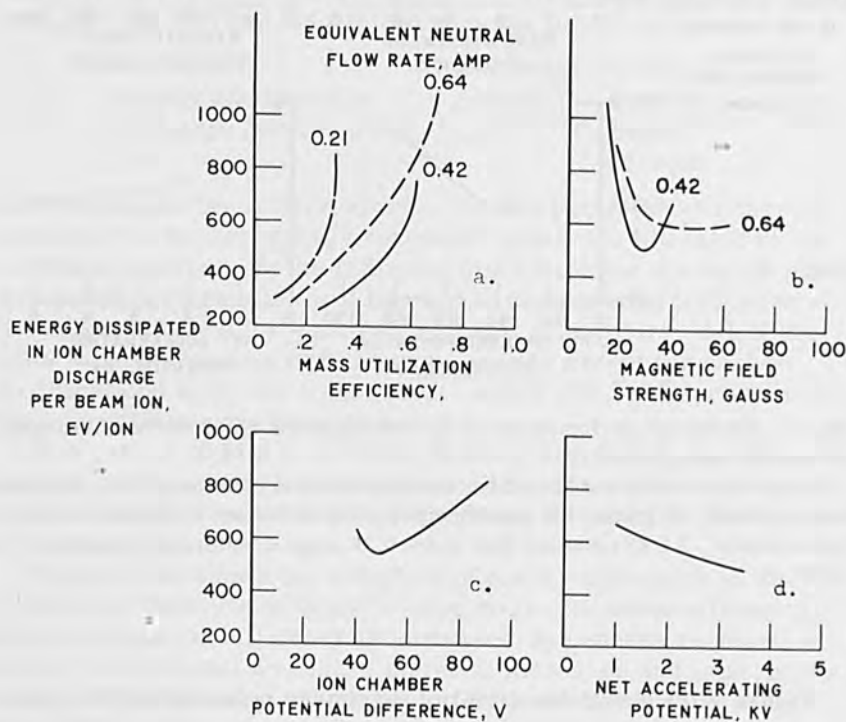


Fig. 3. Discharge performance of 10-cm-diameter ion source using krypton.

Parameters when not specified: mass utilization efficiency, 0.53; magnetic field strength, 25 gauss; ion chamber potential diffusion, 50 V; net accelerating potential, 2.5 kV; neutral flow rate, 0.42 amp; accelerator potential, -2000 V.

losses with increasing mass utilization efficiency for several neutral flow rates. The flow rates that gave the best performance (low discharge power) were found to be equivalent to 0.35 to 0.5 amperes of singly ionized atoms. This range proved to be slightly lower than the 0.56 amperes estimated from the flow scaling relation mentioned previously. Mass utilization efficiencies up to 70% were obtained with krypton.

Figure 3(b) demonstrates the effect of magnetic field on the energy dissipated in the discharge for two neutral flow rates. An equivalent neutral flow rate of 0.42 amperes with a mass utilization efficiency of 53% resulted in a sharp increase in discharge losses when the minimum-loss field strength was exceeded. This trend is characteristic of a chamber operating off-design for the particular material being ionized. Similar characteristics are obtained for mercury with unusual flow distributors or very short ion chambers. Increasing the equivalent neutral flow rate to 0.64 amperes yielded a curve (dashed line) similar to that shown in Fig. 2(b) for mercury.

Figures 3(c) and (d) display the discharge loss trends for krypton with ion chamber potential difference and net accelerating potential, respectively. As mentioned previously, the qualitative trends are similar to those of mercury (Figs. 2(c) and (d)).

Argon gave results that were qualitatively similar to krypton and, hence, mercury. The effects of the electrical parameters on source output were more pronounced in that the optimum values of these parameters required to maximize output were quite well defined.

Figure 4(a) shows the energy dissipated in the discharge as a function of mass utilization efficiency for two argon flow rates. The parameters held constant are again given on the figure. The lowest level of discharge loss at a given mass utilization efficiency was obtained at a neutral flow rate 1.0 ampere. This flow rate had been estimated at 0.8 amperes from the flow scaling relation. The maximum mass utilization efficiency that was obtained with argon was 50%. This limit occurred very nearly at the cathode emission limit. A greater discharge power capability would probably allow a slightly higher mass utilization efficiency with this gas.

Figures 4(b), (c), and (d) show the trends of ion chamber losses with magnetic field strength, ion chamber potential difference, and net accelerating potential, respectively. The results are similar to those obtained with mercury (Fig. 2) and krypton (Fig. 3).

After the above data had been obtained, a modification was made to the source in an effort to eliminate the effects of the ion chamber

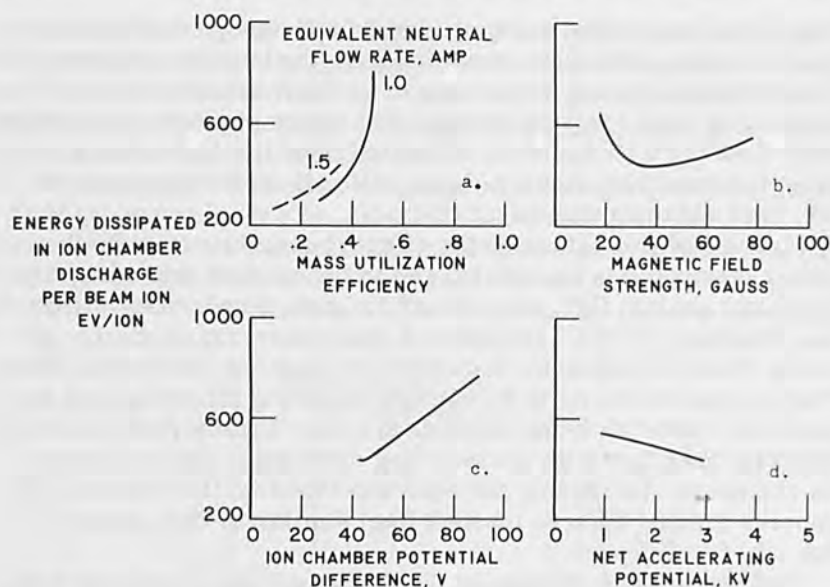


Fig. 4. Discharge performance of 10-cm-diameter ion source using argon.

Parameters when not specified: mass utilization efficiency, 0.4; magnetic field strength, 30 gauss; ion chamber potential difference, 50 V; net acceleration potential, 2.5 kV; neutral flow rate, 1.0 amp; accelerator potential, -2000 V.

power (filament emission) restriction. An annular ring was placed upstream of the ion extraction system to reduce the exhaust beam diameter from 10 to 5 cm. The neutral flow rate was also reduced to 25% of the former values to match the reduction of accelerator area. This modification allowed the maximum discharge power per unit beam area to be raised by a factor of four without changing the filament emission limitation. The internal ion chamber geometry was not altered. A slightly higher mass utilization efficiency (55%) could be attained using argon with this configuration.

Nitrogen was the first diatomic gas tested in the program. The effect of small variations in electrical parameters became of increasing importance with this gas.

The neutral flow rate estimated from the previously mentioned scaling relation was 2.2 amperes of monatomic nitrogen. This flow rate was used as a starting point even though the annular plate had been used to reduce the flow area. (The annular plate was used for the balance of the program with lighter gases.)

Figure 5(a) shows the effect of mass utilization efficiency on ion chamber discharge losses for two values of neutral flow. The lowest discharge losses for a given mass utilization occur at flow rates corresponding to about 3.0 amperes of atomic nitrogen ions.

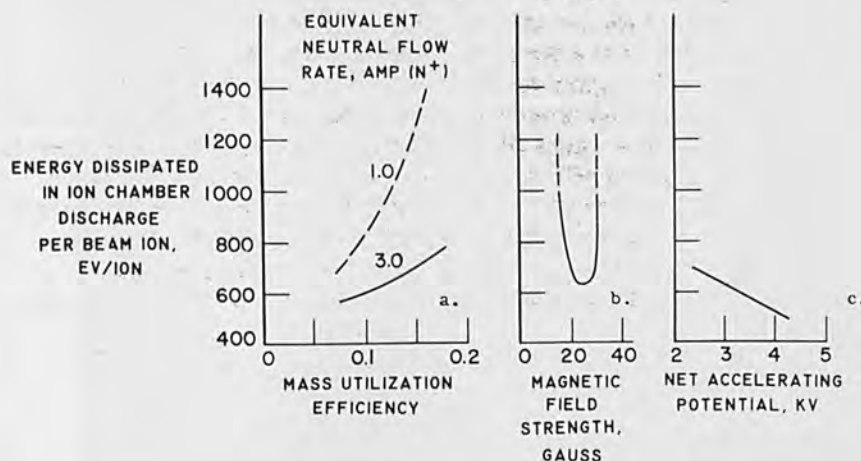


Fig. 5. Discharge performance of 10-cm-diameter ion source using nitrogen.

Parameters when not specified: mass utilization efficiency, 0.12; magnetic field strength, 20 gauss; ion chamber potential diffusion, 60 V; net acceleration potential, 2.5 kV; neutral flow rate, 3.0 amp (N⁺); accelerator potential, -2000 V.

The mass utilization was calculated assuming that the exhaust beam was composed of singly charged atomic nitrogen ions (N⁺). The possible error in this assumption is indicated by measurements made by Nelson L. Milder at the Lewis Research Center with a quadrupole mass spectrometer utilizing a 5-cm diameter electron-bombardment ion source. These measurements indicate that, between discharge potentials of 50 to 90 V, 80% of the exhaust beam is composed of atomic nitrogen ions. The 10-cm diameter ion source has also been shown to be an extremely effective molecular fragmentation device [8]. However, no special effort was made to determine the percentages of N⁺ or N₂⁺ in the exhaust beam during this investigation. The best mass utilization efficiency obtained, assuming all N⁺, was 23% (not shown on the figure). The

actual mass utilization is probably slightly above this value due to the presence of some N_2^+ ions.

Figure 5(b) shows the marked effect of magnetic field strength on the operation (discharge losses) of the source. Above 32 gauss and below 15 gauss, the discharge was extinguished. Figure 5(c) shows the general trend of discharge loss that has been typical of all types of ions with increasing extraction potential.

The operation of the source on both helium and hydrogen was very critical with regard to values of electrical parameters. Beam currents of about 0.35 amperes were obtained with both helium and hydrogen but at flow rates of about 25 and 31 amperes, respectively. The mass utilization efficiencies for both gases were, therefore, below 2%. The maximum mass utilization was severely hampered in both cases by the emission limit of the cathode. Accelerator impingement currents recorded throughout the program were consistently between 1 and 2% of the ion-beam current for every gas tested.

CONCLUDING REMARKS

The electron-bombardment thruster offers a simple and reliable ion source for a variety of experiments. The accelerating potential can be varied to provide a wide range of particle exhaust velocities, and the characteristics of the source allow it to be optimized for ionization of various gases.

The present study indicated that gases with atomic weights greater than that of argon can be ionized in a conventional ion source at mass utilization efficiencies of at least 50%. The reduction of mass utilization and the increasingly critical dependence of source operation of small ranges of electrical parameters with decreasing atomic weights are typical of nonoptimum source operation. Effects of this nature occur with the conventional mercury source either when it is operated at too low a propellant flow or when the chamber geometry is altered significantly from the optimum. It is anticipated that the mass utilization of any of the gases tested could be improved by careful attention to flow and geometric configurations. Even without optimizing the source for a given gas, the utilization can be improved if larger cathode emission currents are possible. Larger discharge power requirements might demand a thruster cooling system, but this would not be a serious problem with a laboratory experiment.

REFERENCES

1. P. D. Reader, "Investigation of a 10-Centimeter-Diameter Electron-Bombardment Ion Rocket," NASA TN D-1163, 1962.
2. P. D. Reader, : "An Electron-Bombardment Ion Rocket with a Permanent Magnet," AIAA Paper 63031, 1963. (See also *Astronautics and Aerospace Eng.* 1, No. 9, Oct. 1963, p. 83)
3. W. R. Kerslake, "Charge-Exchange Effects on the Accelerator Impingement of an Electron-Bombardment Ion Rocket," NASA TN D-1657, 1963.
4. W. R. Mickelsen, and H. R. Kaufman, "Status of Electrostatic Thrusters for Space Propulsion," NASA TN D-2172, 1964.
5. N. L. Milder, and W. R. Kerslake, "Evaluation of Filament Deterioration in Electron-Bombardment Ion Sources," NASA TN D-2173, 1964.
6. P. D. Reader, "Experimental Effect of Propellant Introduction Mode on Electron-Bombardment Ion Rocket Performance," Proposed NASA TN.
7. N. L. Milder, "Comparative Measurements of Singly and Doubly Ionized Mercury Produced by Electron-Bombardment Ion Engine," NASA TN D-1219, 1962.
8. D. C. Byers, W. R. Kerslake, and J. S. Grobman, "An Experimental Investigation of Heavy Molecule Propellants in an Electron-Bombardment Thruster," Proposed NASA TN.

DESIGN OF ION OPTICS ON DIGITAL COMPUTERS†

ABSTRACT

In a recent communication,¹ a procedure for the design of axially symmetric ion optics was proposed. In the meantime, the proposed method has been worked out in detail.

It is based on the following principle: Any Gaussian optical system can be described by a matrix relating final trajectory radii and slopes to the corresponding initial values. The constants appearing in the matrix are related to the focal lengths and the locations of the principal planes. Some of these magnitudes can be represented by a properly chosen trajectory. After such a trajectory has been prescribed, the paraxial-ray equation is solved for the potential required to produce the chosen trajectory. Choosing by trial and error from a suitable family of trajectories, a potential distribution is finally found which is physically realizable and generates most or all of the desired optical constants. From the potential distribution so obtained, the rest of the optical properties are computed. In the final step, the potential distribution is used to compute electrode shapes and electrode potentials.

A description of the theoretical considerations and the presentation of sample results obtained by computer calculations are the subject of this paper.

*Defense Systems Dept., General Electric Co., Santa Barbara, Calif.

†For reasons beyond the author's control, the full paper could not be made available for publication. The paper will be submitted at a later date to the Journal of The Electrochemical Society. (Ed.)

¹G. Rohringer, "Numerical Design Axially Symmetric Electrostatic Ion Accelerators," *Rev. Sci. Instr.*, **34**, 2, 198-199, 1963.

Index

- Aberration disk, 127
- Absorber, 16
- Absorption, curves, 14
 - exponential, 54
 - coefficient, 57
 - of beta rays, 56
- Activation energy, 184
- Acrolein, 358
- Alloys, Al-Mg-Si, 789
 - Cu-Al, 793
 - Cu-Be, 793
 - heat-resistant, 430
 - 4130, 691
 - steels, 428
 - Ti-Co, 467
 - titanium, 422
 - TZM, 651, 730, 731
 - X-2, 699
- α -Tocopherol acetate, 398
- Alumina, 328
- Aluminum, 20, 35, 36, 159, 237
- Amplifier, Darlington, 327
- Analysis, isotopical, 798
- Angular distribution, 12, 35, 36
 - of scattering, 12
- Anomalous transmission, 42
- Anticathode, 801, 926
- APC system, 319, 322
- Aperture, 52
- Aperture diaphragm, 127
- Argon, 142, 784
- Atomic number, 41, 56
- Atomic number correction, 808
- Attenuation, 41, 46
 - coefficient, 41, 43, 51, 52
 - curves, 55
 - fading, 57
 - graphical representation, 58
- Automatic control, 269, 312
- Automation, of electron-beam processes
 - for microelectronics, 293
 - positioning control, 322
- Avogadro's number, 811
- Axial astigmatism, 349
- Backscattering, 61
 - electrons, 345
 - influence of, 258
- Backscatter tail, 17
- Bandwidth, 342
- Barium, 822
- Barium stearate, 778
- BCD numerics, 313
- Beam, electron, 3
 - power and evaporation rate, 250
 - energy, 16
 - deflection, 318
 - high-perveance, 96
 - ion, 3
 - structure of, 96, 108
 - traveling, 210
- Beam programming, 328
- Beam transmission, 921
- Benzene, 394
- Beryllia, 328
- Beta ray, 7, 56
 - absorption of, 56
- Beta-ray gauges, 61
- Boltzmann-Poisson equation, 889
- Boltzmann theorem, 884
- Bottom temperature, 165, 170
- Bragg angle, 178
- Brillouin field, 90
- Butadiene, 356
- Butyltin trichloride, 241

- Cadmium, 241
- Capacitance, 238
- Capacitor, 317
- Capillary emitters, 878
- Carbides, thorium, 455
 - uranium, 455
 - uranium-thorium, 455
 - zirconium, 455
- Carnot efficiency, 72
- Casting, 415
- Casting flaws, 440
- Cathode, bolt, 739
 - cold, 133-155
 - field emission, 200
 - hollow, 134
 - indirectly heated, 734
 - spherical, 134
- Cathode cavity, 149
- Cathode diameter reduction, 748
- Cathode ray oscillograph, 112
- Cathode ray tube, 167
- Cathodolysis, 234
- Cavity emitter, 880
- Ceramics, 328, 400
 - casting flaws, 440
 - electron-beam melting, 368, 439
 - microstructure of, 448, 451
 - substrate, 330
- Cesium, 74, 858, 877
- Chamber, cloud, 7, 8, 9
 - evaporation, 248
 - machining, 266
 - melting, 443
- Channel capacity, 343
- Characteristic image, 783
- Charge, 40
 - distribution, 40
 - space, 40
- Chemical homogeneity, 432
- Chemical rockets, 853
- Cholesterol, 398
- Chromium, 159
- Circuit, 353
- Classical physics in space propulsion, 915-935
- Cobalt, 174, 462
 - distribution in beam-affected zone, 177
- Coefficient, of spherical aberration, 126
 - sticking, 226
- Cold cathode, 133-155
- Collector work function, 73
 - properties, 77
- Compositions of ultra-high-strength steels, 677
- Compression rig, 574
- Contamination, 191, 192, 221
- Control, electronic set-up, 327
- Copper, 159
 - atoms, 858
 - electrode, 858
 - oxide, 794
- Coronene, 394
- Corrosion, Ti-Co alloys, 473
- Crossover, position, 116
- Crystal, equipment, 413
 - growing, 367
- Crystal detector, 150
- Current density, 39, 43
 - profile, 98
- Current flow, 40
- Curves, intensity, 23
 - Spencer's, 20
- Cutting, 275
- Cyclic heating, 496
- Deep welding, 599
- Deflection, horizontal, 112
 - vertical, 112
 - X-Y, 325
- Dibutyl phthalate, 921
- Dielectric constant, 234
- Dielectric characteristics, 235
- Diffelen Ultra, 394
- Differential pumping, 63, 786
- Diffoil, 394
- Diffraction, 42
 - effects, 42
 - maxima, 12, 14
- Diffusion, coefficient, 184, 832
 - couples, 831
- Diode operation, 82
- Discharge, Schüler, 137
- Dissipation factor, 238
- Distribution, of electrons, 14
 - Maxwell-Boltzmann, 79
- Distribution maps, 787
- Dose, depth, 15, 21
 - depth-, functions, 22, 26
 - radiation, 17
 - residual, 15
- Dow Corning 510 fluid, 234
- Drilling, 274
- Duoplasmatron ion source, 384, 390, 893
 - discharge characteristics, 902
- Effect, Cerenkov, 6
 - of electric field, 559
- Ramsauer, 6

- Effects of flux, 93
- Effects of geometric and voltage scaling, 91
- Effects of thermal velocity, 104
- Efficiency, Carnot, 72
- Electrical power, 72
- Electrical resistivity, Ti-Co alloys, 472
- Electric propulsion, 916
- Electric rocket, 859
- Electromagnetic deflection, 234
- Electron, absorption, 33
 - cooling effect, 210
 - distribution, 13
 - emission, 75, 893
 - emission noise, 346
 - emission source, 246
 - energy, 19, 20
 - energy range, 6, 28
 - mass, 10
 - nonthermal, 88
 - optical considerations, 349
 - path, 7, 9
 - scattering, 7, 8, 9
 - scattering angle, 9
 - secondary, 19
 - thermal, 88
 - travel, 8, 10
 - unit source, 19
- Electron beam, affected zone, 176, 178
 - temperature, 162
 - characteristic parameters of, 109
 - cylindrical, 86
 - evaporation of, 382
 - focus, 167
 - for line heating, 505
 - high-power-density, 108, 184
 - information storage, 339-364
 - joint efficiency, 615
 - low energy, 231
 - nonthermal recording, 341
 - nonvacuum weld, 645
 - parameters, 654
 - penetration, 650
 - parallel, 46
 - physics of, 1-186
- Electron-beam attenuation, 53
- Electron-beam automation system, 304, 308
- Electron-beam bombardment damage, 486
- Electron-beam casting, 415
- Electron-beam control, 247
- Electron-beam cutting, 275, 283
- Electron-beam drilling, 275
- Electron-beam furnace, 20 kw, 442
 - 200 kw, 431
 - 1700 kw, 386, 387
 - multichamber, 428
 - radial heater, 406, 410
- Electron-beam joining, 585
- Electron-beam machines, 207, 306, 308
 - Balzers, 734-758
 - characteristics of USSR, 595
 - Hamilton Standard, 713
 - nonvacuum, 634
 - Sciaky Brothers, 606-633
- Electron-beam machining, 381
- Electron-beam melting, 367, 382, 415, 428, 430, 432, 434, 439, 459
- Electron-beam micromachining, 269
- Electron-beam microprobe, 297
- Electron-beam mode discharge, 133
- Electron-beam processes, 295, 368
- Electron-beam processing, 303
- Electron-beam properties, 294
- Electron-beam recording, 339-364
- Electron-beam scanning, 6
 - 1 system, 287
- Electron-beam surface treatment, 278
- Electron-beam technology, 305
- Electron-beam thickness gauge, 6
- Electron-beam weld conditions for UHS steels, 680
- Electron-beam welding, 587-733
 - of 250 series maraging steel, 704
 - of 4130, 691
 - of D6-AC, 694
 - of D36, 720-722
 - of H-11 steel, 699
 - of TZM, 728, 729
 - of X-2, 699
 - of brazed refractory joints, 713
 - of nuclear components, 655-673
 - in USSR, 587-605
- Electron-beam weld pass, 159
- Electron-beam weld properties of ultra-high-strength steels, 674
- Electron-beam weld settings, for nonvacuum welding, 654
 - for Sciaky machines, 610-614
 - for ultra-high-strength steels, 671-680
- Electron-beam zone melting, 278, 367
- Electron bombardment in source, 927
- Electron diffusion, 801
- Electron gun, 5C PIA Superior Electronics, 354
- ALCATEL, 442
- assembly, 553
- Bricka and Bruck, 245

- Electron gun, Pierce, 245
 - Ring, 506
 - Steigerwald, 245
 - triode, 370
 - with ion separation, 740
- Electron imaging, 297
- Electron-ion cloud, 140
- Electron lattice, 159
- Electron micrograph, 744
- Electron microprobe, concentration determination, 825
 - detection limits, 813-824
- Electron microscope, scanning, 191, 297
- Electron microscopy, 41
 - transmission, 527
- Electron penetration, 5-7, 210, 344
- Electron probe, 378
- Electron retarding field, 82
- Electrostatic deflection, 266
- Electrostatic recording, 341
- Emission current, 118
- Emitter, capillary, 877
- Emitter work function, 73
- Energy, 6
 - conversion, 71
 - depth-dose function of, 25
 - dissipation, 12, 13, 16
 - kinetic, 159
 - lateral distribution of, 20
 - loss, 13, 21
 - measurement, 31
 - spectrum of backscattered electrons, 64
 - threshold, 24
- Energy contribution, backscattered electrons, 34
 - penetrating electrons, 34
- Energy conversion, 71
- Energy level diagram, 860
- Energy range for electrons, 59
- Engine construction, 871
- Enthalpy, 16
- Entropy of activation, 843
- Equation, Boltzmann-Poisson, 889
 - paraxial, 105
 - Richardson-Dushman, 884
 - Saha, 887
 - Saha-Langmuir, 883
- Equipotentials, 872
- Errors, with electron gun, 102
- Evaporation equipment, 384, 411, 412
- Evaporation rate, 169
 - control, 251
- Exhaust velocity, 854
- Expellant, 855
- Expellant velocity, 918
- Experiments, of Gentner, 15
 - Gruen, Olde, and Brannen, 20
 - Kanter, 25
 - Schönland, 25
- Exponential decay, 53
- Fabrication of electronic components, 231
- Faraday cage, 47, 112, 116
- Faraday cup, 63
- Fermi level, 82, 83, 860
- Fick's law, 832
- Field emission cathode, 200
- Filament growth, 227-229
- Film adhesion, 218
- Film rupture, 166
- Fission products, 394
- Floating zone technique, Nb crystals grown by, 521
- Fluorescence, foil, 7
 - gas, 7
 - probe, 7
 - screen, 7
- Flux density, 766
- Fluxmeter, 800
- Formula, Davisson-Calbick, 105
 - Rutherford, 10
- Fotoceram, 285
- Fractive toughness, 704
- Free electrons, 149
- Free energy, 429
- Frequency resonance, 149
- Function, excitation, 18
 - Langmuire, 105
 - normalized, 27
 - transmission, 27
- Gauge, Beta ray, 60
 - ionization, 353
 - single-scatter, 7
 - thickness, 6
- Gaussian, angular current density, 132
 - current, 112
 - curves, 121, 128
 - diagram, 832
 - distribution, 103, 121, 122, 131
 - function, 110
 - integral, 832
 - population, 816
 - shape, 88
- Germanium tetranitride, 239, 240
- Granite, 797
- Gun, telefocus, 110
- Gybberrallic acid methyl ester, 394

- Half-value width, 17
- Heat-resistant alloys, 430
- Heats of activation, 843
- Helium, 42
- High impedance, 134
- High temperature testing, 567-581
- Hole current, 154
- Hollow cathode, 134
- Hydrocarbon molecules, 226
- Hydrogen, 42
- "Ignited" mode, 82
- Image plane, 110
- Increased sensitivity method, 813
- Industrial point of view, 431
- Interconnection matrix, 328
- Intermetallic diffusion, 819
- Ion, acceleration, 868
 - argon, 191
 - characteristic, 790
 - characteristic image, 790
 - chlorine, 795
 - copper, 793
 - current, 886
 - emission, 791, 851-935
 - individual, 790
 - iron, 794
 - multicharged, 791
 - nickel, 794
 - optics design, 936
 - positive, 129, 138
- Ion beam, 3
- Ion beam in microelectronics, 189
- Ion beam technology, 384
- Ion bombardment, 151
- Ion engine, 8, 857, 858
- Ionization, 15
 - cross section, 393, 799, 806
 - ion engine, 80
- Ion propulsion, 851-935
- Ion separation, 117
- Isotope, 783
- Isotopical analysis, 798
- Joining, electron beam, 585
- K-Spectrum of krypton, 783
- Kerr cell, 273
- Kjeldahl technique, 461
- Knudsen flow, 890
- Kovar, 298
- Krypton, 782
- Lactoflavin, 398
- Langmuir probe, 47
- Larmor frame, 88
- Larmor frequency, 77
- Laser, 273
- Lattice atoms, 159
- Lattice electrons, 159
- Laue spots, 512
- Law, Bothe, 800, 804
 - cosine, 172
 - exponential attenuation, 44, 50
 - Fick, 832
 - Lenard, 800
 - similitude, 46
 - Thomason-Whiddington, 209, 254
 - Whiddington's, 158, 171
 - Wiens, 578
- Lead, 159
- Lepidolite, 796
- Lithium, 794, 796
- Logarithmic, 46
- Logic system, 324
- Luminescence, 21
- Magnetic lens, 112
 - spherical aberration, 125
- Magnetic tape, 264
- Magnox, 818
- Mass, 57
 - absorption coefficient, 57
- Mass density in electron-beam affected zone, 162
- Mass separation, 389
- Mass spectrograph, 392
- Mass spectrometer, quadrupole, 921
 - time to flight, 921
- Materials processing, 367
- Matrix packaging, 328
- Maxwell-Boltzmann limit, 80
- Maxwellian velocity distribution, 917
- Measurements, by Agu, Burdett, and Matsukawa, 31
 - by Finckewirth, 47
 - by Gruen, 16
 - by Reimer, 41
 - by Reimer, Hall, Inone, and Zeitler, 43
- Measuring, current density distribution, 113, 117
 - diaphragm system, 115
 - mass density, 161
 - microwave system, 150
- Melting, alloy steels, 428, 434
 - automatic control, 445
 - ceramics, 439-459
 - deflection systems, 433
 - regulation system, 447

- Melting, semi-continuous, 432
 - titanium, 415
 - titanium alloy, 422
 - uranium carbide, 439
 - with radial heaters, 409
 - zirconium, 419
- Melting speed, 430
- Melting zone, 470, 513
- Memory, large-scale, 363
- Mercury ion source, 897-913
- Mercury supply, 900
- Mercury vapor lamp, 765
- Message director, 324
- Metal halides, 239
- Microbond, 332
- Microcircuits, 231
- Microelement electrical properties, 286
- Microformation of filaments, 220
- Micromachining, by argon ion bombardment, 194-202
 - by sputtering, 191
- Micromachining apparatus, 265, 372, 378
- Microminiaturization, 205, 304
- Micromodule, 231
- Microprobe, 6, 177
 - Cambridge Instruments, 177
 - Philips Electronic Instruments, 177
- Microscopic, 72
- Microstructure, ceramics, 448
 - nonvacuum welds, 644
 - Ti-Co alloys, 467
 - vacuum and nonvacuum welds, 644
 - weld of TZM, 651, 730, 731
 - welds, 599⁺
- Microthermal, 328
- Microwave diagnostics, 149
- Microwave test cavity, 149
- Microwelding, 305
- Model, Vine and Lauria, 209
- Molecular arrival, 232
- Molybdenum, bend ductility, 545
 - deformation, recrystallization, and welding, 541-547
 - hardness, 543
 - welding of plate, 502
- Monomers, 223, 234
 - acetylene, 223
 - acrylonitrile, 223
 - aniline, 223
 - butadiene, 223
- Monoscope, 353
- Monte Carlo calculation, 209
- Multichamber furnace, 382
- Multilayer crossover, 329
- Neutralization, 871
- Nickel, 13, 41, 159
- Niobium, composition, 524
 - interstitial levels, 525
 - resistivity ratios, 526
 - tensile data, 580
- Nitrides, 794
- Nonthermal, 341
- Normalized, depth-dose functions, 22
 - functions, 27
- Nuclear reaction, 853
- Nucleus, 8
- Optical coupling, 274
- Optical system for TV, 763
- Optical transparency factor, 141
- Organo-metallics, 239
- Oxidation, 292
 - experiments, 478
 - Ti-Co alloys, 473
- Oxides metal, 428
- Packing density, 343, 344
- Paraxial equation, 105
- Particle dissipation, 24, 40
- Penetration depth, 160
- Perveance of beam, 89
- Phase boundary studies, 462
- Phenomena, scattering, 10
- Photocell, 273, 325, 327
- Photochemical methods, 205
- Photodiode, 326
- Photoelectric tape, 314
- Photomultiplier, 47
- Pinpoint analysis principle, 800
- Planar cathode, 148
- Plasma, 7, 877
 - beam, 370
 - beam technology, 398
 - jet, 399
 - microjet torch, 399
- Polyether, 394
- Polymerization, 220, 352
 - rate of, 363
- Polymerization agent, 238
- Polyphenylether, 394
- Polystyrene, 237
- Potassium acid phthalate, 778
- Power density, 108
- Power density on weld spot, 750
- Power requirements, 246
- Preferred emission directions, 200
- Primary electrons, 345
- Propellant, 853

- Propulsion, 853-935
 - electrostatic, 916
 - ion, 853-935
 - mission, 925
 - space, 915
- Pulse height analyzer, 816
- Pyrocarbon, 822
- Quantitative analysis, 801
- Quantitative electron microanalysis, 799-812
- Quantitative evaluation, 82
- Quartz, 274
 - sealed, 831
- Radial heaters, 409
- Radiation damage, 824
- Radiation flux density, 766
- Radiation losses, 6, 72
- Radioactive isotope, 56
- Radioactive tracing techniques, 185
- Raster, 192, 221, 297
- Ray, X, 6
 - Beta, 7
- Rays, "Knoten," 129
- "Faden," 129
- Recording, 341
 - electron-beam, 341, 348
 - high-speed, 347
- Remelting, 428
- Resistivity ratios, for tungsten, 560
- Resistors, 242, 317, 318
- Resolving power, 782, 786
- Resonance frequency, 149
- Richardson formula, 75
- Richtstrahlwert, 109, 123, 124
- Ring cathode, 506
- RMS, 343
- Rocket, 853
- Ru-Ta system, 826
- Saha equation, 881, 888
- SAP, 818
- Scanning control, 271
- Scanning current, 111
- Scanning electron microscope, 191, 192, 220
- Scanning electron microscopy, 191, 297
- Scanning rate, 357
- Scattering, 10
 - angle, 34, 38, 137
 - most probable angle, 33, 34
 - back, 7, 10, 38, 61
 - coherent, 45
 - Scattering, cross section, 11
 - distribution of, 13
 - electron-electron, 10
 - fountain type, 7
 - phenomena, 10
 - plural, 11
 - single, 10
 - with deep penetration, 12
 - Scattering coefficient, 32
 - Screen mesh transparency, 149-155
 - Secondary emission, 152
 - Sensitizers, 241
 - Servo amplifier, 307, 308
 - Servo drive, 307
 - Signal-to-noise power, 343, 347
 - Silicon, 36, 192
 - Silicon oil, 921
 - Silicon photodiode, 273
 - Silver, 819
 - Similitude, 37
 - laws, 46
 - mechanics, 37
 - Skull furnace, 415
 - Slag, 433
 - Sodium chloride, 795
 - Solid solubility limit, 817
 - Solid-state growth, 506, 514
 - Space charge, 77
 - effect, 80
 - Spherical aberration, 349
 - coefficient, 126
 - Spherical cathode, solid stainless steel, 142
 - surface emitter, 138
 - Spot diameter, 5 mils, 300
 - 1-2 mm, 247
 - 1-50 μ , 377
 - 2-70 μ , 206
 - 10 μ , 287, 353
 - Stainless steel, 430
 - 304, 147
 - solid sphere cathode, 142
 - Statistical fluctuation, 10
 - Steel, 428
 - 250 series maraging, 704
 - composition of, 435
 - oxygen content, 436
 - stainless, 430
 - ultra-high-strength, 4130, 691
 - D6-AC, 694
 - H-11, 699
 - X-2, 699
 - Sticking coefficient, 226
 - Stopping power, 29
 - Styrene, 234

- Suboxides, 428
- Substrate, 209
- Substructure, F.Z. grown Nb, 521-539
- Subtractive process, 311
- Surface emitter, 148
- Surface ionization, 853
 - engine, 871
- Surface polymer, 353
- Surface temperature, 163
- Surface temperature and damage, 495
- Surface tension, 919
- Surface topography, 294
- Tantalum cathode, 925, 926
- Tantalum cavity emitter, 886, 888
- Tantalum filament, 897
- Tape control, 315, 323
- Target, dense, 10
 - X-ray, 6
- Telefocus gun, 117
 - current density distribution in the crossover, 118
- Temperature, bottom, 165
 - surface, 163
- Temperature distribution, steady-state, 213
- Tensile properties, Ti-Co alloys, 471
- Tension rig, 572
- Tetraethylorthosilicate, 296
- Tetraethyl-lead, 269
- Tetraethyl-tin, 296
- Tetrode system, 114
- Theory, of beam generation, 109
 - Hermann's optical, 86
 - Langmuir-Saha, 74
 - Lazarus, 849
 - Rutherford, 41
 - Shotky, 78
 - Spencer, 20
 - Spencer and Fano, 19, 20
 - due to Swazin, 846
- Thermal damage, 115
 - etching, 231
 - micromachining, 379, 380
 - processes, 298
- Thermal time constant, 210
- Thermionic cathode, 343, 350, 370
- Thermionic converter, 72, 73
- Thermionic diode, 71
- Thermionic emission, 74
- Thermionic energy, 71
- Thermodynamic results, 844
- Thermoplastic recording, 341
- Threshold, 13
- Tin, 212
- Titanium, 461
- Titanium carbide, 819
- Titanium cobalt alloys, 464
- Tomatadine, 398
- Track cutting, 211
- Track formation, 205
- Transmission, 32
 - electron microscopy, 527
- Triode gun, 117
- Triphenyl silanol, 296
- Trustor, 918
 - electron bombardment, 925
- Tungsten, 13, 159
 - analysis, 556
 - cathode recrystallization, 745, 746
 - cesium emission from, 863
 - effect of bombardment heating, 481-502
 - hairpin filament, 304
 - hexacarbonyl, 239, 240
 - indirectly heated cathode, 734
 - reflectance of, 767
 - single crystal, 200
 - target flux density, 760
 - zone refining, 550
- TV, closed-circuit, 759-773
- Ultra-high-strength steels, 674
- Ultra high vacuum, 384
- Uranium, 799, 819, 821
- Uranium carbide, 455
- Uranium-Zirconium monocarbides, 821
- Vacuum deposition, 245
- Vanadium, 830
- Vapor density, 167
- Variable gradient, 502
- Voltage current curves, 78
- Voltage scaling, 97
- Volume diffusion, 821
- Wehnelt diagram, 739
- Wehnelt electrode, 118, 119, 247
- Weld, program, 325
- Welding, 319
- Weld pass, 159
- Wien's law, 578
- Wood's metal, 921
- Work function, 73
- Worktable, control, 306, 310
- Writing speed, 359, 360
- X-photon, 801
- X-ray, 6, 259
 - absorption, 801
 - intensity, 20

INDEX

945

- X-ray, intensity in alloy, 803
 - intensity ratios, 825
 - levels, 14
 - line characteristic, 827
 - microanalysis, 161, 185
 - microanalyzers, 177
 - target, 7
 - Ti-Co alloys, 468
- X-ray emission analysis, 781, 813
 - limitation of, 782
- X-ray fluorescent analysis, 778
- Zinc, 241
- Zircaloy-2, 511
- Zirconium, 502
 - iodide, 503
- Zone melting, 278, 367
 - Ti-Co alloys, 470
 - ZR, 513
- Zone refining, 503, 513
 - field-aided, 550
 - results of, 561
 - sublimate analysis, 559

