

ELECTRON AND ION BEAM SCIENCE AND TECHNOLOGY

FOURTH INTERNATIONAL CONFERENCE

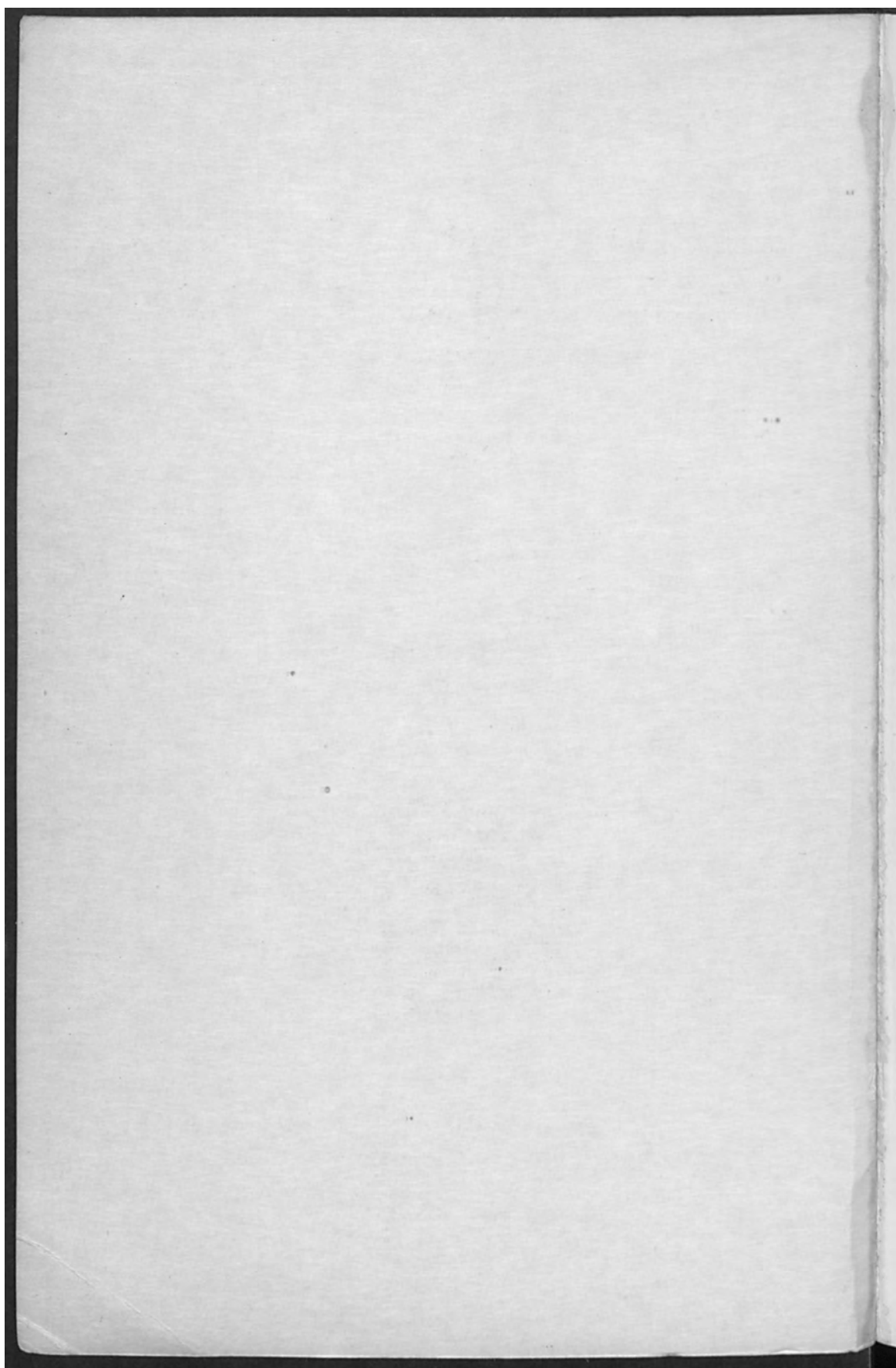
Edited by

Robert Bakish
Bakish Materials Corp.
Englewood, New Jersey



ELECTROTHERMICS AND METALLURGY DIVISION

THE ELECTROCHEMICAL SOCIETY, INC., 30 East 42 Street, New York, N.Y. 10017



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Edited by

Robert Bohn
Bohn Material Corp.
Englewood, New Jersey

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PREFACE

The writer became involved in organizing conferences in the Electron and Ion Beam field and editing their respective proceedings shortly after his professional involvement in it in 1958. The aim of these conferences, similarly to other science and technology conferences, is to survey the field at a given junction and to present the status of the technology as reflected by the newest theoretical advances on the one hand and industrially adopted innovations on the other. At no time were these conferences intended to provide a barometer of the economic climate and, much less, of social problems, unrelated to either the technology or the economic conditions. Yet spontaneously, without making any attempts whatsoever, the IVth International Electron and Ion Beam Conference, in addition to presenting the status of the technology as manifested by criteria given above, also indirectly reflected the difficult economic times and even the troubled life in some of our academic institutions.

To begin with, there was a rash of cancelled papers--with cancellations taking place from a few days preceding actual presentation, to notices at the actual session at which papers were to be presented, to no notice at all. In all the years that I have had anything to do with conferences, I have never seen this. Economies introduced by corporate managements were the prevailing reason, and academic unrest and patent problems caused at least one cancellation each.

Second, several of the individuals who made the presentations subsequently notified the writer that because of additional corporate economies they could not afford to prepare and submit the manuscripts.

Cancellation brought about by academic unrest and some of the correspondence which followed reveal a point referred to above. Let me quote from correspondence with a scheduled speaker: "Although you will no doubt tend to blame me for my failure to attend, the blame must lie with the National Administration which is currently playing football with the basic science and technology effort of this country and with irresponsible news media who have worked so hard to whip up mass hysteria on our campuses."

The status of electron and ion beam technology in 1970, directly, and national economics and even campus unrest, if you will, indirectly, were all part of the IVth Conference.

With papers withdrawn, essentially for the reasons referred to above, from the 54 papers in the program of the meeting, only 39 are to be found in the record; these include three papers submitted as late news items.

The conference papers were edited by the writer and their sequence rearranged in keeping with past conference proceedings and in order to present a more coherent volume. They are arranged in

four sections as follows:

1. Electron and Ion Beam Physics and Related Phenomena
2. Electron Beams in Welding and Materials Processing
3. Electron and Ion Beams in Micro Electronics
4. Electron Beams in Radiation Processing

The first section on physics of electron and ion beams, as has been the case in prior conferences, is a compilation of papers dealing with different aspects of the physics of electron and ion beams. Except for this common denominator there is little else that could be used to group these papers together.

The second section is devoted essentially to welding and contains papers bridging the gap from electron beam welding mechanism related topics to welding problem areas and hardware manufacturing. As there were only three papers dealing with processing or studying materials with the help of electron beams, these have been included in the welding section. One can interpret this as a reflection of substantially reduced financial support and corresponding reduction of research effort in materials.

The third section on micro electronics begins with a review paper on the subject, followed with the papers arranged in two groups. The first group deals mainly with electron beams as device manufacturing and control tools, while the second group explores the use of ion beams for carrier implantation.

The last section presents the papers considering electron beams as ionizing radiation in industrial processing.

It is hoped that the reader will find the IVth Electron and Ion Beam Conference record informative and useful and meeting the conference objectives. We at The Electrochemical Society have already made plans for the Vth meeting and look forward to seeing you again and greeting you. This time it will be in Houston, Texas, May 7 to 12, 1972. I do hope most if not all of the problems which caused us grief in 1970 will be long forgotten by then.

R. Bakish

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INTRODUCTORY REMARKS

As chairman and the individual responsible for this and the three preceding Electron and Ion Beam Conferences, I wish to extend the Society's and my personal welcome to all of you. I would hope that your effort in attending this meeting will be well rewarded by the benefits which you will derive from it. Renewal of old acquaintances, making of new ones, and taking a good pulse on the rate of development of this important technology are to me perhaps the essence of these periodic meetings.

It is most appropriate in these opening remarks to express sincere thanks to all the contributors to the conference; to be sure, without them there would not be a conference.

Permit me to add a few comments about things which interest you. First, it was my intention to distribute to all of you at the time of registration the complete record of this conference. Most regrettably a considerable number of the contributors let me down and failed to meet the original manuscript submission deadline. As our work must of necessity take priority over obligations to meetings, I can understand how this can happen. I would not like, however, to delay extensively the publication of the record of the conference, and wish to request your help in receiving the manuscripts. I hope you will submit them to me here, but if this is not the case, mail them promptly upon your return. Delay in submitting the manuscripts will result in delay in publication of the proceedings, and I am certain none of you would like that.

With our economy being what it appears to be at the moment, we have run into a number of cancellations, but these will be announced at the beginning of each session.

Enjoy the meeting and enjoy your stay in Los Angeles.

Thank you.

R. Bakish
Los Angeles, May 11, 1970

Investigation of a Laser Irradiated Electron Source

C. Andelfinger, K. Biehl, E. Buchelt, W. Ott, G. Siller

Institut für Plasmaphysik, Garching

Introduction

For our electron ring experiment we want to have a 2 MeV electron source which gives a current of 400 A in an acceptance Δ of maximum 30 eV and cm. This corresponds to a minimum brightness (normalized to the momentum of the electron)

$$B = \frac{I}{e^2 \cdot eU \cdot (1 + eU/2mc^2)} = 3 \cdot 10^8 \frac{A}{\text{sterad} \cdot \text{cm}^2 \cdot \text{eV}}$$

SECTION I

The existing electron sources with laser or field emission give values of the brightness of at least one order of magnitude below our knowledge.

AND

RELATED TOPICS

The aim of the experiment described here is the development of a compact electron source, using the high power densities available with giant pulse lasers. The device is similar to laser production of plasma. We are here, however, only interested in the electron current from such a source.

Experiment

The beam of a Q-switched ruby laser (8 Joule, 37 ns) was focused on a tantalum plate (Fig. 1). The focal length of the lens was 15 cm, between this plate and a collector electrode a voltage up to 60 kV was applied. The current oscillogram (Fig. 2) showed an electron pulse, the beginning of which coincided with the laser pulse. This electron pulse is followed by another much higher pulse which corresponds to a short-circuit between tantalum plate and collector by an expanding Ta-plasma cloud. To protect the device a delay line was used to switch off the charging voltage before short-circuit. Fig. 3 shows an example of the electron current pulses which were received in this way with a higher voltage and a higher laser energy than in Fig. 2.

SECTION I
ELECTRON AND ION BEAM PHYSICS
AND
RELATED TOPICS

Investigation of a Laser Irradiated Electron Source

C. Andelfinger, K. Büchl, E. Buchelt, W. Ott, G. Siller

Institut für Plasmaphysik, Garching

Introduction

For our electron ring experiment we want to have a 2 MeV electron source which gives a current of 400 A in an emittance ϵ of maximum 50 mrad cm. This corresponds to a minimum brightness (normalized to the momentum of the electrons)

$$B = \frac{I}{\epsilon^2 e U (1 + e U / 2 m_0 c^2)} \approx 3 \cdot 10^{-2} \frac{\text{A}}{\text{sterad cm}^2 \text{eV}}$$

The existing electron sources with hot cathode or field emission give values of the brightness at least one order of magnitude lower (according to our knowledge).

The aim of the experiment described here is the development of a competing electron source, using the high power densities available with giant pulse lasers. The device is similar to laser production of plasma. We are here, however, only interested in the electron current from such a source.

Experiment

The beam of a Q-switched ruby laser (8 Joule, 17 ns) was focused on a tantalum plate (Fig. 1). The focal length of the lens was 15 cm. Between this plate and a collector electrode a voltage up to 60 kV was applied. The current oscillogram (Fig. 2) showed an electron pulse, the beginning of which coincided with the laser pulse. This electron pulse is followed by another much higher pulse which corresponds to a shortcircuit between tantalum plate and collector by an expanding Ta-plasma cloud. To protect the device a delay line was used to switch off the charging voltage before shortcircuit. Fig. 3 shows an example of the electron current pulses which were received in this way with a higher voltage and a higher laser energy than in Fig. 2.

The height of the electron current pulses were strongly increasing with the laser energy in the focus: Fig. 4. The electron current depended also on the spot size of the laser beam. We reached pulses with amplitude of up to 1000 amperes with extremely good focusing.

Emittance measurements

We measured the emittance of the central part of the electron beam. The emittance box consisted of a 1.5 mm thick metal plate with an array of 0.3 mm diameter apertures. At a distance of 2 cm behind the plate a film was mounted, wrapped in a 10μ aluminium foil. Fig. 5. The photometric curve of the exposed film has to be corrected according to the nonlinear density of the film and to the finite angular acceptance of the apertures. The evaluation was done similar to that described by Allison¹⁾. In Fig. 6 the curves for 1/2 and 1/4 of the maximum phase density are drawn. The area within the 1/2-curve contains about 1/2 of the current and the 1/4-curve about 3/4 of the current to the emittance box which was 17 amps. With these values of the emittance and the current one gets a normalized brightness of

$$B_{1/2} = 10 \cdot 10^{-2} \frac{A}{\text{sterad} \cdot \text{cm}^2 \cdot \text{eV}}$$

$$B_{1/4} = 3 \cdot 10^{-2} \frac{A}{\text{sterad} \cdot \text{cm}^2 \cdot \text{eV}}$$

Discussion

These results for the beam brightness are preliminary and have to be refined. Furthermore it is not enough to have a high brightness in the center of the beam. So we have to optimize also the electrode configuration. Nevertheless we think that these results are encouraging. We are now beginning to work with a 200 kV Blumlein-installation, see Fig. 7, and want to measure the beam emittance time resolved.

Further development of this electron source will of course also depend on the development of laser technique, regarding repetition rates and reproducibility.

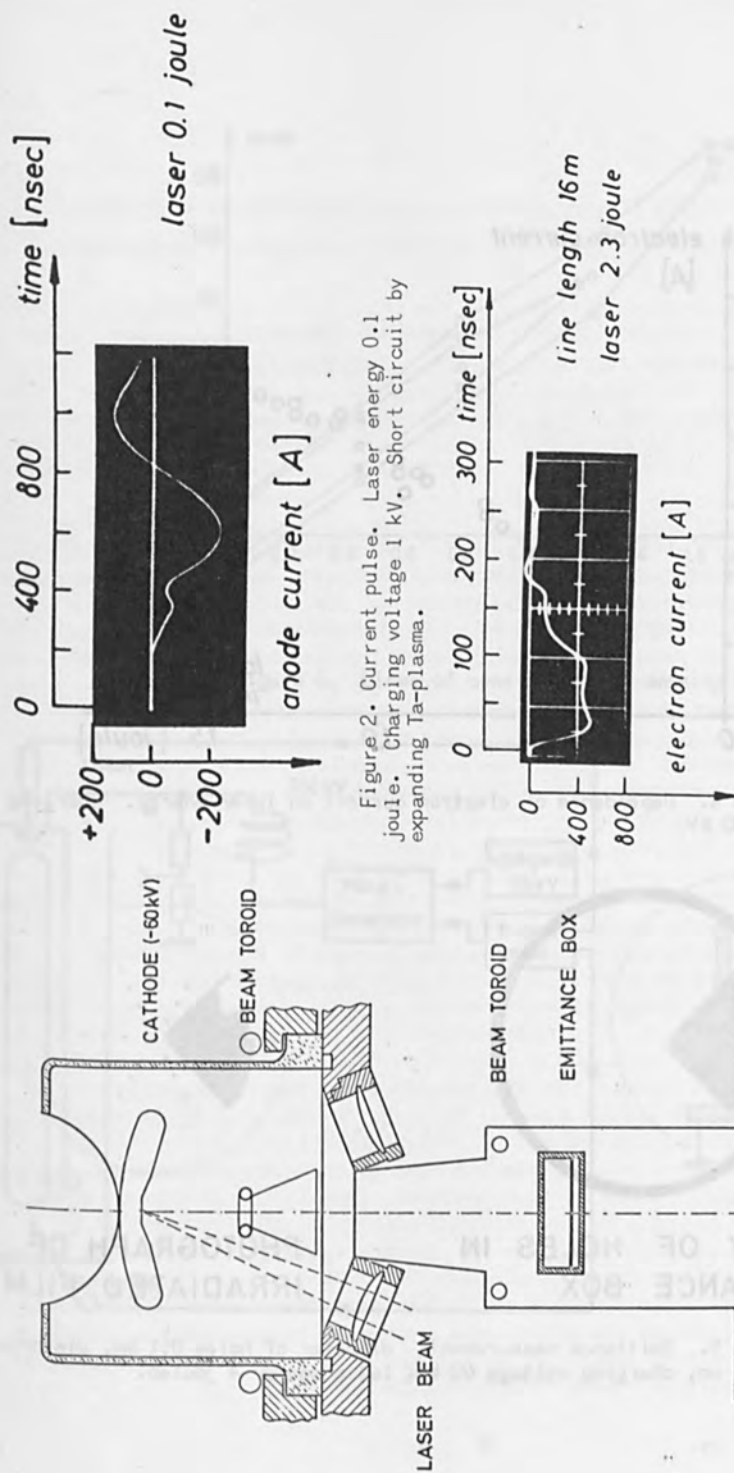


Figure 2. Current pulse. Laser energy 0.1 joule. Charging voltage 1 kV. Short circuit by expanding Ta-plasma.

Figure 3. Electron current pulse. Laser energy 2.3 joules. Charging voltage 30 kV. The voltage is switched off about 100 ns after laser pulse.

Figure 1. Electron source

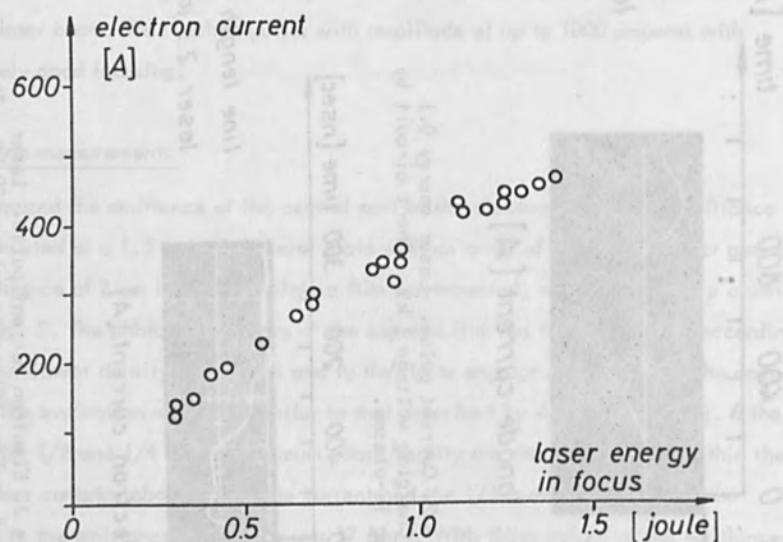


Figure 4. Dependence of electron current on laser energy. Charging voltage 30 kV.



ARRAY OF HOLES IN
EMITTANCE BOX



PHOTOGRAPH OF
IRRADIATED FILM

Figure 5. Emittance measurement: diameter of holes 0.1 mm, distance to film 2 cm, charging voltage 60 kV, laser energy 4 joules.

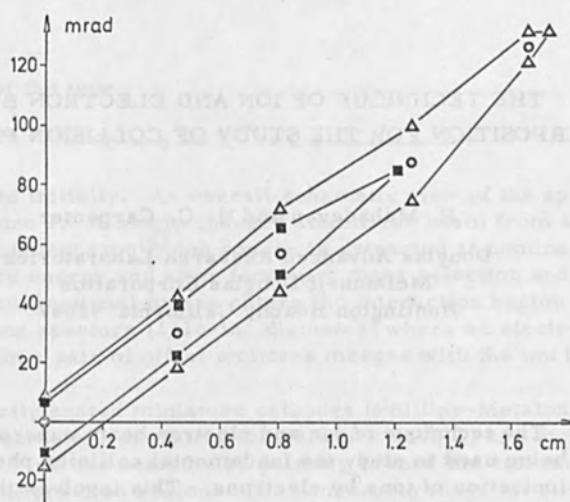


Figure 6. Lines of constant phase density

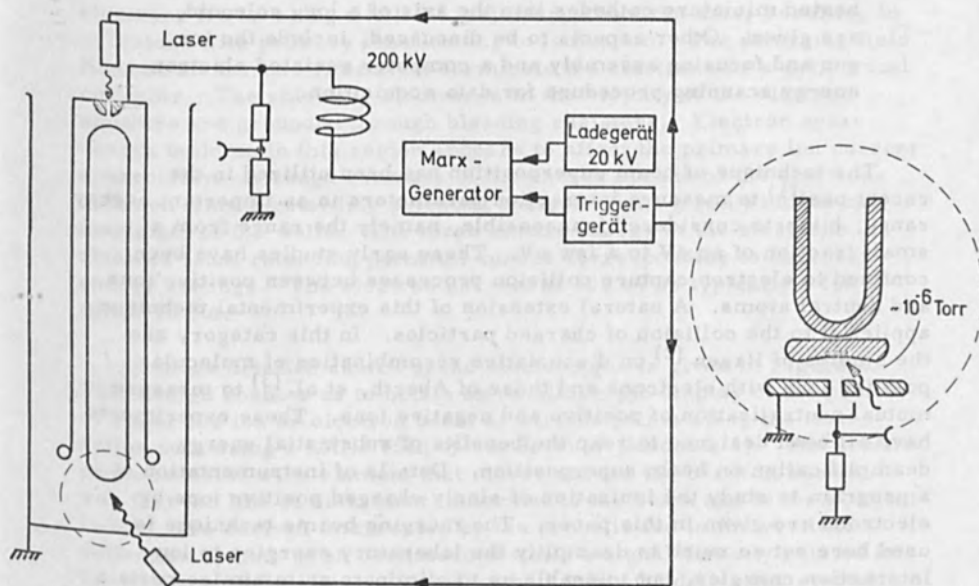


Figure 7. 200 kV device

THE TECHNIQUE OF ION AND ELECTRON BEAM
SUPERPOSITION FOR THE STUDY OF COLLISION PHENOMENA

P. Mahadevan and H. C. Carpenter
Douglas Advanced Research Laboratories
McDonnell Douglas Corporation
Huntington Beach, California 92647

The technique of ion and electron beam superposition is being used to study the fundamental collision phenomenon of ionization of ions by electrons. This involves the flight along identical linear paths of confined beams of electrons and heavier particles such as the positive ions of the atmospheric species. Design features of an electron gun which enables us to draw electrons from two, offset, indirectly heated miniature cathodes into the axis of a long solenoid, are given. Other aspects to be discussed, include the ion gun and focusing assembly and a computer assisted electron energy scanning procedure for data acquisition.

The technique of beam superposition has been utilized in the recent past⁽¹⁾ to measure interaction parameters in an important energy range, hitherto considered inaccessible, namely the range from a small fraction of an eV to a few eV. These early studies have been confined to electron capture collision processes between positive ions and neutral atoms. A natural extension of this experimental technique applies it to the collision of charged particles. In this category are the studies of Hagen⁽²⁾ on dissociative recombination of molecular positive ions with electrons and those of Aberth, et al.⁽³⁾ to measure mutual neutralization of positive and negative ions. These experiments have all been designed to reap the benefits of substantial energy deamplification on beam superposition. Details of instrumentation of a program to study the ionization of singly charged positive ions by electrons are given in this paper. The merging beams technique is used here not so much to deamplify the laboratory energies to low interaction energies, but to enable us to eliminate or minimize certain inherent problems associated with the space charge of intense electron beams and also to take advantage of the property of forward peaking of the multicharged product ions formed in collision.

Reactions of the type



are being studied initially. An overall schematic view of the apparatus is shown in Figure 1. A singly charged atomic ion beam from an electron bombardment type⁽⁴⁾ ion source is extracted at nominally 1 KeV laboratory energy and after focusing, mass selection and collimation by conventional means enters the interaction region through a limiting aperture (1/16-in. diameter) where an electron beam drawn from a pair of offset emitters merges with the ion beam.

Two indirectly heated miniature cathodes (Phillips-Metalonics, Type B, 3 mm diameter, operating temperature 1100°C) are mounted approximately 15° off the beam axis, pointing toward the entrance aperture to the interaction region. The converging magnetic field lines in this region due to the solenoidal coil transport the electrons towards the aperture plate. The fraction of the electrons that are aimed at the aperture are transported into the interaction chamber. The solenoidal magnetic field along the axis being uniform, the electrons travel along a helical trajectory about the axis, resulting in collisions with primary ions leading to ionization. The diverging field lines at the far end effectively terminate the electrons on a cylindrical collector. The shielding electrodes in the vicinity of the electron emitters are grounded through bleeding resistors. Electron space charge build up in this region appears to affect the primary ion current appreciably. A rough estimate of the energy spread of the electrons obtained from a retarding potential analysis gives a full width at half maximum of 0.7 volts. The mean kinetic energy of the electrons is obtained from retarding potential curves corresponding to different energy settings. The electron current at 100 eV is typically a few times 10⁻⁶A.

A more detailed sketch of the electron gun is given in Figure 2. This design enables us to obtain an oscilloscopic display of the profile of either the ion or electron beam at various points along the interaction path using a DANFYSIK Beam Scanner (Model 511). The profile probe is a fine wire element that moves across the beam in such a way that the line of movement is normal to the beam and to the element itself. The current intercepted by the probe is fed to the vertical deflection system of an oscilloscope while a signal directly related to the position of the element is fed to the horizontal deflection system. The normal sweep frequency of the probe element is 15 to 20 Hz. It is also possible to use the probe in a nonoscillating mode with a variable d. c. voltage for spatial calibration. Typical profiles of the

ion and electron beams are shown in Figure 3 (a and b) while the spatial profile of the beams is shown in Figure 4.

Data Acquisition

The system is set up on a semi-automated data acquisition mode. At the present time, the relative ionization efficiency for various primary ions is obtained directly as a function of the laboratory energy of the electrons. The range of electron energies is from zero to about 300 eV. Figure 5 is a block diagram of the data acquisition system. As the preset range of electron energy is repeatedly scanned, the relative signal which is the ratio of the product ion (X^{++}) current to the ionizing electron current is stored in a multi-channel analyzer. This ratio of the product ion current (proportional to the output of the electron multiplier) and the total electron current monitored at the electron collector is obtained directly using a log divider circuit⁽⁵⁾. The signal level in the multichannel analyzer builds up with the number of scans and the efficiency curve is displayed on an oscilloscope or plotted out as required. It is possible to make a more detailed examination of any part of the ionization efficiency curve by presetting the required energy range. It is also possible to check for short term instabilities in the various tuning parameters and for excessive drifts in the ion or electron current by "scanning" the normalized product signal at a fixed electron energy from an external source. Each such "scan" provides one data point per channel of the analyzer. Drifts or instabilities would show up as wide variations in signal intensity from successive channels. Such checks are made from time to time. Consistency checks are also made by scanning the electron energy range with (a) the ion beam turned off and (b) the electrons turned off. In the latter case, since the cathodes are not emitting, the cathode potential alone is varied. No defocusing of the primary ion beam has been observed due to this voltage variation. Beam modulation and synchronous signal detection are not adopted since random noise gets rejected in the signal averaging mode of operation of the multichannel analyzer.

We have, so far, investigated the relative ionization efficiencies of a few rare gas ions and also the atmospheric species O^+ and N^+ .

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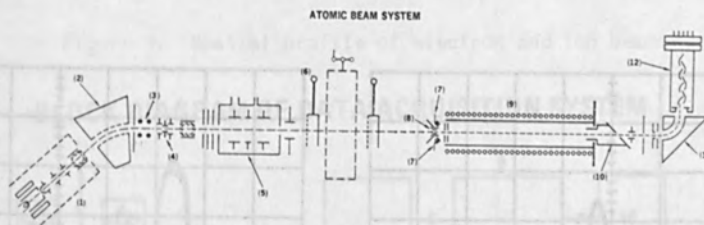


Figure 1. Over-all view of superposed beam apparatus

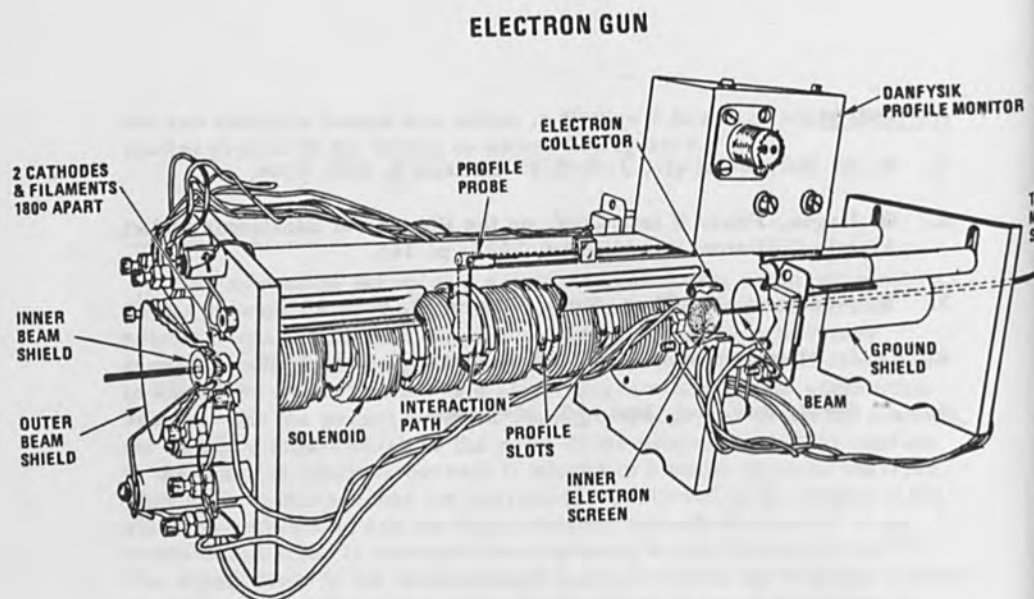


Figure 2. Electron gun and beam profile monitor

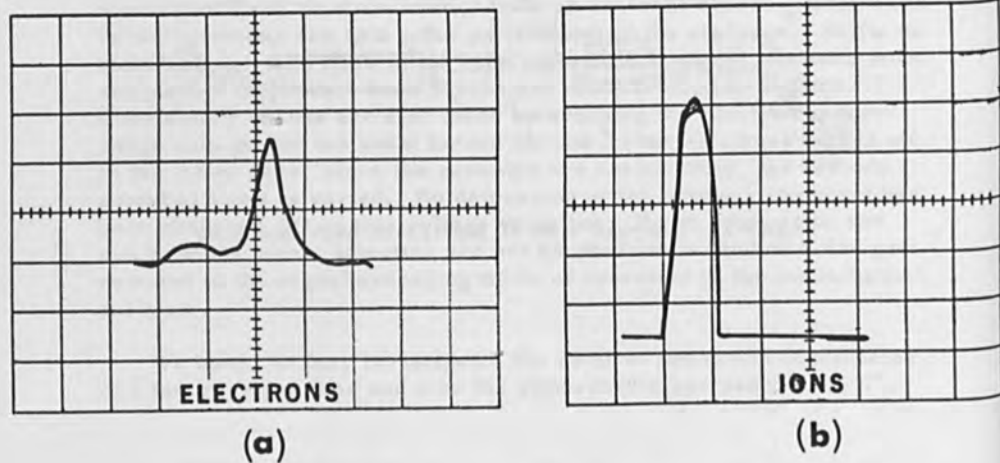


Figure 3. (a) Profile of a He^+ beam at 1.1 keV energy; (b) profile of electron beam at 50 eV energy.

OR

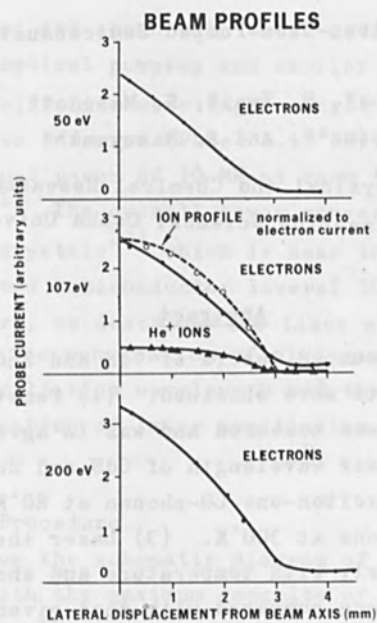


Figure 4. Spatial profile of electron and ion beams

BLOCK DIAGRAM OF DATA ACQUISITION SYSTEM

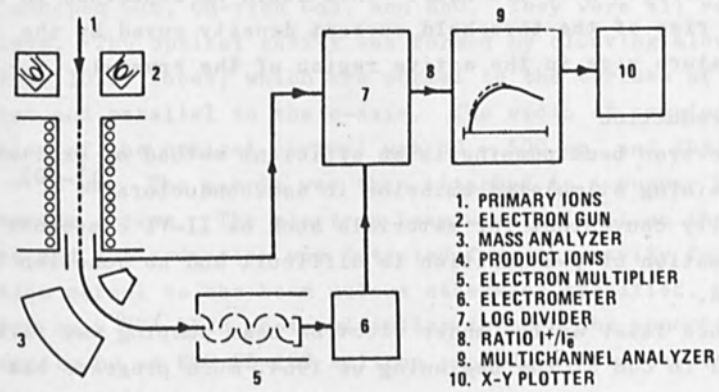


Figure 5. Block diagram of data acquisition system

Electron-Beam-Pumped Semiconductor Lasers

S. Namba*, S. Iwai*, K. Masuda**, K. Kawabe**,
H. Aritome**, and A. Masuyama**

* Institute of Physical and Chemical Research, Saitama, Japan
** Faculty of Engineering Science, Osaka University, Osaka, Japan

Abstract

Electron-beam-pumped lasers of CdS and ZnO were investigated. The following results were obtained. (1) Penetration depth of electrons into CdS was measured and was in agreement with Bethe theory. (2) The laser wavelength of CdS and ZnO shifted from the wavelength of exciton-one LO-phonon at 80°K to that of exciton-two LO-phonons at 300°K. (3) Laser threshold increased approximately linearly with temperature and showed much weaker temperature dependence compared with that given by the exciton-one LO-phonon theory. (4) Time delay of more than 100 nsec was observed in Cd-rich CdS crystals. This was well explained by the trapping model in which traps act as saturable optical absorbers. (5) Time duration of laser output pulse increased with excitation and of the order of 100 nsec. This could be explained to be due to the rise of the threshold current density caused by the temperature rise in the active region of the crystal.

1) Introduction

Electron beam pumping is an efficient method of excitation for obtaining stimulated emission in semiconductors.¹⁾⁻²⁰⁾ It is especially convenient for materials such as II-VI compounds in which the formation of p-n junction is difficult and no junction laser is reported.

Since laser action under electron beam pumping was first reported in CdS at the beginning of 1964, much progress has been made in its wavelength range, power output, and efficiency. In Table 1, semiconductor laser materials under electron beam pumping

reported at present are shown with their oscillation wavelengths, as well as those by optical pumping and carrier injection through p-n junction. The oscillation wavelength ranges from ultraviolet (ZnS: 0.3291 μm) to infrared (PbSe: 8.55 μm).

The peak output power of 10 MW at room temperature is reported in CdS crystals.²¹⁾ The overall power conversion efficiency reaches 26.5% in Cd-rich crystals²²⁾ which is near the upper limit of electron-beam-pumped semiconductor lasers (30%)²³⁾.

In this report, we describe the laser oscillation in CdS and ZnO crystals: the penetration of electrons, temperature dependences of oscillation wavelength and threshold, and the time delay and time duration of laser oscillation.

2) Experimental Procedure

Figure 1 shows the schematic diagram of experimental system. An electron gun with the maximum capacity of 22 mA at the voltage of 40 kV was used. This corresponds to the maximum current density of 7 A/cm². The pulse width and the repetition rate of the electron beam were 100~500 nsec and 20~200 Hz, respectively.

Three kinds of crystals were used for obtaining laser oscillation: undoped CdS, Cd-rich CdS, and ZnO. They were all vapor-grown platelets. The optical cavity was formed by cleaving along two parallel (10 $\bar{1}$ 0) faces, which are normal to the surface of the platelet and parallel to the c-axis. The width of samples (i.e. the dimension of the optical cavity) was 50 ~ 500 μm , and their thickness was 1 ~ 50 μm . The sample was then attached to a copper heat sink with vacuum grease. The electron beam was incident on the platelet surface and the radiation was detected from the cavity face in the direction normal to the beam unless otherwise specified. It was detected by a RCA 1P28 photomultiplier through the monochromator, or directly by an EGG SD-100 silicon photodiode and observed by a dual-beam oscilloscope.

3) Results and Discussions

3.1 Penetration depth of electrons

The electron penetration depth in CdS crystal was measured as a function of electron energy by the microscopic method referring to the luminous figure adopted by Ehrenberg and King.²⁴⁾

An electron beam was focused to impinge upon a flat cleaved face of the CdS single crystal through a hole of about 40 μm in diameter in the aluminum foil and the resultant luminous glow was photographed through the microscope with the magnification of 100. The exposed film was developed and then traced by a microphotometer.

Figure 2 shows an example of the distribution of energy loss of electrons with depth which was converted from the microphotometer trace of the photographed film of luminescence at 80°K. The electron energy was 25 keV. For the determination of the distribution of energy loss per unit length ($-\frac{dE}{dX}$), it was necessary to assume that the luminous intensity from any elemental volume in the glow is proportional to the energy dissipated in the volume. In Fig. 2 the integrated area of the curve of $-\frac{dE}{dX}$ was normalized to give the incident energy E_0 , and the curve was integrated to give the dissipated energy ($E_0 - E$). The depth at which the dissipated energy is equal to the incident energy was defined as the ultimate range R_u . A theory for the total electron penetration depth was first presented by Bethe.²⁵⁾ The total range R_B may be calculated from

$$R_B = \int_0^{E_0} \left(\frac{dE}{dX} \right)^{-1} dE \quad (1)$$

where

$$-\frac{dE}{dX} = \frac{4\pi N q^2}{m v^2} \ln \left[\frac{mv^2}{I} \right] \quad (2)$$

$$N = \frac{Z \rho}{A N_a} \quad (3)$$

where N is the number of electrons per unit volume of the material, q , m , and v are the charge, the mass, and the velocity of an electron, respectively, I is the average excitation potential of atoms, Z the atomic number, A the atomic weight, ρ the mass density of the material, and N_a the Avogadro's number. The Bethe range is

shown by the solid line in Fig. 3 for comparison with the experimental results. The agreement is satisfactory. The range R_u increases with energy as $R_u = 1.4 \times 10^{-2} E_0^{1.75}$, where R_u is in μm and E_0 in keV. It is noted from this experiment that the diffusion of the exciton or ambipolar seems to be unimportant at this temperature when the beam voltage is sufficiently high.

3.2 Laser oscillation threshold

The threshold condition of an electron beam pumped laser was first presented by Klein and discussed in the case of GaAs.²⁶⁾ Here, the threshold condition of CdS laser is discussed on the basis of the expression given by Klein.

The carrier generation rate Z excited by an electron beam is given by

$$Z = \frac{J}{q} \frac{1}{\varepsilon} \left(\frac{d\varepsilon}{dX} \right) \quad (4)$$

where J is the beam current density, q the electronic charge, and ε the average energy for creating one electron-hole pair. For the CdS crystal the value of ε is 7.5 eV.²⁷⁾ Laser action occurs when the generation rate exceeds the threshold value Z_{th} determined by the internal loss. At very low temperatures, Z_{th} is given by

$$Z_{th} = \frac{8\pi n_0^2 (h\nu)^2 \Delta(h\nu)}{h^3 c^2 n_k} \left(\alpha + \frac{1}{L} \ln \frac{1}{R} \right) \quad (5)$$

where h is the Planck's constant, c the light velocity in vacuum, $h\nu$ the emitted photon energy, $\Delta(h\nu)$ the half line width of emission spectrum, n_k the quantum efficiency, n_0 the refractive index, α the absorption coefficient, L the cavity length, and R the reflectivity of the cavity face. Figure 4 shows the threshold carrier generation rate calculated from Eq. (5) as well as the carrier generation rate calculated from Eq. (4) by using the experimental energy loss per unit length given in Fig. 2.

3.3 Temperature dependence of laser wavelength

Figure 5 shows the temperature dependence of the laser wavelength for undoped CdS. The wavelength increased with $(1.3 \pm 0.1) \text{ \AA}/^\circ\text{K}$

as the temperature increased in the range between 80°K and 250°K. This corresponds to the energy change of $-(6.5 \pm 0.5) \times 10^{-4}$ eV/°K. This is a little larger than the temperature dependence of the band gap change (-4.9×10^{-4} eV/°K).

Our observed values of the laser wavelength at 80°K and 290°K closely correspond to reported values of 4933 Å at 77°K and 5280 Å at 300°K which were assigned to the annihilation of the free exciton with emission of one LO-phonon²⁸⁾ and with emission of two LO-phonons,²⁹⁾ respectively. At temperature above 250°K, the temperature dependence of the laser wavelength was much larger than 1.3 Å/°K. These results were explained by the change of the laser transition from exciton-one LO-phonon to exciton-two LO-phonons. In Cd-rich CdS crystals, similar temperature dependence was observed.

Figure 6 shows the emission spectrum under and above threshold for ZnO at 82°K. The spontaneous emission was detected in the direction parallel to the bombarded surface. Three peaks (1), (2) and (3) were observed. The separation between each two peaks was equal to the energy of one LO-phonon (0.073 eV). The peak (1) and (2) correspond to the radiations due to the annihilation of the free exciton accompanied by the emission of one and two LO-phonons, respectively.²⁸⁾ The lasing occurred at the wavelength corresponding to the annihilation of the free exciton with emission of one LO-phonon at 82°K.

Figure 7 shows the temperature dependence of the laser wavelength of ZnO. Here again, the laser oscillation wavelength shifted from the value corresponding to the line of the exciton-one LO-phonon to that of the exciton-two LO-phonons.³⁰⁾ But at temperatures between 130°K and 170°K, simultaneous emissions of two laser lines were observed. The temperature dependence of spontaneous emissions detected in the direction normal to the bombarded surface are also shown as (1') and (2') in Fig. 7. The line (1) and (2) are the temperature dependences of the peak (1) and (2) in Fig. 6. The peak of the spontaneous emission detected in the direction parallel to the bombarded surface shifted toward

longer wavelength side compared with the peak detected in the direction normal to the bombarded surface, presumably due to the internal reabsorption of emission.

3.4 Temperature dependence of laser threshold

Figure 8 shows the temperature dependence of the laser threshold for undoped CdS, Cd-rich CdS and ZnO. It increased approximately linearly with temperature.

The temperature dependence of the laser threshold in case of free exciton-one LO-phonon is theoretically treated by Mashkevich et al.³¹⁾ and Haug³²⁾ and is shown by dotted lines in Fig. 8. The calculated threshold values increase steeply above about 50°K because the internal loss increases steeply with the growing LO-phonon population which is approximately of the form $\exp(-\frac{k\omega_{LO}}{kT})$ where $\hbar\omega_{LO}$ is the energy of the LO-phonon.

The observed change of lasing transition from the exciton-one LO-phonon to exciton-two LO-phonons shown in Fig. 5 and Fig. 7 might be one of the origins of the weak temperature dependence of the threshold.

3.5 Time response of laser oscillation

(3.5.1) Time delay

In III-V compound laser diodes, a time delay of about 1 nsec³³⁾ and in some cases of about 100 nsec³⁴⁾ are observed between the input current pulse and the laser emission pulse. Internal Q-switching is also observed in GaAs laser diodes.³⁵⁾ For II-VI compound lasers, little has been investigated about its time behavior. Since the life time of exciton is of the order of 1 nsec or less,³⁶⁾ the time response up to 1 nsec or less might be expected. It is very interesting from the point of exciton kinetics to investigate the time behavior.

Recently the time delay of about 1 nsec in optically pumped laser emission in Cd(SeS) crystals has been reported by Keune et al.,³⁷⁾ and is explained as the time required to redistribute electron-hole pairs uniformly through the sample thickness to form excitons.

We observed a long time delay of larger than 100 nsec in vapor-grown Cd-rich CdS laser pumped by an electron beam at 80°K.³⁸⁾

Figure 9 shows the output waveforms detected by SD-100 silicon photodiode for various excitation levels. Figure 9-a is that under threshold, Fig. 9-b just above threshold and Fig. 9-c and Fig. 9-d are those above threshold. At the threshold a time delay between the input pulse and the start of laser emission was larger than 100 nsec. The delay decreased as the excitation level was increased. Qualitatively, this result may be explained as it needs some quantity of excitation to start to lase. But it may be necessary to consider some mechanism to explain the large time delay of more than 100 nsec. The electron-hole pair generation rate is about $10^{26} \text{ cm}^{-3} \text{ sec}^{-1}$ at the threshold in our case. Therefore, the density of generated excitons is about 10^{17} cm^{-3} , assuming the exciton life time of 1 nsec.³⁶⁾ Because the maximum allowed density of excitons before exciton break-up is about 10^{18} cm^{-3} in CdS,³⁹⁾ it is not necessary for the generated carriers to be redistributed to form excitons before starting to lase.

Fenner proposed the trapping model to explain the time delay in GaAs diode laser in which the traps would provide a large amount of optical absorption.⁴⁰⁾ In this model lasing is prevented until the traps are sufficiently filled by the injection current and the absorption is reduced. Following the Fenner's model, rate equations of the electron-hole pairs and traps are given by

$$\begin{cases} \frac{dn_t}{dt} = B(N_t - n_t)N - \frac{n_t}{\tau_e} & (6) \\ \frac{dN}{dt} = \frac{V \cdot J}{e \cdot d} - \frac{N}{\tau} & (7) \end{cases}$$

where n_t is the number of filled traps per unit volume, N_t the number of traps per unit volume which act as optical absorbers, B the capture possibility, τ_e the characteristic trap emptying time, N the number of generated minority carriers per unit volume, τ the minority carrier life time, e the average ionization energy, V the beam voltage, J the current density, d the penetration depth of

electrons where the energy loss per unit pass length of electrons $-\frac{dE}{dX}$ was approximated by qV/d . From Eqs. (6) and (7), one can obtain the expression for N_t as a function of time, assuming $\tau \ll \tau_e$.

The instantaneous optical absorption coefficient $\alpha(t)$ caused by traps and the threshold current density $J_{th}(t)$ are given by

$$\frac{\alpha(t)}{\alpha_0} = \frac{N_t - n_t}{N_t} \quad (8)$$

$$J_{th}(t) = \frac{1}{\beta} (\alpha(t) + \alpha_f + \frac{1}{L} \ln \frac{1}{R})$$

$$\equiv \frac{1}{\beta} (\alpha(t) + G) \quad (9)$$

where α_0 is the absorption coefficient caused by traps, α_f the residual absorption coefficient caused by other than traps, β the constant given by $\beta = \frac{V}{e} \cdot \frac{h^3 C^2 \eta_t}{8 \pi \epsilon \eta_s^2 (h\nu)^2 A(h\nu)}$. Substituting the expression for $\alpha(t)$ to Eq. (9) and solving for the delay as a function of the current density J , we find

$$t_d = \frac{\tau_e}{A \cdot J + 1} \ln \left[\frac{A \cdot J + 1}{A \cdot J} \left(\frac{J}{J_{th}} - 1 \right) \frac{G}{\alpha_0} + \frac{1}{A \cdot J} \left(\frac{A \cdot J + 1}{A \cdot J_{th} + 1} - 1 \right) \right]^{-1} \quad (10)$$

where $A = \frac{B \cdot \tau \cdot V \cdot \tau_e}{e \cdot d}$, J_{th} the threshold current density at infinite delay. The time delay was calculated from Eq. (10) by substituting the parameters $J_{th} = 1.2 \text{ A/cm}^2$, $\tau_e = 0.5 \text{ } \mu\text{sec}$, $A = 2.9 \text{ cm}^2/\text{A}$, $G/\alpha_0 = 0.3$ as shown by a solid line in Fig. 10. $G/\alpha_0 = 0.3$ means that the traps induce additional 3.3 times absorption loss. The experimental values plotted in Fig. 10 were explained by this trapping model.

The fact that such a long time delay was not observed for undoped CdS and much depended on the crystals suggests that defects or some impurities are related to this long time delay. According to this model the time delay tends to zero as α_0 reaches zero.

In CdS crystals it is reported that Cd interstitials act as double donors and it's first ionization energy lies at 0.07 eV from

the conduction band.⁴¹⁾ So, these may act as optical absorbing centers because the energy difference between the valence band and this level corresponds closely to the photon energy within 3 meV. But it requires further investigation to identify its origin of the traps which act as optical absorbing centers.

(3.5.2) Time duration of laser output pulse

The duration of the laser oscillation in electron-beam-pumped semiconductors is much shorter than the duration of an input pulse and usually of the order of 100 nsec, while their durations are the same in the case of the spontaneous radiation.

We show that this is due to the increase of the threshold current density because of the temperature rise in the active region of the crystal during excitation.

Figure 11 shows an example of the output pulse shape of the light pulse detected by the photodiode in undoped CdS ($300 \mu\text{m} \times 10 \mu\text{m} \times 2 \mu\text{m}$) pumped by the 25 keV electron beam at 80°K. The laser emission decreased approximately linearly with time and then stopped after about 250 nsec from the start of oscillation. The oscillation wavelength shifted toward longer wavelength at the rate of $0.14 \text{ \AA/nsec} \cdot \text{A/cm}^2$ during excitation. The output pulse width of laser oscillation increased with increasing excitation as shown in Fig. 12.

These effects were investigated by the following methods.

(a) Transient temperature in the crystal

The rate of heat diffusion in the crystal may be described by the thermal relaxation time constant $\tau_{th} = L^2/k$ where L is the thickness of the sample, k the thermal diffusivity $k = K/c\rho$, K being the thermal conductivity and $c\rho$ the specific heat per unit volume. At 80°K for CdS crystals $K = 1 \text{ watt/cm}^2\text{K}$, $\rho = 4.8 \text{ g/cm}^3$, $c = 0.21 \text{ joule/g}^\circ\text{K}$,⁴²⁾ and then $k = K/c\rho = 1 \text{ cm}^2/\text{sec}$. As $\tau_{th} = L^2/k$ is 1 μsec for the crystal of the thickness of 10 μm , the heat

diffusion may be neglected for pulse lengths of up to 1 μ sec. For a constant heat input W per unit area distributed in a thickness d , the temperature rise is simply given by

$$\Delta T = \frac{W \cdot t}{\rho \cdot C \cdot d} \quad (11)$$

$$W = V \cdot J \quad (12)$$

From Eqs. (11) and (12), we obtain

$$\Delta T = \frac{J \cdot V \cdot t}{\rho \cdot C \cdot d} \quad (13)$$

Using the value of $qV/d=10^8$ eV/cm as shown in Fig. 2 (this is equivalent to the assumption $d=2.5$ μ m) and neglecting the temperature dependence of specific heat, we obtain the rate of the temperature rise of $0.10^\circ\text{K/nsec}\cdot\text{A/cm}^2$. As shown in Fig. 5 the wavelength shifted with temperature at the rate of $1.3 \text{ \AA}/^\circ\text{K}$, and then we finally obtain the wavelength shift of $0.13 \text{ \AA/nsec}\cdot\text{A/cm}^2$ at the beam voltage of 25 kV. This is in good agreement with observed value of $0.14 \text{ \AA/nsec}\cdot\text{A/cm}^2$.

(b) Double-pulse experiments

When the two successive pulses as shown in Fig. 13 were applied to the sample, the threshold current density for the second pulse increased. This increase may be considered to be due to the temperature rise in the active region of the crystal. By adjusting the height (above and under threshold) and the width of the first pulse, one can obtain the temperature dependence of the threshold current density similar to the curves shown in Fig. 8. In this case the temperature in the active region at the beginning of the second pulse can be estimated by the wavelength shift during the first pulse. The temperature dependence measured in this way closely, but not exactly, coincided to that obtained by changing the heat sink temperature. The difference is supposed to be mainly due to the fact that the threshold depends not only on the crystals, but even on the bombarded place of the same sample.

When the laser oscillation for the first pulse was quenched, laser oscillation could be again observed for the second pulse when the current density of the second pulse was above the threshold which is higher than that for the first pulse.

(c) Dependence of output pulse duration on the current density

The output power density of laser oscillation is given approximately by

$$P = \eta' \cdot V (J - J_{th}(T)) \quad J > J_{th} \quad (14)$$

where η' is the overall power conversion efficiency, a fairly slowly varying function of temperature and of current density, $J_{th}(T)$ is the threshold current density at temperature T . As shown in Fig. 8, $J_{th}(T)$ can be represented in the temperature range of 80°K and 200°K by

$$J_{th}(T) = J_{th}(80^\circ K) \left(1 + \frac{\Delta T}{T_0}\right) \quad (15)$$

where $J_{th}(80^\circ K)$ is the threshold current density at 80°K and ΔT the temperature rise above 80°K, T_0 the constant.

Substituting the Eqs. (13) and (14) into (15), we obtain the expression for the light output power density as a function of time

$$P = \eta' V \left(J - J_{th}(80^\circ K) - \frac{J_{th}(80^\circ K) \cdot V \cdot J}{T_0 \cdot \rho \cdot c \cdot d} t \right) \quad (16)$$

It is seen that the intensity falls to zero linearly with time. The time t_w corresponding to the thermal quenching is given by

$$\begin{aligned} t_w &= \frac{T_0 \cdot \rho \cdot c \cdot d}{J_{th}(80^\circ K) \cdot V} \left(1 - \frac{J_{th}(80^\circ K)}{J} \right) \\ &= t_{max} \left(1 - \frac{J_{th}(80^\circ K)}{J} \right), \quad J > J_{th}(80^\circ K) \quad (17) \end{aligned}$$

The value of t_w does not exceed the value of $t_{max} = \frac{T_0 \cdot \rho \cdot c \cdot d}{J_{th}(80^\circ K) \cdot V}$.

This dependence on the current density J is shown in Fig. 12, in which the value of $T_0 = 60^\circ K$ was used which was obtained from double-pulse measurement described above. The value of t_{max} is calculated to be 500 nsec in this case.

The quenching of laser oscillation were well explained by the temperature rise in the active region of the crystal.

These effects are also observed in the case of diode lasers. But in the case of electron beam pumping, about 2/3 of the incident energy is dissipated nonradiatively due to the collision of electrons with lattice,⁴³⁾ transient temperature rise effect is strengthened compared with the case in diode lasers even at 80°K.

4) Summary

For electron-beam-pumped lasers of CdS and ZnO, the following results were obtained. 1) The penetration depth in CdS crystal was given by $R_u = 1.4 \times 10^{-2} E_0^{1.75}$, where R_u is in μm and E_0 in keV, in agreement with Bethe theory. 2) The laser oscillation of these crystals occurred at the wavelength corresponding to the line of free exciton-one LO-phonon at 80°K, and to the line of free exciton-two LO-phonons at room temperatures. Between these temperatures gradual transition between the two, or simultaneous emission of both of them were observed. 3) The threshold current density increased approximately linearly with temperature in the temperature range of 80°K and 250°K. This could not be explained by the free exciton-one LO-phonon theory, the theory giving much steeper temperature dependence. Two LO-phonon emission or some other decay mechanisms should be taken into account for explaining the observed weak temperature dependence. 4) A long time delay of larger than 100 nsec was observed in Cd-rich crystal. This delay was well explained by the trapping model by Fenner, but the origin of traps must be further investigated. 5) Laser oscillation wavelength shifted toward longer side during excitation and the oscillation was quenched after about 100 nsec. These effects were explained by the temperature rise effect in the active region of the crystal.

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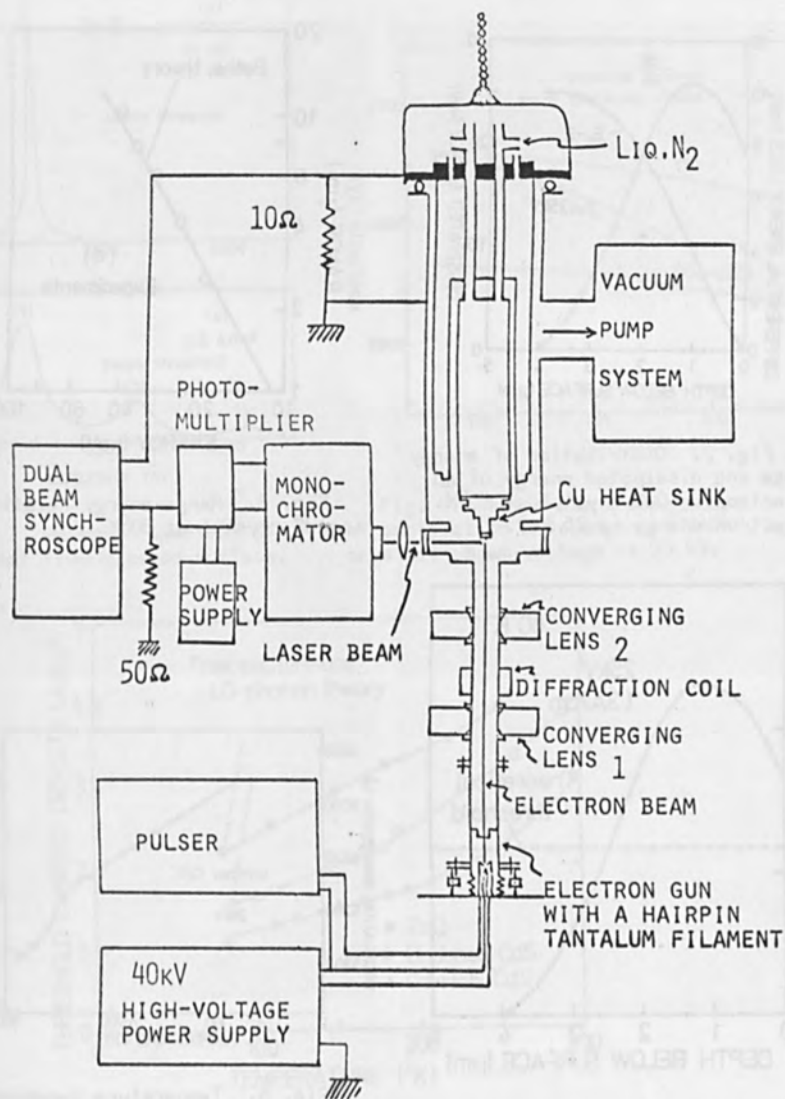


Fig. 1. A schematic diagram of experimental system

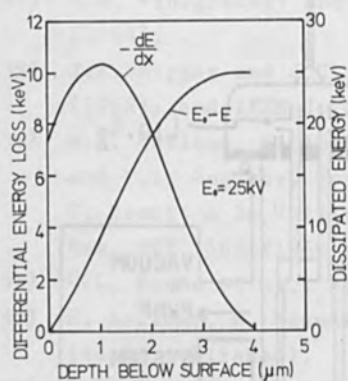


Fig. 2. Distribution of energy loss and dissipated energy of an electron in CdS crystal at 80°K. Electron energy is 25 keV.

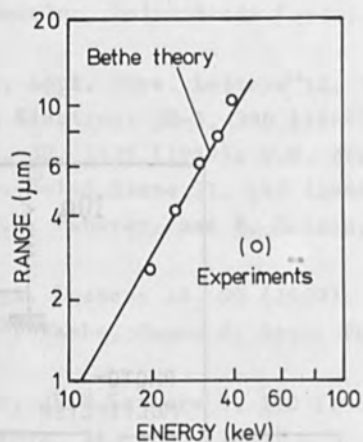


Fig. 3. Range-energy relation in CdS crystal at 80°K.

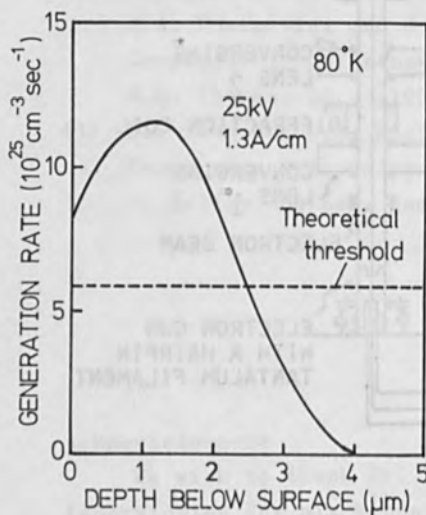


Fig. 4. Carrier generation pattern in CdS crystal at the threshold condition at 80°K.

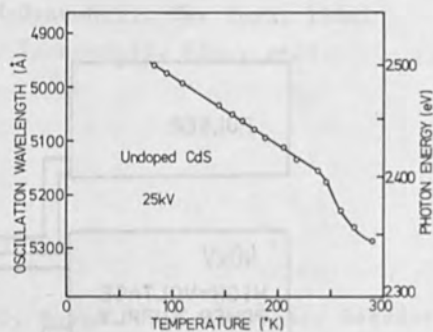


Fig. 5. Temperature dependence of oscillation wavelength of undoped CdS crystal. Beam voltage is 25 kV.

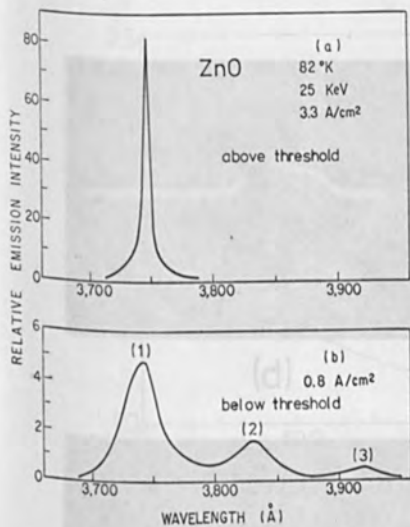


Fig. 6. Emission spectrum of ZnO crystal from cleaved surface.

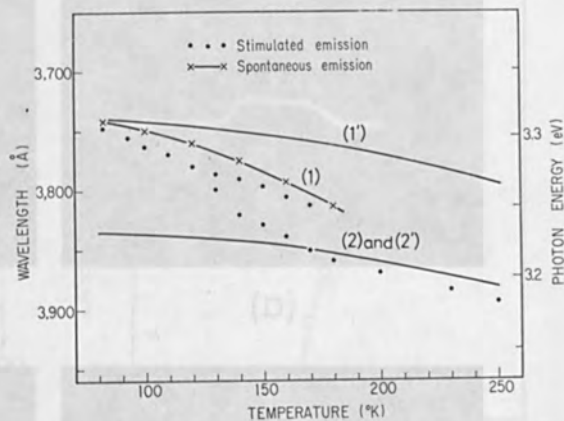


Fig. 7. Temperature dependence of oscillation wavelength of ZnO crystal. Beam voltage is 25 kV.

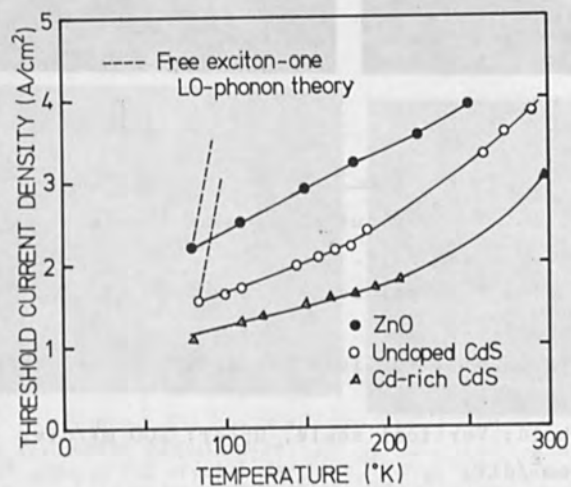


Fig. 8. Temperature dependence of threshold of ZnO (cavity length 200 μ m), undoped CdS (300 μ m), and Cd-rich CdS (220 μ m) crystals. The beam voltage was 25 kV.

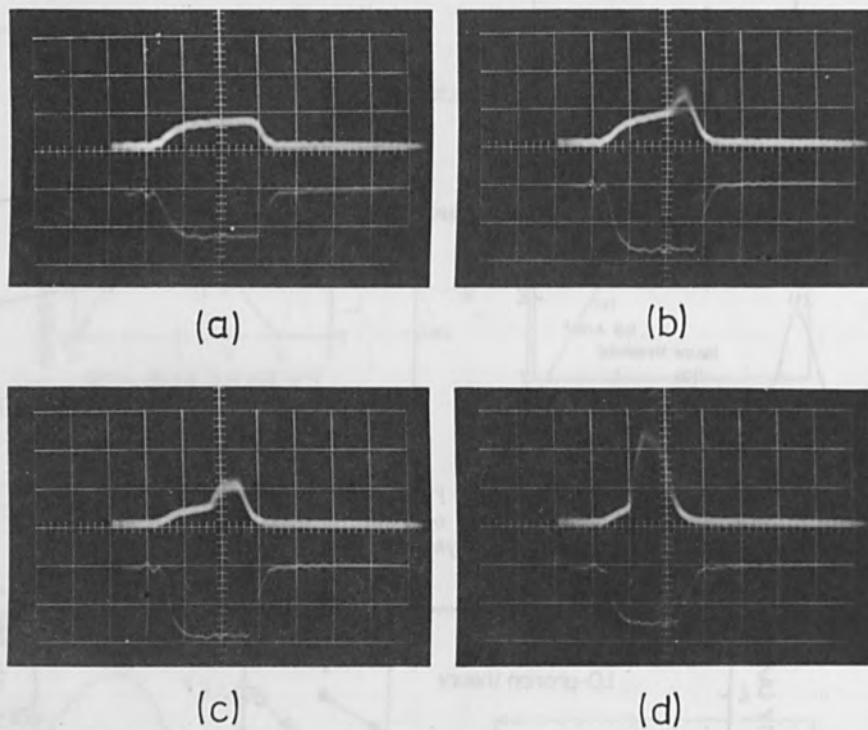


Fig. 9.

Output of photodiode for various excitation levels.

Horizontal scale: 0.1 $\mu\text{sec}/\text{div}$.

- (a) under threshold; Vertical scale, upper: 50 mV/div, lower: 1 A/cm²/div.
- (b) just above threshold; Vertical scale, upper: 100 mV/div, lower: 1 A/cm²/div.
- (c) above threshold; Vertical scale, upper: 200 mV/div, lower: 1 A/cm²/div.
- (d) above threshold; Vertical scale, upper: 500 mV/div, lower: 2 A/cm²/div.

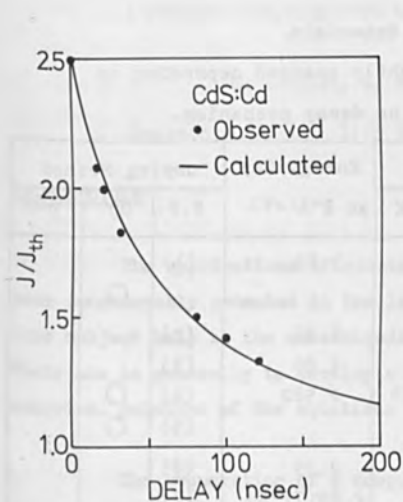


Fig. 10. Experimental and theoretical curves for the delay vs. threshold overdrive at 80°K at the beam voltage of 25 kV. Theoretical curve: $A = 2.9 \text{ cm}^2/\text{A}$, $J_{th} = 1.2 \text{ A/cm}^2$, $G/\alpha_0 = 0.3$, $\tau_e = 0.5 \text{ usec}$.

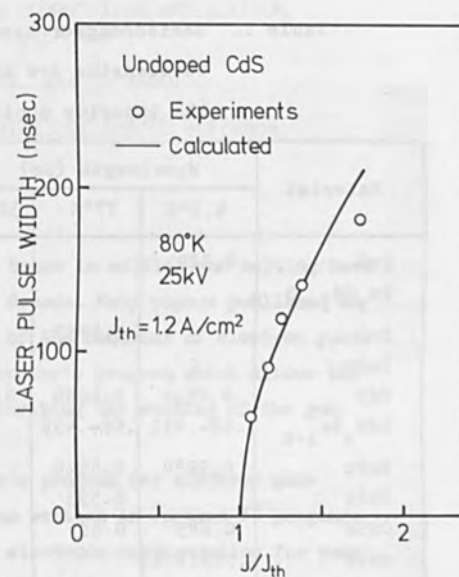


Fig. 12. Dependence of the laser pulse width on the threshold overdrive at 80°K at the beam voltage of 25 kV.

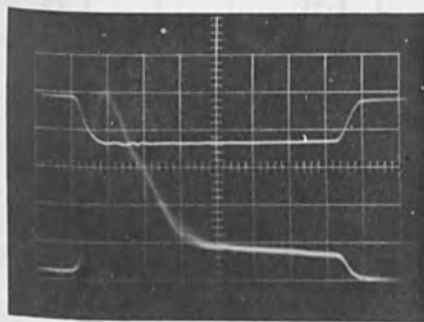


Fig. 11. Laser output pulse shape of undoped CdS crystal detected by a photodiode at 80°K. Horizontal scale: 0.1 sec/div. Vertical scale, upper: 2 A/cm²/div, lower: 20 mV/div.

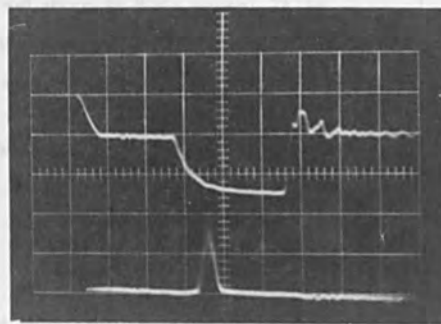


Fig. 13. Laser output of CdS crystal detected by a photodiode at 80°K. Horizontal scale: 0.2 $\mu\text{sec}/\text{div}$; vertical scale, upper: 0.7 A/cm²/div, lower: 20 mV/div.

Table 1. Semiconductor Laser Materials.

Wavelengths are slightly changed depending on the impurity doping or decay mechanism.

Material	Wavelength (μm)			Energy Gap at 0°K(eV)	Pumping Method		
	4.2°K	77°K	300°K		E.B.	Op.	p-n
ZnS	0.3291			3.82	(1)		
$\text{Zn}_x\text{Cd}_{1-x}\text{S}$						○	
ZnO		0.3757		3.30	(2)	**	
ZnSe				2.80	(3)		
CdS	0.4910	0.4960	0.528	2.582	(4)	○	
$\text{CdS}_x\text{Se}_{1-x}$	.68-.491	.69-.496			(5)	○	
ZnTe	0.5280	0.5310		2.39	(6)		
GaSe		0.593		2.09(77°K)	(7)		
CdSe	0.685	0.69		1.84	(8)	○	
CdTe	.785(10°K)			1.605	(9)	○	
$\text{GaAs}_{1-x}\text{P}_x$	.84-.64	.83-.64			(10)		○
GaAs	0.82	.83-.84	0.89	1.52	(11)	○	○
$\text{Ga}_{1-x}\text{Al}_x\text{As}$		.84-.64	.90-.78				○
$\text{In}_x\text{Ga}_{1-x}\text{As}$	.84-3.5						○
InP		0.907		1.42			○
$\text{InP}_x\text{As}_{1-x}$	.91-3.5						○
CdSnP_2		1.0100		1.22(77°K)	(12)		
GaSb	1.53			0.81	(13)		○
InAs	3.0			0.42	(14)	○	○
Te	3.72			0.34	(15)		
$\text{Hg}_{1-x}\text{Cd}_x\text{Te}$	3.8-4.1					○	
PbS	4.26			0.29	(16)		○
$\text{PbS}_{1-x}\text{Se}_x$		4-6.5 (90°K)			(17)		
InSb	5.35- 4.85			0.235	(18)	○	○
PbTe	6.41			0.19	(19)	○	○
$\text{Pb}_{1-x}\text{SnTe}$	6.4- 28					○	○
PbSe	8.55			0.15	(20)		○

A PROGRAM FOR ELECTRON GUNS COMPUTATION APPLICATION
TO A 500 KW POWER GUN

G. SAYEGH, M. ALAIS, and J. SAGOT

Société ALCATEL - 91 - BRUYERES LE CHATEL - France

INTRODUCTION

The applications of electron beams in welding and melting have been considerably promoted in the last decade. Many papers published on this subject help to the understanding of the behavior of electron guns. Their aim is generally to develop a computer's program which allows the numerical solution of the equations describing the working of the gun.

The preparation of a computer's program for electron guns started at ALCATEL in 1965. This program written in Fortran IV language for CDC 6600, led to the definition of electrode configuration for many welding and melting guns. Several guns were fabricated and are in use now in some French and foreign industrial plants. The power of these guns varies from one to several hundreds of k. W.. One of the last realizations by ALCATEL is a 500 kW power gun.

A description of the program is given in this paper. It is followed by the analysis of the configurations of several power guns capable to supply 500 kW. At last some practical realizations are presented and conclusions are drawn from the comparison of the theoretical results with the actual behaviors of these guns.

1.- THEORETICAL CONSIDERATIONS

An electron gun is constituted of an emitting surface (cathode), an accelerating electrode (anode) and one or several focusing electrodes which enable to act upon the characteristics of the beam. The user of an electron gun is interested by the power of the beam, its shape in the gun and the current density repartition in the different sections of the beam.

To estimate the intensity of the current emitted by the cathode one needs to know the potential repartition in the gun and especially near the emitting surface. The presence of space-charge modifies these potentials which can be determined by successive approximations. To evaluate the current density repartition in the beam, it is necessary to compute the electron trajectories in the gun (2nd order partial differential equation) and to know the currents transported by each of the elementary tubes delimited by two successive trajectories.

1.1.- RESOLUTION OF POTENTIAL EQUATION

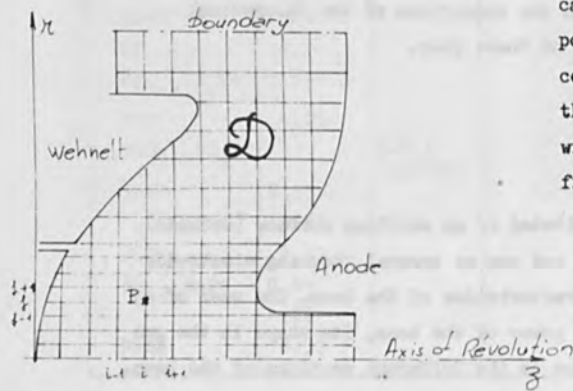
The electric potential V at a point (r, z) of an axially symmetric gun, in presence of space-charge $(\rho(r, z, V))$, satisfies POISSON's equation

$$(1) \quad \frac{\partial^2 V}{\partial r^2} + \frac{1}{r} \frac{\partial V}{\partial r} + \frac{\partial^2 V}{\partial z^2} = - \frac{\rho}{\epsilon_0}$$

ϵ_0 is the dielectric constant of free space.

The potential V is defined on all points of the boundary delimiting the region of integration $\mathcal{D}$ either by potentials of the electrodes or, for the region far from the emitting zone, by simple

calculation of the repartition of potential in a plane or cylindrical condenser. It is known furthermore that in space charge flow the current will so adjust itself that the electric field at the cathode is zero.



The solution of equation (1) in the region $\mathcal{D}$ is obtained by a classical method : The region is divided into rectangular meshes (there can be several dimensions of the mesh in the r direction) and the points are located by their co-ordinates (i, j) . The partial differential equation (1) is approximated by a set of finite difference equations relating the potential V_{ij} to its neighbouring potentials ($V_{i+1,j}$, $V_{i-1,j}$, $V_{i,j+1}$, $V_{i,j-1}$...). This set of algebraic equations is solved by iteration methods and relaxation procedures. Thus instead of a continuous solution of V one gets discrete values of V at all mesh points situated in the region $\mathcal{D}$.

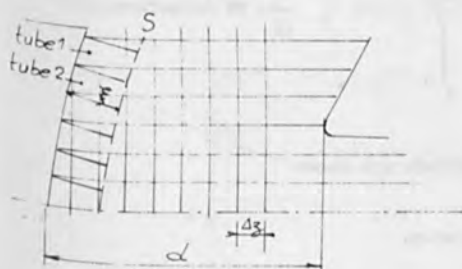
Space-charge density ρ is a function of the potential. To estimate the potential in the gun one should use successive approximations until the desired convergence and accuracy criterion is attained. Let us point out that the number of iterations necessary to obtain an accuracy on V of 0.01 by using over relaxation technique, is about 100 iterations.

1.2.- CATHODE EMISSION

In power guns the electron flow is limited by space-charge density. To estimate the emission current one can use the following

artifice, proposed first by DEIMEL

(1) and used by several authors who studied this problem (2), (3), (4) (5).



Let us consider a virtual surface S parallel to the cathode and situated at the distance ξ from it. The electron beam is decomposed into 10 to 20 elementary concentric tubes. Each tube is

delimited by two successive trajectories. Let us suppose furthermore that the part of the tube included between the cathode and the surface S can be assimilated to a plane diode. Then the flow in the diode is limited by space-charge and the current density on the emission surface is given by the CHILD-LANGMUIR law :

$$(2) \quad J = \frac{4}{9} \epsilon_0 \sqrt{\frac{2e}{m_0}} \frac{V_K^{3/2}}{\zeta^2}$$

J is the current density per unit area

e and m_0 are the charge of the electron and its mass at rest

V_K electric potential of the electrode located at ζ from the cathode.

V_K is estimated for all the tubes from potentials of the neighbouring mesh points. V_K is not identical for all tubes and the current density at the cathode is not uniform. One can consider this density as constant for every tube. The current of the beam is the sum of currents transported in all the tubes. It is function of the arbitrary distance ζ . It has been established, in agreement with HAMZA (3), that for guns working as diode (no difference of potential between the cathode and the focusing electrode) the actual current emitted by the cathode concords quite well with the calculated one if ζ verifies the following inequalities :

$$(3) \quad \left. \begin{aligned} 0.15 < \frac{\zeta}{d} < 0.3 \\ 2 \Delta z < \zeta < 5 \Delta z \end{aligned} \right\}$$

d is the distance between cathode and anode

Δz is the scaled mesh dimension

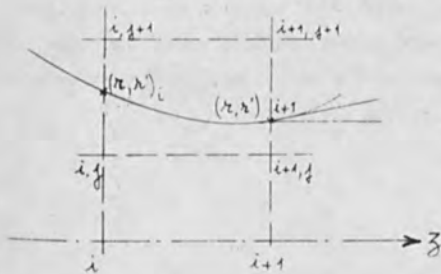
1.3.- EQUATION OF THE TRAJECTORIES

The partial differential equation defining the trajectory of a non relativistic electron $r(z)$ in an axially symmetric gun and an electric field $\left(\frac{\partial V}{\partial r}, \frac{\partial V}{\partial z} \right)$ can be derived from LORENTZ force equations.

$$(4) \quad \frac{d^2 r}{dz^2} = \frac{1 + \left(\frac{dr}{dz} \right)^2}{2V} \left[\frac{\partial V}{\partial r} - \frac{dr}{dz} \frac{\partial V}{\partial z} \right]$$

The estimation of electric field components $\left(\frac{\partial V}{\partial r}, \frac{\partial V}{\partial z} \right)$ at any point of the integration region can be easily done from the potentials of the neighbouring mesh points. The integration of equation (4) is then accomplished by RUNGE-KUTTA's method (6) using four approximations and an integration step equal to the mesh dimension in the z direction.

The RUNGE-KUTTA's method allows the estimation of $r(z)$ and $r'(z)$ at a point situated at abscissa $(i+1)$ from $r(z)$ and $r'(z)$ at abscissa (i) but it implies the knowledge of potentials and fields, not only of the neighbouring mesh points but also of some intermediate points between i and $(i+1)$. A special subroutine carries out this estimation. Initial conditions at the cathode are fixed from the characteristics of space-charge flow which indicate that trajectories follow paths normal to the emission surface. One computes, by degrees, all the trajectories forming the beam in the integration region. It can be shown that the error committed on the ordinate of the trajectory is over-estimated by $\frac{1}{120} \left(\frac{\Delta z + \Delta r}{2} \right)^5$, which is insignificant.

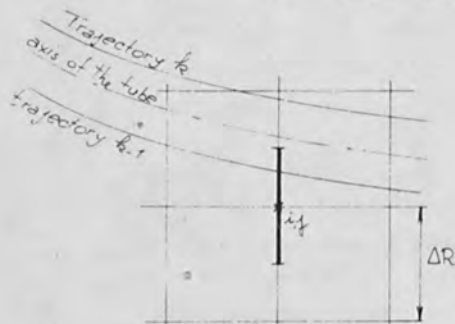


1.4.- SPACE-CHARGE REPARTITION IN THE GUN

Space-charge density at a point P (i,j) of a gun in which the current density is J and the electron velocity v is given by :

$$(5) \quad \rho = \frac{J}{v}$$

The velocity of an electron at P (i,j) is easily estimated from the electric potential at this point V(i,j). To evaluate the current density at every mesh point of the region, one needs to know the position of every elementary tube composing the beam with regard to this point.



Influences zones, of Δr width, are traced around each mesh point (i,j). When an elementary tube goes through this zone it contributes to increase the current density at (i,j) proportionally to the current carried by the tube and to the distance that separates the axis of the tube from the mesh point. A subroutine evaluates the current density

supplied by each elementary tube according to the nature of the mesh point (5) (regular, irregular, special...) and sums up the total current density at every point.

Space-charge is estimated after each trajectory computation and are introduced in the last POISSON equation in order to get a new solution with a better accuracy. One can accelerate the convergence by using at the N^{th} iterations all the solutions, previously determined; modulated by an under-relaxation coefficient (2), (3).

1.5.- BLOC DIAGRAM AND RESULTS

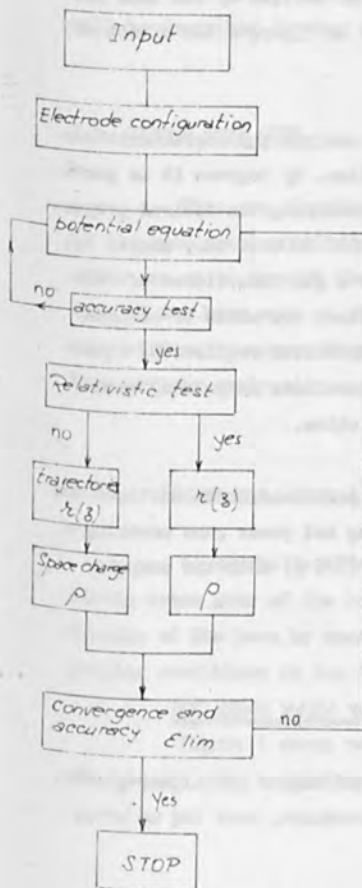
All equations are scaled to render them dimensionless (5).

The program computes, with a pretty good accuracy, equation (1) without space-charge (LAPLACE equation). Trajectories and space charge repartition are defined and one seeks a solution to POISSON's equation with an accuracy better than ε . Trajectories are determined once more from the new electric potential repartition and space-charge densities are calculated. They are introduced, next, in the last POISSON equation and one seeks a new solution with an accuracy better than $\frac{\varepsilon}{2}$. The program ends when the convergence criterion ε_{lim} , an input parameter, is attained.

When the accelerating voltage is over 30 kV, the program takes into account the relativistic corrections on the mass (trajectories eq. 4) and on the velocity (space-charge eq. 5).

The computer analysis furnishes the following characteristics :

- A map of electrode configuration in the gun and the nature of the mesh points in the integration region.
- LAPLACE solution with a given accuracy and electron trajectories without space charge.



- POISSON's solution with an accuracy better than ϵ_{im} and electron trajectories with space-charge
- current density and space-charge at all mesh points.
- perveance of the gun and powers for different accelerating voltages.

The effect of space-charge on the characteristics of the beam in a power gun is very important and it is essential to take them into account. Space-charge increases the section of the beam and reduces the cathodic emission. One can see on figure 2 the effect of space-charge on the beam.

This program is commonly used to analyze the characteristics of the beam of a given electrode configuration. By degrees it is possible to define an electrode configuration furnishing the desired properties of a beam. The time of execution on a CDC 6600 is very short. Let us point out, for instance, that to analyze a gun constituted of 1650 meshes with a beam composed of 16 trajectories, one needs 75 seconds to get an accuracy better than 1 %. The computation requires 92 iterations for LAPLACE's solution and 7 major iterations comprising each 20 minor iterations (2) for POISSON's solution.

Experimental results, concerning perveance, beam sections and current density, obtained on several welding and power guns covering a wide range of powers are in good agreement ($\pm 5\%$) with the computer analysis of electrode configuration.

2.- SOME CONFIGURATIONS OF THE ELECTRODES OF 500kW POWER GUN

Industrial requirements for guns of higher and higher powers, in order to refine ingots of increasing dimensions, have led us after

the construction of a 200 kW and 385 kW guns to design a 500 power gun. The characteristics of a dozen of electrode configurations, able to furnish this power, have been analyzed by the computer program. The guns analyzed can be grouped into two categories :

- Pierce type guns,
- Guns with extracting grid.

The most interesting configurations, which have been analyzed, will be described further down.

2.1. PIERCE TYPE GUNS

They are classical Pierce Guns, whose powers are regulated by the accelerating voltage. The electrode configuration should be chosen so as to furnish the desired power and the required characteristics of the beam such as the diameter of the beam, its aperture and current density repartition at the cross over. Two variants of this type will be analyzed.

2.1.1.- Pierce gun (type A)

We sought in this variant to obtain 500 kW and an electrostatic cross over of the least section in order to get an easy concentration of the beam by means of magnetic lenses and to ensure good pumping conditions in the different parts of the guns.

Figure 1 shows schematically the electrode configuration of the gun and some trajectory plot in presence of space-charge.

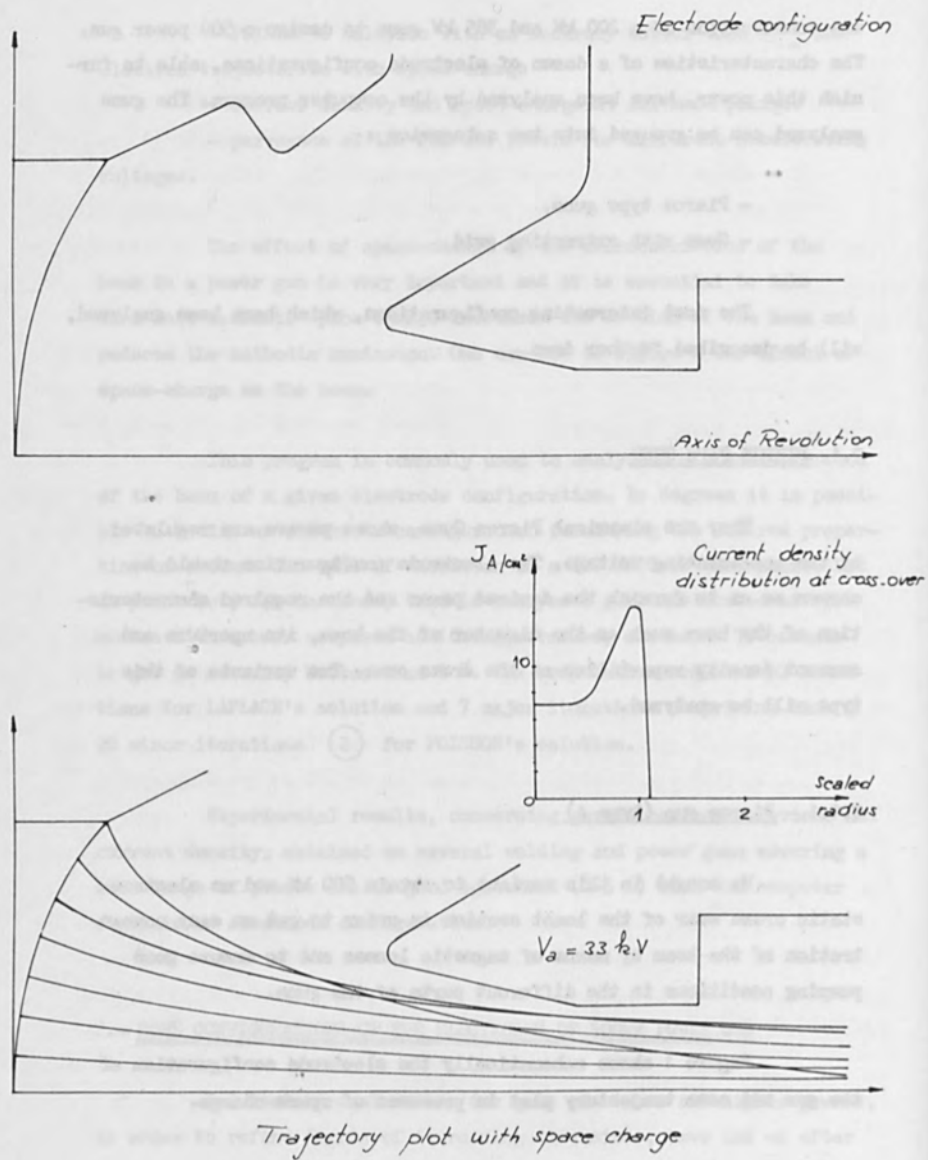


Fig. 1. 500 kW Power Gun (Type A)

There is scarcely a real cross over in a power gun. As a matter of fact, trajectories cross each other and their slopes are not null at the minimum beam section. Nevertheless one can define a quasi cross over at the minimum section of the beam characterized by the diameter and the aperture of the beam.

In the gun type A, the diameter of the cross over is 8 mm and the aperture is 4×10^{-2} radians. The current density is quite uniform at the cross over and the perveance of the gun is $2,5 \times 10^{-6}$. One should therefore operate at an accelerating voltage of 33 kV in order to get 500 kW.

The following comments can be made on this gun.

- the small distance that separates anode from wehnelt (11 mm) may favour electric discharge between these electrodes. If one takes into consideration results published by STAPRANS (7) on discharge in a low power electron gun, he can say that 11 mm are sufficient to avoid discharge at accelerating potentials less than 35 kV, but it was hazardous to extrapolate STAPRANS's results to very high power guns. Experience acquired on discharge in high power guns, since the design of type A, allows us to affirm now that the probability of discharge is very small in the gun A when the accelerating potential is less than 35 kV.

- To diminish the heating power of the cathode and its thermal deformations, it is interesting to use the smallest surface of emission capable of producing the desired power.

2.1.2.- Pierce gun (type B)

In this variant we imposed a distance of 20 mm between the anode and the wehnelt and an emitting surface smaller than type A (ϕ 40 instead of ϕ 45).

Figure 2 shows schematically the electrode configuration and the trajectory plot taking into account space charge and neglecting them. One can remark the great difference which exists between the characteristics of the beam when space-charge is considered in the computation. It is surely very erroneous to neglect space-charge in the determination of the characteristics of the beam in a power gun.

Cross over diameter is 20 mm and the aperture of the beam is 2.5×10^{-2} radians. The calculated perveance is 1.91×10^{-6} . To supply 500 kW, the accelerating voltage of the gun should be 37 kV.

We contemplate to use a X-ray probe in order to analyze experimentally the melting bath in the crucible. This implies to work at a constant accelerating voltage for various powers. We planned to vary the power of the gun by a bias potential applied on the wehnelt. The bias potential regulates the intensity in the gun, thus its power, without acting on the accelerating voltage. It alters the potential repartition in the gun, especially near the emitting surface which leads to the modification of the trajectories and the emission of the cathode. Figure 3 shows the characteristics of the beam for two bias of - 1.1 kV and - 1.85 kV and an accelerating potential of 37 kV. When a bias is applied, the surface S defined in 1-2, in order to estimate the current emission, is replaced by several surfaces located at $\xi_1, \xi_2, \dots$ from the cathode (the shortest distance $\xi \neq 0.1$ d



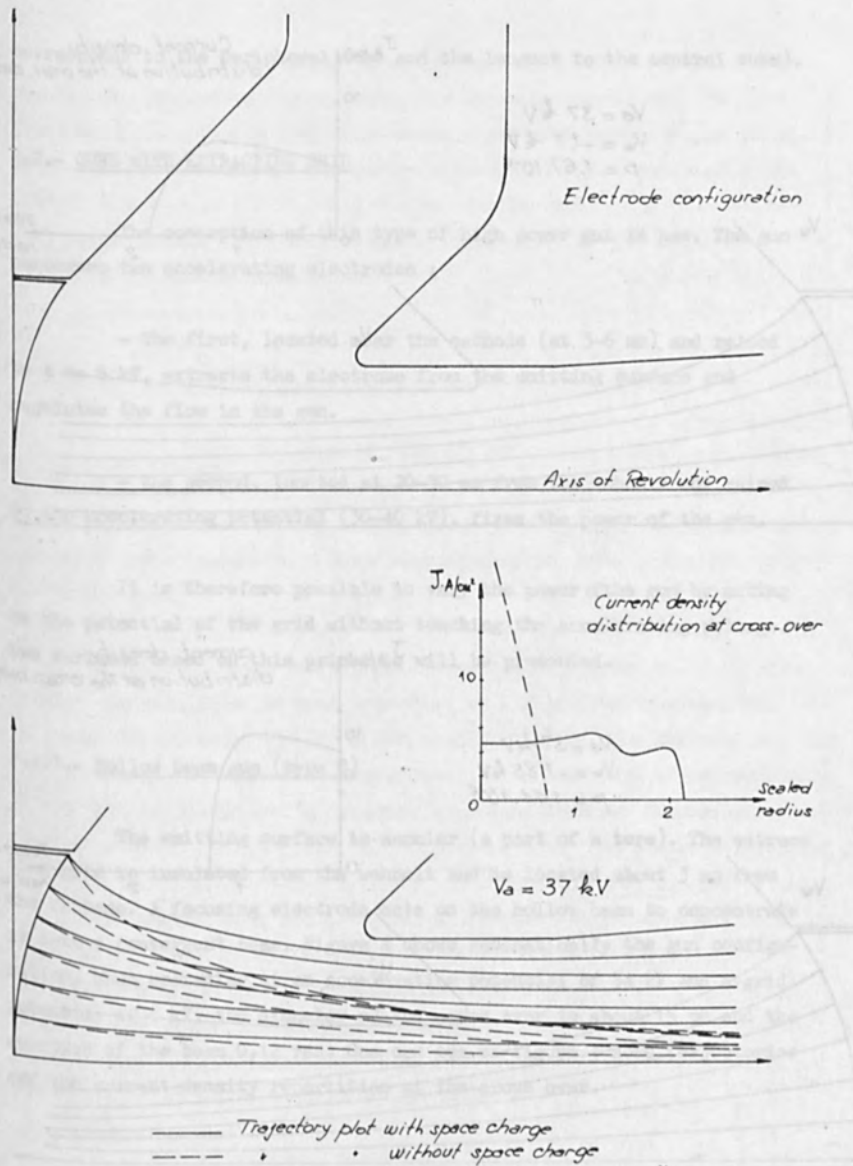


Fig. 2. 500 kW Power Gun (Type B)

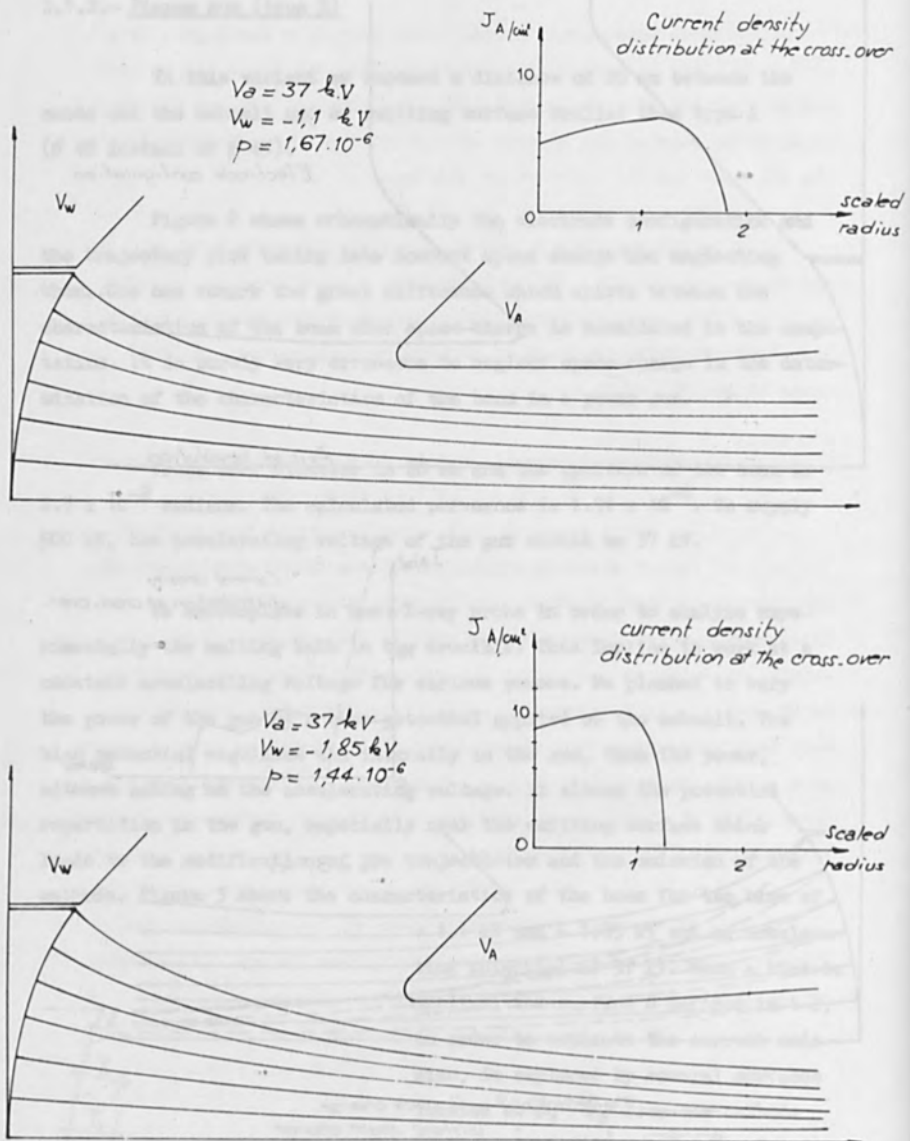


Fig. 3. 500 kW Power Gun (Type B). Bias Potentials.

corresponds to the peripheral tube and the longest to the central tube).

2.2.- GUNS WITH EXTRACTING GRID

The conception of this type of high power gun is new. The gun possesses two accelerating electrodes :

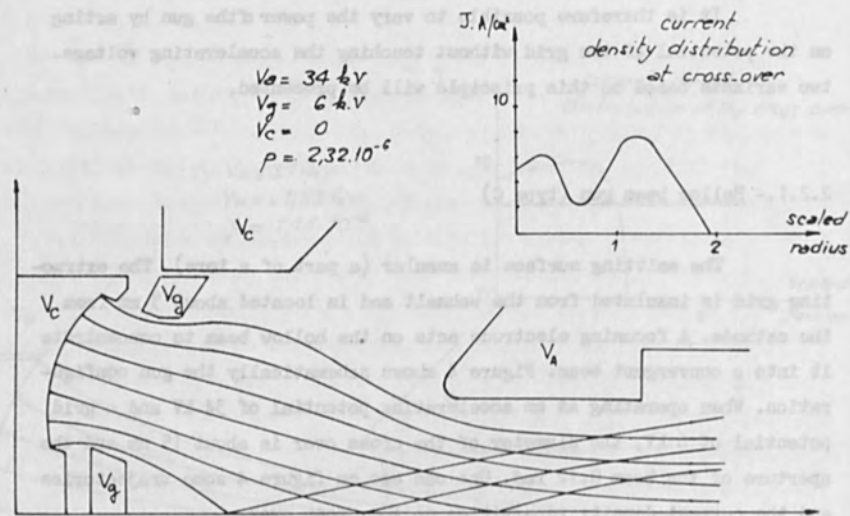
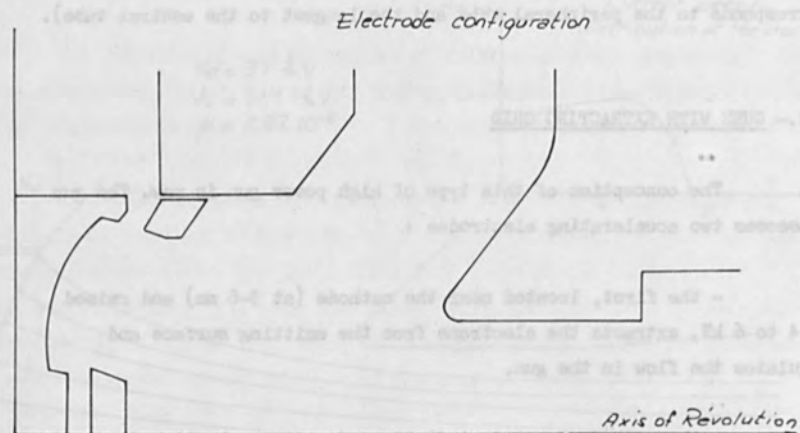
- the first, located near the cathode (at 3-6 mm) and raised to 4 to 6 kV, extracts the electrons from the emitting surface and regulates the flow in the gun.

- the second, located at 20-30 mm from the wehnelt and raised to the accelerating potential (30-40 kV), fixes the power of the gun.

It is therefore possible to vary the power of the gun by acting on the potential of the grid without touching the accelerating voltage. two variants based on this principle will be presented.

2.2.1.- Hollow beam gun (type C)

The emitting surface is annular (a part of a tore). The extracting grid is insulated from the wehnelt and is located about 3 mm from the cathode. A focusing electrode acts on the hollow beam to concentrate it into a convergent beam. Figure 4 shows schematically the gun configuration. When operating at an accelerating potential of 34 kV and a grid potential of 6 kV, the diameter of the cross over is about 15 mm and the aperture of the beam 0.12 rad. One can see on figure 4 some trajectories and the current density repartition at the cross over.



Trajectory plot with space charge

Fig. 4. 500 kW Power Gun (Type C)

It has been possible to render the electrostatic interaction between the extracting region (comprised between cathode and grid) and the accelerating region (comprised between grid and anode) negligible, by choosing appropriately the electrode configuration. Consequently the form of the beam in the extracting zone remains unchangeable for different powers (characteristic of a space-charge flow in the gun), but varies in the accelerating zone.

2.2.2.- Multiple cathode gun (type D)

The emitting surface is composed of several elementary cathodes (7 to 13) located on a spherical surface. An extracting grid is placed in front of each cathode forming with it an elementary gun. The wehnelt and the anode focus the multiple beam into a principal beam of desired characteristics.

Figure 5 shows schematically the gun configuration. The permeance of the gun, when the grid potential is 6 kV and the accelerating potential 35 kV, is 2.12×10^{-6} . Its power is 500 kW. The diameter of the cross over is 18 mm and its aperture 0.2 radians. The concentration of the beam on the target by magnetic lenses is delicate because of its great aperture.

This gun is a juxtaposition of several elementary guns. To regulate the power of the gun, for a constant accelerating voltage, one can either vary the grid potential or switch up some of the elementary guns. As for type C, the shape of the beam is unchanged in the extracting region for the various powers. Figure 5 shows some trajectory plot and the characteristics of the beam at the cross over.

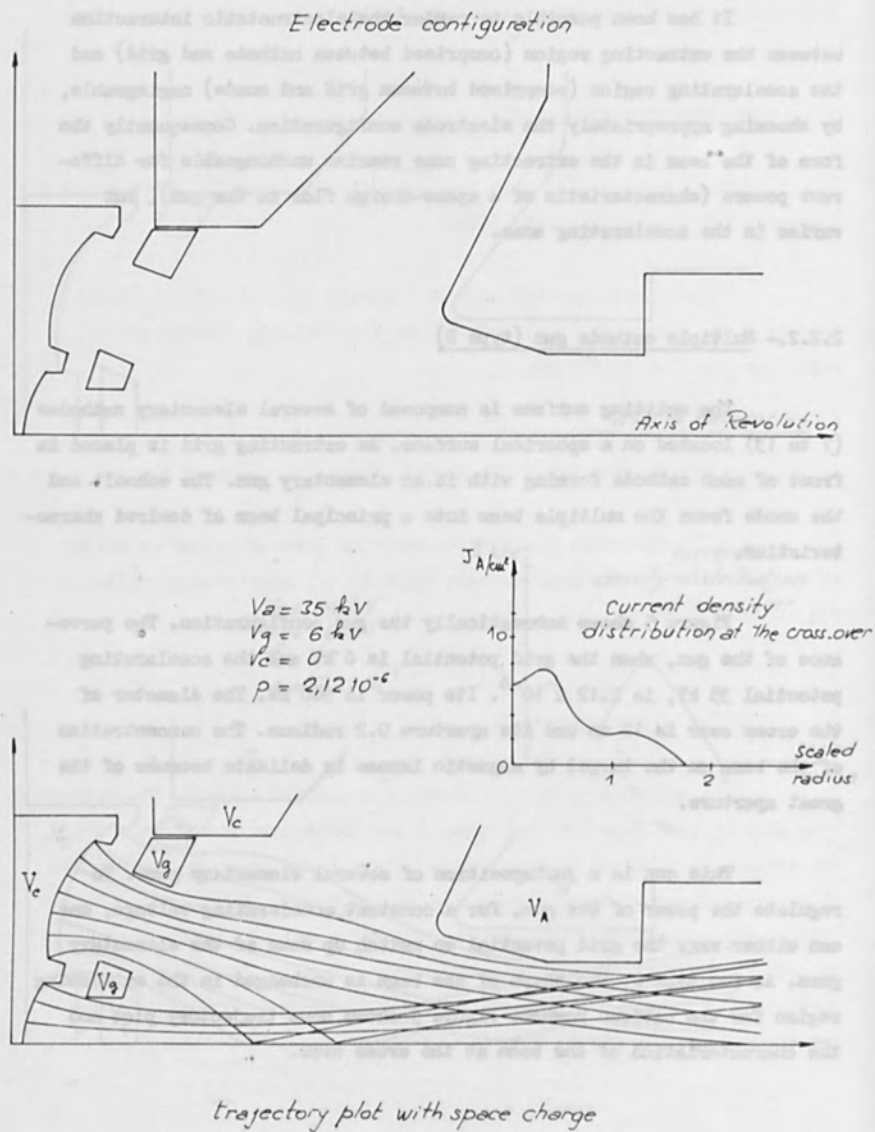


Fig. 5. 500 kW Power Gun (Type D)

3.- ELECTROMAGNETIC PART OF GUN B

The electron beam covers two meters before reaching the target. Behind the anode it diverges naturally because of the space-charge. It is necessary to concentrate it on the target by means of magnetic lenses. In B type gun two magnetic lenses separated by a diaphragm (figure 6 and 7) are used to concentrate the beam. The gun is thus divided into three compartments which are pumped separately to get the desired vacuum.

3.1.- SHAPE OF THE BEAM BELOW THE ANODE

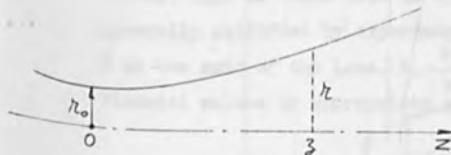
To determine the shape of the beam below the anode one can obviously extend the integration region to the desired zone, but this will increase considerably the number of mesh points and rise the cost of the computation.

To estimate the shape of the beam in this region it is possible to use SPANGENBERG's results (8) which allow us to evaluate the radius of a paraxial beam r at an abscissa z from the radius of the

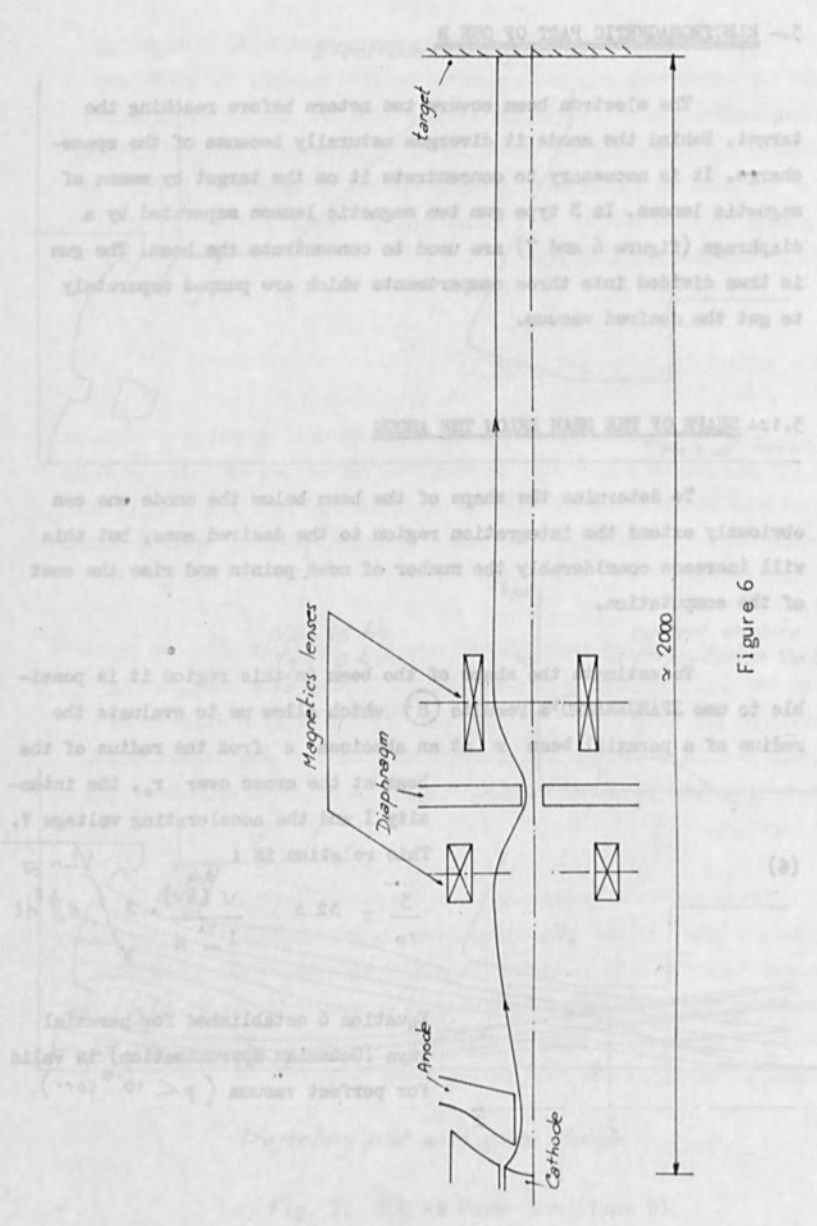
beam at the cross over r_0 , the intensity I and the accelerating voltage V .

This relation is :

$$(6) \quad \frac{z}{r_0} = 32.3 \frac{V^{3/4} (kV)}{I_m^{1/2} A} \times 2 \int_0^{\sqrt{L_n \frac{r}{r_0}}} e^{t^2} dt$$



Equation 6 established for paraxial rays (Gaussian approximation) is valid for perfect vacuum ($p < 10^{-9}$ torr).



It is represented by a graph which gives $\frac{r}{r_0}$ as a function of $\frac{z}{r_0}$. When the pressure is $> 10^{-9}$ torr equation (6) over-estimates the radius of the beam. SPANGENBERG has verified that the radius of the actual beam is only 0.6 time the calculated radius by equation (6) when the vacuum is about 10^{-7} torr. The lowest pressure in a power gun is about 10^{-5} torr. The analysis of experimental results on power guns (40 kW, 200 kW, 385 kW) is in accordance with diameters calculated by (6) if they are multiplied by 0.6.

3.2.- SHAPE OF THE BEAM BEHIND THE MAGNETIC LENSES

In the neighbourhood of a magnetic lens, the equation defining electron trajectory should take into account the components of the magnetic field. When the axis of the lens is aligned with that of the electrodes, equation (4) becomes in an equipotential electric zone :

$$(7) \quad \frac{d^2 r}{dz^2} = \frac{1 + \left(\frac{dr}{dz}\right)^2}{2[\phi - k^2 A^2]} \left\{ \frac{dr}{dz} \frac{\partial A}{\partial z} - \frac{\partial A}{\partial r} \right\}$$

$$k^2 = \frac{e}{2m_0}$$

A is the magnetic vector potential

The estimation of the magnetic vector potential in shielded lenses, such as those used in electron guns, is very difficult. One is generally satisfied by experimental measurements of the induction field B on the axis of the lens. A, $\frac{\partial A}{\partial r}$ and $\frac{\partial A}{\partial z}$ are deduced from the experimental values by appropriate analytical developments.

The actual behavior of electron trajectories in the vicinity of the magnetic lenses in power guns is not entirely defined by equation (7). As a matter of fact positive ions issued from the ionization of gas molecules in this region of the gun alter considerably the trajectories and it is very complicated to estimate, quantitatively, this alteration. Practically the determination of the shape of the beam in this region is made by using simplified calculations of paraxial trajectories (Gaussian approximation).

It is possible to evaluate the section of the beam at different abscissa if one knows the electron optical characteristics of the magnetic lens (focal distance, principal plane, aberrations...). The electron optical characteristics of magnetic lenses have been studied in details by several authors (9), (10). There exist several charts (10) which give all the characteristics of the lens from its geometric configuration, its excitation and the accelerating voltage of the beam. Experimental results on the shape of the beam between the anode and the diaphragm in a 385 kW power gun agree quite well with those evaluated by Gaussian optics.

Concerning the shape of the beam near the target, it has been established experimentally that positive ions liberated from the metallic vapors play a great role in the characteristics of the beam. They neutralize the electronic space-charge and concentrate the beam like an electrostatic lens. This is equivalent to a decrease of the focal distance of the lens or an increase of its excitation. When theoretical excitations are diminished by 15 % to 20 % the calculated section of the beam on the target is in good agreement with the actual section.

4.- PRACTICAL APPLICATIONS

Several guns of various powers (40 kW, 100 kW, 200 kW, 385 kW) have been fabricated at ALCATEL. Some of them have been used

for several years and have accomplished the refinement of several hundreds ingots. The description of a 200 kW and 500 kW guns will be given further down.

4.1.- 200 kW POWER GUN

This power gun equiped a KUHLMANN's factory in FRANCE since 1967. It worked for more than 5000 hours and refined more than 500 ingots (1000 mm length and 60 - 150 mm diameter) of molybdenum, niobium, vanadium and hafnium.

Photo 1 shows a general view of the installation. One can distinguish the working enclosure (center), the pumping canalisations (right) and the commanding board (left). Photo 2 shows the technical pit, the hydraulic jack and the ingot container. On photo 3 is presented the gun with the focalisation and deflecting coils which surmount the working enclosure. Photo 4 shows a detailed view of the cathode and the wehnelt.

The most interesting technical characteristics of this gun are the following :

- main power 380 kVA
- main voltage 380 V triphase
- power of the gun 200 kW for 30 kV
- pumping unit of the working enclosure :
 - diffusion pumps 2 x 11000 lit/s
 - Root's pumps 2 x 1600 m³/h
 - mechanical pumps 2 x 200 m³/h
- cooling water 25 m³/h

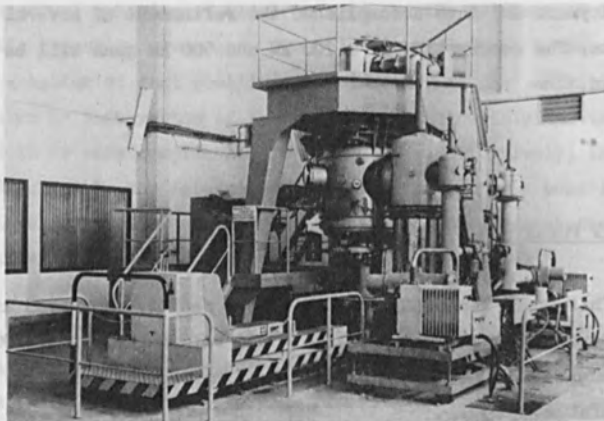


Photo No. 1. General View of the 200 kW Power Gun

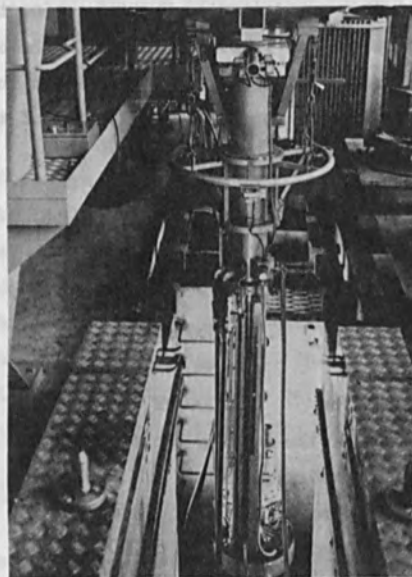


Photo No. 2. A View of the Pit and Container of the 200 kW Power Gun

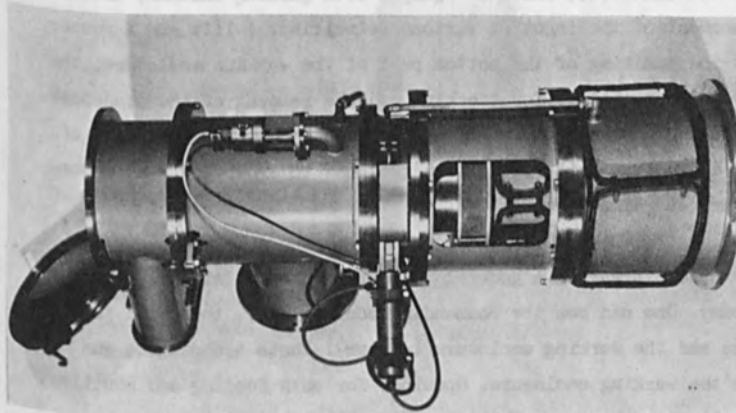


Photo No. 3. 200 kW Electron Gun with Focusing and Deflecting Coils.

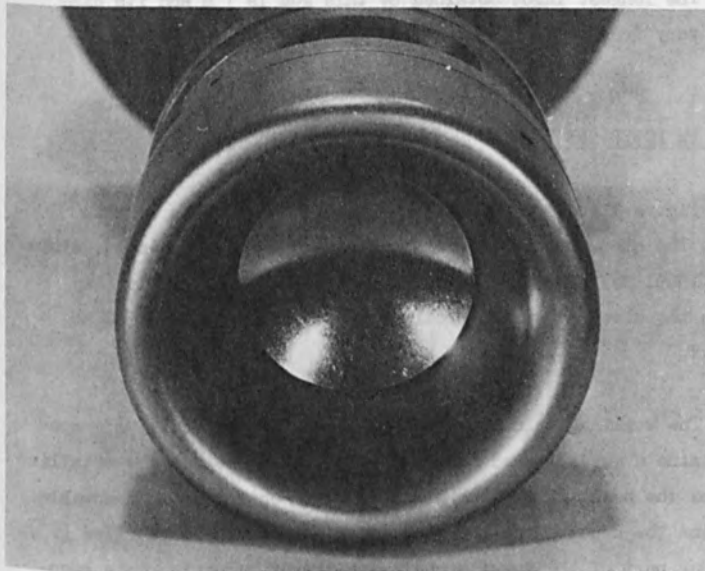


Photo No. 4. Cathode and Wehnelt of the 200 kW Power Gun.

The longest ingot obtained by this gun is 1 m and its diameter may vary from 60 to 150 mm

4.2.- 500 kW POWER GUN

Figure 7 presents schematically the installation of this power gun. The electrode configuration of this gun has furnished, since September 1969, 385 kW under 40 kV. It will be equipped shortly (May 1970) with the electrode configuration type B which supplies 500 kW under 37 kV.

The working enclosure (10) made of mild steel of 15 m³ capacity, contains a water cooled condenser (6) which receives the metallic vapors from the bath. The bottom of the working enclosure is removable and sustains the crucible support (3) and a detachable clack-valve (13). On the upper part of the working enclosure there are appropriate openings for the granule supply (5), for the metal feeding device (11) and for the visual observation of the bath. The crucibles (3) are made of steel with an inner jacket of copper. A hydraulic jack (9) passing through the container (4), which is equipped with guiding columns, allows the displacement of the ingot at various velocities. A lift and a runner (8) permit the handling of the bottom part of the working enclosure, the transport of the container for stocking and the removal of the condenser (6) for cleaning. Several pumps (7) ensure good vacuum in every part of the gun. A deflecting coil (12) allows the beam to sweep the bath according to a predetermined program.

Photo 5 presents a general view of the installation (electron gun put down). One can see the commanding board (left), the metal feeding device and the working enclosure (center). Photo 6 shows the gun mounted on the working enclosure. Openings for bath feeding and scuttles for observation can be seen. Photo 7 shows the technical pit, one can

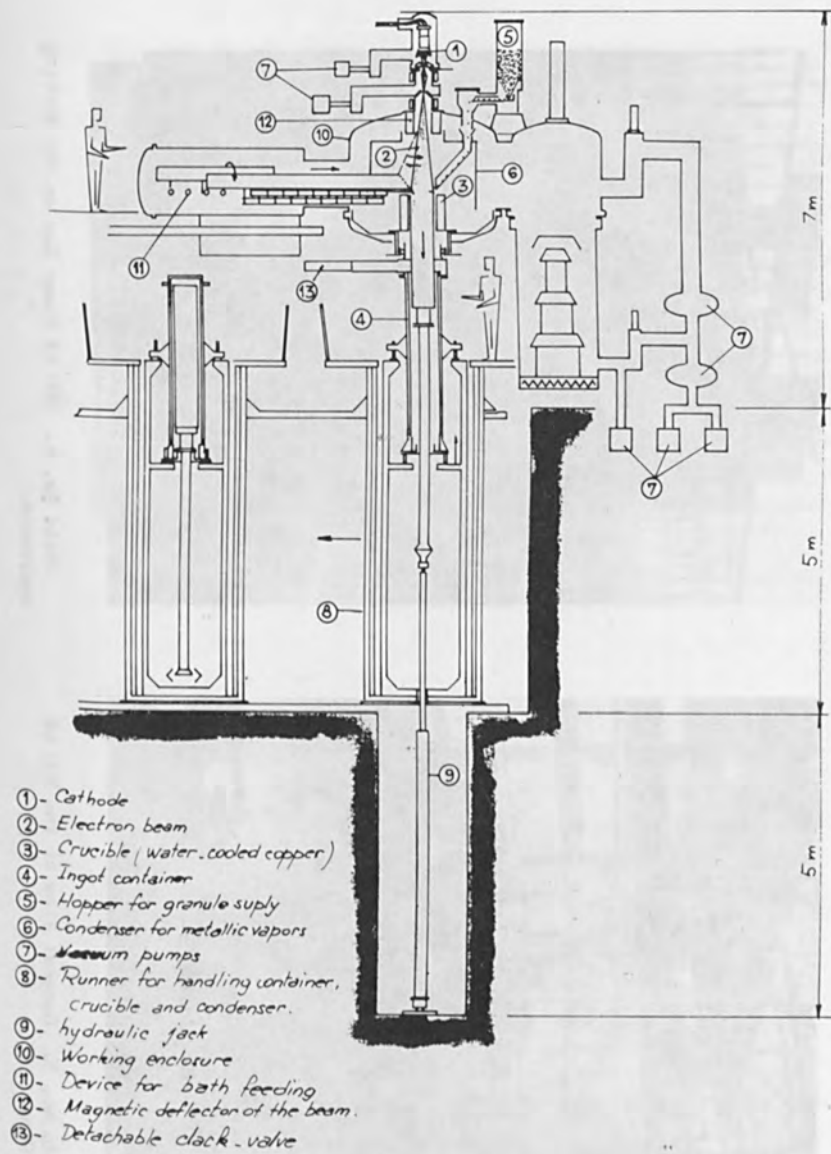


Fig. 7. 500 Power Gun

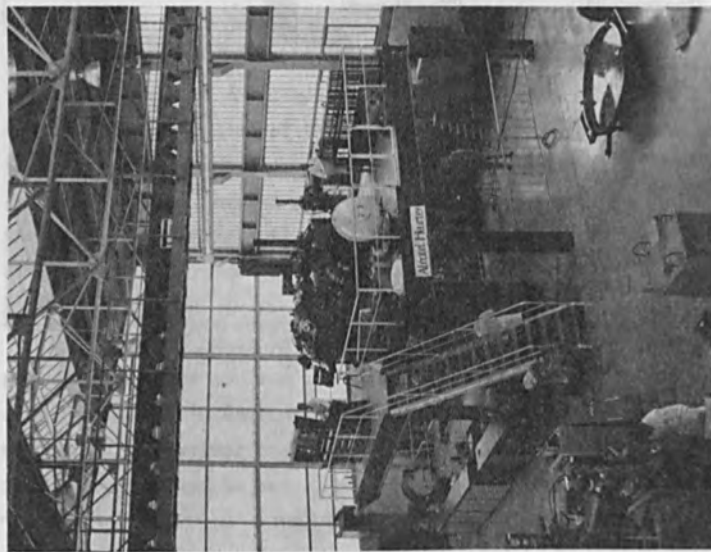


Photo No. 5. General View of the 500 kW Power Gun.

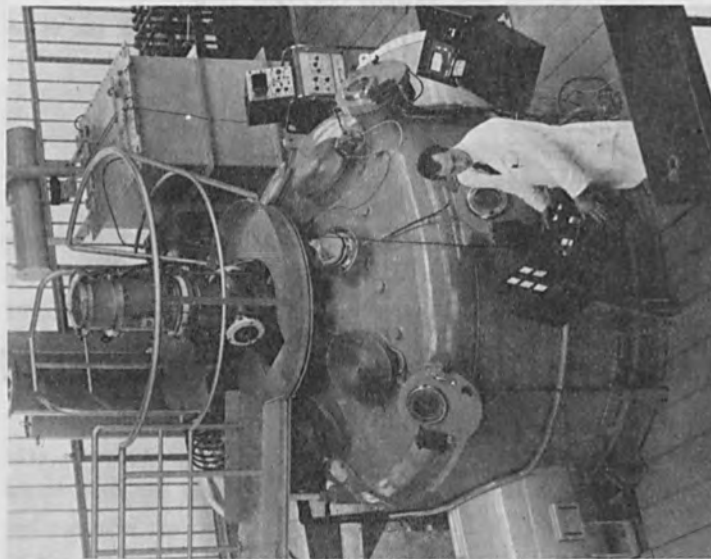


Photo No. 6. 500 kW Power Gun on the Working enclosure.

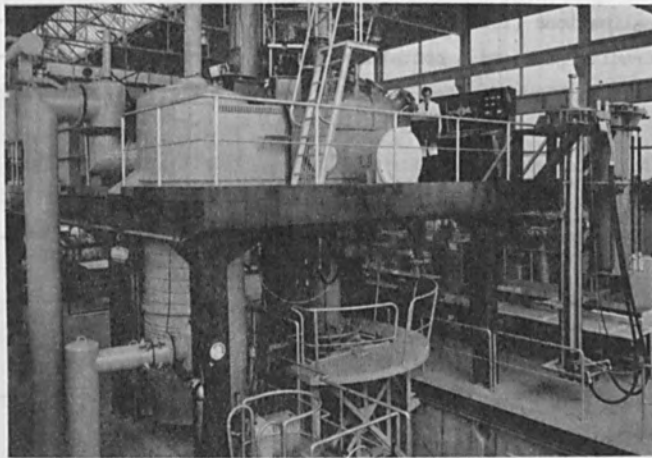


Photo No. 7. Partial View of the 500 kW Power Gun

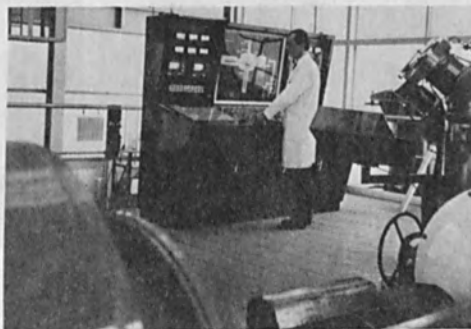


Photo No. 8. Commanding Board

distinguish the container, the lift and the runner (center) and some pumping canalizations (left). At the right hand side one sees a disassembled hydraulic jack and a container. Photo 8 represents the commanding board with a visualization of all orders given to the power gun.

The metal feeding device (11) can provide 4 ingots of 2 m length and 300 mm diameter, which allows the moulding of purified ingots of 2.2 m length and a diameter depending upon the treated metal.

METAL	max. diameter of ingot (mm)	Weight of ingot in (kg)
Ta, W	125	12 400
Mo, Nb	250	12 1.000
Ti, Zr	350	12 1.500
Fe	500	12 3.500

The most interesting technical characteristics are the following

- main power 850 kVA
- main voltage 390 Vt triphase
- max. power of the gun 610 kW under 40 kV
- pumping unit of the working enclosure
 - Diffusion pumps 95.000 lit/sec.
 - Root's pumps 1.000 m³/h + 8000 m³/h
 - mechanical pumps 2 x 180 m³/h
- pumping unit of the gun
 - diffusion pumps 1.200 lit/sec. + 2.500 lit/sec.
 - mechanical pump 25 m³/h



Photo No. 9. Refined Titanium Ingot ($\varnothing = 250$ mm, $l = 2.500$ mm) in a 385 kW Power Gun.

- water-cooling 50 m³/h
- The cathode, made up of W, is heated by an electron beam of about 4,5 kW.

In first meltings it was possible to obtain an ingot of Ti of 2.50 m length and 250 mm diameter (photo 9). The specific consumption is about 2 kWh/kg of treated titanium. This consumption will probably decrease when the electrode configuration (B) will be put into service.

CONCLUSIONS

The characteristics of electron beams can be evaluated accurately in the electrostatic part of a power gun by analyzing numerically the equations defining the behavior of the space-charge flow in the gun. This estimation is quite rapid, not expensive and permits us to choose the electrode configuration capable of furnishing the desired beam characteristics.

The computer program has been used to define the electrode configuration of a 500 kW power gun. Several variants have been analyzed and two types of gun have been described : That of classical Pierce type with the possibility of applying a bias potential at the focusing electrode in order to regulate the power of the gun and those, of a new conception, possessing two electrodes ; one serving to current extraction and the other to electron acceleration. In the two types one can vary the power of the gun without acting on the accelerating potential.

It is possible to estimate the diameter of the beam behind the anode by using the electron-optical characteristics of magnetic lenses established for paraxial rays (Gaussian approximation). In the vicinity of the target, positive ions concentrate the beam and diminish the theoretical focal distance of the lens. A good agreement with experimental

results was obtained when the theoretical excitations of the lens are decreased by about 15 - 20 %.

Finally two power guns of 200 kW and 500 kW have been described. The agreement between the theoretical analysis and many experimental results permits us to rely on the computer program for the estimation of the actual behavior of power guns. From now on one can envisage the design of power guns superior to 1.000 kW. This will probably bring about a sufficient decrease of the cost price of treated metal, which will enable it to be used in the refinement of ordinary metals such as steel. As a matter of fact the refinement of steel improves considerably its fatigue behavior which may be advantageous in many applications.

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IONIZATION PHENOMENA
INSIDE a DUOPLASMATRON EXPANSION CUP

L. BEX - P. BERNARD - C.E.N. Saclay

INTRODUCTION

The Saturne Synchrotron of the Saclay Atomic Research Center is normally utilized for accelerating protons to an energy of 3 BeV or deuterons to an energy of 2.35 BeV.

In order to enlarge the Physics possibilities with this accelerator we intend to accelerate polarized ions (protons and deuterons).

For this purpose, the first task to undertake is the design of a polarized protons or deuterons source as intense as possible.

It is on this problem that we shall report here.

Taking advantage of the fact that the source we are trying to design will be pulsed, it is possible, starting from an existing ion source, to raise the current from a few μA to a few tens μA , by using the expansion cup of a duoplasmatron as an ionizer. (1)

We shall therefore analyse the ionization phenomena that occur in the expansion cup ; then describe the general proposed set up to realize a high ionization efficiency, the injected atoms polarization being conserved. Finally we shall give our first experimental results.

I - CONSTITUTION of a POLARIZED PARTICLES SOURCE (Fig. n° 1)

Polarized deuterons or protons beams are usually obtained from the flight ionization of a polarized atoms stream which is produced in the following manner : the atoms created in a discharge tube called dissociator drift through

a blast pipe and a limiting edge diaphragm, at the exit of the opening the atom beam has a small diverging angle. The unselected atoms are eliminated by the pumping system. (2)

The emerging beam enters a sextupole magnet that focuses atoms in the $m_J = +\frac{1}{2}$ state and defocuses the atoms in the $m_J = -\frac{1}{2}$ state for hydrogen (fig. n° 4).

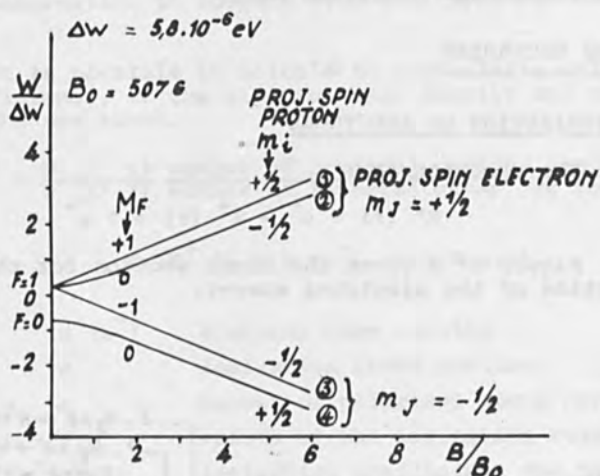


Fig. 4 - Hyperfine structure of the hydrogen atom

After their passage through the sextupole magnet the atoms are either in the {1} or {2} states, for which the spin projections values are $m_i = +\frac{1}{2}$ or $m_i = -\frac{1}{2}$.

The purpose of the transitions is to interchange populations in the different states in order to increase the protons polarization rate. For example states {2} and {4} may be interchanged, and after transitions states {1} and {4} only remain with an equal population, in that case the beam atoms are all in the $m_i = +\frac{1}{2}$ state.

Atoms are afterwards injected into the ionizer which is maintained at the acceleration potential. One can see on

fig. n° 1 an assembly of a classic polarized atoms production together with our new ionizer project.

Dr. BEURTEY's polarized atoms source may presently provide an atomic beam of 5.10^{16} atoms per second focused in a 1 cm^2 area. A complete ionization of such a beam would give a polarized deuterons or protons D.C. beam of 8 mA. In fact, up to this date, the ionization efficiency, with conventional ionizers, would limit the polarized beam to a few μA . Our purpose is to study a new type of ionizer in order to increase the intensity of the polarized beam.

II - IONIZATION MECHANISM

II.1. - IONIZATION by ELECTRONS

The reaction is :

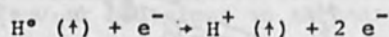


Figure n° 5 gives the cross section for the reaction as a function of the electrons energy.

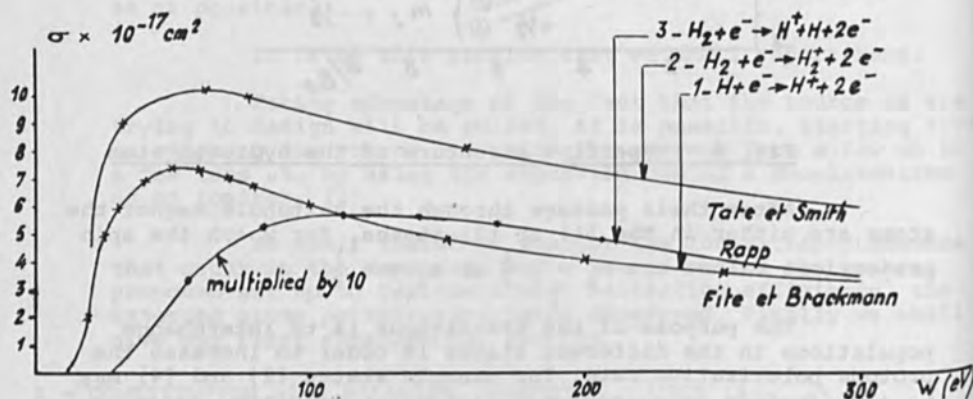
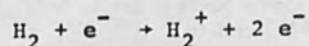
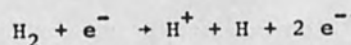


Fig. 5 - Cross section for the ionization reaction hydrogen by electrons (3,4,5)

The cross section is maximum for an electrons energy of 60 eV. Figure n° 5 shows the cross sections for two other possible ionization reactions of the hydrogen molecule.



and



which are interesting to compare with the first one (see section IV.2.2.).

It is possible to calculate an approximate value ϵ of an ionizer efficiency, if the electron beam density and the reaction cross section are known.

$$\epsilon = \frac{N(\text{H}^+)}{N(\text{H}^0)} \frac{\text{number of produced protons per second}}{\text{number of injected atoms per second}}$$

$$N(\text{H}^+) = n(\text{e}^-) \cdot \sigma \cdot n \cdot V \cdot \eta(B)$$

$n(\text{e}^-)$	electron beam density
σ	ionization cross section
n	number of polarized atoms per cm^3
V	volume of the ionization region
$\eta(B)$	ionization coefficient due to the magnetic field which increases the electrons trajectory ($\eta(B) > 1$)

$$N(\text{H}^0) = nv$$

v speed of the polarized atoms

$$\epsilon = \frac{n(\text{e}^-) \cdot \sigma \cdot V \cdot \eta(B)}{v} \quad |1|$$

In order to increase the ionization coefficient, the electrons intensity should be large and their energy should be such as to have a maximum cross section. The electron intensity will be space charge limited for a given difference of potential.

II.2. - IONIZATION by CHARGE EXCHANGE

There exists also the possibility of making a charge exchange reaction between the positive ions of an ionizing gas and the polarized atoms to be ionized.

If helium is used as an ionizing gas, the following reaction has to be considered :



The ionization potential for the hydrogen atom is smaller than for helium :

$$E_1 (\text{He}) = 24.6 \text{ eV}$$

$$E_1 (\text{H}) = 13.5 \text{ eV}$$

In this case the cross section of the reaction is maximum at small or zero energy of the He^{+} ions. The maximum for the cross section of the reaction is $\sigma = 7.5 \cdot 10^{-17} \text{ cm}^2$ (6) (fig. n° 6). It is the same as for ionization by electrons. The use of deuterium as ionizing gas could give good results for the ionization of hydrogen as the ionization potentials are the same and as there is a resonance in the charge exchange reaction. The maximum cross section for the reaction



is obtained for a small kinetic energy of the ionizing ions, and is of the order of $4 \cdot 10^{-15} \text{ cm}^2$ (fig. n° 6) (15) i.e. 50 times larger than the one for ionization by electrons. The ionizer we are proposing is able to realise this type of reaction.

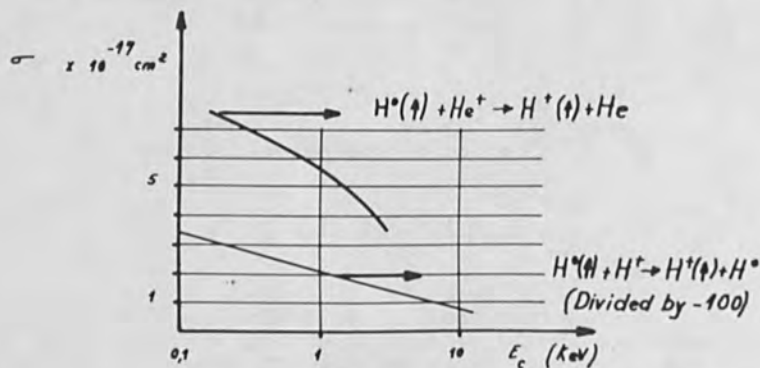


Fig. 6 - Cross section for ionization by charge exchange

III - CLASSICAL ELECTRONS IONIZERS

Apart from Dr. BEURTEY's proposition given at the Strasbourg Conference in 1968, all ionizers in operation until now to produce polarized ions are using an electron beam for ionization. They are made of an electron emitting cathode with an accelerating grid. (8) (9). The polarized atoms are injected through the electrons region where a strong magnetic field is maintained to preserve the ions polarization during the ionization reaction. The electron current is limited by space charge and does not get beyond 0.2 A.cm^{-2} with 200 eV extraction potential. It is useless to increase this voltage, as the maximum cross section for ionization of hydrogen is obtained around 60 eV as seen above.

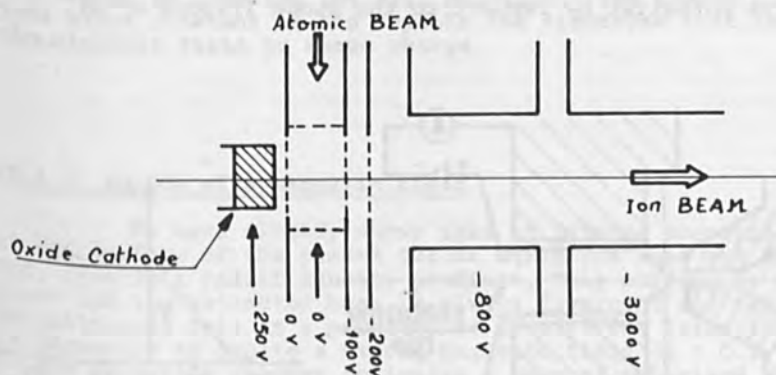


Fig. 7 - Classical electrons ionizers

Application of formula |1| to the scheme of figure n° 7 gives, for the ionization coefficient at an energy of 200 eV, with an electron current of 0.2 A.cm^{-2} for a volume of 1 cm^3 and an atomic speed of 2.10^5 cm.s^{-1} :

$$\epsilon = 6.4.10^{-4} \eta(B)$$

We intend to show that by using the plasma of a Von Ardenne source it is possible to increase this rate of ionization.

IV - NEW IONIZER

IV.1. PHENOMENA in the EXPANSION CHAMBER

The plasma jet from the hole in the anode is made of electrons and positive ions. Both types of reaction considered above can occur : ionization by electronic collision or by charge exchange.

As GAUTHERIN from Orsay (10), we have found (11) that there is a large electron density in the expansion chamber. This gives rise to the possibility of a very high ionization efficiency. To get a measure of the electron current that can be obtained in this chamber, we have insulated its wall (12) and measured the current it collected as a function of its potential by respect to the anode (figure n° 8).

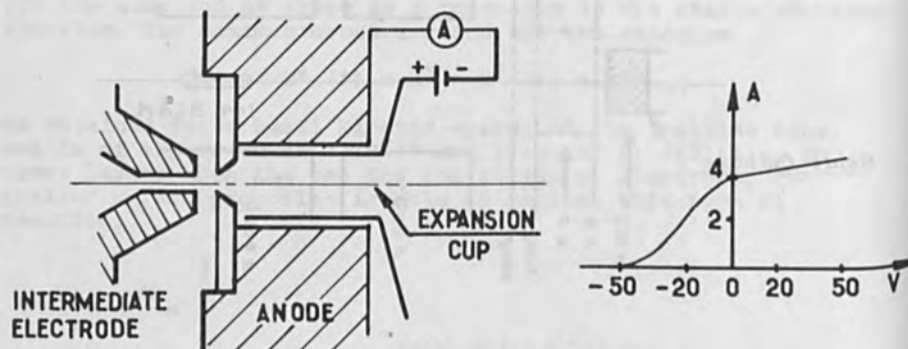


Fig. 8 : Electron current

This measure was carried out with hydrogen and an arc intensity of 30 A.

The electron current is no more limited by space charge and offers large possibilities for ionization, as shown below.

IV.1.1. Action of an electric field

If the wall of the chamber is biased to the same potential as the intermediate electrode, the electrode current vanishes. This shows that the maximum energy of the electrons is given by the difference of potential between the anode and the intermediate electrode. When the bias is at the anode potential the electron stream is nearly maximum. A positive bias will increase the energy of the electrons and it is possible to choose the one corresponding to the maximum cross section for ionization. Under this condition, but when this occurs, a degradation of the beam emittance occurs, as it will be shown later.

The ions seem to come from a region where the potential is larger than the one of the anode (potential mountain) (13). This region sits between the anode and the intermediate electrode. By biasing the wall negatively the energy of the ions stays constant as one repels the electrons that cancel the electric field by space charge.

IV.1.2. Action of a magnetic field

We have already shown that an intense magnetic field at the surface of the plasma out of which the ions are extracted, creates a radial density gradient. This surface is no more plane and the extracted beam is always divergent and has a large emittance. This is a problem, as we will see later that it is necessary to create a strong magnetic field ($B > 0.1$ Tesla) in the expansion chamber to ionize a beam of polarized particles while preserving its polarization.

However, if the density is not uniform, focalisation will be hard to attain. In particular, the so called PIERCE extraction (14) will not give the optimum result.

On the other hand, the magnetic field will increase the pathlength of the electrons, and so doing the ionization efficiency. It will also prevent particle loss on the wall of the chamber. It is interesting to know the Larmor radii of the electrons and the positive particles in the expansion chamber as a function of their energy in a magnetic field of $B = 0.1$ T.

E_{eV}	$R(e^-) \text{ (cm)}$	$R(H^+) \text{ (cm)}$	$R(He^+) \text{ (cm)}$
0,01	$3,37 \cdot 10^{-4}$	$1,44 \cdot 10^{-2}$	$2,88 \cdot 10^{-2}$
0,1	$1,06 \cdot 10^{-3}$	$4,56 \cdot 10^{-2}$	$9,13 \cdot 10^{-2}$
1	$3,37 \cdot 10^{-3}$	$1,44 \cdot 10^{-1}$	$2,88 \cdot 10^{-1}$
10	$1,06 \cdot 10^{-2}$	$4,56 \cdot 10^{-1}$	$9,13 \cdot 10^{-1}$
100	$3,37 \cdot 10^{-2}$	1,44	2,88

Up to 10 eV the radii for the three kind of particles are smaller than the diameter of the chamber. There will be little loss on the walls, if one assumes that the energy gained by the polarized protons in the expansion chamber is smaller than 10 eV.

If the axial magnetic field is slightly decreasing toward the extraction electrode, an ionized particle in the expansion chamber will only be able to move into the region of weak magnetic field, i.e. toward the extraction electrode. This while assuming that there is no accelerating electric field. It can indeed be demonstrated that :

$$\frac{dw_{//}}{dt} = - \frac{1}{2} \frac{1}{B} \frac{\partial B}{\partial z} w_{//}^2$$

$$\frac{d(w_{\perp}^2)}{dt} = \frac{1}{B} \frac{\partial B}{\partial z} w_{//} w_{\perp}^2$$

w : particle speed vector

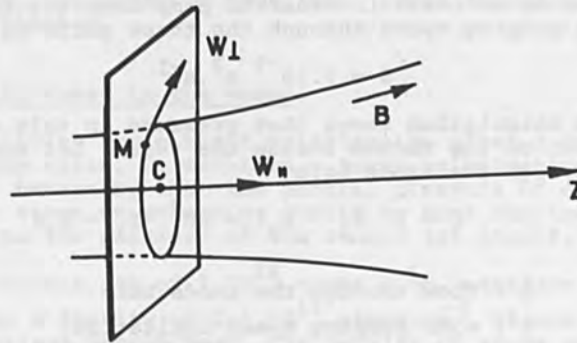


Fig. 9 : charged particle in a decreasing magnetic field

When the particle is ionized, the velocity component parallel to the z axis is zero. It can only increase for decreasing fields. We have thus imagined an ionizer making use of those possibilities of ionization while keeping an eye on the contingency of magnetic field and low pressure to keep the particles polarization and the beam emittance within acceptable values.

IV.2. DESCRIPTION and TECHNOLOGY of the IONIZER

The ionizer is made of a classical duoplasmatron source and of an expansion chamber for the plasma (fig. 2). Its different elements will have to be studied with due consideration to polarization conservation and ionization efficiency.

IV.2.1. Diameter of the anode hole

This diameter is imposed by considerations of pressure and electron flow in the chamber. If it is too large the gas flow is big and the pressure inside the plasma expansion cup becomes too large. The mean free path of the particles gets smaller and risk exists that depolarisation could occur. Also, sparks tend to appear between the electrodes of the cup.

If the hole is too small, the electron flow decreases and ionization is less efficient. We have used as many

openings as we could in order to pump down the ionization zone and the pumping speed through the three ports is about

$$S = 2.10^{-2} \text{ m}^3 \cdot \text{s}^{-1}$$

A quick calculation shows that pressure in this zone is hardly dependent on the vacuum inside the vessel but mostly on the flow through the anode hole.

$$Q = S (P_2 - P_3) = KD^2 (P_1 - P_2)$$

Q : flow through the anode hole

S : zone pumping speed ionization

P_1 : pressure inside the source

P_2 : pressure inside the ionization zone

P_3 : pressure inside extraction zone

D : diameter of the anode hole

$$P_2 = \frac{SP_3 + KD^2 P_1}{S + KD^2}$$

Knowing the orders of magnitude of the different parameters, it is easy to show that P_2 varies faster with D than with P_3 .

Let us take as an example an ionizer (hole diameter 0.8 mm), working with helium with a flow

$$Q = 1.13 \cdot 10^{-3} \text{ p.m}^3 \cdot \text{s}^{-1} \text{ (}^1\text{)}$$

and a pressure inside the extraction zone

$$P = 6 \cdot 10^{-3} \text{ p (}^2\text{)}$$

One finds for the pressure inside the ionization zone

$$P_2 = 6.3 \cdot 10^{-2} \text{ p}$$

(¹) $1 \text{ p.m}^3 \cdot \text{s}^{-1} = 7.5 \text{ torr} \cdot \text{l} \cdot \text{s}^{-1}$

(²) $1 \text{ pascal} = 7.5 \cdot 10^{-3} \text{ torr}$

At this pressure, the mean free path for helium atoms is 40 cm, one order of magnitude larger than the dimensions of the chamber.

IV.2.2. Pumping speed in the vessel

Particles lose their polarization after a collision against the cup walls. To maintain a large polarization efficiency, it is necessary that the partial pressure of depolarized hydrogen atoms or molecules should be kept negligible with respect to the pressure of the atomic jet itself.

An atomic jet of $5 \cdot 10^{16}$ atoms. s^{-1} on a surface of 1 cm^2 corresponds to a density of $2.5 \cdot 10^{11}$ atoms. cm^{-3} . If one wants to get 90 % polarized proton beam, the density of atoms or molecules in the vessel has to be ten times smaller than the one of the atomic jet. It corresponds to a partial pressure of hydrogen of $P = 10^{-4}$ p. Knowing the flow of hydrogen and its acceptable pressure in the vessel it will be possible to predict the size of the pumping group.

The presence of molecular hydrogen is not very bothering because the ionization of hydrogen gives mostly H_2^+ and few H^+ (see figure 5).

IV.2.3. Magnetic field in the chamber

The conservation of the polarization of the particles during ionization imposes us an axial magnetic field of 0.1 Tesla in the expansion cup. Furthermore this magnetic field should decrease slowly enough in the extraction zone to preserve the polarization. The three coils B_1 , B_2 , B_3 create this required axial magnetic field (fig. 3).² But, unfortunately, according to what has been said in IV.1.2., the presence of the magnetic field on plasma surface causes an increase of emittance.

Those coils are completely inside the vacuum, and to avoid out gassing, are made out of a copper spool wound with aluminium ribbon covered with alumina.

IV.2.4. Electric field in the expansion cup

The wall of the expansion cup is made of two cylindrical electrodes for electrical polarization, P_1 and P_2 . Those metallic electrodes are insulated from each other and from the anode, by a deposit of alumina on the external surface.

To prevent any spurious outgassing of hydrogen and to insure good surface conductivity, it is recommended to

have them gold coated. The jet of polarized atoms enters the cup perpendicularly to its axis, through a 8 x 10 mm rectangular opening in P_1 . The unionized particles get out through a square opening of 10 x 10 mm, and are pumped. The electrode P_1 has been provided to allow the energy of the electrons to be changed. The electrode P_2 is used to stabilise the surface of the plasma out of which the ions are extracted, or, in other words, to act upon the emittance of the beam.

Here is a table of the electron currents collected by the various electrodes.

Magnetic field	Electrodes	Electrodes voltage by respect to the anode	Electrons current
$B = 0.1 \text{ T}$	Extractor	+ 100 V	1.3 A
	P_1	(+ 70 V	0.6 A
		(+ 150 V	1 A
	P_2	- 30 V	0.1 A
$B = 0$	Extractor	+ 100 V	0.3 A
	P_1	+ 70 V	2.4 A
	P_2	- 30 V	0.1 A
$B = 0.1 \text{ T}$	Extractor	- 20 kV	0 A
	P_1	(+ 70 V	1 A
		(+ 150 V	1.2 A
	P_2	- 30 V	0.1 A
$B = 0$	Extractor	- 20 kV	0 A
	P_1	(+ 70 V	2.4 A
		(+ 150 V	2.5 A
	P_2	- 30 V	0.1 A

From this table, it can be deduced that the total current of electrons getting out of the anode hole is not space charge limited and reaches values close to 3 A. When the magnetic field is off, the electrode P_1 gets most of this current. When the magnetic field is turned on, the electron current collected by P_1 goes down to 1 A, as the electrons are confined to the center of the expansion cup. In that case, it is likely that the missing electrons oscillate in the ionization zone and are finally captured at the end of the chamber on the anode.

Knowing the electron current it is possible to make an estimate of the ionization coefficient and to establish a comparison with the classical ionizers.

IV.3. CAPABILITIES of the IONIZERS

In paragraph 3, we made a calculation of the ionization coefficient for a classical ionizer using electrons.

The ionizer we are proposing allows an increase of a factor of 10 at least for the current of electrons ; the ionization efficiency is multiplied by the same amount ; this ionizer allows the charge exchange reaction as well. The formula established on paragraph II.1. for the calculation of the ionization efficiency is valid for the latter reaction, and reads :

$$\epsilon = \frac{N (I^+) \cdot \sigma' \cdot V \cdot \eta' (B)}{v}$$

Let us take as an example two sources, one using helium and the other one deuterium. The first one will deliver a He^+ ions current of 70 mA, or $1.1 \cdot 10^{18}$ ions.s⁻¹.

$$\epsilon = 4 \cdot 10^{-4} \eta' (B)$$

This value is comparable with the one obtained by classical ionizers if one supposes $\eta' (B) = \eta (B)$.

The second one can provide us with a current of D^+ of 100 mA, or $1.6 \cdot 10^{18}$ ions.s⁻¹.

$$\epsilon = 3.2 \cdot 10^{-2} \eta' (B)$$

This value appears to be bigger than the one obtained by ionization by electrons but let us notice that

η (B) is unknown and probably bigger for electrons than for ions.

V - FIRST EXPERIMENTAL RESULTS

We give here the nominal parameters of the duoplasmatron ion source working with helium.

V.1. NOMINAL PARAMETERS

Pulse length	600 μ s
Repetition rate	0.5 Hertz
Helium flow	$1.13 \cdot 10^{-3}$ p.m ³ .s ⁻¹ or 40 atm.cm ³ .h ⁻¹
$V_{\text{cathode}} - V_{\text{anode}}$	- 200 V
$V_{\text{intermediate electrode}}$	((- 100 V
- V_{anode}	(
Arc current	35 A
Cathode power supply	($V = 2.8$ V ($I = 30$ A
Anode aperture	0.8 mm
Extraction voltage	20 kV
Helium beam current (He^+)	70 mA

V.2. BEAM EMITTANCE

The emittance of the beam is estimated by measuring the angular divergence of a pencil beam issued from a 0.2 mm diameter hole set in the extraction electrode. The following table shows the measured angle versus magnetic field and bias voltage on P_1 and P_2 electrodes.

B (Tesla)	P ₁ (Volt)	P ₂ (Volt)	α (radian)
0	0	0	$> 10^{-1}$
0.1 T	0	0	$9.2 \cdot 10^{-2}$
0	+ 70	- 30	$> 10^{-1}$
0.1 T	+ 70	- 30	$7.2 \cdot 10^{-2}$
0.1 T	- 90	- 110	$7.2 \cdot 10^{-2}$
0	- 90	- 110	$4 \cdot 10^{-2}$

Measurements are made at 20 kV extraction voltage.

A large molecular hydrogen flow has been injected (large with respect to the weak polarized atomic stream not used for this measurement) and biased voltages on P₁ and P₂ have been adjusted in order to get the largest ionization efficiency together with acceptable emittance value and 0.1 Tesla magnetic field.

The results are :

$$V_{P_1} = + 70 \text{ V}$$

$$V_{P_2} = - 30 \text{ V}$$

$$\alpha = 7.2 \cdot 10^{-2} \text{ rd}$$

V.3. RESIDUAL HYDROGEN GAS

Without any hydrogen gas feeding, the mass analysis of the beam shows that we have protons in addition of the ionized helium beam (He⁺). This proton current has been decreased by using purified helium gas. The approximate rate of the intensities is 500 μ A/protons with 50 mA He⁺ ion beam.

When introducing a polarized atom beam of $5 \cdot 10^{15}$ atoms per second in the expansion cup we have observed an instantaneous burst of the hydrogen pick of a few per cent.

But we can also see a slow increase of the hydrogen pick of 20 % due to an accumulation of hydrogen in the expansion cup. This problem will be solved by increasing the pumping speed through the extractor, and by pulsing the polarized atomic beam.

VI - CONCLUSION

Calculation of the ionization efficiency makes us confident that it will be possible to produce a pulsed polarized ion beam of a few tens μ A current.

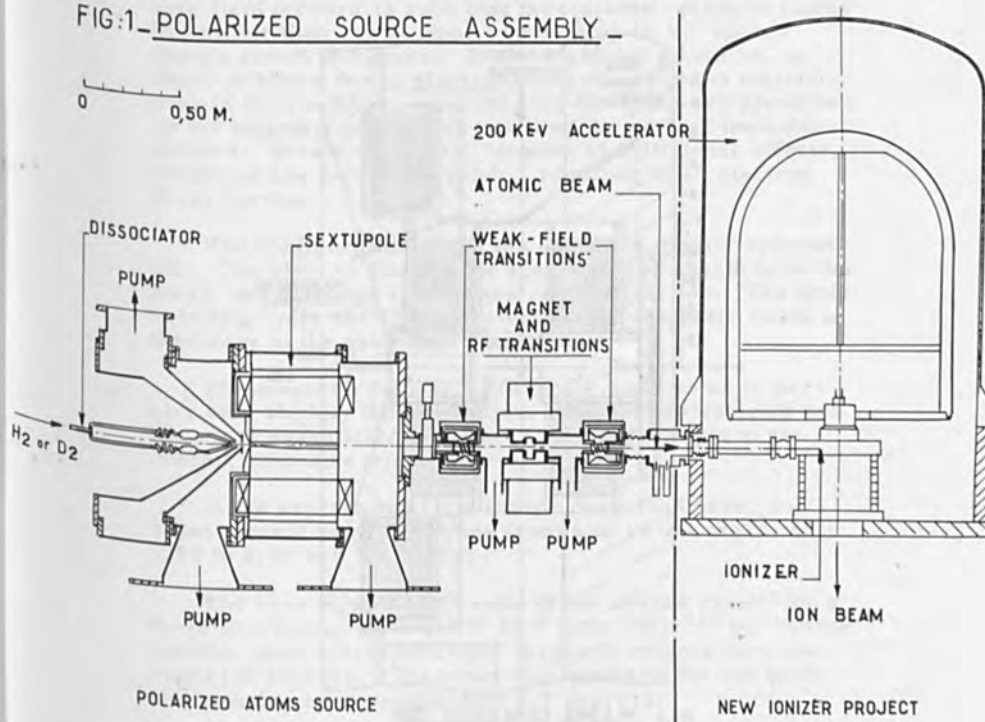
Experimental developments have however shown that attention had to be given to the elimination of H^+ impurities.

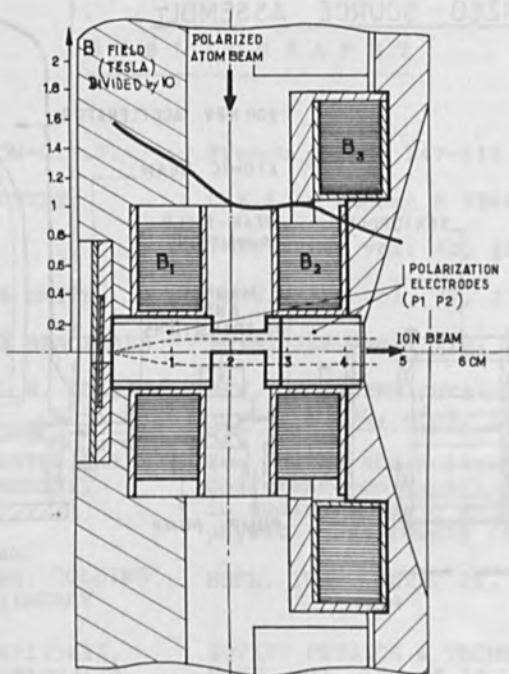
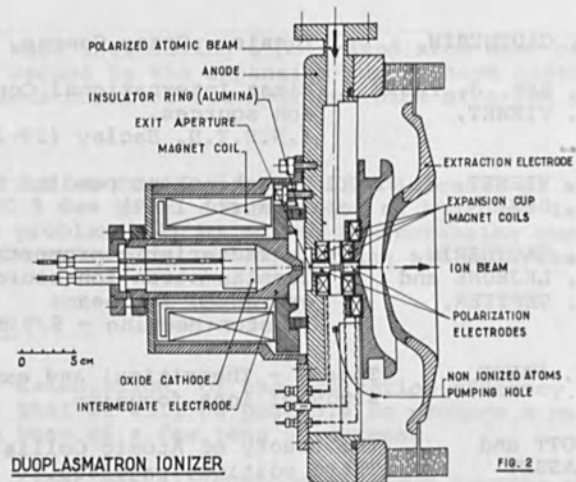
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FIG:1 - POLARIZED SOURCE ASSEMBLY -





A CESIUM ION SOURCE WITH LARGE PERVEANCE AND HIGH DEGREE OF COLLIMATION

Rudolf X. Meyer
Plasma Research Laboratory
The Aerospace Corporation
El Segundo, California 90045

Abstract

A cesium ion source is described, in which cesium ions--generated by contact ionization on a hot tungsten surface--are electrostatically accelerated in a space-charge sheath formed on this surface. The space charge is mainly negative, and is maintained by means of electrons emitted from a thoriated tungsten filament (cathode) which is located adjacent to the contact ionization surface. A magnetic field is maintained between two shaped pole pieces that are designed in such a manner as to result in a field which is exactly parallel to the contact ionization surface. The magnetic field strength is such that the electron cyclotron radius is smaller than, or at most comparable to the space-charge sheath thickness. In the idealized condition, in which diffusion due to electron/ion collisions and instabilities in the sheath is neglected, the electrons are prevented by the magnetic field from reaching the contact ionization surface. In fact, however, because of collisional effects, electrons are lost to the anode, resulting in an electron drain current.

The configuration of the ion source is axially symmetric. The electric field in the space-charge sheath is in the axial, the magnetic field in the radial direction. The drift resulting from the crossed electric and magnetic fields is therefore in the azimuthal direction.

The magnetic field is sufficiently weak so as to perturb only slightly the ion motion. The ion trajectories are therefore essentially rectilinear, at a right angle to the contact ionization surface.

In the experimental ion source described here, ion beam current densities range from 4 to 14 mA cm⁻², at 1.50 to 2.00 keV ion energy.

The very large aspect ratio of the source results in a large perveance, up to 2.2×10^{-6} Amp Volt^{-3/2} per source module, with correspondingly larger perveance for clustered ion sources. The mean divergence of the ion beam was found to be approximately 1.5 degrees.

In this paper, in addition to briefly summarizing experimental results obtained in testing this new ion source, an analysis of the collisionless electron and ion motions in the space-charge sheath region is developed.

The computed charged particle trajectories are those resulting from the combined action of the magnetic field (which can be assumed to be constant in the space-charge sheath region) and the electric field. The latter is computed self-consistently from the charged particle motions and the resulting net space-charge distribution. The analysis resembles the calculations which have been carried out for electron trajectories in magnetrons and crossed-field devices, but in addition takes into account the self-consistent motion of the positive ions.

Two different models for the injection of the electrons into the space-charge sheath are considered. These two models, referred to as the quasi-cycloidal and the Brioulin case, respectively, correspond to two opposite limits of the ratio of the filament distance to the space-charge sheath thickness. Here also, the considerations are similar to those relating to microwave tube devices.

The distribution of the electrostatic potential in the sheath is shown to be nearly independent of the particular assumption made concerning the electron injection into the sheath.

Based on this analysis, ion and electron current densities, particle trajectories, and electric potential distribution in the space-charge sheath of this ion source are computed.

Introduction

The characteristic feature of the new ion source described in this paper is the acceleration of the ions in a space-charge sheath. This sheath, which contains a net negative space charge, in effect serves as the accelerating electrode and replaces the ion-optical structures used in more conventional sources. Since such structures are often subject to erosion from ion sputtering, a potentially important advantage is gained if they can be eliminated.

A schematic representation of the space-charge-sheath ion source is shown in Fig. 1. Cesium vapor is admitted to the ionizer assembly (I) which consists of an annular manifold for distributing the vapor and of a porous tungsten plug. The cesium diffuses through the tungsten and is ionized at the surface (Langmuir-Kingdon effect).

The ionizer assembly is contained between two magnetic pole pieces (P_1 and P_2) which are shaped in such a manner as to produce a magnetic field (H) exactly parallel to the ionizer surface. A heated filament (F) is located in the magnetic gap, and is at a negative potential with respect to the ionizer. In the absence of collisions, the electrons that are emitted by the filament are free to move along magnetic field lines and thereby impart the filament potential to the magnetic field surface intersecting the filament. The resulting electric field is perpendicular to the ionizer surface and accelerates the ions in the small gap between the ionizer and the magnetic field surface.

The arrangement of the ionizer, pole pieces, and filament is annular with axis of symmetry (A-A) parallel to the ion-beam (B), which has, therefore, a hollow core. As a result of mutually perpendicular electric and magnetic fields which are in the axial and radial directions, respectively, the electrons drift in the azimuthal direction as schematically indicated in Fig. 1b. An additional electron source is needed to compensate the positive space charge in the beam; in the experimental arrangement, this source is conveniently provided by secondary electrons resulting from the impingement of the ion beam on the beam collector.

The geometrical configuration of the space-charge-sheath ion source is quite similar to the one of the Hall-current accelerators investigated for applications in electric propulsion of space vehicles (Ref. 1). However, the accelerating region in Hall-current accelerators extends over many Debye lengths, with the result that the acceleration of the ions takes place in a space-charge neutral plasma, whereas in the present ion source the acceleration is confined to the space-charge-sheath region. In Hall-current devices, the plasma is unavoidably in contact with the insulators which confine the plasma, resulting in wall losses by momentum and energy transfer, whereas in the space-charge-sheath ion source, the plasma is only in contact with the ionizer itself.

Apparatus and Experimental Data

In this section, we give a brief summary of the principal laboratory measurements carried out with a prototype of this ion source. Additional data are contained in Ref. 2.

The experimental apparatus used is shown in Fig. 4. In operation, the ion beam extended upwards from the ionizer and was intercepted by a liquid nitrogen cooled collector plate shown near the top of the vacuum vessel. Provisions were made to allow changing during test runs, without breaking the vacuum, the axial distance between ionizer and filament, and the adjustment of the magnetic pole pieces.

The ionizer, which was resistance heated, consisted of a porous tungsten ring with a 1.516 inch inner diameter, an outer diameter

of 2.290 inch, and a nominal emitting area of 13.8 cm^2 . Great care was taken to maintain the emitting surface as nearly planar as possible, and to align it accurately with the magnetic lines of force.

The ion beam current was measured at the collector plate intercepting the beam. A high-transparency double grid was used to eliminate the effect of secondary electrons.

The measured ion beam current is plotted in Fig. 2 for typical test conditions^{*}), as a function of the cesium vapor pressure in the reservoir (at a magnetic field strength of 2060 Gauss in the center of the magnetic gap, accelerating potential of 1500 Volt, ionizer temperature of 1150°C , and an axial distance of 2.03 mm between ionizer surface and filament) and also as a function of the magnetic induction (at 21.8 mm Hg cesium vapor pressure; other data as stated above).

Conditions for a typical test are summarized in the Table below.

Accelerating potential, Volt	1500
Accelerating gap (ionizer/filament axial separation), mm	2.03
Ionizer temperature, $^\circ\text{C}$	1150
Filament temperature, $^\circ\text{C}$	1835
Magnetic field strength in center of gap, Gauss	2060
Reservoir cesium vapor pressure, mm Hg	21.8
Ion current, mA	82-113 ^{**})
Electron drain current to ionizer, mA	50-55 ^{**})
Beam divergence (mean half-angle), degrees	1.5

The maximum ion current obtained in a series of tests was 195 mA, at 2000 Volt accelerating potential, corresponding to a permeance of $2.18 \times 10^{-6} \text{ Amp Volt}^{-3/2}$.

Analysis of Ion and Electron Motions[†])

(a) Basic Equations

We give here a rather detailed account of the motions of ions and electrons and of the electric space charge field in the accelerating region, i.e., in the region between the ionizer and the virtual cathode.

^{*}) Because the surface condition of the tungsten ionizer varied somewhat between tests, the ion currents measured under otherwise identical conditions (2060 Gauss magnetic induction, and 21.8 mm Hg vapor pressure) did not entirely agree with each other.

^{**}) Depending on ionizer surface condition.

[†]) A brief account of some of the results of this section has been given previously in Ref. 3.

This region turns out to be a double space charge sheath, with a positive net charge density near the ionizer, and a negative net charge density near the virtual cathode. The ions and electrons move under the influence of the magnetic and electric field, the latter being determined in turn by the charge densities set up by the ions and electrons (self-consistent electric field calculation).

The following approximations are made in the analysis:

1. The magnetic field in the accelerating region is assumed to be constant. (In the experimental device, the applied magnetic field varies as a function of the radius, deviating from the mean by approximately ± 15 percent; this variation is neglected in the analysis; in the axial direction the variation is much smaller.) The magnetic fields induced by the ion current and the electron drift current can be neglected, the former being about 10^{-5} , the latter about 10^{-3} times the applied field.
2. Since the ion cyclotron radius (≈ 20 cm) is large compared with the length of the accelerating region (≈ 1 mm), the effect of the magnetic field on the ions can be neglected. The motion of the ions is approximately rectilinear, at a right angle to the ionizer surface.*)
3. The thermal energy of the ions (≈ 0.1 eV) emitted from the ionizer, and of the electrons (≈ 0.15 eV) emitted from the filament which forms the cathode, is negligible compared with the energy gained by the accelerating potential (500 to 2000 eV). Likewise, the kinetic energy associated with electron motion parallel to the magnetic field, which is of the order of 50 eV in the experiment, is negligible.
4. The collision frequency for effective 90° scattering due to cumulative small-angle Coulomb scattering is of the order of $5 \times 10^4 \text{ sec}^{-1}$, which is small compared both with the electron cyclotron frequency ($\approx 5 \times 10^9 \text{ sec}^{-1}$) or with the frequency associated with electron drift around the axis of symmetry of the device ($\approx 5 \times 10^6 \text{ sec}^{-1}$). Collisions are therefore neglected.
5. Trapped particles are neglected.

Taking the x-axis perpendicular to the ionizer surface, i.e., in the direction of the ion beam, the y-axis in the azimuthal direction, and the z-axis in the radial direction, we have

$$\frac{\partial}{\partial t} = \frac{\partial}{\partial y} = \frac{\partial}{\partial z} = 0 \quad (1)$$

* Since the ions can be assumed to be emitted with zero velocity from the ionizer, the trajectories have, however, a cusp at this point.

For a cold, collisionless electron gas,

$$(\vec{V}^{(j)} \cdot \nabla) \vec{V}^{(j)} = -\frac{e}{m_-} (\vec{E} + \vec{V}^{(j)} \times \vec{B}) \quad (2)$$

where $\vec{V}^{(j)}$ is the velocity of the j^{th} stream, and where the meaning of the other symbols is conventional. In general, it is necessary to consider a left- and a right-going electron stream (i.e., a stream in the direction of the negative and positive x-axis, respectively). Equation (2) holds separately for each stream.

The electric field $\vec{E}$ is in the x-direction, and the magnetic induction $\vec{B}$ in the z-direction. Designating the electron velocity components by $u^{(j)}$, $v^{(j)}$, $w^{(j)}$, we have $w^{(j)} = 0$, and, from Eq. (2),

$$u^{(j)} \frac{\partial u^{(j)}}{\partial x} = -\frac{e}{m_-} \left(-\frac{\partial \phi}{\partial x} + v^{(j)} B \right) \quad (3)$$

$$\frac{\partial v^{(j)}}{\partial x} = +\frac{e}{m_-} B \quad (4)$$

where $\phi = \phi(x)$ is the electrostatic potential. A simple interpretation for the last equation can be found by introducing the vector potential $\vec{A}$ for the magnetic field, and the canonical momentum $\vec{P}^{(j)}$, defined by (e.g., Ref. 4)

$$\vec{P}^{(j)} = m_- \vec{V}^{(j)} - e \vec{A}$$

Since $A = A_y = xB$, we find

$$\frac{\partial (\vec{P}^{(j)})_y}{\partial x} = m_- \frac{\partial v^{(j)}}{\partial x} - e B = 0$$

Thus, the y-component of the canonical momentum is seen to be a constant of motion.

If $n^{(j)}$ is the number density of the electrons for the j^{th} stream, the continuity equation for the electrons reduces to

$$\frac{\partial}{\partial x} (n^{(j)} u^{(j)}) = 0. \quad (5)$$

We note that Eqs. (3) to (5) are invariant with respect to a change in the sign of $u^{(j)}$. Due to this symmetry of the right- and left-going electron streams, it suffices to consider, for instance, only the right-going stream (with particle density n_-) with the proviso, of course,

that the total negative space charge density is $2n_-$ in those cases where two electron streams are present.

For the positive ions, since u_+ and ϕ vanish at $x = 0$,

$$\frac{1}{2} m_+ u_+^2 = -e\phi \quad (6)$$

$$\frac{\partial}{\partial x} (n_+ u_+) = 0. \quad (7)$$

The last equation to be satisfied is Poisson's equation,

$$\frac{\partial^2 \phi}{\partial x^2} = -\frac{e}{\epsilon_0} \left(n_+ - \sum_j n_-^{(j)} \right) \quad (8)$$

where the summation is extended over the several electron streams which may be present. Equations (3) to (8) determine completely the electron and ion motion and the electric field.

It is convenient to introduce the ratio $\delta = x_1/r_{-1}$ where x_1 is the gap between ionizer and virtual cathode, and where

$$r_{-1} = \frac{1}{\omega_-} \left(\frac{-e\phi_1}{\frac{1}{2} m_-} \right)^{1/2}$$

is the cyclotron radius for an electron of energy $-e\phi_1$. ($\omega_- = eB/m_-$ is the cyclotron frequency, and $\phi_1 = \phi(x = x_1)$ is the filament potential, which is also the potential at $x = x_1$.)

It will also be found convenient to introduce the dimensionless quantities

$$x' = \frac{x}{x_1}, \quad y' = \frac{y}{x_1}, \quad \phi' = -\frac{\phi}{\phi_1}, \quad j'_+ = \frac{j_+}{j_{+0}}, \quad j'_- = \frac{j_-}{j_{-0}} \quad (9)$$

where $j_+ = en_+ u_+$ and $j_- = en_- u_-$ are the ion and electron current densities^{*} respectively, and where

$$j_{+0} = \frac{4}{9} \epsilon_0 \left(\frac{2e}{m_+} \right)^{1/2} \frac{(-\phi_1)^{3/2}}{x_1^2} \quad (10)$$

^{*} j_- is taken positive and is the current density for the right-going electron stream.

is the Child-Langmuir current density for the ions (i.e., the current density which would apply for space-charge limited conditions, if no electrons were present). Similarly, j_{-0} is the corresponding expression for the electrons, and represents the current density in the absence of positive ions and without a magnetic field. We note that

$$j_{-0} = \left(\frac{m_+}{m_-} \right)^{1/2} j_{+0}$$

(b) Injection of Electrons into the Beam

In general, the trajectories of the electrons between cathode and beam are quite complex, due to the three-dimensional nature of the motion. However, in the two special instances referred to below as the quasi-cycloidal and the Brioullin case, respectively, relatively simple solutions can be obtained. The two cases correspond to opposite extremes: in the quasi-cycloidal case, the spacing between filament and beam is small compared with the distance x_1 ; conversely, in the Brioullin case, this spacing is large. The assumption of the so-called Brioullin planar flow is often made in computations relating to microwave tube devices (Ref. 5).

The distinction between these two modes of electron injection is made clearer if we briefly consider a simpler, but closely related problem, in which a single electron initially located in the plane $x = x_1$ is released at time $t = 0$ with zero initial velocity, and moves under the influence of a spatially homogeneous, but time-varying electric field $E = E(t)$. As before, it is assumed that the electric field is in the x -direction. We assume that the electric field initially is zero, $E(t = 0) = 0$, and, after a time interval T reaches a constant value $E(t \geq T) = E_T$.

The electron trajectory $[x_-(t), y_-(t)]$ in the x, y plane can be calculated for any prescribed, suitably well behaved function $E(t)$, and the result expressed in terms of quadratures.

The following results corresponding to two limiting cases are of interest for the purpose at hand:

1. $T \ll 1/\omega_-$, i.e., the electric field is assumed to reach its final value E_T in a time short compared to an electron cyclotron period. In this case, the electrons describe a cycloidal path, with a maximum excursion of

$$\frac{2eE_T}{m_- \omega_-^2}$$

from the plane $x = x_1$.

2. $T \gg 1/\omega_-$. The final electron trajectory (i.e., for $t \geq T$) is rectilinear, at a distance of

$$\frac{eE_T}{m_- \omega_-^2}$$

from the plane $x = x_1$ from which the electron was launched.

In either of these cases, the final electron trajectory is quite independent of any particular time dependence assumed for the applied electric field in the interval $0 < t < T$.

Considering once more the actual case at hand, i.e., the motion of an electron gas in a magnetic and self-consistent electric field, we shall examine the two limiting cases referred to above. In the first case, electrons are emitted at zero velocity from the plane $x' = 1$ (the projection along magnetic field lines of the actual cathode), and fall towards the ionizer. If the magnetic field is sufficiently large, the electrons will return to the vicinity of the plane $x' = 1$, resulting in two regions $0 < x' < x_2$ and $x_2 < x' < 1$, where the first region is occupied by ions only. In the second region, account has to be taken of a right-going and a left-going electron stream, as well as of the positive ions.

In the case of Brioullin injection, there are again two regions. Next to the ionizer exists a region which turns out to be inaccessible to the electrons; in a second region adjoining the plane $x' = 1$ both ions and electrons must be considered, the latter moving parallel to the y axis at the usual crossed-field drift velocity.

(c) Quasi-Cycloidal Electron Motion

Considering first the region $x_2 < x < x_1$, we find

$$\frac{d^2 \phi}{dx^2} + \frac{1}{\epsilon_0} \left(\frac{j_+}{u_+} - \frac{2j_-}{u_-} \right) = 0$$

from integrating Eqs. (5) and (7), and substituting into Eq. (8). The factor 2 appears due to the symmetry of the two electron streams. Integrating Eqs. (3) and (4) with the boundary conditions $u_- = v_- = 0$ at $x = x_1$ yields an expression for u_- . Substituting u_- and similarly u_+ [from the energy equation for the ions, Eq. (6)] into the previous equation, we find, in terms of the dimensionless quantities introduced earlier

$$\frac{d^2 \phi'}{dx'^2} + \frac{\frac{4}{9} j'_-}{(-\phi')^{1/2}} - \frac{\frac{8}{9} j'_-}{(1 + \phi' - \delta^2 (1 - x'))^{1/2}} = 0 \text{ for } x'_2 < x' < 1. \quad (11)$$

from a somewhat lengthy, but straightforward calculation.

In the region $0 < x' < x'_2$, no electrons are present and the ions are unaffected by the magnetic field. If we assume space-charge limited operation of the ion source, the Child-Langmuir equation applies in this region. Hence

$$\phi' = \phi'_2 \left(\frac{x'}{x'_2} \right)^{4/3} \quad \text{for } 0 \leq x' < x'_2 \quad (12)$$

where ϕ'_2 is the non-dimensional potential at $x = x_2$. It also follows that

$$j'_+ = \frac{(-\phi'_2)^{3/2}}{(x'_2)^2} \quad (13)$$

The boundary conditions imposed on the electrostatic potential are

$$\phi' = -1 \quad \left. \vphantom{\phi'} \right\} \text{at } x' = 1 \quad (14a)$$

$$\frac{d\phi'}{dx} = 0 \quad \left. \vphantom{\frac{d\phi'}{dx}} \right\} \quad (14b)$$

the latter condition resulting from space-charge limited operation of the electron source. Where the two regions are joined, matching conditions requiring that both the potential and its first derivative be continuous are imposed, i.e.,

$$\phi' \Big|_{x'_2 - 0} = \phi' \Big|_{x'_2 + 0} \quad (14c)$$

$$\frac{d\phi'}{dx} \Big|_{x'_2 - 0} = \frac{d\phi'}{dx} \Big|_{x'_2 + 0} \quad (14d)$$

The last condition means that we assume that there is no surface charge accumulated in the plane $x = x_2$. The assumption can be justified by an expansion in fractional powers of $x' - x'_2$, and is analogous to the situation at the turning points in the Ferraro-Rosenbluth cold-plasma sheath model (Refs. 6-8).

Equation (11) is a nonlinear differential equation for ϕ' , with two singularities. One of the singularities occurs at $x' = 1$, where the square root in the last term vanishes. This singularity is associated

with the vanishing of the x-component of the electron velocity at the plane where the electrons are injected [cf. the equation preceding Eq. (11)]. A second singularity occurs at the turning point of the electrons, $x' = x_2'$ where the x-component of the electron velocity and therefore the same square root again vanish. The location of the second singularity, as well as the two parameters j_+ and j_- , are a priori unknown, and must be determined from the solution of Eq. (11) and its boundary conditions.

Equation (11) has been integrated numerically on a digital computer. It is convenient to eliminate the explicit dependence on δ by introducing a new variable of integration $x'' = \delta(1 - x')$ and the parameters $j_+'' = (4/9\delta^2)j_+$ and $j_-'' = (4/9\delta^2)j_-$. Equation (11) then becomes

$$\frac{d^2 \phi'}{dx''^2} + \frac{j_+''}{(-\phi')^{1/2}} - \frac{2j_-''}{(1 + \phi' - (x'')^2)^{1/2}} = 0 \text{ for } 0 < x'' < x_2'' \quad (15)$$

The integration starts at the origin, ^{*}) satisfying the boundary conditions $\phi' = -1$ and $d\phi'/dx'' = 0$ at $x'' = 0$. It is necessary to first guess the value of the parameter j_+'' . (A value of j_-'' must also be assumed, but this does not amount to a second guess, since Eq. (15) and the boundary conditions no longer contain δ ; any choice of j_-'' will generally produce the required solution for some value of the magnetic parameter δ and is therefore of interest.)

The numerical integration proceeds until $1 + \phi' - (x'')^2$ vanishes again. This determines x_2'' , ϕ_2' and $(d\phi'/dx'')_2$. The procedure then must be repeated by successive approximation of j_+'' , until the matching condition

$$j_+'' = \frac{\left(\frac{d\phi'}{dx''}\right)^2}{4(-\phi')^{1/2}} \bigg|_{x'' = x_2''} \quad (16a)$$

is satisfied. The value of δ which corresponds to the particular solution is obtained from

$$\delta - x_2'' = -\frac{4}{3} \frac{\phi'}{d\phi'/dx''} \bigg|_{x'' = x_2''} \quad (16b)$$

^{*}) $\phi'(x'')$ has a branch point at the origin, but can be expanded (cf. Appendix) as a function of $(x'')^{2/3}$. The numerical integration is started at a slightly positive value of x'' , the gap being bridged by a power series.

Equations (16a) and (16b) are easily derived from Eqs. (12), (13), and (14c, d).

The trajectories of the electrons in the x, y plane are found from

$$\frac{dy'}{dx'} = \frac{v_-}{u_-} = \mp \frac{x''}{(1 + \phi' - (x'')^2)^{1/2}}$$

[obtained from integrating Eqs. (3) and (4)]. Hence, with a slight change of variables,

$$y' = \pm \frac{1}{\delta} \int \frac{x''}{(1 + \phi' - (x'')^2)^{1/2}} dx'' \quad (17)$$

where the constant of integration can be chosen arbitrarily.

In order for the electrons not to intercept the anode (a condition referred to in electron devices as "cut-off"), we must have δ larger than some critical value $\delta_{\text{cut-off}}$. From setting $x' = u_- = \phi' = 0$ in the expression

$$\frac{1}{2} (u_-')^2 = 1 + \phi' - \delta^2 (1 - x')^2$$

where $u_-' = (-e\phi_1/m_-)^{-1/2} u_-$; it follows that

$$\delta_{\text{cut-off}} = 1 \quad (18)$$

The computed ion and electron normalized current densities are plotted in Fig. 3. It may be noted that the current densities become infinite as cut-off is approached. The physical reason for this is easy to see, since at cut-off the electron turning points are forming a virtual cathode immediately adjacent to the anode, resulting in an infinite ion current density (which in turn requires an infinite electron current density).

For increasing magnetic field strength, i.e., increasing δ , the excursion of the electrons from the plane of injection tends to zero, resulting in an ion current density which approaches the Child-Langmuir limit. This is verified by the fact that the computed j_+ curve tends to 1 as δ becomes infinite.

(d) Brioullin Electron Motion

In this case, a simple closed form solution for the electrostatic potential can be obtained. Since the electrons originate at the cathode at potential ϕ_1 with negligible kinetic energy, and since $u_- = 0$,

$$\frac{1}{2} m_- v_-^2 = e(\phi - \phi_1)$$

in the interval $x_2 \leq x \leq x_1$. Since, therefore, $v_- = 0$ at $x = x_1$, integration of Eq. (4) gives

$$v_- = -\frac{eB}{m_-} (x_1 - x)$$

Substitution of v_- into the previous equation and introduction of the primed variables results in

$$\phi' = -1 + \delta^2 (1 - x')^2 \quad \text{for } x_2' \leq x' \leq 1 \quad (19)$$

In this case, therefore, the ion dynamics plays no role in the determination of the electrostatic potential. On the other hand, the electron density as a function of x' is determined by the ion dynamics [Eqs. (6) and (7)] and Poisson's equation [Eq. (8)].

The expression for ϕ' given by Eq. (19) is to be matched continuously and with a continuous first derivative to the corresponding expression for ϕ' given by Eq. (12). An elementary calculation for the dimensionless distance x_2' from the anode to the interface between the two regions, gives

$$x_2' = \frac{1}{2} \left(-1 + \sqrt{9 - \frac{8}{\delta^2}} \right) \quad (20)$$

(Only the positive root is physically meaningful.) The equation for the critical magnetic field strength at which cut-off occurs [Eq. (18)] is unchanged.

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Appendix

Expansion in the Neighborhood of the Singularity at $x'' = 0$

With the change of independent variables $w = (x'')^{2/3}$, Eq. (15) transforms to

$$\frac{d^2 \phi'}{dw^2} - \frac{1}{2w} \frac{d\phi'}{dw} + \frac{9j_+'' w}{4(-\phi')^{1/2}} - \frac{9j_-'' w}{2(1 + \phi' - w^3)^{1/2}} = 0 \quad (A-1)$$

Setting

$$\phi' = \sum_{i=0}^n a_i w^i \quad (A-2)$$

it follows from the boundary conditions at $w = 0$, that $a_0 = -1$, $a_1 = 0$. Substituting the expansion into Eq. (A-1), and comparing coefficients of equal powers of w yields

$$\left. \begin{aligned} a_2 &= \left(\frac{9j_-''}{2j_+''} \right)^{2/3} \\ a_3 &= \frac{1}{10} - \frac{9}{20} j_+'' \end{aligned} \right\} \quad (A-3)$$

Expansion (A-2), with $n = 3$, was used to compute ϕ' and $d\phi'/dx''$ at a small distance Δ from the singular point $x'' = 0$, and thereby to provide initial conditions for the numerical integration of Eq. (15). A simple, although of course only partial check of the computer program and of

the accuracy of the above expansion was obtained by observing that the results for ϕ' are virtually unaffected if Δ is changed to some other small positive value.

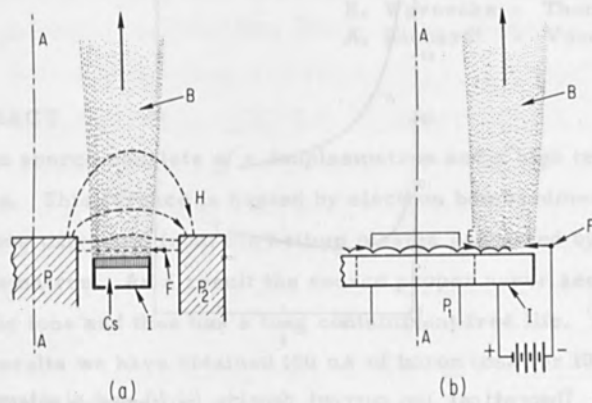


Fig. 1. Schematic of Space-Charge-Sheath Ion Source: (a) radial cross section, (b) side view.

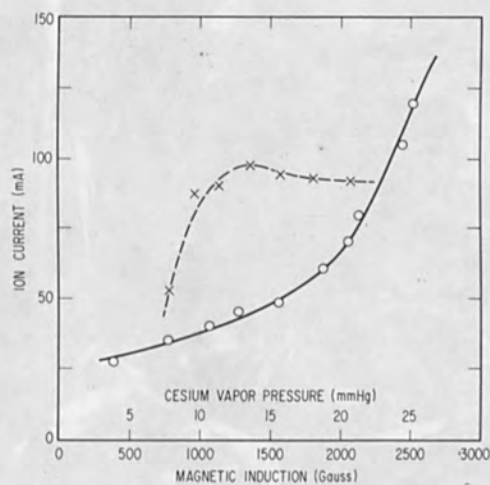


Fig. 2. Measured ion current vs. cesium vapor pressure in reservoir (solid curve), and vs. magnetic induction (dotted curve).

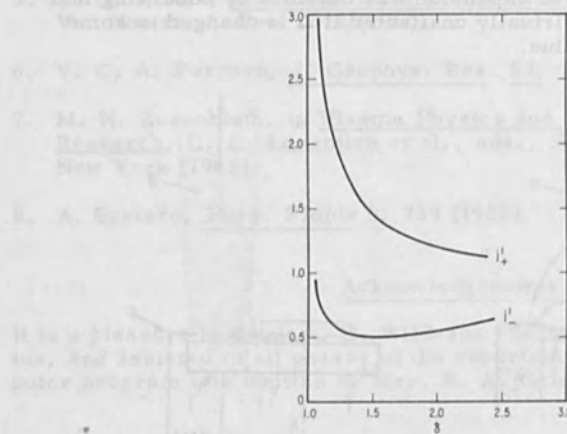


Fig. 3. Theoretical ion current density (j_+') and electron current density (j_-') vs. magnetic field parameter.

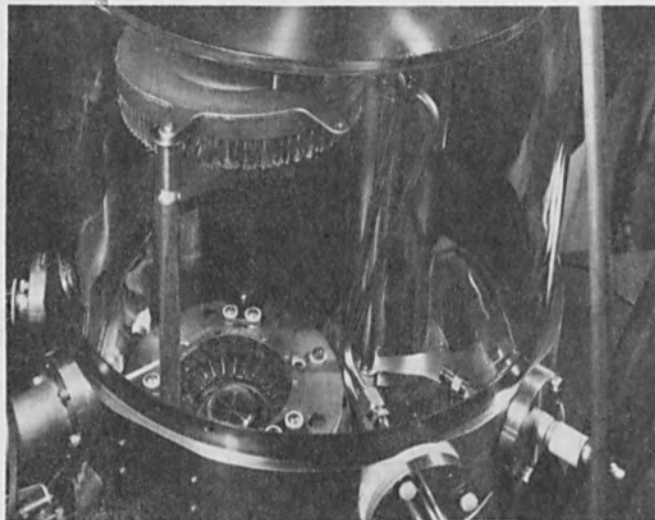


Fig. 4. Experimental ion source and beam target plate.

PRODUCTION OF HEAVY IONS USING THE PLASMA
JET FROM A DUOPLASMATRON SOURCE

R. Warnecke - Thomson-CSF
A. Richardt - Veeco S.A.

ABSTRACT

The ion source consists of a duoplasmatron and a high temperature furnace. This furnace is heated by electron bombardment using electrons extracted from the helium plasma generated by the duoplasmatron. As a result the source proper never sees the metallic ions and thus has a long contaminant free life. Among first results we have obtained 100 μ A of boron ions for 100 hours of continuous operation.

(A) INTRODUCTION

We have developed a new use of the duoplasmatron 1) to produce ion beams from various solid elements, including metallic elements. The principle of a duoplasmatron is illustrated in Fig. 1. A low pressure arc is produced between a cathode K (oxide cathode) and an anode A which is at a potential of about a hundred volts with respect to the cathode. An intermediate electrode G, of a conical shape, is connected directly to the anode through a resistance R, and self-biases automatically to a potential intermediate between the cathode's and anode's producing a first pinching effect on the discharge. The plasma of which the density has thus been increased diffuses with the non-ionized gas in the area GA where there is a strong axial magnetic field B. The existence of this field brings a new increase of the plasma density as a result on one hand of a magnetic pinching effect, on the other hand of the post ionizing effect of accelerated and focused electrons that

originate from both the cathode and the plasma. One thus obtains, at the level of the anode, a plasma of very small dimensions (diameter of the order of 1 millimeter) and a high degree of ionization (density of the order of 10^{14} ions/cm³).

(B) USE OF AN EXPANSION CUP

When one wants to decrease the initial divergence of the ion beam, one must decrease the density of the plasma at the level of the exit hole of the source. This is done by prolonging the anode by use of an expansion cup centered on the beam axis. This cup acts as an electrostatic shield with respect to the extraction electrical field (Fig. 2).

The plasma issuing from the source diffuses in this chamber and presents, face to the extractor, an emissive surface of which - for equal discharge and extraction voltage characteristics - the shape and the density depend only upon the geometry of the cup and the extractor. However while this manner of operating allows for an improvement of the optical qualities of the beam at the level of the extraction zone, it also brings, - all other things being equal - a sizeable reduction of the extracted ion current. As shown by G. Gautherin 2) however, one can get around this disadvantage (due mainly to losses of electrons on the walls) either by using a cup with insulating walls or by creating a small axial magnetic field in the expansion volume.

(C) PHENOMENON OF POST IONIZATION. APPLICATION TO THE PRODUCTION OF HEAVY IONS

Recent works have shown that in the usual arc and pressure conditions in the ionization chamber of a duoplasmatron (for example $I_{\text{arc}} = 5\text{A}$, $P = 10^{-1}$ torr), the plasma that diffuses in the expansion cup is made of three types of particles:

(1) Electrons:

which come from the cathodic plasma and are accelerated by the positive potential difference V_p that exists between this plasma and the walls of the cup. Having energies of between 50 eV and 100 eV, these electrons constitute at the entrance of the expansion cup a very intense beam (approx. 1 A) with a high ionizing power (Fig. 3).

(2) Ions:

that come from the anode zone.

(3) Neutral particles:

made of, in the main, atoms or molecules of gas G_1 introduced in the source which come out of this source without having been ionized.

In the middle of the cup, the residual pressure P_{RI} of the non-ionized gas G_1 has in the normal utilization conditions of the source, a value comprised between that of pressure P_i (about 10^{-1} torr) in the ionization chamber, and that of pressure P_e (about 10^{-6} torr) in the vacuum chamber to which the source is connected. When the value of P_{R2} (which is of course a function of the size of the anode hole, and the exit orifice of the cup) reaches the 10^{-4} torr region, the number of neutral particles that are ionized becomes important, hence the high ionization rate which characterizes this type of source. It is this phenomenon of "post-ionization" which we use to produce ions of various elements, in particular ions of metallic elements. The principle, illustrated by Fig. 4 is as follows:

The duoplasmatron is fed by a gas G_1 (preferably a neutral gas, argon, or even better helium). We introduce in the expansion cup the element G_2 in the form of a gas or a vapour. Experiments

have shown that the larger the difference $P_{R2} - P_{R1}$ (between the values of the partial pressures of gas or vapour G_2 and of gas G_1), the richer the extracted beam will be in ions G_2^+ . (The impurity rate defined by the ratio I_1/I_2 of the currents carried by the 2 species of ions G_1^+ and G_2^+ can at the limit become zero).

All other things being equal, this ratio depends upon the nature of elements G_1 and G_2 , but in the case where the adjustment of the flow of gas G_2 alone (or the rate of vaporization of G_2) is not sufficient to obtain a sufficiently pure beam of ions G_2^+ , one can always eliminate the G_1^+ ions with magnetic filtering.

The advantages of this method are multifold:

- (1) due to the small conductance of the anode hole and the high ratio P_1/P_{R2} , the retrodiffusion effects are small; it is thus possible to keep a reasonable pressure in the expansion cup without having as a consequence an entrance of an important quantity of gas or vapour G_2 in the ionization chamber of the source.

Because of this fact, it is possible to produce without difficulties ions of chemically active elements (oxygen or carbon compounds for example) that could not be introduced directly in the source without risks of damaging the cathode or contaminating the walls of the ionization chamber.

- (2) What we have described in (1) is valid whether for a gas or for a vapour. In the latter case, in opposition to what happens in classical sources, if the vapour condenses partly on the cold wall of the anode, the exit orifice of the source is continuously cleared by the plasma jet that comes out of it, and thus has no tendency to clog.

- (3) As a result of the small volume of the extraction zone plasma vapour, a moderate heating power (a few hundred watts) is enough to bring the expansion cup and the reservoir that holds specimen G_2 to the evaporation temperature of G_2 .
- (4) Lastly, due to the use of a switching mechanism for the expansion-cup/reservoir, it is possible to rapidly switch (some 30 seconds) from one type of ions to another with memory effects reduced to a minimum, and without having to modify the arc regime in the source.

(D) HEAVY ION SOURCE, TYPE SDM

The SDM 3) is an original ion source that can be used to produce ions of almost all elements, and in particular those belonging to columns III and IV.

Fig. 5 gives the schematic representation of the SDM and the switching system to change quickly from one type of ions to another. The cathode of the duoplasmatron is made of a nickel sheet covered with oxides and spiraled in the inside of a nickel reflector. The lifetime of this cathode is better than 300 hours. One sees that in the main the SDM source is made of a duoplasmatron combined with a furnace (9) with two lateral openings; one to receive the support (8) of the heating element made of two tungsten filaments (12), the other to give passage to a mobile subassembly, including one post ionization chamber (10) and one crucible (11). In this crucible is placed the specimen that is to be evaporated so that this vapour can then be ionized by the electrons of the plasma coming from (4). An auxiliary coil (19) is placed in the body of the furnace creates a magnetic axial field a few tens of gauss which increases the efficiency of

the post ionization. Thermal insulation is provided by a series of tantalum shields (13).

The furnace is of OFHC copper, chromed outside, and water-cooled. Depending upon the nature of the products to be evaporated, the crucibles are stainless steel, tantalum, molybdenum or tungsten. The switching mechanism has a magazine fixed to the furnace body by a vacuum valve used for isolation purposes.

This magazine holds a removable stainless steel barrel with four chambers; in the inside of each chamber is a crucible/post-ionization chamber set. A motor drives the rotation of the barrel bringing to the orifice of the furnace the chamber that holds the specimen from which one wishes to obtain ions.

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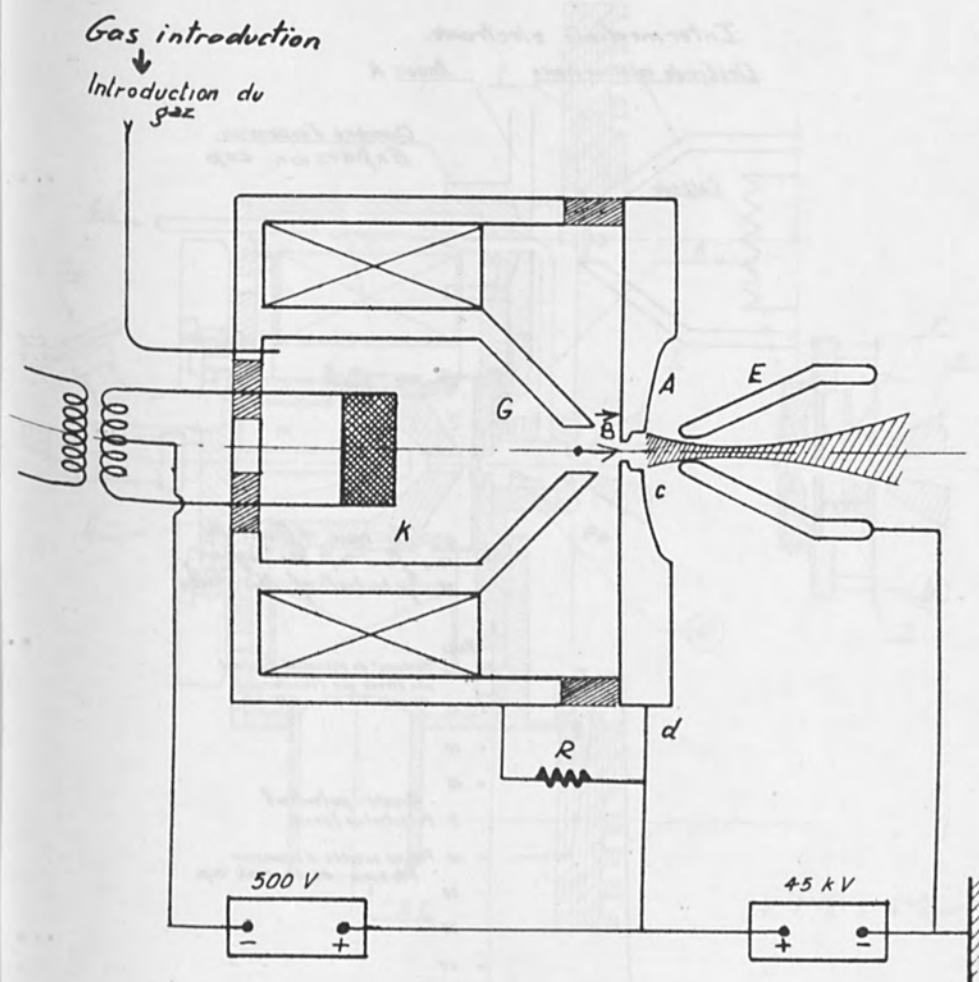


Figure 1

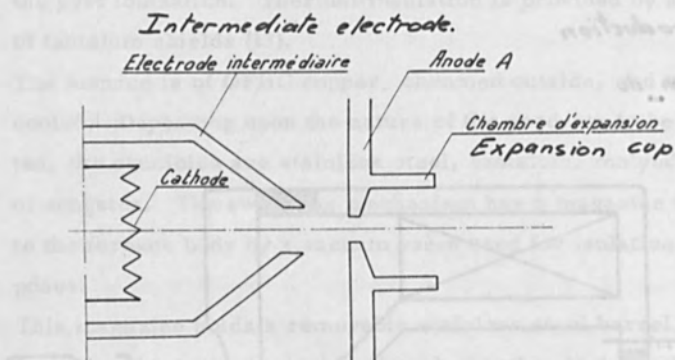


Figure 2

The more difficult the ions flow is, the higher the potential of this hill.

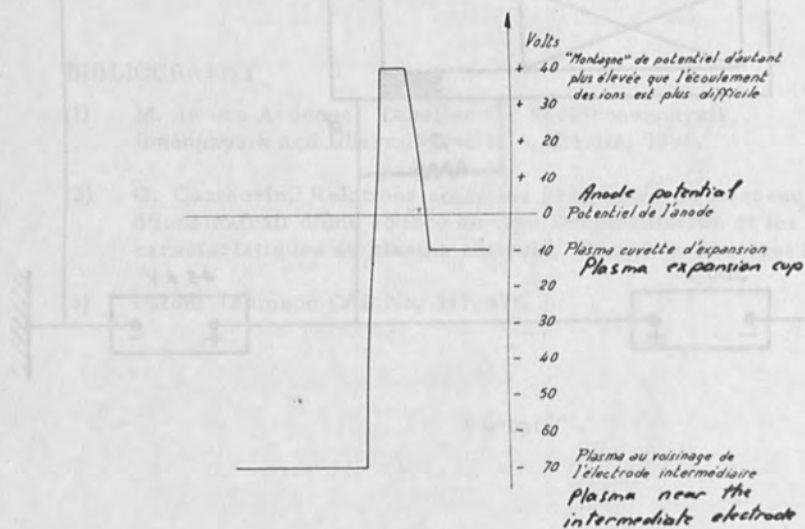


Figure 3

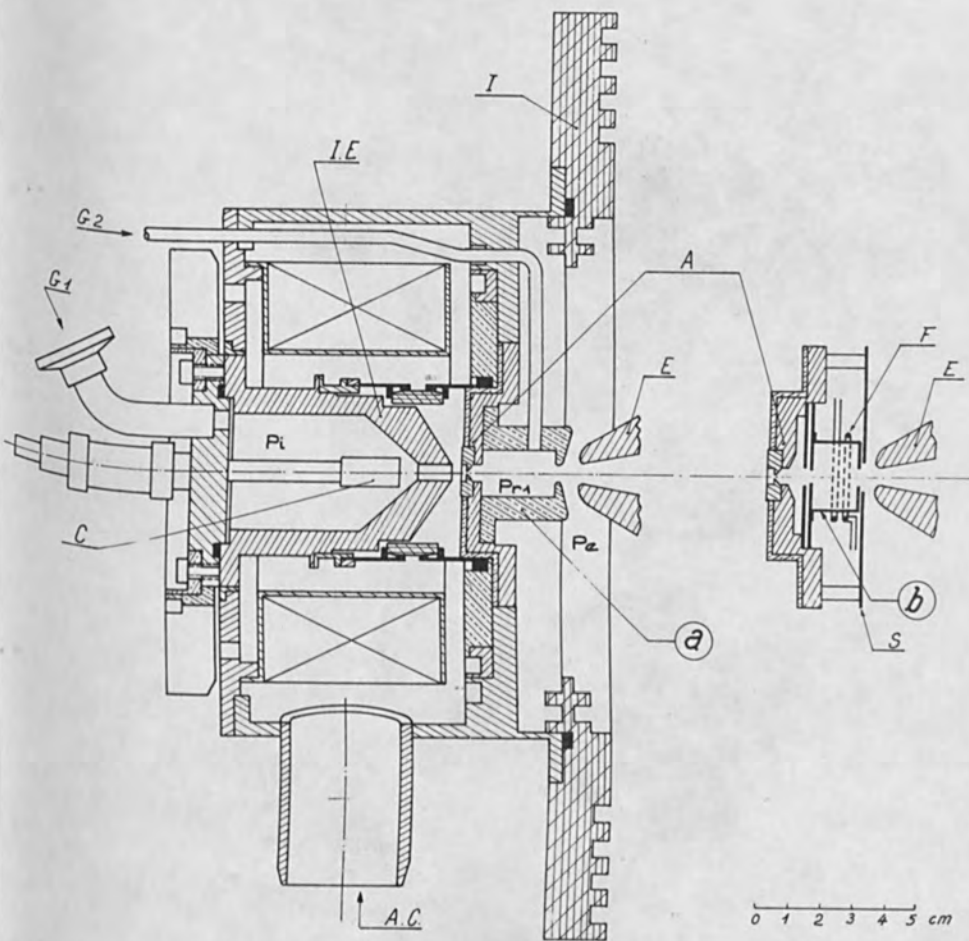
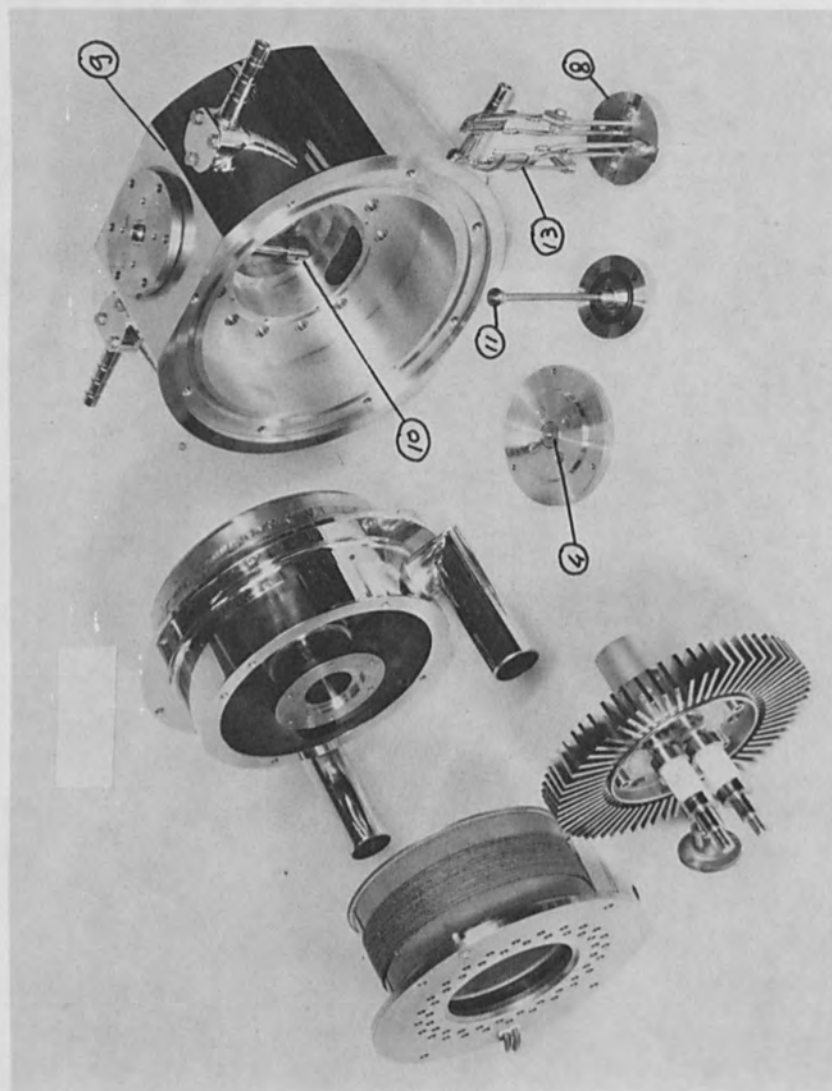


Figure 4



On the Behavior of Electron Beams in Weak Vacuum

H. Schiller, U. Heisig, and G. Jägersch.

Contributed by the Forschungsinstitut Manfred von Ardenne,
Dresden / GDR.

Introduction

It is known from physical and technological reasons that electron beam processing plants have to be operated in the high vacuum range, and especially at a pressure of 10^{-5} Torr.

SECTION II

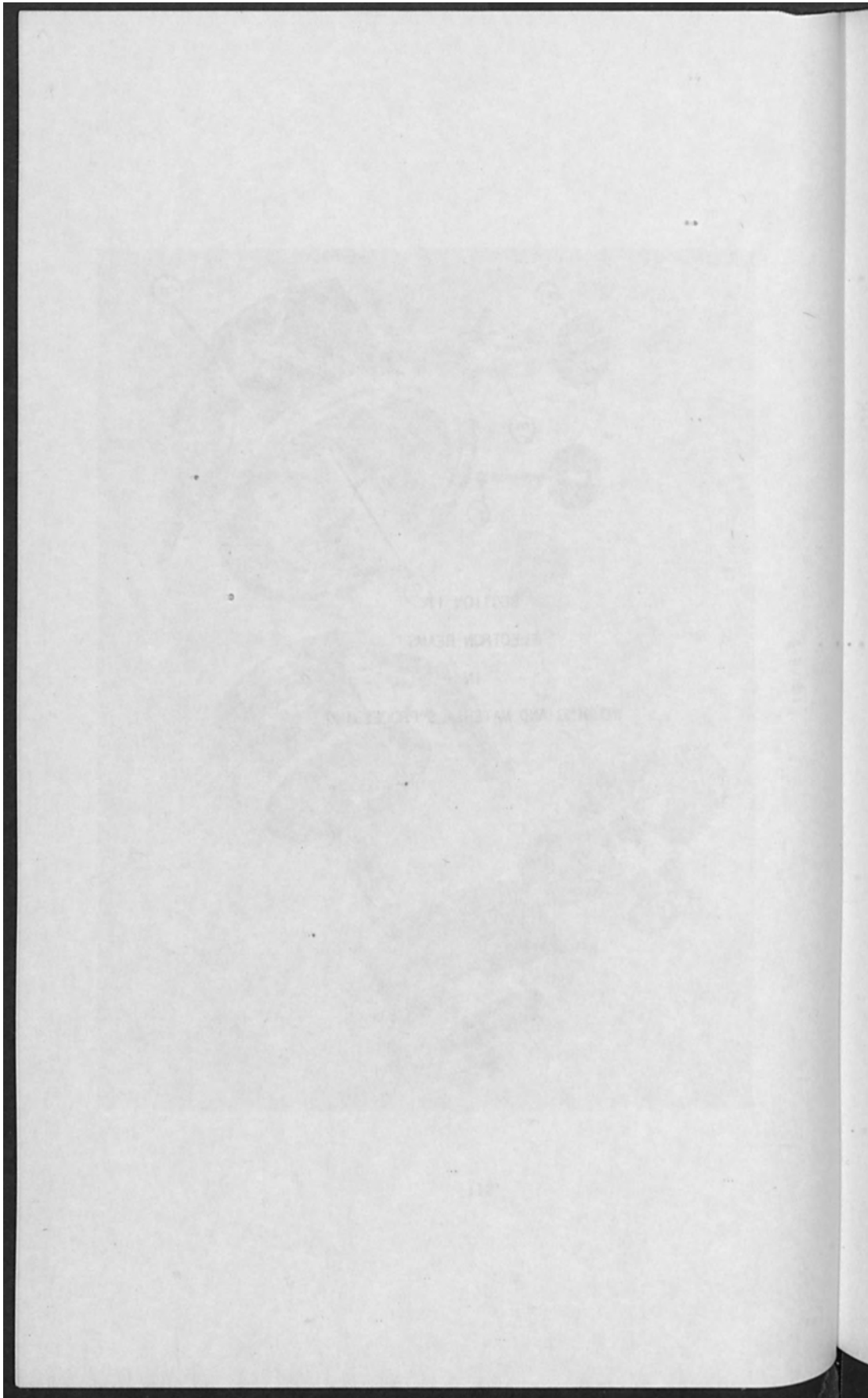
ELECTRON BEAMS

IN

WELDING AND MATERIALS PROCESSING

... of the beam properties, and that of the results of the welding process, are changed by the electron beam and the electron beam welding process between electron and molecules of the material, the generated plasma and a certain extent of the developed vapors.

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



On the Behaviour of Electron Beams in Weak Vacuum

S.Schiller, U.Heisig, and G.Jaesch.

Contributed by the Forschungsinstitut Manfred von Ardenne,
Dresden / GDR.

1. Introduction

For known physical and technological reasons the electron guns of electron beam processing plants have to be operated in the high vacuum range, and especially at a pressure of $\leq 10^{-4}$ Torr.

However, most of the known electron beam processes, such as electron beam welding, melting or evaporation may be carried out more profitable also at a higher pressure level. In such a case the electron beam must pass a distance of weak vacuum without critical alteration of the essential properties demanded by the respective technological process.

Changes of the beam properties, and thus of the results of processes carried out under weak vacuum, are caused by scattering processes between electrons and molecules of the residual gas, the generated plasma and - to a certain extent - of the developed vapours.

With electron beam welding, the pressure-depending effects on the possible depth of welding have been dealt with in various papers [1], [2], [3], [4]. Due to a reduced energy transfer to the workpiece and the broadening of the electron beam the depth of welding penetration reduces with increasing pressure. In addition, there is a deterioration of the depth-to-width ratio of weld profiles. Fig.1 shows this known effect with various electron beam parameters.

Another example concerning the influence of the residual gas is the alteration of focusing conditions in electron guns for melting furnaces. Here a definite gas intake in the gun reduces the power loss in the latter through filament beam production [5]. In this case a sub-pressure of $3 \cdot 10^{-4}$ to $5 \cdot 10^{-4}$ Torr has to be maintained in an intermediate chamber. The cited examples clearly reflect the increasing interest in further information with reference to pressure-dependent alterations of scattering, total energy losses, energy transfer and distribution of power density on the workpiece encountered with electron beam processing plants.

2. Working Pressure Limits of Electron Processing Plants

The propagation of an electron beam remains undisturbed if there are neither energy losses nor deflections from the initial path caused by collisions with molecules or ions. Such effects may be neglected if the distance l is less than the gas kinetics mean free path of electrons l_λ .

The field of path length and pressure below the straight line $l = l_\lambda$ of Fig. 2 represents that range where the effects of the residual gas may be neglected. Among other things, this includes all high vacuum electron beam processing plants.

In case electron beam scattering in residual gas reaches a magnitude with a mean scattering angle equal to or larger than 10° , the beam has just passed a path length l_x which is known as Lenards's parallel drop path [6]. As a rule, such a scattered beam should not be used for technological processes. The field between the straight line l_λ and l_x denotes a range of limited effects caused by the residual gas.

In this field are included electron beam plants which operate in the weak vacuum range up to normal atmospheric pressure.

The influences caused by a residual gas atmosphere are due to the known interactions between the electrons of the beam and the gas molecules. Such interactions are:

- elastic scattering without energy release on the fields of force of the atomic nuclei;
- non-elastic scattering with energy release between the electrons of the beam and the extranuclear electrons of the atoms; and
- velocity scattering due to discrete energy losses caused by quantum jumps during interactions with the atomic shell.

The fundamentals of these processes have been studied both theoretically and experimentally. As a result, the proportions of a scattered electron beam compared to a non-scattered configuration may be estimated in dependence of the parameters influencing the scattering process:

pressure, gas type, accelerating voltage and beam path length [7] (see compilation in ref. [8]).

However, such an estimate does not permit quantitative predeterminations with respect to the function of electron beam processing plants under the restricting influence of the residual gas. These effects are too intricate.

Especially it is difficult to estimate quantitatively the influence of the plasma on the focusing of the beam.

In addition, it should be pointed out that even the scattered components of the beam contribute to the process.

As to the practical application of electron beam processing plants, informations concerning the scattering angle, the current proportion in a certain angular range and the amount of discrete energy losses as given by the various scattering theories are of limited importance. Our main interest deserves the overall energy loss, the energy transfer to the workpiece, and the power density distribution of the electron beam in dependence of the pressure.

3. Energy losses along the beam path and energy transfer to the workpiece.

3.1 General

With an electron beam process it will be impossible to determine the energy transfer to the workpiece simply by measuring the electron beam power $p = U_B \cdot I_B$. This applies to high vacuum processes as well. When measuring the beam current with the aid of a cage, it is generally known that reflected and backscattered beam electrons, thermally emitted electrons and electrons as well as ions generated by the ionization of metal vapours have to be considered, too. Testing conditions for the determination of electron currents in high-power guns by means of a cage are well-known [9]. Such measurements were employed to determine the electric efficiency η_{EL} of welding guns, where

$$\eta_{EL} = \frac{I_{A1}}{I_B} ;$$

with I_{A1} = the beam current bombarding to the workpiece and I_B = the current in the electron source.

As compared with the electric efficiency η_{EL} , the efficiency of power or energy transfer η_Q to the workpiece is still more interesting. We obtain

$$\eta_Q = \frac{Q_2}{Q_1} ;$$

where Q_2 is the energy absorption of the workpiece which is equal to the energy absorption of a calorimeter without cage. Q_1 denotes the energy of the electron beam bombarding to the workpiece.

Contrary to the electric efficiency η_{EL} , the efficiency of the energy transfer η_Q to the workpiece is markedly reduced by the portion of backscattered electrons [10].

Other authors have calculated the efficiency of the energy transfer with electrons incident on tungsten, copper and aluminium and found values for η_0 in the order of 0.7, 0.77 and 0.9 [11]. It should be pointed out, however, that these theoretical values were obtained without the effects of power density on the energy transfer. However, we have found the power density influences these values essentially. Therefore we have measured also with different power densities.

Calorimetric and cage current measurements were performed and compared. Fig.3 gives a schematic presentation of the test set-up used. The following parameters were measured:

I_B Beam current in the electron source (beam power $p = U_B \cdot I_B$)

I_{A1} Beam current bombarding to the workpiece (measured with the aid of a cage as shown in Fig. 3b)

Q_1 Energy of the bombarding beam (measured by means of a calorimeter with cage - see Fig. 3b)

I_{A2} Electron current from workpiece to ground (measured without cage and back-up voltage acc'd. to Fig. 3a)

Q_2 Energy absorption of a calorimeter without cage and without back-up voltage = energy absorption of a workpiece.

3.2 Measurements in High Vacuum

In order to check the validity of the test set-up it was necessary to perform measurements in high vacuum. Some characteristic results with different arrangements and

sub-pressures of $\leq 10^{-4}$ Torr are shown in Fig.4 . During the measurements the power density of the beam was altered by changing the focusing conditions; i.e., by changing the lens current I_L . Qualitative considerations with reference to the range of power densities used for the measurements may be seen from the schematic illustration of penetration profiles obtained with Ni-Cr steel (rate of feed $v_s = 2$ m/min.) .

Based on these tests we came to the following conclusions:

3.2.1 Independent of focusing conditions - and with the three applied pressure stage diaphragms - the electric efficiency $\eta_{EL} = I_{A1}/I_B$ of the employed ES 150/10 electron gun is ≈ 0.9 .

3.2.2 According to calorimetric determination, the incident energy $Q_1 = I_{A1} \cdot U_B \cdot t$; with I_{A1} being the current measured by means of a cage-type target.

The incident energy Q_1 may thus be obtained by a simple cage-current measurement; a method which does not depend on focusing conditions.

3.2.3 Energy absorption Q_2 of the workpiece essentially depends on focusing conditions. With a defocused electron beam, i.e., a beam of low power density, the major part of the incident electron energy is not converted into heat; mainly due to backscattering and other secondary effects. With copper as target material, the measured efficiency of the energy transfer $\eta_Q = Q_2/Q_1$ in this defocused range is about 0.8 and agrees well with the theoretical values given in [11].

In the range of high power densities used for electron beam welding, the efficiency of the energy transfer η_Q increases with the power density. Under the specified conditions, the efficiency in the point of accurate focusing is $\eta_Q = 0.98$.

This high value of energy transfer can be related to the so-called 'deep welding effect' [12]. The proportion of backscattering electrons becomes smaller corresponding to penetration.

- 3.2.4 The electron current I_{A2} from the workpiece to earth shows a behaviour similar to Q_2 . Deviations between I_{A2} and $Q_2/U_B \cdot t$ can be traced back to the fact that the various test set-ups resulted in a different coverage of the secondary effects.

3.3 Measurements in weak-vacuum

Measurements have been carried out in dependence on the pressure level p both on a defocused beam - i.e., a beam having a relatively low power density - and on a focused beam of very high power density. The other parameters are the same as shown in Fig.4. The applied values of lens currents are marked in Fig.4.

3.3.1 Test results with a defocused beam (see Fig.5)

- Owing to the interactions with the residual gas, the values of the incident beam current I_{A1} , of the incident energy Q_1 , and of the energy Q_2 transmitted to the workpiece will reduce with rising pressure p .
- Up to about 10 Torr, the calorimetrically determined incident energy Q_1 equals $I_{A1} \cdot U_B \cdot t$; that means, up to this pressure range the incident energy may be determined by means of a simple cage current measurement. We can point out, that both calorimetric and cage current measurements gave the same results.
- Up to a range of about 10 Torr, the efficiency of the energy transfer remains practically constant at $\eta_Q \approx 0.8$. (Note: higher pressure levels were not included in the measurements)

3.3.2 Fig. 6 gives the measured results of the incident energy Q_1 on the workpiece and the associated energy transfer Q_2 as a function of the pressure with a focused beam. Moreover, this illustration shows the decrease of penetration depth of welds d obtained with Ni-Cr steel. - Measurements of this depth were carried out with an electron beam power of 3 kW, and a working distance of $a = 10$ mm. Fig.6 further includes the theoretical ratio of the non-scattered portion of the electron current I to the incident electron current I_0 .

These tests and comparisons led to the following conclusions:

- The expected unidirectional decrease of both the incident energy Q_1 and the energy transmitted to the workpiece, Q_2 , is confirmed of up to more than 10 Torr.
- With adequate power, and conditions as specified in this paper, the efficiency of the energy transfer, $\eta_Q = Q_2/Q_1$, remains at about 0.98 even at higher pressure levels.
- The energy Q_2 transmitted to the workpiece is larger than the expected values based on the theoretical ratio I/I_0 of elastic scattering. Hence, a considerable part of the scattered electrons also reaches the workpiece.
- With increasing pressure it is found that penetration of welds, reduces only at higher pressure levels well above the expected theoretical I/I_0 ratio of elastic scattering. Electrons scattered through small angles in the region of the focal spot diameter obviously contribute to establish a high penetration effect.

3.3.3 Filament beam production in the electron gun may take place under certain conditions of pressure. This is seen characterized by an additional focusing effect. Test results shall be cited in order to demonstrate the effect of such a filament formation on the electric efficiency η_{EL} . Measurements of the incident electron beam energy, $I_{A1} \cdot U_B \cdot t$, were first carried out at a pressure of 10^{-4} Torr, and with a working distance of $a = 100$ mm. Then - with the beam power kept constant at $I_B \cdot U_B = 8.7$ kW and with $U_B = 100$ kV, the energy Q_2 transmitted to the workpiece was determined as function of the pressure level. The lens current was constant. The obtained results are shown in Fig.7. In a certain pressure range, $Q_2 = Q_2(p)$ becomes larger than Q_2 at 10^{-4} Torr. In other words, additional focusing due to filament beam production increases the energy output of the electron gun as compared to the output obtained at 10^{-4} Torr. In consequence, filament beam production reduces the energy loss in the gun. With the mentioned example, this reduction of the energy loss due to filament beam production amounts to 7 per cent, max. The effect of filament beam production is utilized in practice with our high power electron guns.

4. Alteration of power density distribution in electron beams.

Apart from the incident energy and its transfer to the workpiece, it is the distribution of power density in the bombarding beam which is of significance for electron beam processes in production plants.

Its determination becomes possible by various methods; especially with the aid of suitable diaphragm and probe arrangements.

Diaphragm measurements result in direct distribution curves provided the diaphragm diameter has been kept very small as compared to the diameter of the electron beam spot. However, such conditions are hardly achieved in practice, especially with beams having a small spot diameter. Hence it will be necessary to resort to comprehensive test set-ups with additional imaging systems [13].

Deriving the distribution of power density from a beam is also possible by means of probes which - as compared to the electron beam spot - are either of a larger [14] or a smaller diameter [15], [16]. Frequently, a wire probe is rapidly moved relative to the electron beam to measure the current flow into the probe. These tests confirmed that - for electron sources with small area cathodes and neglected interactions with the residual gas - the distribution of the power density $p(r)$ in the central zone is of normal Gaussian distribution. In this range, the following equation applies:

$$p(r) = p_0 \exp - (r/\sigma)^2 .$$

With a given spot diameter of such an electron beam, this generally refers to a diameter of 2σ [14] which means that 63 per cent of the incident beam power concentrates on the workpiece within the given diameter.

In this context, it should be pointed out, that the mentioned test methods are liable to fail with extremely high power densities or beam powers as the heating of the probes is too high and because the secondary effects increase. Owing to the latter, application of these methods seems to be crucial especially in the higher pressure ranges is uncomfortable. But there are still other possibilities to derive qualitative information concerning the distribution of the power density directly from the beam action. With reference to the latter it is

assumed that the applied work of the electron beam per area unit - e.g., eroding of sublimable matter or melting of metals - is almost proportional to the incident beam power. Moreover, the mutual influence of the individual area units hit by the beam have to be kept small. These conditions are met by applying materials which sublime or have a low thermal conductivity. Examples of such influences are the known penetration profiles of welds in Ni-Cr steel shown in Fig.1 and the penetration profiles in graphite and oak wood shown in Fig.8 . Under these conditions it will be possible to determine the distribution of the power density $p(r)$ from the boundary curves $h(x)$ of such a profile.

Assuming a normal Gaussian distribution of $p(r)$, it is then possible to read off an 'electron beam diameter' of 2σ directly from the boundary curves.

If the beam is used to burn-through either conducting or nonconducting foils, the width b of the burn-through trace the passing power P_D , and the incident power P_A on the foil may be used to determine a beam action diameter 2σ based on a Gaussian distribution of the power density $p(r)$.

This is achieved by means of the following equation:

$$P_D = \frac{2 (P_A + P_D)}{\sigma \sqrt{\pi}} \int_0^b dx \exp -(x/\sigma)^2 .$$

Fig.9 shows these beam action diameters 2σ for the various target materials as a function of the pressure level. In the range of minor interactions with the residual gas, these action diameters will coincide within a factor of 2, - a result, which is surprisingly good if we bear in mind that the mentioned conditions are only roughly met. In the higher pressure range, i.e., pronounced interactions

with the residual gas, the action diameters for various materials diverge by more than a factor of 10. This divergence clearly reveals that in the range of residual gas influences according to Fig.2 the assumption of a Gaussian distribution of the electron beam does not hold. Hence it is suggested to utilize the following distribution to obtain an analytical description of the distribution of electron beam power density under conditions of residual gas effects:

$$p(r) = p_{01} \exp -(r/\tilde{\sigma}_1)^2 + p_{02} \exp -(r/\tilde{\sigma}_2)^2 .$$

The first addend with $\tilde{\sigma}_1$ describes the distribution of the non-scattered beam current component.

The second addend with $\tilde{\sigma}_2$ refers to the portion of the scattered beam current.

Hence:

$$p_{01} = f(\text{pressure}) \quad p_{01}(10^{-4} \text{ Torr}) = p_0$$

$$p_{01} \longrightarrow 0 \text{ for high pressure levels}$$

$$p_{02} = f(\text{pressure}) \quad p_{02}(10^{-4} \text{ Torr}) = 0$$

$$\tilde{\sigma}_1 = \text{constant} \quad \tilde{\sigma}_2 = f(\text{pressure}) .$$

With high vacuum, this function reduces to the normal Gaussian distribution. In the range of extreme residual gas effects, i.e., with 'complete diffusion', it is only the second addend which describes the power density.

The weld profiles shown in Fig.1 demonstrate that the description by two addends is more suitable in a certain pressure^{range} than a simple distribution.

Fig.10 is a photograph of the electron beam equipment which serves for measurements. Maximum voltage amounts 150 kV and max. power is 12,5 kW.

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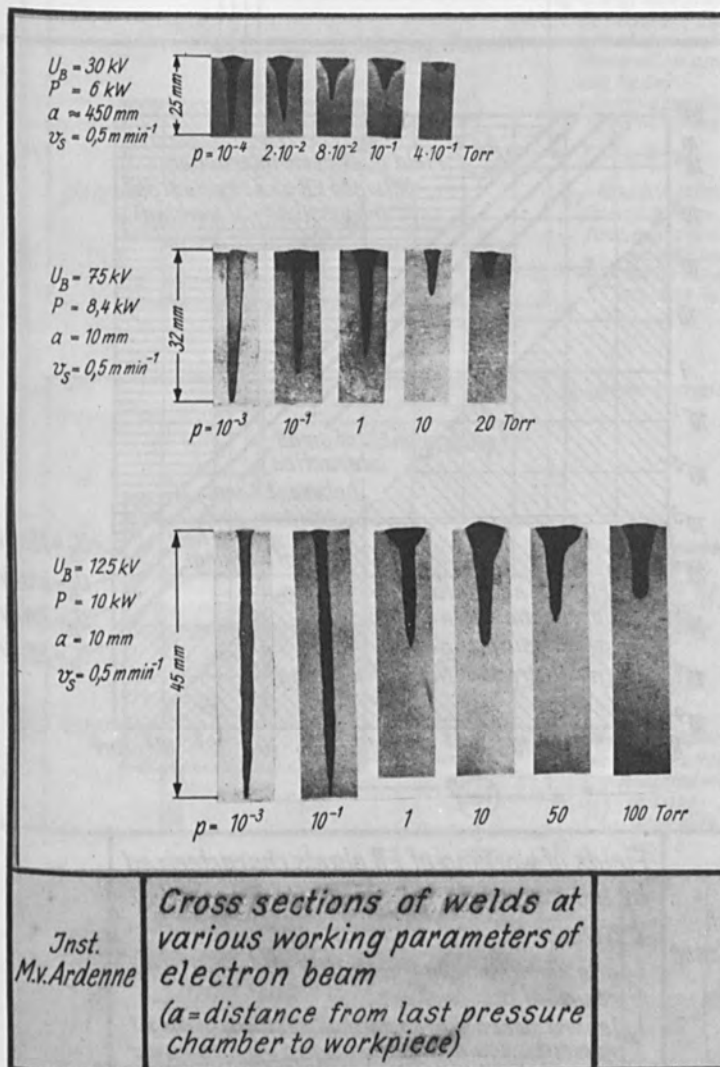


Figure 1

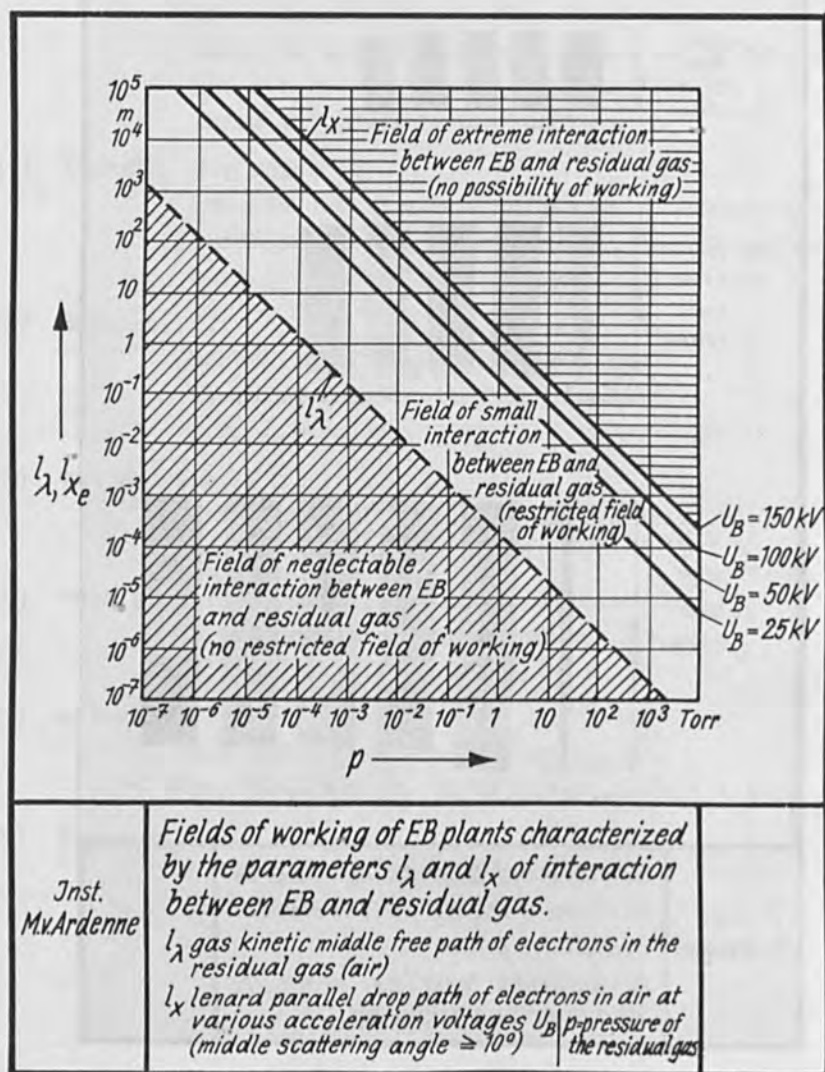


Figure 2

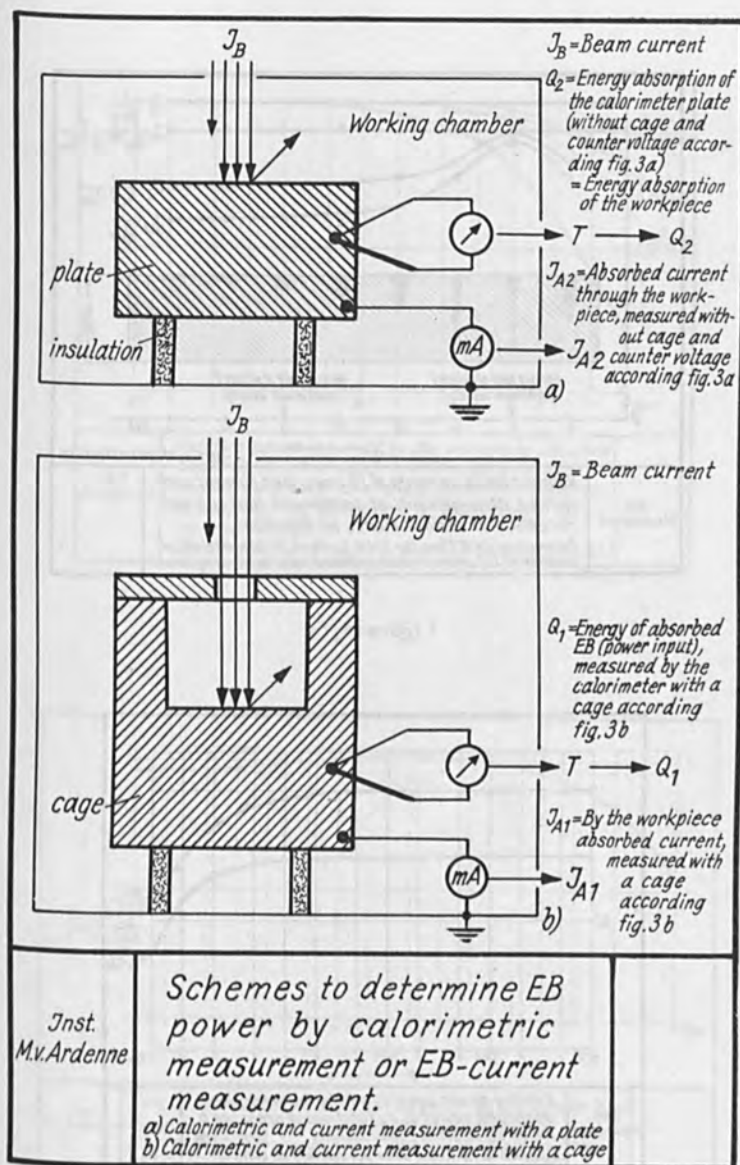


Figure 3

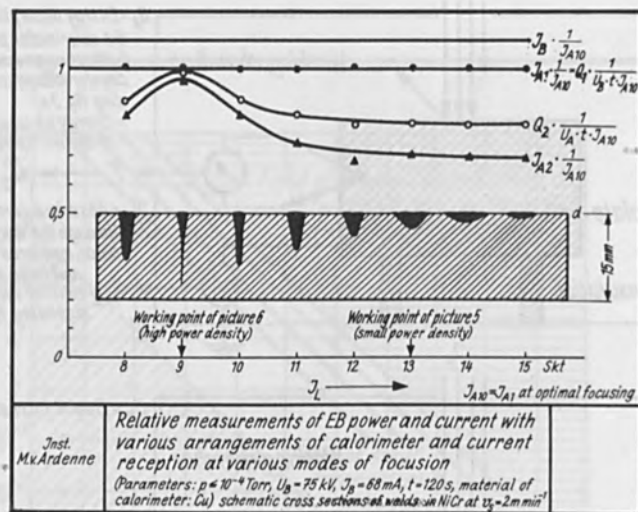


Figure 4

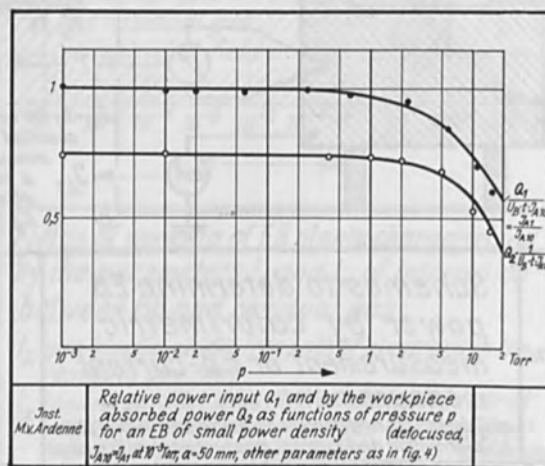


Figure 5

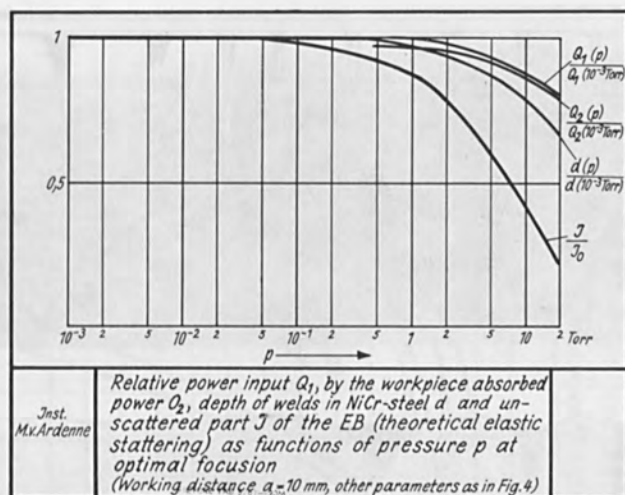


Figure 6

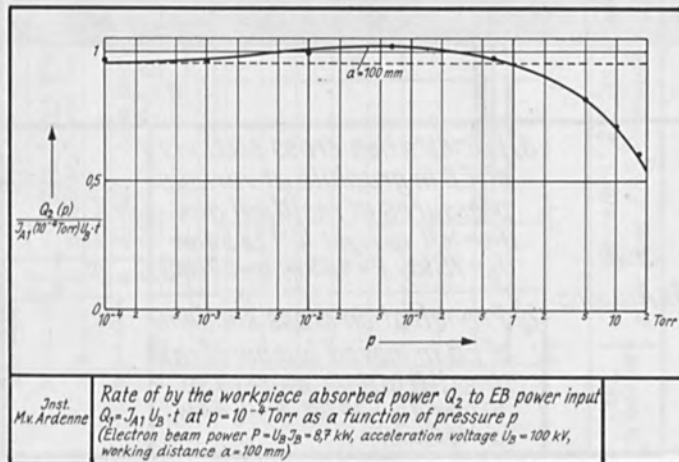


Figure 7

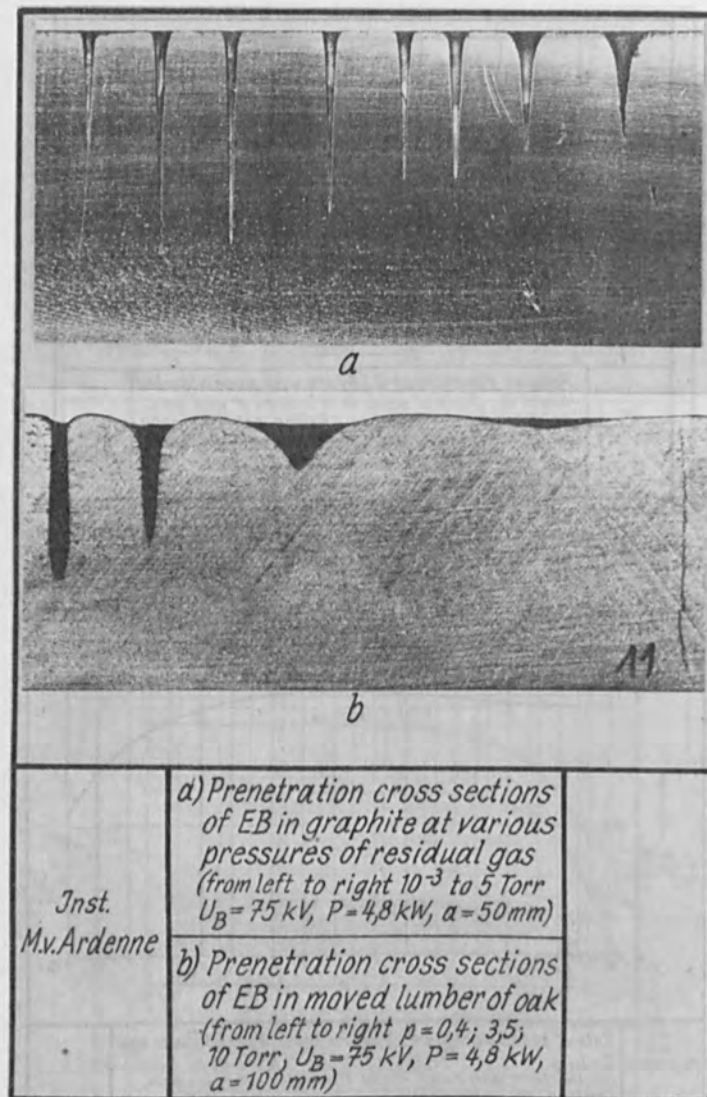
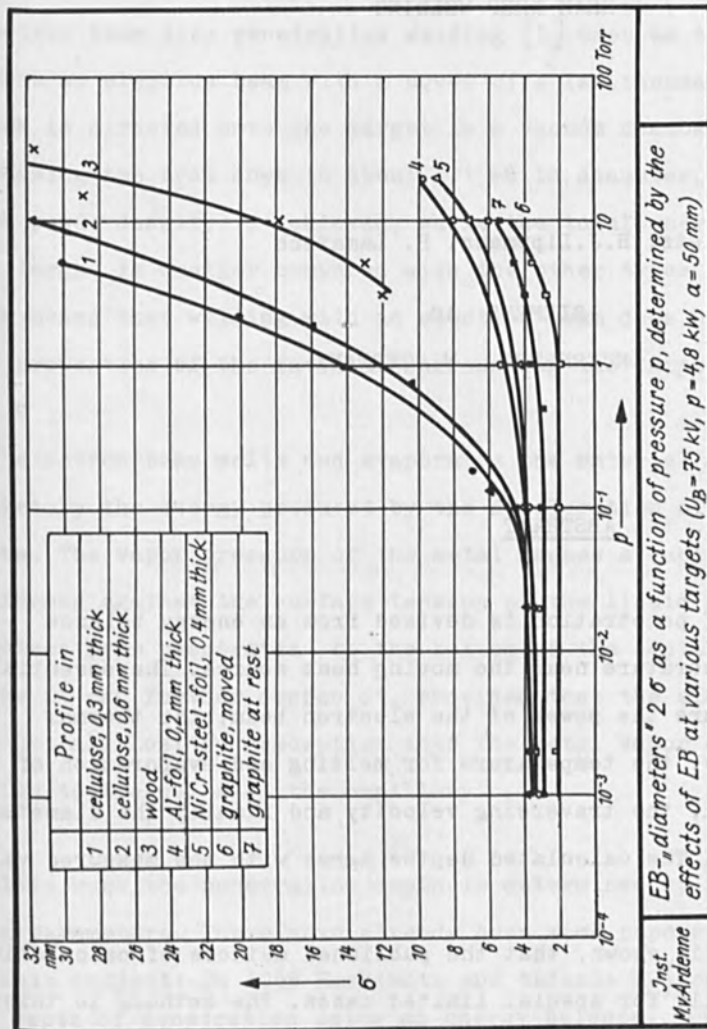


Figure 8



EB-diameters' 2σ as a function of pressure p , determined by the effects of EB at various targets ($U_B = 75$ kV, $p = 4.8$ kW, $\alpha = 50$ mm)

Figure 9

CONTRIBUTION TO THE DEPTH OF PENETRATION IN ELECTRON
BEAM DEEP WELDING

H. Dietz, H.J.Lippmann, H. Lamatsch

SIEMENS AG
NUERNBERG W.-GERMANY

ABSTRACT

The depth of penetration is devised from an energy balance and the temperature near the moving heat source. The essential parameters are the power of the electron beam, the thermal conductivity, the temperature for melting and evaporation of the material, the traversing velocity and finally the diameter of the beam. The calculated depths agree with the measured values.

Moreover it is shown, that the published methods of calculation are only valid for special limited cases. The methods in this paper will show a deeper understanding of the different methods of calculation of the depth of penetration and their validity.

1. INTRODUCTION

Electron beam deep penetration welding [1] uses as the heat source an electron beam with a power of a few thousand Watts, which is directed onto the target in a vacuum chamber. By focussing the beam down to about 0,1 mm in diameter, an extremely high power density is achieved, while the total energy put into the target is smaller compared with the other types of welding. This means that welding with an electron beam does not change the properties of the material surrounding the target.

The electron beam melts and evaporates the material at the target by the energy produced by the deceleration of the electrons. The vapor pressure of the metal causes a capillary to be formed against the surface tension of the liquid metal. The electron beam penetrates to the bottom of the capillary and there it can further deepen it, provided that the electrons are not all lost by absorption into the metal vapor or dissipation to the walls of the capillary.

In this work the penetration depth is determined by the essential parameters. There have already been some papers written on this subject: In 1965 Hashimoto and Matsuda [2] calculated the depth of penetration using an energy balance; however the results achieved theoretically have been two or three times

greater than the measured values. Beckert and Neubert [3] were able to improve the results achieved with the energy balance by considering the effect of the reflected electrons. In both works the energy balance is based upon the temperature distribution, which is only valid for large electron beam diameters, but not for the particularly important case of diameters $< 1\text{mm}$. The work of Meyer, Scheffels and Steigerwald [4] uses the correct approximation of temperature distribution for small beam diameters, but does not consider the reflected electrons. We now want to put forward a new theory of electron beam deep penetration welding, combining the important points of the above mentioned works.

2. THE ENERGY COMPONENTS OF ELECTRON BEAM WELDING

In order to facilitate the calculation, the cross-section of the electron beam will be taken to be a square of side b [2,3]. The melting-zone will also be taken as a square of side b (figure 1), and the form of this zone beneath the surface will be taken as a prism. The main assumption is that the extent of the melting-zone on the surface of the material is precisely the same as that of the electron beam. The capillary filled with metal vapor will at first be neglected. Later it will be shown why the calculation of the energy components is still valid, according to figure 1.

Let the traversing velocity of the electron beam be v , and let the energy in the prism (see fig.1) be W_e in the time $t=b/v$. This energy melts and then partially evaporates the target (components W_s and W_d), a part W_{st} is lost by radiation of heat, and finally an important part W_λ is removed by conduction of heat. The energy balance then is

$$(1) \quad W_e = W_s + W_d + W_{st} + W_\lambda$$

The electron beam is accelerated through a potential U and has a current, given by J . The energy put into the prism in fig. 1, in the time $t=b/v$, is then

$$(2) \quad W_e = UJ \cdot \frac{b}{v} \cdot \eta$$

Here η represents the efficiency of the energy transformation [3]. η gives therefore the fraction of the kinetic energy of the electron beam, which is transformed into heat in the prism of fig. 1. η is influenced by the energy component, which is lost by the reflected electrons. Of these the true secondary electrons and those elastically reflected are of little interest. We find [3]

$$(3) \quad \eta = 1 - 0.7B$$

if the small effect of X-rays is ignored. β is the fraction of the reflected to the total incident electrons. The equation is valid for a range of the potential for acceleration from 30 kV to 150 kV. For 150 kV, and iron as target material, η is found to be 0,8.

For the energy components for melting (W_s), evaporation (W_D) and for radiation of heat (W_{st}) we find

$$(4) \quad W_s = \frac{1}{2} b^2 \rho (c_p \cdot (T_L - T_\infty) + H);$$

$$(5) \quad W_D = \frac{1}{2} b^2 \rho \delta H_v$$

$$(6) \quad W_{st} = b^2 \cdot \frac{b^2}{V} \cdot \epsilon \delta_0 \cdot T_L^4$$

ρ and c_p are density and specific heat, respectively; H and H_v the latent heats of melting and vaporisation per unit mass; T_L the melting point temperature; ϵ is the density of blackness; and δ is the ratio of the evaporated mass to the molten mass. (δ_0 is the Boltzmann constant.)

One finds with such small diameters and high powers of the electron beam, that all three components of energy are very small compared with W_e . Therefore in the energy balance (1) W_λ is the important value.

3. DETERMINATION OF w_λ

The heat conducted away from the sides of the prism (area A) in the time $t=b/v$ is

$$(7) \quad w_\lambda = \lambda A \cdot \frac{b}{v} \cdot \frac{\partial T}{\partial r}$$

Area A can be calculated according to fig. 1, where the side adjacent to the molten zone is neglected. In order to determine the temperature distribution we considered the work-piece as a plate with infinite dimensions in the x,y-directions and with the thickness h in the z-direction. The plate is moved in the x-direction with a constant velocity v. The linear heat source in the plate is stationary relative to the vacuum chamber, and from $t=0$ gives a constant power P. Before $t=0$ the plate is at temperature T_∞ . The temperature T (x,y) in a point (x,y,z) of the plate, after a sufficiently long time is [5]

$$(8) \quad T(x,y) = T_\infty + \frac{P}{2\pi\lambda h} e^{\frac{yx}{2\kappa}} K_0\left(\frac{vr}{2\kappa}\right); \quad r = \sqrt{x^2 + y^2}; \quad \kappa = \frac{\lambda}{\rho c_p};$$

λ, ρ, c_p and κ are heat conductivity, density, specific heat and temperature conductivity, respectively, of the target material, K_0 is the modified Bessel-function of second kind and of order 0. For K_0 there are two different approximations

$$(9) \quad \begin{aligned} K_0 &\approx \sqrt{\frac{\pi}{2z}} e^{-z} && \text{for } z > 2 \quad (\text{Case I}) \\ K_0 &\approx \ln \frac{1}{z} && \text{for } z < 0,1 \quad (\text{Case II}) \end{aligned} \quad z = \frac{vr}{2\kappa}$$

κ for steel is in the order of $0,1 \text{ cm}^2/\text{sek}$. For example there is

$$(10) \quad \kappa = 0,1 \text{ cm}^2/\text{sek}; \begin{cases} v \geq 5 \text{ cm/sek}; r \geq 1 \text{ mm} & : z \geq 2,5 \text{ (Case I)} \\ v \leq 1 \text{ cm/sek}; r \leq 0,2 \text{ mm} & : z \leq 0,1 \text{ (Case II)} \end{cases}$$

Case I is for "high traversing velocities" and "large beam diameters"; and on the other hand case II is for "low traversing velocities" and "small beam diameters". With the approximations (9) in the temperature distribution equation (8) we find for the energy W_λ

$$(11) \quad W_\lambda = 2,5 \cdot (T_R - T_\infty) \cdot \frac{hb^2}{V} \cdot \left(\frac{v}{2\kappa} + \frac{1}{b} \right) \quad (\text{Case I})$$

$$(12) \quad W_\lambda = \frac{2,5 \cdot bP}{\pi v} \quad (\text{Case II})$$

Here T_R means the temperature for $r=b/2$, at the boundary of the melting zone. This will be explained later.

4. CALCULATION OF THE PENETRATION DEPTH IN CASE I

In the energy balance (1), the components W_s , W_D and W_λ are proportional to h , according to equations (4), (5) and (11). By neglecting the very small effect of the radiation of heat, we have [3]

$$(13) \quad h = \frac{UJ\eta}{2.5 (T_L - T_\infty) \cdot \left\{ 1 + 2z \cdot \left(1.4 + \frac{H + \delta H_v}{2.5 c_p (T_L - T_\infty)} \right) \right\}}; \quad z = \frac{yb}{4\kappa} > 2$$

$z > 2$ means relatively large beam diameters. Conversely one can ignore the small capillary of metal vapor. Therefore T_R is here the melting-point temperature T_L . We see that h is proportional to UJ .

5. CALCULATION OF THE PENETRATION DEPTH IN CASE II

Since the beam diameters according to equation (10) are very small, the components W_s , W_D and W_{st} in the energy balance (1) can be neglected. However W_λ does not depend on h , according to equation (12), and so we cannot calculate the penetration depth from the energy balance. From the temperature distribution (8) we get, with the approximation (9), in case II,

$$(14) \quad h = \frac{UJ}{2\pi\lambda (T_s - T_\infty)} \cdot \ln \frac{1}{z}; \quad z = \frac{yb}{4\kappa} < 0.1$$

With the small beam diameters of case II we identify here b with the diameter of the capillary of metal vapor, and therefore T_R is the temperature of vaporisation T_s of the target material. Again h is proportional to UJ .

6. EXAMPLE: WELDING OF WORK PIECES OF HIGH-TEMPERATURE STEEL

$$(15) \quad \left\{ \begin{array}{l} UJ = 1000 \text{ VA} ; \eta = 0,8; \\ \lambda = 0,12 \text{ cal/cm} \cdot \text{sek} \cdot \text{Grad}; \rho = 7,8 \text{ g/cm}^3 ; \\ c_p = 0,115 \text{ cal/g Grad}; K = 0,134 \text{ cm}^2/\text{sek}; \\ H = 65,5 \text{ cal/g}; H_v = 1600 \text{ cal/g} ; \\ T_L - T_\infty = 1500^\circ\text{C}; T_s - T_\infty = 2800^\circ\text{C} ; \delta = 2,5 \cdot 10^{-2} ; \end{array} \right.$$

We find from (13) and (14)

$$(16) \quad \left\{ \begin{array}{l} h = \frac{4,24 \text{ mm}}{1 + 3,29 z} ; \quad z = \frac{vb}{4K} > 2 ; \\ h = 0,903 \text{ mm} \cdot \ln \frac{1}{z} ; \quad z = \frac{vb}{4K} < 0,1 ; \end{array} \right.$$

The two functions are shown in fig. 2. The dotted line indicates the range $0,1 < z < 2$. The value z is characteristic for both cases.

The comparison between the measured and calculated values of h in three tests gives the following table:

UJ	v	b	z	h (measured)	h (calculated)
$8,1 \cdot 10^3 \text{ VA}$	1,5mm/sek	0,1mm	$2,8 \cdot 10^{-3}$	52 mm	48,3 mm
$7,5 \cdot 10^3 \text{ VA}$	2 mm/sek	0,1mm	$3,7 \cdot 10^{-3}$	40 mm	37,5 mm
$5,4 \cdot 10^3 \text{ VA}$	2 mm/sek	0,1mm	$3,7 \cdot 10^{-3}$	30 mm	27 mm

A more refined consideration of this subject would have to distinguish between the electron beam diameter, the melting zone diameter and also that of the capillary of metal vapor. In order to do this one would have to know the energy distribution of the cross-section of the beam, and also have a precise concept of the "temperature T_R of the boundary".

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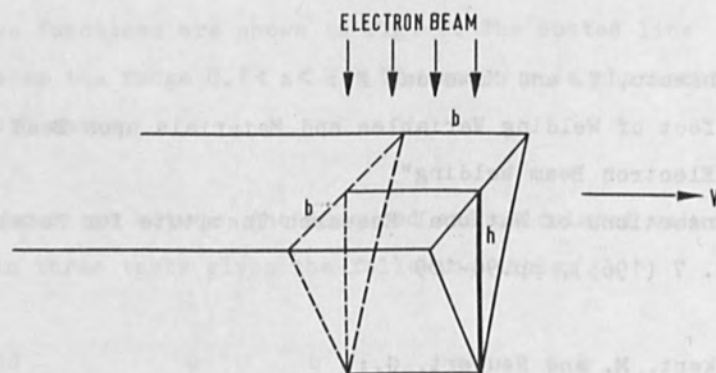


Figure 1

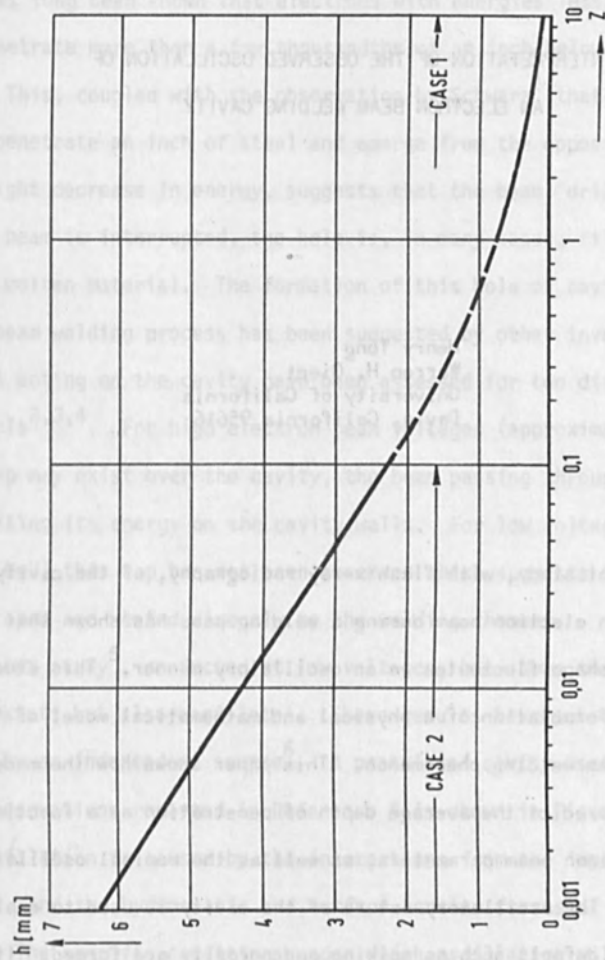


Figure 2

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A Study of the Dynamics and the Energy Balance of the
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INTERPRETATION OF THE OBSERVED OSCILLATION OF AN ELECTRON BEAM WELDING CAVITY

[5] Taylor and Taylor

"Formulation of Heat in Welding"
1965

Henry Tong
Warren H. Giedt
University of California
Davis, California 95616

Abstract:

A recent study, with flash x-ray radiography, of the cavity formed by an electron beam during a welding pass has shown that the cavity shape fluctuates in an oscillatory manner. This study led to the formulation of a physical and mathematical model of the electron beam welding phenomenon. This paper shows how the model is used to predict the average depth of penetration as a function of the electron beam parameters, as well as the nominal oscillation frequency. The oscillatory nature of the cavity is used to explain how welding defects such as spiking and porosity are formed. It is anticipated that this new model will lead to techniques which will minimize these defects.

I INTRODUCTION

It has long been known that electrons with energies less than 150 keV cannot penetrate more than a few thousandths of an inch below a metal surface. This, coupled with the observation by Schwarz¹ that the electron beam can penetrate an inch of steel and emerge from the opposite side with only a slight decrease in energy, suggests that the beam "drills" a hole. After the beam is interrupted, the hole is, in many cases, filled by an inflow of molten material. The formation of this hole or cavity during an electron beam welding process has been suggested by other investigators². The forces acting on the cavity have been assessed for two different proposed models^{2,3,4}. For high electron beam voltages (approximately 150 keV) a small cap may exist over the cavity, the beam passing through this cap and depositing its energy on the cavity walls. For low voltages (approximately 35 keV) this cap has not been observed which suggests that the cavity is open and vented directly to the welding chamber.

A recent study⁵, conducted at low voltages, has shown that the cavity not only exists but also oscillates. These results have subsequently been confirmed by an independent source⁶. A postulated cavity oscillation sequence for the observations reported in Reference 4 is shown in Figure 1. The cavity oscillation is caused by the interactions from the impulse of vaporizing metal, and the hydrostatic and surface tension forces of the molten material. The specific conditions under which oscillation will occur are not clear but undoubtedly are related to the electron beam power density and possibly to the voltage. The objective of what follows will be to present

a qualitative interpretation of the observed cavity oscillation phenomena. This will, hopefully, provide some insight about the conditions for which the degree of oscillation will have unacceptable effects on the fusion zone.

II Cavity Oscillation

As shown in Figure 1 the dimensions of the cavity fluctuate between some minimum and maximum depth. The interface between the molten fluid and the virgin metal, though, oscillates to a much lesser degree. Nevertheless this oscillation of the interface will result in a fusion zone which will exhibit an irregularity at its base as shown in Figure 2. This irregularity may be a slight ripple as in 2a or a more pronounced spiking as in 2b. The degree of spiking is determined by the welding parameters, the properties of the fusion material and possibly its homogeneity.

The forces that act on the cavity walls have been discussed by other authors and one can conclude that the predominant forces are a) impulse force due to the evaporation, b) surface tension forces, c) the hydrostatic forces of the molten fluid, and d) the reaction forces arising from the motion of the cavity through the fluid. The electron pressure can be shown to be negligibly small.

We turn now to the formulation of a physical model⁷ which will incorporate the observed oscillation features of the cavity. A great deal of insight into the phenomena can be gained by comparing the sequence of cavity shapes shown in Figure 1 with the cavities formed by the entry of small spheres into water (see, for instance, references 8 and 9). The nature of the latter cavities depends on the entry velocity and the diameter of the sphere. The four different cases which have been observed are illustrated in Figure 3. In each case the period immediately after entry is characterized by the formation of a surface cavity and a spray of water above the surface. In the first case (associated with low entry velocities and large

diameters), the surface cavity is eventually separated, by a deep closure, into a surface cavity and a subsurface cavity. The time of closure is dependent upon the entry conditions and the characteristics of the fluid. In the second case, no closure occurs so that only the surface cavity is observed. In the third case a different kind of closure is observed, namely a surface closure. A surface closure is associated with high entry velocities and small diameter spheres. This occurs when the surface tension of the fluid in the spray is sufficient to pull the spray in radially causing the cavity to be sealed off and a bubble to form at the surface of the fluid. The last case is a combination of surface closure and subsurface closure; the former invariably occurs first so that the initial cavity is separated into two subsurface cavities. Whenever a deep closure occurs a hydraulic jet is observed to emanate from the point of closure. In the case of a surface closure followed by a deep closure the hydraulic jet from the deep closure has often been observed to puncture the surface bubble. The referenced water entry studies have shown that a surface closure is most likely to occur for small spheres entering at high velocities.

The above phenomena can be compared with the electron beam welding phenomena since the reaction effect of the evaporation process effectively forms a body (the cavity) which penetrates into the fluid (the molten metal). Thus one has a physical model for the electron beam cavity penetration process.

At equilibrium the momentum transfer due to evaporation is equal to the vapor pressure. If evaporation occurs into a complete vacuum the momentum transfer is equal to $1/2$ of the vapor pressure. Since a pressure buildup in the cavity is necessary in order for the cavity to exist, the pressure in the cavity will be between 0.5 and 1.0 of the vapor pressure as determined for the cavity surface temperature. In the water entry phenomena, the driving force for penetration of the sphere was the inertia of the sphere.

The electron beam cavity is essentially massless so that its effective driving force is the vapor pressure times the cross-sectional area on which it acts. It should be noted that, although the bubble formed by a surface closure may be thin enough for the beam to penetrate, a deep closure blocks the beam from the lower cavity, hence removing the driving force.

Here, then is a possible explanation for the fundamental difference between high and low voltage welding. High welding voltages are characterized by small beam diameters and high power densities. In terms of the analog, this is represented by small spheres entering water at high velocities. These conditions enhance the formation of a high spray and its subsequent contraction which may cause a surface closure. It is apparently this surface closure which was observed in Reference 4.¹ It is believed, though, that the bursting of the electron beam welding cap is caused by the hydraulic jet from a deep closure.

In any event the retarding forces for both the sphere and the electron beam cavity penetration are the hydrostatic forces, surface tension forces and the "drag" due to the motion of the cavity through the fluid. These forces are shown in Figure 4 acting on the base of the cavity. Since the vapor pressure is a function of the surface temperature which in turn is determined to large degree by the electron beam power density and the welding speed, one would naturally expect that electron beam power and a characteristic dimension of the cavity or electron beam would be important variables. Naturally, material properties would also be important. As noted by Schwarz¹, the cavity penetration speed is exceptionally fast when compared with practical transverse speeds so that insofar as the penetration time is concerned, the welding speed is not of significance. Then applying Newton's second law to a hypothetical massless body (which represents the cavity) penetrating into a fluid, the following differential equation is

obtained. *

$$\left[\frac{1}{2} \left(\frac{A_1}{A_2} \right) \left(1 + \frac{A_1}{A_2} \right) \right] \rho \left(\frac{dx}{dt} \right)^2 + \rho g x = P_{ex} \quad (1)$$

$$\text{with } P_{ex} = P_v - \frac{2\sigma_s}{a}$$

where A_1 = cross-sectional area of cavity

A_2 = cross-sectional area of fluid around cavity

ρ = density of fluid

x = distance from top surface of workpiece

g = gravitational constant

P_v = vapor pressure

σ_s = surface tension

a = beam or cavity radius

Equation (1) can be readily integrated to yield

$$T = 2 \left[\frac{f(A_1 A_2) h}{g} \right]^{1/2} \left[\left(\frac{P_{ex}}{\rho g h} \right)^{1/2} - \left(\frac{P_{ex}}{\rho g h} - 1 \right)^{1/2} \right] \quad (2)$$

$$\text{where } f(A_1/A_2) = \frac{1}{2} \left(\frac{A_1}{A_2} \right) \left(1 + \frac{A_1}{A_2} \right) \quad (3)$$

The variable T is the time required for the cavity to penetrate to the mean depth of the fusion zone h . After the cavity reaches the depth h , a closure occurs; i.e., the cavity fills up. The inertia of the fluid motion caused by closure causes a delay between when closure is initiated and when the cavity can once again penetrate. Although the penetration time can be

* The details of the derivation of this equation and its solution are presented in Reference 7.

calculated the closure time cannot. Because of the symmetry frequently associated with oscillatory phenomena, it is reasonable to surmise that they are of the same order of magnitude. Finally, experimental evidence shows that A_1 is of the same order of magnitude as A_2 so that

$$f \left(\frac{A_1}{A_2} \right) \approx 0 \quad (1)$$

Thus if equation (2) is used to calculate the penetration time, the oscillation frequency will be given by

$$\text{Hz} \approx \frac{1}{2T}$$

The above oscillation frequency is a uniform oscillation. The oscillation or spiking patterns observed in fusion zones will have this frequency and it may be modulated by other resonant frequencies with occasional irregularities.

III Thermal Model of Process

A second model, based on energy considerations will now be presented. Due to the oscillation of the cavity, the electron beam energy is alternately deposited deep in the workpiece or near the surface of the workpiece as shown in Figure 5. Thus, any analysis of the energy transfer characteristics which depends on an energy source, be it a surface or a line source, is not consistent with the observed physical phenomena. Even though the cavity is oscillating between wide limits, it is expected that the variation of the melt-solid interface will oscillate within much smaller limits. This interface should then represent a better boundary condition for a quasi-steady analysis since it is a reasonably stable boundary and its temperature is fixed. To

idealize the problem, it shall be assumed that the weld zone has a large depth-to-width ratio so that energy transfer in a direction parallel to the incident electron beam is negligible. For tractability, it shall be assumed that the radius of the fixed temperature boundary is constant although this is not a necessary assumption. The energy transfer mechanisms are than as indicated in Figure 6. It has been show^{10,11} that energy losses due to radiation and evaporation are small so that they can be ignored. Then considering only the forward half of the welding zone, the energy transfer model is as defined in Figure 7. The input energy comes from the electron beam and is some fraction ϵ of its total power; the energy losses are due to thermal conduction in the forward direction and the convection of fluid in the reverse direction. For constant average material properties, conservation of energy yields

$$\epsilon \left(\frac{P}{4.186} \right) = 2\rho Uah \left[\frac{c_p}{2} (T_s - T_m) + H_{sf} \right] + Q \text{ (conduction)}$$

where P = beam power

U = welding speed

h = average depth of fusion

c_p = specific heat capacity

T_s = surface temperature of melt layer

T_m = equilibrium melting temperature

H_{sf} = heat of fusion

The heat conduction term can be obtained from the solution of a constant temperature cylinder moving through a solid¹². The complete energy solution then becomes¹³.

$$\left(\frac{4.186}{\epsilon P}\right) h = \left\{ S \left[2k(T_s - T_m) + 4\rho a H_{sf} \right] + k(T_m - T_\infty) \left[\int_0^{\pi/2} D(\theta, S) d\theta \right] \right\}^{-1} \quad (4)$$

with

$$D(\theta, A) = e^{-S \cos \theta} \left\{ S \cos \theta (1 - \cos \theta) I_1(s) + \frac{1}{K_0(S)} + 2 \sum_{n=1}^{\infty} \frac{\cos n \theta}{K_n(S)} \right\} \quad (5)$$

$$S = \frac{y_a}{2\alpha}$$

where α = thermal diffusivity

k = thermal conductivity

I_n, K_n = Bessel functions of first and second kind.

From equation (5) it is observed that the integral in equation (4) is a universal function of the parameter S . Equation (4) relates the average depth of penetration to the beam power, S , and material properties. The surface temperature T_s is approximated through the use of available, surface tension and vapor pressure data, and from this, the typical curves shown in Figure 3 were calculated.

IV Discussion

From the preceding development, it was not intended to suggest that the electron beam welding cavity oscillates for all welding conditions. Indeed for shallow welds, it is expected that the surface tension forces will be sufficient to maintain a relatively stable cavity. Likewise one should not expect that closure results in a complete filling of the cavity. It seems almost impossible that the fluid in a 6 to 9 inch deep weld with a width of only 1/8 inch could oscillate the full depth of the weld. In this case if there is any oscillation it is probably restricted to the base region of the weld.

What can be stated with certainty is that for moderately deep welds, say about 1 inch, the cavity does oscillate and it is this oscillation that is responsible for some of the base region defects which have been observed in the fusion zone. In particular, it can be observed from Figure 1 that the gradual recession of the melt-solid interface with each oscillation of the cavity results in a ripple at the base of the weld. The function P_{ex} increases as the power density is increased so that from equation (2) it is observed that the penetration time decreases or alternatively the spiking frequency increases. Thus for any given welding speed and average depth of penetration the frequency increases with power density. To reduce spiking, one of two conditions seem highly desirable--either a steady state cavity or a very large oscillation frequency. The former would be most probable for low power densities and wide welds and the latter for high power densities with very narrow welds. Somewhere in between a maximum degree of spiking occurs and this operating region, which has not been quantitatively defined, should be avoided.

For high welding speeds and attendently high freezing rates at the back side of the melt, porosity and cold shuts are likely to be formed. The porosity arises from the turbulent motion of the fluid, due to the oscillation of the cavity, which entrains gas bubbles. The high freezing rates trap these bubbles before they can "float" to the surface. Similarly, from Figure 1, one can see that at some period during the cavity oscillation the electron beam cannot, because of a high cooling rate, keep the lower wall of the cavity molten. Then when closure is completed a cold joint may be formed.

The vapor pressure is an important parameter affecting both the oscillation frequency and the depth of penetration, the latter in an indirect fashion by affecting the surface temperature of the melt layer. The curves shown in Figure 8 were obtained using the vapor pressure of the predominant species and the difference in welding depth for the various types of aluminum are due to thermal property changes. If the alloying elements have a vapor pressure which is substantially greater than the primary material, some adjustment should be made. To appreciate the magnitude of the effect of vapor pressure the curves shown in Figure 9 were calculated. The thermal properties were fixed and only the vapor pressure was changed by varying the amount of magnesium alloyed with aluminum. At any given temperature, magnesium has a vapor pressure which is several orders of magnitude greater than that of aluminum so that small percentages of it have a significant effect on the net vapor pressure. The net vapor pressure was calculated using Raoult's rule¹⁴. It can be seen from equation 4 that the maximum depth of penetration will occur for $T_s = T_m$. This would be the case for an infinite vapor pressure or direct sublimation. The relative effect of magnesium content in aluminum is most notable at large values of Ua approaching a negligible

magnitude at small values of U_a .

In order to obtain a fusion zone with minimal irregularities of the root one can make multiple passes so that the spikes overlap. Alternatively, since the spiking frequency is, for our approximations, independent of the welding speed and is primarily a function of depth of penetration, low welding speeds will tend to compress the spikes together to form a smooth root. Finally, a low power density beam will have a wider cavity so that each spike will be wider and again the overlap will result in a smoother root. The last two conditions are really related since lower power densities would require lower welding speeds to obtain the same average depth of penetration as can be obtained with a high power density beam. Low power densities have the disadvantage of depositing a larger total power into the workpiece and increasing the possibility of thermal distortion so that some compromise may be necessary.

V Summary and Conclusions

A qualitative discussion of the oscillatory nature of an electron beam welding cavity based on models for the metal flow and energy flow was presented. Analyses of the hydrodynamics of the fluid oscillation and the energy transfer characteristics were reviewed. From the results of these analyses, it was noted that the oscillation frequency and the average depth of fusion could be calculated. It was shown that the oscillations are the primary cause of the observed spiking phenomena and of porosity in the fusion zone. It was concluded that low electron beam power densities and low welding speeds should reduce the severity of the spiking although this will also result in a greater degree of thermal distortion. Further studies will be necessary in order to define in an unambiguous manner,

quantitative measures of the degree or severity of spiking. Studies need also be conducted to determine welding procedures that will reduce the degree of spiking to acceptable limits.

Acknowledgment

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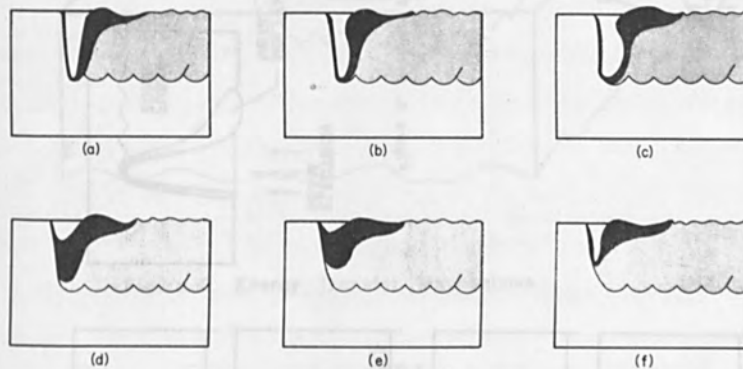


Figure 1. Cavity Oscillation Sequence for 1100 Aluminum

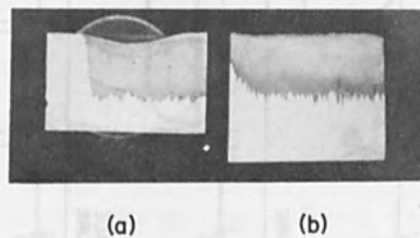


Figure 2. Typical Spiking

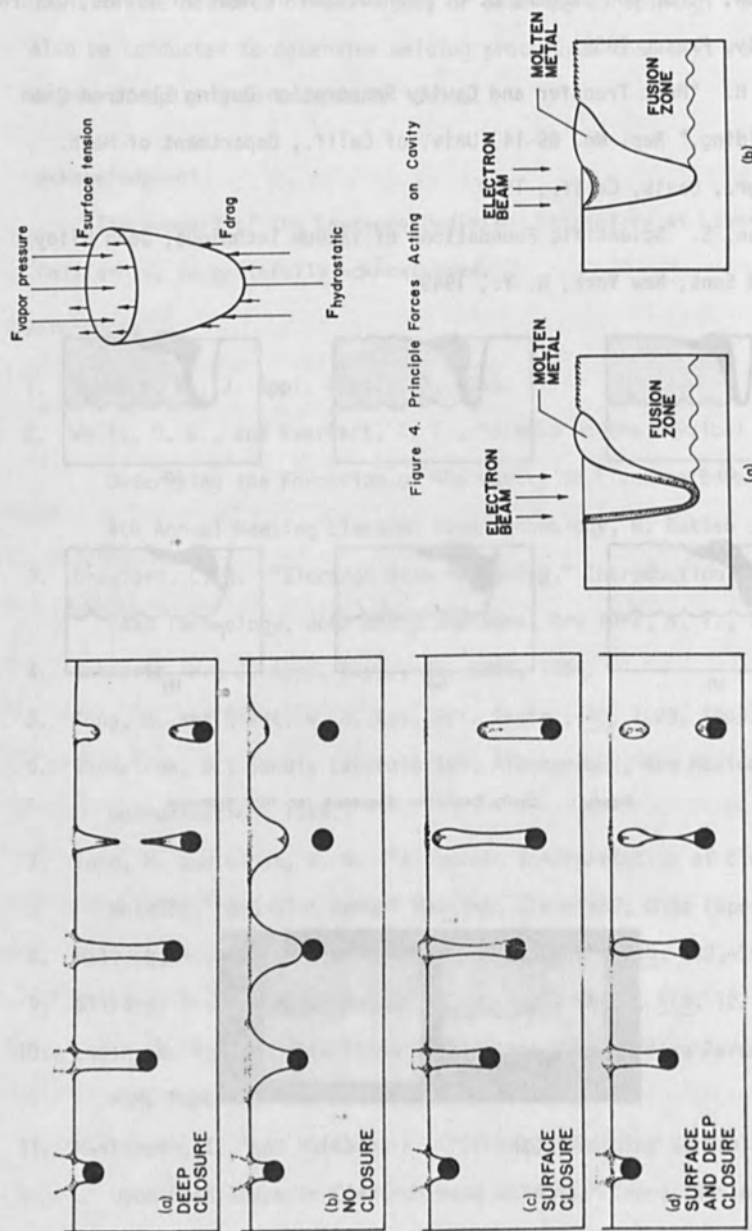


Figure 4. Principle Forces Acting on Cavity

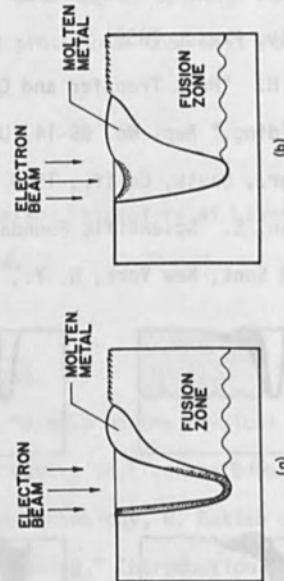


Figure 5. Electron Beam Energy Deposition

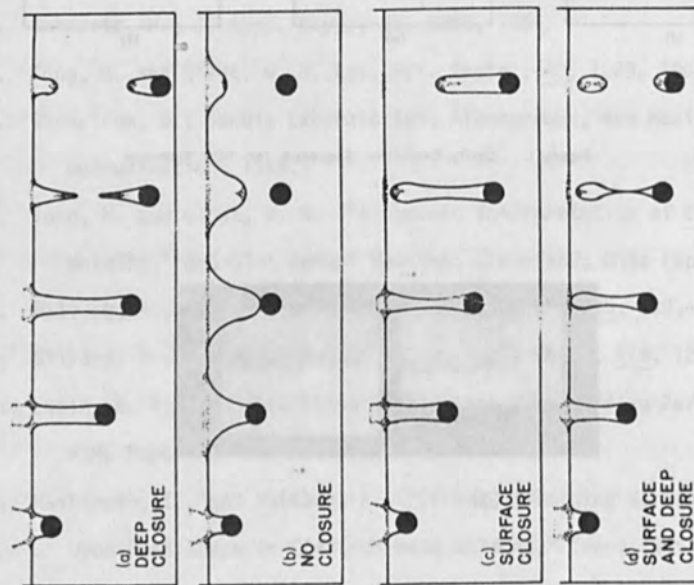


Figure 3. Cavities Formed by Entry of Projectiles into Water

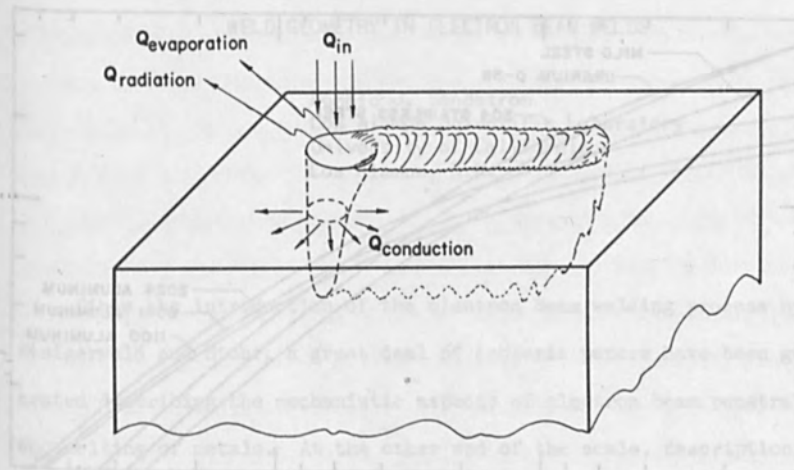


Figure 6. Energy Transfer Mechanisms

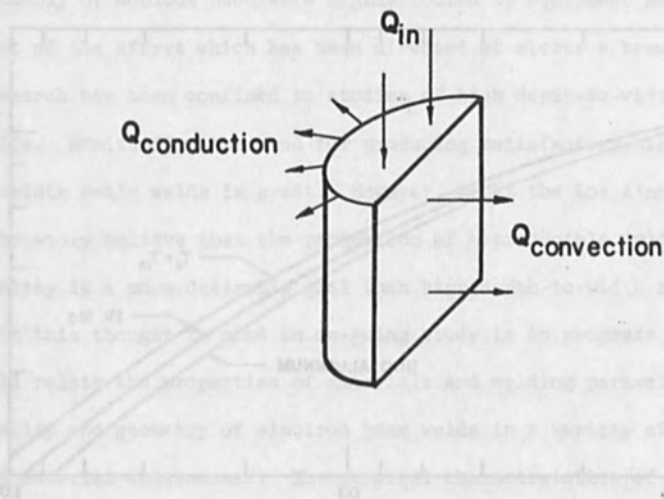


Figure 7. Energy Transfer Model

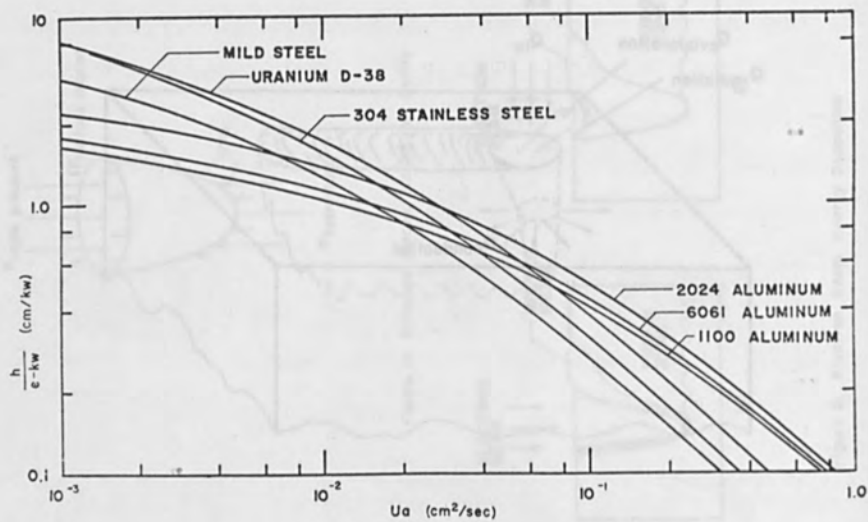


FIGURE 8. THEORETICAL DEPTH OF PENETRATION

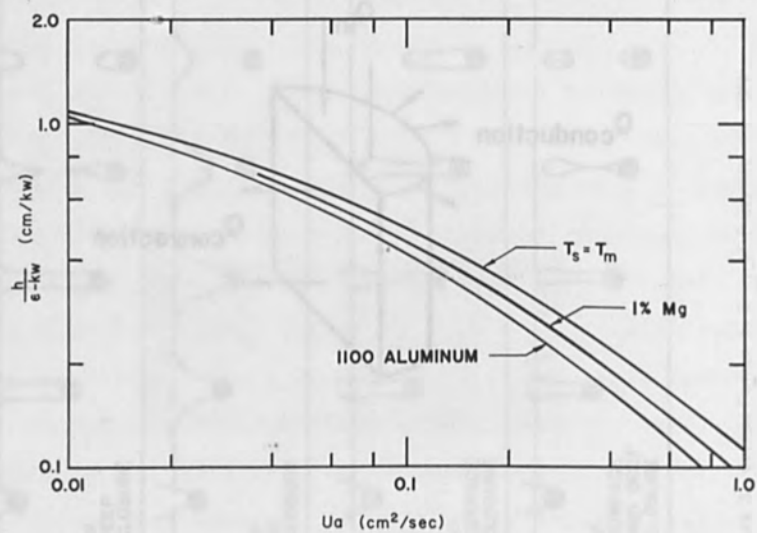


Figure 9. Effect of Vapor Pressure

THE RELATIONSHIP OF BEAM PARAMETERS TO
WELD GEOMETRY IN ELECTRON BEAM WELDS

Donald J. Sandstrom
Los Alamos Scientific Laboratory
University of California
Los Alamos, N.M.

Since the introduction of the electron beam welding process by Steigerwald and Stohr, a great deal of esoteric papers have been generated describing the mechanistic aspects of electron beam penetration and melting of metals. At the other end of the scale, descriptions of such bizarre possibilities as joining beryllium to tungsten with 100% joint efficiency and penetrating stacks of box beams to produce an assembly of dubious need were highly touted by equipment manufacturers. Most of the effort which has been directed at electron beam welding research has been confined to studies of high depth-to-width ratio welds. Admittedly, the need for producing satisfactory high depth-to-width ratio welds is great. However, we at the Los Alamos Scientific Laboratory believe that the production of reproducible welds of high quality is a more desirable goal than high depth-to-width ratios per se. With this thought in mind an on-going study is in progress which hopefully will relate the properties of materials and welding parameters to the quality and geometry of electron beam welds in a variety of materials and material thicknesses. The physical characteristics of electron beam welds cannot be completely described in terms of the usual beam parameters of current, voltage, and travel speed. A fourth parameter -

beam spot size - must be defined in order to characterize the geometry of electron beam welds. This fourth parameter allows one to determine the power density in a weld and from this determination it allows one to make some prediction as to weld geometry. **

Figure 1 (702835) consists of a schematic representation of the processes which occur when a material is electron beam welded. From the standpoint of welding, there is little or no concern for the x-rays which are produced by the impinging electron beam. The point of vital interest is the amount of energy actually dissipated in the work piece. The back scattered electrons will not be recoverable from the standpoint of measurement, but from the standpoint of energy transfer must be assumed to contribute all of their energy to the work piece. The secondary electrons will, of course, be recoverable by biasing the work with a voltage on the order of 100 volts. The impinging electron beam will produce localized melting, transformation within heat affected zones, and actual ionization of some of the material being welded.

Instrumentation has been developed for use in probing the electron beam in order to optimize the focusing characteristics and to measure the beam size and density. Figure 2 (697712) consists of a schematic of the device which has been developed at Los Alamos for this measurement. An aluminum vane which contains various diameters of W wires at its extremities is rotated through the electron beam. The rotation of this vane is accomplished by the 1800 rpm synchronous motor. The aluminum vane does not intersect the beam, but the W wires which are at the vane ends do. (The diameter of the W wire should be kept small in order to obtain accurate measurements.) The current that is picked

up by the wires intersecting the beam is sampled and applied to an oscilloscope. A Faraday Cup is located below the rotating wire scanner and the total beam current is collected in this cup and in this manner a check on the accuracy of the meters on the welding machine is made. The collector plates, in between which the vane rotates, were set at the same potential as the vane in order to collect the back scattered and secondary electrons.

The signal which is picked up by the probe is shown in Figure 3 (703749). This oscilloscope trace was made on a beam sharply focused at a gun-to-work distance of 1.25 inches with beam power of 20 kV at 60 mA. The larger trace is the signal "picked-up" by a 2 mil diameter W wire, whereas the smaller signal represents the signal measured by a 1 mil diameter wire. The diameter of the beam can be readily calculated from this trace and for these parameters the beam diameter is approximately 15 mils.

Figure 4 (703747) consists of a plot of beam current versus beam diameter for various voltages. The data plotted on these curves shows that the beam diameter is linearly dependent upon the beam current for constant voltage. As would be expected, the beam diameter increases with increasing current probably due to charge repulsion effects.

Figure 5 (6917129) is a plot of the beam diameter versus the accelerating voltage for constant current welds. This dependence is obviously exponential in form. The plots shown in the two figures demonstrate how the beam diameter, as measured, is affected by the two important parameters of accelerating voltage and beam current. Schwarz¹ has derived an equation for beam diameter based upon electron optics

theory. This equation is of the form that the beam diameter is proportional to the current divided by the voltage all to the $3/8$ power. This equation indicates that the beam diameter is exponentially dependent upon both beam current and voltage. The data generated at Los Alamos does not yield an equation of the form shown by Schwarz, but rather of the form shown in Figure 6 (705517). This equation shows linear dependence of beam diameter with current and a reciprocal exponential relationship with the accelerating voltage.

Figure 7 (6917242) is a drawing of the stigmatism produced in a beam at 10 kV and 40 mA which has been sharply focused at 1.25 inches. Changing the gun-to-work distance to 1.0 inches or 1.5 inches resulted in the change in stigmatism which was shown. The point of this figure is to show that the symmetry of a beam generated in a Pierce type gun with a neutral field cathode is quite good.

The effect of variations in weld parameters on the characteristics of welds has been investigated using a flash radiographic technique which has been described by Tong. Figure 8 (6911793) consists of a photograph of the Field Emission Corporation Flash X-ray unit which has been "married" to the 7.5 kW Sciaky Electron Beam Welder at Los Alamos. Experiments were performed on two different types of materials using power levels which yielded two different modes of melting in the material. Figure 9 (6912370) is an artists sketch which was produced from the flash radiographs produced on an aluminum bar which was welded in the high power density mode. This work substantiated the "cavity penetration" mechanism proposed by Tong and co-workers.² The variations in cavity characteristics show the instability of the cavity in various portions of the weld. The area designated No. 1

shows complete cavity collapse with a large root defect. The other areas show various degrees of cavity instability.

A technique which has been used for evaluating the characteristics of penetration in welds is shown in Figure 10 (703748). This technique involves the static radiography of a welded plate to yield the root characteristics and penetration developed in the weld.

Using this technique in combination with conventional metallographic techniques can give a reasonably accurate representation of welds in both the longitudinal and transverse directions. Figure 11 (700289) shows the longitudinal and transverse geometries developed in welds which were made in stainless steel. These welds were all made at the same energy input per linear inch of weld, but at radically different levels of power density as measured by beam diameter measurements. This figure graphically shows the meaninglessness of describing electron beam welds in terms of joules per inch of weld.

Hashimoto and Coworkers³ and Henry Tong⁴ have developed equations relating the physical properties of various metals to their welding characteristics, especially as characterized by depth of penetration measurements. The work which we have done has attempted to solve Hashimoto's equation for the materials which we have considered. Figure 12 (700179) is a tabulation of the physical properties assumed to be important for electron beam welding. The equation which has been evaluated is this equation which was developed by Hashimoto and is shown in the figure. This equation has been solved for the penetration which should be obtained using the known values for voltage, current, and beam diameter. The calculated penetration was then plotted against the measured penetration and the results are shown in Figure 13

(703746). The results obtained for the aluminum are in good agreement with those predicted by the equation, however, for the materials with lower thermal conductivity the calculated penetration exceeds the measured penetration whereas for the higher conductivity material the calculated penetration is less than the measured penetration. In all fairness, it must be pointed out that the values of lower penetration were produced using low power density welding techniques which were described earlier.

Conclusions:

A number of conclusions have been drawn from the work which has been performed at Los Alamos both as a result of the experiments previously described and from experience with the fabrication of piece parts. A summary of these conclusions is shown as follows:

1. For thin gauge material (0.030 in. or less) minimal drop-through and weld spatter is obtained using low voltage-high current welding techniques. These welding parameters automatically establish broad focus low power density welding condition.
2. The problem of root porosity and cold shuts is aggravated by high depth-to-width ratio welds in materials of high thermal conductivity. High power density appears to aggravate the problems of "spiking" due to the cavity formation inherent to the process.
3. Precise penetration and control of root characteristics in partial penetration weldments of thicker materials can be best achieved using multipass procedures in which the power density is kept low.
4. The mechanism of melting in electron beam welds appears to be strongly dependent upon the power density of the electron beam. High power density welds produce melting by the cavity penetration

technique and low power density welds appear to produce melting by a conduction mechanism.

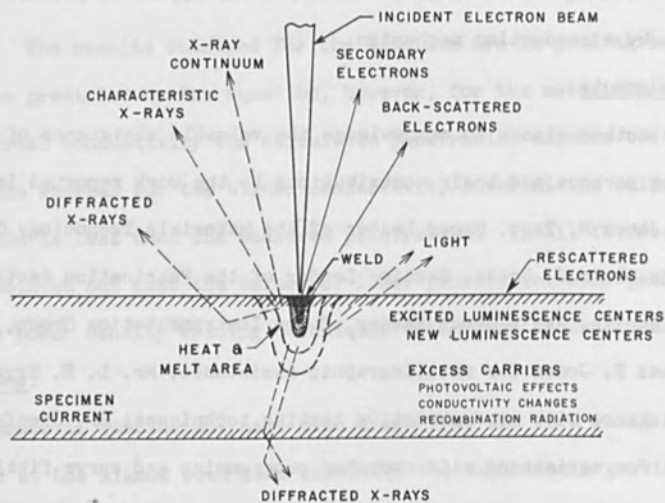
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PHYSICAL PROCESSES THAT OCCUR
DURING ELECTRON BEAM WELDING

Figure 1

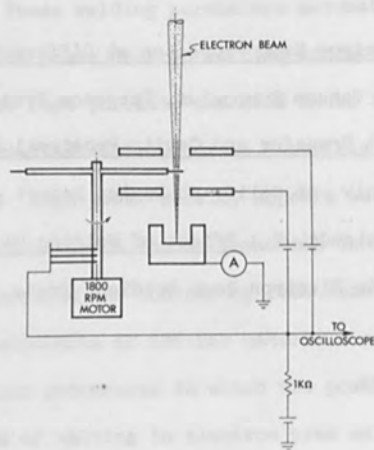
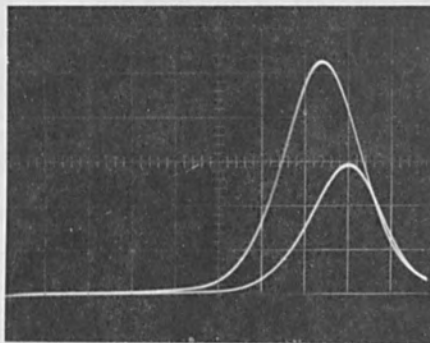


Figure 2



OSCILLOSCOPE TRACE OF A 20kV, 60mA ELECTRON BEAM AT SHARP FOCUS.

VERTICLE SENSITIVITY = 2 mA / cm

HORIZONTAL SWEEP SPEED = $5\mu\text{sec/cm}$

Figure 3

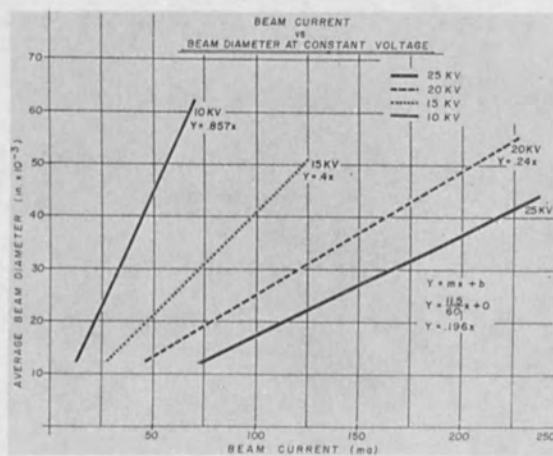


Figure 4

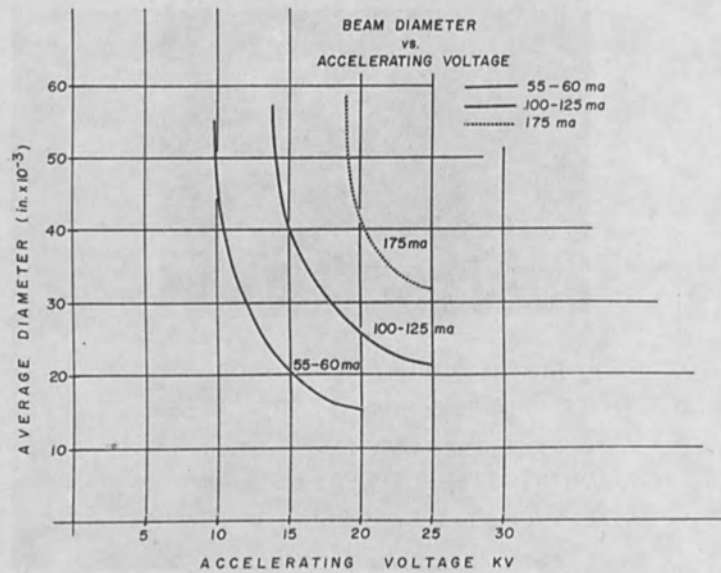


Figure 5

AN APPROXIMATION FOR THE BEAM DIAMETER
AS DETERMINED AT L.A.S.L.

$$d \approx k \frac{i - i_0}{v^a}$$

k = CONSTANT

WHERE :

- i = beam current in ma
- i₀ = beam current at the intercept
- v = accelerating voltage
- d = diameter in $\frac{1}{1000}$ inch.

Figure 6

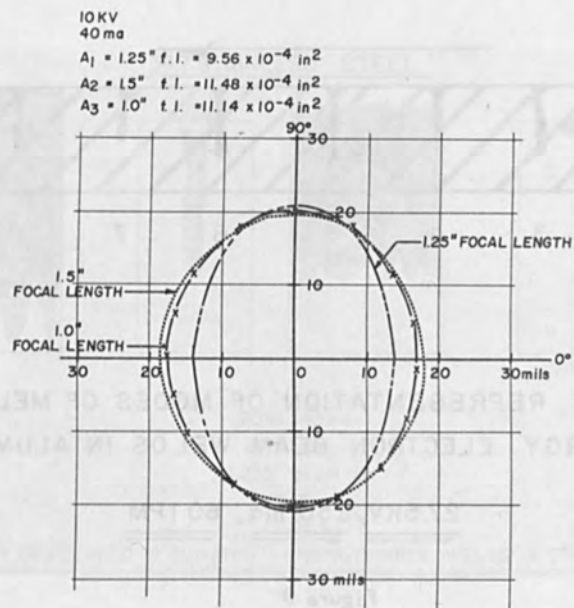


Figure 7

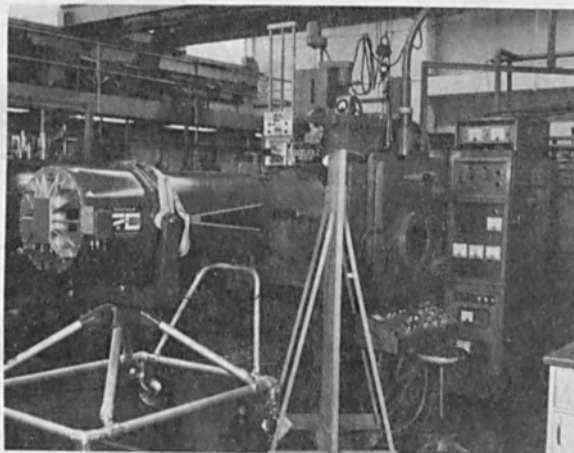
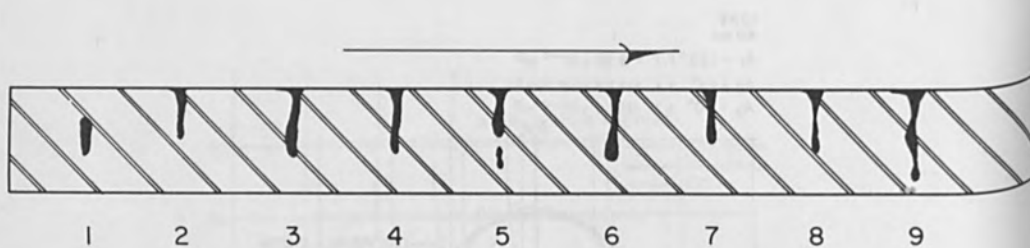


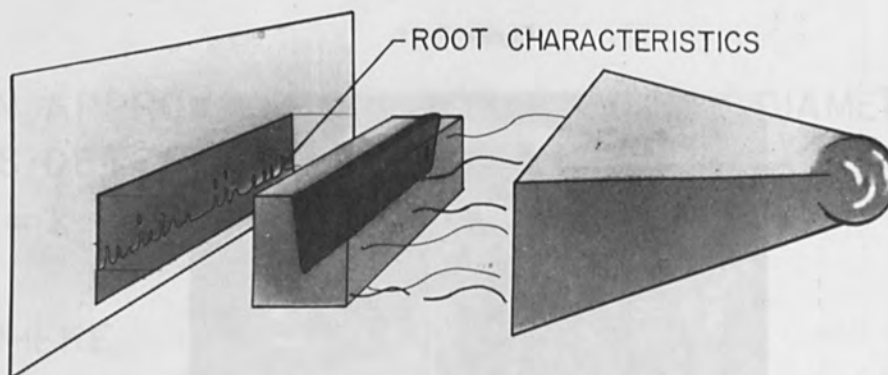
Figure 8



SCHEMATIC REPRESENTATION OF MODES OF MELTING IN
HIGH ENERGY ELECTRON BEAM WELDS IN ALUMINUM

27.5KV, 250 ma, 60 IPM

Figure 9



STATIC RADIOGRAPHIC TECHNIQUE FOR EVALUATING
PENETRATION AND ROOT CHARACTERISTICS OF E.B.
WELDS

Figure 10

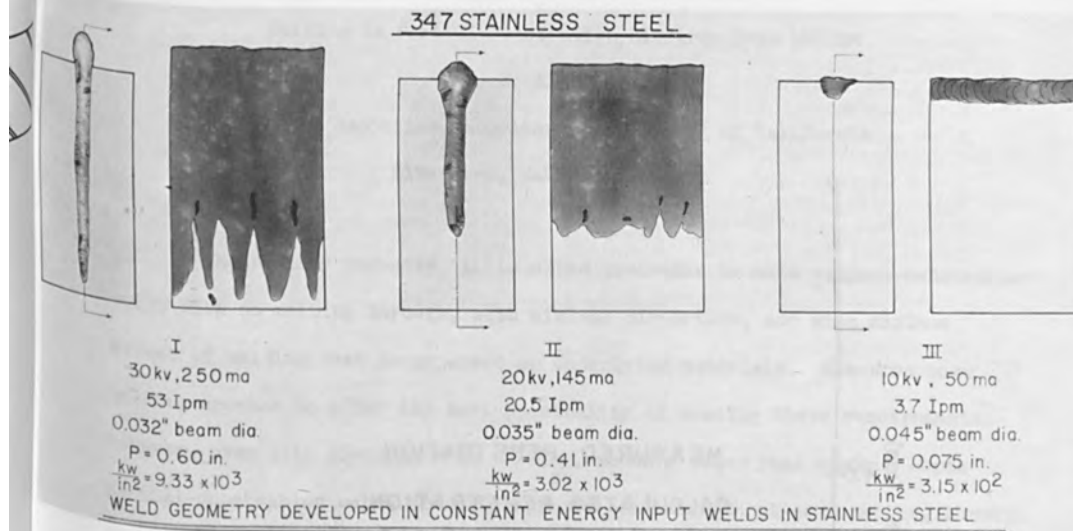


Figure 11

PHYSICAL CONSTANTS FOR SOME METALS

METAL	C (cal/g°C)	ρ (g/cc)	K (cal/°C sec cm)		$\alpha = K_m \rho / C_p$	T_m °C	H (cal/cc)	$C_p T_m + H$ (cal/cc)	$\frac{T_m}{(C_p T_m + H)}$
			20°C	M.P.					
ALUMINUM	0.22	2.7	0.53	0.53	0.9	660	255	640	1.0
CARBON STEEL	0.11	7.9	0.13	0.07	0.08	1500	510	1980	0.8
18-8 STAINLESS STEEL	0.12	7.9	0.04	0.07	0.07	1460	(510)	(1900)	0.8
COPPER	0.09	9.0	0.94	0.80	0.10	1100	455	1343	0.8
URANIUM	0.03	19.07	0.065	0.10	0.175	1130	376	1022.5	1.11

$$\frac{EI}{V} = 2.1 \times 10^4 \phi (C_p T_m + H) \left\{ 1 + 5K \left(\frac{1}{\phi} + \frac{V}{2\alpha} \right) \frac{T_m}{C_p T_m + H} \cdot \frac{1}{V} \right\}$$

E = ACCELERATING VOLTAGE KV
I = BEAM CURRENT ma
V = WELDING VELOCITY cm/sec
t = WELD PENETRATION cm
 ϕ = BEAM DIAMETER cm

Figure 12

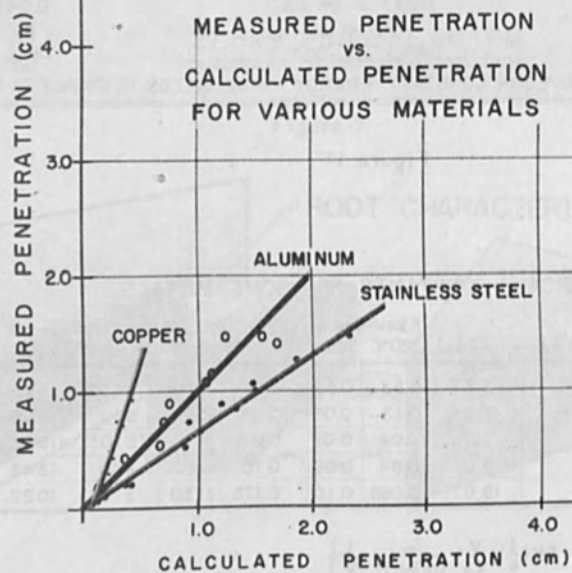


Figure 13

Spiking in Partial Penetration, Electron Beam Welds*

R. E. ARMSTRONG

Lawrence Radiation Laboratory, University of California

Livermore, California 94550

In the nuclear industry, it is often desirable to make maximum-penetration welds with no melting through, with minimum distortion, and with minimum effect of welding heat on adjacent or underlying materials. Electron beam welding appears to offer the best possibility of meeting these requirements. However, even with electron beam welding, we have found that maximum depth partial-penetration welds are difficult to achieve without some melting through because the root of an partial-penetration electron beam weld is not uniform but is very irregular. These irregularities in the root are what we have chosen to call "spikes". Figure 1 shows typical spikes in longitudinal sections through the center of electron beam welds in (a) aluminum, (b) uranium, and (c) stainless steel. Many spikes have voids in their lower portions because the molten metal following the beam does not fill them completely. In some cases, the molten metal will fill the spike hole but not fuse to the sides, leaving a condition similar to a cold shut in a casting. These cold shuts seriously reduce the strength of the weld at the root. This study was made to determine the effect of each of the controllable machine parameters on spiking, so that we might develop a welding procedure that would give maximum penetration with minimum spiking.

* Work performed under the auspices of the U.S. Atomic Energy Commission.

We selected depleted uranium for this study to obtain additional experience in the electron beam welding of this material. The uranium metal used for this study was furnished by the AEC Y-12 plant at Oak Ridge, Tennessee. A total of 650 welding coupons, each 3 in. long, $3/4$ in. wide, and 0.335 in. thick, were cut from two wrought salt bath annealed plates, with the 3-in. dimension taken in the rolling direction. Each coupon was numbered and its position in the plate recorded so that material variations could be eliminated or accounted for.

The welding equipment used was a 5-yr. old, 150-kV, 6-kW, hard-vacuum electron-beam welder with most of the commercially available attachments, including a variable-frequency circle generator. We have made some modifications, including digital meters to supplement the standard kilovolt and milliamperage meters and a digital meter to show the amperage supplied to the focus coil.

The joint design used was a straight butt weld without a step and no backing material. Coupons were first electropolished in phosphoric-chromic acid solution to remove oxide, and then placed in a special fixture that allowed us to weld twelve samples at one pumpdown and also ensured that the surface of each sample was at the same height in the welding chamber. The chamber was evacuated to 5×10^{-4} Torr or less, and each end of each pair was tack-welded to prevent the joint from opening during welding. The twelve welded pairs were then fractured by applying pressure in a testing machine to the face side of the weld. The center 1 in. of each fractured surface was then photographed at $8\frac{1}{4}X$ for record and evaluation.

The six welding variables we believed to have the greatest effect on welding and which can be machine-controlled are: voltage, beam current, travel speed, focal-spot position, circle size, and circle frequency. Three additional variables that may have a lesser effect on welding are; the distance of the work-

piece from the focus coil, the age of the filament, and the degree of vacuum.

In this study, the distance of the workpiece from the focus coil was held constant and a minimum vacuum of 5×10^{-4} Torr was reached before welding was started.

The effect of filament age was not determined.

The first three machine parameters--voltage, beam current, and travel speed--determine the heat input to the work. The combination of these can be expressed in joules per inch. The last three machine parameters--beam focal spot position or spot size, circle size, and circle frequency--control the concentration of heat in the work.

When we originally planned the study, we hoped to use statistical analysis to separate the individual effects on spiking of each machine parameter change. Further study showed that even in a continuous weld the spiking varied in such a random fashion that we could find no spiking characteristic that changed only as machine parameters were changed. The alternative was to use visual comparison as the final method of evaluation. The photographs of each weld are available for future study and can be evaluated statistically at any time a useful quantity is found.

We first checked the effect of heat input parameters on spiking. The voltage, travel speed, and total heat input were varied as shown in Fig. 2. The beam current is not specified., because it depends on the voltage and travel speed if a specific heat input value is desired. The circles designate the samples for which photographs are shown in this talk and they represent only a few of the many photographs taken.

The results of the study of the effect of heat input on spiking show that the changing of voltage, current, and work travel speed has little individual effect on spiking when the same depth of penetration is maintained. Fig. 3

shows the spiking at the extremes of our tests. The 110 and 150-kV welds at the same heat input are quite similar in penetration depth and spiking characteristics and so are the fusion and heat-affected zones. As the travel speed is slowed from 60 to 30 in./min., the fusion and heat affected zones become wider and the penetration and spike length decrease. When we increase heat input from 3400 to 5000 joules/in. at 30 in./min., we increase the penetration and we also increase the spiking.

After this study was completed, we made samples to check the effect of heat concentration parameters on spiking by varying the focus level above or below the work surface and by rotating the beam in circles of varying diameter and at several rotation frequencies. At first, the focus level was selected by visually focusing the beam on tungsten targets set at different levels. This method did not give satisfactory results, as I will explain later when I discuss beam focusing. The experiment was repeated by selecting a set of parameters that gave considerable spiking with the focus level at the work surface and then raising the focus level above the work surface by increasing the focus coil current by specific increments. The actual level of focus was not determined. Fig. 4 shows the effect of raising the beam focus position above the work surface by increasing the focus coil current. We can see that the spiking decreases but so does the penetration.

Fig. 5 shows the effect on spiking of deflecting the beam in a circle. Again, the spike length is reduced but so is the penetration.

Fig. 6 shows a weld made with the beam both defocused and rotated in a circle and with the beam power increased to get deep penetration. There is still considerable unfused area below the root, yet one spike penetrates completely through and one is almost through. The cross section shows a broad fusion zone

with a round root and a large heat-affected zone. We can expect high shrinkage and a large heat input to the work with these settings and we can also see that we get a considerable undercut on the surface because of the large molten pool formed during welding.

The size and position of unfused areas or cold shuts change when spiking is reduced by softening the beam. In a surface-focused weld, the cold shuts usually occur in the root area of a weld and are probably formed when the molten metal following the beam fills a spike hole but freezes before it bonds to the sides of the hole. When the beam is softened, these cold shuts increase in size and extend up the sides of the fusion zone. Fig. 7 shows examples of cold shuts in the fusion zone of a weld. They can reduce the strength of a weld more seriously than one might expect because we have found that they act as fracture initiation points in less ductile materials.

When we studied the test results, we suspected that spiking might be affected by how closely the penetration approached the root surface of the weld. As the penetration increases, the unfused area under the weld becomes hotter and should be easier to penetrate. A small variation in beam power might cause a large variation in penetration and put a practical limit to the maximum depth of penetration we could expect to obtain without spiking through.

Four weld samples were prepared with 1-in.-long steps milled in the root side to give six material thicknesses decreasing from 0.335 to 0.235 in. in 0.020-in. thick steps. These samples were welded at 110 kV, 40 in./min travel speed, and 20 to 23-mA beam current with surface focus. When we examined the fractured surfaces of the samples, there was no apparent difference in spiking or total penetration depth as the fusion zone approached the undersurface of the plate. Fig. 8 shows the samples made at 20 and 23-mA on the 0.335 and 0.235-in.

thicknesses. It appears that almost full penetration is feasible if we can find a method of eliminating spiking.

The conclusions reached from this study is that spiking appears to be a direct function of power density. Reducing the power density will reduce spiking but reduces penetration as well. Increasing power to recover the penetration loss increases the power density and brings back the spiking. Spiking is at a maximum when a deep weld with a narrow fusion zone is made because of the high power density required.

What must be done to reduce spiking and still get a narrow, deep weld? We do not know at the present time. First, we must understand the action in the pool and cavity and what causes the spikes. The work just described by Henry Tong is a tremendous step in this direction. We have taken some preliminary high-speed color motion pictures at 10,000 and greater frames per second looking along the beam into the pool and we can see that the cavity is in violent motion at incredibly high speeds. We can now see part way down into the cavity and there is no evidence of a cap as proposed by some investigators. The cavity walls appear to be very unstable and come together as Dr. Tong described. I had hoped to have many more films by now to observe the effect of material and parameter variations but due to several non-technical difficulties the program has been delayed. I expect to be able to find clues to the cause of the cavity instabilities and thus determine methods to stabilize the action.

During the course of this work, we made a very interesting study of beam focusing. In the heat input study, the beam was focused at the surface of the work by the standard method of visually focusing the beam on a tungsten block. The disadvantage of this method is that the block heats up very quickly at high power and the outline of the incandescent spot is hard to determine. The high

power beam also drills a hole in the block and the level changes. These conditions make this method of focusing difficult to repeat and very operator-dependent. Since the position of the focal point of the beam has a decided effect on spiking and penetration, accurate focusing is essential.

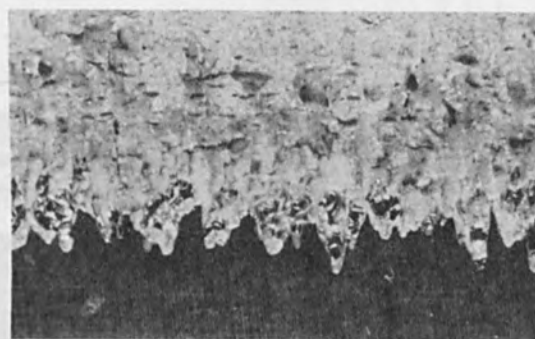
The focus coil amperages required to get a sharp visual focus was recorded for the welds made at 125, 130, and 135-kV and these values were plotted against the beam current for each of the welds made at these voltages. The object was to check the accuracy and reproducibility of the standard visual method of focusing. The operator took special pains to get the focus as sharp as possible by rotating the beam in a circle and focusing to a sharp line, instead of a point. The focus coil values for each weld are plotted in Fig. 9. There is not only an overlap between the different voltages, but there is a considerable spread between the focus coil settings for the same voltage and amperage. From our study of the effect of focal spot position on spiking, we know that a change of 10-mA in focus coil current makes a detectable change in spiking and penetration. Some of the values in this plot appear to vary as much as 30 mA. This variation casts doubt on the accuracy of the standard visual focusing method, especially if we intend to use focal spot position as a means of controlling spiking and penetration.

A focusing study was made at various voltages and amperages, using both tungsten and OFHC copper as target materials. We found a variation in focus coil currents similar to, although not as great, as that shown in Fig. 9. The beam was visually focussed three times at each point, and there was as much as 15mA difference between the three focus coil values. When sections of the curves were repeated at a later date, we found the shapes of the curves to be the same but they were displaced above or below the previous curves. We suspect that

this was caused by aging or replacement of the filament. This shift probably accounts for the large spread in focus coil values in Fig. 9. When copper was used as a target material, less focus coil current was required at the same power level than when tungsten was used. This may be because the ion stream from copper is greater and the space charge neutralization more complete. It is obvious that accurate focusing is a problem and that a more positive and reliable means of determining focus is required.

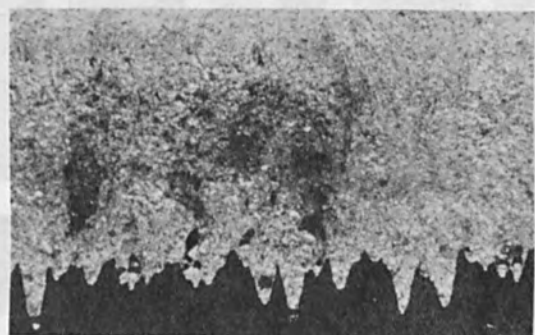
Adams and Sanderson of the British Welding Institute have done some excellent work in this field as has Don Sandstrom of the Los Alamos Scientific Laboratory. They pass a tungsten wire rapidly through the beam and display the resulting current signal on an oscilloscope. When the beam focus is at its sharpest, the current signal and the trace on the oscilloscope will be at a maximum. This method allows setting the focal spot at exactly the same position each time, eliminating the effect of material and operator judgement. We have modified this technique by using a special circuit to sweep the beam over a wire at controlled time intervals. We have found this method to be sensitive to the 60-cycle and the 360-cycle ripple on the high voltage to the electron gun, especially at lower currents. If we sample the beam when the voltage is at the high point, we get a different focal point than if we sample the beam when the voltage is at the low point. We have refined our signal generator so that we can deflect the beam at the same selected point on the voltage curve each time and are installing a dual-trace oscilloscope to coordinate the beam trace with the power supply voltage trace. This appears to be not only an accurate method for electronic focusing but also a good method for detecting changes in the quality of the operation of the machine by noting any change in focus coil current required to get a sharp focus with the same machine settings.⁵

Thank you.



(a)

0000
0001
0002
0003
0004
0005



(b)

0006
0007
0008
0009



(c)

0010
0011
0012
0013
0014
0015

0016
0017
0018
0019
0020

Figure 1

Table 1. - Heat input parameters investigated.

Joules per inch	110 kV	125 kV	130 kV	135 kV	150 kV	Inches per minute
3800		x	x	x		60
3400		x	x	x		
3200	(x)	x	x	x	(x)	
3000	(x)	x	x	x	(x)	

3800		x	x	x		50
3400	x	x	x	x	x	
3200	x	x	x	x	x	
3000	x	x	x	x	x	

4400	x				x	40
4000	x	x			x	
3800	x	x	x	x	x	
3400	(x)	x	x	x	(x)	
3200			x			
3000			x			

5000	(x)				(x)	30
4800	x				x	
4200	x				x	
3800	x				x	
3400	(x)				(x)	

(x) Photographs reproduced in report.

Figure 2

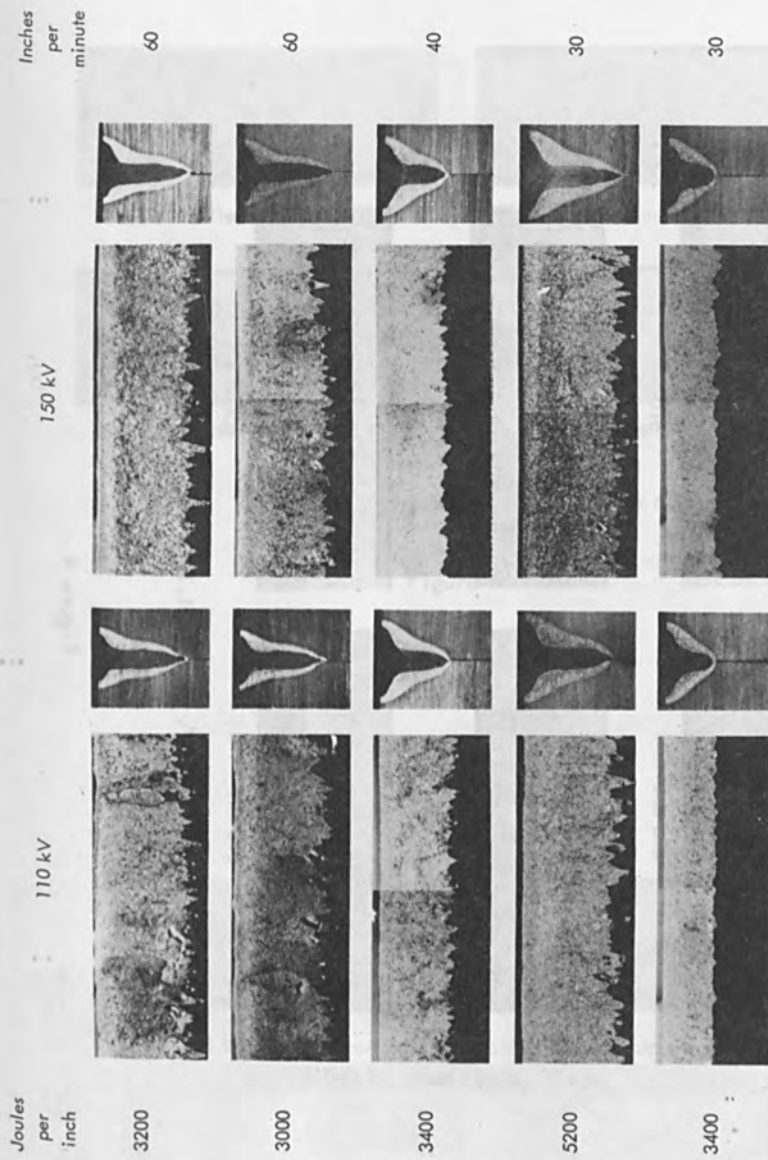


Figure 3

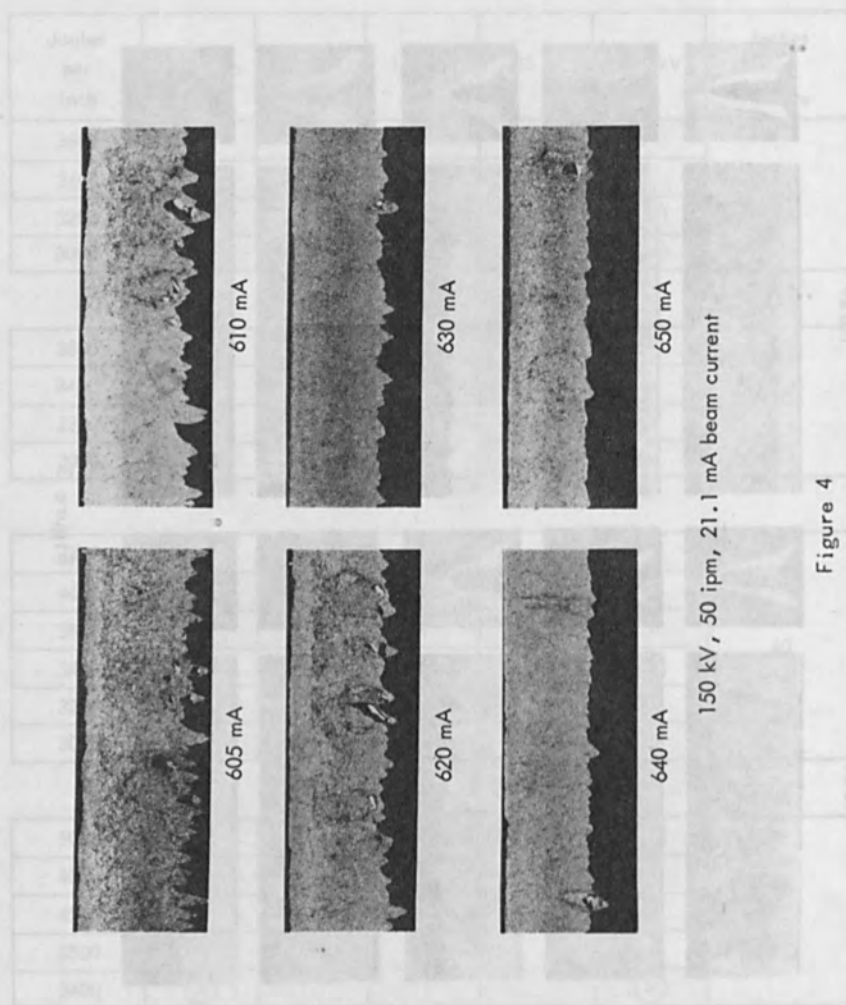


Figure 4

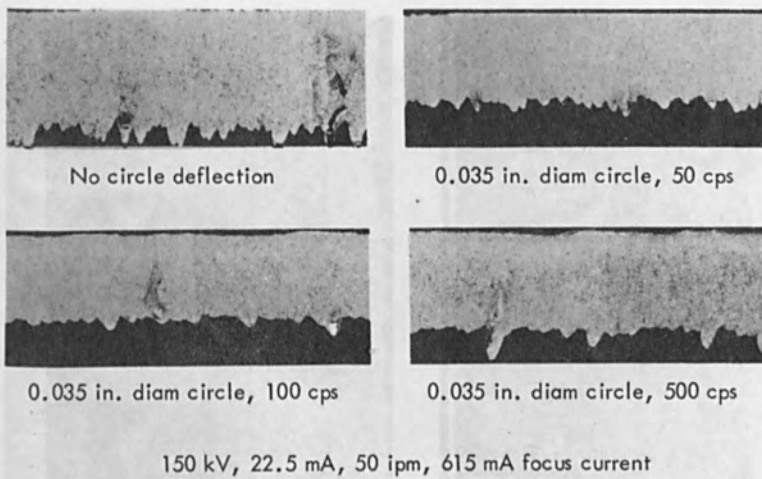


Figure 5

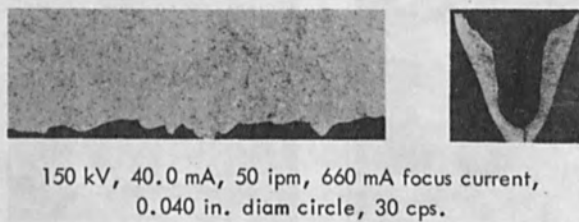


Figure 6

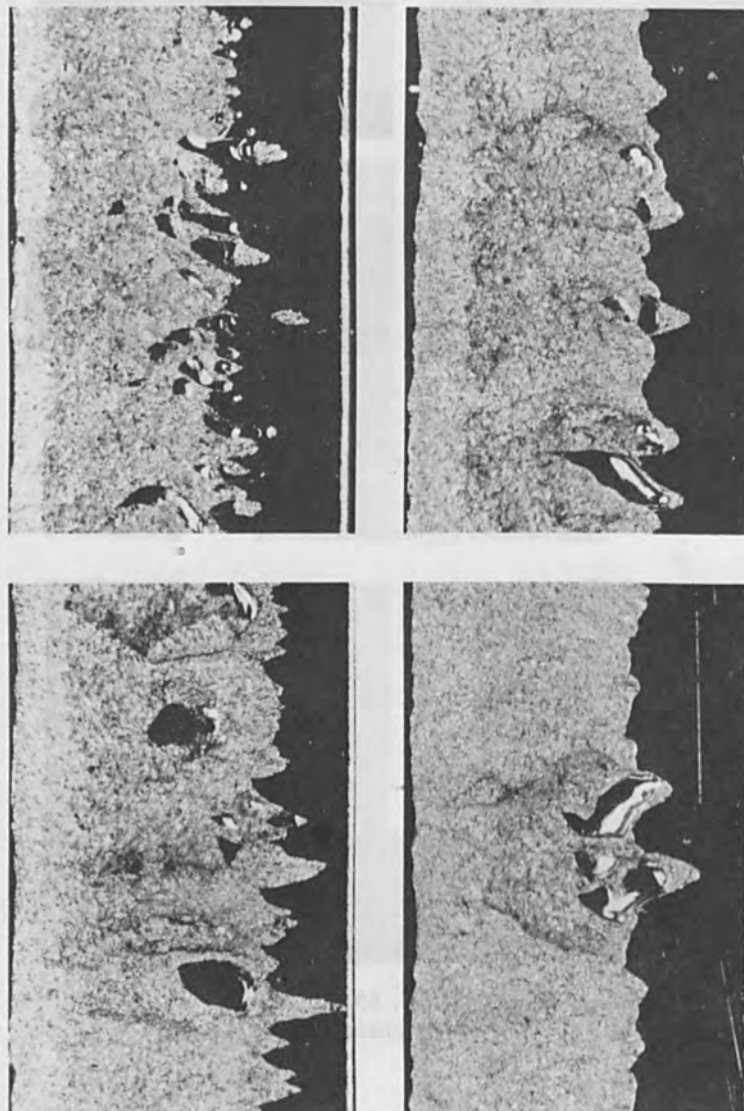


Figure 7

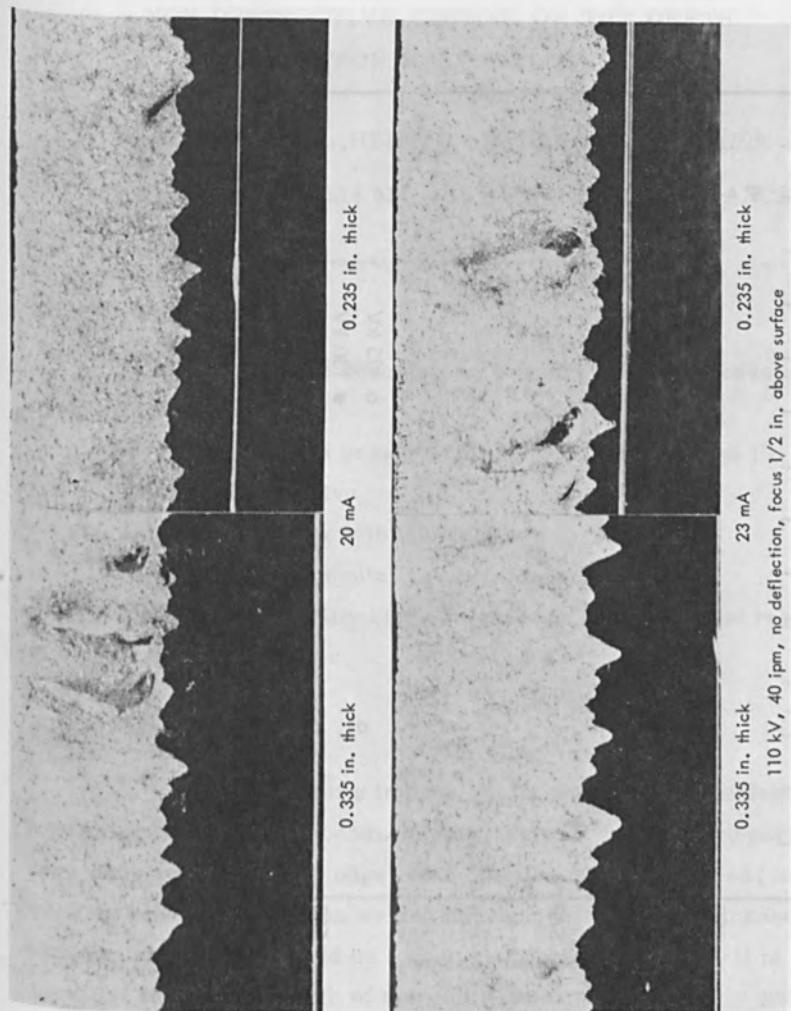


Figure 8

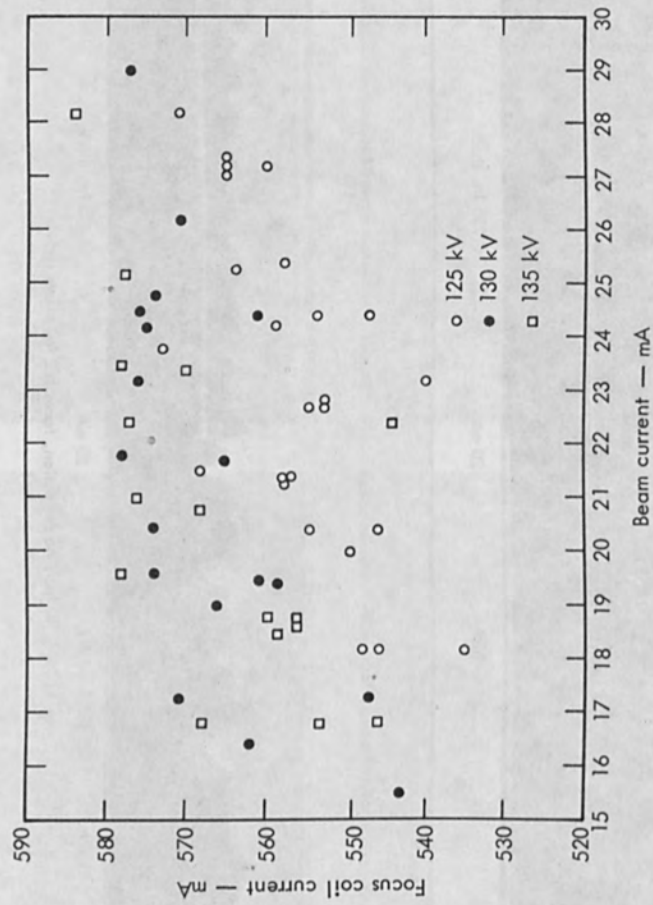


Figure 9

NON DESTRUCTIVE TESTING OF THE DEPTH
IN ELECTRON BEAM WELDS

M.BONCOEUR - L.HEINTZ - M.LAVAUD - M.RAPIN

C.E.A. Boite postale 511 - 15 - PARIS 15ème (FRANCE)

ABSTRACT

The authors describe an original testing process of the depth of the welds.

This method presents the following advantages :

- 1) It is non destructive
- 2) It is simultaneous with the welding.

The experimental set up is described.

The sensibility of the methode is derived of the results of a series of experiments.

INTRODUCTION

It is frequently impossible to visually test the depth of a weld made by electron beam welding. This is so when two parts have been welded edge to edge, on a fraction of the thickness, or when an assembly has been welded throughout its entire thickness with the bottom of the weld no longer visible after welding. It is essential to know the depth of the weld, for any insufficiently welded assembly may break causing serious accidents. It is why we tried to find a non-destructive method that would enable us to ascertain that the depth of the weld made was as required by the operator.

I - PRINCIPLE OF METHOD

A pure metal wire, a few hundredths of a millimetre in diameter, is placed along the joint of the parts to be welded at the required minimum welding depth, as shown in diagram of figure (1). When the electron beam hits the target, a bubble of molten metal is formed. If the depth of the molten area is adequate, the test wire becomes diluted in it. This dilution occurs in a short lapse of time and is helped by high temperature and the active mixing of the liquid metal. The ejection of metal vapour from the bottom of the liquid bubble to the free surface also makes easy the rise of tracer element to the surface of the weld seam.

The depth reached by the molten area can thus be related to the detection of the tracer.

- a) either on the surface of the weld after welding
- b) or during welding, in the plasma formed by the impact of the electrons on the metal.

This detection can be performed by optical spectrographic means. The tracer element is either detected or not. If it is not detected the desired depth has not been reached, and if it is detected this depth has been reached or exceeded. If the depth must be controlled more accurately, for instance, between a minimum and maximum depth, two wires of dissimilar tracer metals may be respectively placed at the desired levels. The detection of the two metals takes place, in exactly the same way as before.

II - TESTING OF WELD DEPTH AFTER WELDING

II.1 Apparatus

There are two distinct operations, i.e. electron beam welding and testing by emission spectroscopy, as will be seen from

the diagram in figure (2). The first samples were welded under low vacuum, with a SCIAKY machine fitted with a low voltage SV 2 gun. The other samples were welded a machine fitted with a high voltage HAMILTON triode gun. The welding parameters used with both the JARREL-ASH spectrograph of 3.40 meters focal length with photographic recording. The used grating had 1.200 lines per mm (30,000 lines per inch). The utilization of a high performance spectrograph was necessary because of the complex spectrum emitted by the welded metal. The operating conditions are shown in table (2).

II.2 The samples

The welded metal is a uranium alloy containing 0.2 per cent vanadium. The tracer metal is copper, it was mainly chosen on the assumption that its dissolution in the uranium was homogenous and that its concentration was less than 300 ppm at every point of the weld seam. In view of this, the copper was not expected to modify the characteristics of the U-0.2%V. alloy to any appreciable extent. The first tests were carried out on a sample composed of two cylinders of 124 mm outside diameter with a socket 4 mm deep. An 0.15 mm diameter copper wire was placed along the joint at a depth of 4 mm. The two cylinders were electron beam welded to a depth of approximately 4.5 mm. The first results being successful, we completed them by undertaking a second series of tests on flat samples similar to the one shown on figure (3), i.e. the height of the socket is 3 mm, the copper wire 0.15 mm diameter and the expected welding depth 4.5 mm. In order to find out the minimum length of copper wire that must be dissolved so as to be detected on the surface and, conversely, the minimum length of wire that must not be dissolved for an absence of copper to be detected on the surface of the weld seam, copper segments varying in length and spacing were placed as shown in the diagrams of figure (3).

II.3 Results

The micrographs of figure (4) represent cross sections through two welds, i.e. the copper wire did not melt in the case of the insufficiently deep weld and it did where the weld is correct. We looked for correlation between the presence of copper on the surface of the weld seam and the existence in the welds seam of an area where the copper was diluted in the uranium - indicating that the weld had penetrated to the tracer wires. Penetration faults were simulated by breaks in the copper wire. To verify this correlation, longitudinal macrographs along the median plane of the weld were made on samples after spectrographic determination of the copper content on the surface of the weld seam. Figures (5), (6) and (7) represent the macrographs of the three welds. On each one we have marked the analytical points and above we have indicated the copper contents determined at each of these points. The upper limit of the copper contents observed in the base metal is equal to 40 ppm and is indicated by a thick unbroken line on figures (5), (6) and (7). Uncertainty as to the position of the points and to copper contents are ± 1 mm and ± 10 ppm respectively. The base metal, and above the molten metal, may be seen on the macrographs. In the molten metal, copper rich areas show up in light colouring where the copper wire segments were. These areas are heterogeneous and copper concentration which is greatest at the bottom of the weld seam, diminishes towards its surface. The copper contents, determined by electronic micro probe, at the bottom of the weld seam and at 0.5 mm from the surface, are about 1.68 per cent and 0.03 per cent respectively. A comparative examination of the copper contents on the surface of the weld seam and the macrographs show that :

- 1) the copper wire has melted on a level with the spots where

the emission spectrography has detected a copper content exceeding 40 ppm. Consequently the depth of the weld was more than the thickness above the copper wire and the weld is correct.

2) there is no copper wire on a level with the spots where the emission spectrography has detected copper contents less than or equal to 40 ppm. This would denote an insufficiently deep and therefore incorrect weld. Figure (5) shows an abnormal spot, which is probably due to an error in determination or a surface fault.

It is also observed that a penetration fault covering a length of 3 mm is detectable under our experimental conditions. The dilution of a copper wire segment 2 mm long is sufficient to be detectable on the surface. The examination of a micrographic cross-section passing through an analytical point shows that disturbances on the surface of the surface of the weld are not significant. The method is therefore non-destructive.

III - TESTING OF WELD DEPTH DURING WELDING

The testing just described is made after welding. If a weld is incorrectly done, it is necessary to place the part back in the welding machine and effect all adjustments again. It would be better to have a method in which the depth reached could be tested directly during welding and with possible modification of the welding parameters should the weld not be correct.

III.1 Plasma formed during welding

During electron beam welding, a plasma appears over, the welding area, as will be seen from figure (8). A well known phenomenon of welders, is the modification of light emitted by the plasma when an over deep weld breaks through to an underneath support of a different composition. This change is due to the presence

in the plasma of atoms from the support. Likewise, if the electron beam meets a tracer located in the base metal, its atoms will appear in the plasma formed.

III.2 Experimental methods

The alteration in the composition of the plasma suggests two spectrographic methods to test the depth of a weld, i.e. detection of the tracer element atoms (a) by emission spectroscopy or (b) by atomic absorption spectrophotometry.

III.2.1. Emission spectroscopy

The principles of the experimental method are outlined on figure (9). A beam of the light emitted by the plasma crosses a window, transparent to the analytical line of the tracer element used. It enters a direct reading spectrograph whose photomultiplier delivers an electrical signal proportional to the intensity of the analytical line.

This signal may be recorded as a function of time on a potentiometric recorder.

III.2.2. Atomic adsorption spectrophotometry

One of the experimental devices that can be used for this second method is shematized in figure (10). The beam from a lamp emitting the spectrum of the tracer element goes through the plasma above the molten area. The initial intensity I_0 of the selected characteristic line of the tracer element is reduced to value I if atoms from it are present in the plasma. Then the light enters a monochromator adjusted on the selected line. A photomultiplier delivers a signal proportional to I . Both these methods have the advantage of being non-destructive and instantaneous. Ultimately, if the welding parameters can be regulated by the penetration monitoring,

these two methods enable production to be automated. Nevertheless the atomic adsorption spectrophotometry method is more sensitive and requires less complicated and less costly apparatus as well as less qualified staff. It should be given preference over the emission spectrography method for routine testing.

III.3 Experimental results

We used the atomic adsorption spectrophotometry method for the preliminary tests. We wished to verify whether a small variation of tracer element content in the base metal fed to a change in the plasma composition, resulting in an alteration of its optical density, for a characteristic wave length of the tracer. We used a 1.5 kW type CL1 ALCATEL gun placed in a low vacuum enclosure. The samples were various uranium cylinders with 5, 35, 80 and 120 ppm of copper. The targets were bombarded for time t until a constant optical density of the plasma was obtained, read on a curve similar to that which may be seen in figure (11). Time t is related to the specific power required to melt the metal and to the mass of the targets. For a low power of about 250 watts concentrated on 1 mm^2 and a uranium target target of about 2.5 cm^3 , this time is in the order of 30 sec. With a high powered gun, time t is in the order of a few hundredths of a second and determination is instantaneous. On figure (12) the optical density d of the plasma, for the characteristic wave length of copper is plotted, as a function of the copper content in the targets. We observe :

- 1) that copper present in a target may be detected by atomic absorption spectrophotometry
- 2) that a relationship exists between the optical density d of the plasma for the characteristic wave length of copper, and the copper content of the target, enabling calibration to be effected ;

3) that the detection threshold of copper is very low and the method very sensitive.

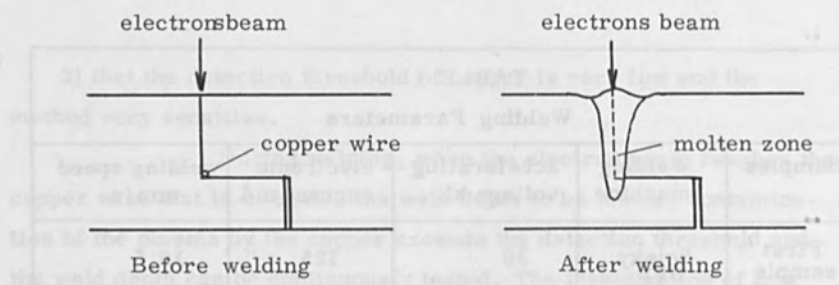
During welding, when the electron beam reaches the copper wire that is to enable the weld depth to be tested, contamination of the plasma by the copper exceeds the detection threshold and the weld depth can be continuously tested. The investigation of this technique is not quite terminated.

IV - CONCLUSION

Our results show that the depth of an electron beam weld may be tested during or after welding by diluting a tracer wire placed at the required depth. After welding, this metal is detected on the surface of the weld by emission spectroscopy. Furthermore, the depth of the weld can be tested continuously during welding through tracer detection by atomic adsorption spectrophotometry. This second method has the advantage of submitting the welding parameters to the continuous testing of the weld depth. The two methods show common advantages : they are both non destructive, reliable and sensitive. This type of testing could also be envisaged, say, for microplasma or T.I.G. welding processes.

TABLE 1 Welding Parameters				
Samples	Welding machine	accelerating voltage kV	electronic current mA	welding speed mm/s
First sample	Sciaky SV2	20	125	16,5
A	Hamilton	140	11	22
B	Hamilton	140	11,5	25
C	Hamilton	140	11	22

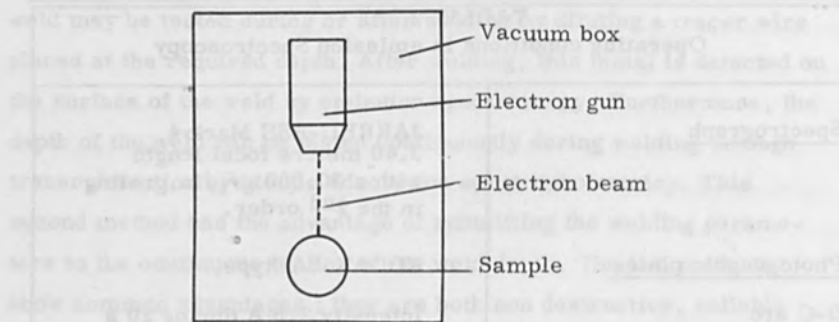
TABLE 2 Operating conditions in emission Spectroscopy	
<u>Spectrograph</u>	JARREL-ASH Mark 4 3.40 meters focal length with a 30.000 gr./in.grating in the 2nd order.
<u>Photographic plates</u>	KODAK SA1 type.
<u>D-C arc</u>	Intensity 2.5 A during 20 s Sample as cathode Graphite counter-electrode Analytical gap : 4 mm.
<u>Controlled atmosphere</u>	10% H2 in N2 gas send in the analytical gap at the rate of 5 liters per minute.
<u>Analytical lines</u>	Cu 3,247.54Å /U 3,247.06Å After background correction.



Testing of weld depth after welding : principle of method

FIGURE 1

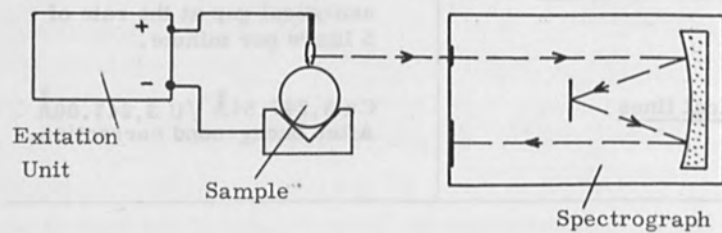
1 st Operation Welding.

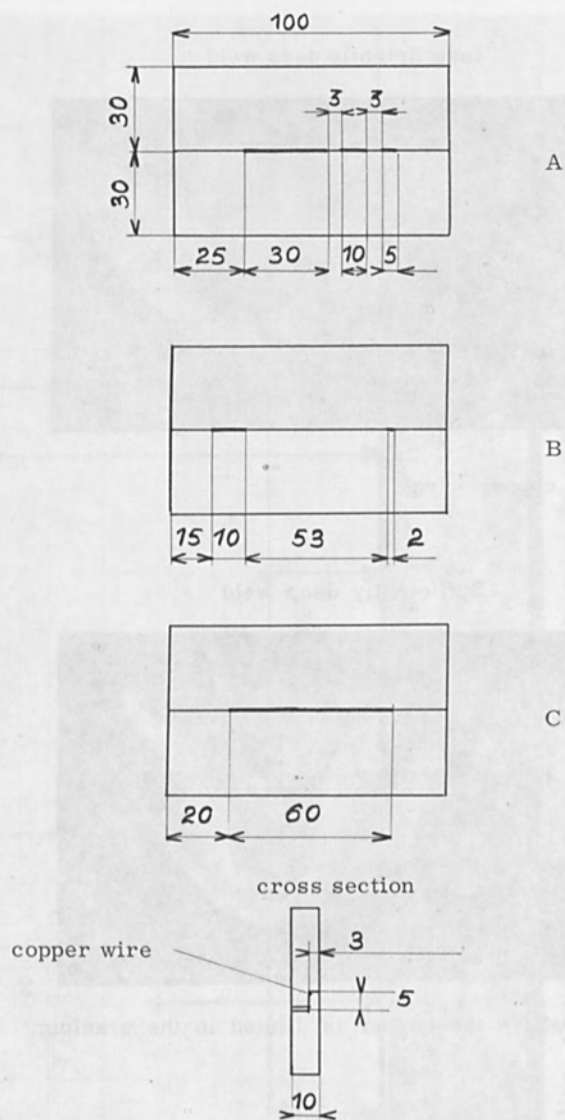


Testing of weld depth after welding : apparatus

FIGURE 2

2nd Operation Testing

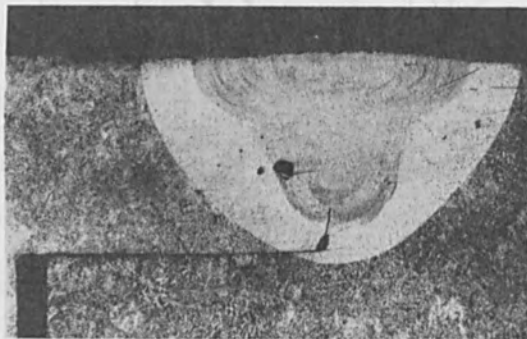




Testing of weld depth after welding : flat samples.

FIGURE 3

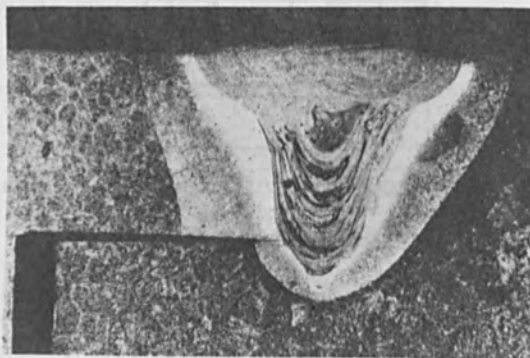
Insufficiently deep weld



copper wire

X 11

Sufficiently deep weld



X 11

area where the copper is diluted in the uranium

Micrographs of cross sections through two welds

FIGURE 4

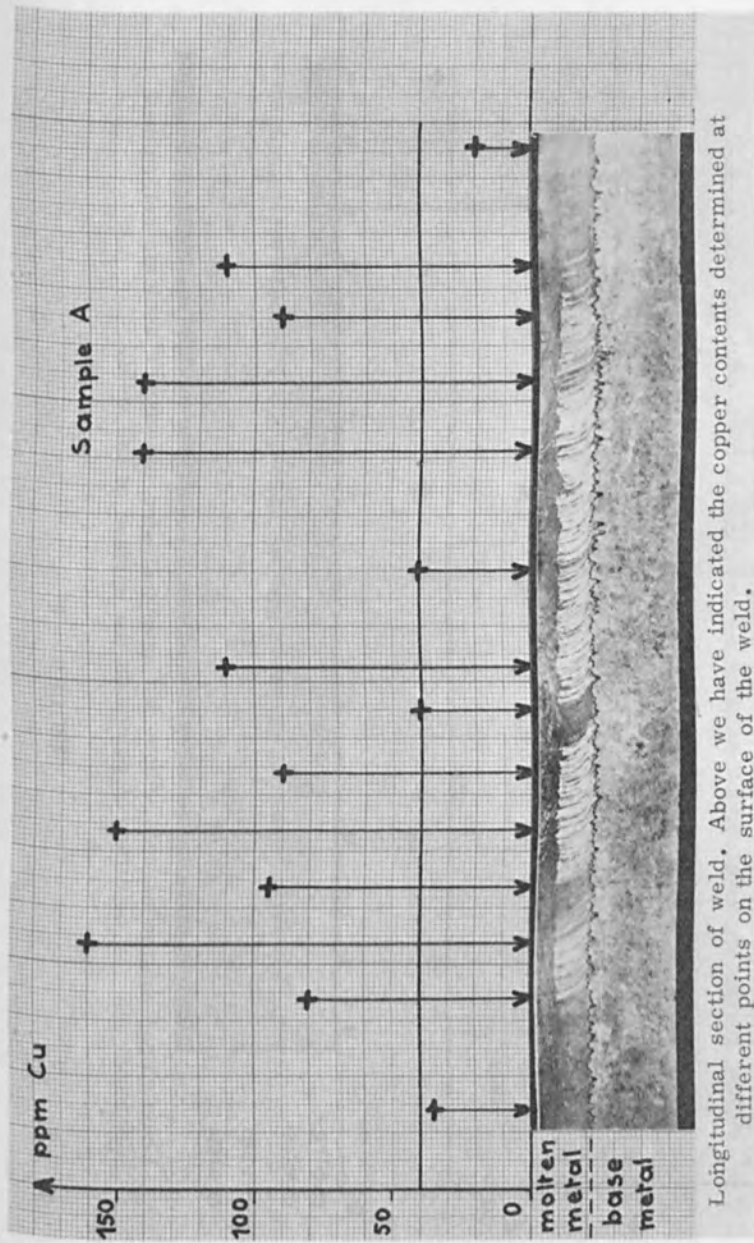
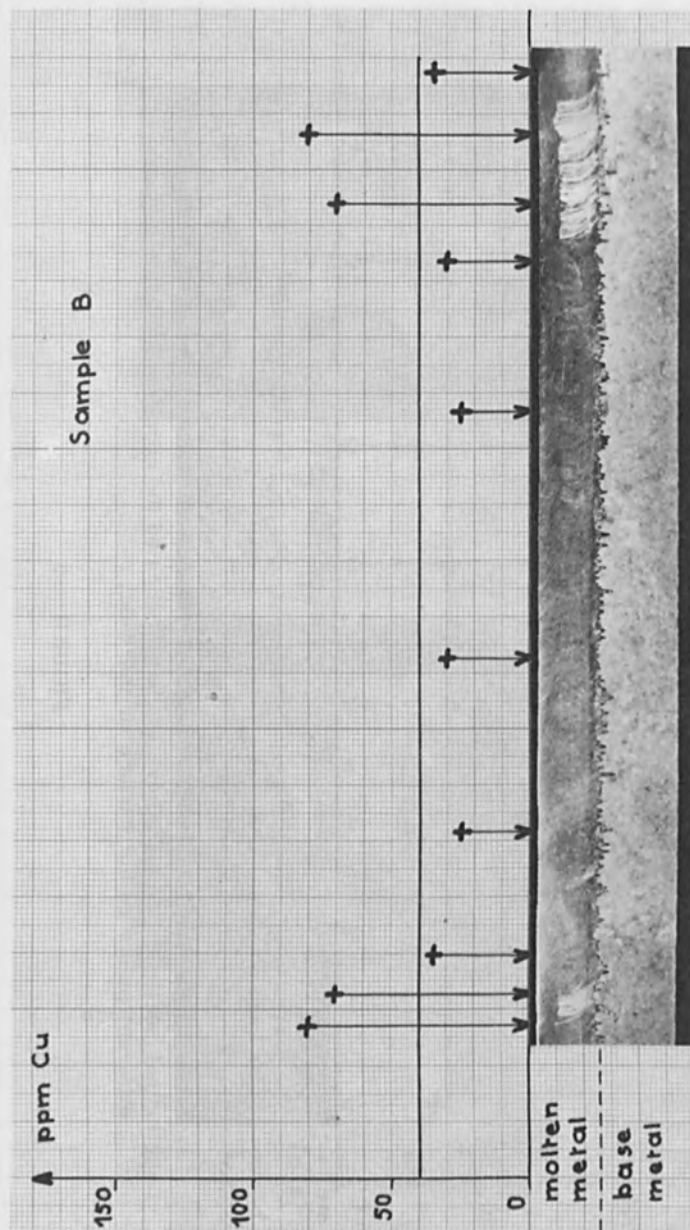
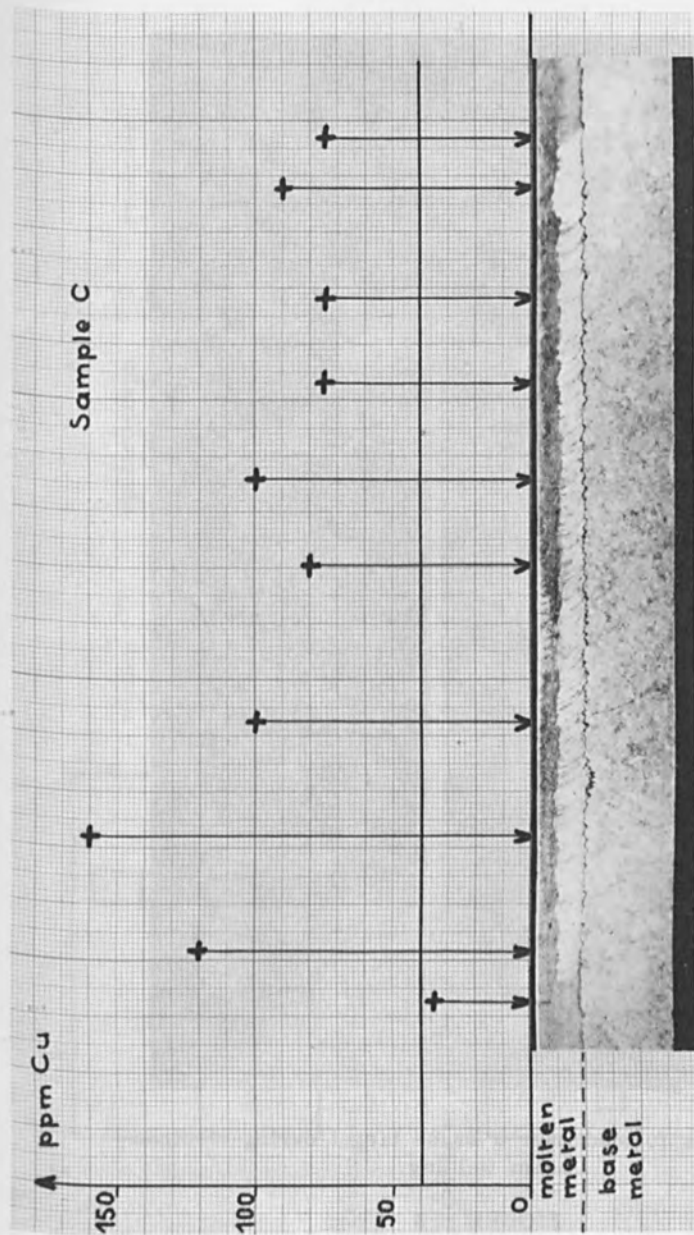


FIGURE 5



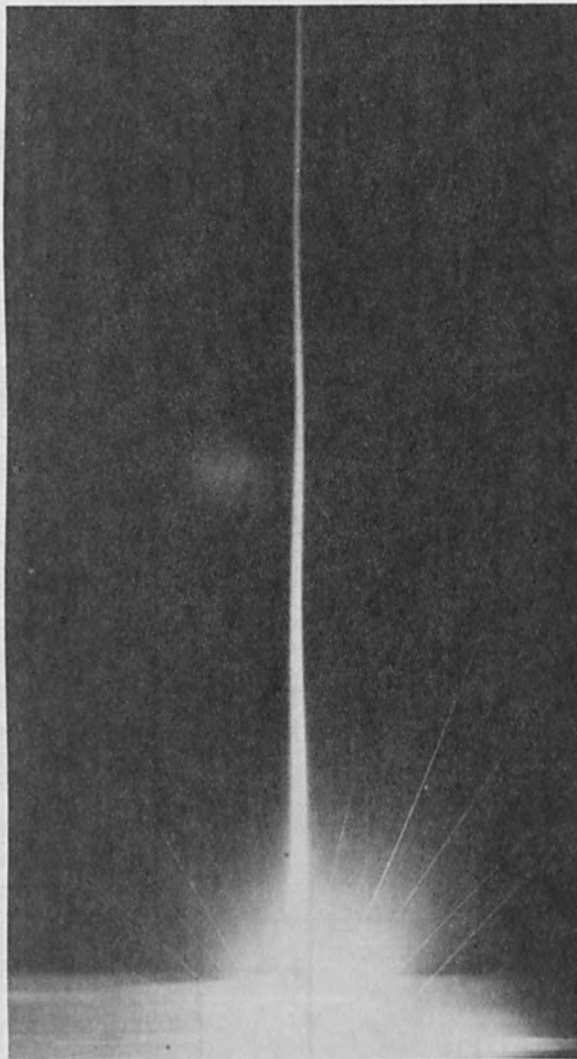
Longitudinal section of weld. Above we have indicated the copper contents determined at different points on the surface of the weld.

FIGURE 6



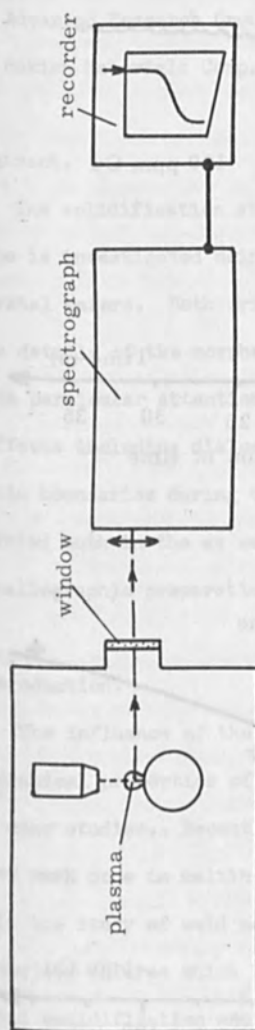
Longitudinal section of weld. Above we have indicated the copper contents determined at different points on the surface of the weld.

FIGURE 7



Plasma formed over a brass target during electron beam welding.

FIGURE 8



Testing of weld depth during welding with emission spectroscopy - Principle of the experimental method.

FIGURE 9

Testing of weld depth during welding with atomic absorption spectrophotometry. Principle of the experimental method.

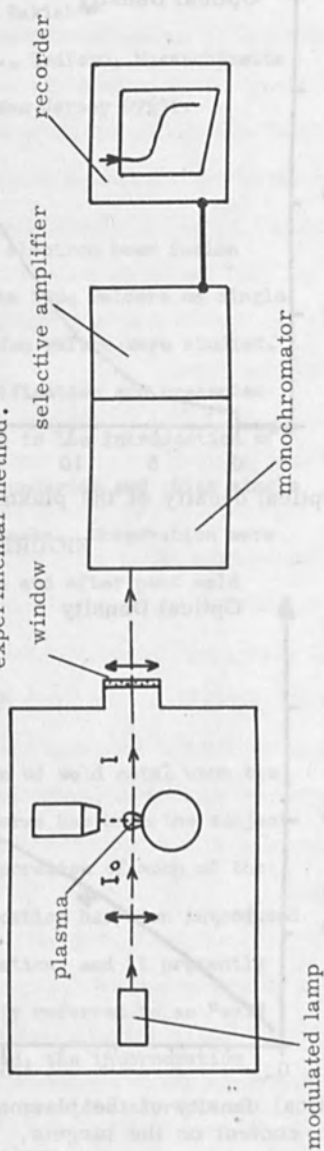
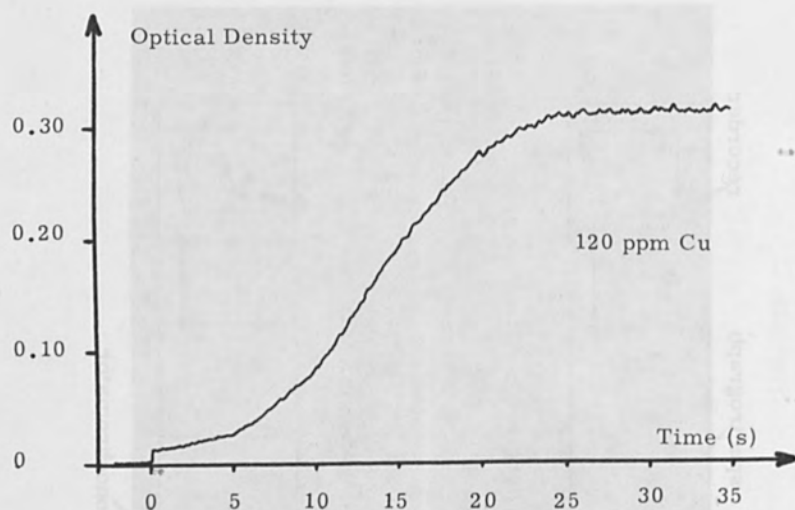
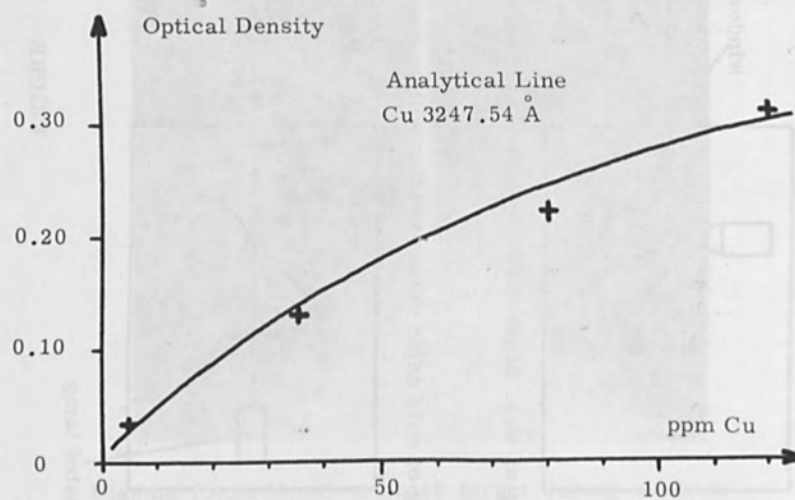


FIGURE 10



Optical density of the plasma as a function of time

FIGURE 11



Optical density of the plasma as a function of the copper content on the targets.

FIGURE 12

SOLIDIFICATION STRUCTURE OF EBW SINGLE CRYSTALS

D. Passoja* and R. Bakish**

* Advanced Research Group, EBTEC Corp., Bedford, Massachusetts

** Bakish Materials Corp., Englewood, New Jersey 07631.

Abstract.

The solidification structure in an electron beam fusion zone is investigated using beam on plate type welders on single crystal wafers. Both oriented and random wafers were studied. The details of the morphology of solidification are presented with particular attention being devoted to the introduction of defects including dislocations, sub boundaries and high angle grain boundaries during the welding process. Observations were carried both in the as welded condition and after post weld metallographic preparation.

Introduction.

The influence of the solidification of weld metal upon the mechanical properties of welded structures has been the subject of many studies. Recently^{1,2,3)} incorporation of much of the past work done in melting and solidification has been introduced into the study of weld metal solidification, and it presently comprises an area which has been loosely referred to as "weld metal solidification mechanics". Indeed, the incorporation has proved to be meaningful, for it has provided a practical extension of our present welding technology.

Fundamental problems still remain to be solved, however, for it appears that there is much to be studied on the atomic scale before it is possible to understand all of the structure-properties relationships required to describe the physical metallurgy of electron beam welds. Obviously, it would be desirable to assemble a detailed description of solidification on the atomic level and, through use of suitable models, to predict phenomena on a macroscopic scale. One of the most important defects to be considered on the atomic scale is the dislocation. Of particular importance is the role of dislocation upon:

- a) the mechanical properties of metals during and after welding, and
- b) the mechanism of solidification of weld metal.

It would appear that these are two of the more important considerations if we are to make progress toward understanding the physical metallurgy of electron beam welding.

The following work is a study of solidification of electron beam welds in ultra-pure single crystals of aluminum and zinc. By using ultra-pure metal single crystals of known orientations, it was hoped that the role of dislocations in solidification could be studied.

During planar growth from the melt, solidification (i.e., epitaxial growth of the melt) proceeds along "easy growth" directions as dictated by the surrounding solid substrate. It has been⁴⁾ shown that these directions are $\langle 100 \rangle$ in FCC and BCC crystals, whereas these directions are $\langle 0001 \rangle$ directions in HCP crystals. Growth proceeds along any one direction so long as the temperature

gradient resolved along any particular crystallographic direction is a maximum. Both crystallographic and kinetic factors seem to influence the appearance of any one growth direction.

Observance of this phenomenon is by no means restricted to liquids. For example, Figure 1 shows an interface of $\langle 100 \rangle$ orientation of CVD (chemically-vapor deposited) tungsten. The growth form in this case is a pyramid whose faces are bounded by $\{110\}$ planes; this growth form and direction is precisely that which is observed in dendrites of solidified metal crystals⁵⁾ *.

Experimental.

Single crystals of high purity aluminum and zinc were prepared by sparkcutting and cleaving the respective crystals. Chemical etching was used to reveal the orientations of the aluminum crystals. $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$ orientations were welded with "bead on plate" welds being made in the crystal faces. Various welding directions were used on each crystallographic face.

Zinc crystals were cleaved at liquid nitrogen temperature in order to expose a clean (0001) type surface. Welding was performed with the following crystallographic relationships:

Electron beam $\parallel \langle 0001 \rangle$

Welding direction $\parallel \langle 11\bar{2}0 \rangle$

All crystals were welded on a W-2 Hamilton-Zeiss electron beam welder. The following welding parameters were used:

	<u>Voltage</u>	<u>Current</u>	<u>Welding Velocity</u>
Zinc	90 KV	1 ma	35 ipm
Aluminum	120 KV	4 ma	30 ipm

Undercutting of the fusion zone in the zinc crystals was commonly observed and, due to the high thermal stresses, cleaving of thin crystals would often occur after welding.

The as-welded crystals were observed with optical and scanning electron microscopes in order to study the as-welded structure in the fusion zone. Some cross sections of the welds were made in order to confirm that the electron beam had penetrated deeply into the crystal during welding.

Welds were also made on zinc crystals that had been vapor coated with silver on one half of the exposed (0001) crystal face. Pulsed-type welds were made in these cases so that the role of the influence of an impurity on solidification could be observed.

Results.

FCC - Aluminum Single Crystals: welded with the $\langle 100 \rangle$, $\langle 110 \rangle$ and 111 directions parallel to the electron beam.

Fusion Zone: The microstructure of solidification was relatively featureless. Weld ripples were present in all welded samples; however, no structural features other than the ripples themselves could be observed in the fusion zone. (See Figure 2.)

Heat Affected Zone: The microstructural features observed in these areas were slip traces parallel to $\langle 110 \rangle$ directions. Slip takes place in FCC crystals on (111) planes; hence, one would expect to observe slip traces in $\langle 110 \rangle$ directions, since a (111) plane would intersect a (100) face at 45° or in a $\langle 110 \rangle$ direction. Cross-slip was also evident as indicated by the wavy slip traces present along with the straight slip traces in these regions. One would also expect

to see this phenomena, since the stacking fault energy is very high ($> 100 \text{ ergs/cm}^2$) and screw dislocations are known to cross-slip easily in this system.

Hexagonal - Zinc: Surface ripples were observed in the fusion zone in all of the welded crystals. At 50 - 100 x the ripples generally appeared featureless; however, closer inspection at higher magnification revealed that three distinctly different ripples were present. These three types were:

- 1) Featureless -- smooth ripples having no observable structural features at 10,000x
- 2) Faceted -- ripples containing crystallographic facets observable at 1,000x
- 3) Ledges -- terraced ripples having distinct ledge-like growth features.

Featureless: Figure 3 shows featureless ripples often observed within the fusion zones. These ripples were without structural details at 10,000x; i.e., it was not possible to observe any crystallographic details at this magnification.

Faceted: Figure 4-A shows a typical faceted ripple. Usually, these regions could be found as transition regions between two ripples like those shown at B. Higher magnification revealed that these facets contained fine steps whose heights were on the order of 300 - 700 Å (Figure 5). It was possible to trace out steps which traversed several facets. The facets, of course, represented different crystallographic faces of the crystal; the steps, however, had a common crystallographic origin. Facets could usually be observed in ripples which deviated a large degree from a (0001) plane. Usually, these

facets bounded a ripple which was growing as a ledge parallel to a (0001) plane. This was not always the case, however, for when ledge-type ripples deviated drastically from (0001), a faceted growth form would appear.

Ledge Growth: The ledge growth structure is shown in Figure 4-B. Ledge heights provided a means by which to make a logical sub-division of the different types of ledges observed.

Ledges - 3000 - 1500 Å: Figure 6 shows coarse ledges which were contained within a ripple. Seldom was there any crystallographic faceting observed on any one ledge; rather, the ledges appeared to be influenced by the ripples themselves. Apparently, the shapes of the coarse ledges were strongly influenced by growth kinetics within the weld ripples, since these ledges were observed to be influenced by the position of the liquid - solid interface during solidification. For example, the arrow at A - A' in Figure 7 shows how the ledges were influenced by the position of the liquid - solid interface during their growth. Growth was interrupted at A - A'; However, ledge growth continued after the interruption. The influence of the interruption is clearly seen on the ledge growth morphology shown in Figure 5.

Ledges - 200 - 500 Å: Within the coarse ledges, finer ledges were also observed (Figure 8). Usually, these were approximately 300 - 400 Å in height. It would appear that the coarse ledges were composed of many fine ledges, and bunching of the fine ledges would produce the coarse ledges.

Influence of the substrate on the ripple growth is shown in Figure 9. A + B - B' is a twin which traverses both the solid single

crystal substrate and the fusion zone. The twin is shown to be traversing the weld ripples, but it does not create any noticeable change from one side of the twin boundary to the other.

Growth sources: Perhaps the most significant observation was that it was possible to locate growth sources from which ledges would originate. Figure 9-C shows one such source. Higher magnifications revealed that both double and single spiral growth sources (Figure 10) were present. These growth sources were parallel to the basal (0001) plane and were generally located in a region close to the outer edges of the fusion zone.

One growth source, shown in Figure 11, had generated ledges which migrated along two ripples. When the orientation of the ripple deviated more than a few degrees from (0001), faceted growth occurred (E); along the other ripple, however, ledge growth prevailed.

Figure 12 shows a growth source and details of fine ledges along the sides of the source. There would often appear to be some type of impurity at the finer ledges; however, the impurity type was never identified. Regular, reproducible faceting was never observed along the sides of the ripples; rather, some type of facets were present. A generalized shape was, however, difficult to ascertain.

Discussion:

The experimental observations presented above have great similarity to experimental observations made when investigating the structure of the liquid-solid interface by decanting. Generally, it was found that in these experiments ^{6,7)} decanting the liquid in front of the liquid - solid interface would produce a terraced surface on the solid interface.

It was not known whether this terraced structure was indicative of the true structure of the liquid - solid interface present during solidification, or whether it was the structure of a solidified thin film of liquid which had adhered to the solid after decanting⁸⁾ and had subsequently solidified. Indeed, these works indicated that no terraced structures were present on aluminum interfaces. In lead, tin, and zinc, however, when the solid interface approached a close packed plane, the steps became larger and flatter and the terraces were occasionally centered around some projection⁹⁾. The origin of these projections was not known.

The results of this work coincide nicely with many of the observations previously obtained in decanting experiments. Indeed, the surface of the weld pool is a free surface during solidification. Previous work in zinc¹⁰⁾ has indicated that during planar growth the interface is parallel to the $\{0001\}$ planes. Our observations indicate that spiral dislocation growth sources are parallel to $\{0001\}$ and these sources can always be found as an origin of ledges.

It is postulated, therefore, that these growth sources might well control the observed $\{0001\}$ preferred growth direction. Indeed, a growth mechanism of this type is quite possible. The undercooling required to cause growth in those directions would be greatly reduced because the requirements for two-dimensional nucleation were eliminated.

The presence of both coarse and fine steps within a ripple is believed to be due to "traffic jams" during the growth of single steps at the surface. Formation of macroscopic steps have been discussed by Frank^{11,12)}. He suggested that impurity adsorption at the front of a growing ledge would tend to impede the ledge and, thus, slow its

growth. Ledges moving behind the impeded step would subsequently be slowed and, thus, step bunches would form. Our observations show that the coarse steps, indeed, are composed of an accumulation of smaller steps of the order of 300 Å or so. The 300 Å represents the limit of resolution of the SEM; thus, it is not possible to see steps smaller than the one of 300 Å.

It was felt that impurity adsorption at the liquid - vacuum interface, for thermodynamic reasons, would raise the impurity level at the surface of the melt and, thus, increase the frequency at which the coarse steps were observed. By vapor coating one-half of a zinc crystal with silver and pulse welding at various points on the surface, we were able to create macroscopic steps within a solidified region. Figures 13 and 14 show optical micrographs of the uncoated and coated regions, respectively. The formation of coarse steps in the vapor coated regions are striking experimental evidence of the influence of impurity adsorption on coarse step formation. In light of this finding, the formation of coarse ledges is thought to occur due to impurity adsorption near the surface of the liquid. For this reason the true structure of the liquid - solid interface is not necessarily that which has been observed in these experiments. Rather, the impurity levels near the surface of the melt probably increase the frequency at which the coarse ledges are observed in the last material to solidify.

Growth sources have previously been observed in several experiments¹³⁾ and are known to occur due to the intersection of a dislocation with the growth front. Double spiral growth sources arise when two screw dislocations of opposite sign intersect the growth front¹⁴⁾. The exact mechanism for the formation of the sources is not known, but two possibilities

have been forwarded:

- 15)
1) Forty has observed that concentric circles (growth sources) are left behind when impurity particles are incorporated into the growth front;
- 16)
2) Chalmers has suggested that the supersaturation of vacancies present during solidification probably acts as source of dislocations by vacancy condensation during cooling.

Both of these observations indicate that some dislocation sources are available during solidification; however, neither observation suggests an exact mechanism by which dislocations of a certain type generate the growth sources observed in these experiments. Particularly, for double growth sources in the $\langle 0001 \rangle$ direction two screw dislocations of $\bar{b} = c [0001]$ and $\bar{b} = c [000\bar{1}]$ (or an equivalent) are required to operate. This type of dislocation is stable in the hexagonal lattice; however, it is less stable than a $[11\bar{2}0]$ type of dislocation.

For lack of better terminology, the word faceted ripples was used to describe ripples having distinct crystallographic features, but without ledges parallel to the basal plane. Presence of these facets is thought to be due to some thermal etching which occurred upon cooling from the welding temperature, as well as kinetic influences.

If one considers a weld ripple to be a growing crystal then the surviving surfaces of the crystal after solidification are the slowest growing faces. Essentially, the fastest growing surfaces "grow out" of the crystal and the slowest growing faces survive. The faceted growth morphology is thought to be a manifestation of this phenomena.

Conclusions.

Observations of weld ripples by scanning electron microscopy have revealed features of solidification in zinc and aluminum.

Ultra-pure single crystals of known orientations were used to reveal the details of the solidification process.

FCC - aluminum -- structureless weld ripples were present, planar growth obtained.

Hexagonal - zinc -- three types of ripples features were observed:

- a) Featureless -- showing no structural detail at 10,000x
- b) Faceted -- distinct, irregular faceted ripples composed of fine terraces of 300 - 700 Å.
- c) Ledges -- composed of both coarse ledges of 3000 - 1500 Å, and finer ledges of 200 - 500 Å.

Growth of the ledges was observed to originate from double spiral growth sources; the plane of these sources was parallel to (0001).

* The authors would like to express their gratitude to Dr. C. Lundin for supplying the sample of CVD tungsten.

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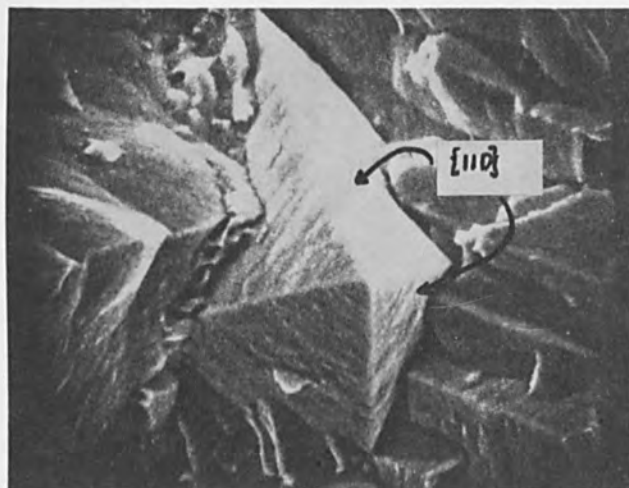


Figure 1.
CVD tungsten growth interface. The pyramid tip points in a $\langle 100 \rangle$ direction with the faces being composed of $\{110\}$ planes.

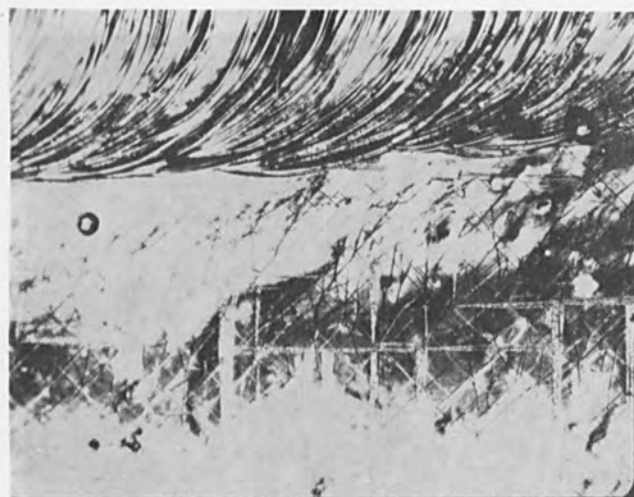


Figure 2.
EB weld made in an aluminum single crystal: orientation of the electron beam is EB $\parallel 100$; weld direction $\parallel \langle 010 \rangle$ 80x.

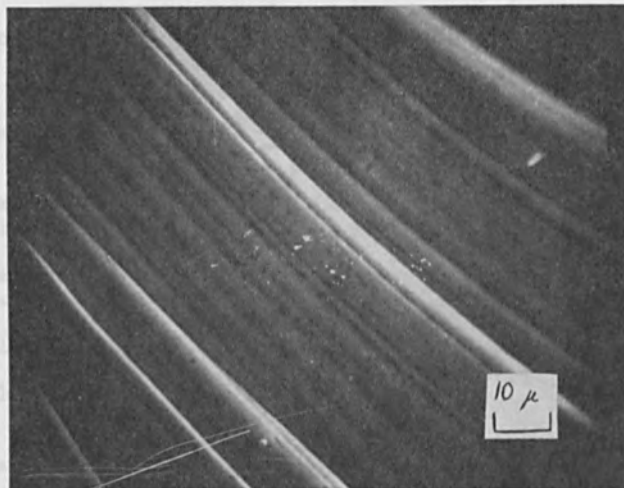


Figure 3.
Featureless weld ripples observed in zinc. The dark patches are liquid film deposits resulting from the cleaning solution used after welding.

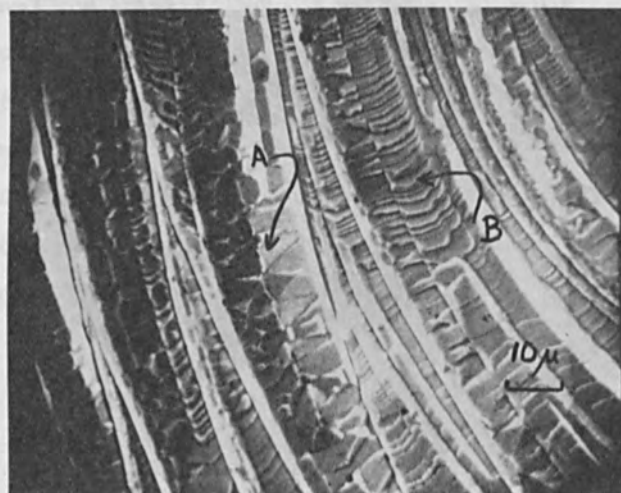


Figure 4.
Two types of weld ripples -- Faceted, at A and Ledge, at B.

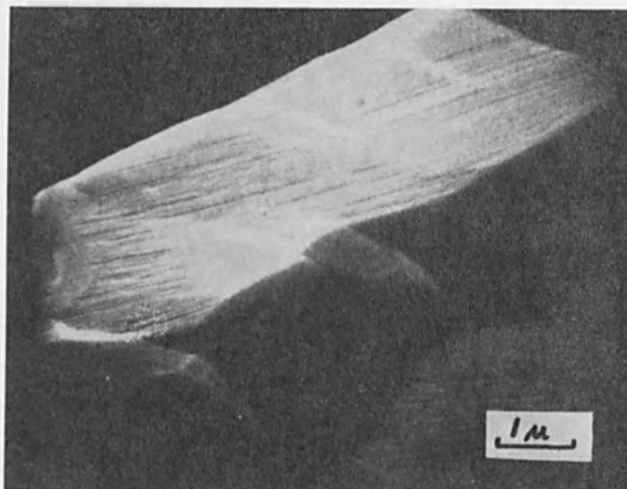


Figure 5.
Fine ledges present on the faceted ripples in zinc. Note that the ledges are all crystallographically oriented.



Figure 6.
Coarse ledges observed in zinc.

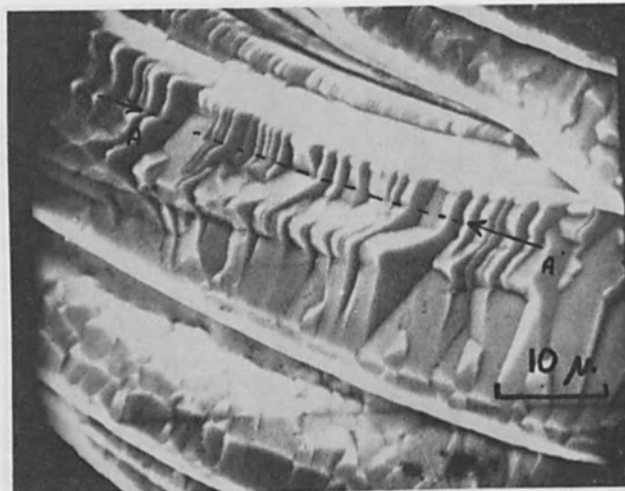


Figure 7.
Interruption of coarse ledge growth at A - A' in zinc.

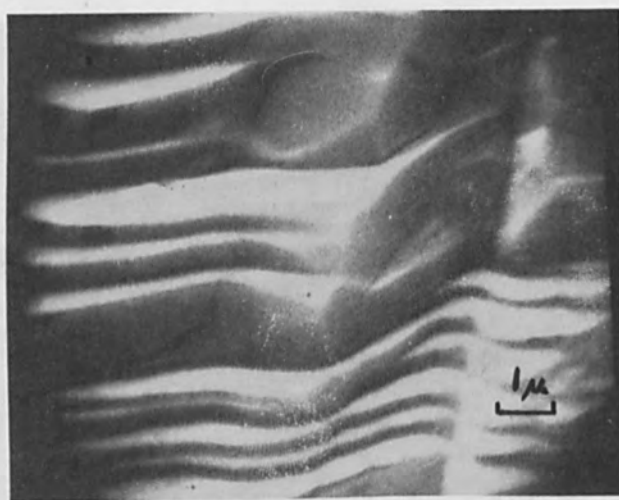


Figure 8.
Fine steps on coarse ledges within a weld ripple in zinc.

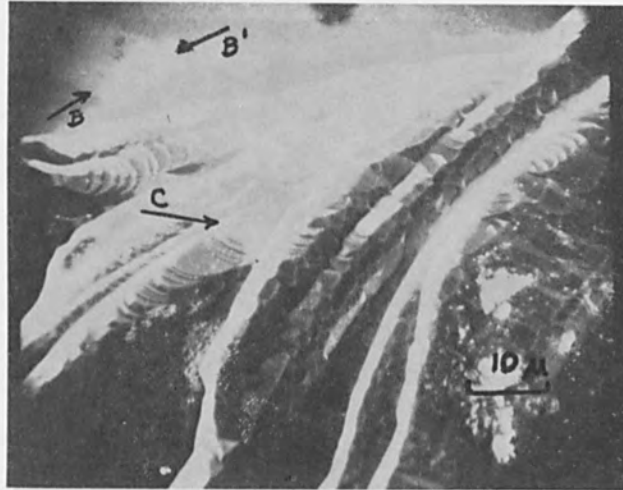


Figure 9.
Twin at B - B' traversing substrate and fusion zone proving epitaxy exists between substrate and weld zone. Note growth source at C. There is no noticeable growth change across the twin boundary.

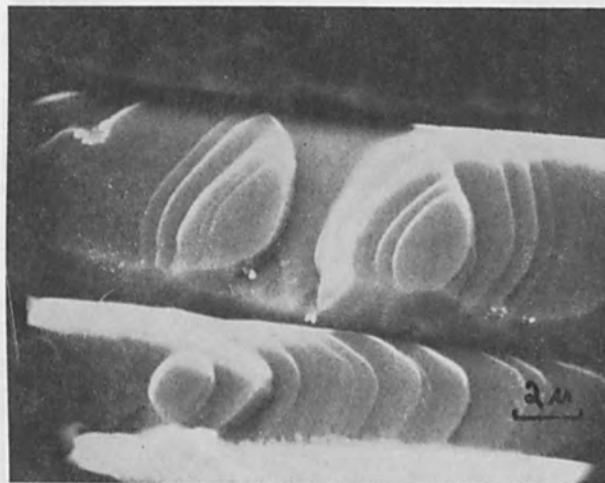


Figure 10.
Double and single growth sources shown to be generating "ledges" within a ripple.

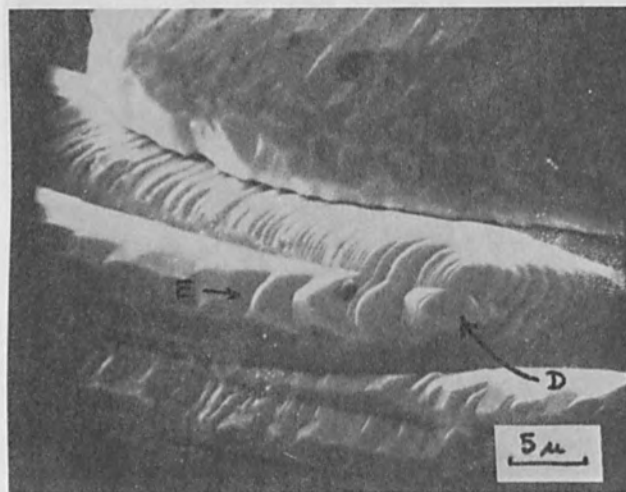


Figure 11.
Single growth source generating ledges along two ripples at D. Ledge to facet transition shown at E.



Figure 12.
Double growth source showing ledge generation with fine ledges at the side of the source.

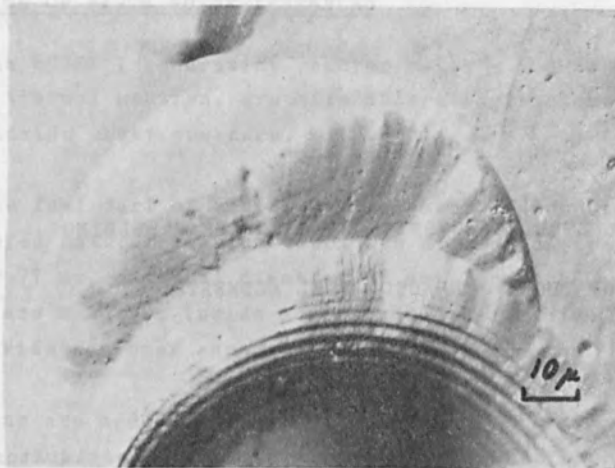


Figure 13.
Optical micrograph of a pulsed weld showing evidence of very fine ledge growth.



Figure 14.
Micrograph showing the same general area as 13 in a vapor coated crystal. Note coarse ledge growth.

TECHNICAL STUDY CONCERNING ELECTRON BEAM WELDING
OF BUGEY I REACTOR FUEL ELEMENTS

MM. Marc MARQUET Commissariat à l'Energie Atomique
Département de Développement
des Eléments Combustibles
Centre d'Etudes Nucléaires de Cadarache

Marcel MICHEL Société Industrielle de Combustible
Nucléaire

Bernard SAVORNIN Compagnie pour l'Etude et la Réalisation
de Combustibles Atomiques

Summary

Electron beam welding has been agreed for the natural uranium magnesium-canned fuel elements assembly of the CO₂-cooled and graphite moderated BUGEY I reactor. Problems came from electron beam welding process (vacuum, accurate beam position) and seam characteristics (low fusion temperature, high thermal conductivity and vapor pressure, heterogeneous geometries). Non destructive processes control tightness, health, penetration.

1 - DEFINITION OF PARTS TO BE ASSEMBLED

The BUGEY I generating reactor, with a 500 MW electric power, is a thermal neutron, graphite moderated reactor, cooled by carbon dioxide under pressure.

The fuel part of the BUGEY I element is composed of a lightly alloyed natural uranium tube. Its dimensions are : internal diameter 77 mm, external diameter 95 mm, length 540 mm. Circular grooves are machined inside and outside the uranium tube, in order to provide a proper anchoring of cans on uranium.

Cans are made of a magnesium alloy (0,7 % zirconium). They include, outside for the external can, inside for the internal can, trapezoidal fins shaped like a V which enable an adequate cooling by carbon dioxide under pressure. These fins have following dimensions : height 8 mm, length about 35 mm. The can body is 2 mm thick.

Two magnesium alloy plugs (thickness : 5 mm) are welded at the ends of both cans. Each element, thus made up, is placed in a graphite jacket (external diameter 220 mm, thickness 28 mm, length 604 mm) through a zirconium alloy holder and four centering pieces which are part of external can (fig. 1). The graphite jackets are then piled up in the vertical channels of the reactor which contains about 12 000 fuel elements.

2 - SELECTION OF THE ELECTRON BEAM WELDING PROCESS

The stress with which the cartridge rests on its holder is the sum of its weight and of stress due to the cooling gas pressure loss. As a matter of fact, the cooling gas runs downwards. The intensity of stress in the plug is obtained through the distribution of this stress on the plug.

The tungsten inert gas welding of the plug on cans is not deep (fig. 2) (pénétration inferior to 1,5 mm). The welded area should, preferably, not support the assembly. This is equivalent to a reduction of the support surface, therefore to an increase of the intensity of stress in the plug. This intensity of stress is then too high to induce the plug to stand creep satisfactorily, for the 20 000 hours during which the fuel element has to remain in the reactor. The development of our studies led us to bind both cans to fuel by an intermediate aluminium layer. A good binding has to be performed under vacuum. Logically, we were led to the electron beam welding process. This process enables to obtain a sufficient depth so that the whole cartridge extremity, including the welded area, may be used to hold the cartridge. Indeed, it is then possible to machine the welded area in order to obtain a plain face. Under these conditions, the obtained intensity of stress. (25 g/mm²) enables to stand creep satisfactorily during 20 000 hours.

Taking these considerations as a basis, such a welding was studied, in order to produce a tight welding, the depth of which would be as constant as possible, and would not exceed the plug thickness, namely 5 mm.

3 - PERFECTING OF THE ELECTRON BEAM WELDING TECHNIQUE

3.1 - Welding under rough vacuum

The first magnesium welding tests were achieved under high vacuum, the residual pressure inside the enclosure ranging about $5 \cdot 10^{-5}$ Torr. The wall and equipment pollution by vaporized magnesium led progressively to extend the pumping duration this being not very compatible with the aim to achieve mass production. By achieving welding at various pressures, ranging from 10^{-5} to 10^{-2} Torr, we underlined the possibility to weld under rough vacuum, with results equivalent to those obtained under high vacuum. The use of geared pumps enables to reach rapidly (less

than 2 minutes) a $5 \cdot 10^{-3}$ Torr pressure ; this pressure was chosen as working pressure under manufacturing conditions.

Moreover, it was necessary to obtain, in the cartridge, a vacuum ranging between same values. Channels were machined on the plug cylindrical parts, in order to avoid defect formation in the weld, coming from an internal overpressure in the cartridge.

The number and shape of these channels were determined taking into account both following factors :

- the offered conductance has to be sufficient, so that internal pressure may be lowered at the level of enclosure pressure within a reasonable time.

- the magnesium volume taken away from the can-plug interconnection area has to be low enough to avoid the formation of a very grooved bead, during welding.

The diagram on figure 3 shows that a small number of channels is enough for conductance to be sufficient and to enable an equalization of pressures between the welding enclosure and the cartridge inside part, in about 3 minutes.

Moreover, we want to point out, as an example, that 0,2 mm deep grooves obtained by drifting or milling, regularly spaced 2 mm apart on the plug circumference, correspond to a sufficient low matter removal for the over mentioned defect (fig. 4) not to appear.

3.2 - Finding the welding parameters

Taking a well known comparison element, one may consider that it is more difficult to obtain a safe welding bead in magnesium than in stainless steel.

Indeed, some physical properties may cause a bad filling by liquid metal, of the hole produced when the beam is passing on.

The low melting point, vaporization heat, and density, enable the formation of a hole showing :

- an important ratio depth/width (at least twice higher than in stainless steel with same power density), therefore not very favourable to a good filling.

- correlatively, a great lateral surface favourable to a liquid metal cooling, the latest being all the more rapid as material thermal diffusivity itself is high.

Moreover, solidification is accelerated by evaporation, since the liquid vapour pressure is higher than the pressure in the enclosure (2 Torr at 650°C). This may involve the formation, in the bead, of shrinkage cavities and on the surface, various defects such as a median furrow, the depth of which is unacceptable. Our tests showed that it was possible to avoid such defects by adopting a relatively low welding speed ; the latest was finally determined as being about 0,3 cm/s⁻¹. Correlatively, necessary power is ranging about 700 W, to obtain the desired penetration.

If one increases the welding speed, to keep same penetration, power has also to be increased. Experiment has shown that melting bath was unstable. Figure 5 gives a typical example of this unstability.

3.3 - Use of a vibrated beam

It appeared that although low speed welding enables to obtain a safe bead, the latest is still very thin ; it is, namely

in the shape of a triangle, the height of which is long compared with the base (fig. 6). This could lead to 2 mm losses of welded parts for an angular displacement of the beam axis reaching 0,2 mm compared with the joint plane. Taking the diameters of pieces to be welded into account, the elimination of this alignment defect of the beam axis on the joint plane could not be reasonably considered, in an industrial manufacture, just by achieving mechanical improvements concerning a very precise machining of cartridge ends and rotating feed devices.

The problem was solved very satisfactorily by giving to beam a vibration perpendicularly to the joint plane, which leads to a broadening of the welding bead lower part. The determination of the vibration frequency, amplitude and shape formed the object of a particular investigation :

- Frequency and amplitude

The amplitude is calculated so that the bead width measured on the surface of pieces is always about 3 mm. This leads to two possible frequency ranges, according to the control possibilities of the operating electronic beam generator :

- . a so called "low frequency" range (3 to 20 Hz)
the vibration amplitude being inferior to the spot diameter
- . a so called "high frequency" range (1000 Hz) the
amplitude being superior to the spot diameter
(fig. 7 and 8)

- Shape

It is useful for controlling the repartition of energy received on the bead width. In low frequency, the use of a common

signal sinusoidal shape results in the formation of an irregular welded area; the latest is eliminated by using a signal, the type of which is represented on figure 9 ; the time during which this signal is maintained on zero is adjusted. In high frequency, the sinusoidal vibration is suitable, the best appearance being obtained when amplitude is about twice the spot size which can be seen.

As a conclusion, the following main welding parameters were selected :

- vacuum in the enclosure : 5.10^{-3} Torr
- average linear drift speed of pieces : $0,3 \text{ cm/s}^{-1}$;
this speed is programmed to keep a constant penetration.
- power about 700 W, with progressive set up for starting
(without or with bath heating-up period)
- beam vibration : 15 Hz or 1000 Hz according to the
type of beam used.
- speed programming associated to power decreasing at the
end of the cycle, in order to avoid formation of a
crater.

Figure 9 gives the vibration shape of the electron beam, the power and speed diagrams, and a macrography of the welded area obtained for a 15 Hz frequency ; figure 10 gives the same information for a 1000 Hz frequency.

Figure 11 shows a BUGEY fuel element after welding and machining of ends.

4 - INDUSTRIAL APPLICATION AND NON DESTRUCTIVE TESTINGS

4.1 - Non destructive testings

We had then to be in a position to achieve an industrial manufacture in reasonable operating conditions.

First of all, various non destructive testings were perfected for these manufactures. The tightness checking of the completed cartridge is achieved by helium immersion and detection by means of a mass spectrometer.

Moreover, an X ray radiography method was developed to check penetration and possibly the good welding state. But this method is limited, as regards sensitivity. On the other hand, a very suitable process for immersion ultra-sonic penetration checking by oblique beam just after welding and before machining again, was achieved.

These works formed the object of developments parallel to the welding investigation.

4.2 - Industrial application

The observations taken during the above mentioned welding tests, as well as the out of pile tests on cartridges led us to specify a welding penetration included between 3 and 5 mm, this distance being counted from the plug face which has been machined again.

It was therefore important to make sure that under these normal industrial operating conditions, the $3 \begin{smallmatrix} + 2 \\ - 0 \end{smallmatrix}$ mm limit values could be maintained.

The refuse threshold at 3 mm was revealed by the ultra-sonic diagram ; the fact that penetration had not been extreme was revealed by X ray radiography (appearance of uranium inclusion in the welding).

A statistical variance survey of welding parameters on fifty cartridges, namely 200 weldings, was conducted with systematic parameter recording, ultra-sonic, radiographic and macro-graphic checking of obtained weldings.

It showed that, to avoid underpenetration as well as overpenetration, it was suitable to work inside the power band included between following values : $650 \text{ W} \pm 40 \text{ W}$, which corresponds to a $\pm 6 \%$ maximal possible power variation. This allowed power variation leads to a ± 100 volts possible variation on high voltage, namely about $\pm 0,6 \%$.

Industrial machines were therefore designed in order to provide an accurate control of voltage, focusing parameters and possibly of power, to enable the operator to check continually that he remains in the allowed range.

A preliminary series of 300 cartridges, namely 1200 weldings, was achieved in confirmation of the manufacturing process. Less than 3 % cartridges were rejected because of welding.

Lastly let us remark that, besides the previously mentioned accuracy requirements, the investigated industrial machines were designed to comply with cadence requirements, namely 200 weldings per working unit. The adopted machines own an intermediate vacuum chamber whereas the gun, which is independantly pumped, is under high vacuum, in order to reduce the enclosure pumping times. The gun remains permanently under an about 10^{-5} Torr high vacuum, whereas the enclosure vacuum on the piece is about 10^{-2} Torr before welding. The enclosure is equipped with necessary mechanisms to enable to achieve at least two weldings together, without breaking vacuum. The gun is provided with easily approachable and removable protection mechanisms in order to avoid pollution by magnesium condensations.

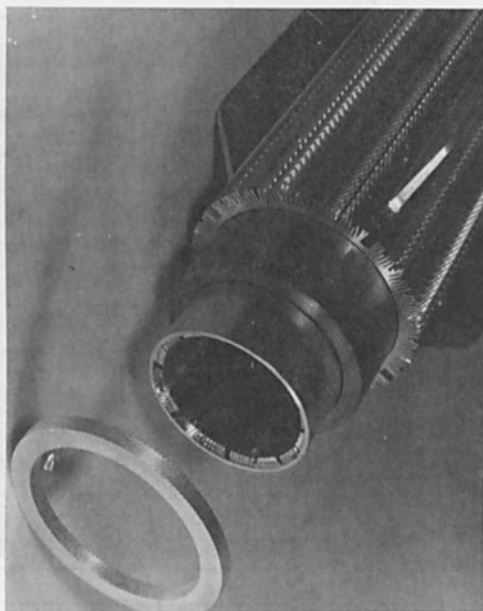


Fig. 1. General view of the fuel element.

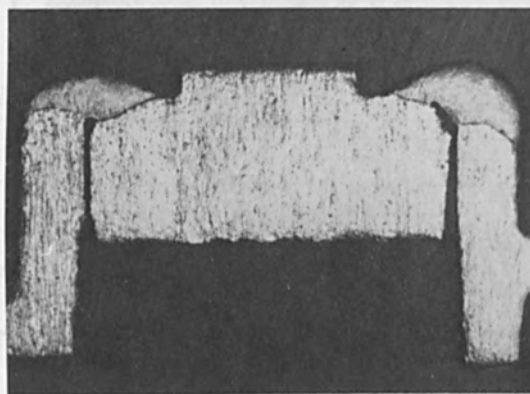


Fig. 2. Macrograph of a TIG welding.

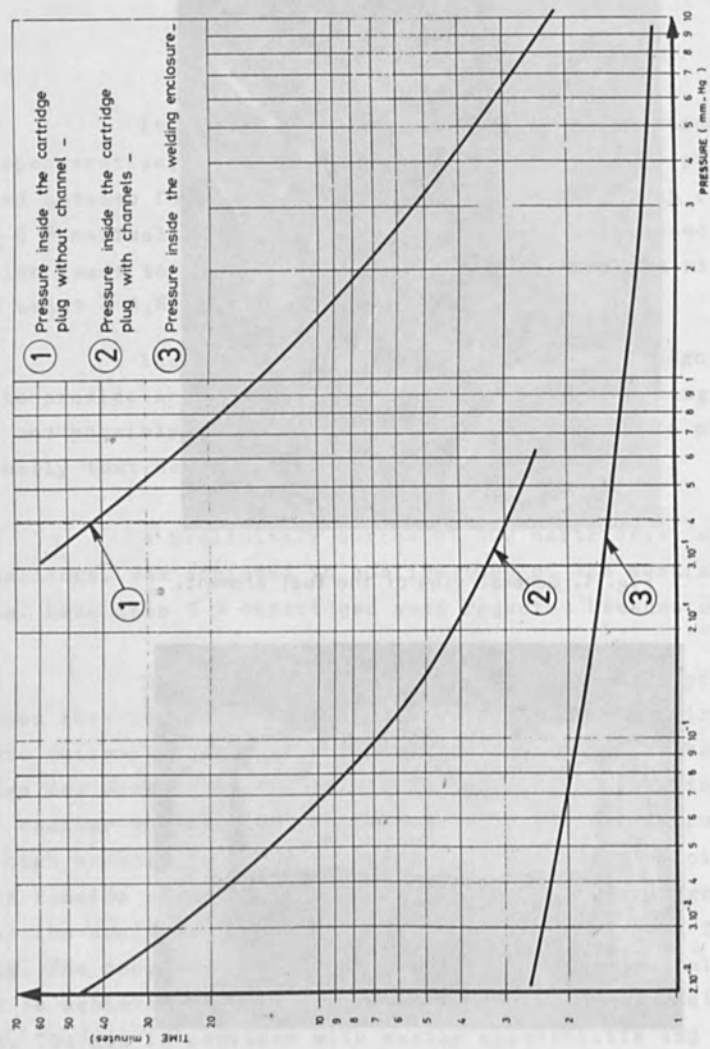


Figure 3.

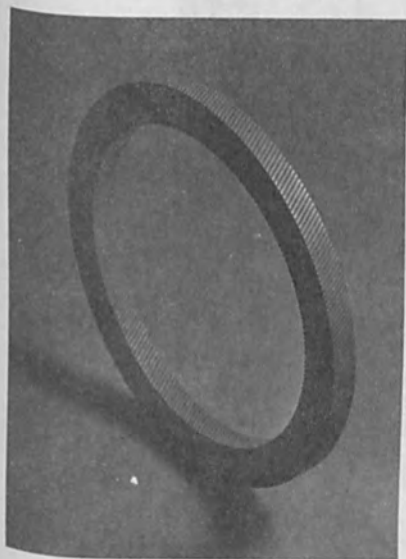


Figure 4.

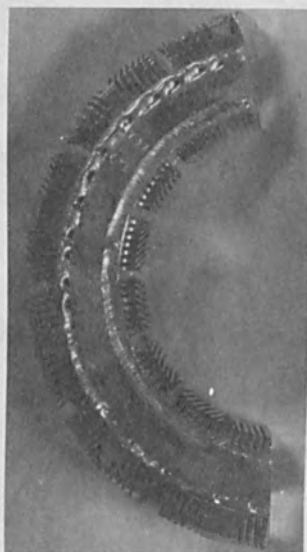


Fig. 5. Typical view of an unstable bath ($1000 \text{ w} \cdot 0,6 \text{ cm} \cdot \text{s}^{-1}$).

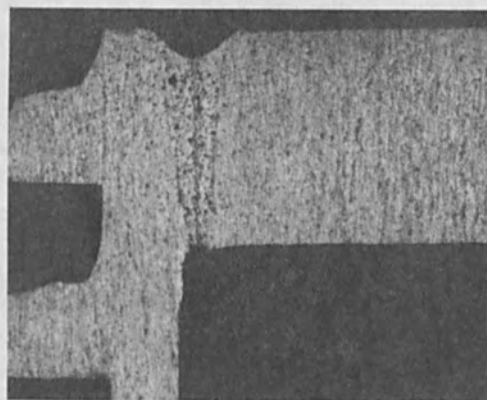


Fig. 6. Macrography of an E.B welding with no vibration of the beam.

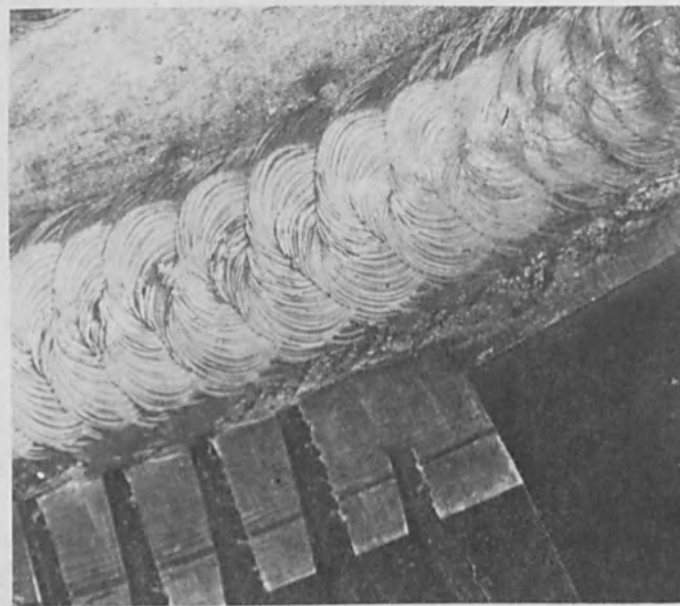


Fig. 7. Solidification of a "low frequency" welding.

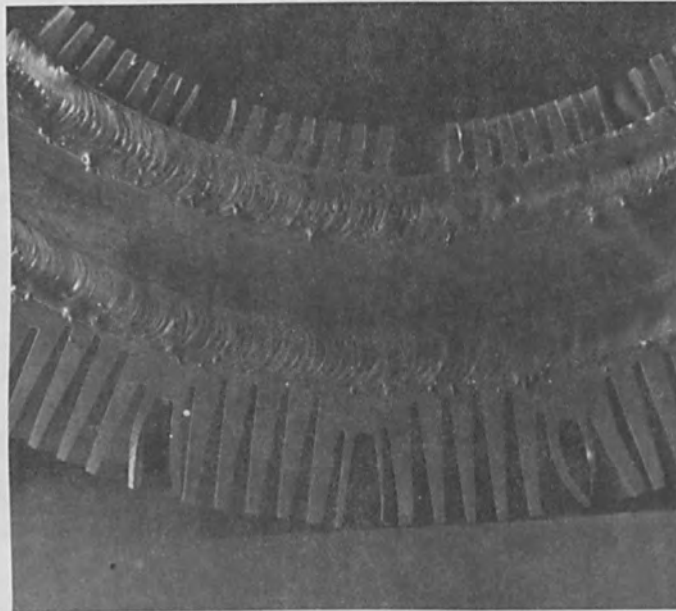


Fig. 8. Solidification of a "high frequency" welding.

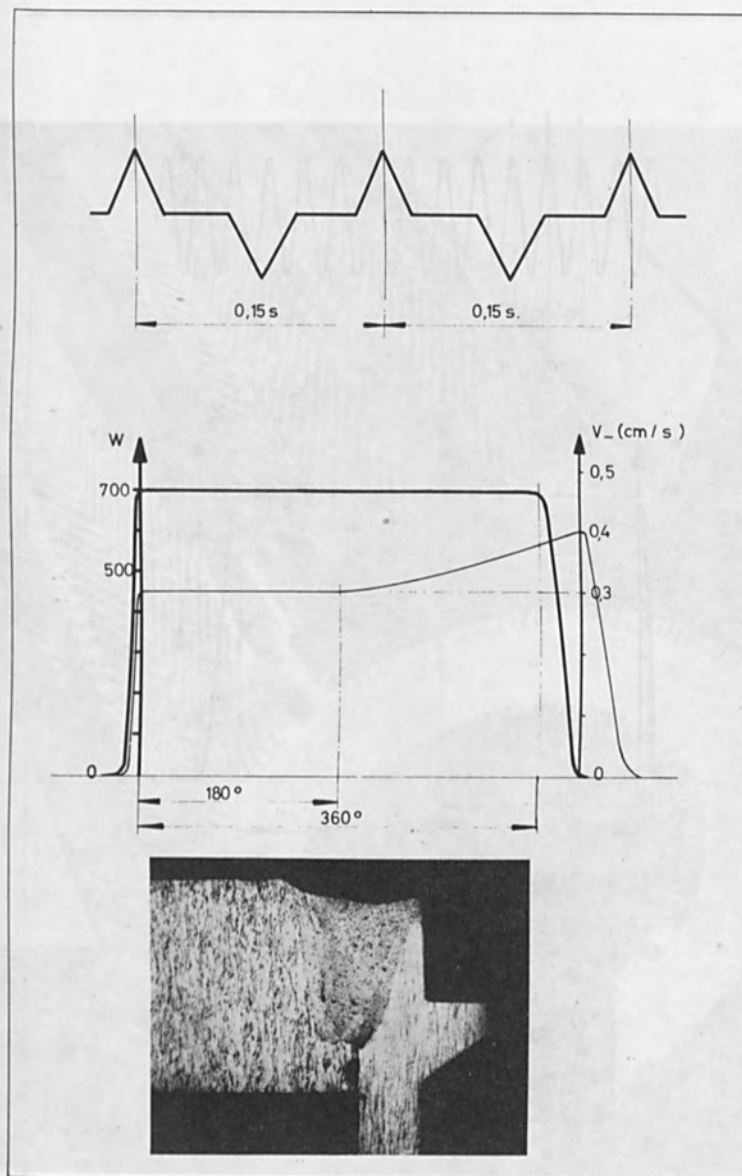


Figure 9.

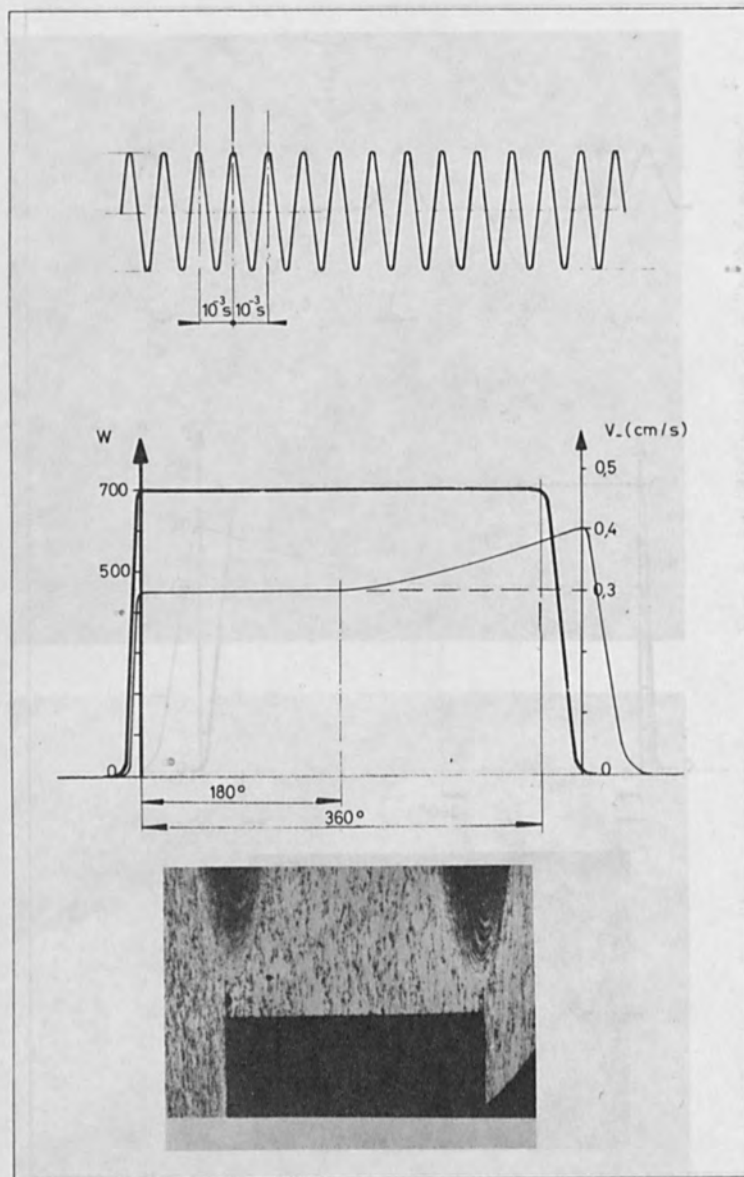


Figure 10.

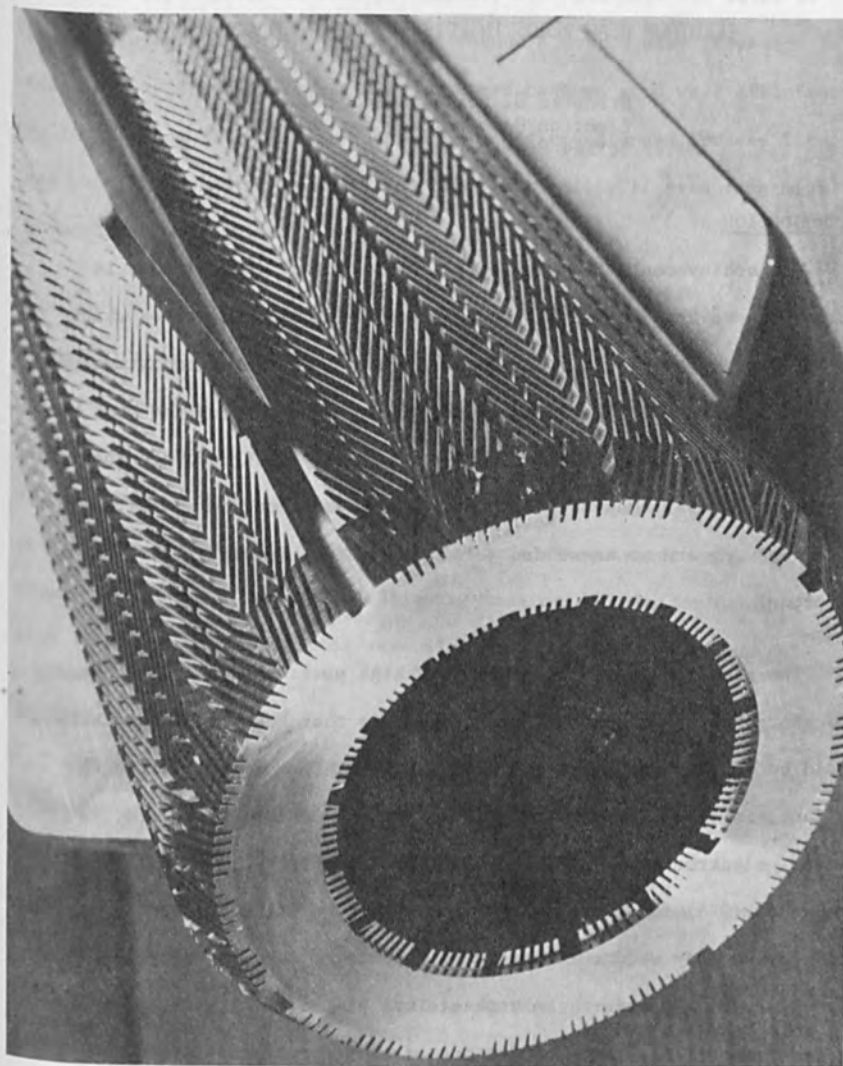


Fig. 11. Welded and tooled fuel element.

EXPERIMENTAL, AS-WELDED, PROPERTIES OF OUT-OF-VACUUM
ELECTRON BEAM WELDS IN 2219-T87 ALUMINUM ALLOY*

Frederic D. Seaman, Manager
Electron Beam Welding Systems
Sciaky Bros., Inc.

INTRODUCTION

The achievement of a high efficiency weldment is desirable in structures related to the space, defense and atomic industries which are characterized by their efficient use of materials. Such structures usually involve welding. Such welded joints must reflect the same high mechanical efficiency that is characteristically designed into the rest of the structure. Many structures of aluminum cannot be heat treated after welding and so as-welded joint properties, in particular, become important.

The electron beam processes offer high power densities and, hence, low energy inputs. This situation suggests that heat-treated materials could be welded by an electron beam with a minimum loss of strength. In fact, in one study of the 2219-T87 aluminum alloy, as-welded, in-vacuum, electron beam welds broke at 49-51,000 psi while single pass arc welds (GTA) failed below 45,000 psi-average 40,000 psi.⁽¹⁾ However, the size of many weldments precludes the use of the in-vacuum electron beam process. Therefore, an emphasis was placed in this study on as-welded properties produced under out-of-vacuum conditions.

*Work performed while author was associated with Westinghouse Electric Corporation.

This paper presents some preliminary strength data for welds in 2219-T87 aluminum made by the out-of-vacuum electron beam process. In addition, weld soundness was also observed because good weld efficiency implies freedom from defects as well as high strength and process latitude has been investigated to establish the practicality of producing high strength welds consistently.

DESCRIPTION OF THE EQUIPMENT

The Westinghouse portable out-of-vacuum equipment used in this program is shown in Figure 1. The 12 KW (150 KV) high voltage power supply, diffusion pumps, and the electron optical system have been packaged into a single compact, mobile piece of equipment weighing about 350 pounds. Two other stationary machines of similar design with 6 KW and 9 KW capacities were also used during this program.

Principle of Operation

In this type of equipment (Figure 2), electrons are released from the cathode of the gun in a region of high vacuum and they are focused through differentially pumped orifices by a combination of electrostatic and electromagnetic electron optical systems. They are subjected to increasing pressure and ultimately pass through the exit orifice into an atmosphere that is at ambient pressure. A more complete description of the principles through which beam scattering is minimized and of the basis for other design features will be found in References 2 and 3.

Effluent Gas Flow from Beam Exit Nozzle: A positive pressure is applied at a special gas protection orifice which is located just below the exit orifice of the gun, producing a flow of gas out of the exit orifice toward the work area. Such a flow directs contamination emanating from the work away from the pumping orifices. A portion of this gas also is drawn into the column. The exact balance between the portion that flows into the gun and the effluent gas flow from the gun depends upon the size of the orifices. This balance was shifted several times during the program. During these tests we found that certain rates of flow produced one type of weld while different flow rates produced a distinctly different appearance in weld cross section. We also found that, within certain limits, a narrow weld with high mechanical efficiency could be produced.

MATERIAL

Aluminum alloy 2219-T87 was selected because comparative arc welding data is available from other aerospace programs. Nominally the alloy has an ultimate tensile strength of 46,000 psi (12% elongation). The 2219 alloy has been used for large complex structures where the narrow bead, minimum heat effect, and low distortion characteristics of electron beam welding would be technically and economically attractive, but which are too large to fit into conventional vacuum chambers. The composition of the alloy is listed in the following table:

TABLE I

NOMINAL COMPOSITION OF 2219 ALUMINUM⁽⁴⁾

	<u>Si</u>	<u>Fe</u>	<u>Cu</u>	<u>Mn</u>	<u>Mg</u>	<u>Zn</u>	<u>Ti</u>	<u>V</u>	<u>Zr</u>	<u>Other</u> <u>Total</u>
Typical Percent	0.20	0.30	5.8/	0.20/	0.02	0.10	0.01/	0.05/	0.10/	0.15
(Maximum unless otherwise stated)			6.8	0.40			0.02	0.15	0.25	

All welds produced were in 0.250 inch thick material, except where specifically noted.

The helium gas (-90°F Dew Point) was employed as an effluent gas flowing from the beam exit nozzle and also as a shielding gas applied to the surface of the weld by the device shown in Figure 3.

EXPERIMENTAL TECHNIQUE

Control of the effluent gas flow was accomplished by a precision flow meter calibrated at the gas pressure that was delivered to it from the helium gas cylinder. The value recorded experimentally represented all of the gas that was delivered to the gas protection orifice located just below the exit orifice of the gun. This value was then related to an observed change in weld configuration and reported in terms of units of flow, or "transition flow rate"*, above or below such a change. The measured flow quantities varied somewhat because of a shift in balance between the gas flowing out of the gun and the gas flowing toward the pumps. A direct measurement of the gas impinging on the workpiece during welding could not be made.

* The "transition" or "transition flow rate" is the minimum helium flow rate at which a narrow weld could be produced for a given beam nozzle diameter.

In addition to the effluent gas flow described above, the welds were protected by an envelope of inert gas. The envelope was generated by a diffuser shield (Figure 3). Flow rates were, typically, 120-170 CFH.

The welds produced in this program were subjected to radiography at the sensitivity level of 2-TT**. For bead-on-plate welds, which were used to check porosity or process latitude, only the upper surface of the plate was scraped to remove heavy mill oxide. This was done to improve metal flow. The oxide was left on the under surface to provide a source of hydrogen from moisture and hydrocarbons entrapped in it. In reading the radiographs, data were taken from a 1-1/2" length of weld. The first 1/2" of weld was disregarded as was the last 1/2" of the weld. This was done to avoid end effects caused by the entry of the process onto the plate, and by its termination in the plate. Heavily undercut (gouged) welds were not subjected to individual interpretation if it appeared from the film that the joint thickness was beyond the limits of radiographic sensitivity. All visible porosity was noted by recording the number, the distribution, and the size of the pores.

Welds used to produce mechanical data were cleaned on both sides of the joint by shaving the surface with a sharp blade. Two specimen configurations were used. All mechanical test specimens were cut from

** That is, using an appropriate penetrameter with a thickness equal to 2% of the specimen being radiographed and adjusting the radiographic procedure so as to define a hole in the penetrameter with a diameter equal to twice the penetrameter thickness.

12-inch long square butt welds. However, data relating to process latitude and the occurrence of porosity was gathered from short bead-through-plate welds. The butt welds were prepared by milling the edges and typically exhibited gaps of 0.010 inches.

A stainless steel weld fixture was employed for holding the workpiece during the welding of the joints for tensile tests. Backup gas was used with this fixture. Hold-down to the fixture was not required on low speed butt welding of any material thicknesses. At high welding speeds (over 100 ipm) it was necessary to clamp the material to the backup. Tacking also was necessary on all 1/4" plate butt joints to assure consistent joint opening and thus weld configuration. Tacks were made by using the welder to fuze short lengths on either end of the joint. Bead-through plate welds were fixtured in such a manner that the tooling did not influence the temperature of the workpiece appreciably.

All mechanical testing was accomplished on a Wiedemann Mark G, 60,000-pound capacity, screw-driven universal test machine. Tensile elongation was measured by means of a deflectometer which translates the mechanical motion of the cross head to an electrical output which, along with load, can be recorded to give an autographic load-elongation curve. The tests were run at a cross head speed of 0.005 inch/inch/minute through the 0.2 percent offset yield and then increased to 0.05.

Power measurements represent the actual power applied to the workpiece surface as measured by directing the beam into a Faraday cage.

RESULTS

Prior to this program, tests with a 6 KW welder established several procedures adoptable to 0.250 inch aluminum. The beam width was not measured during the establishment of these procedures. No attempt was made to control weld width.

Weld Strength Study On Wide Weld Configurations

From these early tests, a procedure that represented the lowest total heat input (kilojoules/inch of weld/inch of thickness) to the workpiece, and which also produced the radiographically acceptable weld quality, was devised. A series of 12 inch long weld joints was made in the 1/4" material using the selected procedure. The procedure was as follows:

Power Transmitted To Work	Work Distance	Weld Speed	Heat Input
5.1 KW	9/16 inch	45 IPM	27 Kj/in./in.

The resulting welds exhibited cross sections similar to Figure 4. All welds produced under these conditions met the appropriate radiographic inspection requirement without exception. A typical X-ray and weld cross section is shown in Figure 4. These weld panels were cut and tensile specimens removed. Mechanical tests were made with the weld bead present and also with the bead shaved to base metal thickness.

The average ultimate strength of the joints in the as-welded condition without the weld buildup was 42,500 psi. Weld buildup that occurred with the selected parameters was a maximum of 0.020 inch total on the face and root. Even this minor amount, however, increased aver-

age strength by approximately 2000 psi.

Weld Strength Study On Narrow Welds

The full capabilities of the 12 KW Westinghouse out-of-vacuum welder were then applied by increasing the power as near to the 12 KW maximum as the 280 inch per minute, (the maximum speed of available manipulators) would permit. Based on earlier corporate tests, (5) an attempt was made to control the environment between the beam exit nozzle and the surface of the workpiece. This was done by controlling the effluent gas flow values to produce a narrower weld than that shown in Figure 4. These flow values varied from time to time because of changes in the equipment. A typical flow was 120 CFH (helium) into the welder. Some of this exited from the welder to create the condition noted above and the remainder was pulled into the low pressure region column of the welder. The permissible range of gas flow rates required to produce acceptable welds was about 2 CFH. Excessive flow had to be avoided since such flows left a rough bead or even reduced penetration (by chilling at long work-gun distances). Accelerating voltage was maintained at 150 KV. Distance between the exit nozzle and the workpiece was reduced to 1/4", although this change alone was not capable of producing narrow welds if gas flow was not controlled.

Using this more powerful welder, the speed-power combination obtained at maximum positioner speed resulted in the lowest heat input experienced in the program (8.35 Kj/in./in.) and narrower welds. Narrow welds were also produced at 10.8 Kj/in./in. and 12 Kj/in./in. A cross section from this series of welds is shown in Figure 5. Depth-

to-width relationships observed in these welds were as follows:

TABLE II

DEPTH TO WIDTH RATIOS

<u>Weld Power to Work (KW)</u>	<u>Weld Speed (IPM)</u>	<u>Width (Inches)</u>	<u>Depth/Width Ratio</u>
5.1	70	0.150	1.67
7	140	0.135	1.85
9.75	280	0.100	2.5
10	240	0.110	2.27

A gun to work distance of 1/4 inch was the maximum at which the weld narrowing could be achieved by gas flow adjustment. Welds produced at this point were very similar in appearance to those produced at 1/8 inch work distance as shown in Figure 5.

As will be discussed in another section of this report, whenever the narrow bead phenomena was achieved on the 1/4 inch plate material, very low porosity levels were also observed. Thus, high-quality, high-speed and low-energy welds were produced for tensile evaluation.

Tensile data for these joints is shown in Table III. Two results are evident. First, a number of the weld specimens showed the high strength (47,000-49,000 psi) expected⁽¹⁾ based on the low heat input involved. Second, a wide range of strengths was evident between welds made at similar heat input values and also in samples in the same weld. This strength variation is attributed to the contour of the weld face. When welding with the narrow beam phenomena, the weld surface often contains ripples, some of which are below the original surface, causing a notch. The weld cross section shown in Figure 6 illustrates this

point. The figure shows the undercut effect and grain orientation of weld number 504. This weld had low test values apparently related to premature failure. Weld number 508 is shown in Figure 7. This weld was produced at similar conditions and heat input levels, but had higher measured tensile strength. This weld had a surface without the weld edge effects.

TABLE III

AS-WELDED TENSILE STRENGTH OF 1/4 INCH 2219-T87 ALUMINUM
WELDED BY THE OUT-OF-VACUUM ELECTRON BEAM PROCESS

Weld No.	Weld Energy (KW)	Speed (IPM)	Heat Input (KJ/in/in)	Ult. PSI	0.2% YS PSI	% Elongation	
						1 In.	2 In.
420-1	9.75	280	8.35	42,900	- - -	5	2.5
-2				27,700	46,900	4	2
421-1	9.75	280	8.35	41,000	- - -	4	2
-2				40,300	- - -	4	2
504-1	9.75	280	8.35	41,700	39,800	- -	1.5
-2				37,700	- - -	- -	1.5
508-1	9.75	280	8.35	49,200	40,500	- -	1
-2				47,500	40,600	- -	1
465-1	6.3	140	10.8	45,080	40,850	- -	3
-2				46,950	40,920	- -	3
467-1	6.3	140	10.8	47,190	41,570	- -	2.5
-2				46,950	40,920	- -	3
-3				44,160	40,960	- -	2.5
470-1	7	140	12	45,720	41,090	- -	2.5
-2				40,190	40,110	- -	1.5

In making these longer joints, it was found that the narrow bead phenomenon could be readily produced in a series of plates.

Process Latitude Study

These results prompted a study of the narrow weld process latitude with respect to gas flow and welding speed for any given power input. 150 KV was used as the accelerating voltage in this series. Gun-to-work distance was established at 1/8 inch for these tests.

As a starting point for these studies, the types of welds that can be produced by varying speed and exit gas flow over a wide range at a given power level were classified as shown in Table IV:

TABLE IV
WELD CATEGORIES

<u>Name</u>	<u>Description</u>
Type A	<u>Wide Welds</u> , Penetration through the full workpiece thickness (Figure 4).
Type B	<u>Wide Welds</u> , Incomplete penetration of the workpiece.
Type C	<u>Narrow Welds</u> , Penetration through the full workpiece thickness (Figure 6).
Type D	<u>Narrow Welds</u> , Incomplete penetration of the workpiece.
Type E	<u>Narrow Welds</u> , Undercut (gouged) upper bead surface; penetration through the full plate thickness.
Type F	<u>Narrow Welds</u> , Undercut alternating with regularly spaced sectors of normal or excessive crown on the upper surface of the bead; incomplete or marginal penetration of the workpiece.

A plot (Figure 8) was made with the exit nozzle gas flow as the vertical axis and welding speed as the horizontal axis for the 7 KW (150 KV) process and 1/8" gun-work distance. Each weld was entered on this plot using one of the above symbols (Table IV) which described its character (i.e., whether it was a wide weld such as Figure 4, a

narrow weld, Figure 5, or some other configuration). The various weld categories appeared to fall into specific areas and approximate boundaries were sketched in with dotted lines. Probably these lines represent transition zones rather than abrupt configuration changes.

This sorting process revealed the boundaries (Figure 8, Area A-B-C) within which one may expect to obtain the type of weld that resulted in high breaking strengths in Table III.

From a practical point of view, Figure 8 suggests that at 7 KW, the narrower weld process (Area A-B-C) for 1/4 inch aluminum is more tolerant of a variation in speed than it is of a variation in gas flow. The shape of the area A-B-C suggests that tolerance for gas flow variation might be greater in the higher speed regions. The boundaries are actually reflecting decreased ability to assure that a given weld configuration will result. The location of these boundaries suggests that the optimum gas flow was 2 to 4 CFH above the "transition flow rate" at low speeds (115 to 130 ipm) and 5 to 7 CFH at high speeds (130 to 140 ipm).

Similar plots were made for the 1/4 inch thick material at 5 KW, 4 KW and at 3 KW. The narrow weld process area corresponding to ABC in Figure 8 decreased in its speed dimension and flow rate dimension until at 3 KW no acceptable narrow welds could be produced in 1/4 inch plate at any combination of settings. Figure 9 illustrates the effect of welding power on narrow weld process latitude. At these low-power

tests, orifice wear was negligible. Under these conditions, the absolute value of the metered gas input could be measured. It was found to be 88 ± 1.3 CFH.

Returning to the 7 KW (510 KV) process, the effect on process latitude of increasing plate thickness from 1/4" to 1/2" was observed (Figure 10). It would appear that the maximum thickness at which a narrow weld could be made, using 7 KW, with any assurance, was between 3/8" and 1/2".

Figure 8 is a composite of several experimental series wherein the change from a wide to a narrow weld cross section actually occurred at different gas flows. This variation resulted from changes in nozzle configuration that were either part of the overall program or that resulted from wear. Accordingly, the gas flow data, Fig. 8, was plotted using the transition flow rate as a common reference point to relate one experimental series to another. In order to determine if the transition was, in fact, unaffected by increased process speed (or by changes in power level), sets of data closely bracketing the transition were selected. These sets come from two nozzle conditions as indicated in the following table:

TABLE V

<u>Nozzle</u>	<u>Power</u>	<u>Speed</u>	<u>Gas Flow to Produce A</u>	
			<u>Wide Weld</u>	<u>Narrow Weld</u>
A	3	45	86 CFH	88-1/2 CFH
	4	60	85	87-1/2
	5	80	85	87-1/2
	7	127.5	85	87
		Approximate transition		86-87 CFH
B	7	145	117	123
	7	157	115	120
		Approximate transition		117-120

This review of available information does not indicate that changing power level has any effect on the relationship between flow and weld configuration. Nor does the data indicate that changing welding speed for a given power level affects the flow rate at which transition takes place.

An X-ray comparison of the 6 KW (wide) and 10.5 KW (narrow) welds from which the tensile data was obtained has suggested that there was a relationship between the occurrence of porosity and the welding procedure.

Porosity Study

In order to display any relationship which might exist between porosity and the major process variables of power, welding speed, gas flow, workpiece thickness, and working distance, the existence of porosity in a particular weld was marked on reproductions of the process parameter plots as used in the preceding sections. A vertical cross was made over the symbol of those welds which showed porosity. The symbols of clear welds were left unchanged. Figures 11, 12 and 13 represent these data for a 7 KW (150 KV) procedure operating on plates of increasing thickness.

The most remarkable result (Figure 11) is that no porosity was evident as soon as the process yields what is called a narrow weld. Even where the narrow welding process becomes marginal, as with thicker material, the level of porosity is low. Most of the narrow welds in 1/2" thick material (Figure 13) produced with 7 KW of power would meet conventional aerospace porosity standards and would easily meet the ASME Code for Unfired Pressure Vessels.

It seems that even some of the wide welds which were produced can be, and are, free of porosity. There is some evidence in Fig. 12 (7 KW data for 3/8" plate) that reducing the speed about 10% to 15% below the line C to E will produce porosity free wide welds. In this respect, wide welds perform in much the same way as conventional arc welds.

The number, distribution, and the size of the pores was noted for all welds. However, it was felt that the sample size (length of the weld) was too small to attempt to draw conclusions regarding trends in these effects. Therefore, only the presence or absence of porosity in the sample area is reported.

DISCUSSION

Controlling the environment (gas composition) between the exit nozzle and the surface of the workpiece by means of a controlled jet of gas from the beam exit orifice to the workpiece does, in fact, alter the appearance of the weld (Figure 4 compared with Figure 5). This is the most significant observation that can be made from the data presented in this paper. This paper is not intended to detail the mechanism by which the transition from the wide mode to the narrow mode may be accomplished. This was the subject of another paper (Reference 6). As discussed there, the electron beam has not shown a relationship between the observed change in the shape of the weld and any characteristic of the beam as delivered by the machine (i. e., peak power density or cross sectional area). Thus, it was postulated that the shield gas replaces the air between the beam orifice exit and the workpiece and may be displacing metal vapor from the beam path. Both mechanisms would reduce scattering.

The increase in the properties of welds produced when conditions are established such that a narrow-bead high-efficiency weld is obtain-

ed seems to be well supported by the data. Welds which are termed wide (Figure 4) and which require considerably more energy per inch (27 Kj/in.in.) average 42,500 psi breaking strength. Narrow-bead high-efficiency welds produced at one-half or one-third that energy (8.35 - 12 Kj/in./in.) can exhibit eight percent greater strength if the weld bead is free of undercut. This substantial increase in strength as energy per inch of weld (and weld width) is reduced bears out the original premise of the program -- that the high welding efficiencies possible with a beam of electrons could be used to join heat treated aluminum alloy 2219-T87 with a minimum sacrifice of strength in the weld joint.

Although the process does not achieve the very high depth-to-width ratios reported for other processes, it is a great improvement over other conventional fusion welding processes.

Of considerable practical importance in the operation of the process is the observation that the gas flow at which the weld shape transformed from wide to narrow falls on a straight line. In other words, this critical gas flow is constant and unaffected by speed and power. Therefore, while this gas flow may be the key to the narrow welding process, it is not a fundamental variable in the process, but a machine parameter.

Separating the various areas shown on Figure 8 by boundaries actually provides a two-dimensional manifestation of the narrow weld process and shows how it is affected by the gas flow and speed. The validity of these observations is enhanced by the fact that the

suggested boundaries lend themselves to reasonable explanation. As is the case with any fusion process, welds exhibiting lack of penetration become more prevalent as speed is increased, without regard to gas flow. Such a region is shown as D-B-C-E area at the right of Figure 8. Changes in gas flow complicate this phenomenon but still permit an explanation that is not too far removed from arc welding experiences. For example, as gas flow is increased above the transition line, a narrow weld that was not fully penetrating the plate can be expected to increase in penetration as shown by the slope of the line B, D. In simple language, the high speed gas-jet is blowing the liquid metal out of the joint and the heat source is affecting the bare metal more directly. In the range of low gas flow (below line TR in Figure 8) where wide welds predominate, the gas flow apparently chills the work-piece so that gas flow must be reduced if the speed is increased in order to maintain full penetration. This explains the negative slope of C-E. Shielding gas has a similar effect in the arc welding processes. As noted previously, the actual lines represent, only approximately, the location of the transition which is probably not very abrupt.

An extension of the line D-B down to A (Figure 8) on the transition-gas-flow-line encloses the area of acceptable high efficiency, narrow welds. Above line A-B excessive gas blows metal from the joint. This is harder to do as the process and solidification rate increase from A to B. Thus the line A-B has a positive slant and the process enclosed by A-B has an increased tolerance for excessive gas flow as speed increases.

The significance of the line T-R has been noted previously in the discussion and appears to relate to the effect of the effluent gas jet from the beam exit nozzle on the environment helium, the exit nozzle and the workpiece.

At point A (Figure 8), the low speed end of the process, it appears that a direct transition from gouging to cutting might occur, because the process has slowed to a point where gravitational effects begin to have an influence on bead contour equal to that of the effluent gas. In other words, at very low process speeds, the liquid metal probably falls out of the slot as a result of excessive energy input regardless of the effects of the gas flow. In fact, plots of tests at 5 KW showed such an effect. When this happens, a threshold speed is thereby established, below which (for a given power) narrow welds cannot be made.

The reduction in process latitude that results from reduction of welding power for a given plate thickness (Figure 9) can be best explained by reviewing the appearance of the 3 KW (150 KV) welds produced in 1/4 inch thick plate. The upper bead surface of those welds with penetration and those that lacked penetration showed intermittent undercut with alternating sections of normal or excessive crown. This condition defines a type F weld (see Table IV) though the degree was not as severe as often encountered at higher power levels. Apparently the rate of heat input is sufficiently high, and the rate of translation along the joint sufficiently slow, that by the time penetration is achieved, the balance necessary to maintain appropriate melting conditions resulting in a narrow weld cannot be achieved.

The overall effect of increasing plate thickness is much like reducing power. To get full penetration of this thicker plate at the 7 KW power level requires, of course, a slower welding speed and this reduces process latitude (Figure 10). Nevertheless, successful narrow welds were produced in 1/2 inch thick plate at this relatively low power level and could be easily produced at slightly higher power levels.

No general conclusions should be drawn from the results suggesting the elimination of porosity as the process transforms from a wide-weld process to a narrow-weld process because the results stemmed from a survey of available specimens rather than an experiment designed to determine the cause of the phenomenon. The effect may have, in fact, been caused by the characteristic cross section of the weld itself. For example, porosity may have had no chance to form because narrow welds are characterized by a very small degree of underbead penetration. Greater penetration results in a gouged weld. The most likely source of porosity - forming hydrogen in these tests is the mill scale on the underside of the plate. Wide welds with their greater penetration interact with this scale to a much greater degree. Wide welds exhibit much more porosity than narrow welds which seems to confirm the hypothesis. Therefore, if there were another source of hydrogen, one that was equally available to both the narrow and wide bead process, it is possible both would exhibit porosity. On the other hand, gouged welds are also greatly over-penetrated. Therefore, they should exhibit the greatest reaction with the scaled underside of the plate. However,

gouged welds did not show porosity (as far as could be determined from radiographs made in the presence of the characteristically deep undercut).

The presence of certain porosity free wide welds (Figure 8) and the absence of porosity in the initial 3 KW and 4 KW procedures appear to relate to a common denominator - welding speed. Apparently these wide weld processes are slow enough to retain metal in the molten condition long enough for gas or vapor voids to float out. Reducing the speed about 15% below that at which penetration is barely sustained (referred to as the bench mark speed earlier) permits this to happen. This particular mechanism for producing radiographically sound welds is not compatible with or related to the freedom from porosity observed in the narrow welds.

As noted in Reference 6, the answer to freedom from porosity in narrow welds may lie in the geometric distribution of the metal during the unique melting process that takes place. In general, the liquid metal is always in the form of a thin layer from which gas or voids could easily escape. This configuration is maintained until freezing by the formation of a long narrow "funnel" behind the weld (Figure 14). This phenomena has also been reported in continuous casting where it has been observed to be a requirement for sound castings.

CONCLUSIONS

Although the work reported in this paper represents the first systematic attempts to characterize the narrow bead phenomenon, it appears that certain broad conclusions can be drawn.

1. There was a distinct increase in the as-welded breaking strength of butt welds made in 1/4" 2219-T87 aluminum when the procedure was varied to cause the weld cross section to become narrow and reduce the heat input per inch of weld.
2. When the high-strength, narrow-bead configuration was achieved, there was a marked reduction in the occurrence of porosity.
3. A controlled flow of helium from the exit gas nozzle of the welder was necessary to produce narrow welds over a variety of power levels and weld speeds. Presumably the controlled helium flow rate replaced the air between the electron gun and workpiece. In addition, the controlled exit gas flow may be displacing metal vapor from the beam path. Both actions would reduce scattering.
4. For the particular beam density that was available, an analysis of the latitude of the narrow-bead, high-strength, procedure with regard to process speed and exit nozzle effluent gas flow indicates that:
 - a. A speed threshold exists for any given procedure (i.e., power-thickness combination) and below this speed, narrow welds cannot be obtained.

- b. The tolerance for variances in speed can be considerable, but as the power is reduced or thickness is increased, this tolerance disappears.
- c. The tolerance for variances in gas flow appears to be critical (about 1% of the total flow into the system) and is reduced as power is reduced or thickness increases.

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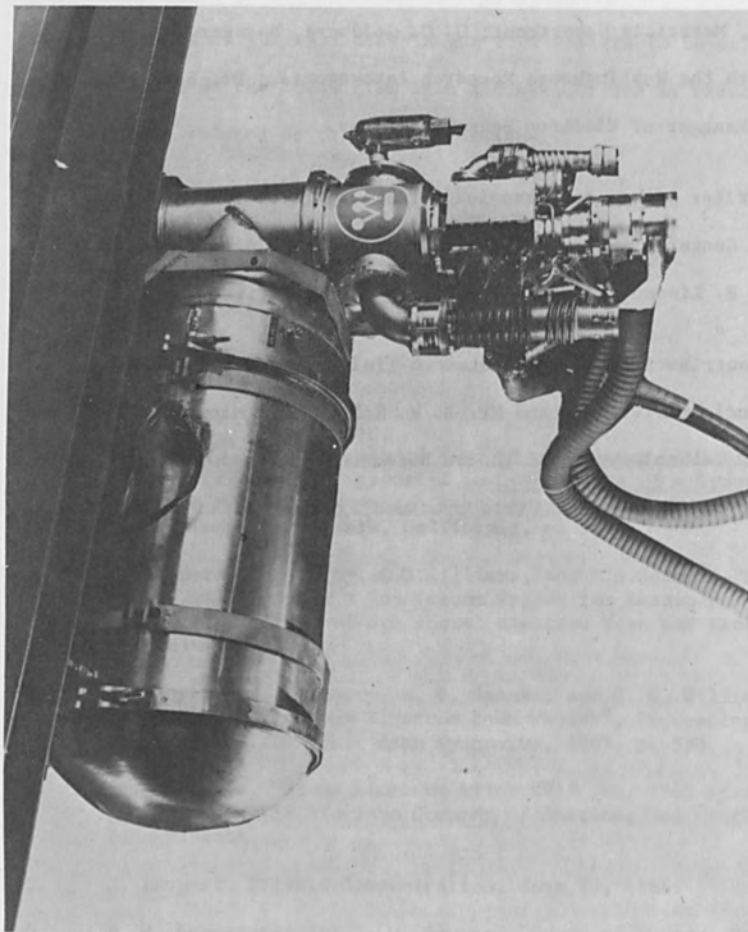


Fig. 1. Portable 12 KW (150 KV) out-of-vacuum welder

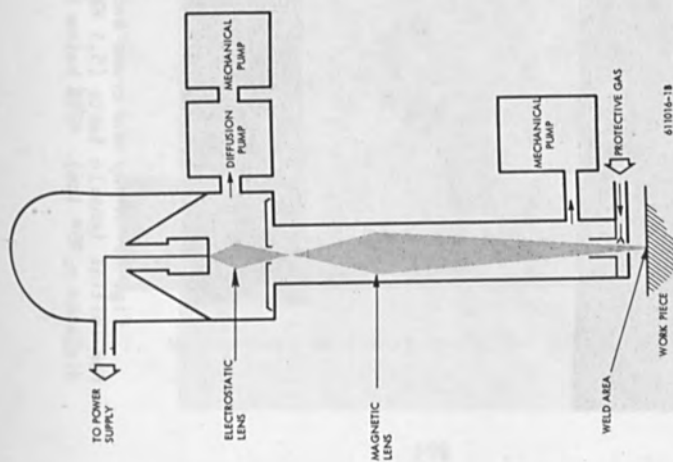


Fig. 2. Principles of operation of an out-of-vacuum electron beam welder.



Fig. 3. Typical diffuser shield for providing inert gas coverage to the area around the point of beam impingement.

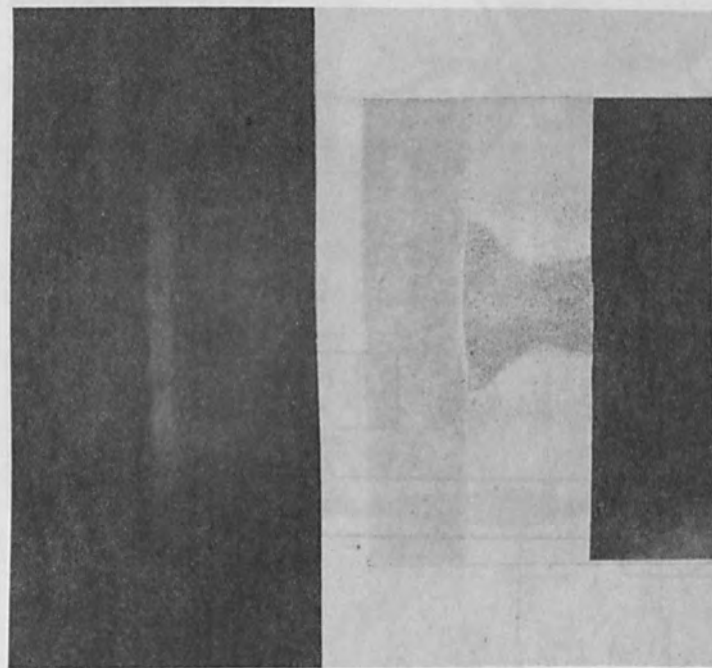


Fig. 4. X-ray and cross section of weld used for initial tensile tests (5.1 KW - 9/16 work distance - 45* ipm) *25% below bench mark speed.

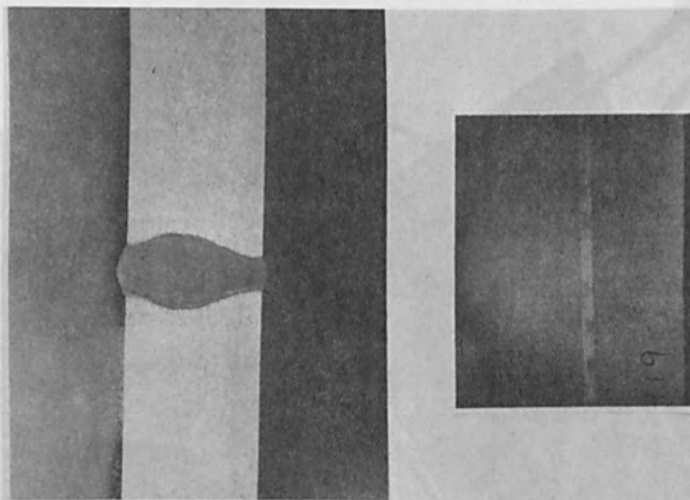


Fig. 5. 8.35 Kj/in narrow-bead out-of-vacuum weld (280 ipm 1/8 gun-work distance).

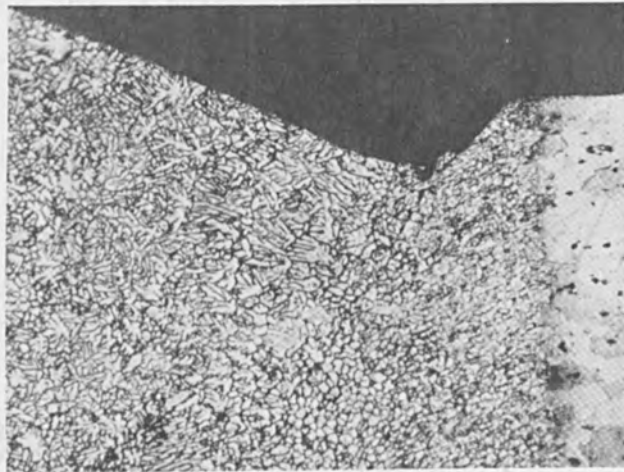


Fig. 6. Narrow-bead weld weakened by undercut



Fig. 7. Narrow-bead weld that exhibited high tensile strength

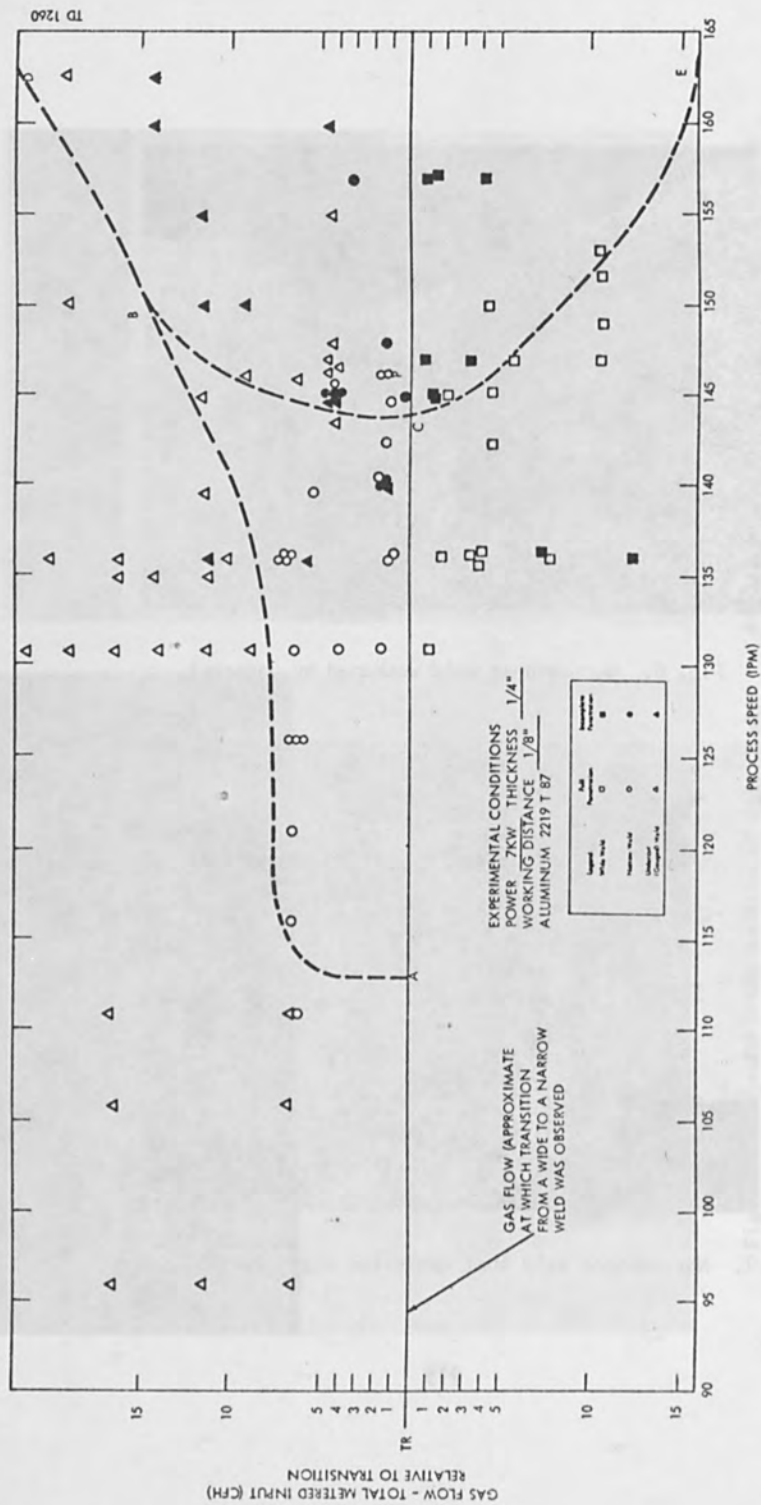


Fig. 8. Process-parameter-plot of narrow weld with welding speed and gas flow as independent variables (7 KW-150 KV, 1/8 work distance, 1/4" thick aluminum).

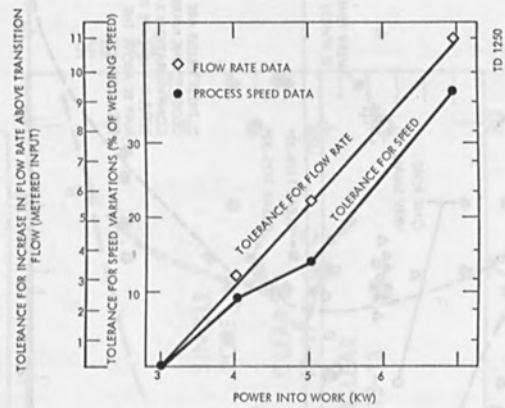


Fig. 9. Effect of weld power level on process latitude for a given material (thickness 1/4").

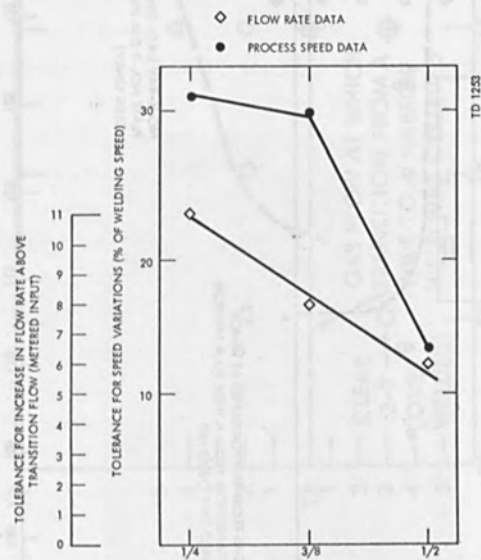


Fig. 10. Effect of material thickness on process latitude for a given power level (7 KW).

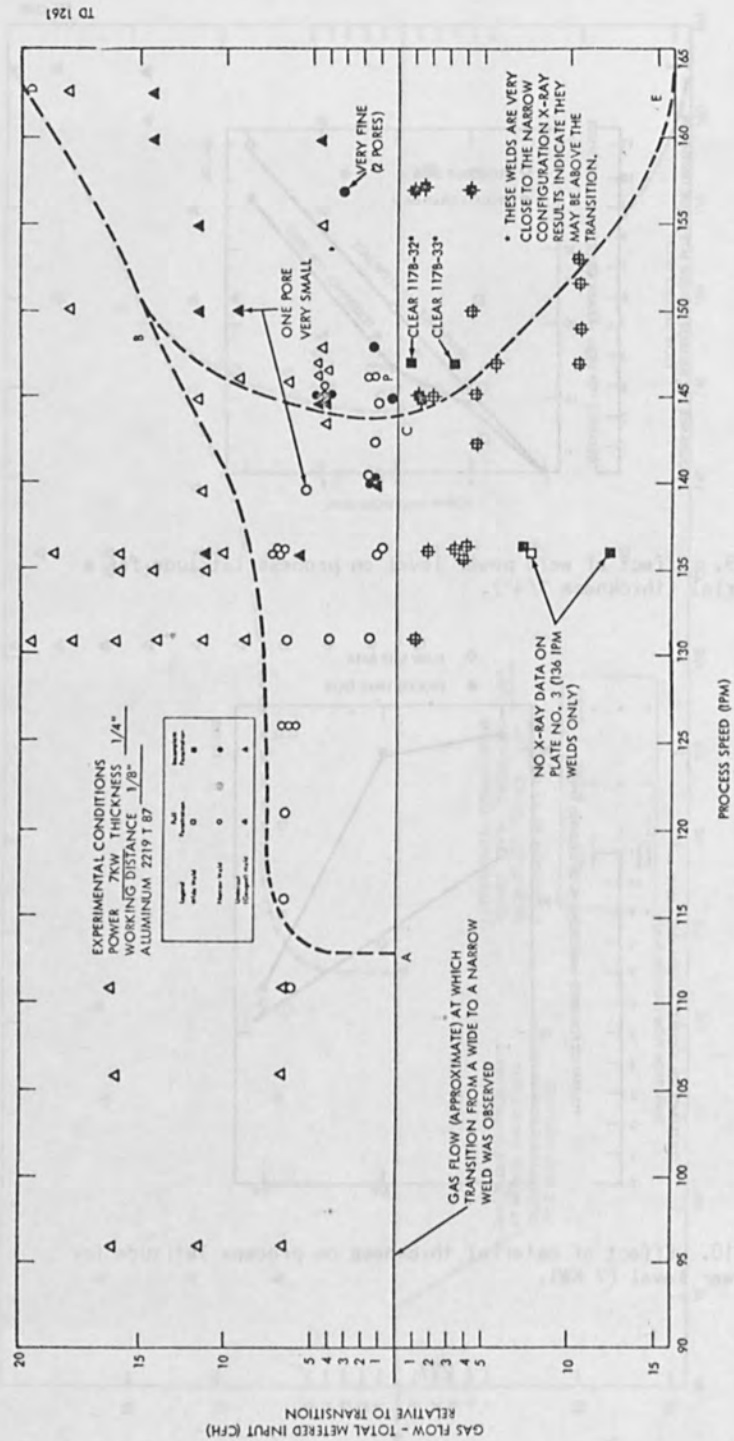


Fig. 11. Occurrence of porosity in welds on 1/4" plate.

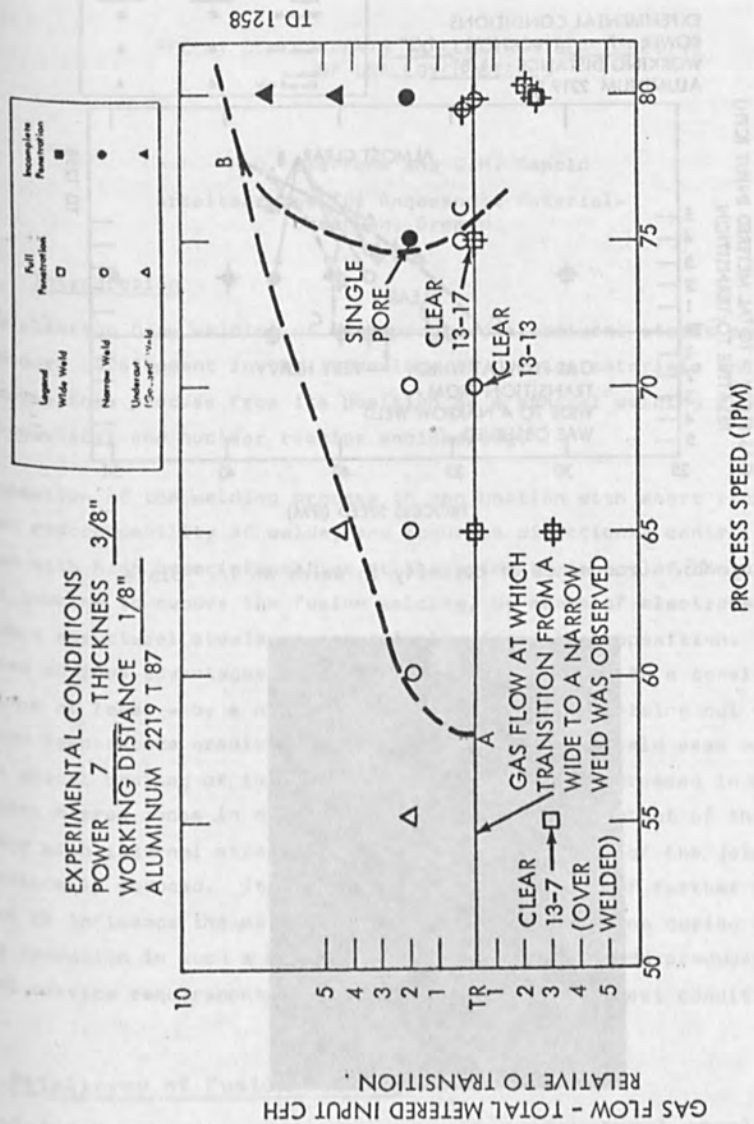


Fig. 12. Occurrence of porosity in welds on $\frac{3}{8}$ " plate.

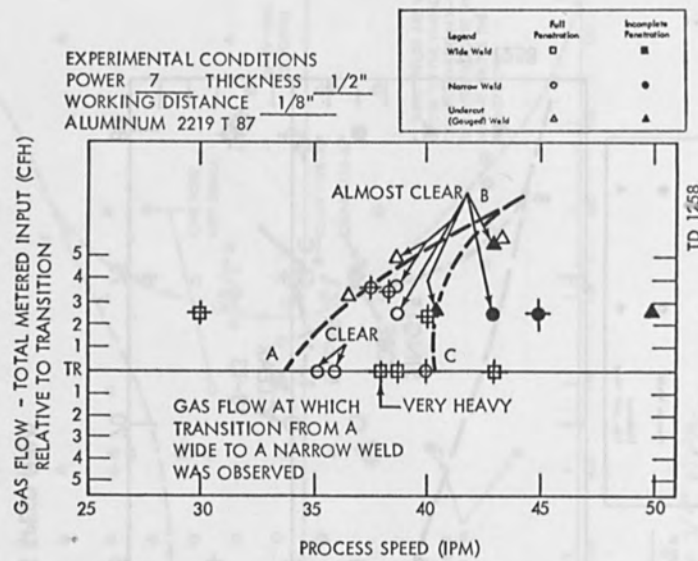


Fig. 13. Occurrence of porosity in welds on $\frac{1}{2}$ " plate.

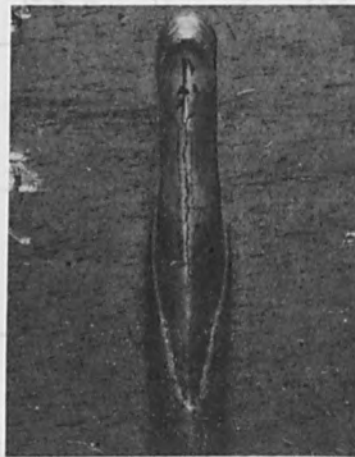


Fig. 14. Metal flow pattern in narrow welds.

RECENT DEVELOPMENTS IN ELECTRON BEAM WELDING OF UNALLOYED STEELS

H.D. Steffens and G.H. Sepold

Arbeitsgruppe für Angewandte Material-
forschung, Bremen

1. Introduction

The electron beam welding of mass-produced structural steels represents a modern development in the technology of joining materials and thus removes this process from its position as a special welding process for aeronautical and nuclear reactor engineering.

Automation of the welding process in conjunction with short cycle times, good reproducibility of welds, and accurate directional control of the beam with high beam intensities at the point of impact of the electrons all combine to render the fusion welding, by means of electrons, of low-carbon structural steels an economically feasible proposition. However, these obvious advantages have hitherto been offset - to a considerable extent at least - by a number of unsolved problems arising out of the steep temperature gradient across the region of the weld seam owing to the abrupt cooling of the weld pool. This causes increases in hardness across narrow zones in dependence upon the carbon content of the materials along with internal stresses so that the loadability of the joint is drastically reduced. It is, therefore, the purpose of further research work to influence the metallurgical processes occurring during the welding operation in such a way as to ensure that the welds produced will meet service requirements as well as more stringent test conditions.

2. Metallurgy of Fusion Welding

Included in the investigations were two structural steel grades whose composition is given in Table 1.

Steel grade	C	Si	Mn	P	S	Cr
CK 45 corres. to AISI 1050	0,44	0,22	0,66	0,022	0,03	0,15
C 22 corres. to AISI C1020	0,19 to 0,24	0,22	0,42	0,014	0,022	-

Table I: Chemical Composition of welded steels

These two steel grades differ mainly in respect of their carbon content. While C 22 steel, welded by conventional techniques, will require no post-heat treatment, such will definitely have to be carried out on grade CK 45 immediately after the welding operation. Problems arising in conjunction with the EB-welding of unalloyed structural steels were already discussed in two earlier papers [1,2], particular attention being drawn therein to the inadmissibly high hardness values and internal stresses associated therewith, and to the susceptibility of certain regions to cracking. In the EB-welding of CK 45, these disadvantages are even more pronounced so that the application of EB-welding to that material is rendered questionable unless satisfactory steps are taken to overcome them. Thus it may be assumed that suitable adjustment of welding conditions, particularly in conjunction with circular beam deflection, will produce a larger and hence more slowly cooling weld pool. The recording of temperature gradients, in relation to both location and time, on the top side of the weld seam by means of thermocouples - assuming uniform heat dissipation in all directions of the metal from a cylindrical heat source - did not, however, provide confirmation of the above assumption as will be noted from Fig. 1 in which temperature gradients Nos. I to III are plotted; these were obtained at various distances from the weld pool. The slope of curve No. I which resulted when applying a deflected beam at a circular frequency of 50 Hz and at an amplitude of 1 mm, corresponds with that of curves No. II which were obtained when employing an undeflected beam. Within the 5 to > 10 seconds range, curves No. II exhibit a considerably steeper gradient than do, for example, temperature curves No. III which were obtained after pre-heating the material to 200°C. Since the temperature functions of a deflected beam and those of a "static" beam tally

with one another, it is reasonable to assume that the types of structure occurring in the region of the weld seam will also be similar. This was substantiated by micrographs and hardness measurements across the weld zone.

The influence on structural transformation of temperature changes occurring in time can be noted, in principle, from TTT-diagrams. After a brief holding period at the austenitic stage between 800° and 900°C, the material cools at various cooling rates - curve No. 1 in Fig. 2. The encircled figures indicate Vickers hardness values at room temperature. Due to the very fast heating and cooling rates inherent in EB-welding, the validity of the TTT-diagrams is rendered dubious since at a higher austenitizing temperature there is a shift of the ranges within which ferrite and bainite form as indicated by broken lines in Fig. 3. Such shifts are taken into account by maximum temperature-cooling time diagrams which have not yet been established for very high and steep temperature gradients. A comparison of Figs. 2 and 3 on the basis of approximately identical temperature gradients for C 22 and CK 45 steel grades suggests that larger proportions of martensite and bainite may be expected in the case of the higher-carbon content steel grade, i.e. CK 45, whereas the structure of the weld seam in C 22 steel is composed of ferrite with a lattice-type distribution, of densely packed bands of perlite, and of bainite and martensite - Fig. 4. This joint distinguished itself by increased ductility even though hardness and V-notch impact strength levels were not entirely satisfactory [1].

3. Heat-Treatment of Welded Structural Steel

Greater toughness of the welds can, for example, be achieved by normalizing the workpieces so that a re-formation of grains within the region of the weld takes place. In many cases, however, stress-free annealing of the components suffices to relieve welding stresses. Extended tempering at still lower temperatures brings about a partial relieving of internal stresses and greater resilience of the joint. This was established on a heat-treated steel component with a carbon content of 0.24%.

This type of steel is ordinarily used without post-heat treatment in the construction of boilers and similar apparatus. When welding it by means of the electron beam, the required minimum strength was not obtained in the joint so that a post-heat treatment involving annealing for a period of one hour at temperatures between 200°C and 600°C became necessary. Subsequent tempering at 400°C produced the desired strength values with Charpy-V-notch impact strength rising to 3.5 kpm/cm², for example, and even exceeding 20 kpm/cm² at a temperature of 550°C, Fig. 5. By contrast, the Charpy-V-notch impact strength values obtained at room temperature dropped markedly at a test temperature of 0°C.

Heating the components immediately prior to the welding operation leads to less abrupt cooling of the weld seam so that quenching proceeds considerably less rapidly, which was demonstrated by Fig. 1. Macroscopically, these weld seams are distinguished by a raised top bead and root as well as by a greatly reduced number of small pores and cracks which are considered unavoidable on structural steels with carbon contents in excess of 0.2%. Microscopically, a less steep temperature gradient affects the structural composition in such a way as to reduce the proportion of martensite and to increase that of bainite as evidenced by a micro-section. In terms of strength this means improved resilience of the joint when subjected to tensile and bending stresses as well as increased V-notch impact strength. The latter increases from 1.5 kpm/cm² to 8 kpm/cm² when pre-heating the material from 20°C to 300°C and ties in with a corresponding decrease in hardness in the region of the weld seam from HV 0.2 = 420 kp/mm² to HV 0.2 = 250 kp/mm² on steel grade No. 1020, Fig. 6. The considerably greater decline in hardness in the case of slightly pre-heated specimens as against specimens subjected to post-heat treatment is evident. However, one drawback to the pre-heating of components is the need for rather elaborate fixtures inside the chamber which must be equipped with non-heat-conducting jaws. Over and above that, exact positioning of the weld seam in relation to the beam during evacuation of the chamber is not always assured due to heat distortion.

A further possibility of reducing hardness within the region of the weld seam consists in carrying out the post-heat treatment with a de-focussed electron beam. On unalloyed steel sections containing 0.18% of carbon it was possible by this procedure to reduce hardness peaks of HV 0.2 = 420 kp/mm² in dependence upon the thickness of the section to HV 0.2 = 200 to 250 kp/mm². This method is, however, uneconomic on larger lot sizes. On the other hand, consideration might be given to installing a second electron beam generator within the chamber for the purpose of effecting such heat treatment. Over and above that this technique is only feasible on sections up to a maximum thickness of 5 mm as otherwise the lower regions of the weld seam will not be reached by heat conduction. In any event, this kind of post-treatment of the seam should endeavour to avoid re-melting of the weld surface as this will increase internal welding stresses and will lead to cracks within the re-melted zone. This was particularly observed on unalloyed steels with a carbon content in excess of 0.2%.

In general, EB welded joints with carbon contents in excess of 0.2% are rather susceptible to cracking, the shape of the component also exercising an appreciable influence on the formation of cracks so that post-heat treatment of the type described can hardly be recommended.

4. Welding with Interlayers

The joining of unalloyed structural steels containing more than 0.2% of carbon requires a post-heat treatment. Thus, the heat-treated steel grade No. 1050 exhibits hardness values in the weld seam of more than 600 kp/mm² from which excessive brittleness within the region of the weld must be inferred. In these circumstances, a post-heat treatment which will not, however, remove pores and cracks, is indispensable. By melting the high-carbon workpiece flanks and simultaneously fusing the interlayer, intermingling of the materials takes place, Fig. 7. The illustrations designated 'a' are micro-sections through welds made in CK 45 without insertion of interlayers whilst 'b' and 'c' are micro-sections through welds made in CK 45 and C 22, respectively, with a 250 μ m

thick pure iron foil inserted in the joint in each instance. Decreasing carbon contents within the region of the weld may be inferred from the structure of the weld seams. Verification of this is provided by hardness curves across the weld zone and by micro-analyses, Fig. 3. Decline and uniform distribution of the carbon content are directly apparent from these. A low carbon content in the weld seam led to a considerable fall-off in hardness, with hardness in the heat-affected zone being slightly higher. The insertion of a 100 μ m thick foil did not yield any improvement in hardness levels so that an interlayer of minimum thickness must be postulated. Identical results were obtained from experiments on C 22 steel, Fig. 9. Here, the carbon concentration curve is merely indicative of a more uniform distribution of carbon within the seam than in the base metal whilst hardness curves across the weld zone provide a more accurate picture of ductility within the region of the weld seam. The result of this technique is a relatively soft joint which can be improved by furnace-annealing. Furthermore, carbon diffusing from the base material into the weld seam produces regions of greater strength which serve to raise the overall strength of the joint if necessary. Further experiments are designed to vary the carbon contents of the interlayers by inserting metal sections with a higher carbon content so as to obtain welds of optimum strength and toughness.

Latest investigations are concerned with applying the iron interlayer to the workpiece by metal spraying. While this technique requires greater initial expenditure in respect of preparation of the components, it is nonetheless more economical if one considers, for example, that the amount of scrap resulting from blanking out an annular sheet metal insert for a pipe joint is not inconsiderable.

Initial experiments carried out in conjunction with EB welding and involving the application of pure iron interlayers by plasma-spraying and in the presence of argon as shielding gas, can be judged positively even though a certain degree of porosity does not appear to be altogether unavoidable to begin with. It is assumed, however, that results can be improved using an electron beam deflected at a high frequency thereby

enabling this combined plasma-application and electron-beam technique to become technologically useful. It will be noted from Fig. 10 and 11 that the formation of the weld and the hardness curve across such a joint indicate a relatively soft structure in combination with an almost flawless seam. Hardness peaks of up to 450 kp/mm^2 occur within a very narrow zone and, due to the supporting effect of the adjacent regions, are not expected adversely to affect the overall strength of the joint.

5. Summary

The electron beam welding of unalloyed structural steel is attended by a multiplicity of metallurgical problems which are due to the unusually high intensity involved in this process. This intensity produces steep temperature gradients across the weld seam, high internal welding stresses and, in dependence upon the carbon content, embrittlement resulting in a joint with inadequate mechanical-technological properties.

The procedures proposed to solve these problems involve pre-heating and post-heat treatment of the workpieces. Welding with thin interlayers of pure iron is discussed and, finally, a new technique of applying interlayers by metal spraying is proposed with a view to improving the economy of the electron beam welding process as applied to unalloyed structural steels.

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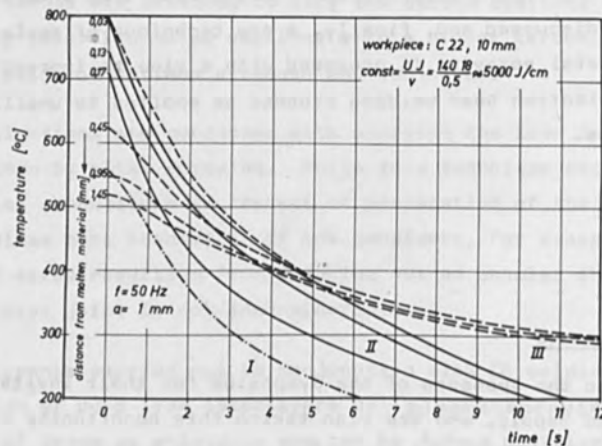


Fig. 1. Local time-temperature curves

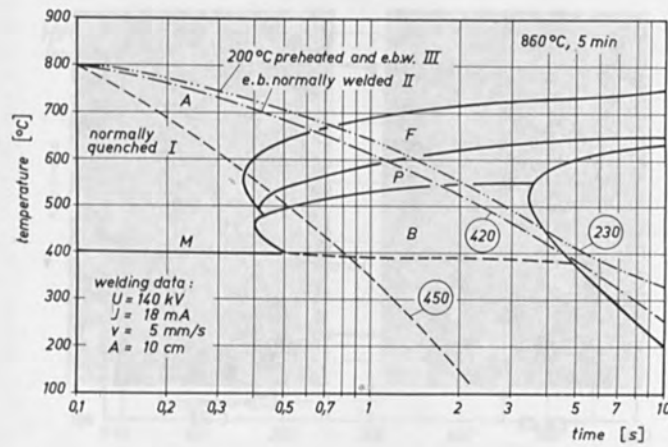


Fig. 2. Continuous cooling TTT-diagram for Ck 45, 10 mm thick

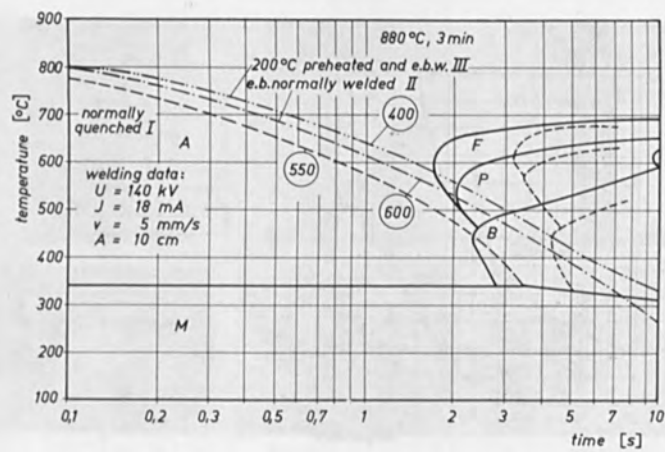


Fig. 3. Continuous cooling TTT-diagram for Ck 45, 10 mm thick

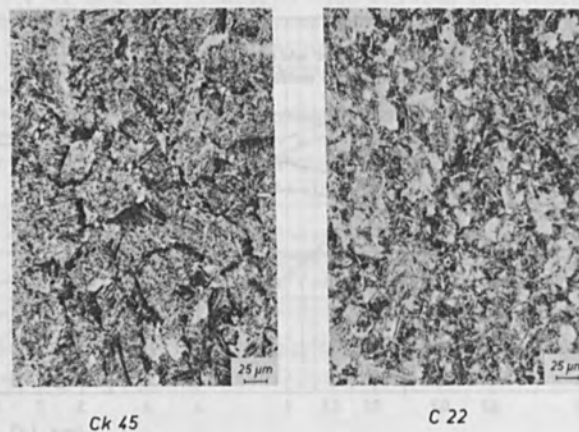


Fig. 4. Microstructure of e.b. welded steels with different C-content.

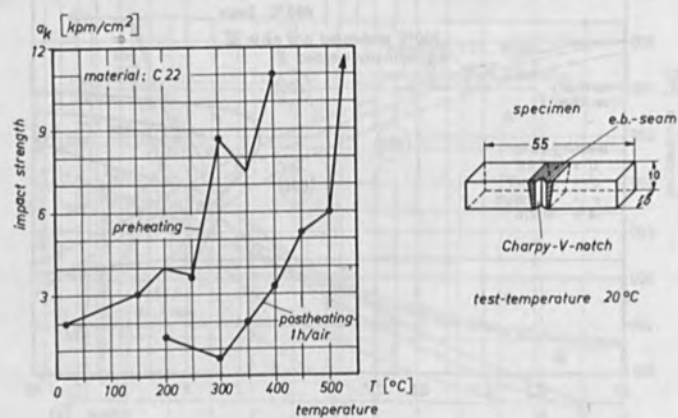


Fig. 5. a_K -values of preheated and postheated e.b. welded C 22.

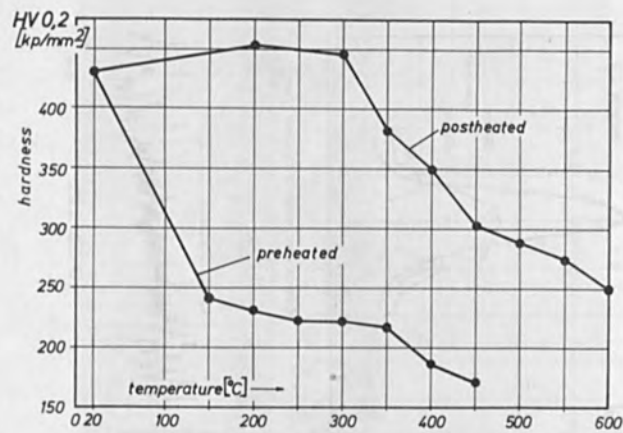


Fig. 6. Preheating and postheating effects on Vickers hardness of e.b. welded C 22.

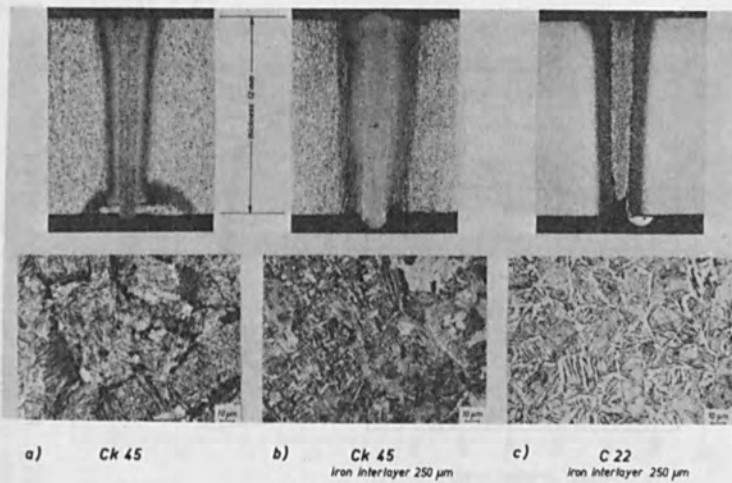


Fig. 7. e.b. welded structural steel with foil interlayers, material Ck 45.

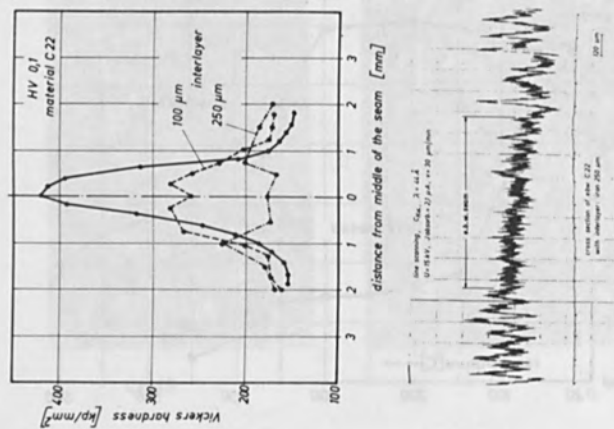


Fig. 9. Carbon distribution and microhardness of e.b. welded Ck 45.

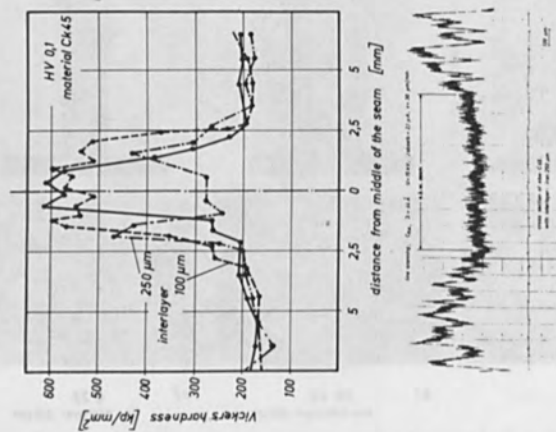
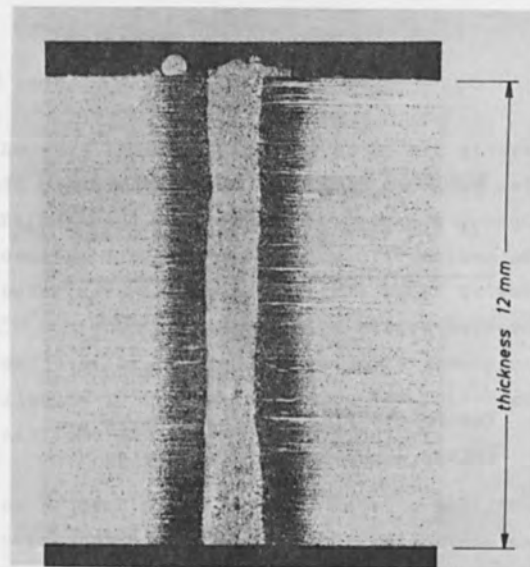


Fig. 8. Carbon distribution and microhardness of e.b. welded C 22.



C 22

Fig. 10. Cross section of a plasma-sprayed and e.b. welded joint, material C 22.

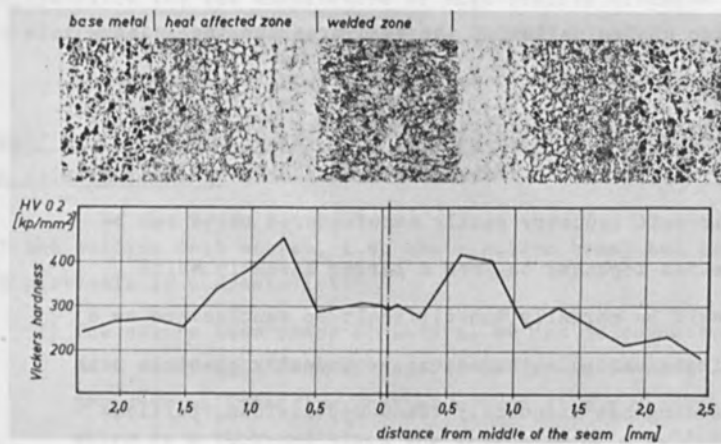


Fig. 11. Micro-hardness of a plasma-sprayed interlayer, fusion welded by e.b.

ELECTRON BEAM WELDING OF TITANIUM
IN THE GERMAN AEROSPACE INDUSTRY

Lutz DORN,
Vereinigte Flugtechnische Werke -
Fokker GmbH, Bremen / W.GERMANY

SUMMARY

Electron beam welding guarantees the best protection from contamination of the weld area and the least amount of welding damage. By welding in vacuum embrittlement of reactive metals like Titanium caused by reaction with absorbed gases is avoided. In the aircraft industry easily manufactured parts can be welded together to form a larger assembly which would be normally too difficult to manufacture as a single unit. In the aerospace industry electron beam welding has frequently found application, particularly for the construction of compressed gas or fuel containers.

1. Introduction

The steady increase in the efficiency of modern aircraft, missiles and spacecraft requires the use of highly advanced materials, designs and manufacturing processes. The designer's requirements for lightweight construction are met by the utilization of high-strength lightweight materials. In the temperature range between room temperature and 450°C high-strength titanium alloys have the highest specific strength of all currently available aerospace material. Therefore, titanium is increasingly used for load-bearing structural elements in airframe and launcher construction.

The processing properties of titanium require modification of conventional working procedures or introduction of new manufacturing techniques. In the field of joining methods particular emphasis is to be placed on electron beam welding. The difficulties encountered during the initiation of this method have been overcome as experience in handling the process grows. Today this method has become indispensable for the manufacture of high-quality titanium components for airframes, aero-engines, missiles, and satellites.

2. Specific Characteristics of the Electron-Beam Welding Method for Joining Titanium

For the welding heat source, i.e. the electron beam, two properties are particularly characteristic:

- 1) The entire beam power of several kW can be concentrated on a very small area of approx. 0.1 mm in diameter. This produces an intensive thermal effect at the welding joint which is a thousand times the intensity of a welding arc and even ten thousand times that of an oxy-acetylene flame, Fig. 1.

- 2) The electron beam welding process is generally performed under a vacuum pressure of $<5 \cdot 10^{-4}$ mm Hg. to minimize dispersion of the electron beam by air particles. At this pressure less oxygen and nitrogen is present than in inert gases of 99.99% purity, Fig. 2.

Because of the low heat input into the workpiece, shrinkages are reduced about ten times as compared with arc welding, Fig. 3. Improved accuracy in shape and size of the components will provide new possibilities for the use of welding techniques on high-precision parts. In production the process is used to avoid difficult machining or forming operations by splitting up complex workpieces into simple elements which are joined by electron beam welding. The possibility of repairing components showing cracks, pores or damage due to wear, without the danger of distortion, allows a reduction in the 'percentage defective' during production and a decrease in downtimes caused by spare parts procurement.

The steep temperature drop on both sides of the beam restricts changes in the heat treatment condition to narrow zones, Fig. 4. Rapid cooling of the welding joint, coupled with the possibility of single-run welding, considerably reduce adverse metallurgical effects frequently occurring in conventional welding processes such as coarse-grain formation and segregation phenomena. Mechanical property tests performed on several alloy types have demonstrated a tensile strength for electron-beam welds which usually is comparable to that of the base material, Fig. 5. The ductility in the seam area has been found to be somewhat reduced, although the decrease is much less compared with conventional methods. The high fatigue strength of the electron-beam weld structure is substantially reduced in the presence of pores and surface notches in seam surfaces not properly finish machined, Fig. 6.

The high purity of high-vacuum electron beam welding is of considerable benefit in titanium processing because this metal has a high affinity to atmospheric gases at increased temperatures. Hydrogen, oxygen and nitrogen will immediately be absorbed by molten titanium. Hydrogen embrittles the material at contents above 250 ppm. Oxygen and nitrogen also cause embrittlement at contents above 0.25% [8] .

3. Investigation of the fatigue strength of electron beam welded thin sheet titanium

Machining of electron beam welds is difficult in the case of thin sheets as used in aerospace. The influence of the seam structure and of post welding treatment of the weld seam on the ^{fatigue} behaviour of 2 mm thick sheet specimens of the titanium alloys Ti-5Al-2,5Sn, Ti-8Al-1Mo-1V and Ti-13V-11Cr-3Al was investigated. Figure 7 shows the σ/N -curves for 50% survival probability.

Unwelded Ti-5Al-2,5Sn and Ti-8Al-1Mo-1V have a greater fatigue strength than Ti-13V-11Cr-3Al. This reflects the different crystalline structure of the α - and β -alloys. As a rule, the fatigue strength/tensile strength ratio is higher for hexagonal than for body centred cubic metals. The fatigue values for TIG butt-welds, however, do not show any influence of the alloy composition. Shot-peening of the weld surface does not cause any significant change in the fatigue strength, since fracture originates from internal defects, predominantly from individual pores.

Electron-beam joined butt-welds show disappointing behaviour as compared with that of TIG-welded seams. Fracture originates from notches or from weld spatter defects in the weld root. Shot-peening of the weld surface on both sides rounds off edge notches and improves the fatigue strength to the TIG-weld level. At the same time the built-in tensile stress in the weld seam is replaced by

a compressive stress at the surface. In this case, fracture originates from edge notches on the weld upper surface.

The fatigue strength behaviour of electron beam welded fillets was studied on a 3 mm thick flat test specimen with transverse stiffeners of the same thickness welded onto both sides and was compared with a correspondingly shaped milled specimen, Fig. 8. The electron beam welded fillets were manufactured with a filler-metal insert of the same material in order to obtain the largest possible concave fillet radius. Although the electron beam welds were subsequently shot-peened, the fatigue strength of the welded test piece is considerably less than that of the milled specimen.

The purpose of the tests was to determine the weld quality achieved under present day operating conditions. While for static loading the load-carrying capacity of electron beam welds is comparable to that of the base material, the fatigue strength is unsatisfactory. Therefore, further development of methods and equipment seem to be necessary to improve the surface structure of electron beam welded joints to that which can be achieved under laboratory conditions.

4. Examples for Electron Beam Welding in the Aircraft Industry

Electron beam welding provides for aircraft manufacturers the possibility of designing simple elements which can be joined by electron beam welding to form a costly, complex integral assembly.

The frame, shown in Figure 9, of the VAK 191 B fighter is a milled forging of Ti-7Al-4Mo. The web protruding from the frame is attached by electron beam welding. Manufacturing the entire part from solid material would have involved considerable extra costs because of the larger quantity of material required and additional machining work.

For reasons of weight-saving a study was made into the manufacture of a tie-down fitting out of Ti-5Al-2,5Sn instead of high-strength steel, Fig. 10. Whilst TIG-welding produced an inadmissible distortion, electron beam welding resulted in a usable tight-tolerance part. However, this version was not adopted for the production aircraft because of the large manufacturing effort compared with the weight saving actually achieved.

Because of the reduced possibility of gas embrittlement, electron beam welding was considered for the production of titanium alloy bleed air piping, which is subject to high mechanical and thermal stresses, Fig. 11. The longitudinal pipe seam was electron beam welded, whereas the circular seams, which are subject to only half the load caused by internal pressure, were made by shielded arc welding. Dynamic internal pressure tests confirm the strong correlation between load capacity and weld surface structure. After optimizing the welding conditions, the test pipes supported more than 200,000 load cycles with a reciprocating stress amplitude of 24 kgf/mm².

The problems in titanium processing naturally result in an increased 'percentage defective'. Considering the high cost of these titanium parts, the possibility of repair using electron beam welding is of particular importance. For example, the wall thickness of a titanium frame did ^{not} attain the required minimum dimension. After this area had been cut away, a correspondingly shaped plate was welded in and then finished, Fig. 12.

5. Examples for Electron Beam Welding in Space Technology

In the space industry the advantages of electron beam welding are used to obtain extremely light-weight structures. The weight of the propellant tank shown in Fig. 13 is reduced by half upon replacing

the TIG welded aluminum structure by an electron beam welded titanium construction. (Initial TIG welding of the titanium alloy resulted in embrittled, coarse-grained welds.) Electron beam welding of such large-sized components involves some difficulties which hardly affect smaller parts. For instance, unstable component parts can scarcely be clamped or set up with the same precision as small parts, Fig. 14. Special beam controls are also necessary for welding the diaphragm segments since the contour of the diaphragm is not exactly circular, Fig. 15. The constant rotary movement of the diaphragm during welding results in a variable welding speed, in a change in the working distance from the gun and in a change of the depth of metal to be welded. The necessary changes in lens and beam current of the electron gun are automatically corrected by means of a servo-mechanism. The tank must withstand 24.8 atm gauge pressure, which corresponds to a diaphragm load of about 70 kgf/mm².

The propellant tank shown in Fig. 16 consists of two similar hemispherical shells out of Ti-6Al-4V, which are welded together by electron beam welding. The tank wall thickness of 0.5 mm is reinforced at the joint area by a collar which is turned down to 0.8 mm wall thickness after electron beam welding. The collar facilitates clamping together of the two shells in a work support. In addition, after machining the seam a completely notch-free surface is obtained. Pressures applied in the bursting test exceeded 75 kgf/cm². Under repeated loading the weld supported the required 100 cycles between 0 and 43 kgf/cm². The leakage test with helium and the functional test under space environmental conditions also proved the functional qualification of the component.

The satellite pressurized gas tank shown in Fig. 17 is made by electron beam welding of two copy-turned hemispherical shells in Ti-6Al-4V. The required absence of splashes in the interior necessitates a special technique: first, protruding recesses at

the inner wall are welded together, then the outer wall is welded after the gap to the outside has been filled with filler material. This first test tank withstood the calculated burst pressure of 590 atm gauge pressure.

The manufacturing technique for light-weight fuel tanks, which was developed for space projects, has already been applied to 'terrestrial' constructions: spherical fuel tanks for PORSCHE racing cars, made from Ti-6Al-4V, with 300 mm diameter, 0.3 mm wall thickness and 400 g weight we have also manufactured by use of electron beam welding.

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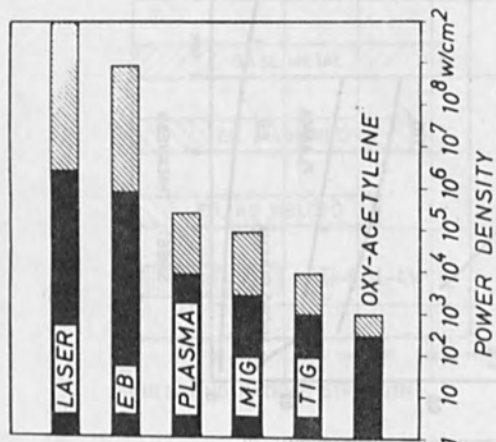


Fig. 1. Comparison of the power densities of various heat sources for welding [1].

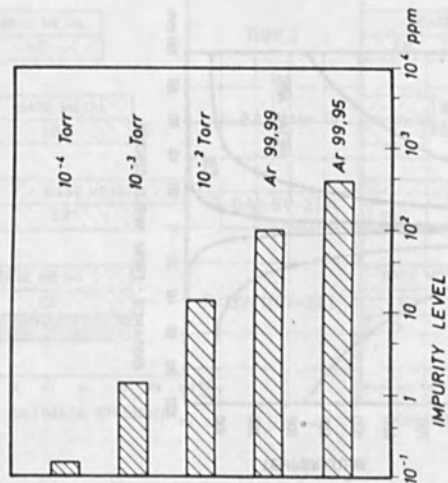


Fig. 2. Comparison of the impurity level of argon shielding gas and vacuum at different pressures.

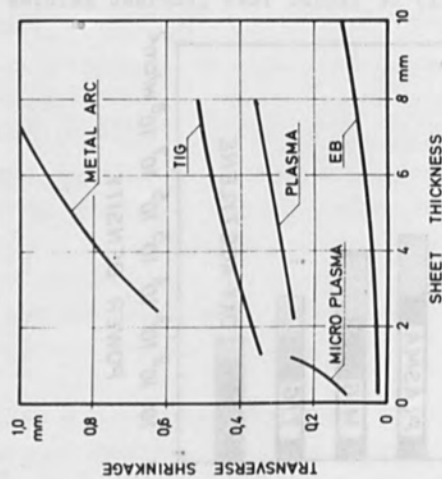


Fig. 3. Transverse shrinkage as a function of the sheet thickness for different welding methods [2].

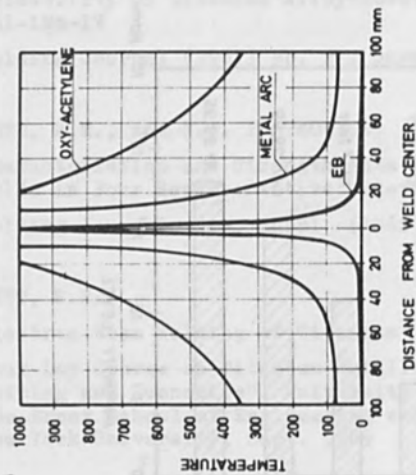


Fig. 4. Maximum temperatures transverse the seam for various welding methods [3].

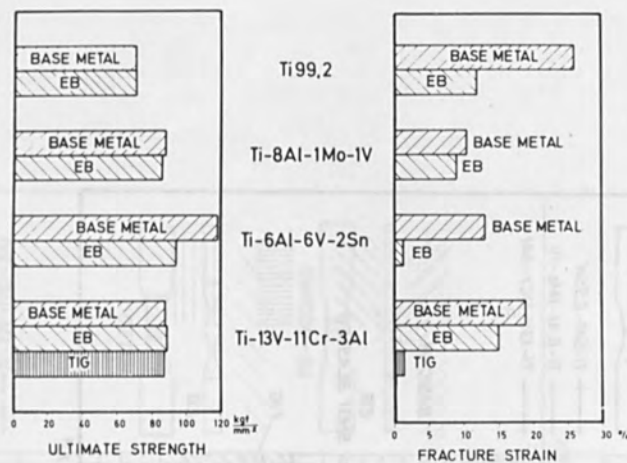


Fig. 5. Strength and strain data of welded and unwelded titanium material obtained in tensile tests [4, 5, 6].

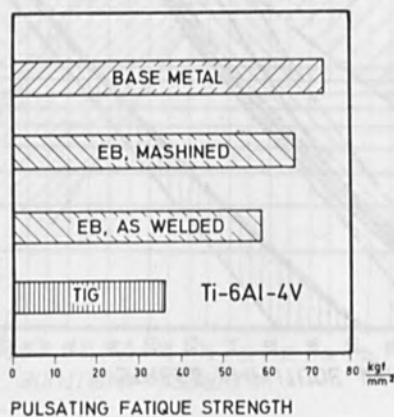


Fig. 6. Fatigue strength of 12,5 mm thick Ti-6Al-4V in welded and unwelded condition (tension-tension-test) [7].

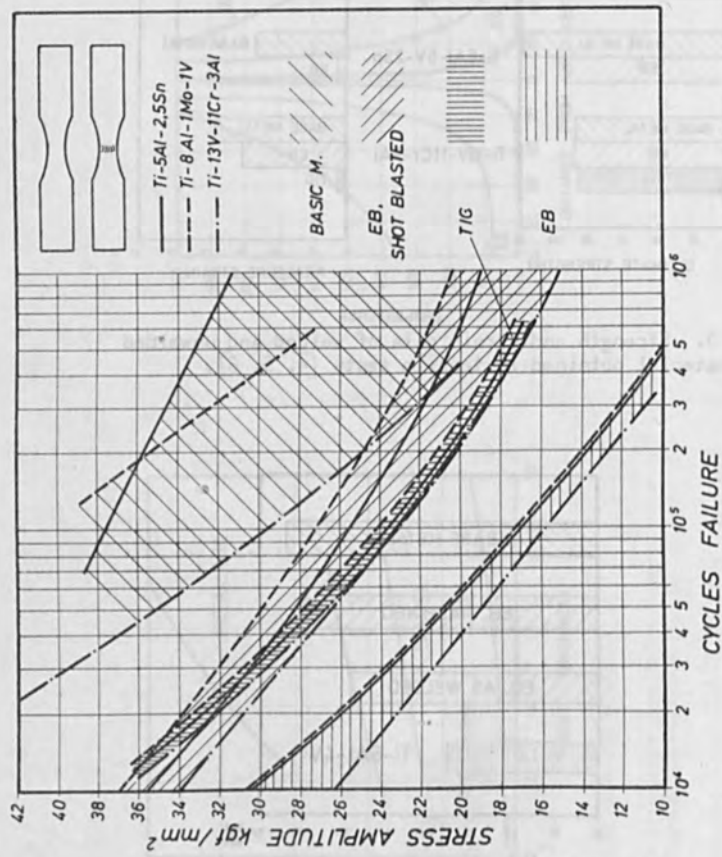


Fig. 7. Comparison of fatigue strength of unwelded as well as electron-beam welded TIG welded specimens (tension-tension-test).

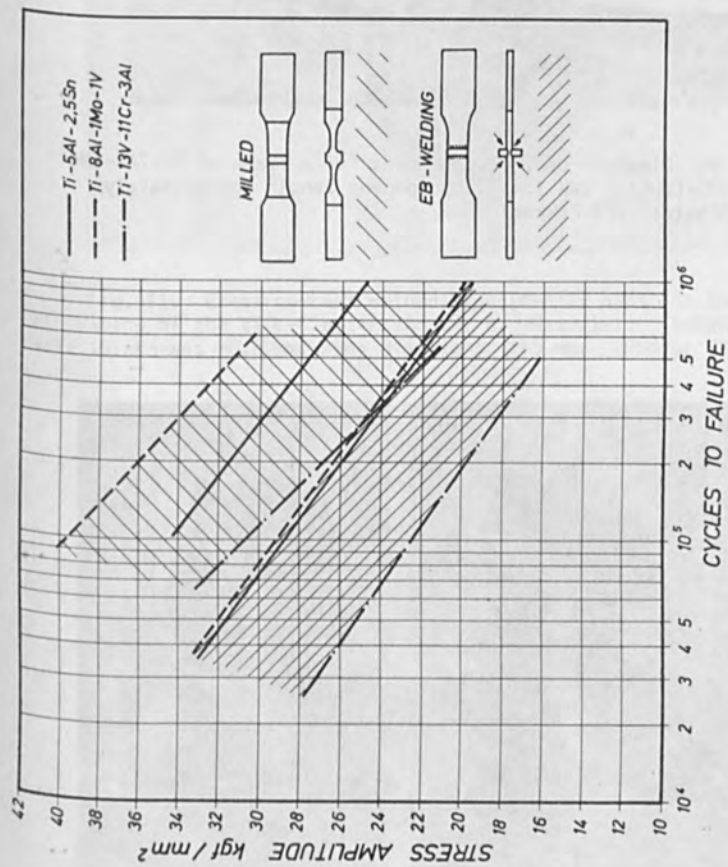


Fig. 8. Comparison of fatigue strength of flat specimens with milled and electron-beam welded transverse stiffeners (tension-tension-test).

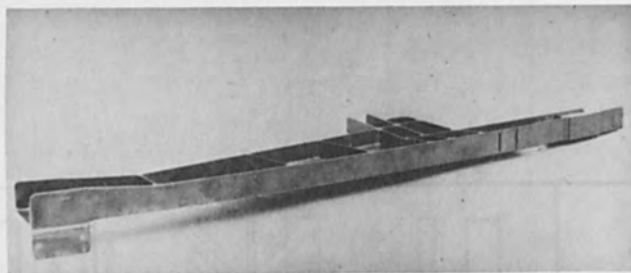


Fig. 9. Electron-beam welded joint for a frame of Ti-7Al-4Mo for the VTOL-fighter VAK 191 B. Component length approximately 750 mm. (Photo: VFW-Fokker)

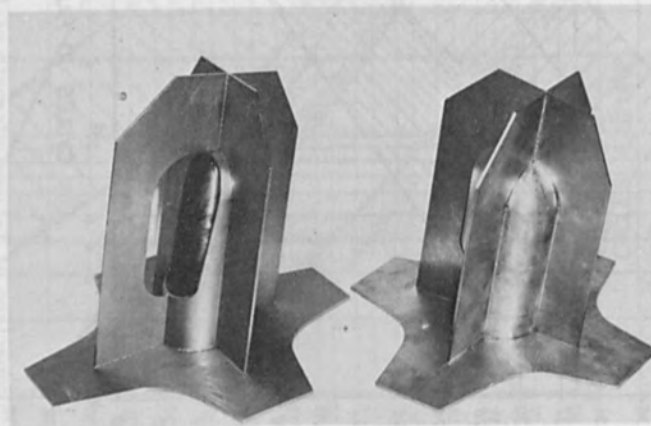


Fig. 10. Electron-beam welded tie-down fitting for TRANSALL C-160 transport aircraft. Material Ti-5Al-2,5Sn, wall thickness: 2 mm, Height: 300 mm. (Photo: VFW-Fokker)



Fig. 11. Electron-beam welded experimental pipe for the bleed air piping of the VTOL-fighter VAK 191 B. Material: Ti-5Al-2,5Sn, wall thickness: 0.8 mm, pipe diameter: 115 mm. (Photo: VFW-Fokker)

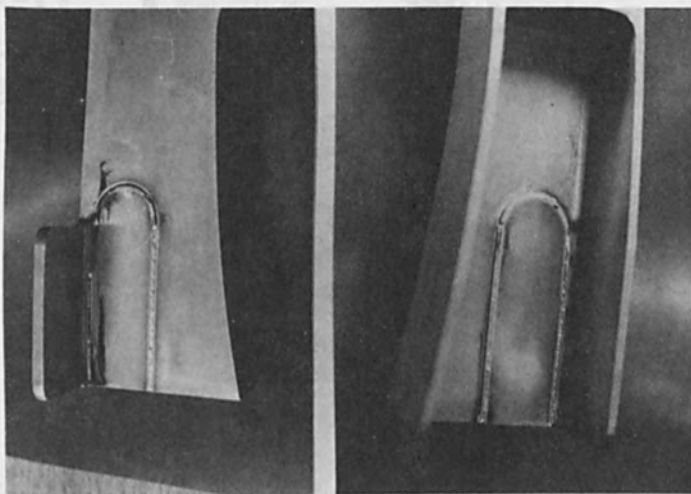


Fig. 12. Electron-beam repair welding on a frame of Ti-7Al-4Mo before finishing. (Photo: VFW-Fokker)

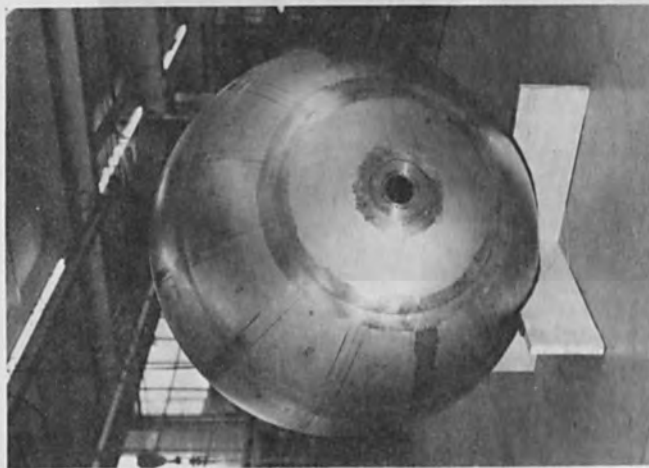


Fig. 13. Electron-beam welded propellant tank design for the third stage of the ELDO-A launcher after machining the welds. Material: Ti-13V-11Cr-3Al, wall thickness: 1.5 mm. (Photo: VFW-Fokker)

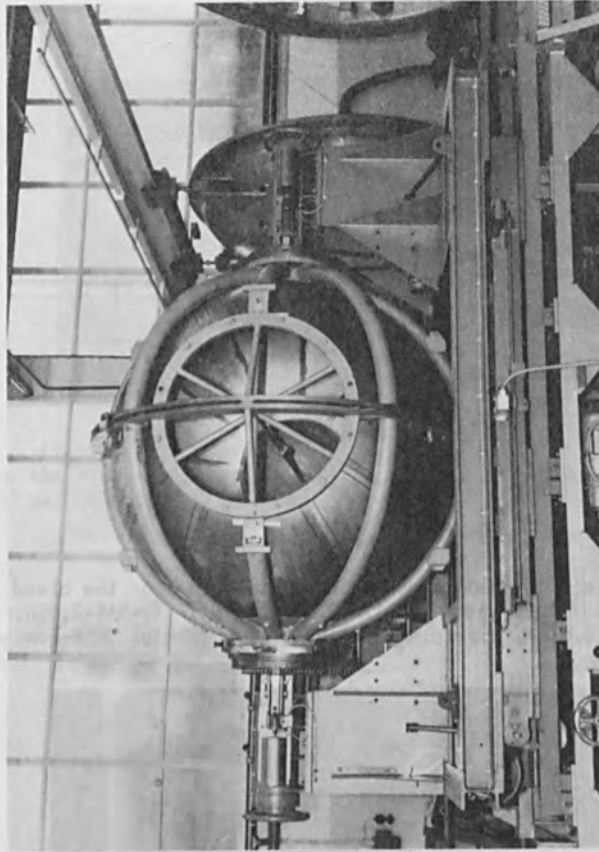


Fig. 14. Holding fixture for welding together the spherical shells of the propellant tank. The electron beam penetrates to the joint through the gap between the flanged rings. (Photo: VFW-Fokker)

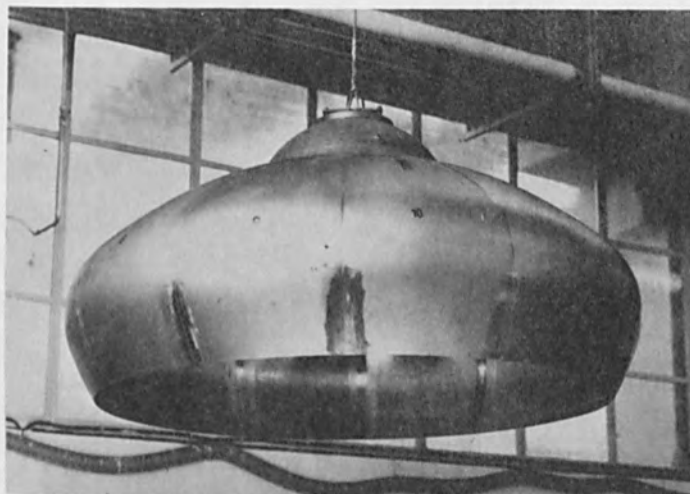


Fig. 15. Propellant tank diaphragm to separate oxidizer and propellant. Segment seams are ground off where they contact the spherical tank. (Photo: VFW-Fokker)

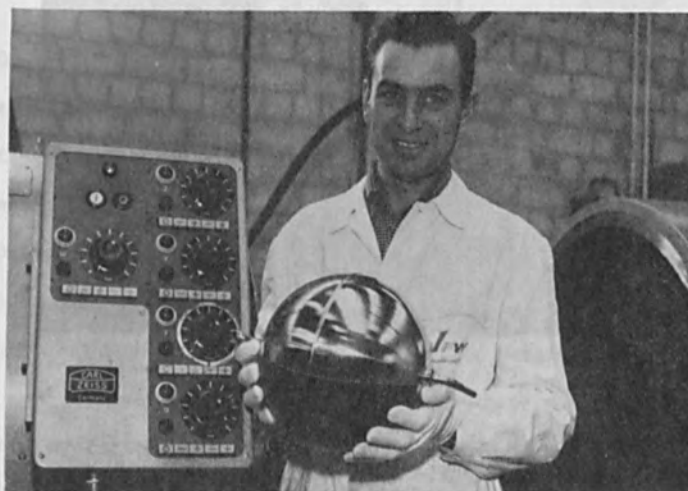


Fig. 16. Electron-beam welded satellite propellant tank for INTELSAT III. Material: Ti-6Al-4V, wall thickness: 0.5 mm, diameter: 258 mm. (Photo: VFW-Fokker)



Fig. 17. Electron-beam welded satellite pressurized-gas tank for TD-2. Material: Ti-6Al-4V, wall thickness: 6 mm, diameter: 460 mm. (Photo: VFW-Fokker)

ISOTOPE ENCAPSULATION BY ELECTRON BEAM WELDING

R. F. Boolen

Battelle Memorial Institute
Pacific Northwest Laboratory
Richland, Washington

ABSTRACT

One of the principal elements of the radioisotope research and development program at Hanford has been development of heat and radiation source fabrication technology. This includes electron beam welding development, a part of which comprises the content of this paper. $^{147}\text{Promethium}$, $^{239}\text{plutonium}$, and $^{241}\text{americium}$ were encapsulated. Both low and high power density electron beams, with open handling techniques heretofore thought too hazardous to be used, were employed to encapsulate certain of these radioisotopes, mainly in refractory metal claddings.

A successful method of encapsulating these highly toxic alpha and beta emitters without excessive contamination of electron beam equipment was found. The seriousness of low level contamination,

which is inevitable on primary welds, within the welder, depends upon the type of welder used. It was found that radioisotope contamination in high voltage machines did not reach the light optics or gun--allowing normal maintenance and adjustment of these parts.

Leak testing of capsules was done through the use of helium mass spectrometers and certain back pressurizing techniques were used to insure the validity of the method.

The use of a number of different metals and alloys was investigated. Electron beam test welds were made for each cladding material and these were evaluated through metallographic techniques.

A heat source of $^{147}\text{promethium}$ was incorporated in the Radioisotjet (microthruster) for the AEC and NASA. The critical welds were made by electron beam. Radiation sources made of $^{147}\text{promethium}$ were encapsulated for use in In-Phantom Dosimetry studies. Many $^{147}\text{promethium}$ capsules were constructed for compatibility tests of cladding and core. A heat source made up with $^{147}\text{promethium}$ as the fuel was used as a thermal preconditioner for the Advanced Manned Strategic Aircraft (AMSA) test program.

Future work in encapsulation of heat and radiation sources will be the development of specialized equipment for producing large numbers of capsules. The equipment will be made up mostly of soft-vacuum (partial vacuum) and out-of-vacuum welders. Laser welders will be used to encapsulate thin walled liners in packaging radioisotopes--with no welding equipment in the cell.

INTRODUCTION

One of the principal elements of the Radioisotope Research and Development program at Hanford has been development of fabrication technology. This involves the adaptation of nuclear fuel technology to radioisotope heat and radiation source fabrication. The technology includes electron beam welding development and processing, particularly adapted to the encapsulation of promethium (Pm) and other radioisotopes for heat and radiation sources. It was recognized at the outset of the program in 1966 that refractory metals would be the most likely materials to be used in encapsulation of radioisotope sources and that welding processes most applicable to these materials should be used.

We were well experienced in welding most of the refractory metals due to our involvement of several years in the fabrication of test assemblies for reactor cores, ceramics, and high temperature research. We also recognized that the relatively new electron beam welding process held great promise as the best process for welding refractory metals, and combined the additional advantages of sealing evacuated capsules, and welding with a much reduced heat energy input, as compared to gas tungsten arc welding.

Equipment

We began working with a 2 kilowatt, 150 kilovolt electron beam welder. This machine, which is shown in Figure 1, was tested and found to be extremely useful in producing welds in refractory claddings of limited thickness. Figure 1 shows externally all the equipment

necessary for electron beam welding. At the left is a cabinet containing the low voltage supply transformers. To the right of this, in the rear, may be seen a part of the high voltage transformer. In front of these is the high voltage desk for controlling the transformer output. To the right are the filament and bias control console and pulsing unit. In the center, the gun and optics column sit atop the welding chamber. The binocular optics--with up to 20X magnification--allowed precise positioning and welding of thin wall and small capsules. At the extreme left may be seen an inert gas cylinder, with disconnected hose atop the transformer cabinet. This was consistently used for backfilling the chamber after welding--both to provide helium for leak testing and provide inert rapid quenching to speed work throughput.

Our work with the 2KW machine proved to be so successful that a larger 6KW machine was procured with which to do increased development in the encapsulation of radioisotopes. This machine is shown in Figure 2. Compared to the 2KW machine, this welder--in addition to having increased welding capacity--has precise voltage regulation (within $\pm 1\%$ regardless of load condition) and automatic up and down slope for the beginning and finish of a weld. It also has a swing-away console, providing access to a pair of glove ports in the front of the chamber.

For the welding of containers of radioisotopes, a number of precautions must be taken. These are necessary because of the very toxic nature of nearly all radioisotopes.

A cut-away view of the welder (Figure 3) serves to illustrate some of the features which are used to advantage in welding radioisotope capsules. Notice that the beam can be made to pass a relatively long distance below the deflection coil area to weld a capsule. This means that contaminating backstreaming ions and spatter are remote from the welding head.

The roughing line can be valved to a high efficiency building exhaust converting the welder to either a glove box, where the glove ports are used, or to an open faced hood when the door is open. There is also a separating vacuum valve that allows work to be done on the gun (such as changing filaments) without breaking the box vacuum or containment.

DISCUSSION

Handling Problems of ^{147}Pm

^{147}Pm was the first radioisotope we encapsulated with the electron beam welding process. This radioisotope presents radiation handling problems very different from that of any other radioisotope. Those required to handle it or work in glove boxes contaminated with it, must follow careful handling procedures. These personnel must also be familiar with the peculiarities of promethium in order to safely work with it. Table I is a listing of some comparative data which describes the major handling and contamination hazards of this isotope.⁽¹⁾

TABLE I
Comparative Isotope Data

Isotope	Half Life year	Specific Activity Ci/g	Radiation Particles	Toxicity MPC (Ci/cm)	
				in air	in water
Pm-147	2.6	928	beta	6×10^{-8}	6×10^{-3}
Pm-146	4.5	545	beta-gamma		
Pu-238	89.0	17	alpha-gamma	2×10^{-12}	1×10^{-4}
Pu-239	24,360.0	0.61	alpha-gamma	2×10^{-12}	1×10^{-4}
U-Natural	4.5×10^9	--	alpha	1×10^{-11}	

When first asked to provide recommendations of needs in welding of ^{147}Pm , the author requested that every effort be made to fabricate the fuel core in the form of dust-free pellets. There are several good reasons why this is desirable; (1) the presence of Pm compounds in welds can cause porosity and weakening of the seal joint, (2) the presence of Pm in a weld zone surface presents a contamination hazard, and (3) the handling radiation dose is reduced to a minimum where diffusion of Pm dust particles is prevented. In the case of (1) and (2), it was known from prior work with nuclear fuel forms that loading of radioactive material in any form except a dust-free pellet, in a can or tube will result in some contamination of the weld joint. In practice, only a relatively dust-free pellet can be prepared. The case for (3) is exemplified if one realizes that as little as one milligram of promethium, spread over a hood floor as an oxide, will produce a surface dose rate of about 1R/hr.

The method of encapsulating highly toxic alpha and beta emitters without excessive contamination of electron beam equipment consists of

densifying the compound into a dry solid pellet, or combining it with a matrix material to form a dry solid composite pellet. Special filters may be used where the formation of a dust-free pellet is not possible, or is not desirable.

Gamma and Beta Dose Problems

Distance is a very important factor with promethium as it is mostly a low energy beta emitter and such particles are easily shielded.* Table II indicates the dose rates to be expected from two different power sources when shielded by typical cladding thicknesses. Considering only the shielding effect, we see in the first example of the Table that an increase in refractory metal cladding, in this thickness range, results in a corresponding percentage reduction of total dose rate-- and that the addition of a super metal, in this case Hastelloy, results in no significant reduction of dose rate.

Figure 4 indicates the serious dose rate problem when handling of ^{147}Pm sources which must be done at close proximity. The dose rate from a 65 watt source, when shielded by 60 mils of rhenium is almost one-half an R/hr. This means that handling of a source at this range by one person would be limited to only a few hours per month. Note that most of the dose rate comes from the impurity, $^{146}\text{promethium}$, rather than from $^{147}\text{promethium}$.

* The beta radiation from $^{147}\text{promethium}$ is stopped by about 65 mg/cm² of any absorber, so the primary hazard except at very close range is from secondary bremsstrahlung radiation.

TABLE II

DOSE RATES FROM PROMETHIUM HEAT SOURCE CAPSULES

Source of Radiation	Dose Rate, mR/hr				
	50 cm		105 cm		From 20 Watt Source 60 mil Ta
	30 mil Re	From 60 Watt Source 51 mil Re	Re+25 mil Hast.	From 20 Watt Source 60 mil Ta	
Pm^{147}	17	2.4	2.2	0.1	
Pm^{146}	42	38	34	3.5	
Pm^{148m}	2.8	2.5	2.5	9.0	
Eu^{154}	9.9	8.9	8.7	2.4	
Total Calculated	72	52	47	15.0	
Total Measured	72	52	48	14.5	

RATIOS OF ACTIVITY TO Pm^{147} ACTIVITY,
60 WATT CAPSULE

Pm^{146}	2.2×10^{-7}
Pm^{148m}	4.2×10^{-9}
Eu^{154}	2.5×10^{-8}

Contamination Control and Hazards

Fortunately promethium, if ingested, is not a bone seeker nor is it known to be concentrated in any part of man's digestive chain. Less than 0.1% of promethium ingested is known to be taken up by any internal organ. The greatest hazard comes from inhalation, where about 25% remains as body deposition, mainly in the lungs, bone, and liver. Toxicity limits for promethium are much lower than for plutonium on a Curie basis, but on a weight basis, because of the high specific activity of promethium, they are fairly close.

To complicate the situation somewhat, the very low beta energy of Pm cannot be detected by the customary GM tubes. A special mica window probe instrument is necessary to properly survey Pm surface contamination. In addition, the survey must be accomplished in a low radiation background area which frequently requires taking smears and moving to a low radiation background for counting. Wet surfaces are also unsatisfactory because of the low penetration of the beta radiation.

Decontamination is most easily accomplished with detergents and water. Organic solvents have not been used successfully in large scale decontamination.

The unique properties of Pm, which are its high specific activity and intense low energy beta radiation, result in the need for shielded glove boxes and transfer containers, specialized radiation monitoring equipment and careful attention to operating procedures.

Facilities For Loading, Storage, and Transfer In Welding

A full range of facilities for loading, storage, and transfer of capsules and components was made available during most of the programs. These included hot cells, both shielded and unshielded glove boxes, and fume or open-face hoods. A portable, self-contained radioisotope glove box proved to be a great aid in testing capsules prior to insertion in the electron beam welder.

The high level radiochemistry cells made available for loading and assembly of some radioisotope capsules were generally the same ones which had also been used for densification and formation of the dry radioisotope material into pellets.

The welding engineer or welder must take part in planning the procedure for core processing; for it is here that loading and handling must be done without excessive contamination of the seal weld joint, if high integrity welds are to be made.

It was found that joints designed for a press fit should not be used unless a gas pressure relief hole is included in the design. The diametral fit-up between cap and container for EB weld joints may be allowed to vary between only one or two mils clearance without ill effects.

Since capsules must normally be evacuated for welding with electron beam, and electron beam welders are not expendable, it was found best to pre-pump the radioisotope filled capsule in an expendable chamber, such as a dessicator jar, with a high efficiency

filter system between the pump and the capsule. Such a procedure is a cheap method of determining the contamination potential of a given capsule.

Materials of Interest

Most cladding materials with which the welding personnel have been concerned are metals of the refractory type. One class of capsules comprised aluminum as the cladding. Claddings fall into two categories of use; primary and secondary. Since the primary cladding is in contact with the radioisotope, the choice of this material depends mainly on its chemical and thermal compatibility with the radioisotope to be used. Depending on the intended use of the capsule, the specific generation of gamma radiation (by bremsstrahlung) may also play an important part in selection of the primary cladding. Mechanical strength is of secondary importance for this first jacket or cladding. Liners which would form a complete shielding around a promethium core, although tried in simulated assemblies, have not been used in actual encapsulation--mainly because simulation has shown several concepts to be awkward and unworkable in a remote assembly situation.

Materials which have been electron beam welded in order to study their weldability in preparation for use as primary claddings, are shown in Table III. Of these, only aluminum, tantalum, rhenium, tungsten-25Re, Haynes-25, and 304L stainless steel have actually been used by us as primary claddings in promethium encapsulation.

TABLE III

PRIMARY CLADDING MATERIALS

ALUMINUM	TUNGSTEN
TANTALUM	TUNGSTEN-25 Re
TANTALUM-10 TUNGSTEN	HAYNES 25/L-605/WF-11(Co-Cr-Ni)
MOLYBDENUM	HASTELLOY ALLOY C
MOLYBDENUM-0.5% TITANIUM	HASTELLOY ALLOY C-276
TZM (Mo-0.5% Ti-0.1% Zr)	304 L STAINLESS STEEL
RHENIUM	316 L STAINLESS STEEL

COMPATIBILITY CAPSULES

Capsules in various couples (Sm_2O_3 or $^{147}\text{Pm}_2\text{O}_3$ to metal cladding) were needed to assess the service compatibility of $^{147}\text{Pm}/^{147}\text{Pm}_2\text{O}_3$ with the candidate cladding metals in certain heat source designs. General requirements in welding these capsules called for seal welds to be made with joint penetration equal to wall thickness and leak tight to less than the minimum detectable amount (back-pressurizing technique described later).

Electron beam welding was selected for closure of compatibility capsules because of its several advantages; a high vacuum atmosphere during welding improves the quality (purity) of weld metal in refractory materials. A vacuum is desirable in the finished capsule to eliminate or reduce impurity gases which would otherwise be a factor in compatibility. Low heat input is desirable to prevent volatilization of

$^{147}\text{Pm}_2\text{O}_3$ during welding of refractory metal capsules ($^{147}\text{Pm}_2\text{O}_3$ melts at 2300°C , which is several hundred to 1000°C less than some of these refractory claddings).

^{147}Pm Contaminated Sm_2O_3 /Haynes-25 Couple

This capsule, designed for approximately 7 grams of radioisotope material, was fabricated to indicate feasibility of the chosen seal welding process and to aid in determination of compatibility of ^{147}Pm /Haynes-25 couples densified by pneumatic impaction.⁽²⁾ This first promethium capsule was successfully fabricated only after considerable difficulty. Both the capsule fabrication and testing, however, provided valuable information in excess of the planned investigation. It was desired that the capsule be evacuated and heated to out-gas prior to fusion welding. Electron beam welding was chosen to accomplish this.

A simulated capsule of the required design, loaded with Sm_2O_3 in an out of cell facility, was first successfully sealed by welding with a low voltage EB welder with no difficulty. The ^{147}Pm contaminated- Sm_2O_3 capsule was then loaded by step pressing Sm_2O_3 powder in-can in the cell facilities which had been contaminated during processing of ^{147}Pm . The outer surfaces of this unsealed capsule were decontaminated to a non-smearable level only after considerable difficulty. After decontamination the assembly was securely clamped and sealed in a shielded container, and transferred to a gloved chamber where GTA tack welds were made in the periphery joint of the capsule. After making the tack welds at the peripheral joint, the unsealed capsule was

transferred to a low voltage EB weld chamber. A satisfactory seal weld was made at the capsule weld joint, however, internal parts of the weld chamber were contaminated with up to 60,000 dpm, smearable.

Pm₂O₃-Haynes-25 Couples (Capsules)

In order to circumvent the problem encountered with the first capsule a modified design of the capsule lid was chosen to mechanically seal in the fuel material, allowing partial decontamination and secondary encapsulation in a non-contaminated cladding. This modified design is shown in Figure 5. The procedure for this capsule called for partial decontamination following step press loading, pressing the fritted cap in place to make the mechanical seal, and then inverting and inserting the couple into a secondary stainless steel cladding of similar design.

The procedure for this design, following manual GTA welding all around the periphery, was to transfer the assembly to the low voltage EB welder, heat and outgas the capsule and seal weld the plug into the vent hole with the electron beam. Three capsules, each containing approximately 7 grams of ¹⁴⁷Pm₂O₃ were successfully assembled and seal welded in the manner described above. The cross section of a typical ¹⁴⁷Pm capsule of this type, in the as-pneumatically impacted condition is shown in Figure 6 (macrophoto).

Joint Design

An additional requirement in electron beam welding is location of the joint in such a manner that, when complete penetration through the joint occurs due to clearance between the faying surfaces, the

electron beam cannot enter and damage the core material. Figure 7 shows the design most often used to satisfy this requirement.

Processing

A special technique for obtaining a loaded, non-contaminated capsule was developed; prior to loading a capsule, the cladding was wrapped with two layers of aluminum foil. The outer layer was curled over the top edge to protect the weld zone from contamination, as shown in Figure 8. The capsule and plug were placed in clean plastic vials and inserted into the cell containing the sintered Pm_2O_3 pellet. A fresh sheet of paper, along with a clean pair of forceps contained in a plastic bag, was placed in the cell. The clean forceps were used to place the capsule on the paper, and the pellet was inserted by means of a different pair of forceps. In each step clean forceps were then used to insert the metal plug into the capsule, to remove the outer layer of foil, and to place the capsule into a clean lead holder in the air lock. The lead shielded portion of the air lock was then rotated 180° to expose the capsule at the rear side of the cell where contamination checks are made, any minor decontamination necessary is done, and the capsule lid is fitted into place.

These capsules, which were made later in the program, were seal welded in our 2KW high voltage welder. It is significant that even though appreciable levels of $^{147}\text{promethium}$ contamination were often found in the chamber--no contamination of the gun or light optics occurred. This means that the seriousness of low level contamination, which is inevitable on primary welds, within the

welder, depends upon the type of welder used. It was found that radioisotope contamination in high voltage machines does not reach the gun--allowing normal maintenance and adjustment of these parts.

After welding of promethium filled compatibility capsules, vapor deposition of a trace amount of $^{147}\text{Pm}_2\text{O}_3$ in the heat shield area of the electron beam welder was found, as anticipated. Contamination problems with the vacuum chamber were minimized by covering the major surfaces exposed to 147 promethium deposition with aluminum foil, disposable after use.

Leak testing of capsules sealed in vacuum was performed by the use of two tests; (1) a low power microscope was used to examine the welds for any cracks or holes which might be visible, and (2) a technique known as the "back pressurizing" technique⁽³⁾ was consistently used. Essentially the latter test consists of placing the capsule within a chamber which is then evacuated to several hundred Torr or less, held for a certain length of time, pressurized with helium for a controlled time, then removed and placed in the leak detector chamber where the pressure is reduced to a level compatible with helium mass spectrometer sensing, which then is used to determine whether the capsule leaks or not. Of course a leaking primary capsule will normally be found by smear tests (using a suitable detector) before either of the two tests described above are run.

RADIOISOJET HEAT SOURCE CAPSULE

The first major project for which the program was designed was a 'radio-isojet' heat source for the AEC and NASA. This was used as a microthruster designed for attitude control and stationkeeping of certain spacecraft.

Original design of the radioisotope powered microthruster, shown in Figure 9, was based on the design of an existing electrically powered microthruster. This was done to maximize the interchangeability of parts, to provide for direct comparison with the electrically powered thruster, and to minimize costs. The constraints of the design of the capsule heat source imposed by thruster operating conditions and interchangeability included:

- (a) Fuel form: $^{147}\text{Pm}_2\text{O}_3$ having demonstrated compatibility with the cladding.
- (b) Power: Nominal 60 watts thermal.
- (c) Shielding: 50 mils minimum of re-fractory metal (radioisotope capsule cladding) for Bremsstrahlung radiation control and heat resistance.
- (d) Inner thruster body: 25 to 30 mils of Hastelloy Alloy X (HX).
- (e) Encapsulation: double rhenium cladding.
- (f) Capsule dimensions (maximum): 1.145 inches diameter x 2.745.

Other specifications which might be required, as determined during the course of development would also apply.

The choice of radioisotope capsule cladding was based in part on fuel-primary clad compatibility. Compatibility studies at PNL have shown that Pm_2O_3 can be adequately contained in a variety of metals at 100°C for several months or longer.⁽⁴⁾ Metal-metal compatibility also plays an important role when multi-clad capsules are to be considered. The final choice of pure rhenium as the radioisotope capsule cladding resulted from a blend of the desired shielding, compatibility, physical properties, etc.; requirements known at the time of decision.⁽⁵⁾

Because of limited availability, toxicity, and radiation generated from Pm_2O_3 , most of the welding and fabrication studies in which a core material was required were performed using the non-radioactive Sm_2O_3 as a stand-in. $^{147}\text{Pm}_2\text{O}_3$ was used in the encapsulation of two 1/3 scale capsules to verify results which had been obtained using Sm_2O_3 (or in some cases Nd_2O_3). The smaller capsules served to reduce material costs and availability problems in providing an onsite timely thermal test to prove promethium encapsulation feasibility, and to verify weld integrity and chemical compatibility.

The encapsulation process for the two 1/3 scale capsules was prototypic of the full scale microthruster capsule. Specifications called for a length-to-diameter ratio equivalent to that of the designed full scale capsule. Both 1/3 scale capsules were refractory metal clad, one rhenium and one tantalum.

The encapsulation development program for this specific application and concept study resulted in a process which included double encaps-

sulation of the fuel and assembly of the encapsulated heat source into the inner and outer microthruster components using both GTA and EB welding.

Welding development was required for three areas of the work:

- (a) Seam and seal welding of 1/3 scale cladding and capsules.
- (b) Seal welding of full scale cladding and the final capsule and
- (c) Seal welding and assembly of radioisotope components.

The latter area of work was not designed for EB welding, and is therefore not included in this article.

Material

Two supplies of rhenium were procured for the program. Both were sintered powder (powder metallurgy processed and rolled) products. The first consisted of a small amount of 0.030 inch thick sheet. Results of welding this material were less than satisfactory due to excessive porosity. Only one vendor could be found who would supply the other rhenium, which was needed for full scale capsule welding development and construction. This material consisted of rhenium of 99.99% purity in the form of containers (0.030 inch wall thickness--0.060 inch thick bottom) and 0.060 inch thick caps. This material was also a powder metallurgy product and results with it were similar to those in 1/3 scale work. The seam welds in the cans, as fabricated in the vendor's plant were made by a semi-automatic gas tungsten-arc (TIG) process.

Radiographic and fluorescent penetrant tests indicated gas porosity and poor penetration in portions of seam welds in all the containers, and poor penetration in all container lid periphery welds. Ultrasonic testing confirmed lack of weld penetration of up to 50% of the joint thickness. Porosity and other discontinuities can be seen in the radiographs of the container seam welds (Figure 10).

Metallography of samples taken from the containers showed the parent metal to be in a well annealed condition.

Two bars of Hastelloy Alloy X were procured for the inner thruster body. The first bar was rejected because of a center defect (shrinkage crack) running its full (6 ft) length. Ultrasonic, dye penetrant, and metallographic testing of the good bar and sections thereof were done to assure integrity of the material.

Tantalum and arc-cast molybdenum were procured for fabrication of simulated test workpieces.

Design and Development

A thin pre-formed disc, of the same material as the container, was designed for placement over the Pm_2O_3 load to aid in decontamination and to act as a barrier to electron beam impingement on the pellets during seal welding of the vent hole.

The final design was based on the use of pressed and sintered Pm_2O_3 pellets, GTA welding of the periphery joints, and EB seal welding of the vent hole at one end of each of the first and second claddings.

As mentioned above, both the EB and GTA fusion processes produced welds in the rhenium which exhibited considerable gas porosity and varying penetration. Sufficient testing to find the cause of porosity in the welds could not be done, but it may be assumed that rhenium heptoxide at the grain boundaries was the major contributor. This volatile oxide has a melting point of 297°C and a boiling point of 363°C.

Vent holes in capsule endcaps were formed by drilling with the electron beam. A continuous beam of approximately 110KV and 6 to 7 ma beam current for two to three seconds was required to form a hole in 0.030 inch thick material. This technique invariably caused cratering on both sides in rhenium material. Such extreme cratering was not experienced previously when drilling holes in W or Ta. By using the EB welder pulser (above parameters approximately equal) at a frequency of 45 cps and a pulse width of 5 msec the cratering was eliminated.

Closure of outgas holes was developed and later accomplished by manually controlling the magnitude and sweep of a deflected beam of approximately 110KV (no load) and 5 ma beam current. The outgas vent hole was fitted with a parent material spike in each case, to prevent cratering and excessive impingement of the beam on inner components.

Loading and Welding

The primary claddings were loaded by the process previously described. The foil covered full scale container was loaded inside the hot cell using clean, new forceps at each step (Figure 11).

A heavy copper chill block, close fitting for each capsule, was used to prevent overheating and volatilization of Pm_2O_3 during both GTA and EB welding. Following GTA welding, vent holes were closed in the electron beam welder to provide the desired vacuum. When insulated by vacuum, as during pumpdowns in the GTA inert gas weld chamber, the full scale capsule glowed red hot from self heat, as shown in the lead photograph.

A circumferential square butt joint design was chosen for joining the inner thruster body sheath (tertiary cladding) to the top dome (end cap). These components were fabricated from fully tested bar stock. A view of the completed inner thruster assembly (held in cooling chill block) is shown in Figure 12. The electron beam weld is evident near the flanged end. Prior to making this weld a 2 ma electron beam with a transverse deflection amplitude of 1/32 inch at 60 hz and 115KV potential and a weld speed of 30 ipm was determined to be optimum after sectioning and examining several sets of simulated parts. The cross section of a typical weld is shown in Figure 13. Cracks, which were noted in the end cap HAZ only, were not of serious consequence to the integrity of the joint.

Helium leak testing of capsules was performed following each seal weld--all welds and claddings were found to be leak tight to less than 10^{-6} atm cc/sec, He. Visual examination and ultrasonic testing were also used to assure final integrity.

WELDING FOR THE AMSA RADIOISOTOPE APPLICATION PROGRAM

This is a description of development and fabrication welding which ultimately produced a flight-qualified 65 watt $^{147}\text{Pm}_2\text{O}_3$ heat source for the Air Force. A thermal preconditioner, for which this capsule was designed, is a temperature maintenance device for an inertial measuring unit

(IMU). The IMU is targeted for use on the advanced manned strategic aircraft (AMSA), hence the capsule, for short, has been dubbed the "AMSA capsule".⁽⁶⁾ The AMSA radioisotope capsule development program was divided into three phases:

Phase I - Fabrication of six to ten capsules loaded with Sm_2O_3 to evaluate the impact rupture resistance of capsules prepared by designed fabrication processes. These included both pressed and sintered pellets as well as pneumatically impacted capsules.

Phase II - Fabrication and testing of twenty capsules containing Sm_2O_3 fuel simulant to qualify a final design.

Phase III - Fabrication and delivery of a single 65 watt Pm_2O_3 capsule for use by the U.S. Air Force in tests of this prototype thermal preconditioner.

A sketch of the capsule as designed appears in Figure 14. The inner clad is tantalum while the outer is a Co-Cr-Ni alloy.

Two core assembly procedures were considered. One was a slip-fit design, using pressed and sintered pellets (primary process), and the other was a pneumatically impacted capsule design (alternate process). All required development and fabrication (slip-fit assembly) were accomplished essentially as planned.

Table IV (Impact Test Data and Post-Impaction Evaluation) provides an over-all summary of the individual tests. This Table provides information regarding impact conditions and post-impaction inspection for every capsule. The weld inspection and comments are listed in the last column. Impact testing⁽⁷⁾ was performed by Sandia Corporation (AEC), Albuquerque, New Mexico.

TABLE IV

Impact Test Data and Postimpact Evaluation

AMSA Capsule No.	Impact Angle	Velocity ft/sec	Target Material	Results	Postimpact Weld Inspection and Comments
1	22-1/2	414	LV granite	intact	90% pent. - excellent weld
2	not tested	-	-	-	Destructive evaluation
3	22-1/2	400	ES granite	intact	80% pent. - weld root voids
4	45	396	LV granite	intact	45% pent. - weld reject
5	0	396	ES granite	intact	95% pent. - excellent weld
6	67-1/2	398	ES granite	intact	95% pent. - excellent weld
7	45	406	ES granite	rupture	50% pent. - reject
8	0	403	ES granite	intact	70% pent. - reject
9	67-1/2	407	ES granite	intact	40% pent. - reject
10	90	410	ES granite	intact	75% pent. - weld root voids
11	not tested	-	-	-	Destructive evaluation
12	0	494	ES granite	intact	90% pent. - excellent weld
13	22-1/2	494	ES granite	rupture	90% pent. - excellent weld
14	45	479	ES granite	rupture	85% pent. - excellent weld
15	67-1/2	479	ES granite	intact	95% pent. - excellent weld
16	0	485	ES granite	intact	95% pent. - small voids
17	22-1/2	485	ES granite	rupture	95% pent. - excellent weld
18	45	487	ES granite	rupture	95% pent. - small voids
19	45	381	ES granite	rupture	95% pent. - excellent weld
20	90	487	ES granite	intact	95% pent. - ~0.030 in. void
21	45	329	ES granite	intact	95% pent. - excellent weld
22	not tested	-	-	-	Destructive evaluation
23	not tested	-	-	-	Destructive evaluation
24	22-1/2	429	ES granite	intact	90% pent. - excellent weld
25	not tested	-	-	-	Destructive evaluation
26	35	388	ES granite	rupture	90% pent. - 0.010-0.015 in. voids
27	45	429	ES granite	rupture	95% pent. - excellent weld
	30	630	soil	intact	
28	45	385	ES granite	rupture	95% pent. - excellent weld
29	55	388	ES granite	rupture	85% pent. - excellent weld
30	35	374	ES granite	intact	95% pent. - excellent weld
31	45	374	ES granite	rupture	95% pent. - excellent weld
32	45	323	ES granite	rupture	85% pent. - 0.010-0.015 in. voids
33	45	392	concrete	intact	90% pent. - excellent weld
34	not tested	-	-	-	Destructive evaluation
35	not tested	-	-	-	Wind tunnel test - WPAFB
36	45	312	ES granite	rupture	95% pent. - excellent weld
37	45	285	ES granite	intact	95% pent. - 0.030 in. void
38	-	-	-	-	Not loaded
39	45	678	concrete	intact	85% pent. - small voids
40	45	650	concrete	intact	Not sectioned

Development Welding

Development efforts were aimed at establishment of procedures which would (1) assure the success of seal welding of the inner (primary) tantalum cladding, which was designed to be in contact with the pellet (Sm_2O_3 or Pm_2O_3), and (2) assure the required quality of the outer cladding.

Development of seal welding by electron beam (2KW EB welder) was based on three major steps:

- . Desired penetration
- . Elimination or minimizing of defects, and
- . Reproducibility.

Test weld workpieces (Figure 15A and B) were machined from certified material to simulate the primary cladding joints and evaluate weldability of the material. Fit-up requirements of the initial design (Figure 15A) could not be met using normal machining procedures, therefore, the design shown in Figure 15B was used for final processing.

Primary Welds

Since the seal weld in the tantalum primary cladding is mainly for contamination containment, achievement of a consistently successful seal weld was made the important consideration, rather than maximum joint efficiency (penetration). Release of radioisotopic material during welding is, of course, highly undesirable because of equipment contamination, formation of weld inclusions, and degradation of seal joint integrity.

It was determined through experimentation⁽⁸⁾ that a maximum of 50% joint penetration could be achieved without undue risk of blowout during welding. Weld cross sections were taken from capsules loaded with samarium oxide which were fabricated according to the initial joint design. Figure 16 shows the cross section of an optimum joint, as designed for 50% penetration of weld.

Secondary Welds

Initial problems in development of electron beam welding simulated outer cladding joints were (1) elimination of root voids, and (2) reproducible penetration. Surprisingly, no significant cracking of either of the Co-Cr-Ni alloys occurred (initially HS-25*, finally used L-605**).

Test welds were made on simulated joints having a 15 mil step, 100 mils from the inner diameter surface (final design). A finely focused beam, with 30 mils peak-to-peak deflection was used to make fully penetrated (1/4 inch) welds approximately 43 mils wide (Figure 17). The heat affected zone (Figure 18) shows only mild grain boundary precipitation. The peculiar root voids are illustrated in Figure 19. The root voids were not shown to be a significant problem because of undefined strength requirements, and indeed did not appear to contribute to impaction rupture failure as determined by postimpaction evaluation of Phase I capsules (1 through 10).

* Tradename of Union Carbide Corporation, Haynes Stellite Company Division

** Unitemp tradename

A few of the Phase I outer cladding welds exhibited good penetration and an absence of voids, as well as elimination of the "tooth root" configuration at the bottom of the weld. The parameters for these welds (unintentionally differing from the others in beam focus) appeared desirable for subsequent welding development. The requisite change in parameters consisted of defocusing the beam by shifting its focal point upward (consequently increasing spot size at the weld face) and reducing the transverse deflection to 16 mils. Although the new set of parameters resulted in welds of approximately the same width as those previously made, penetration and weld geometry were perfected and voids were minimized. A typical example of the final weld utilizing a 15 mil step 15 mils from the inner diameter surface is shown in Figure 20.

Optimized parameters for Phase II electron beam welding of L-605 alloy were:

<u>Beam Potential (KV)</u>	<u>Beam Current</u>	<u>Transverse Beam Deflection</u>	<u>Face Speed</u>
142.5 (No Load)	13.5 ma	0.016 inch	24 ipm

Postimpaction results and examination of weld cross sections served as a guide for final development. Optimum penetration established for the outer weld was 85% minimum, 100% maximum.

By means of an extensive review of postimpaction weld inspections (Table IV) the nondestructive ultrasonic test was refined to identify both the degree of weld penetration and voids as small as 0.010 inch in size.

The electron beam weld closure of the primary capsule inner tantalum cladding (Figure 21) was determined to be satisfactory by ultrasonic nondestructive testing. Destructive evaluation of the test welds, made before and after welding the fueled capsule, established weld penetration for the inner seal weld to be optimum (50% of joint thickness).

An as-welded AMSA capsule in the final cladding is shown in Figure 22. The weld bead, which comes entirely from the base metal, was later machined flat, as planned. Each seal weld on the actual capsule, both primary and secondary, was preceded and followed by simulated test welds which were both nondestructively and destructively examined for quality assurance.

An ultrasonic test profile of the ^{147}Pm loaded AMSA capsule (Figure 23) was made to verify weld integrity.

ENCAPSULATION OF Pm-147 RADIATION SOURCES
FOR IN-PHANTOM DOSIMETRY STUDIES^{9,10}

Requirements

These capsules were each to comprise a pressed and sintered Pm_2O_3 pellet (15 watts, nominal) doubly encapsulated in type 304L stainless steel. Weld closures were to have 100% or greater penetration at the joint (designed for 100% wall thickness), and have leak rates undetectable by helium mass-spectrometer testing. Two doubly clad capsules of this description were eventually fabricated and used by the Environmental Health Department of PNL. Appearance of the as-welded doubly-clad capsule appears in Figure 24. The capsules were used as radiation sources in the study of dose rates with a man-equivalent phantom (Figure 25).

Development

Electron beam parameters were developed through trial welding of empty test claddings and metallographic examination of sections thereof. No other destructive testing was required.

Test weld workpieces were fabricated with the radial offset joint which was found to be compatible with electron-beam welding. These were used to establish parameters to achieve 100% joint penetration in the loaded capsule. Parameters were:

Beam voltage = 110KV (no load)

Beam current = 1.8 ma

Transverse deflection = 0.008 inch (60 Hz)

Weld speed = 30 inches per minute

The 0.040 inch thick joint was tacked in three places by manual control of the beam. Radiation readings of the as-welded capsule were 100 rad/hr at contact and 2.5 rad/hr at 6 inches. Microphotographs of test weld cross sections were taken; the micrograph of Figure 26 is of a sample joint welded at slightly lower energy input than was used for actual $^{147}\text{Pm}_2\text{O}_3$ encapsulation. Porosity, an example of which may be noted in the photomicrograph, had no significant effect on joint integrity.

ENCAPSULATION OF TARGET ELEMENT CAPSULES FOR PROMETHIUM-146 BURNOUT STUDIES

These capsules were fabricated for a program⁽¹¹⁾ at PNL to establish the technical and economic feasibility of upgrading the properties of promethium for premium applications (such as the heart program) by reirradiation to selectively burn out the gamma-emitting ^{146}Pm isotope.

The capsules were prepared as follows: The pellet, together with a piece of aluminum-cobalt alloy flux wire, was placed in an aluminum can which was then sealed by electron-beam welding. Weld integrity was evaluated by a helium leak check, with external contamination of the welded capsule being below the detectable limit. A total of six capsules were prepared for each of two tests at different sites. The composition and dimensions of the cold pressed pellets and capsules are summarized in Table V. Figure 27 shows the twelve capsules after welding and leak checking.

TABLE V
Target Elements for Phase II ^{146}Pm Burnout Studies

	Savannah River	Oak Ridge
Capsule Material	1100 A1	1100 A1
Capsule Dimensions		
External Length, in.	1.50	0.408
External Diameter, In.	0.625	0.255
Pellet Dimensions		
Diameter, In.	0.500	0.200
Length, In.	0.95	0.25
Pellet Composition		
Pm_2O_3 , wt. %	5	10
Aluminum, wt. %	95	90
Pellet Weight, g	~ 8	~ 0.4
Pellet Density, g/cm^3	2.75	3.02
Pellet Density, % of theor.	~ 93	~ 94

Processing

The cermet pellets were made by thoroughly mixing Pm_2O_3 with aluminum powder. Unlike previous capsules, the weld lip of these containers was not kept free of contaminating promethium particles by a foil wrap. The small size of the capsules made this technique impractical. Rather, a mild steel insert, with a very thin wall inserted into the capsule mouth, was used to prevent such contamination.

The twelve aluminum clad capsules, containing $^{147}\text{Pm}_2\text{O}_3$ -Al cermet pellets, were loaded in an open-faced hood and sealed in vacuum by electron-beam welding as planned. The large (SRP) capsules were in turn encapsulated, in groups of three, in two secondary aluminum capsules.

Successful loading and seal welding of these capsules marked the first time $^{147}\text{Pm}_2\text{O}_3$ pellets were encapsulated in the laboratory without the use of a protective cover disc or foil directly over the pellet.

DEMONSTRATION SAMPLES FOR AN EASE Pu-238 HEAT SOURCE CAPSULE

Preliminary EB welding of workmanship samples for the EASE (Early Apollo Surface Experiment) ^{238}Pu heat source capsule was performed for the AEC. This work was done in anticipation of fabrication of two 15 watt heat sources, one of which was scheduled for use on the Apollo 11 mission. The capsule core was to be 20-50% ZrO_2 with the remainder $^{238}\text{PuO}_2$. Compatibility claddings of TZM, molybdenum, tantalum, and HS-25 were considered.

Since we had already gained much experience in welding tantalum and HS-25, the workmanship samples were of TZM and molybdenum. Figure 28 shows demonstration workmanship pieces of actual cladding with a $^{238}\text{UO}_2$ fuel load to simulate ^{238}Pu . An inner cladding of 0.015 inch wall molybdenum was made up of two hemispheres, which were joined to a cylinder to form the primary container. One hemisphere had a small 1/4 inch diameter hole in one end through which the pelletized core ($^{238}\text{UO}_2$) was loaded, followed by final seal welding with the electron beam, with which all welding was done.

ENCAPSULATION OF $^{241}\text{AMERICIUM}$

Five capsules containing this radioisotope were encapsulated using the 6KW electron beam welder. The 3/4 inch diameter capsule cladding was 316 stainless steel with a weld penetration thickness of 0.080 inch. The design of the capsules, (Figure 29) includes an annealed brass sealing ring which comes into play when the cap is pressed into the container, thereby sealing the lower compartment by means of the spacer. This holds the $^{241}\text{americium}$ pellet against the 0.010 inch thick "window."

Several empty claddings and one loaded with stand-in Sm_2O_3 were welded to establish optimum parameters as follows:

<u>Beam Voltage</u>	<u>Beam Current</u>	<u>Deflection</u>	<u>Focus</u>	<u>Weld Speed</u>
110KV	4.0 ma	0.015 Circle @ 60 hz	at Surface	30 ipm

The capsules were pre-pumped to indicate contamination potential; no contamination was experienced, and both visual inspection and helium leak testing proved the capsules to be of required integrity.

GENERAL WELDING DEVELOPMENT APPLICABLE TO ENCAPSULATION OF RADIOISOTOPES

Because of the very large amount of so-called "waste" created in the production reactors at Hanford, and the radioisotopes available from both the waste and the reactors, we have had many radioisotopes to encapsulate. Most of these have, of course, not been encapsulated by electron beam welding. Those which have been encapsulated include tellurium, $^{210}\text{polonium}$ and $^{85}\text{strontium}$ as well as $^{147}\text{promethium}$. $^{239}\text{plutonium}$ and $^{235}\text{uranium}$ are encapsulated in such large quantities at

Hanford--in experimental fuel elements--that we normally do not consider them a part of our radioisotopes program. A listing of the programs involving these latter two is too large to be included in the scope of this paper, even though electron beam welding has, and is being used in many of them. Over 62 individual capsules containing radioisotopes were encapsulated with the electron beam.

EQUIPMENT FOR RADIOISOTOPE ENCAPSULATION IN THE FUTURE

Several types of equipment will see new and increasing use in the encapsulation of radioisotopes. More electron beam welding, similar to that described in this report, will be done on low level radioisotopes. Much good work has been done at ORNL on in-cell electron beam welding of radioisotopes.⁽¹²⁾

The welding encapsulation of intensely radioactive isotopes, such as ^{90}Sr strontium, ^{137}Cs cesium, and ^{144}Ce cerium will see increasing use, not only of the ORNL techniques, but of a new welding method. This is non-vacuum or out-of-vacuum electron beam welding.

We have seen that electron beam welding is extremely useful for high integrity closure of many radioisotope heat sources. This is especially true when the cladding materials are refractory or high temperature alloys, such as the super-metals now being used to encapsulate space environment heat sources. Electron beam welding of radioisotope heat sources by most fabricators has been to date very limited--limited in most primary encapsulation of radioisotopes because contamination levels or potentials are too high for reasonable adjustment and maintenance of

an electron beam welder. As described in this paper, Battelle-Northwest has done considerable primary encapsulation--by electron beam weld closure--on numerous $^{147}\text{Pm}_2\text{O}_3$ capsules through special fuel formation and handling techniques. Other past and present limitations in using electron beam in heat source encapsulation are the impracticality of remote operation of an electron beam welder and the lack of confidence of many sites in safely using (from an equipment standpoint) an electron beam welder in such work.

Electron beam welding will be increasingly used in radioisotope encapsulation in the future, and its inherent advantages--high energy density and precision alignment for critical applications--will be used in closure welding of heat sources.

It will be used because the electron beam welder can remain outside the cell while passing the beam through differentially pumped orifices in the cell wall (Figure 30) to make EB fusion welds on high level radioactive heat sources within the cell--just as non-vacuum EB welds are being made on non-radioactive assemblies every day.

By use of a differential pumping system, the integrity of the hot cell is not broken, and the electron beam welder is mated to the hot cell to allow this most desirable closure weld process to be used routinely in heat source fabrication. All parts of the system are in current manufacture, and combination of them is all that remains to be done. The EB welder head remains outside the cell for easy, low cost alignment and maintenance.

Soft or partial vacuum electron beam machines will also see increasing use in encapsulation of radioisotopes--particularly in high production applications.

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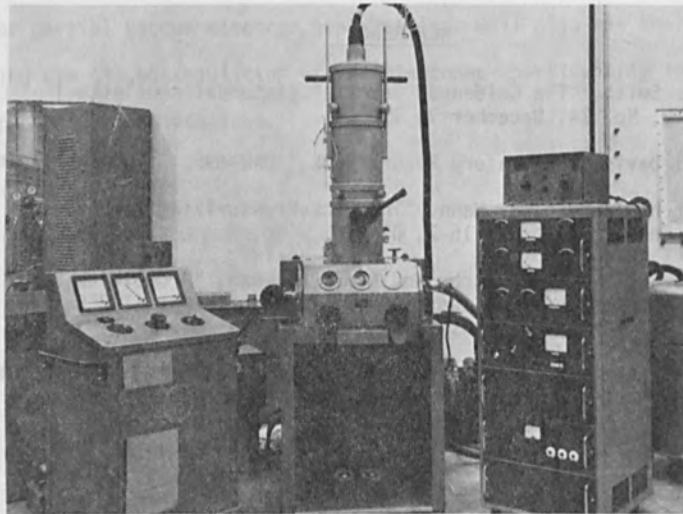


Fig. 1. 150KV, 2KW Electron beam welding machine.

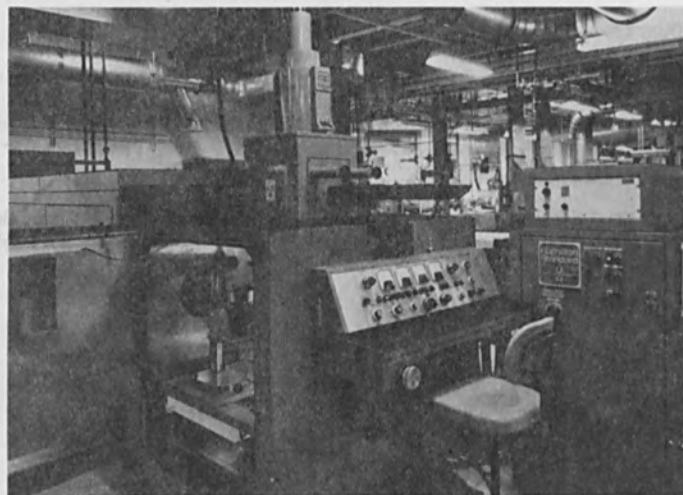


Fig. 2. 150KV, 6KW Electron beam welding machine.

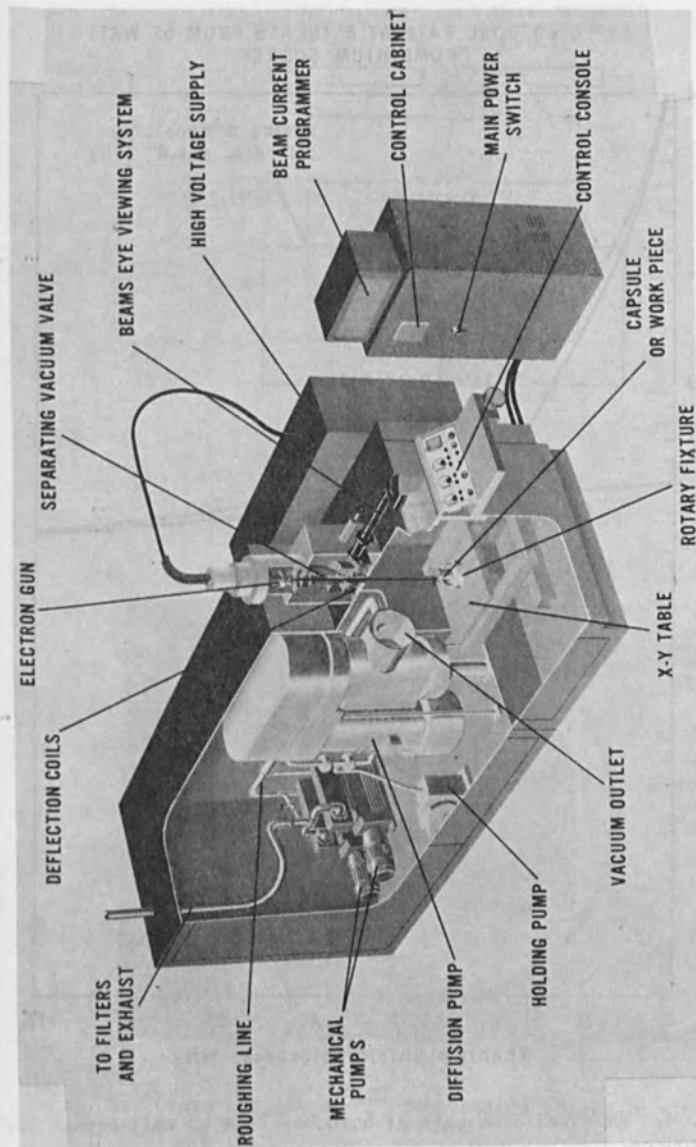


Fig. 3. High density electron beam welder radioisotope encapsulation.

EXPECTED DOSE RATE AT 6 INCHES FROM 65 WATT
PROMETHIUM SOURCE

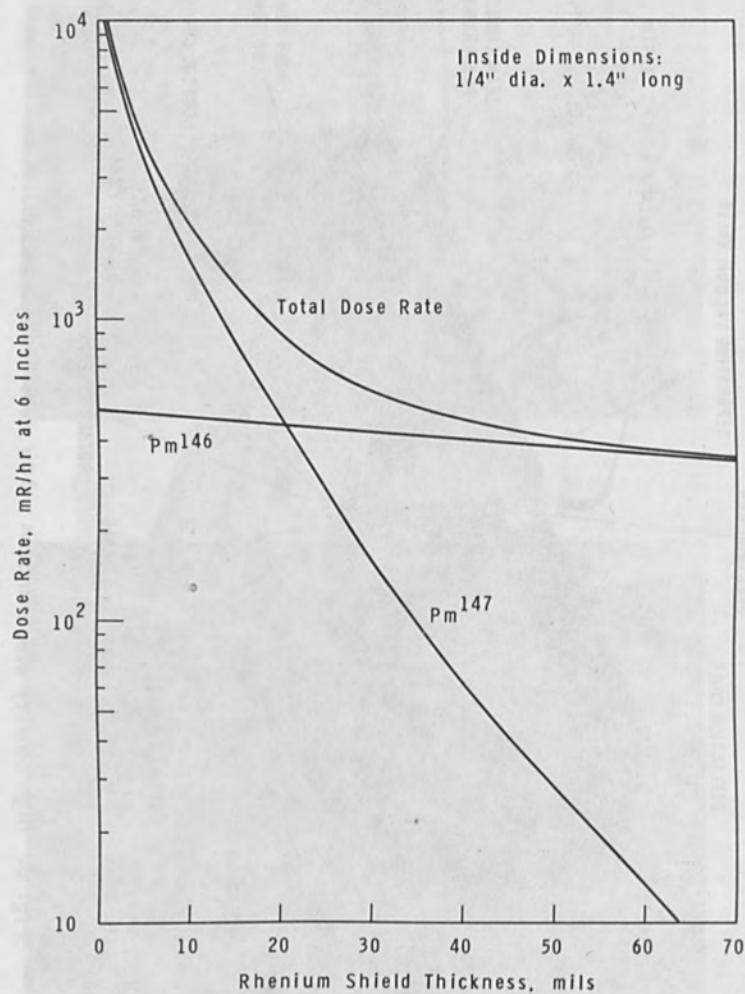


Fig. 4. Expected dose rate at 6 inches from 65 watt promethium source.

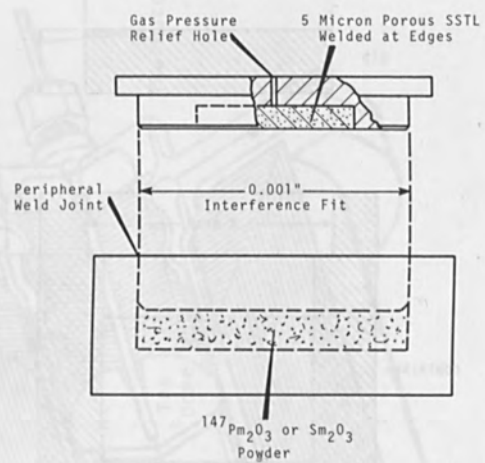


Fig. 5. Pm/Haynes-25 couple design.

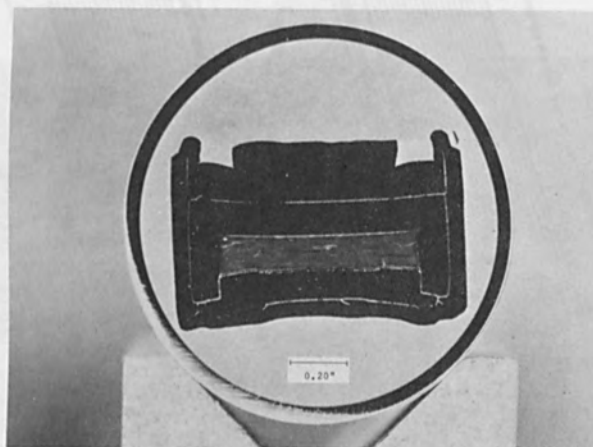


Fig. 6. Cross section of ^{147}Pm -Haynes-25 couple in stainless steel container.

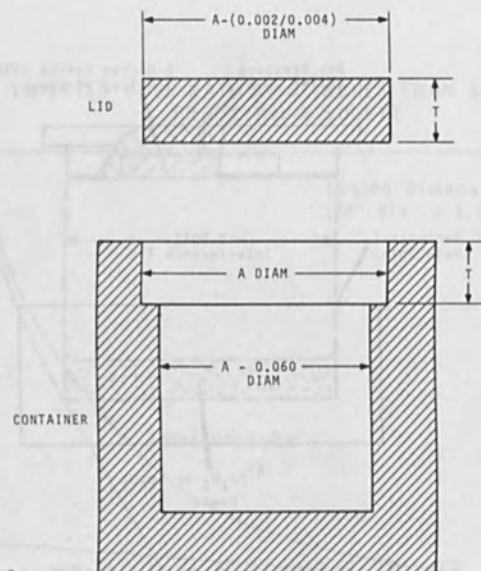


Fig. 7. EB weld joint configuration.

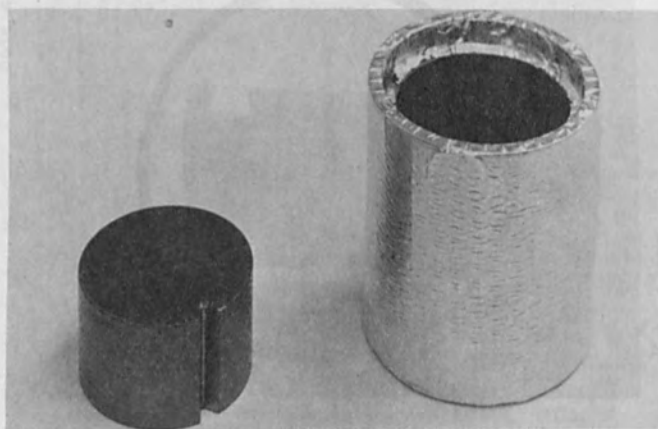


Fig. 8. Foil wrapped capsule and plug spacer.

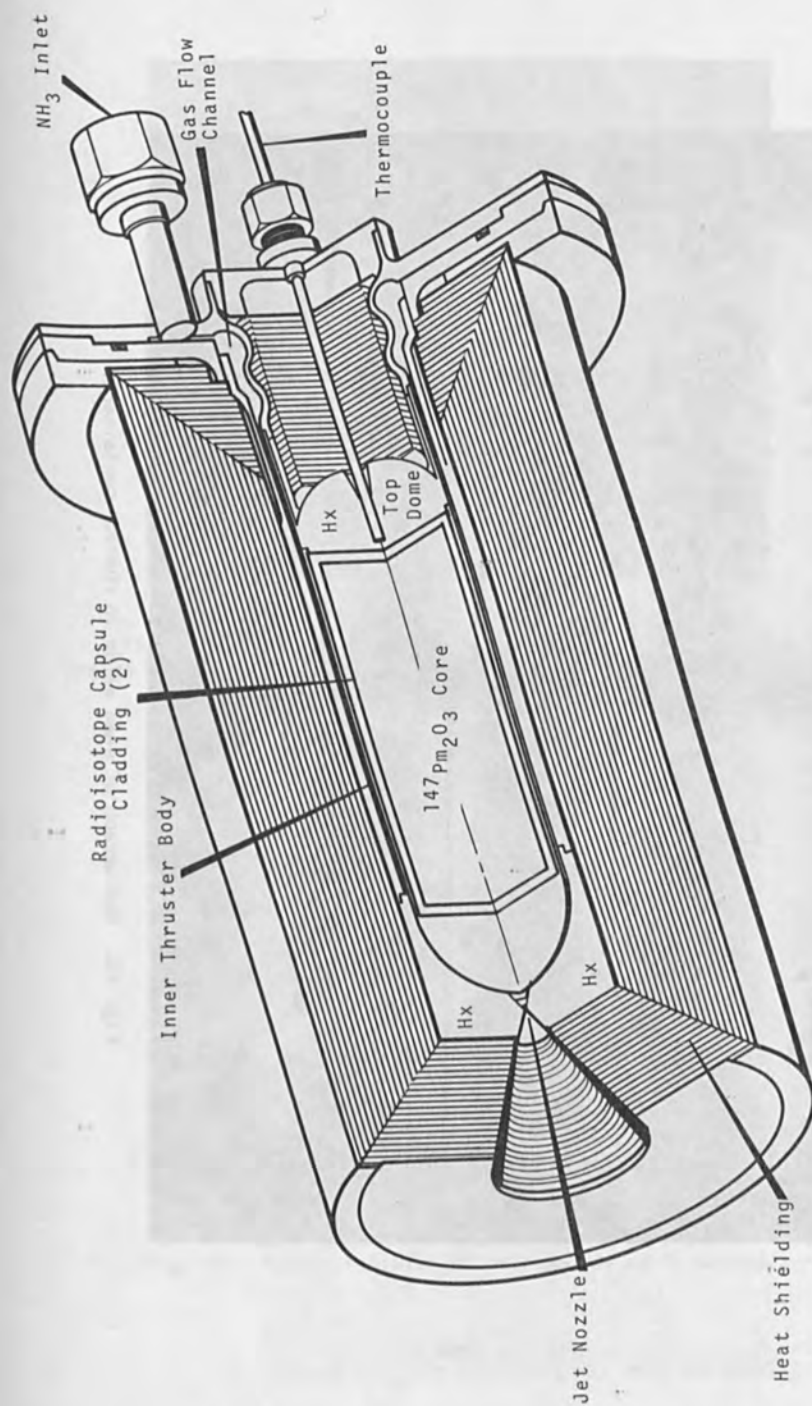


Fig. 9. Radioisotope jet design.

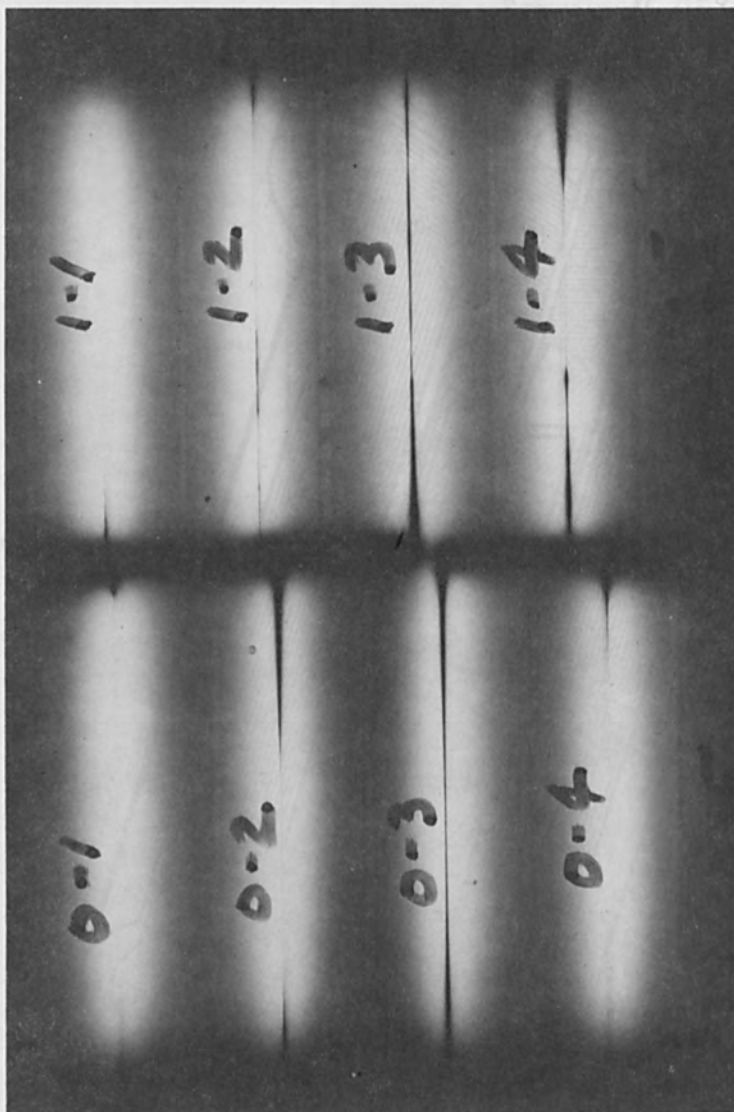


Fig. 10. Radiograph of seam welds in rhenium containers.

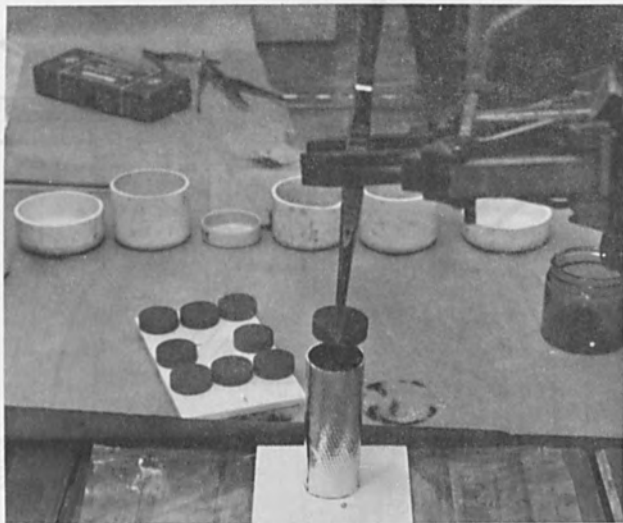


Fig. 11. Loading of $^{147}\text{Pm}_2\text{O}_3$ pellets into full scale cladding inside hot cell.

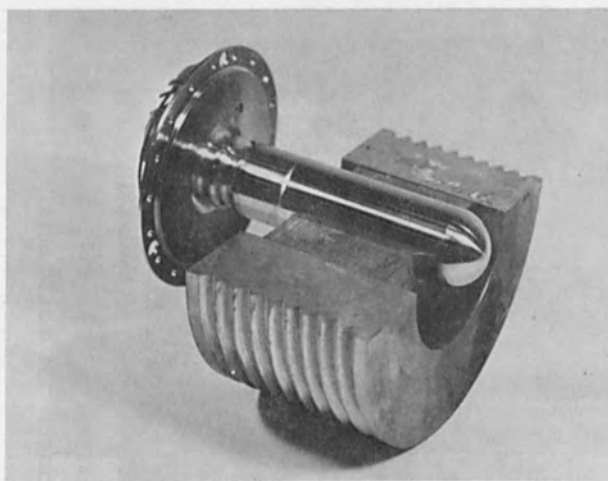


Fig. 12. Actual radioisotope heat source as EB welded.



36X 566-1739B 36X 566-1739A
ETCHANT: 10% OXALIC ACID

Fig. 13. Hx top dome to Hx inner thruster body test weld.

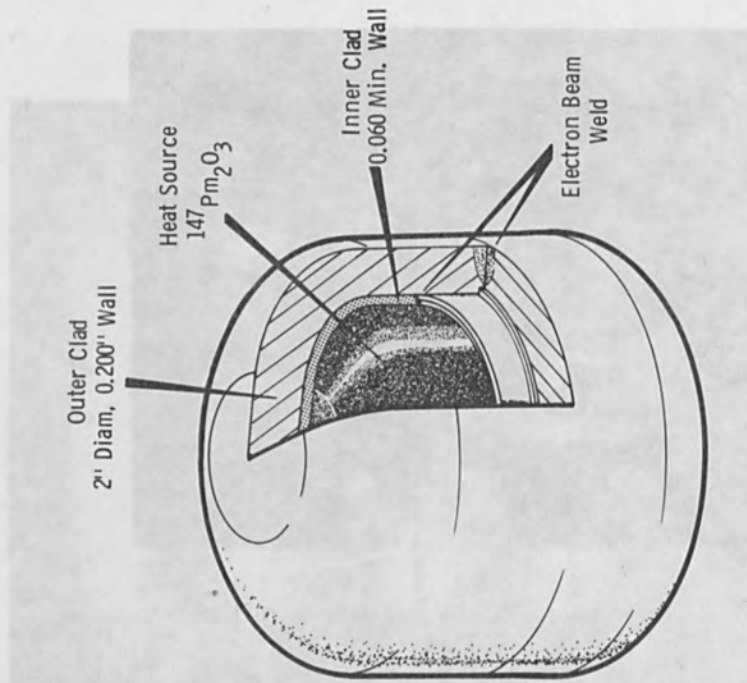


Fig. 14. AMSA Thermal power capsule.

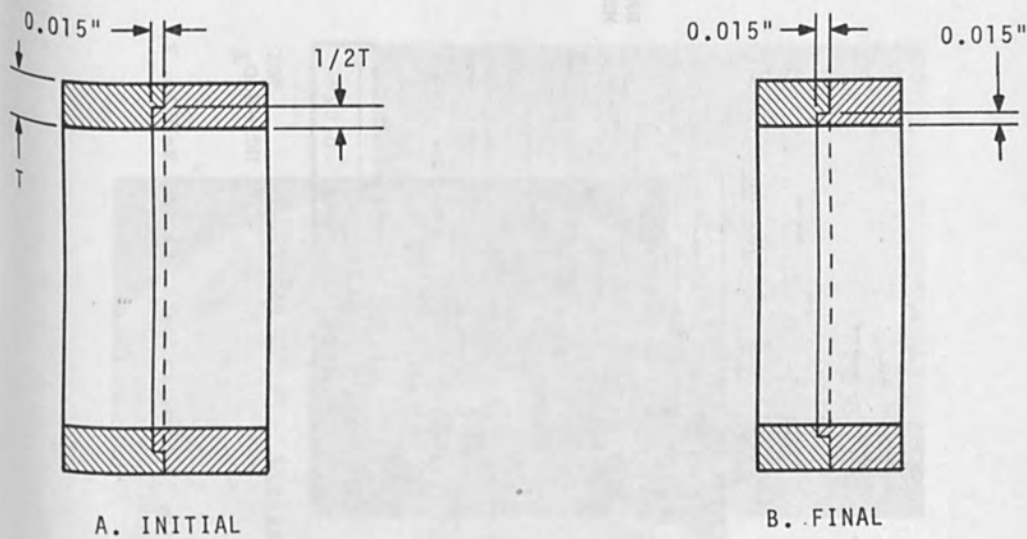


Fig. 15. Design of AMSA cladding weld joints.

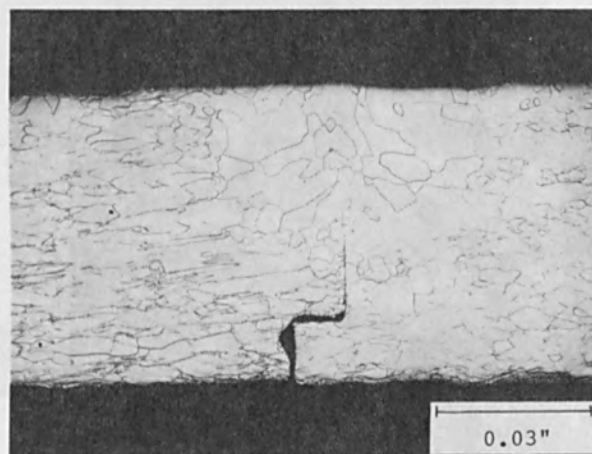
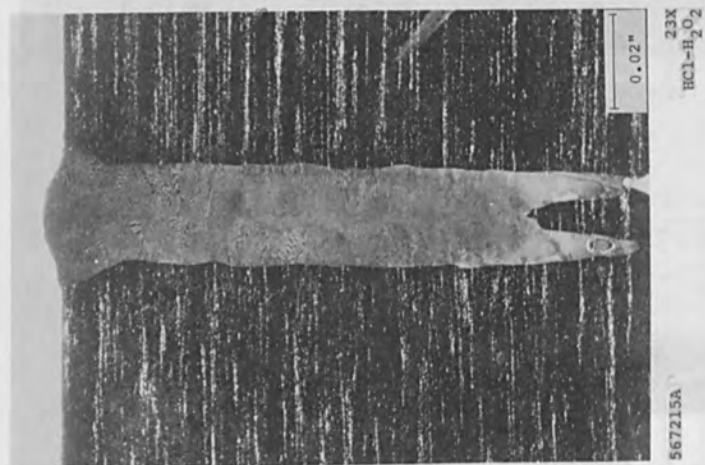


Fig. 16. Electron beam weld in tantalum inner clad 50% joint penetration.



WELD
METAL



BASE
METAL

Fig. 17. Electron beam weld in HS-25, random section, weld No. 2, section C.

Fig. 18. HAZ in HS-25 EB weld. Weld No. 2, section B.



50X

NEG. NO. 567215B

Fig. 19. Root voids and cracks in Co-Cr-Ni EB weld.



Fig. 20. EB weld in L605 phase II secondary cladding showing optimized penetration.

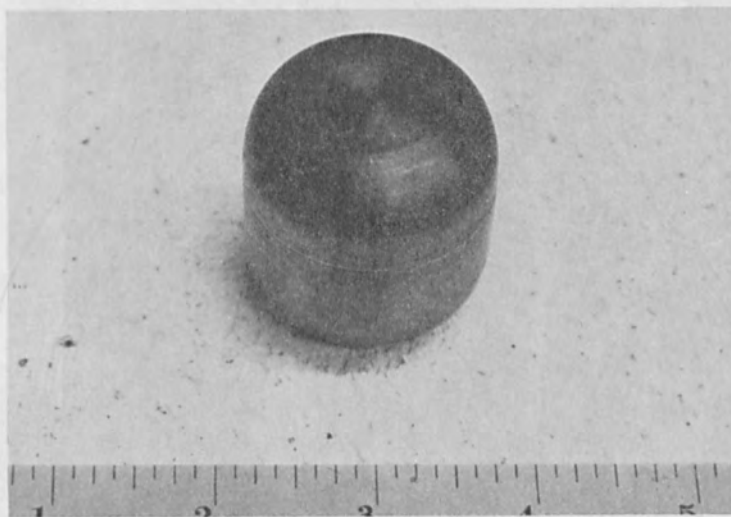


Fig. 21. AMSA primary capsule, as welded.

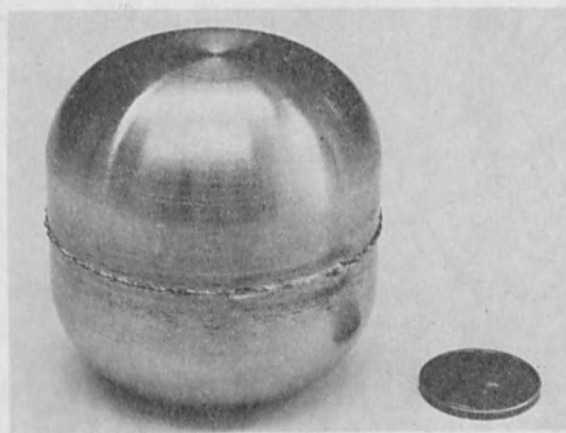


Fig. 22. As EB welded AMSA capsule-Co-Cr-Ni secondary container.

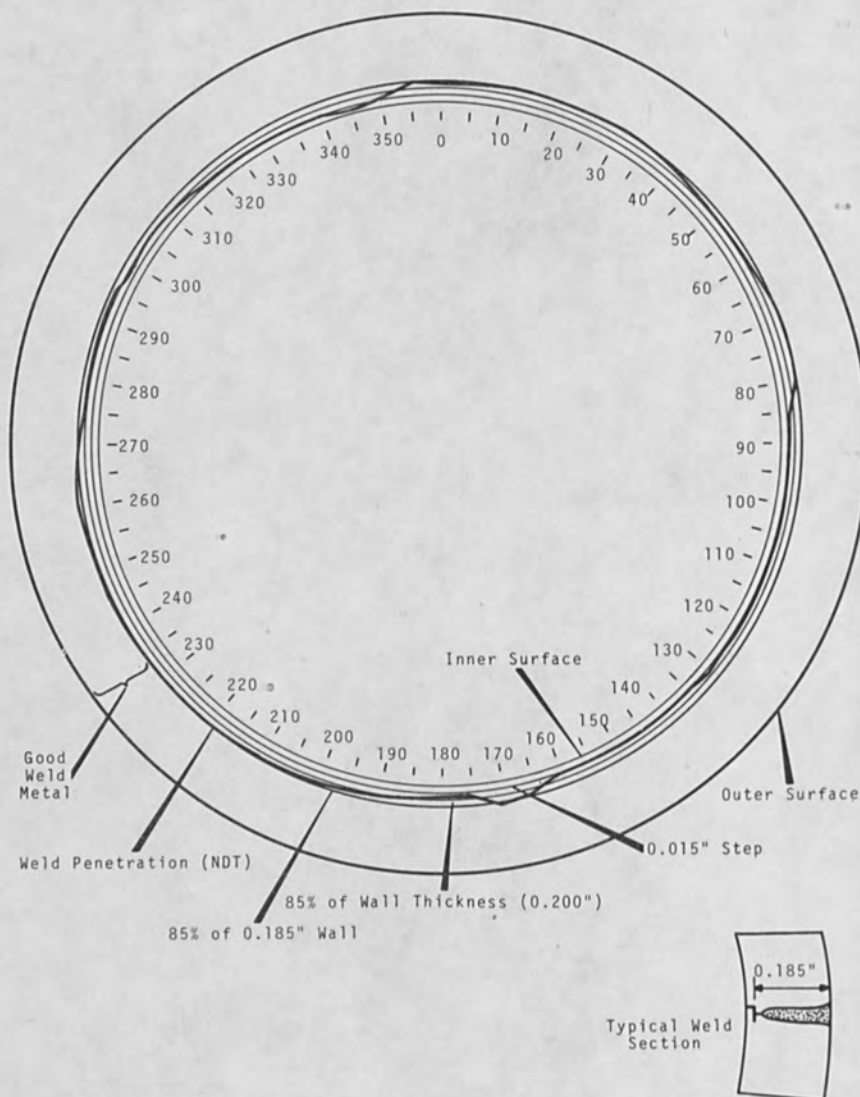


Fig. 23. Ultrasonic test profile of fueled capsule.

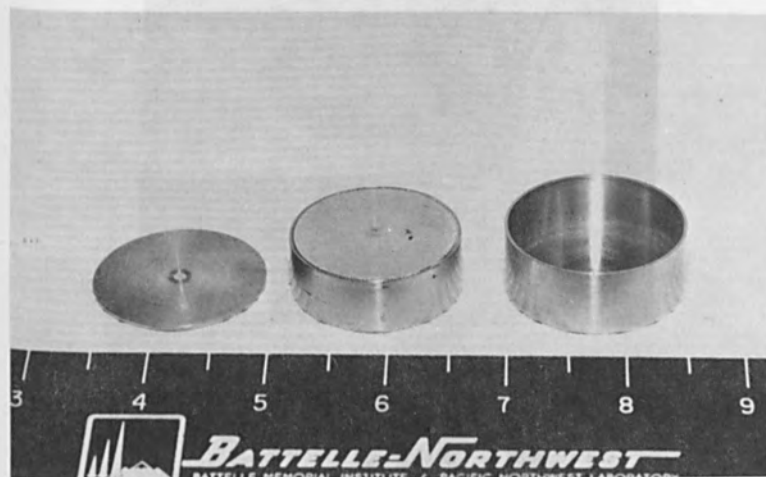


Fig. 24. Primary clad, as-welded promethium-147 source (center) and secondary stainless steel claddings.

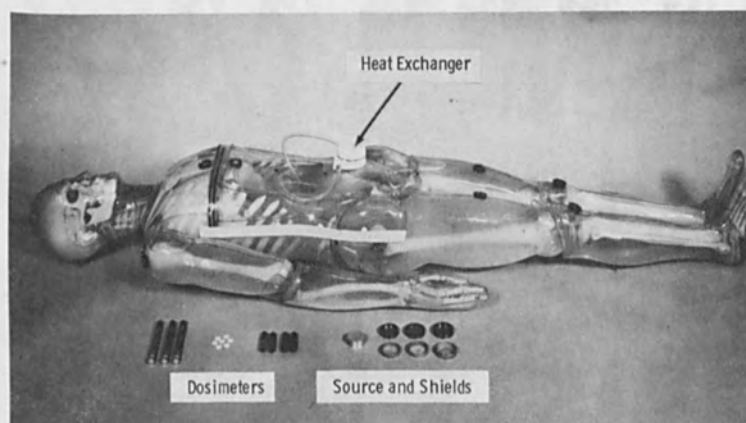
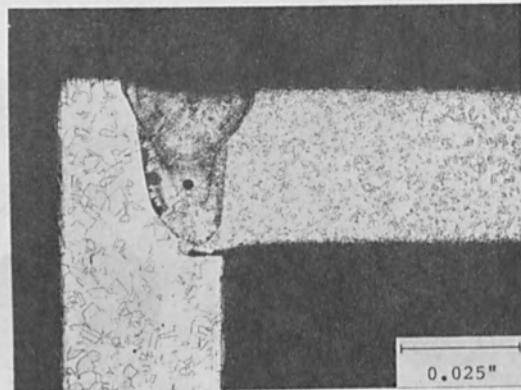


Fig. 25. Remab phantom and ancillary equipment.



4B76

40X

Fig. 26. EB seal weld cross section in 304L stainless steel.

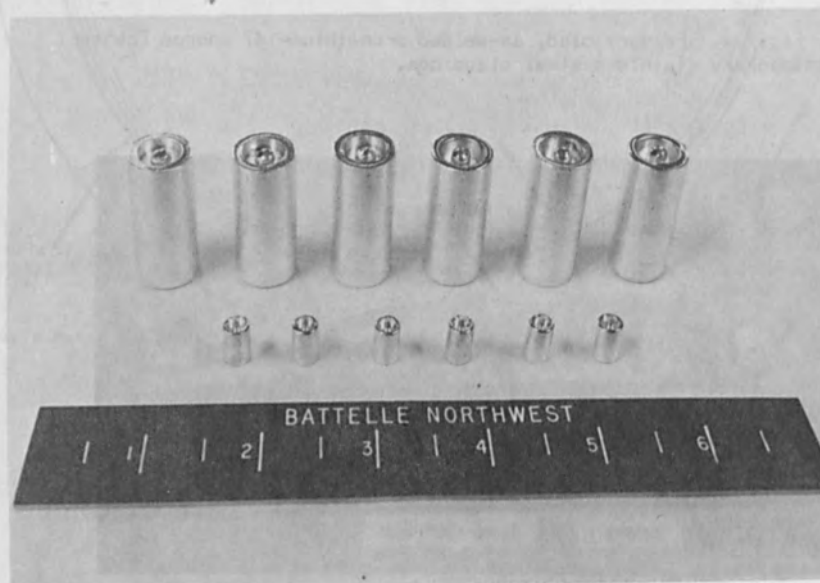


Fig. 27. Aluminum clad promethium or/and SRP capsules-as EB welded.

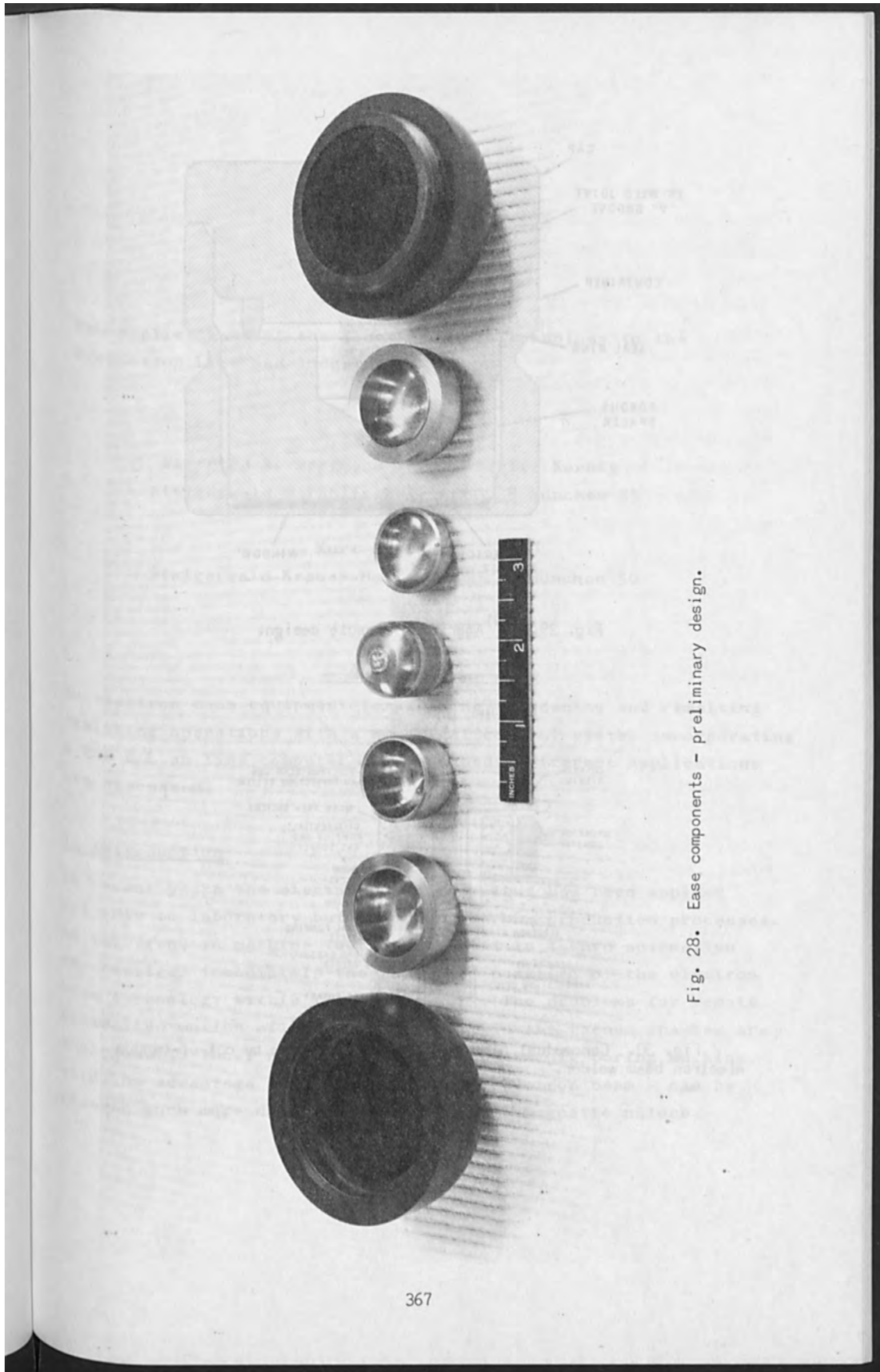


Fig. 28. Ease components - preliminary design.

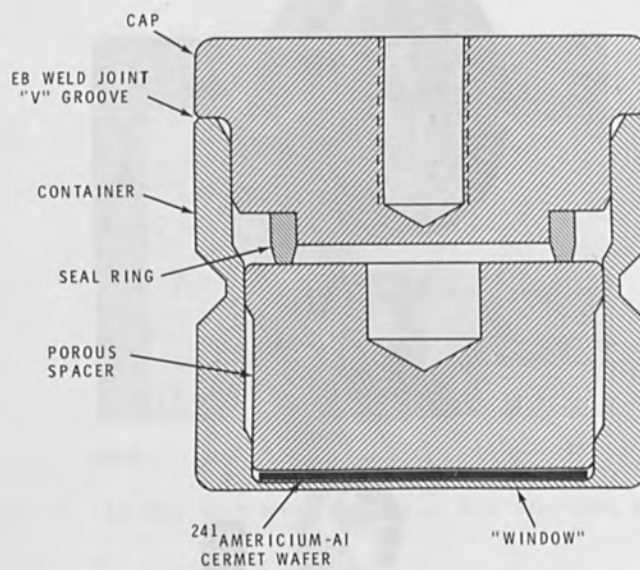


Fig. 29. ²⁴¹Americium capsule design.

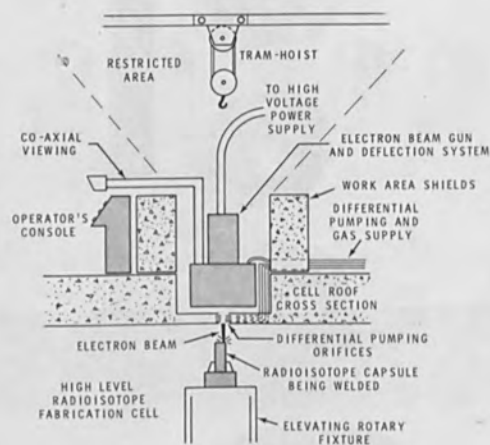


Fig. 30. Conceptual high level capsule closure by out-of-vacuum electron beam welder.

New Applications of the Electron Beam Technology to the Production Line and Industry

Wilfried H. Bergmann and K. Dieter Koenig
Steigerwald Strahltechnik GmbH, 8 München 55
Haderunstr. 1a

Kurt Stoffels
Steigerwald-Krauss-Maffei GmbH, 8 München 50

Abstract:

An electron beam equipment for welding, hardening and remelting finishing operations with a numerical control system incorporating a PDP 8/L on line computer is described. Different applications are discussed.

1. Introduction

In recent years the electron beam technique has been applied not only to laboratory but also to numerous production processes. As the trend in machine tool development is toward automation one realizes immediately the favorable position of the electron beam technology within this frame-work. The problems for remote controlled motion of the workpiece within the vacuum chamber are analogous to those of moving it in a modern jig boring machine, with the advantage that the tool - the electron beam - can be steered much more directly by its electro-magnetic nature.

Therefore the control problem is a twofold one: a conventional electro-mechanical problem of controlling the workpiece in three dimensions and a purely electro-magnetic one, of controlling the beam parameters appropriate to the welding point. Because of this deviation from the similar problem in the numerical control of jig bore machines we developed a special on line control system at Steigerwald Strahltechnik, which makes use of a PDP 8/L on line computer. We followed this approach since we developed not only automatic electron beam welding procedures but also hardening and remelting finishing processes by means of the electron beam.

2. Electron Beam- and Control-System

Figure 1 shows the principal layout of our numerical control system. The particular operating parameters are fed into the PDP 8/L on line computer via a teletype-read-in-punch, which also receives the program in tape form. The signals from the computer are processed through a photo-electric interface and digital-to-analog converters to the individual controllers.

The principal advantage of this layout is: with an appropriate diagnostic instrumentation which determines the actual value of any parameter, a correction can be applied via the computer and the beam steering system for achieving the nominal desired value. Presently this is employed in a track control system. We determine the actual position of the worktable, which moves with an actual accuracy of only about ± 1 mm, in x and y with a precision of $\pm 10 \mu$ and correct for the deviation from the nominal value by appropriate beam deflections. This gives us also the possibility to superimpose any particular track program, which may be necessary for mechanically steering the table. For an illustration we show in figure 2 the brake-down of a welding track of a turbine blade assembly into a sequence of circular and straight line elements.

It is obvious that the above outlined numerical control system steering the inertia-free beam yields the ideal tool for correcting unwanted delays due to the inertia inherent in fast mechanical motions. Thus the computer can for instance adjust the working beam to a constant welding speed in a complicated track pattern with an accuracy of $\pm 0,15\%$ or the welding speed appropriately to variations in the thickness of the workpiece. Such electro-mechanical and photo-electric seam-followers are being developed.

3. Applications

A simplified version of such a track correcting system could possibly supplement a production welding machine for assembly of precision gears as demonstrated in figure 3 and 4.

Another production example for electron beam welding is the fabrication of sawblade bimetal strips, which is demonstrated in the next 4 figures (fig. 5-8).

A novel application of the electron beam technique is the thermal surface treatment. The major advantages are that one heats in a very short time only a very small surface volume of the workpiece to a precisely predeterminate temperature so quickly that the surroundings remain practically at room-temperature. Therefore self-quenching follows immediately. Quenching speeds in the range of many thousand degrees Kelvin per second have been achieved. This process can be applied in two modifications:

- First for local precision hardening, in which the workpiece is given the final shape in the "soft condition". Optimal hardness values are only introduced into the working surfaces and edges. Therefore very close tolerances can be maintained. As an example we show electron beam hardened saw-tooth edges in the next figure 9. Figure 10 demonstrates a film-die, which was machined to final dimensions before electron beam hardening of only the working edges around the holes was performed. Figure 11 depicts a ball bearing surface electron beam treated

to a hardness of 1050 HV₁ after final machining. The interesting metallurgical effect to be achieved by this method of surface treatment are further illustrated by the following figure 12 which shows in a micro cross section of a spheroidal graphite iron in which one observes the transformation of the graphite into solid solution.

In the second modification the workpiece surface is heated to melting temperatures. In this process of remelting finishing special phase transformations in alloys can be achieved which are exemplified in the following pictures. Figure 13 shows a typical micro cross section through a treated surface of a high-temperature steel while figure 14 exhibits the same on an aluminium alloy. Characteristic improvements by this remelting finishing process are demonstrated in the last figure 15, in which the shaft of a valve stem has been treated by remelting finishing to improve its properties under dynamic loading.

The above short description demonstrates that a numerical control system with an on line computer is essential in applying the electron beam technology to welding as well as hardening and surface remelting finishing in commercial tooling processes.

Organisation of EBN 10/4 Computer Control

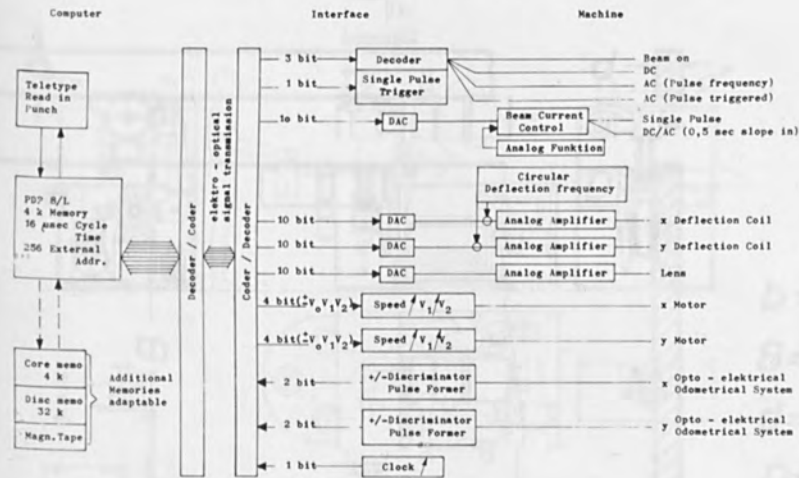


Fig. 1. Organization of computer control.

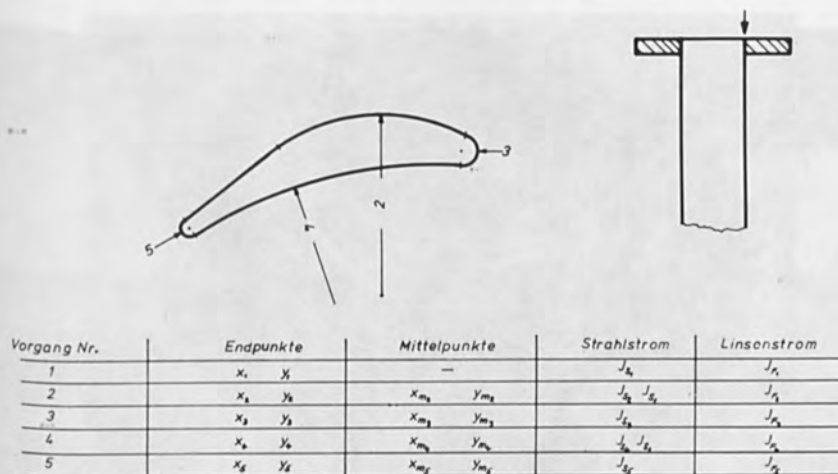


Fig. 2. Coordinate table for NC-controlled electron beam weld of a turbine blade assembly.

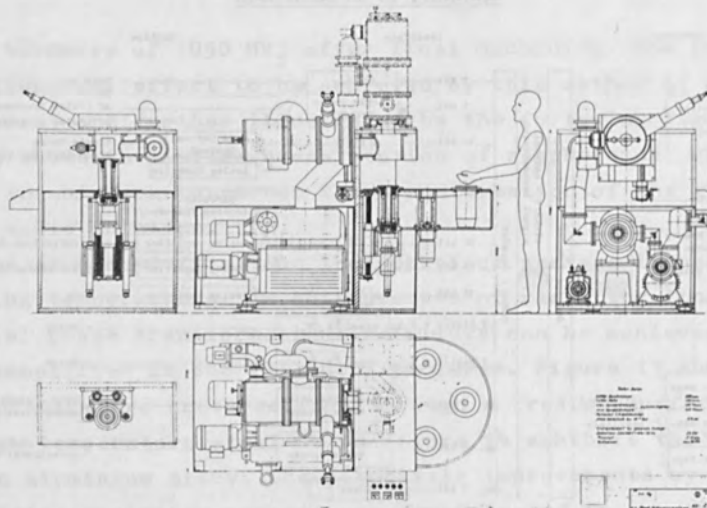


Fig. 3. Production welding machine P3.

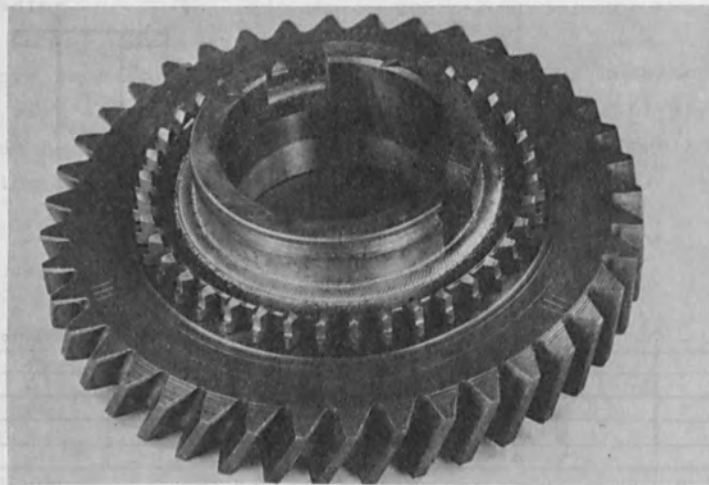


Fig. 4. Precision welded gear assembly.

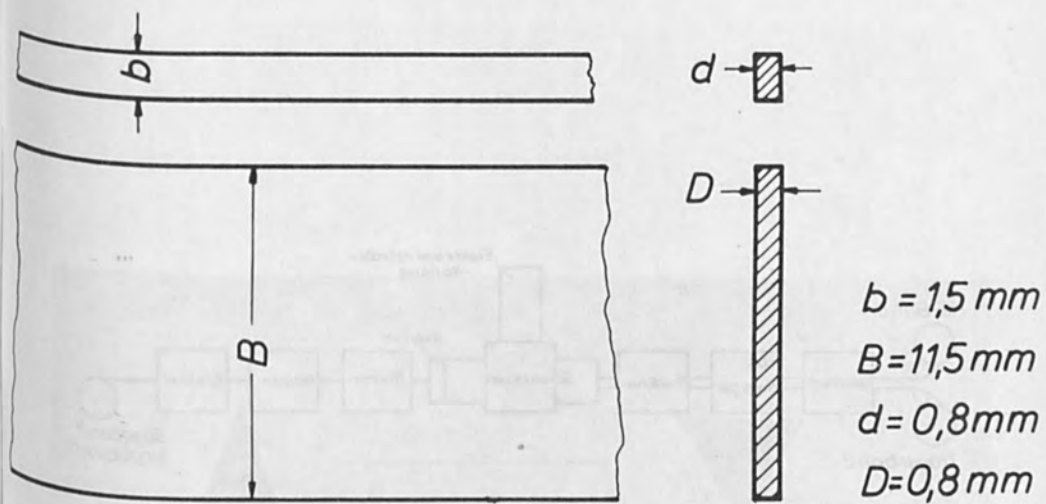


Fig. 5. Dimensions of the bi-metal saw blade assembly.

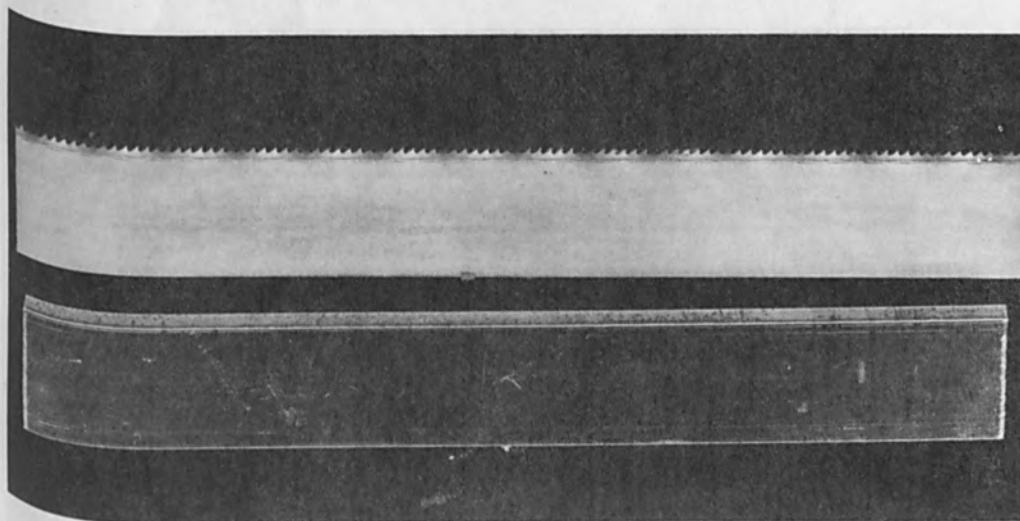


Fig. 6. Bi-metal saw blade assembly.

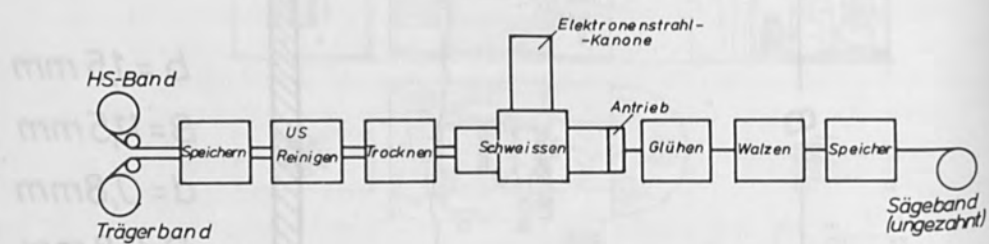


Fig. 7. Scheme of a continuous bi-metal ribbon electron beam welding machine.



Fig. 8. Continuous bi-metal ribbon electron beam welding machine.

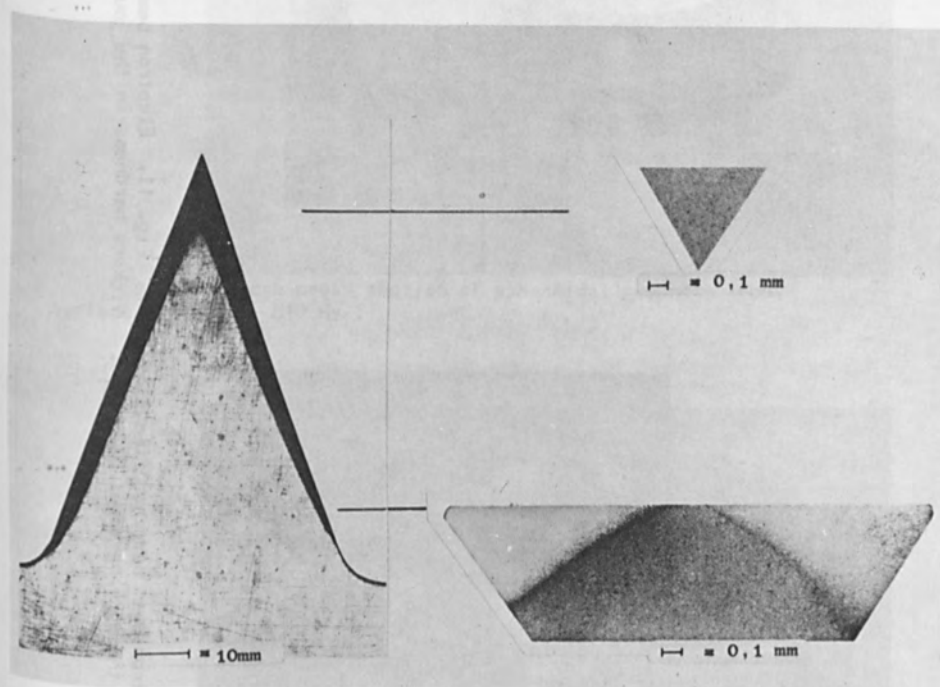


Fig. 9. Electron beam hardened saw-tooth edges. Vickers hardness in the working edge: 950-1000 HV₁.

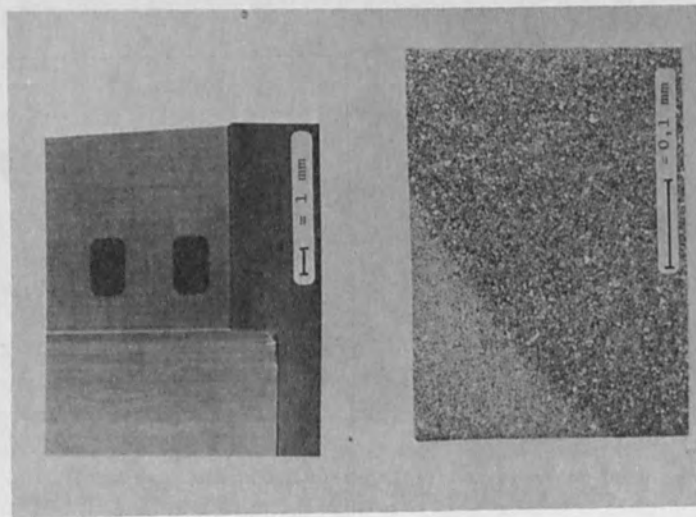


Fig. 10. Electron beam hardened film-die.
Vickers hardness in the working edge: 820 HV_{0,3}.

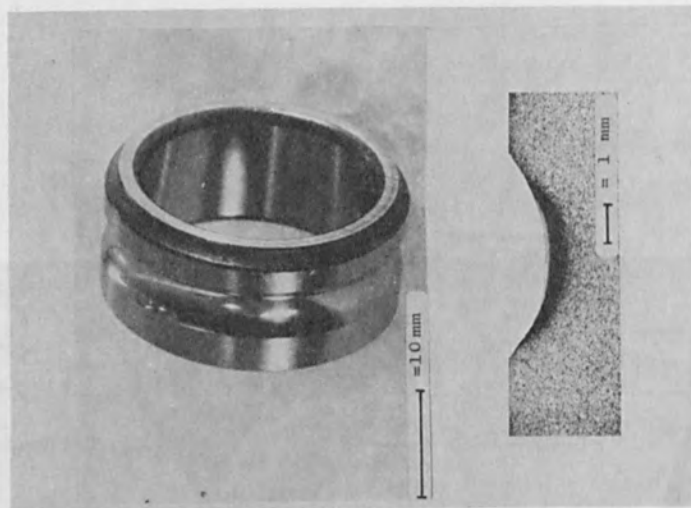


Fig. 11. Electron beam hardened ball bearing.
Vickers hardness in the journal: 1050 HV₁.

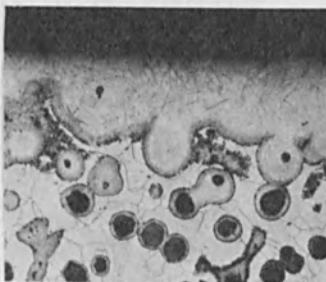


Fig. 12. Micro cross section of spheroidal graphite iron.
Surface hardness: 819 HV₁.

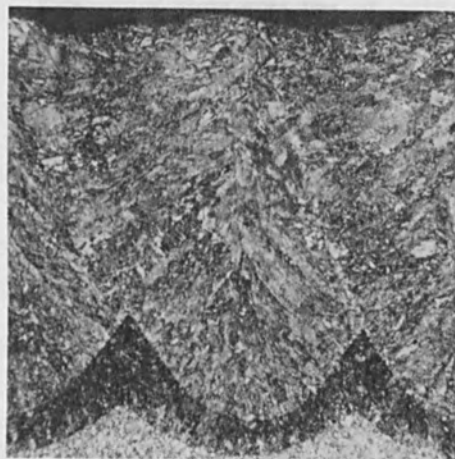


Fig. 13. Micro cross section of remelted surface of 10 CrMo
910 high temperature alloy. Surface hardness: 400 HV₁.

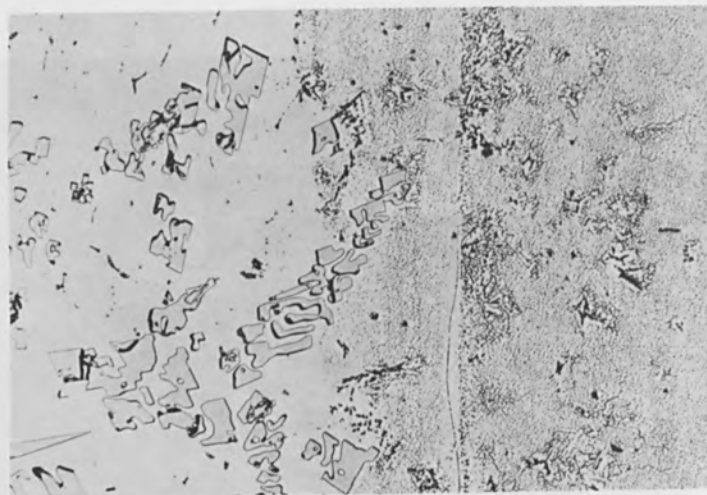


Fig. 14. Micro cross section of remelted surface of Al-6Mn.
Surface hardness: 75 HV_{0,1}.

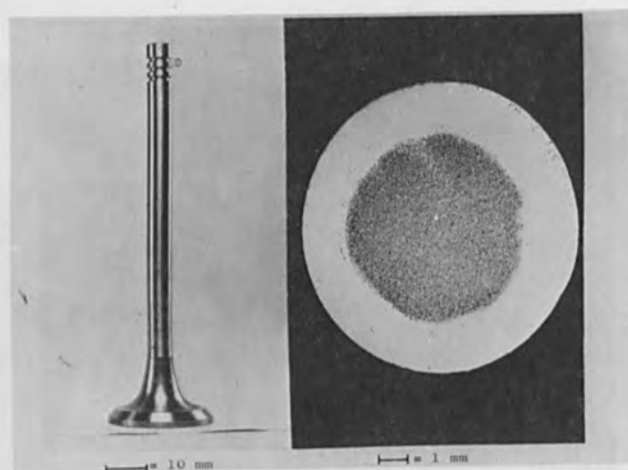


Fig. 15. Remelting finished valve stem in X50CrMnNi229.

New Possibilities of EB-Micromachining

Karl-Heinz Steigerwald, Wilfried H. Bergmann
Karl-Heinrich v. Grote, Wilhelm Scheffels, Alfred v. Walter
Steigerwald Strahltechnik GmbH, 8 München 55
Haderunstr. 1a

W. Edgar Meyer
Steigerwald-Krauss-Maffei GmbH, 8 München 50

Abstract:

An electron beam equipment numerically controlled by a PDP 8/L on line computer für micro-machining is described. The state-of-the-art of its machining capabilities for joining, perforation, cutting and sublimation is discussed.

1. Introduction

Today's situation in precision machining is characterized by the advance of new technologies which try to enter the market directly from the worktable of the scientist and which demand not only higher precision but also smaller and smaller dimensions. Two characteristic representatives of this trend of development are superconductivity and the ultra high frequency and computer technology pushing toward 10 GHz. The full exploitation of the physics implications inherent in both of these applications demands a precision machine tooling capable of manufacturing reliable structures with the characteristic dimension of at least 0.1μ , below the limit of the optical resolution. A number of rather new technologies are competing for this most challenging tooling

problem, mainly ECM and EDM (electro discharge and electro chemical machining) sputter etching the laser and the electron-ion beam technology. The three first processed can handle dimensions down to about 5μ and have entered the field of precision machining of high temperature alloys and micro integrated circuits.¹ YAG-Laser working beams with a wave-length of 1.06μ have been successfully used in the laboratory for machining structures with dimensions equivalent to their optical resolution of about 1μ .² Up to now only the electron beam technique in conjunction with a modification of the photo-resist technique, the electron-resist technique, has been capable of achieving structures of 0.2μ dimension in the laboratory.³ This was possible because the electron beam has only a wave length of a few Angström. This demonstrates that in principal the particle beam technology is the only one which can meet the demands in dimension and precision of the new coming fields of super-conductivity and ultrahigh frequency.

2. Electron Beam- and Control-System

For the above outlined aim three major machining capabilities are necessary:

1. joining, perforating and cutting of thin foils
2. engraving of films
3. refining (for instance ion-beam-implantation, sputtering)

This demands for the first group that we have to improve present days welding, perforation and cutting techniques. Engraving can only be accomplished by sublimation and refining implies in many cases beam implantation, sputtering or other techniques.

At Steigerwald Strahltechnik we developed as a first step toward goal one an electron beam equipment, the EBM 10/4⁴ with on line computer control depicted in figure 1 and 2 which incorporates simultaneously the technical capabilities of a laboratory machine and a pilot plant for production purposes. Figure 3 shows the principal layout of our numerical control system

The particular operating parameters are fed into the PDP 8/L on line computer via a teletype-read-in-punch, which also receives the program in tape form. The signals from the computer are processed through a photo-electric interface and digital-to-analog converters to the individual controllers.

The principal advantage of this layout is: with an appropriate diagnostic instrumentation which determines the actual value of any parameter, a correction can be applied via the computer and the beam steering system for achieving the nominal desired value. Presently this is employed in a track control system. We determine the actual position of the worktable, which moves with an actual accuracy of only about ± 1 mm, in x and y with a precision of $\pm 10 \mu$ and correct for the deviation from the nominal value by appropriate beam deflections. This gives us also the possibility to superimpose any particular track program, which may be necessary for mechanically steering the table.

It is obvious that the above outlined numerical control system steering the inertia-free beam yields the ideal tool for correcting unwanted delays due to the inertia inherent in fast mechanical motions. Thus the computer can for instance adjust the working beam to a constant working speed in a complicated track pattern with an accuracy of ± 0.15 % or the working speed appropriately to variations in the thickness of the workpiece.

3. State-of-the-Art

In the following figures we would like to document the capabilities of our EBM 10/4 for welding, perforating and cutting. The large range of precision joining possibilities should be indicated by figure 4 which shows a tungsten disk welded into a molybdenum cylinder, figure 5 demonstrates a diffusion bond between graphite and molybdenum, figure 6 shows a micro-cross-section of the first weld while figure 7 demonstrates a wiggle-weld on a titanium-aluminium alloy structure used in supersonic aircraft construction.

Another precision welding and perforation experiment is demonstrated in the next figure 8 which shows the profile of an electron beam welded turbine blade assembly with perforated casing.

A further application for joining is exemplified in figure 9 which shows a solder joint performed on an integrated circuit.

The perforation possibilities are demonstrated in figure 10 showing a cross section through a Inconel 600-foil of 1,5 mm thickness perforated with holes of $500 \pm 10 \mu$ diameter. Figure 11 demonstrates a tantalum foil of 50μ thickness perforated with holes of 35μ diameter. The next figure 12 shows 100μ holes in a 100μ thick nickel foil. Figure 13 shows holes of 4μ diameter in a 12μ Hostaphan foil and finally figure 14 shows partially perforated artificial leather with holes of 170μ diameter.

Cutting examples are shown in figure 15, demonstrating a 20μ slit milled into a 0.2 mm stainless steel foil, and in figure 16 which shows a cross shape slit milled into a 0.5 mm thick teflon-foil. The size of the cross is 1 mm.

Engraving examples are demonstrated in the next figure 17 in which 5μ thick copperfilm is engraved into a meander-like shape and in figure 18 showing a tantalum-film of 0.5μ thickness divided by 2μ wide slits.

Engraving can best be achieved by sublimation. For this purpose a special electron beam-equipment demonstrated in figure 19 is needed. The schematic cross section is depicted in the next figure 20.

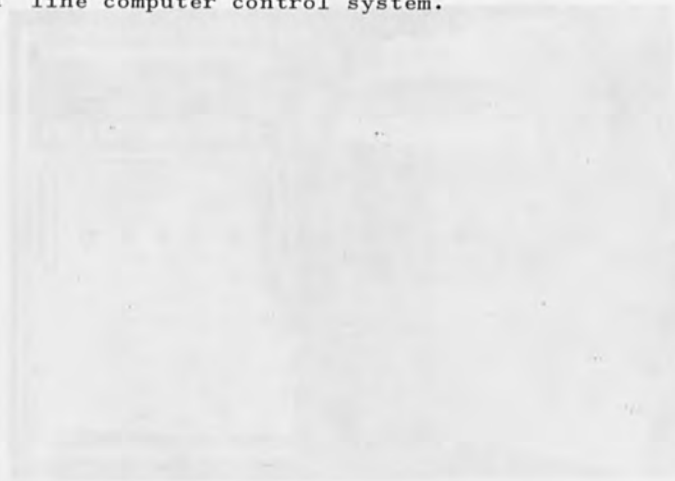
40 kV electrons are applied for a very short time with high power density to the sample. By these means direct sublimation is accomplished without any material becoming liquid. Typical sublimation data are shown in the next slides, figure 21 and 22. Recently we have applied this technique to commercial Nb_3Sn superconductors for subdividing the superconductor into small filaments for intrinsic stabilization. We achieved in the 6μ thick

superconducting film channels from $10 \times 3 \mu^2$ to $20 \times 8 \mu^2$ dimensions, fig. 23.

The same effect of subdividing the superconducting layer was achieved previously by forming recast tracks in the superconducting layer. During melting the stoichiometric ratio was changed considerably (which was analysed by an electron micro probe), destroying the superconducting properties in the recast tracks.

A cross section through such a track is shown in figure 24 while figure 25 shows a number of 30μ wide parallel filaments, spaced about 50μ apart.⁵

Even though the demonstrated examples fall short of the initially outlined aim they demonstrate the potential capabilities of the described technologies. Further developments towards higher resolution are in progress. At the present time we still rely on visual observation. For further diagnoses of the achieved results for instance electron and ion microscopes, scanning microscopes and micro-probes are available and could be adapted in the future to our on line computer control system.



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5. These experiments were in part supported by C.H. Rosner, Superconductive Products, General Electric Co., Schenectady, N.Y. and by J.C. Laurence, NASA LEWIS Research Center, Cleveland, Ohio.

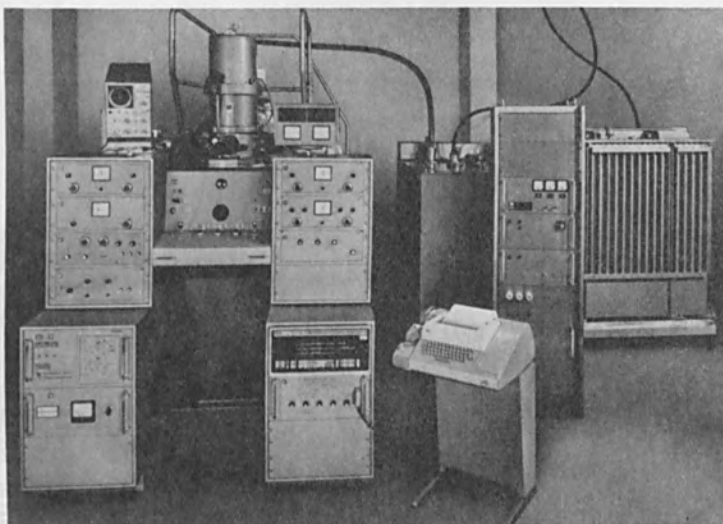


Fig. 1. Electron beam micromachining equipment EBM 10/4.

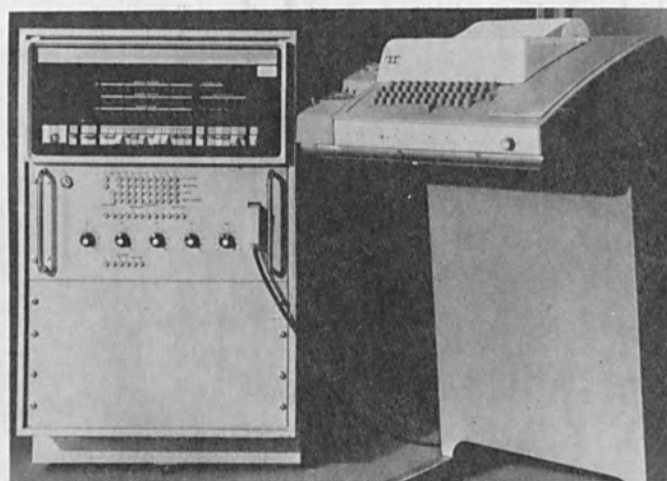


Fig. 2. NC-equipment of the EBM 10/4.

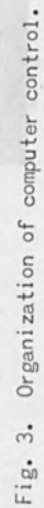


Fig. 3. Organization of computer control.

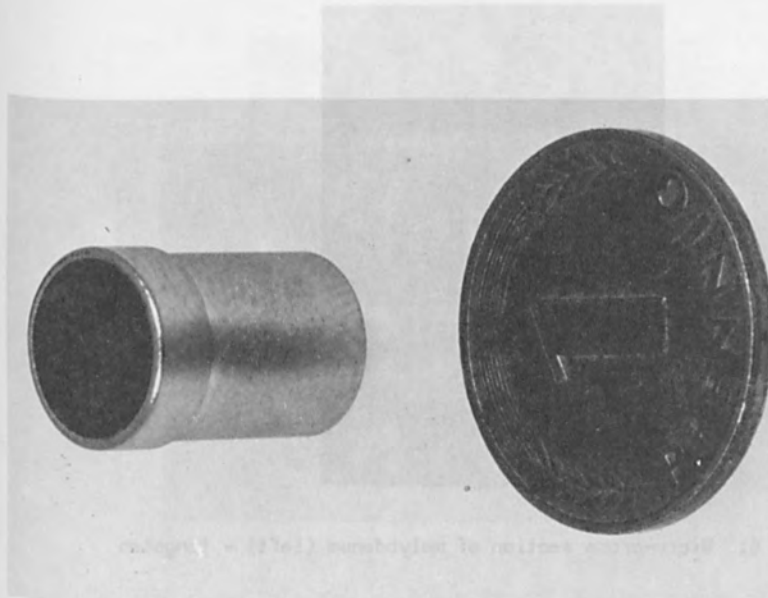


Fig. 5. Graphite disk welded into a molybdenum cylinder.

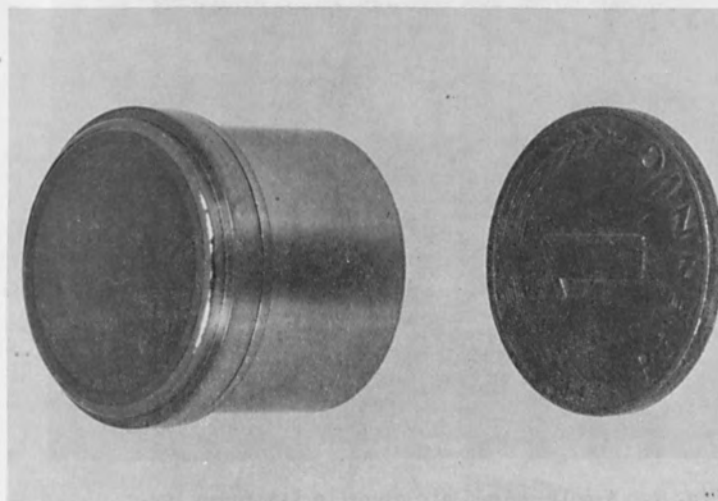


Fig. 4. Tungsten disk welded into a molybdenum cylinder.

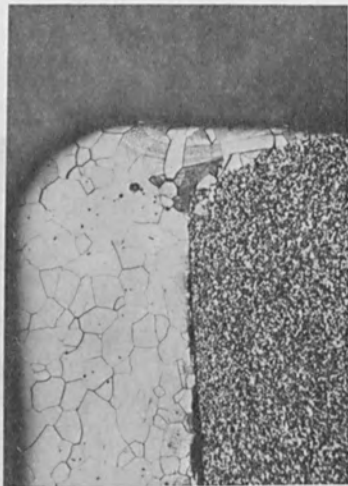


Fig. 6. Micro-cross section of molybdenum (left) - tungsten weld.

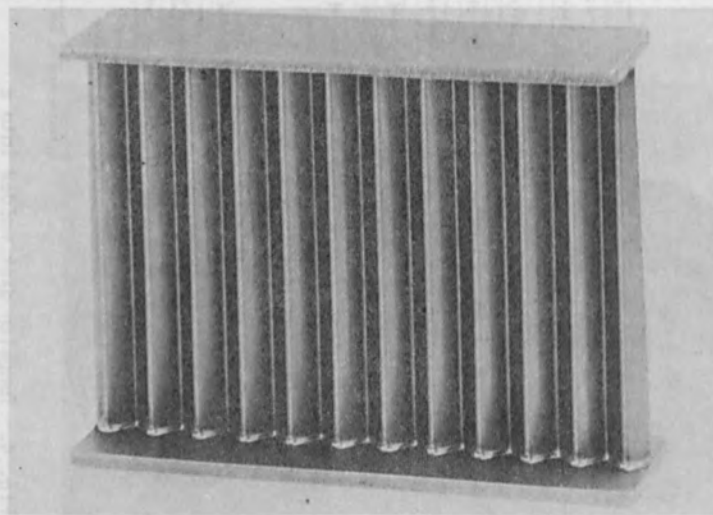


Fig. 7. Wiggle-welded structure in TiAlV64-alloy.

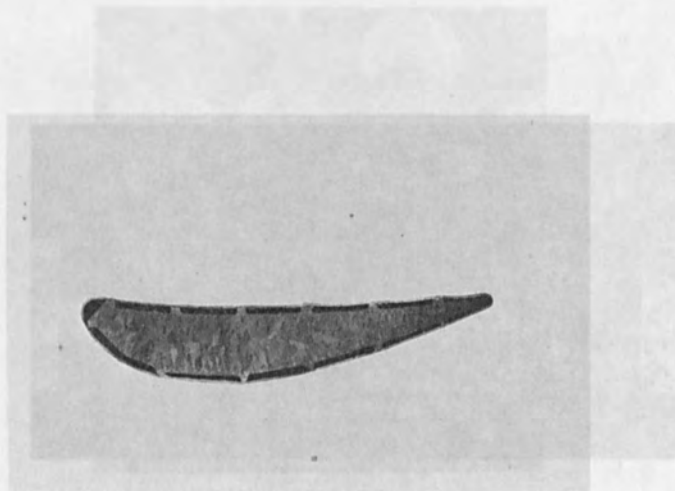


Fig. 8. EB-welded and perforated turbine blade assembly.

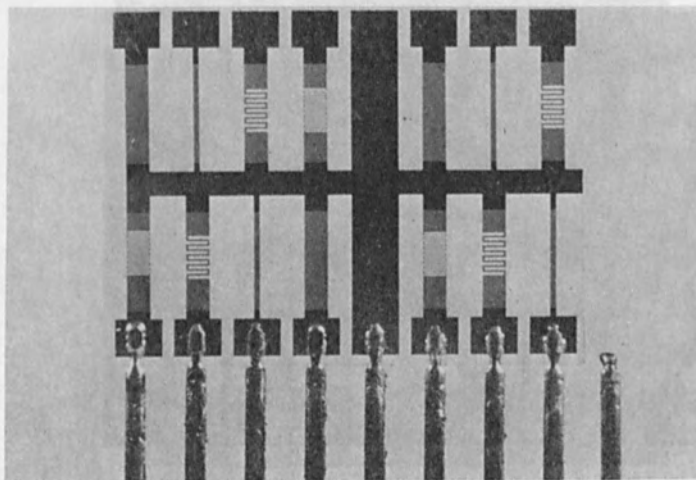


Fig. 9. EB-soldered contacts on an integrated circuit.

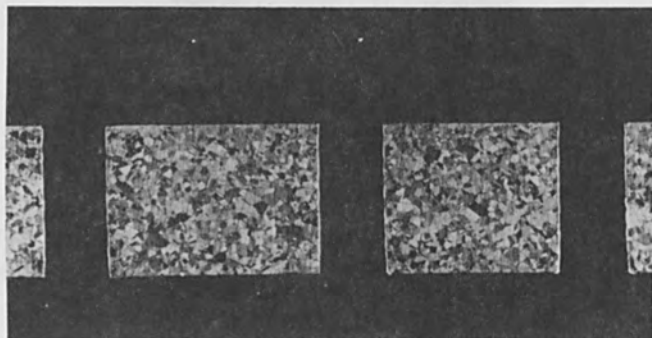


Fig. 10. Cross section through a 1,5 mm thick Inconel 600-foil with holes of $500 \pm 10 \mu$ diameter.

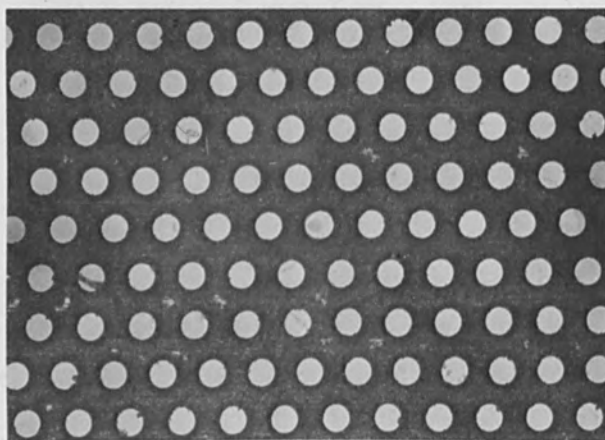


Fig. 11. 50μ thick Ta-foil with holes of 35μ diameter.

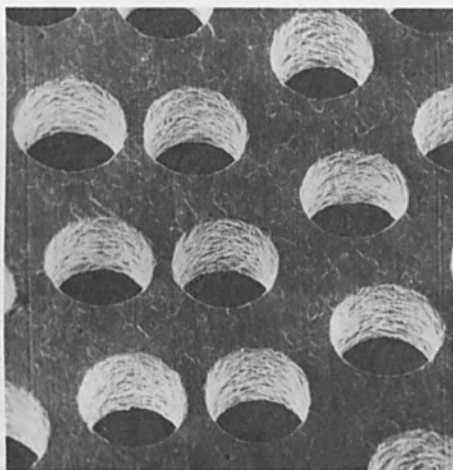


Fig. 12. 100 μ thick Ni-foil with holes of 100 μ diameter.

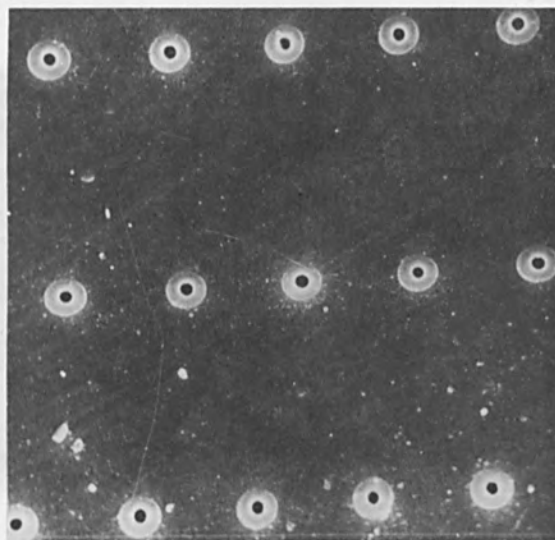


Fig. 13. 12 μ thick Hostaphan-foil with holes of 4 μ diameter.



Fig. 14. Partially perforated artificial leather with holes of $170\ \mu$ diameter.

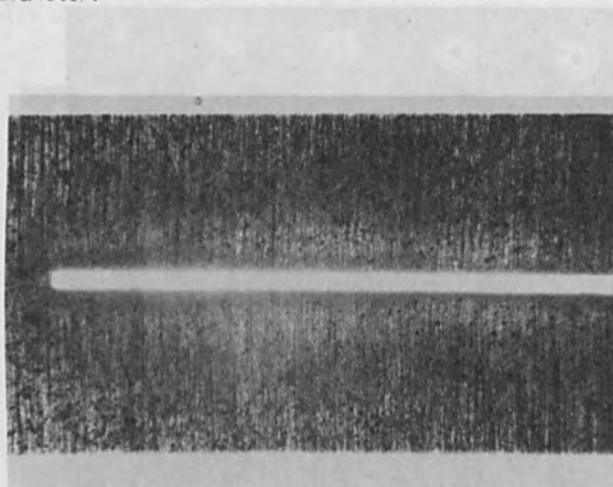


Fig. 15. $20\ \mu$ wide slit milled into $200\ \mu$ thick stainless steel foil.

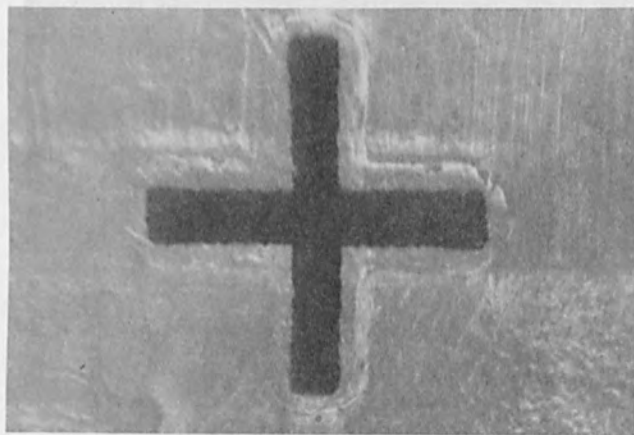


Fig. 16. 1 mm large cross milled into 0.5 mm thick Teflon-foil.

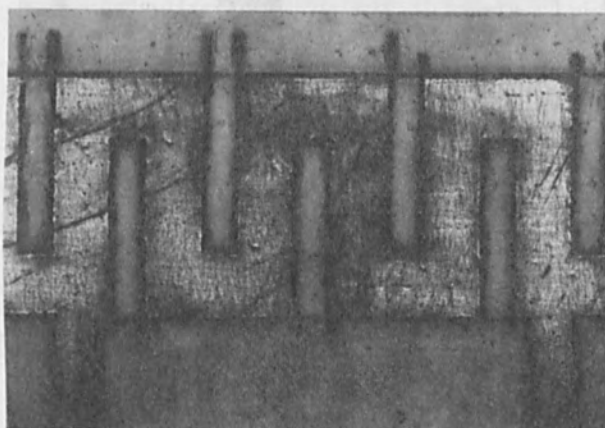


Fig. 17. $100\ \mu$ wide Meander-resistor EB-engraved into $5\ \mu$ copper.

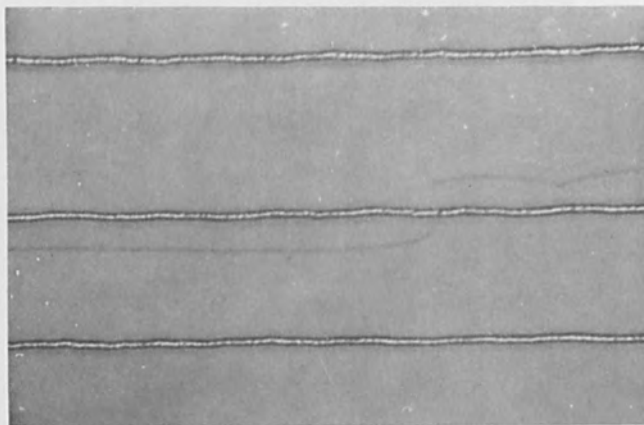


Fig. 18. 2μ wide EB-engraved slits into 0.5μ tantalum.

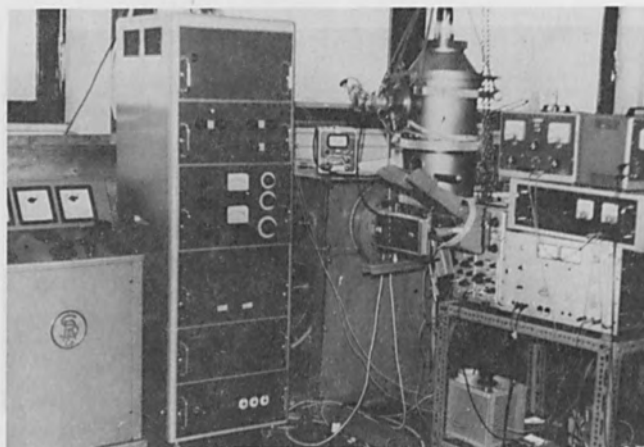


Fig. 19. Experimental equipment for sublimation.

Elektron Beam Parameters for Sublimation

High Voltage	40 kV
Beam Current	20 mA
Focus Diameter	20-30 μ m
Current Density	3000 A/cm ²
Power Density	$1,2 \times 10^8$ W/cm ²
Focus Deflection Velocity	10^4 cm/sec

Fig. 21. EB-parameters for sublimation.

Evaporation Rates for Sublimation

(40 kV, 20 mA, 10^8 W/cm², 10^4 cm/sec)

Tungsten	0,2 g/sec
Magnesium	0,6 g/sec
Graphite	0,6 g/sec
18/8 Cr-Ni (AISI 303)	0,75g/sec
Brass	1,1 g/sec

Fig. 22. EB sublimation rates.

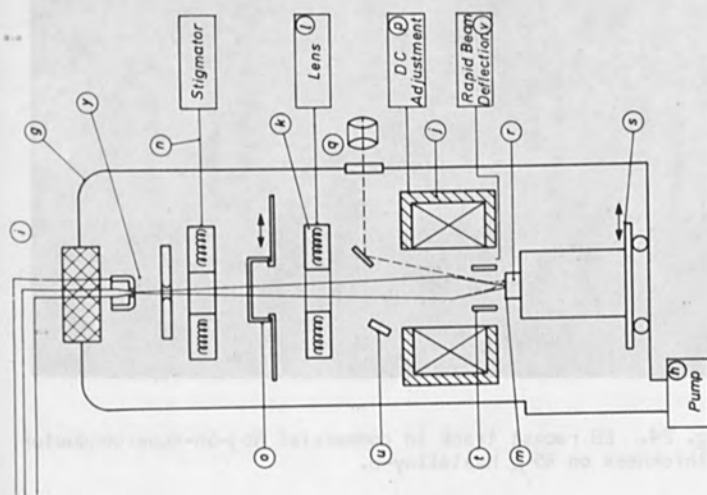


Fig. 20. Schematic of sublimation
EB-equipment.

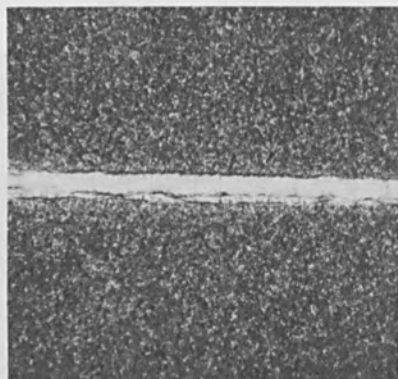


Fig. 23. EB sublimation engraved slit in commercial Nb₃Sn-superconductor 6 μ thick, about 20x6 μ^2 slit cross section.

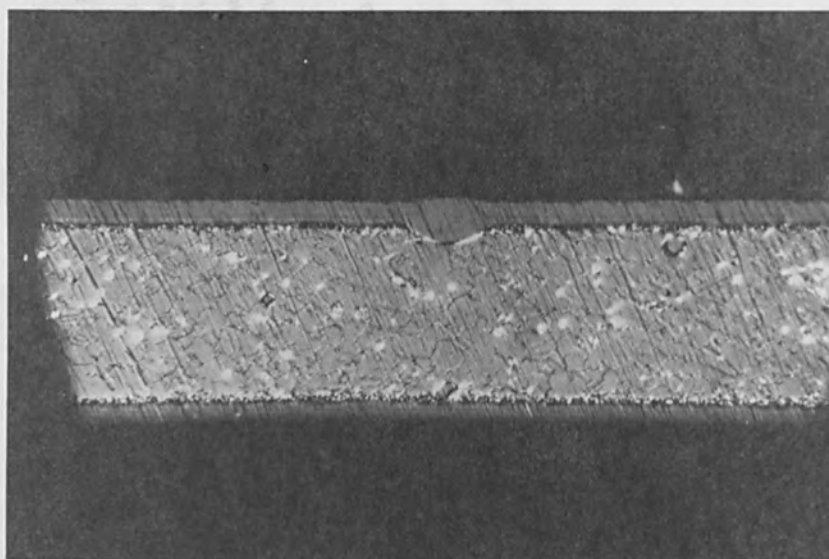


Fig. 24. EB recast track in commercial Nb₃Sn-superconductor of 6 μ thickness on 45 μ Hastelloy-B.

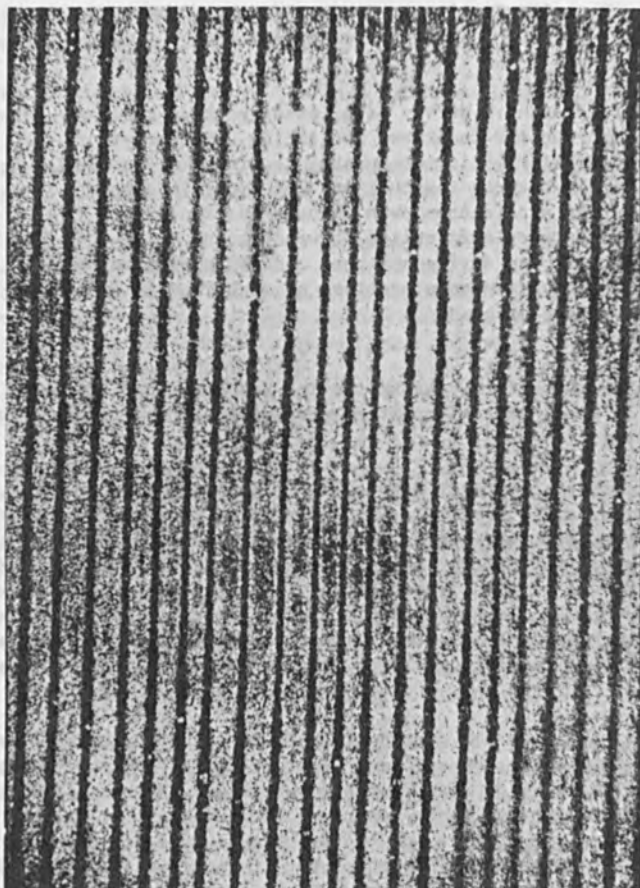


Fig. 25. EB recast tracks $20\ \mu$ wide, spaced $50\ \mu$ apart.

EVAPORATION RATE MONITORING BY SWEEPING
ELECTRON BEAM IONISATION METHOD:
DEPOSITION CONTROL OF THIN AND THICK FILMS

B. Zega

Battelle Institute, Geneva Research Centre
Geneva, Switzerland

Abstract : After a survey of existing methods for evaporation rate and thickness monitoring, a description of a particular ionisation method is given, where discrimination between residual gas and vapor is obtained by sweeping an electron beam through the ionisation chamber. Particularly suited for industrial scale evaporators, the device has been used with electron gun heated sources and some problems arising from this environment are discussed.

During the past years, vacuum coating has continued to expand and this originally small scale laboratory process has conquered many branches of industry.

This expansion is mostly due to two facts : the first is the enormous progress made in vacuum pump engineering during the past 20 years, and the second, more recent and in full evolution right now, is the progress achieved in power electron guns. The industrial scale evaporation, which was originally restricted to easily evaporating materials, like Zn and Se, is now possible and economically attractive even with refractory metals, oxides, etc., owing to the probability offered by electron guns to concentrate large amounts of thermal power on well defined spot.

The progress in vacuum coating techniques is not only quantitative. Methods have also been refined, in order to get better reproducibility and, often, more or less complete automation. Reproducibility means control, and control means measuring. What we have to measure is final thickness of the deposit, or the deposition rate and deposition time. In static, "batch" coating processes, it is often sufficient to know the final thickness; but in more refined cases, some properties of the film may depend on deposition rate : In consequence, the source needs to be continuously monitored. The same is valid for continuous, "in line" coating processes, where rate has to be held constant, and is also true for deposition of composite materials (alloys, cermet, etc.).

Among the many monitoring methods proposed until now, few are worth being considered in connection with industrial coating systems. Saturable sensors, such as the oscillating quartz crystal, in spite of their excellent performances on laboratory use, are to be eliminated.

Some simple, continuously working, non saturable methods, having a particular use, must be mentioned :

- Optical absorption of the deposited film, method limited to transparent or translucent substrates and very thin (1000 Å) coatings;
- Electrical resistivity of the film, even so limited to isolating substrates and conducting coatings;
- Magnetic permeability (permascope) method, limited to non magnetic coating on magnetic substrate or inversely, etc.

In order to find a universal method, which can be used with any evaporant and independent of the substrate, we must seek among physical phenomena, related to the density of the evaporant vapor between the source and the substrate, this density being generally proportional to the deposition rate. In this domain, the following possibilities exist and have been proposed for use :

- Selective optical absorption of the vapor;
- Excitation (by electron) of the vapor, and detection of emitted spectral lines, in visible, UV or X-range;
- Ionisation (also by electrons) of the vapor, and collection of produced ions.

The limited time does not permit to analyse particularities of all three groups. The third possibility, the ionisation method has turned our attention at Battelle Geneva some 8 years ago⁽¹⁾, and since then, we are continuously involved in this direction.

The ionisation gauge, which has been used for half a century for measurement of low pressures, has been successfully introduced in the field of evaporation rate monitoring only during the last 10 years. Fig. 1 shows in a condensed form the evolution of the ionisation gauge used as vapor beam detector under ordinary coating conditions. Six principal types are considered in the table, and in vertical columns, 5 most important criteria permitting an evaluation of the device.

As first criterion has been taken the gauge electrical output. For evident reasons there is an obvious advantage to have an alternating output signal instead of the low level d.c. signal.

But the most important problem to be resolved when working in normal vacuum coating conditions (vacuum $10^{-4} - 10^{-5}$ Torr) is the differentiation between ion currents from the residual gas and those from vapor. A differential gauge gives one possibility, but the output signal is d.c. Furthermore, the gauge electrodes are exposed to the vapor source and receive heavy metallic or dielectric deposits, which cause fail-offs. Perkins⁽²⁾ proposed a version of gauge with heated, self cleaning electrodes. Another possibility has been proposed by Crawford⁽³⁾, using a parallel electron beam, traversing two ionisation, with hidden collectors equipped chambers, one chamber containing the residual gas only, and the other residual gas plus vapor. But the problem how to differentiate two low level d.c. signals remains the same.

A much more exact method was proposed by Haase⁽⁴⁾ and followed by Schwartz⁽⁵⁾: mechanical chopping of the vapor beam before it enters the gauge. By this method, one gets a d.c. signal from the residual gas, and an a.c. signal from the vapor. In combining Crawford's geometry and Schwartz's chopper principle, one can have a very reliable monitoring device, which has been used in our laboratory with success. A mechanical chopper in vacuum may however be a source of trouble sometimes, particularly because of heavy deposit forming on it.

Both mentioned principles have one common inconvenient, when electron gun heated vapor sources are used. Both are unable to distinguish between ions created in the interior of the gauge, and ions originating from the source. It is known that about 1 % of the vapor from electron gun heated sources is ionised, and this represents a fairly high value compared with the internal ionisation (e.g. : Al deposited at a moderate rate of 100 Å/s produces in the gauge an ion current of 10^{-7} A. The rate of 100 Å/s means $100 \cdot 10^{16}$ atoms/cm²/s, and 1 % of this value is 10^{16} ions/s, giving, when single charged, a current of $1.6 \cdot 10^{-3}$ A.

This means that very reliable ion strippers have to be added, preventing ions to enter the gauge. According to our experience, it is not easy to choose a convenient stripper bias, covering very large rate ranges.

A third principle of discriminating between residual gas and vapor, which is simultaneously insensitive to ions from the exterior, is the "sweeping electron beam ionisation" method. It was presented for the first time by the author in 1968⁽⁶⁾ and has been in the meantime successfully used with very different kinds of vapor sources.

Fig. 2 shows the working principle of this gauge: a nearly parallel electron beam of constant intensity is periodically deflected in a given angle through the ionisation chamber. In absence of vapor, residual gas ions are created uniformly along the electron beam's way, and collected by the symmetrically placed ion collector, producing a d.c.

electrical signal in the collector circuit. In presence of vapor, which enters through an unsymmetrically placed diaphragm, an a.c. signal appears, which is easily separated from the d.c. background. It is evident that ions originally contained in the vapor beam can be collected on the ion collector without interfering with the "vapor" signal. A low impedance input is needed in order not to saturate the collector by this parasitic ion current.

The described device ^{*)} has been successfully used in connection with electron guns of the 10 kW range, at evaporation rates of more than 0.5 micron/s, and there was no sign of approaching any saturation. (The lowest limit of sensitivity is situated in the 1 Å/s range!)

Electron gun heated sources are not only sources of ions. They emit a large quantity of secondary electrons and X-rays, which, in their turn, produce photoelectrons on the chamber walls. One must prevent these electrons from entering the gauge and being collected by the electron collector, which, through a feedback circuit, controls the electron emission of the gauge. A collimating tube before the diaphragm, combined with a back cover, is very effective.

In order to study phenomena arising during evaporations by electron guns, a special assembly has been made as shown in Fig. 3 : between a collimating tube and the standard gauge, two deflecting plates are seen, and another collector is installed in the back cover. When the vapor source has been heated with increasing power, positive and negative currents are collected on the conveniently biased plates. In Fig. 4, gauge output signal (evaporation rate) and negative plate current (mostly dispersed electrons) are plotted versus gun input power. The evaporation rate follows a fairly linear slope, then begins to saturate. At the same time, the secondary electron current increases rapidly. This observation is important as it shows that gun power should not be increased above some reasonable limit; yield begins to drop when the vapor density over the molten metal surface reaches the value where the mean path of electrons becomes too short for them to reach the surface. At the same time, the intensity of dispersed electrons increases sharply. Obviously, any reserve of gun power should be used only in parallel with spot size increase or by scanning the molten pool with larger amplitude.

The gun used for this experiment was a 20 kV, 10 kW with 90° deflection (Leibold-Heraeus KA 10/20 FD 90). Evaporated metal was tantalum, contained in a water-cooled Cu-crucible.

As conclusion, we can say that the described principle of vacuum

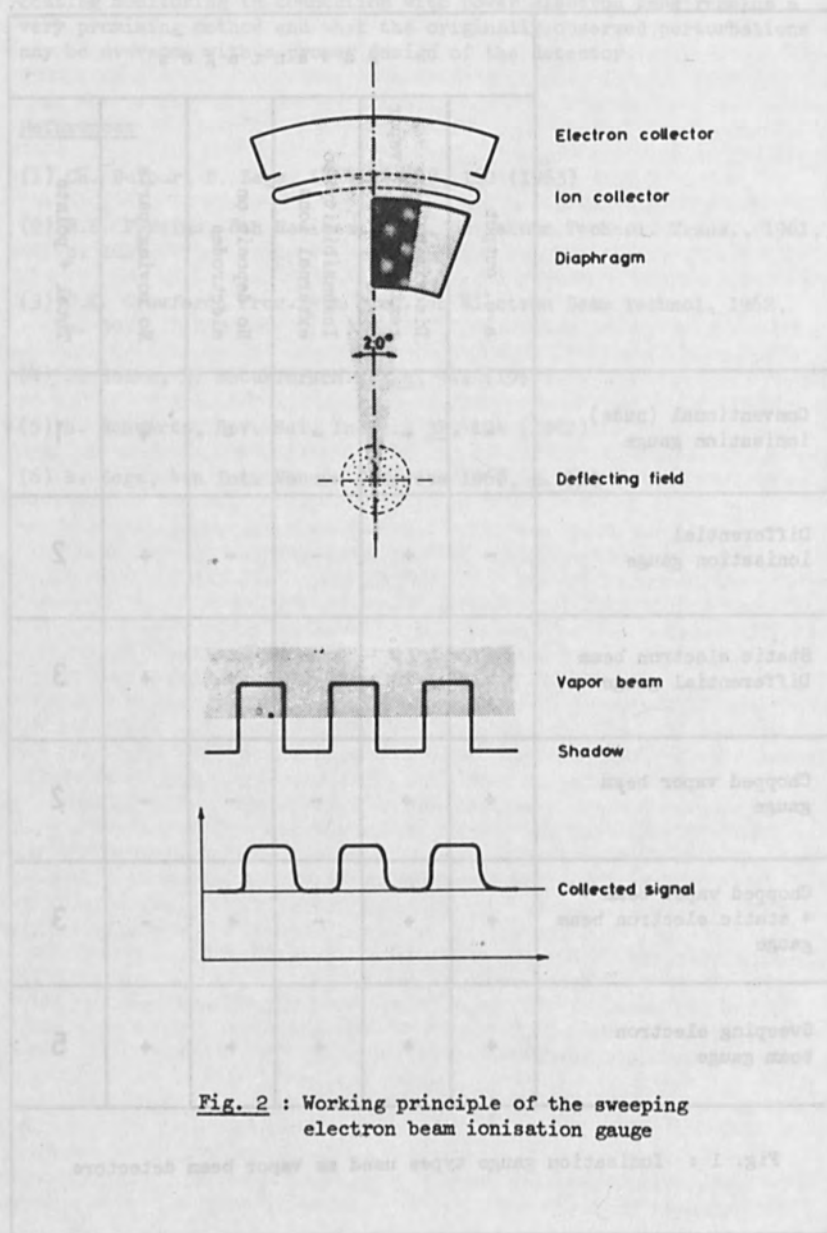
*) On market under the name "NORALP", produced by Nordac S.A., 74-Archamps, France.

coating monitoring in connection with power electron guns remains a very promising method and that the originally observed perturbations may be overcome with a proper design of the detector.

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	Advantages					
	a.c. output	Discrimination residual gas : vapor	Insensibility to external ions	No deposit on electrodes	No moving parts	Total + points
Conventional (nude) ionisation gauge	-	-	-	-	+	1
Differential ionisation gauge	-	+	-	-	+	2
Static electron beam Differential gauge	-	+	-	+	+	3
Chopped vapor beam gauge	+	+	-	-	-	2
Chopped vapor beam + + static electron beam gauge	+	+	-	+	-	3
Sweeping electron beam gauge	+	+	+	+	+	5
Fig. 1 : Ionisation gauge types used as vapor beam detectors						



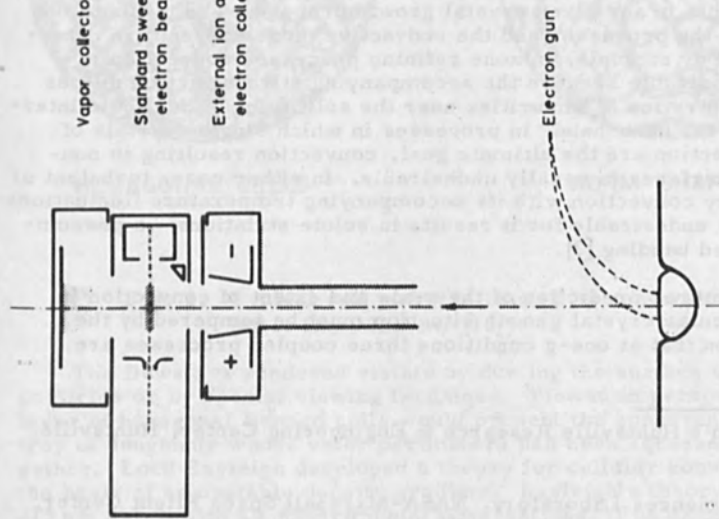


Fig. 3 : Experimental assembly ion- and dispersed electron currents from gun-heated sources

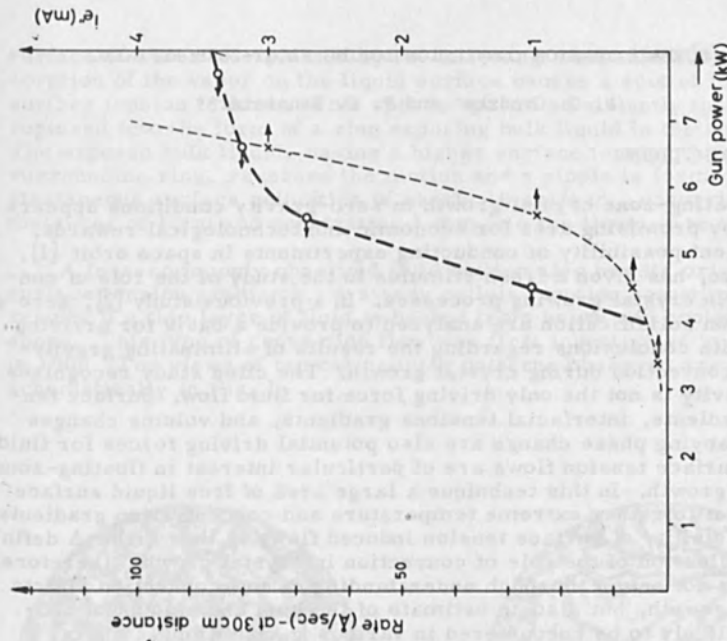


Fig. 4 : Evaporation rate of Tantalum from water-cooled copper crucible, and corresponding dispersed electron current versus gun power

SURFACE TENSION CONVECTION DURING FLOAT-ZONE REFINING

P. G. Grodzka* and T. C. Bannister**

INTRODUCTION

Floating-zone crystal growth in zero-gravity conditions appears as a very promising area for economic and technological rewards. The recent possibility of conducting experiments in space orbit [1], therefore, has given a fresh stimulus to the study of the role of convection in crystal growing processes. In a previous study [2], zero-g effects on solidification are analyzed to provide a basis for arriving at definite conclusions regarding the results of eliminating gravity-driven convection during crystal growth. The cited study recognizes that gravity is not the only driving force for fluid flow. Surface tension gradients, interfacial tensions gradients, and volume changes accompanying phase change are also potential driving forces for fluid flow. Surface tension flows are of particular interest in floating-zone crystal growth. In this technique a large area of free liquid surface is subject to rather extreme temperature and concentration gradients. The possibility of surface tension induced flows is thus high. A definitive evaluation of the role of convection in crystal growth, therefore, requires not only a thorough understanding of how convection affects crystal growth, but also an estimate of the kind and amount of convection likely to be encountered in various g conditions.

The first requirement will not be dealt with in depth. It is sufficient for present purposes to note that convection may or may not be desirable in any given crystal growth process. The ultimate objective of the processor and the convective mode present are determinate. For example, in zone refining processes convection is usually desirable because the accompanying stirring action dilutes the concentration of impurities near the solidifying solid-liquid interface. On the other hand, in processes in which single crystals of high perfection are the ultimate goal, convection resulting in non-planar interfaces is usually undesirable. In either case, turbulent or oscillatory convection with its accompanying temperature fluctuations is usually undesirable for it results in solute striations — a phenomenon called banding [3].

A ventured prediction of the mode and extent of convection in any particular crystal growth situation must be tempered by the realization that at one-g conditions three coupled processes are

*Lockheed's Huntsville Research & Engineering Center, Huntsville, Ala.

**Space Sciences Laboratory, NASA-Marshall Space Flight Center, Huntsville, Ala.

surface of water is explained by the following mechanism [8]. Absorption of the vapor on the liquid surface causes a spot of low surface tension to form. This spot is spread so violently that it is ruptured into the form of a ring exposing bulk liquid in the center. The exposed bulk liquid, having a higher surface tension than the surrounding ring, reverses the motion and a ripple is formed. Instantaneous surface velocities of nearly 10 cm/sec., occurring over distances of 1 cm and penetrating 1 mm into the liquid, are reported.

A less-commonly observed fluid motion also has its origin in surface tension gradients. A steady cellular motion sometimes results if a thin layer of fluid is heated from below and cooled from above. This type of convection flow was first investigated by Bénard in 1900. Top views of typical cellular patterns observed are shown schematically in Fig. 1.



HEXAGONAL CELLS



WORM-SHAPE CELLS

Fig. 1. Typical cell patterns caused by convection
(after Hamalainen, [12])

The flows are rendered visible by dusting the surface with fine particles or by special viewing technique. Viewed in perspective a layer of hexagonal Bénard cells would present the appearance of a tray of doughnuts whose outer perimeters had been squeezed together. Lord Rayleigh developed a theory for cellular convection on the basis of an unstable density gradient. Rayleigh's theory, however, did not agree with all experimental observations. For example,

possible. These are gravity-driven convection, surface tension-driven convection, and crystallization. Each is dependent on the local temperature and concentration conditions. Thus, the degree and manner of occurrence of any or all of the processes is inter-related. For example, the occurrence of compositional cells which have their origin in constitutional supercooling depends on the temperature gradient in the liquid phase [4]. The steeper the gradient the less likely the occurrence of compositional cells. The steeper the external temperature gradient, however, the more likely that convection will result. The internal temperature gradient near the solid-liquid interface, therefore, may be reduced. The presence of a moving solid-liquid interface which is liberating heat, in turn, influences the convective field. The complexity of the problem can be reduced to manageable proportions by postulating mathematical models, the results of which can be compared to experimental data. In this way, the chief or controlling parameters in any given crystal growth process can be identified.

In the following, the nature of studies for floating-zone crystallization currently in progress and projected space flight experiments are described. As a basis for this discussion, however, the physical nature of surface tension-driven convection and considerations specific to floating-zone crystallization utilizing electron beam heating are first described. It is appropriate to add that electron beam heating appears to be the one preferred heating method for space applications. Its advantages for this purpose are considered [5].

SURFACE TENSION CONVECTION

If a free liquid surface experiences a temperature or concentration gradient, a surface tension gradient will result because surface tension is a function of temperature and concentration. Along the surface the liquid will flow from the region of low surface tension to one of high surface tension. Usually this will be from hot regions to colder regions because surface tension generally decreases with increase of temperature. The liquid moves the fastest near the surface, the depth of the disturbance depending on a variety of hydrodynamic parameters. Fluid flow caused by surface tension gradients is called the Marangoni effect. (Several excellent general reviews of surface tension flows are given [6]). The occurrence of "tear drops" in a wine glass is a common illustration of the Marangoni effect. In this case evaporation of alcohol leaves the layer of liquid on the wetted glass wall with a higher surface tension than in the bulk of the liquid. As a result, liquid on the glass walls continuously draws up more liquid from the bulk until a "tear" is formed. When the "tear" becomes large enough it falls back into the liquid. The violent twitching observed on the surface of water saturated with ethyl ether is also attributed to surface tension gradients occasioned by evaporation [7]. The formation of surface ripples during absorption of acetone or methanol vapor on the

cellular convection was found in shallow pools cooled from below; a stable situation according to Rayleigh's theory. Also the flow patterns observed in some instances were opposite to those predicted by Rayleigh's theory. Hot liquid was observed rising below centers of depression, whereas hot liquid would be expected to coincide with elevated regions if the flow were gravity driven (see Fig. 2).

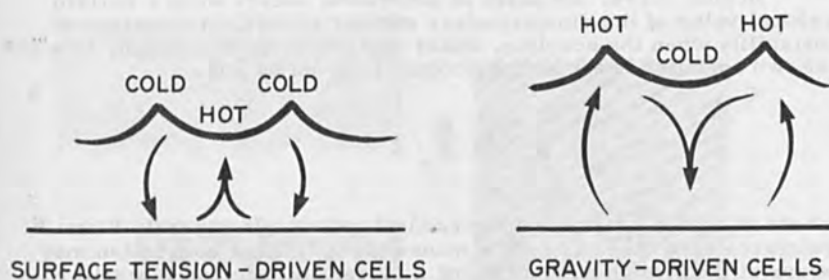


Fig. 2. Fluid flow patterns in Bénard and Rayleigh convection cells

The discrepancies were resolved when both gravity and surface tension were recognized as possible motive forces for fluid flow. Couplings between surface tension and buoyancy were also recognized as a possibility.

In one analysis of cellular convection [9] a stability criterion for surface tension-driven flows based on a dimensionless number B is given. This number known as the Marangoni number expresses the ratio of surface tension to viscous forces and is defined as

$$B = \frac{-\frac{d\sigma}{dT} \frac{dT}{dy} d^2}{\rho \nu \alpha}$$

where dT/dy , ν , α , ρ , and d are vertical temperature gradient, kinematic viscosity, thermal diffusivity, density and thickness of the fluid layer, respectively. The term $d\sigma/dT$ represents the rate of change of surface tension with temperature.

For gravity-driven cellular convection the stability criterion is based on the Rayleigh number, which is given by

$$R = \frac{g\beta \frac{dT}{dy} d^4}{\nu \alpha}$$

where g is the acceleration due to gravity and β the coefficient of expansion.

In both cases, the onset of convection occurs when a certain critical value of the dimensionless number occurs. A criterion of instability when the coupling, under maximum reinforcement, between the two agencies is taken into account is given by [10].

$$\frac{R}{R_c} + \frac{B}{B_c} \geq 1$$

where R_c and B_c represent the critical values. In general, R and B reinforce each other in such a manner that cellular convection may occur at a Rayleigh number R and a Marangoni number B smaller than their corresponding critical values, R_c and B_c .

The similarity between the appearance of Bénard cells and the hexagonal cells attributed to constitutional supercooling (see Fig. 3) is considered in a recent paper [12]. In this paper certain impurity patterns found in thin alkali halide crystals grown from the melt is attributed to segregation induced by microscale cellular convection.

The phenomenon of convective overstability is still another area when further clarification of basic physical processes is needed. When a fluid layer is subjected to an appropriate temperature gradient convection may set in as a stationary, cellular convection or as an oscillating convection with a characteristic frequency [13]. The flow can be pictured as proceeding in one direction for a time, coming to a halt, and then reversing [14] p. 744. Overstable modes result in regular oscillations in temperature and are thus of considerable interest to crystal growers. Regular temperature oscillations have been observed in molten metals contained in horizontal boats across which temperature gradients are imposed in a number of cases [15 and 16]. Overstable oscillation has been the object of a large number of mathematical treatments. The outcomes of these treatments has been somewhat inconsistent with actual observations. In particular, the presence of overstable convection was observed in cases prohibited by theory [17]. A recent treatment, [17], however, establishes that the combined effects of buoyancy and surface tension gradients may lead to overstable oscillations in certain circumstances where oscillations, according to previous theory, would not be expected. The analysis presents criteria for stationary and overstable convective

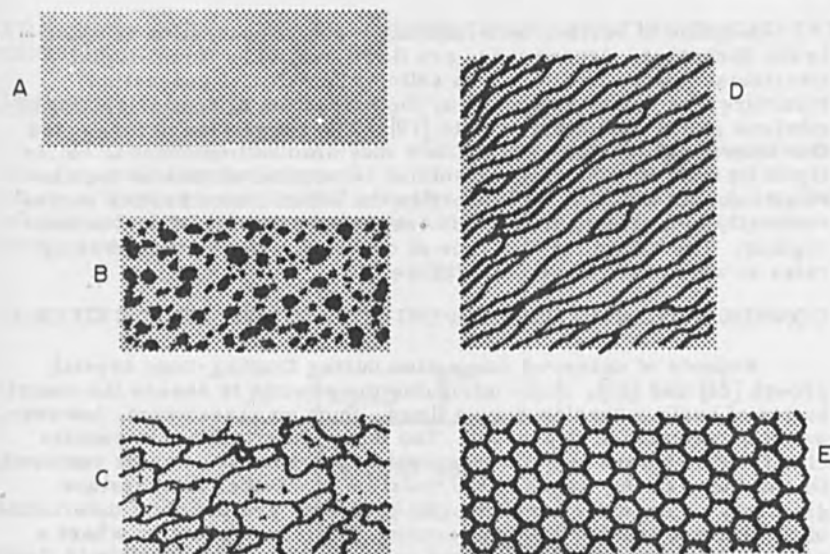


Fig. 3. Illustration of the various morphologies as a function of constitutional supercooling. Supercooling increases from (A) to (E) [11]

modes in terms of R , the Rayleigh number, R/B , the ratio of the Rayleigh number and B the Marangoni number, and a term representing a frequency characterization of the fluid velocity and temperature perturbation. Overstable oscillation is predicted for water heated from above between 0 and 4°C. Interestingly, observable melting and freezing oscillations are actually reported in one experimental study of freezing water in a temperature gradient [18]. No free liquid surfaces were present in the experimental study, thus raising the interesting possibility of liquid-solid interfacial tension involvement.

Mention of various less familiar types of convection is found in the literature. Among these are flows caused by liquid-liquid interfacial tension gradients. A cellular interfacial convection structure was found to accompany the extraction of acetic acid out of ethylene glycol with ethyl acetate [19]. The suggestion is forwarded that interfacial tension-driven flows may also be important at solid-liquid interfaces [20]. Thermosolutal convection results in certain situations. In this type of convection the solute concentration varies vertically in a gravity field. The result is a layering of convection regions. The origin for this type of convection lies in the differing rates at which heat and solute diffuse [21].

CONSIDERATIONS IN DEVELOPING CONVECTIVE MODE CRITERIA

Reports of observed convection during floating-zone crystal growth [22] and [23], make intriguing the attempt to assess the contribution of surface tension driven flows. Such an assessment, however, would be premature at present. Too many uncertain areas require clarification before even the roughest approximations can be ventured. One of these cloudy areas is the question of whether temperature gradients or impurity concentration gradients are the chief determinate of the surface tension gradient causing flow. In a situation where a gaseous impurity is being evolved or absorbed, as is possible in floating-zone processes, it is probably reasonable to assume that the impurity is the chief determinate of the observed flows. Dissolved oxygen and nitrogen are reported to be capable of drastically lowering the surface tension of metals [24]. The probable nature of the impurity present, however, must be considered carefully. Impurities which lower surface tension (positively adsorbed) will tend to cause rippling on free liquid surfaces. Impurities which increase the surface tension (negatively adsorbed), however, apparently cause no fluid flow when they are absorbed from the vapor [8]. A very strongly positively adsorbed impurity — i. e., a condensed monolayer — will inhibit convection dramatically. The presence of a strongly adsorbed impurity is reported to increase the temperature gradient which water is able to sustain without convection from 0.72°/cm to about 180°/cm, [14], p. 743.

Finally, the method of heating may itself be responsible for additional convective modes. Floating-zone crystal growth processes

are limited under one 0-g conditions to materials of high surface tension. These materials are usually metals with high melting temperatures and, therefore, necessitate electron beam or induction heating to create a molten zone. Although induction can introduce considerable stirring in the molten zone, the amount of direct convection resulting from electron beam impact is judged to be negligible. Electron pressures of the order of several dynes per centimeter square are judged to cause negligible distortion of the shape of molten titanium drops [25].

PRESENT AND PROJECTED STUDIES OF CONVECTION IN CRYSTAL GROWTH

Mathematical Models

Mathematical models which will predict melted zone length and freezing interflow face shape as a function of the independent parameters of floating-zone crystal growth are being formulated.* A detailed mathematical model, somewhat as presented in Fig. 4, is currently under development.

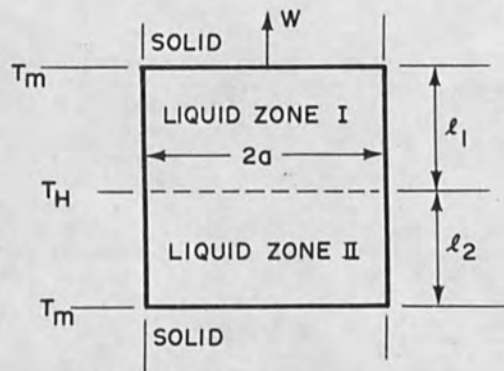


Fig. 4. Initial mathematical model of floating-zone crystal growth

*Dr. Ben Johnson of Lockheed's Huntsville Research & Engineering is responsible for the mathematical formulation.

The melted zone is approximated by two cylinders of height l_1 and l_2 and diameter $2a$. Cylindrical coordinates are being used. The melted zone moves upward (or downward) with an average velocity ω . Both upper and lower solid-liquid interfaces are assumed to be at the melting point of the material. The main heat flow across the freezing interface is the heat of fusion. This heat of fusion is also introduced through the sides of the cylinder. The shape of the freezing interface is determined by the heat transfer characteristics and the remaining boundary conditions. Also, it is necessary to introduce some degree of superheat into the melt. This superheat condition is reflected in the temperature T_H , which is a function of the radius, at the boundary between Zones I and II.

The heat passing through the vertical, side face of Zone II is assumed to be the heat necessary to melt the material that crosses the boundary. It is assumed that the only heat transfer across the boundary between Zones I and II is carried as internal energy by material crossing the boundary. The excess heat carried into Zone I is assumed to be radiated through the side of Zone I. The solution to the general equations governing the system is assumed to be the quiescent solution plus a perturbation. The effect of surface tension is introduced into the model by means of the boundary condition.

$$-\mu \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_r}{\partial r} \right) = \nabla_1^2 \sigma$$

where μ is the viscosity, r the radial coordinate, u_r the radial fluid velocity, σ the surface tension, and

$$\nabla_1^2 = \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial Z^2}$$

where θ and Z are the other two cylindrical coordinates. The former equation represents the balance of shear stress at the free surface. Similar conditions will be defined for Zone II.

Ground-Based and Flight Experiments*

A number of ground-based experiments pertinent to the subject of floating-zone crystal growth in zero-g are being performed by investigators representing both government and industrial concerns. Reference is made to the 1969 Space Processing Meeting, held at the Marshall Space Flight Center, Huntsville, Alabama. In one current study, certain features of mathematical models are checked by conducting contained zone crystallization of an organic compound of low melting temperature. In particular, the length of the melted zone and the shape of the freezing interface are being determined as a function of the rate of heater travel and rod diameter. Studies of

the dependence of impurity banding on the thermal boundary conditions during crystal growth of indium antimonide, In Sb, are also currently underway. Indium antimonide crystals grown from tellurium-doped melt, exhibit rather marked impurity striations. It is reported that striation free indium antimonide crystals result if the crystal growth is conducted in a magnetic field [26] and [27]. The magnetic field acts to dampen melt convection and, hence, temperature oscillations. The Czochralski technique is being used for the present.

To simulate as closely as possible crystallization under zero-g a series of experiments utilizing a drop tower are planned [28]. The influence of zero-g during free fall on melts of high melting points metals will be investigated. In particular, the geometry and shape, grain structure, and other structural features will be studied in metal spheres solidified during free fall. A drop time of one second is attained for a drop length of 16 feet. Development of the tower is currently underway.

One flight experiment on crystal growth is currently under development for a future flight. This experiment is being designed to study the solidification of materials in orbit whose behavior is highly observable and whose handling is relatively simple. The experiment is also designed to visually and photographically observe through a microscope the freezing of transparent materials. Laboratory movies (time lapse) are presently available in which studies [29] are documented on the behavior of such materials as hexadecane, lithium nitrate trihydrate, and water. Bubble formation, convection, nucleation, auto-nucleation, and growth rates are easily observed. (Similar studies have been made in a centrifuge where smaller grain sizes were observed.)

A number of experiments for future, prolonged space orbit flights are now in the planning stage. Among those being studied are a set of experiments on convection in zero-g. These have a two-fold objective. One is to generate new knowledge on convection decoupled from gravity and the other is to provide data for possible crystal growth experiments in space.

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An Investigation of Methods of Preparing
High Purity Vanadium^{*†}

H. A. Beale and R. J. Arsenault
Engineering Materials Group
College of Engineering
University of Maryland

Abstract

High purity vanadium single crystals (resistance ratios in excess of 600) have been grown in an electron beam float zone refiner. Numerous parameters have been varied and their effects on the purification process studied. Also hydrogen and ammonia gases were introduced during the first molten zone pass and pumped back out on subsequent molten zone passes. Most experiments were performed twice, once with, and then without, pedestal rod rotation.

Introduction

In recent years there has been a significant and somewhat concerted effort by a growing number of investigators to produce high purity single crystals of the body-centered cubic refractory metals. The reasons for investigating methods of producing high purity vanadium are basically the same reasons that have led various other investigators to produce high purity single crystals of other b.c.c. refractory metals. However, in the case of vanadium, there is an additional reason for producing crystals of high purity. The radioactive isotopes of vanadium

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have very short half lives and hence the radioactivity caused by irradiation decays rapidly. Therefore vanadium can be studied in conventional surroundings.

Investigations of methods of producing high purity vanadium single crystals are almost non-existent. The calcium reduction technique^(1,2) presents the drawback of severely contaminating the vanadium with the material of the crucible and so eliminates the possibility of doing irradiation studies. A possible reason for the subsequent lack of interest in attempting to produce high purity vanadium could be the theoretical work of Brewer and Rosenblatt⁽³⁾ which indicated it was at least two orders of magnitude more difficult to purify vanadium than niobium. Also Stringer⁽⁴⁾ has calculated that an oxygen partial pressure of less than 10^{-13} torr is required to reduce VO.

High vacuum electron beam zone melting has proven to be an efficient method of purifying some of the b.c.c. metals⁽⁵⁾. However, the literature concerning the high vacuum electron beam zone melting of vanadium is very vague,⁽⁶⁾ and most conclude it is not a practical means of purification⁽⁷⁻⁹⁾. The principal reason for the interest in the removal of oxygen is due to the large tetragonal strain associated with interstitial oxygen. The strain due to oxygen is four times larger than the strain produced by a nitrogen interstitial⁽¹⁰⁾ and could be at least three times larger than a carbon interstitial in the vanadium lattice.⁽¹¹⁾

There are reasons for assuming that if oxygen could be removed, the remaining interstitial impurities would not be difficult to remove. The stable nitride of vanadium, VN, should decompose above 900°C in vacuo,⁽¹²⁾ and it has been stated that nitrogen follows Sievert's law in vanadium.⁽⁸⁾ Therefore nitrogen should be readily removed by high vacuum annealing above 900°C. The case of carbon is not as clear, but Powers and Doyle⁽¹³⁾ state that the addition of carbon to vanadium was difficult to accomplish.

The purpose of this investigation was to consider various

techniques of high vacuum electron beam zone melting as a means of producing vanadium with a low interstitial concentration.

Experimental Procedure

The starting material was obtained from the U. S. Bureau of Mines, Boulder, Colorado, in the form of dendritic crystalline powder. The initial analyses of the powder are shown in Table I. The aggregate purity of the dendritic crystals was also checked by determining the value of the ratio of the resistance at room temperature to the resistance at 4.2°K.

The experimental equipment consisted principally of a modified arc melter, which has been described elsewhere⁽¹¹⁾, and an ultra-high vacuum electron beam float zone refiner. The zone refiner is shown diagrammatically in Fig. 1. Several features make the system quite unique. First, the system incorporates internal heaters. Long strip heaters are strapped to the scanner assembly, making it possible to bake the entire scanner to temperatures in excess of 425°C. Second, a special split mantle is so designed that, in conjunction with the placement of external heaters, it is possible to bake most of the surface area of the 12 in. dia. stainless steel bell jar to 325°C, the viewing ports to 300°C, and the electrical feed-throughs, with their ceramic insulators, to 225°C. This non-uniform temperature distribution greatly increases the life of the electrical feed-throughs and has an insignificant effect on the value of the working vacuum. After baking the system, this mantle can be easily removed since it is split. Third, the variable leak valve, which is not thermally insulated, is preceded by a bakeable valve. The bakeable valve is baked to 200°C but the precision leak valve does not reach temperatures in excess of 75°C. This has greatly increased the life of the sapphire valve seat in the precision valve without affecting the value of the working vacuum. Fourth, the system is equipped with a turbomolecular pump which is usually the only active high vacuum pump during the baking period. The

advantage is that the solid state pumps may be baked while being inoperative. In addition, there is a large valve preceding the turbomolecular pump, so that this pump may be closed off the system when pressure is below 1×10^{-8} torr. This eliminates the possibility of backstreaming of oil or hydrogen through the pump. Fifth, the stainless steel bell jar is equipped with a cooling jacket. The jacket covers approximately 80% of the external surface area of the bell jar. A mixture of pure ethylene glycol and water is used as the coolant and during an experiment, it is possible to maintain the temperature of the coolant at -10°C . Sixth, the system is equipped with four pressure measuring gauges. The first is included in the VacIon pump and is reasonably reliable only to 1×10^{-9} torr. The second is actually a monopole residual gas analyzer. This unit is reliable only to 1×10^{-10} torr in the total pressure mode, so a Keithley Model 610C electrometer is used to facilitate measurements at lower pressures. The substitute electrometer is also used on occasion to help resolve the residual gases. The third is a nude ion gauge located at the base, but in front of the scanner. The fourth is also a nude ion gauge located at the base of the scanner, but this one is located behind. At no time is this last gauge in a direct line to the molten zone. With no filament power and a pressure in excess of 1×10^{-9} , all the gauges usually agree to within a factor of 3x. When a molten zone has been established, the gauge in front of the scanner indicates the highest pressure, the gauge behind indicates lower, the monopole analyzer indicates lower still, and the gauge in the pump indicates the lowest pressure. In a normal experiment however the monopole analyzer is usually maintained in the partial pressure mode. The reason for this is that the emission current for the gauge filament is different for the two modes of operation and it takes considerable time to achieve equilibrium with respect to the temperature of the tube. The value of the total pressure within the system is taken as that indicated by the gauge located behind the scanner.

Since all pressure determinations from ion gauges are usually accepted with suspicion, the values quoted herein as true total gas pressures are therefore conservative in this regard. Unfortunately the stainless steel bell jar is equipped with only two viewing ports and there is a dead space between these ports. When the electron gun is moving in this dead space, it is impossible to view the heat zone. By observing the pressure indicated by the gauge located in front of the scanner, it is still possible to determine if a molten zone exists. Seventh, the motion of the electron gun or filament is by direct drive. The reason for going to direct drive was to make a first attempt to eliminate the banded structures which other investigators⁽¹⁴⁾ have found. This also makes the mechanism simpler, a laudable feature for any mechanism used in ultra-high vacuum work. Eighth, the system incorporates the ability to rotate the pedestal rod, again by direct drive. The traverse motion of the block which holds the pedestal rod is also by direct drive. However, if the pedestal block is moved over 4 inches from bottom, the pedestal rod rotation mechanism uncouples.

Many of the experiments to be described have certain initial steps in common and where deviations occur, they will be noted. First, an ingot, prepared in various ways to be described, is swaged to the desired diameter. The rod is etched in a mixture of HF and HNO₃ before each reduction. The rod is next cut to length, etched again, and installed in the zone melter. The pedestal rod is a seed crystal and all crystals grown during this series of experiments had the same orientation to eliminate a variable. The orientation chosen was approximately 14° from the (110) and 5° from the (110)-(111) symmetry line of a standard stereographic triangle. The chamber is closed and evacuated to approximately 1×10^{-5} torr. If a gas is to be introduced during the experiment, the leak valves are opened to the gas source and the necessary lines are purged with that gas. The precision leak valve is then closed and the entire system is put into the baking

cycle. After baking for approximately 16 hours, with the temperature distribution noted earlier, the pressure is typically 2×10^{-6} torr. At this time the system elements are taken off bake out in a programmed manner. Just prior to bringing the last element, the bell jar, off bake out the pressure is 3×10^{-7} torr. When the pressure drops below 1×10^{-8} , the turbomolecular pump is valved off the system. The system is usually allowed to cool until the surface of the bell jar not covered by the cooling jacket is fairly close to ambient temperature, at which time the pressure is typically 2×10^{-10} torr, with the principal residual gas being hydrogen.

The filaments are usually hand-wound from 0.025 in dia. tungsten wire with the filament diameter being approximately 1.25 in. The diameter of the rods to be zone melted are typically 0.250 in. The next step in the process is to energize the filament without turning on the high voltage. The filament is then made to traverse the length of the rod. It is found that the pressure within the system will rise as much as two orders of magnitude during this operation with about 85% of this difference due to hydrogen. In this mode of operation, the temperature of the rod does not exceed 400°C since the heating is obviously by radiation only. Such a low temperature of course prohibits any significant amount of diffusion within the rod, even of hydrogen.⁽¹⁵⁾ Therefore the hydrogen and other minor gases must be coming from the surface of the rod.

If a gas is to be introduced during the first molten zone pass, the next step is to weld the rod and the pedestal rod together and then shut down all power. The solid state pumps are shut down, the valve to the turbomolecular pump is opened, and the system is allowed to equilibrate. The appropriate gas is introduced to near the desired pressure, a molten zone is established about 3 in. above the position of the original weld and a zone pass is made. No sputtering, i.e., the molten zone spewing gas and, if violent enough, metal particles out of itself, occurs when the first molten zone pass is made at these higher pressures. The rod is then pulled apart

while a molten zone is present and the entire system is put back into the bakeout cycle. The experiment proceeds further from this point almost as if the introduction of the gas had not transpired. For the gases used, the ammonia was commercial grade while the hydrogen was high purity and was passed through a palladium diffusion purification apparatus prior to its introduction into the system. The maximum pressure for which electron beam zone melting is possible was found to be roughly in agreement with other investigators,⁽⁷⁾ but once a fluctuation in the system caused a plasma to develop during a hydrogen experiment and all later experiments were conducted at a slightly lower pressure.

If no gas was to be introduced, a second "light" degassing pass is made in the opposite direction. Pressure during this second pass is usually an order of magnitude lower than for the first pass, but the principal contaminant is still hydrogen. At the conclusion of this pass, the rod and pedestal rod are welded together and then a beam power degassing pass is made. The power used is approximately 60% of that required for the welding process. The temperature at the outside center of the heat zone is probably in the range of 700 to 1300°C. Pressure during this pass is usually in the same range as for the second light degassing pass, but the residual gas composition is now more divided, though hydrogen is still dominant. In the case that a gas had previously been introduced, the pressure is usually one to two orders of magnitude higher and the major contaminant, for the experiments involving hydrogen or ammonia, is by far hydrogen. Also in the case that a gas had previously been introduced, the next operation is a molten zone pass, otherwise a second beam power degassing pass is made.

If no gas had previously been introduced, the initial molten zone pass is always made in the upward direction, starting just below the weld. A single crystal always seems to result, irrespective of the zone speeds used during the experiments. The growth of crystals of the same orientation at this point will always have the

same surfaces exposed to the vacuum atmosphere. The purpose for this is that some investigators⁽¹⁶⁾ believe that the hot portions of the rod above and below the molten zone react readily with the residual gases within the chamber. Hence after the first pass the reactive surfaces exposed were always the same.

If a gas had previously been introduced, the next molten zone pass is always made in a downward direction, since it is not desirable to contaminate the seed. The result of the third molten zone pass has the proper orientation.

The number of molten zone passes for a specific experiment is determined either by previous, similar experiments or by experimental conditions, i.e., the pedestal rotation comes uncoupled as mentioned earlier, the viewing ports become too coated with condensed metallic vapor to allow observation of the experiment (even though a rotating window cover is used) or it becomes seemingly impossible to maintain a molten zone.

For resistivity ratio determinations, the methods of cutting the crystals and welding the leads has been described elsewhere.⁽¹¹⁾ The circuit used is shown in Fig. 2. All connections are made within a grounded aluminum box which also contains the standard resistors, to avoid thermal effects. For measurements at room temperature and at 77°K, the currents used never exceed 0.6 amp to avoid heating the sample. For measurements at 4.2°K, currents as high as 6 amps are used. A simple switch allows reversing the flow of current for each current value so that thermal emf's may be eliminated in a very straight-forward manner. Measurements at 4.2°K are of course made in the presence of a magnetic field. Data is taken as a function of the field values and extrapolated to zero field.

The internal friction apparatus was designed here and was designed to have a much lower background Q^{-1} than other torsional pendulum systems mentioned in the literature.^(6,13) Temperature is controlled by a thermocouple located adjacent to but not touching the middle of the sample and two temperatures are recorded during

an experiment, one from a thermocouple in contact with the stationary lower specimen grip and the other from a thermocouple hanging free near the top of the specimen. The temperature is varied in a programmed manner and data points are observed every 15 min, since it has been verified that equilibrium has been reasonably well established within this interval of time. The system can be evacuated to a pressure of 3μ , but the tests are conducted in a dynamic helium atmosphere of approximately 27μ . All the specimen tested had a 0.050 in. dia. and were tested at a frequency of 4 Hz. The method of specimen preparation has been described elsewhere.⁽¹¹⁾

Experimental Results

The starting material for this series of experiments was obtained from the U. S. Bureau of Mines, Boulder, Colorado, in the form of dendritic crystalline powder. The initial analyses of the powder are shown in Table I. The resistance ratio of two of the largest dendrites in Lot A was determined to be approximately 1000.*

A summary of the first or vacuum series of experiments is shown in Table II. The starting rod for these experiments was obtained by arc melting compacted powder, and then cold swaging the ingot to a 0.250 in. diameter. The most significant result is the reduction in the oxygen concentration. However, the variation in the total pressure during the last molten zone pass results in no consistent variation in the resistance ratio, provided the total pressure was below 4×10^{-9} torr.

The rotation of the pedestal rod appears to result in a contamination of the crystal, i.e., the resistance ratio of the rotation zoned crystal is, in general, less than that of a non-rotated sample. However, increasing the number of zone passes does result in a significant increase in the resistance ratio.

* As determined separately by Dr. R. E. Reed of the Oak Ridge National Laboratory and Dr. D. R. Anderson of the Department of Physics, University of Maryland.

The number of molten zone passes was generally limited to four, because in some cases the zone became unstable, i.e., the surface tension decreased and it was seemingly impossible to continue. However, by swaging a four zone pass crystal bar to a 0.187 in. diameter rod it was possible to increase the number of molten zone passes to a total of eight. Once a problem with the surface tension decreasing occurs, all subsequent passes are made in the upward direction, where as prior to that, the direction is alternated for each successive pass. This was done since the expected method of purification was vacuum distillation. The resistance ratio of the 8 pass crystal was 360 where as the 4 pass crystal had a resistance ratio of 193, and both crystals were zoned at the same total pressure.

From a partial pressure analysis during the last molten zone pass, hydrogen was always found to be the dominant residual gas, with the others being quite varied. If the Orb-Ion pump was shut-off, the total pressure was lower. Hydrogen was still the principal residual gas, but, of the others, CO and CO₂ were more dominant. Interestingly, the resistance ratio was smaller than in experiments where the Orb-Ion pump was used.

A second series of investigations, or the gas addition series, is presented in Tables III and IV. The technique of the addition of a third component to remove a second component and result in a more pure first component is a well known technique for zone melting.⁽¹⁷⁾ Hydrogen was chosen because previous investigations⁽¹²⁾ have shown that hydrogen reduces some of the oxides of vanadium. Hydrogen was also chosen because it exhibits a reasonable attempt to follow Sievert's law in vanadium at low temperatures⁽¹⁸⁾ and at higher temperatures⁽¹⁹⁾. Further, the hydride is not stable at the temperature of molten vanadium.⁽¹⁵⁾ Also, some of the mechanical properties of hydrogenated vanadium have been investigated⁽²⁰⁾ for possible later comparisons. Ammonia was chosen because it should have all the benefit of hydrogen and also nitrogen. Nitrogen is supposed to follow Sievert's law in vanadium.⁽⁸⁾ Other investigators have noted that ammonia reacts

more favorably to reduce the oxides of vanadium than does hydrogen.

(12) However, their work was done at low temperatures. Also the solubility of nitrogen in fairly pure vanadium has just recently been investigated.⁽⁶⁾ In the low temperature investigation, it was also noted that carbon was reduced from the vanadium with the production of HCN gas and it was hoped that the same reaction would prevail at higher temperatures. The zone speed during the introduction of the various gases was always 20 cm/hr.

The addition of hydrogen or ammonia during the first molten zone pass did not improve the resistance ratio or decrease the oxygen content of the as-zoned crystals. In fact the resistance ratio is lower than that of the crystals grown without hydrogen or ammonia additions and the oxygen concentration is greater.

Again, similar to the results obtained from the high vacuum series of investigations, rotation of the pedestal rod results in even smaller resistance ratios and larger oxygen concentrations than without rotation.

At the completion of the molten zone passes for the hydrogen experiment without rotation, an attempt was made to anneal the crystal bar, in situ. However, the total pressure within the system began to increase significantly and the experiment was consequently terminated. The system total pressure after the power was shut down was 6×10^{-11} torr, which is typical, so the problem was apparently not a vacuum leak.

At a later date, the samples for resistance ratio measurements from experiments 8, 10, and 12 were given a short duration, low temperature anneal as a check on the behaviour of experiment 7. The crystals were annealed in the EBFZR by placing them in molybdenum wire baskets and heating with the electron beam. The system total pressure increased two orders of magnitude as each of the crystals, in turn, were heated to approximately 800°C ($\pm 150^\circ\text{C}$). The dominant contaminants were hydrogen and carbon monoxide and, to a lesser extent, carbon dioxide. New leads were welded (the original leads had been etched away prior to annealing)

and the resistance ratios measured. The low temperature anneal resulted in a significant increase in the resistance ratio of the hydrogen rotation zoned sample, had no effect on the ammonia rotation zoned sample, as shown in Table IV, but, the resistance ratio of the ammonia without rotation sample decreased noticeably.

For all the experiments involving ammonia, no problem with the surface tension decreasing occurred. Part of the problem may have been due to the relative impurity of the gas used. During the third molten zone pass, the presence of helium and argon was still detectible in all cases. These gases are never detected after the second "light degassing" pass on the other experiments. Also, all the vacuum pumps on the system were found to pump ammonia gas in a very sluggish manner. In the hydrogen addition case, problems with the surface tension decreasing occurred during the third pass of one experiment but did not reoccur on the fourth pass.

A summary of the third or powder series of experiments appears in Table V. The arc-melted ingot was obtained by directly melting the dendritic crystalline powder, without previously compacting it. The oxygen and nitrogen contents of the arc-melted ingot are comparable to those obtained when a compact is arc-melted. However, there appears to be a greater reduction of the oxygen content during zone melting when the arc-melted ingot is not made from compacted powder. Again, increasing the number of molten zone passes, even though the pressure does not decrease, results in lower oxygen concentrations. A high temperature ($1600^{\circ}\text{C} \pm 150^{\circ}\text{C}$) anneal at high vacuum resulted in an increase in the oxygen concentration and a decrease in the resistance ratio.

The fourth or zoned compact series of experiments is summarized in Table VI. Here, the compacted dendritic powder was placed directly in the EBFZR. The diameter of the compact was 0.75 in., and no focussing plates with such a large aperture were available, so a nude filament was used. The height of a completely molten zone would have been prohibitive under such conditions, so only the surface of the ingot was melted. The technique used involved

getting the surface molten, letting it solidify, translating the filament, and then repeating the process. It was however, possible to melt the compact through its entire cross-section at two places along the length, using this technique. The result was a vanadium compact within a vanadium tube. This ingot, which had a 0.5 in. dia., approximately, was swaged to furnish a 0.250 in. diameter rod.

Again the oxygen concentration decreased with an increased number of zone passes. However, the most significant observation is the high value of the resistance ratio of the annealed crystal, 655, and it should be recalled that annealing at high temperatures and at a high vacuum results in an increase in the oxygen concentration and a decrease in resistance ratio. Further, it should be noted that the last three of the five zone passes were in the upward direction. The sample from the bottom of the crystal bar had the ratio of 655 whereas the sample with the ratio of 232 came from the top.

The internal friction results are shown in Fig. 3. The first sample was taken from a swaged rod and was annealed for 75 minutes at 700°C and at a pressure of 7.0×10^{-9} torr. The sample was analyzed and was found to contain 118 wt. ppm. oxygen and 127 wt. ppm. carbon. The second sample was taken from a zone melted crystal bar, reduced to the proper size for the internal friction experiment in a manner identical to the first sample. It was then annealed for 45 minutes at 700°C and at a pressure of 2×10^{-9} torr. This sample was analyzed and was found to contain 46 wt. ppm. oxygen and 30 wt. ppm. carbon. The value of the room temperature damping which is taken as the background Q^{-1} was less than 0.00015 in both cases. A half-width analysis shows that both curves may be composed of more than one relaxation process, i.e., the activation energy so determined (24 kcal/mole) is less than that for either carbon or oxygen.⁽¹³⁾

Since the spectrum of values exists in the literature for the value of the resistivity of vanadium at room temperature⁽²¹⁻²⁶⁾,

this was measured on the sample used for the resistance ratio determination for experiment #1. The value calculated for the resistivity at 296°K is $22.8 \mu\Omega \cdot \text{cm} \pm 0.3\%$, where the error is principally in the measurement of the lead spacing.

Discussion

This extensive investigation indicates several major points which are contrary to the existing literature concerning vanadium and supports some of the conclusions which have resulted from the investigations of others.

The first major point which is contrary to the literature, is that it is possible to improve the purity of vanadium by ultra-high vacuum electron beam zone melting techniques.^(3,4,7-9) In Fig. 3, it is shown that the zone melting operation removes free interstitial oxygen, as well as free interstitial carbon. However, it is to be noted that the analysis values deviate significantly from the internal friction results. Oliver and Garofalo⁽²⁷⁾ noted similar behaviour for carbon and oxygen in iron, but for much lower concentrations. The absence of nitrogen for the case of the sample from the arc-melted and swaged rod is in keeping with the usually low concentration of nitrogen in vanadium from the arc-melter, the fact that the swaging operation has no tendency to introduce nitrogen, and that the sample was annealed at a total pressure of 7×10^{-9} torr. The large carbon contamination occurring during the arc-melting operation could be due to the vaporization of the vacuum grease used in the sliding members inside the arc-melter.

The vacuum series of investigations did prove that it is possible to reduce the oxygen concentration by zone melting at total pressures of 10^{-9} to 10^{-10} torr. However, variation in the pressure from 10^{-9} to 10^{-10} torr did not result in a corresponding variation in the oxygen concentration. Also, high vacuum annealing (10^{-10} torr) at $1600^\circ\text{C} \pm 150^\circ\text{C}$ resulted in an increase in the oxygen concentration. It was previously proposed⁽¹¹⁾ that the reduction in the oxygen concentration which occurs during zone

melting and not during annealing was the result of the envelope of metallic vapor which surrounds the molten zone.^(16,28) This vapor envelope effectively reduces the partial pressure of oxygen below 10^{-13} torr so that oxygen removal can occur.⁽⁴⁾

A second point which is contrary to the existing literature on zone refining concerns the rotation of the pedestal rod. The results of this investigation clearly indicates that pedestal rod rotation does not result in greater purification as has been found in iron.⁽²⁷⁾ Pedestal rod rotation here results in higher oxygen concentrations and lower resistance ratios. The significance of the increase in the oxygen concentration will be considered along with the subsequent discussion of the gas additions.

A significant point concerning high purity vanadium is that there is not necessarily a correlation between the oxygen concentration and the resistance ratio. Comparing the results in Tables II and V, for approximately the same oxygen concentration, the resistance ratio varies from 193 to 392. This large difference in resistance ratio is probably due to another impurity, which could be carbon. The internal friction results support such an assumption. This observation is again contrary to the literature⁽¹³⁾, where it has been stated that adding carbon to vanadium was difficult.

The gas addition series of experiments (Tables III and IV) is very interesting. The first conclusion here is that the addition of either hydrogen or ammonia at the melting point is harmful, i.e., results in a higher interstitial concentration in comparison to the vacuum series of experiments, though ammonia is not as harmful initially as hydrogen.

The second point is that the rotation of the pedestal rod is again harmful. The hydrogen series and also the ammonia series indicates that, for a constant number of zone passes, rotation is worse, irrespective of the final value of the pressure.

An explanation of the detrimental effects of the gas additions could be based upon the impurities existing as gases. However, if this were the case, then the ammonia experiments should have

resulted as the poorer case, but just the opposite was observed. Therefore it must be concluded that the gases themselves have a detrimental effect in terms of purifying vanadium. This conclusion is further substantiated by considering the results in Table IV. A short duration, low temperature anneal caused significant changes which ideally should not have taken place. The resistance ratio almost doubled for the hydrogenated sample. Earlier it was noted from the literature^(18,19) that hydrogen is supposed to follow Sievert's law in vanadium. If this were absolutely the case, then very little should have happened during the anneal where the hydrogen partial pressure was almost two orders of magnitude higher than during zone melting. The only reasonable explanation for such behaviour is that hydrogen must exhibit retrograde solubility in vanadium. However, the high temperature investigation⁽¹⁹⁾ was limited to temperatures below 1300°C, and therefore this phenomena was not observed.

In the case of nitrogen (ammonia), there was a significant decrease in the resistance ratio compared to the other experiments. This clearly indicates that high vacuum (10^{-10} torr) does not result in a reduction in nitrogen concentration, which is again contrary to the existing literature^(8,12) which states that nitrogen follows Sievert's law in vanadium.

The harmful effects of pedestal rod rotation cannot be easily explained. The usual reason for the use of pedestal rod rotation is to improve mixing within the molten zone. However, before considering the mixing within the molten zone, it is necessary to consider the diffusion of oxygen, carbon, nitrogen, and hydrogen in vanadium.

Powers and Doyle⁽¹⁰⁾ have tabulated the appropriate values for oxygen, carbon, and nitrogen in vanadium at low temperatures from their internal friction and elastic after-effect investigations. Schmidt and Warner⁽²⁹⁾, in their electromigration studies, have determined the same quantities at high temperatures. Their results indicate a similar activation energy in all cases, but their values

for the diffusivities of carbon and nitrogen are greater by a factor of two, which is not uncommon for b.c.c. materials.⁽³⁰⁾ It is more interesting to note that oxygen does not behave in a similar manner.

Butera and Kofstad,⁽¹⁵⁾ in their investigation of the internal friction behaviour of hydrogen in vanadium, list the activation energy for diffusion. Further, they note enough information to allow the calculation of the diffusivity. Their results indicate

$$D_{H-V} = 1.21 \times 10^{-4} \exp \left(\frac{9,050}{RT} \right)$$

where the symbols have their usual significance. If this equation, which is valid at the temperature of the measurements (195°K), is used at higher temperatures, then the appropriate information for both hydrogen and nitrogen in vanadium is known. Using the classical $X^2 = 4Dt$, taking for D the value just below t_{mp} , and t the average time that a given section of crystal bar is molten, then the value of X obtained is greater than the radius of the crystal bar being zoned. Therefore, conventional diffusion of a single free interstitial atom cannot account for the difference between rotation and non-rotation. In addition, the usual approximation is to increase the value of D by at least an order of magnitude for diffusion in the liquid, which is particularly applicable for zone melting due to the minor turbulence which is always present.^(14,31) However, this even further weakens the case for conventional diffusion. The need for pedestal rod rotation to improve mixing in vanadium is of dubious advantage. The question remains, though, as to why the rotation results in harmful effects.

The mixing which does occur must be harmful. If the assumption is valid that the reduction of the oxygen concentration occurs by the decomposition of the oxides at the surface of the molten zone, then it is possible that the oxides are mixed into the molten zone, where their decomposition would not readily occur.

The effect of rotation during the hydrogenation experiments can be readily explained by noting that the results are basically

similar to those of the vacuum series of experiments. There is no appreciable difference between the resistance ratios of rotated and non-rotated samples in the vacuum series and the rotated and non-rotated hydrogen series of experiments.

The difference between the rotated and non-rotated nitrogen (ammonia) series of investigations is somewhat more difficult to explain. The compounding difficulty is to explain why the low temperature anneal resulted in the reduced resistance ratio of the non-rotated sample and the resistance ratio of the rotated sample remained the same. If the nitrogen enters the vanadium interstitially, then, from consideration of the diffusion process, rotation vs. non-rotation should have little effect. If the nitrogen reacts with the vanadium to form a known nitride, then the nitride could become mixed into the molten zone. However, the known nitrides of vanadium are supposed to decompose for temperatures above 800°C in vacua.⁽¹²⁾ Therefore if the nitride formed on the solid surface of the rod, then it should decompose as the molten zone approaches. Nitrogen complexes have been identified in Ta⁽³²⁾ and in Ta-Re alloys.⁽³³⁾ It is conceivable that such may also be the case for nitrogen in vanadium. The existence of complexes in vanadium is not new to the literature as evidenced by the investigations of Edington and Smallman,⁽³⁴⁾ but they attributed the complex to be an oxide, even though they could neither identify nor isolate it. Problematically though, the diffusion characteristics, again indicate an anomaly. The explanation of the greater impurity content with rotation requires the value of the diffusivity to be low, and to explain the significant decrease in the resistance ratio upon a short duration, low temperature anneal, a higher value for the diffusivity is required.

Other anomalous behaviour of nitrogen in vanadium has been noted in the literature. If the nitrogen content is high, it may be removed, obeying Sievert's law very nicely.⁽⁸⁾ Further, again for high concentrations, it will move during electromigration experiments.⁽²⁹⁾ The data of Stanley and Wert⁽³⁵⁾ indicates that

as the concentration is reduced, nitrogen ceases to move during electromigration and fails to obey Sievert's law. Horz has verified both cases^(36,37) and other investigators generally agree. (13,36)

The only seemingly plausible explanation for the results of the present investigation is that at low temperatures the nitrogen is present as an uncombined interstitial. A complex, either with other interstitials or with the vanadium, must exist at higher temperatures. To account for this, the free energy of formation of the complex must change quite drastically with temperature, to allow the stability of the low temperature free interstitial. It should also be noted that the same behaviour is found in the experiment was present in the investigation of Stanley and Wert, but no conclusions seem to have been drawn from it.

The highest purity vanadium, as determined by the largest resistance ratio, was obtained by electron beam melting a compacted rod. The carbon content was low due to the fact that there was no possible exposure to vacuum grease. The total pressures during zoning were higher than the previously mentioned experiments. This implies that for low pressures, the metallic vapor envelope surrounding the molten zone does afford complete protection^(16,28) and also may have the effect of locally reducing the partial pressure of oxygen below 1×10^{-13} torr which is needed to remove oxygen from molten vanadium. However, annealing in the same atmosphere as zoning contaminates the specimen, giving additional strength to the above conclusion, i.e., that the oxygen reduction occurs only as a result of the metallic vapor envelope surrounding the molten zone.

The reason for the increased resistance ratios which result from an increased number of zone passes is not clear. A plausible explanation is that a true zone refining effect is occurring since there is a decrease in the resistance ratio when going from the bottom to the top of a crystal bar, i.e., the direction of zone traversal.

Finally, the present value determined for the room temperature

resistivity is in good agreement with White and Woods.⁽²⁵⁾ That this agreement occurs inspite of the difference in the resistance ratios (280 vs. 7.4) is in keeping with the Debye temperature of vanadium.⁽³⁸⁾

Summary

This investigation demonstrates that it is possible to consistently electron beam zone melt vanadium at total pressures within the 10^{-10} torr range. One of the principal features of the system which makes this possible is the ability to bake the electrostatic pumps and the major part of the system at pressures below 10^{-6} by using the turbomolecular pump.

The experimental results presented are in general contrary to the existing literature concerning interstitial impurities in vanadium. The results clearly indicate that it is possible to reduce the oxygen concentration in vanadium by high vacuum electron beam zone melting. However, changing the total pressure during zone melting from 4×10^{-9} to 2×10^{-10} torr does not result in a corresponding decrease in the oxygen concentration.

A second major experimental finding which again is contrary to the existing literature is that rotation of the pedestal rod here results in an increase in the oxygen concentration and a decrease in the resistance ratio. The literature indicates that pedestal rod rotation should result in greater purification.

Yet another experimental finding which is contrary to the literature is that hydrogen or ammonia gas additions do not aid in purification. They actually result in a contamination of the crystal.

High temperature, high vacuum annealing in general results in an increase in the oxygen concentration and a decrease in the resistance ratio. It is therefore concluded that the removal of oxygen occurs by the decomposition of vanadium oxides at the surface of the molten zone, i.e., a metallic vapor envelope surrounds the molten zone and affectively reduces the partial pressure of oxygen below 1×10^{-13} torr. Since there is no ready

correlation between the resistance ratios and oxygen contents of the various samples tested, it must be concluded that other impurities are present within the crystals. Carbon is the most likely suspect because the internal friction results indicate that carbon is present in the samples. The presence of carbon indicates, again contrary to the literature, vanadium will become easily contaminated with carbon.

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Table I

Analyses of Vanadium Dendritic Crystalline Powder
Supplied by U. S. Bureau of Mines

	Lot A*	Lot B**	Lot C**
Oxygen	110 ⁺	28	18
Nitrogen	10		1
Carbon	20	10	12
Hydrogen	10	43	40
Chromium	<6	16	14
Silicon	<15	24	17
Magnesium	3		<3
Aluminum		36	7
Copper		<5	<5
Manganese		<5	5
Nickel			<3
Lithium			
Sodium			
Potassium			
Chlorides	~200	~200	~200

* Oxygen and hydrogen were determined by vacuum fusion analysis and nitrogen by the Kjeldahl method on an arc-melted button of the vanadium.

** Oxygen, hydrogen, and nitrogen were determined by vacuum fusion analysis on the dendritic crystals.

+ All figures in wt. ppm.

Table II
Vacuum Series

Experiment No.	1	2	3	4	5	6
Compacting Pressure (kpsi)	24	30	30	30	30	
Analysis of Arc-melted ingot						
Oxygen	175	141	141	141	141	153
Nitrogen	14	8	8	8	8	6
Pressure in EBFZR during 1st molten zone pass (torr)	6.0×10^{-8}	4.0×10^{-9}	4.0×10^{-9}	7.0×10^{-9}	7.0×10^{-9}	1.4×10^{-9}
Pedestal rod rotation speed (rpm)	0	0	0	45	45	0
No. of molten zone passes	4	4	4	4	8	3
Pressure in EBFZR during last molten zone pass (torr)	4.0×10^{-9}	3.4×10^{-10}	2.2×10^{-10}	5.5×10^{-10}	6.8×10^{-10}	7.0×10^{-10}
Crystal growth rate (cm/hr)	10	20	20	20	30	10
Analysis of zoned crystal						
Oxygen	46			46		35
Nitrogen						14
Carbon	30					43
Resistivity ratio as-zoned crystals	280	150	86	193	360	

Table III

Experiments No.	Gas Addition Series					
	7	8	9	10	11	12
Compacting Pressure (kpsi)	24	30	30	30	30	30
Analysis of Acr-melted ingot						
Oxygen	175	141	141	141	141	141
Nitrogen	14	8	8	8	8	8
Gas added during 1st pass	H ₂	NH ₃	NH ₃	H ₂	H ₂	NH ₃
Base pressure of system before introducing gas (torr)	6.0x10 ⁻⁸	8.0x10 ⁻⁹	8.0x10 ⁻⁹	1.5x10 ⁻⁹	1.5x10 ⁻⁹	8.0x10 ⁻⁹
Pressure during 1st molten zone pass (torr)*	1.4x10 ⁻³	8.0x10 ⁻⁵	8.0x10 ⁻⁵	6.0x10 ⁻⁵	6.0x10 ⁻⁵	8.0x10 ⁻⁵
Pedestal rod rotation speed (rpm)	0	0	0	30	30	45
Pressure during Beam power degassing	2.0x10 ⁻⁷	9.7x10 ⁻⁹	9.4x10 ⁻⁹	6.0x10 ⁻⁸	8.0x10 ⁻⁹	2.0x10 ⁻⁸
No. of further zone passes	3	3	1	3	3	3
Pressure in EBFZR during last zone pass (torr)	6.0x10 ⁻¹⁰	1.6x10 ⁻¹⁰	1.0x10 ⁻⁹	6.0x10 ⁻¹⁰	7.2x10 ⁻¹⁰	6.5x10 ⁻¹⁰
Crystal growth rate (cm/hr)	8	8	20	20	20	20
Analysis of zoned crystal						
Oxygen	49			68		
Nitrogen						
Resistivity ratio of as-zoned crystals	111	190	86	86	66	80

* Hydrogen pressures shown have been corrected for the ionization constant of the gauge.

Table IV

Annealing Results — Gas Addition Series

Experiment No.	8	10	12
Cold system total pressure before annealing (torr)	6.0×10^{-11}	3.8×10^{-10}	4.2×10^{-10}
Maximum pressure during annealing (torr)	8.0×10^{-9}	9.5×10^{-9}	9.0×10^{-9}
Total annealing time (hr)	2	1.25	1.5
Final total pressure (torr)	3.4×10^{-9}	1.0×10^{-9}	1.8×10^{-9}
Resistivity ratio of annealed crystals	125	160	80

Table V

Experiment No.	Arc-melted Powder Series		
	13	14	15
Analysis of Arc-melted ingot			
Oxygen	153	153	153
Nitrogen	6	6	6
Pressure in EBFZR during 1st molten zone pass (torr)	1.8×10^{-9}	1.7×10^{-8}	1.8×10^{-9}
No. of molten zone passes	3	3	6
Pressure in EBFZR during last molten zone pass (torr)	$<1.0 \times 10^{-9}$	$<1.0 \times 10^{-9}$	$<1.0 \times 10^{-9}$
Crystal growth rate (cm/hr)	10	10	10
Analysis of zoned crystals			
Oxygen	33	18	16
Nitrogen	4	250	
Resistivity ratio of as-zoned crystal		135	
Total annealing time (hr)	33	51	39
Pressure during annealing (torr)	2.5×10^{-9}	3.4×10^{-10}	3.4×10^{-10}
Analysis of annealed crystals			
Oxygen		58	48
Resistivity ratio of annealed crystals	149	100	392

Table VI
Zoned Compact Series

Experiments No.	16	17	18
Compacting Pressure (kpsi)	3.8	3.8	3.8
Pressure in EBFZR	5.5×10^{-8}	5.5×10^{-8}	5.5×10^{-8}
Analysis of Ingot			
Oxygen	89	89	89
Nitrogen			
Pressure in EBFZR during 1st molten zone pass (torr)	2.8×10^{-9}	9.0×10^{-9}	9.0×10^{-9}
No. of molten zone passes	2	5	5
Pressure in EBFZR during last molten zone pass (torr)	2.3×10^{-9}	1.3×10^{-9}	1.3×10^{-9}
Crystal growth rate (cm/hr)	16	13	13
Analysis of as-zoned crystals			
Oxygen	55		
Resistivity ratio of as-zoned crystals			
Total annealing time (hr)	0.75	15	15
Pressure during annealing (torr)	2.0×10^{-10}	2.7×10^{-10}	2.7×10^{-10}
Analysis of annealed crystals			
Oxygen		31	
Resistivity ratio of annealed crystals	342	655	232*

* Value determined by Dr. R. E. Reed of the Oak Ridge National Laboratory.

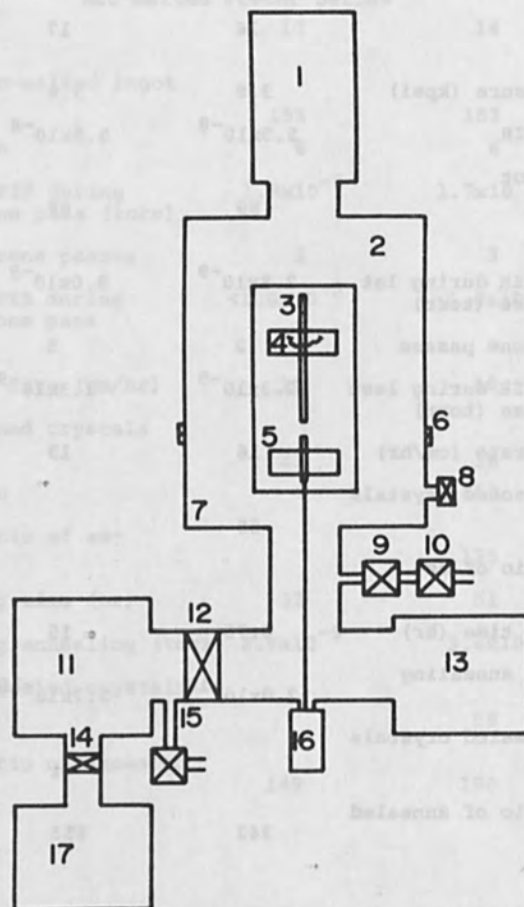
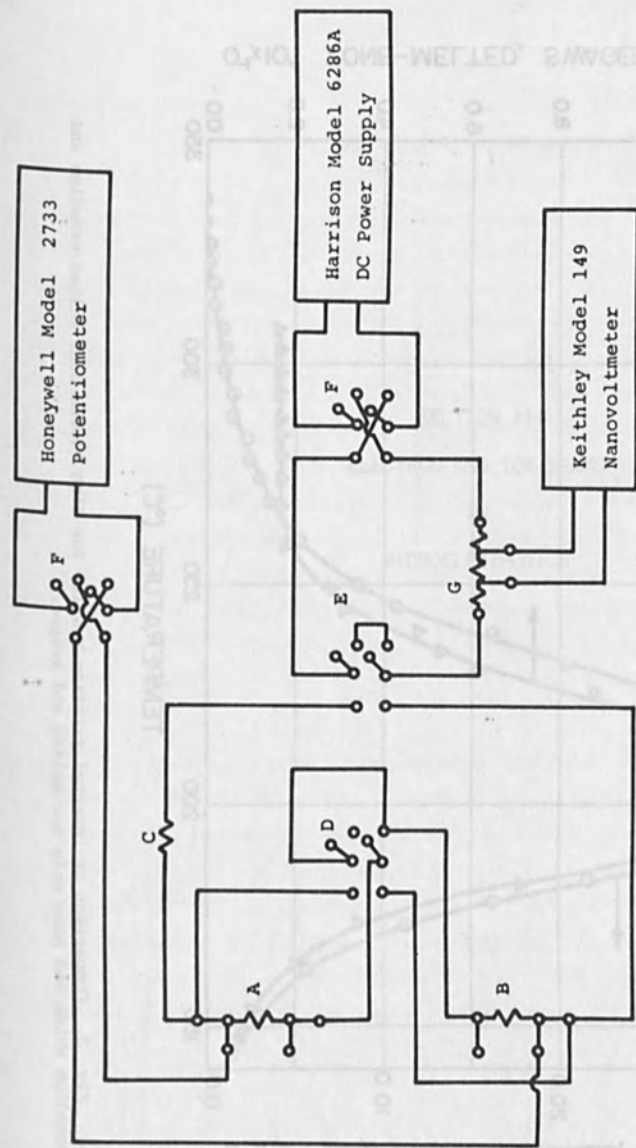


Fig. 1. Diagram of the electron beam floating zone refining system:
 (1) Orb-Ion Pump; (2) Chamber; (3) Rod to be zoned; (4) Filament travel block; (5) Pedestal rod travel block with rotation capability; (6) Wheeler flange on bell jar; (7) Bottom of bell jar (sump); (8) Ports for electrical feed-throughs, ion gauges, and monopole residual gas analyzer; (9) Bakeable valve; (10) Precision leak valve; (11) Turbo-molecular Pump; (12) Valve; (13) Vac Ion Pump; (14) Valve; (15) Valve to atmosphere; (16) Magnetic rotary feed-throughs for direct drive of filament travel block, pedestal rod travel block, and rotation; (17) Mechanical roughing pump.



- A, B Standard Resistors, 5a @ 50 ma
 C Circuit protection resistor
 D Switch for putting standard resistors in parallel or series
 E Switch for by-passing standard resistors
 F Switches to reverse polarity of current and potentiometer
 G Sample

Fig. 2. Circuit used for resistance ratio measurements.

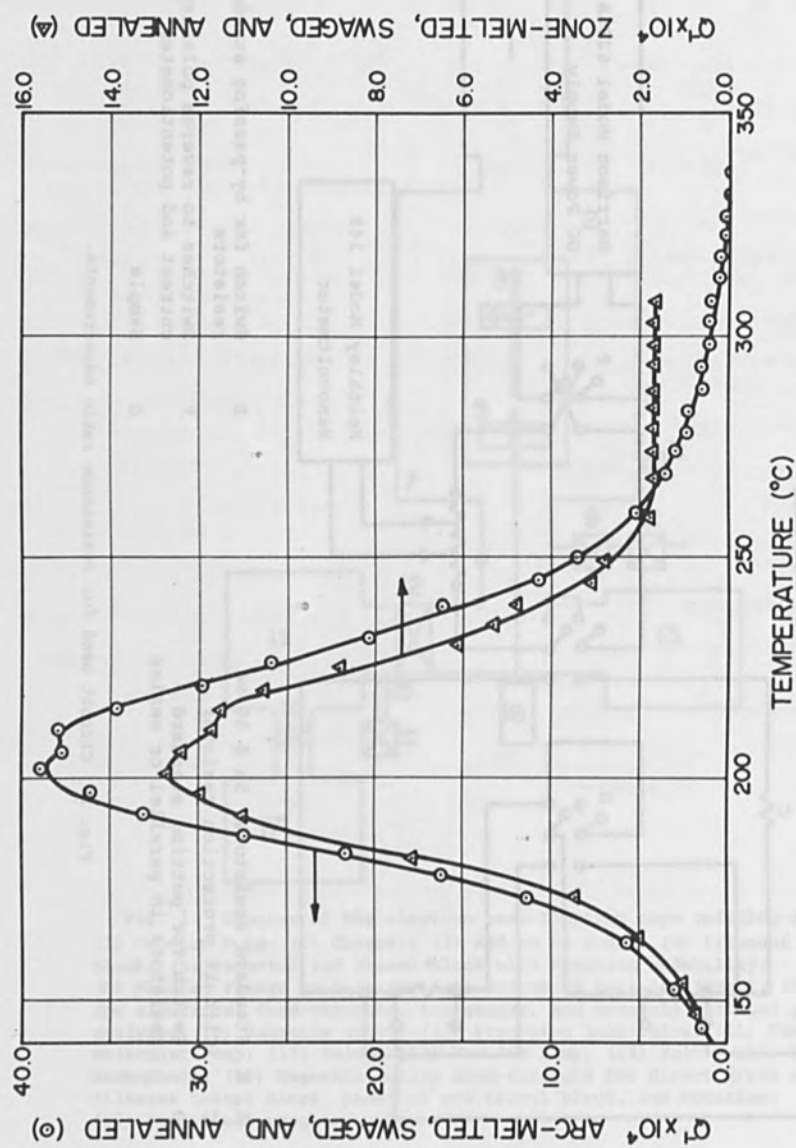


Fig. 3. Comparison of internal friction results for high vacuum zone melted vanadium and vanadium which had been only arc-melted and swaged.

A REVIEW OF ELECTRON AND ION BEAMS FOR MICROELECTRONICS APPLICATIONS

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Hughes Research Laboratories
Malibu, California 90265

INTRODUCTION

The use of electron and ion beams in the microelectronics field has grown rapidly over the past ten years. Fine-focused electron beams were first used for diagnostic tests on integrated circuits and semiconductor devices.¹ As these circuits began to be made to the resolution limit of photolithography, the far greater resolution capability of the electron beam was necessary in order to observe structure details. The ability of an electron beam to reveal qualitatively different types of information than are obtainable by optical methods has proved to be a valuable advantage.² The use of electron beams for implantation doping of

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ELECTRON AND ION BEAMS

IN

MICROELECTRONICS

semiconductors has received attention in recent years and is being used for making devices which are nearing the production stage. Now electron beams are being used in the laboratory for fabricating devices with resolution better than is possible by photolithography, and the focusing of ion beams to very small spots is being studied. Both of these latter techniques are intended as means for fabricating new classes of devices and circuits with superior operating characteristics.

In many areas the beam approach to fabrication of microelectronic devices offers advantages or unique capabilities compared with more contemporary techniques. Some of these advantages arise because of inherent limitations on the photolithographic process, e.g., such fundamental factors as diffraction and lens aberrations. Of course, charged particle beams are also subject to certain physical limitations which restrict their utility; e.g., the current that can be focused into a given spot or the size of the pattern which can be generated is limited. Even though beams of electrons can fabricate advanced devices which cannot be made any other

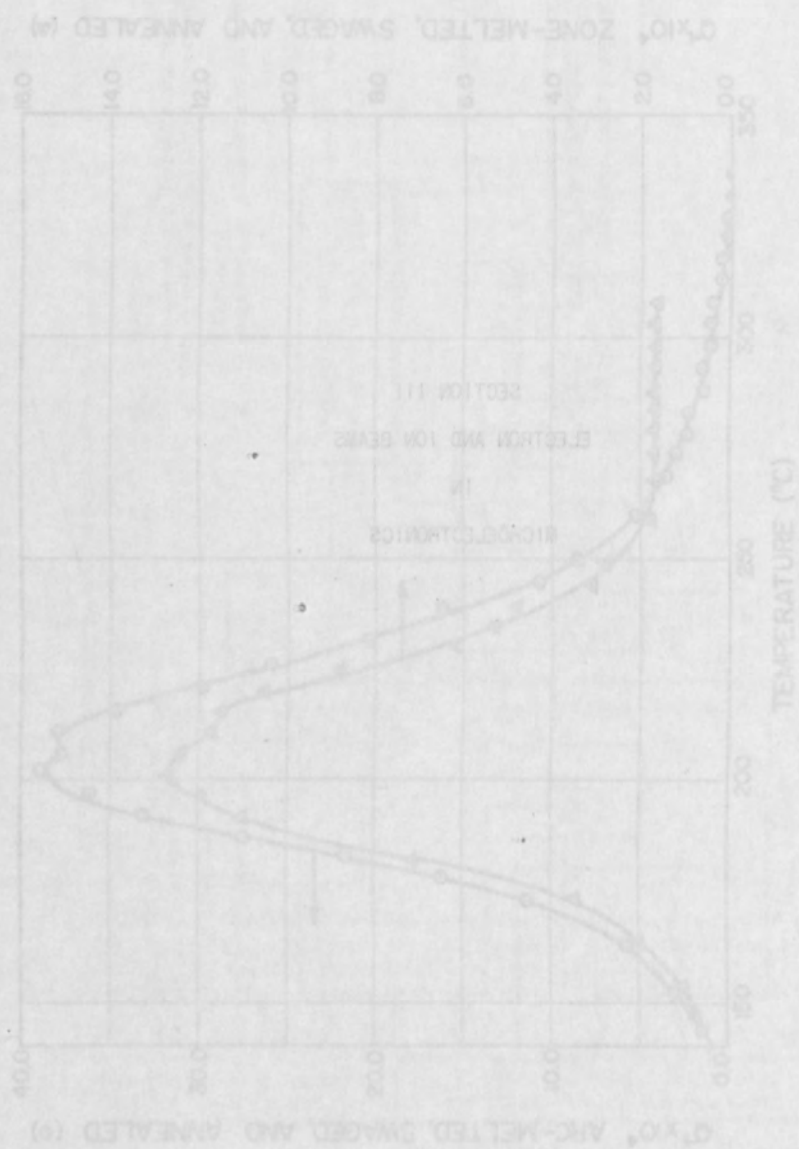


Fig. 4. Comparison of diffusion data for two different diffusion mechanisms: arc-melted, swaged, and annealed (A) and zone-melted, swaged, and annealed (Z). The data were only arc-melted and annealed.

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way, they do not form a magic panacea for the microelectronics industry. Therefore, in order for beams to be used most effectively, it is important to appreciate not only the advantages, but the limitations on their utility for microelectronics processes.

This paper presents a somewhat tutorial review of electron and ion beams as applied to the fabrication of microelectronic devices. It will show some of the types of devices and circuits which can now be fabricated with electron beams and thus indicate the state of the art in this field. The factors influencing the speed of electron beam fabrication (which determines the economics of the process) as well as the resolution attainable (and therefore the device performance gain) are given, as well as the trade off between these two important parameters. The limits on beam process performance by certain physical factors, such as lens aberrations, space charge, and transverse thermal velocities of the electrons are discussed. The use of electron beams as a diagnostic tool has been treated extensively and will not be covered here.

WAYS OF USING BEAMS

Some of the attractive features of electron and ion beams for use in microelectronics are summarized in Fig. 1. Electron beams have been focused to a spot size as small as $5 \text{ \AA}^4$; beams of $100 \text{ \AA}$ diameter are used routinely. Since these dimensions are a small fraction of the wavelength of light, it is clear that beams can "see" and make patterns with much higher resolution than are possible using light. The beams of charged particles can be programmed in intensity and position over wide ranges by electrical signals. In addition, electron and ion beams can interact with surfaces to produce qualitatively different effects than light beams. Electrons can induce carriers into semiconductors and thus locate junctions and other features accurately; the secondary electron emission, x rays, and cathodoluminescence process yields information in the surfaces not found from optical inspection. The injection of ions into surfaces as a means of doping semiconductors is qualitatively different from the diffusion process, yielding unique features to the semiconductor. If these ions

are focused to small programmable spots, it may be possible to carry out selective doping over a monolithic surface without the use of masks.

There are two general ways in which beams can be used to irradiate a surface and create a pattern. These methods involve parallel exposure of all pattern elements at the same time and sequential exposure of one pattern element at a time. The simplest parallel method is merely a flooding of a desired surface by a broad beam; an intercepting mask is interposed between beam and surface to create the desired pattern. A serious disadvantage of this technique arises from the fact that focused beams almost always have non-uniform current density distribution. Thus, only a small portion of the center of the beam can be used if a reasonably uniform density is required at the target; this is, of course, wasteful of beam current, so this technique is rarely used.

A second parallel exposure technique involves the use of a focused beam which is caused to pass through a transmission mask so as to create a demagnified image of the mask on the target.⁵ Since such a mask must be self-supporting, the type of patterns which can be created is limited (e.g., an annular ring cannot be exposed). A parallel exposure method which does not suffer from this latter restriction makes use of a cathode with emission areas made to define the pattern; this object is then imaged onto the target by an image orthocon-type lens system.⁶ A new cathode is made for each new pattern. The parallel exposure technique in general provides shorter pattern generation time (for a given area) than the use of sequential exposure, but it is not as flexible or rapidly programmable for different patterns. In the remainder of this paper we will emphasize the sequential exposure method.

A commonly used sequential beam technique involves focusing the beam to a small spot (diameter d_s) and scanning it over the target in a TV-raster fashion, as illustrated in Fig. 2. Quite uniform irradiation of the surface can be attained if two precautions are taken. First, the beam which will usually show a Gaussian distribution of current density with radius, must be scanned in such a way that the traces

overlap in order to produce a reasonably uniform irradiation. For one percent fine scale variation (between peaks and valleys) the center-to-center spacing should be about 1.5 times the spot radius and the minimum number of lines in a length L will then be about $1.3 L/d_s$. Second, the beam must be scanned over an area larger than the area of the target. This overscan is necessary since the tails of the Gaussian beam distribution will add so as to yield a greater beam irradiation over the center of the target than at the edges. If one percent large scale uniformity is desired, the beam should be over-scanned $\approx 1/N_L$ on each end of the horizontal and $\approx 2/N_L$ on the top and bottom.⁷ While this overscan is obviously not important with tiny beams, the use of 0.1 to 1 cm scanned ion beams for implantation doping requires this correction to attain the necessary uniformity.

The writing time (τ) and bandwidth (BW) can be related to the beam parameters by the equations:

$$\tau = \frac{SA}{I_b} G \quad \text{BW} = \frac{L^2 F}{2d_s^2}$$

The useful interaction of a beam at a surface can be discussed in terms of the number of electrons or ions deposited per second per unit area which is necessary to produce the desired effect. This "sensitivity" factor (S) is measured in coulombs/cm². For example, in order to implant the desired impurity atoms into a semiconductor, roughly 10^{15} ions must be injected per square centimeter of surface area; in this case $S = 10^{15} \times e$ ($e = 1.6 \times 10^{-19}$ coulombs, the electronic charge) or $S = 1.6 \times 10^{-4}$ C/cm². Consider a second example as the exposure of electron resist materials; approximately 5×10^{14} electrons/cm² are required to break the chemical bonds of a typical resist polymer so that it can be developed to the desired degree of contrast. In this case $S = 8 \times 10^{-5}$ C/cm². The smaller value of S , the more sensitive is the process, i.e., fewer charged particles are required to effect the desired surface interaction. In Fig. 2, A represents the area of surface to be irradiated by the beam of current I_b and G a geometrical factor

($G \approx 1.3$) to represent the overlapping* and overscan of the beam (the beam covers a larger area than A). The writing time τ will be discussed more later. The bandwidth is determined by the time required to turn the beam on and off in one beam diameter (to attain the resolution possible with a spot of diameter d_s). F represents the frame rate (i.e., number of complete frames/sec). By using typical numbers in this bandwidth equation, it can be shown easily that an excessive bandwidth (e.g., 400 MHz for $L = 1$ cm, $d_s = 2 \mu\text{m}$) is required with TV scan rates ($F = 30/\text{sec}$). Thus, to achieve good resolution (small d_s), a slow speed flying spot scanner must be used as the modulation technique.

The third method of using a beam for pattern generation is illustrated in Fig. 3. In this case the fine-focused beam is programmed by digital signals to expose selected areas of the desired pattern. Each pattern element is exposed by sequentially stepping the beam along a row, with sufficient number of rows used to make up the pattern element. The deflection current is a staircase with the beam blanked off between step levels. There are four units of time which influence the total writing time of this system. The path time from the last spot on one element to the first spot on the next element is related to the slewing rate of the scan coil and amplifier in the beam deflection system; it will be typically about a microsecond. The time required for a 16-bit address word from the computer to direct the beam to a new location will be about 2 microseconds, and the time required for the deflection current to settle to its next level value on the staircase (so that the beam can be turned on) will be about 2 microseconds. The fourth element of time is that required for the beam to deposit the requisite charge on the surface. It is clear that the actual exposure time per spot ($\tau_s = S/J_s$)

* The use of this factor (> 1) in the above equation suggests that longer writing time is required as a result of the overlapping scan pattern.

Actually, the exposure time will be less, since a greater exposure would be necessary without overlap in order to assure that the target regions exposed by the low density "tail" of the beam receive adequate current.

should be the limiting factor in writing speed; the surface process, deflection system, and computer should be designed as a unit so that the first three elements of time are small compared with the exposure time. It is noted that the bandwidth required for this means of writing will be one megacycle or less. The resolution attainable will be limited by either the digital-to-analog converter or the spot diameter, depending on the system design. The technique of beam writing illustrated in Fig. 3 also offers the advantage (over the raster scan method, Fig. 2) that the beam is scanned only over the areas where it will write.

RESOLUTION

Now let us look at the electron beam process from the points of view of resolution and writing time to see what can be achieved, then we will examine those factors which limit the resolution and speed. Two examples of the type of patterns which have been generated using the sequential digital scan technique are shown in Figs. 4 and 5. Figure 4⁸ shows the programmed electron beam used to expose a positive electron resist (polymethyl methacrylate) which is developed to yield a mask through which metal is evaporated or sputter deposited to yield the desired device. The device illustrated here is an interdigital transducer used to couple to a surface acoustic wave on a piezoelectric crystal at a frequency of about one gigahertz. The transducer is about 0.4 mm square, composed of fingers about 0.5 μm wide and 0.1 μm (1000 Å) thick. Similar patterns with finger width as narrow as 0.1 μm have been generated by a specially designed electron beam using the raster scan method, with a flying spot scanner input.^{9,10}

A somewhat more complex pattern, for a JFET microwave switch,¹¹ is shown as the second example in Fig. 5. In this device the source and drain are diffused into the epitaxial layer which acts as gate. The device conductance is increased by extending the source and drain electrodes by means of an array of interlocking fingers which are fabricated starting with electron beam exposure of resist which covers a gold layer. The developed resist is used as a mask against

ion beam sputtering to define the finger pattern in the gold, which is then used as a mask for implantation of the fingers into the epitaxial layer. The finger pattern and an enlargement (scanning electron microscope picture) of the fingers are shown in Fig. 5. The implanted fingers are about $1.3 \mu\text{m}$ wide, with an edge resolution of $0.1 - 0.2 \mu\text{m}$. A single device involves a finger pattern about 0.4 mm square, with 1180 fingers; the total beam-on time was 118 seconds.

While electron beams can produce fine patterns, with line width and edge resolution smaller than those possible by photolithography, the area of pattern which can be written is limited, and is related, as in the photographic process, to the resolution. The state of the art in these two quantities is shown in Fig. 6, where total outside pattern area is plotted vs resolution (defined here as the minimum line width), with several processes shown for comparison. The heavy solid line shows the area/resolution limit of photolithography due to diffraction and lens aberration.¹² Due to practical tolerances (registration, etching, undercut, diffraction from the optical mask etc), the practical optical limit is given approximately by the heavy dashed line. Several points showing the area and resolution which have been attained in laboratory samples of devices fabricated by electron beams are shown by the circles. In one case (marked W), the basic pattern was created by a scanned electron beam. This pattern was then reproduced by a step and repeat (S & R) process to result in a larger total pattern with only slightly degraded resolution.¹³

It is seen from Fig. 6 that electron beam patterns have been created with resolution which is better by about an order of magnitude than that of photolithography. It should be noted that the circles shown represent typical devices which have not been made with resolution near the ultimate limit of the electron beam process. A limiting factor in resolution of resist materials will be shown later. The resolution limits of collodian¹⁴ and hydrocarbon "contaminant" films¹⁴ are shown by the vertical lines so marked. The number of lines, representing the ratio of pattern height to line width, are shown for reference. This number is limited, in the sequential digital scan technique by the resolution of

the digital/analog converter; 10,000 lines should be possible using high quality converters. The graph in the upper left (plotted to the same abscissa scale as the larger graph) shows the beam current which can be focused into a given spot in two scanning electron microscopes. It is clear that as higher resolution is sought, lower current in the electron beam is available to carry out the fabrication task. This point will be discussed more fully later.

The direction of further technology development will be represented on this chart by movement upward (larger area) and to the left (better resolution). The electron optical factors limiting such progress are discussed later in this paper.

WRITING SPEED

The economics of a beam process will be determined partly by the time required to write the desired pattern; this time must be short enough that fabrication of complex, high resolution patterns can be carried out with the desired edge resolution (e.g., $\leq 0.1 \mu\text{m}$) in a reasonable time (e.g., a few minutes). It will be seen that the exposure times for reasonable patterns are, in general, short enough that cost is not the determining consideration but, rather, long term power supply stability will limit the maximum pattern writing time.

The exposure time per resolution element (i.e., per spot) was shown to be given by $\tau_s = S/J_s$. This time is plotted versus spot current density for several beam processes of interest in Fig. 7. It is seen first that the sensitivity of the beam-type processes varies over wide limits. It is seen that the exposure or process time per spot decreases inversely as the current density is increased. A simple consideration shows that a square pattern of n_s spots will have an edge resolution $R = 1/\sqrt{n_s}$. To attain $0.1 \mu\text{m}$ edge resolution with $1 \mu\text{m}$ line width requires about 10^2 spots per square; 6×10^3 such square areas (about the number in a typical surface wave transducer) requires 6×10^5 spots. In order to expose these areas in one minute using PMM resist, the spot current density must be about 0.1 A/cm^2 . Electron beam densities as high as 10^2 A/cm^2 have been focused into a spot with existing equipment. For ion beams, the corresponding value is $\approx 10 \text{ A/cm}^2$; these have been inert gas ions.

It could be inferred from the discussion above and the curves in Fig. 7 that one should always strive for the highest possible spot current density in designing an electron beam fabrication system. It may also be thought that the system should be operated so as to produce the smallest possible spot. In general these conclusions would be incorrect. There is no need to operate with a spot diameter smaller than the desired edge resolution; to do so increases the number of spots required to cover a given area and thereby the address and stepping times. Sharp lines have been achieved by using an electron beam spot which is about one fifth of the desired line width in PMM resist.

Perhaps of even more importance is the writing speed limit placed by the address and settling times. The sum of these two times per spot (note Fig. 3) is $4 \mu\text{sec}$; this level of time is drawn on Fig. 7 as a horizontal dashed line. It is clear that we will not gain much in writing time by increasing the current density above the $1\text{-}10 \text{ A/cm}^2$ level. That is, using the scan technique shown in Fig. 3, we cannot reduce τ_s below the dashed line.

The characteristics of the electron beam used to write the desired patterns are obviously an important element in this process. The state-of-the-art in focused electron beams is indicated in Fig. 8. This figure shows a plot of spot diameter versus beam current with some superposed light lines which will be discussed later. The heavy dashed lines are of interest here; they show the diameter/current characteristics of a commercial (upper) and a custom-built⁹ scanning electron microscope. The fine diagonal lines on the left of Fig. 8 are drawn at a 1:2 slope and therefore represent lines of constant current density. It is seen that in practice the attainable current density decreases somewhat as the spot size is reduced (i.e., the dashed lines have a lower slope than 1:2). It is seen in Fig. 8 that electron beams have been focused to about $25 \text{ \AA}$ diameter in an SEM and somewhat lower in electron optical systems using field emission.⁴

By combining the appropriate curve ($S = 6 \times 10^{-5} \text{ C/cm}^2$) of Fig. 7 with an I versus d curve of Fig. 8 we can determine the exposure time as a function of resolution. Such a curve is shown in Fig. 9.

This relation is determined in the following way. The writing time for one spot is $\tau_s = S/J_s$ while $J_s = 4I/\pi d_s^2$. In addition, the spot diameter d_s is related to the current I , from the lower SEM line on Fig. 8 as $I = 1.85 \times 10^{-5} d_s^{2.86}$ where d_s is measured in microns. Thus

$$\tau_s = \frac{S}{1.85 \times 10^{-5} d_s^{2.86}} \cdot \frac{\pi}{4} \times 10^{-8} \text{ sec}$$

This curve is plotted in Fig. 9. As an example, the area of a typical element (e.g., the fingers of an acoustic surface wave transducer as shown in Fig. 4) which may be drawn as part of an electron beam created pattern may be about 10^{-5} cm^2 . If the beam diameter is one fifth of the line width (i.e., about $0.1 \mu\text{m}$), a total of 10^6 spots will be required. From Fig. 9, $\tau_s \approx 2 \times 10^{-7} \text{ sec}$ so that the actual exposure time will be $\approx 0.2 \text{ sec}$. In this case however, the address and settling times (see Fig. 3) are $4 \mu\text{sec}$ per spot, or a total writing time of about 4 sec .

The principal point of Fig. 9 is to illustrate in principle the inevitable trade off between speed and resolution. The speed-resolution curve can be lowered by only two means, viz., higher spot current density or increased sensitivity (i.e., lower value of S). As shown by Fig. 8 the spot diameter is fixed principally by the beam current. Since great effort has been spent for many years to improve lenses (i.e., to reduce aberrations), and to increase the emission current density (J_c), it is unlikely that significant improvement in these parameters can be expected. Thus, higher writing speed than shown in Fig. 9 will probably come as a result of research on resist materials. However, unless the address and settling times are reduced correspondingly, lowering the solid line in Fig. 9 by these means will not reduce the total writing time. For other beam processes such as ion implantation doping, where the value of S is fixed by the doping density desired, once the I vs d relation is determined by the beam system design, nothing can be done to improve the speed/resolution tradeoff.

RESOLUTION LIMITS

One of the principal advantages of electron beams as a fabrication tool for microelectronic devices lies in the higher resolution attainable. This higher resolution of the written pattern allows devices to be made which will operate at higher frequency, with higher packing density, etc. The resolution however is dependent on the diameter to which the electron beam can be focused. We saw that the process or writing speed is dependent on the current density in the beam. There are a number of physical effects which limit the attainable spot size and current in a beam of charged particles. These effects include lens aberrations and transverse thermal velocities. There are other effects which limit the size of pattern that can be created (deflection distortions) and which restrict the minimum resolution of pattern (electron scattering in the resist). These problems will be discussed briefly.

It is well known that the electrons of a beam impinging onto a resist material will be scattered through large angles by the atoms of the resist. These scattered electrons will expose the resist through a roughly spherical volume which is larger in diameter than the beam spot diameter. This effect is illustrated in Fig. 10 which shows an SEM photomicrograph¹⁵ of a cleaved layer of thick electron resist (PMM) into which a 500 Å electron beam has been scanned perpendicular to the plane of the photograph. It is seen that the 500 Å beam produced a neck at the surface which is about 2000 Å wide and a cylinder of diameter about 4500 Å, about nine times the diameter of the beam. Since the resist is usually used in a thickness of about 0.2 to 0.5 μm, the exposed line will actually be about 2000 Å wide, or four times the beam diameter. While Fig. 10 shows an extreme example, chosen to illustrate the effect, electron scattering does cause a serious limitation in attainable resolution, relatively independent of beam diameter. Since the exposed hole radius R_e varies with voltage approximately as $V^{1.75}$, it would seem that lower voltage will yield better resolution. However, the lower voltage results in an increase in the electron beam spot diameter. Despite this scattering problem, electron-beam-exposed resist materials have been used to create lines as narrow as 0.1 μm.

The spot current density (J_s) is of course related to the total current in the beam and the spot diameter at the target. The diameter of an undeflected spot is limited by three principal effects, viz., the spherical and chromatic aberration of the focusing lenses and the transverse thermal velocity of the particles. Each of these effects can be considered to give rise to a minimum value of beam diameter, usually called the disc of least confusion. Typical electron trajectories drawn to illustrate these two forms of aberration are shown in Fig. 11 and 12. These effects are treated extensively in the literature on electron optics. Briefly, spherical aberration results from the fact that the focusing fields of a lens are almost always stronger at the outside of the beam than on the axis; electrons entering the lens at different radii are therefore directed toward a focus at different points along the axis. Chromatic aberration refers to the sensitivity of the lens to particle energy; since the electrons always have a spread in energy (due to thermal velocity of emission, power supply ripple etc.), this effect will also defocus the beam.

The electric (or magnetic) fields which are used to deflect an electron or ion beam will cause a distortion of the initially round beam into a non-circular spot; further, even though a linear (with time) deflecting voltage is applied to the deflecting system, the pattern written on the target may not be a faithful reproduction of the input signals. These effects are illustrated in Fig. 13. The deflected spots can be distorted into rather grotesque shapes; those illustrated are typical of coma, a defect typical of off-axis deflecting fields. Even a beam deflected in one direction will exhibit a non-circular spot, due to astigmatism. As electrons are deflected through different angles they will be brought to a focus at different distances from the gun, as illustrated in Fig. 13; thus, the spot size seen on the screen will change with deflection angle. If deflecting voltage are applied so as to create a rectangular pattern on the screen, this pattern will in general appear distorted, as illustrated in Fig. 13. While the area which can be scanned by an electron beam without excessive distortion is an important quantity, unfortunately, very little data on deflection distortion is available.

Figure 14 shows experimental data¹⁶ on an electron beam in a scanning electron microscope with and without distortion correction; the dashed line is calculated from the simple theory of magnetic deflection distortion. It is seen that, with corrections, a distance of almost 1 mm each side of center can be scanned before the beam diameter doubles (an extreme case). For the 0.5 μm spot, this corresponds to total deflection of about 4000 spot diameters.

In addition to beam distorting effects caused by the electric or magnetic fields used to focus or deflect beams, the beams themselves contain, in a sense, the cause or two other forms of distortion. The first of these is the beam expansion resulting from the beam charges. The electrons (or ions) repel each other by the well-known inverse-square law. Thus, an initially convergent beam will reach a minimum radius (not zero) because at some radius the space charge expansion force causes the electrons to expand from the axis. The space charge relation is plotted as the almost-vertical lines on the right side of Fig. 8. It is seen that this space charge expansion effect is usually not the limiting factor in determining spot size, since the current values are too low and the beam voltage too high in those types of beams of interest for the applications discussed here.

A second property of the charged particles in a beam which does (usually) cause a first-order effect on both spot size and current density, is the transverse random component of velocity due to the emission of the particles from a hot surface. Electrons or ions emitted from a solid surface or from a plasma sheath at temperature T_c will in general possess a Maxwellian velocity distribution $f(v_r) = k \exp(-mv_r^2/2kT_c)$. In passing through the subsequent acceleration and focusing systems, this transverse velocity distribution is transformed into a Gaussian distribution of current density in the focused spot. If the convergence angle of the beam toward this focus is denoted by θ , it can be shown¹⁷⁻¹⁹ that the maximum possible current density J_m at this spot is related to the emitter current density J_c by the following equation, which represents the Langmuir limit on current density:

$$J_m \approx J_c \frac{11600 V_o \theta^2}{T_c} = \frac{4}{\pi} \frac{I_b}{d_g^2}$$

where d_g is the "Gaussian" spot diameter, i. e., the diameter of the spot is limited by this transverse thermal velocity effect.

The equations relating the diameter to the beam and lens parameters due to these three effects are:

$$d_s = \frac{1}{2} C_s \theta^3 \quad (\text{spherical aberration})$$

$$d_c = C_c \frac{\Delta V}{V} \theta \quad (\text{chromatic aberration})$$

$$d_d = \frac{15}{\sqrt{V} \theta} \quad (\text{\AA}) \quad (\text{diffraction})$$

$$d_g = \frac{1}{\theta} \sqrt{\frac{I_b}{\frac{\pi}{4} F J_c \left(\frac{11600}{T_c} \right) V_o}}$$

where C_s and C_c are the spherical and chromatic aberration coefficients, respectively, θ is the half-angle of convergence of the beam at the target, I_b and V_o are the beam current and voltage at the target, and J_s and T_s are the current density and equivalent temperature of the electrons from the source. The term d_d refers to the minimum spot diameter as limited by diffraction in the aperture. F is a factor determined by the definition of the diameter of a beam with Gaussian density distribution; if 80% of the current is to be enclosed within a circle of diameter d_g , $F = 0.62$.

Since the independent parameter in these equations is θ , it is useful to plot the total beam diameter versus θ . Beam diameter is taken as the RMS sum of these four quantities. The left graph in Fig. 15 shows such a plot for the electron beam in a scanning electron microscope.²⁰ The values of the parameters used in the calculations of the curves are shown; they are typical of modern commercial scanning electron microscopes. It is seen that the principal effects influencing

spot size are the transverse thermal velocities (i.e., d_g , dependent on beam current) and spherical aberration (d_s). The effects of chromatic aberration (d_c) and diffraction (d_d) are relatively insignificant. This graph predicts a spot size 200 Å at a current of 10^{-11} amps, which is about that shown by the upper dashed curve in Fig. 8 (which was based on experimental data).

The data given above has shown that electron beams can be focused to usable (for fabrication) diameters as low as 200 Å or so, and programmed with tolerable distortion over a 0.2×0.2 cm area. Line width in the resist can be $0.1 \mu\text{m}$ or less.³

Ion beams can also be focused to small spots and deflected over a target surface. If ion beams can be generated and focused to a small enough spot while carrying sufficient current, they can be used in a programmed manner to achieve ion implantation doping without the use of masks. An indication of the expected spot diameter can be obtained in the same way as for the electron beam, as shown in the right hand graph in Fig. 15.

The parameters chosen for the ion beam plot are indicated in Fig. 15. The spherical aberration coefficient was chosen as five times the value used in the electron beam case (i.e., $C_s = Kf$, $K = 10$); this value of K is representative²¹ of the quality of the best electrostatic lenses (which are required to focus ions). The value of the chromatic aberration coefficient was chosen for the same reason.²² Admittedly, a somewhat more realistic value of C_s would be ≈ 100 cm; this value would increase the minimum ion spot diameter to about $1 \mu\text{m}$ for $I = 10^{-9}$ A. The source current density J_s was assumed to be 0.1 A/cm^2 , which is compatible with higher current density ion sources (e.g., duoplasmatrons) but is high for present day lower-energy electron impact ion sources. The 50 kV ion energy was chosen as representative of medium mass ions for simple present applications. These current density and aberration parameters are difficult, but not impossible, to achieve in practice. A minimum spot diameter around $1 \mu\text{m}$ is predicted for realistic values of ion beam current (10^{-9} to 10^{-8} A). Through the use of higher voltage or lower beam

current, smaller spot sizes would be predicted. It is seen that the effect of chromatic aberration is not important in the case studied here and that the diffraction limit (d_d in the electron beam chart) will not appear in the ion beam sizes desired.

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RESOLUTION	- spot size $\ll$ wavelength of light ($\sim 100 \text{ \AA}$ vs $\sim 5000 \text{ \AA}$)
DEFLECTABLE SPOT	- beams positioned electrically - computer control - - deflection ~ 4000 spot diameters vs ~ 300 for laser - - complex patterns, maskless processes
USEFUL INTERACTIONS AT SURFACES	
ELECTRONS	- surface inspection using secondary electrons, cathodoluminescence, induced currents in semiconductors - chemical reactions, electron resist exposure - penetration up to $\sim 10 \mu$
IONS	- semiconductor doping - material deposition and removal by sputtering

Fig. 1. Summary of the principal features of beams which make them attractive for fabrication or diagnostic uses in microelectronics.

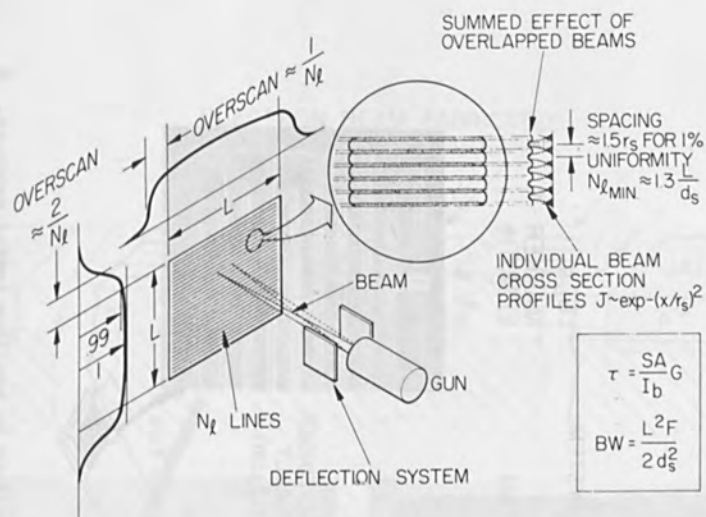


Fig. 2. Illustration of the raster scan technique of writing patterns with a focused beam. The degree of overscan and line overlap necessary to achieve uniformity of beam irradiation is indicated.

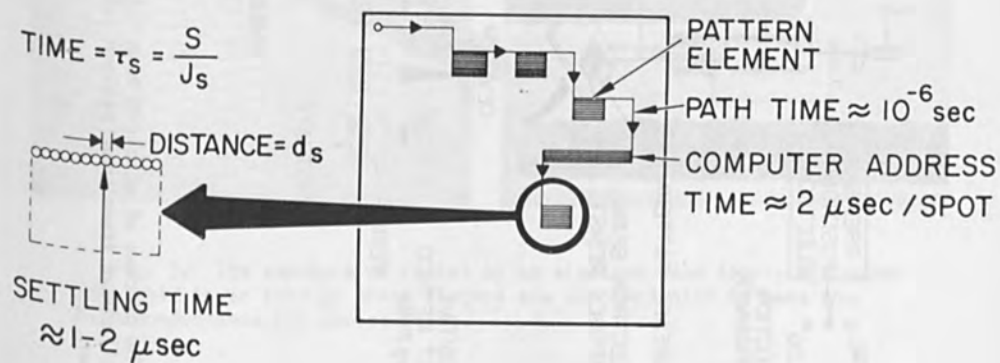


Fig. 3. Illustration of the sequential scan techniques of writing patterns with a focused beam controlled to move stepwise by a digital input signal. The exposure time and the several address and path slewing times are illustrated.

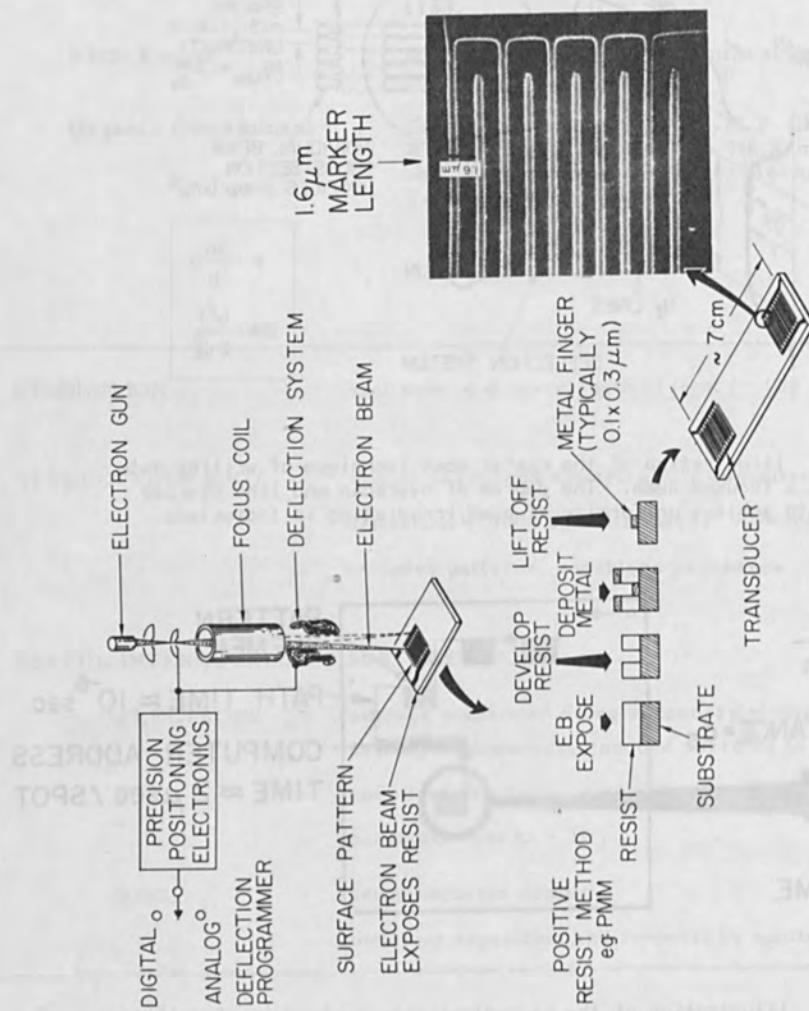


Fig. 4. The process of electron beam fabrication of a pattern by positive resist exposure. A surface acoustic wave transducer pattern fabricated by this technique for operation at approximately 1 GHz is shown.

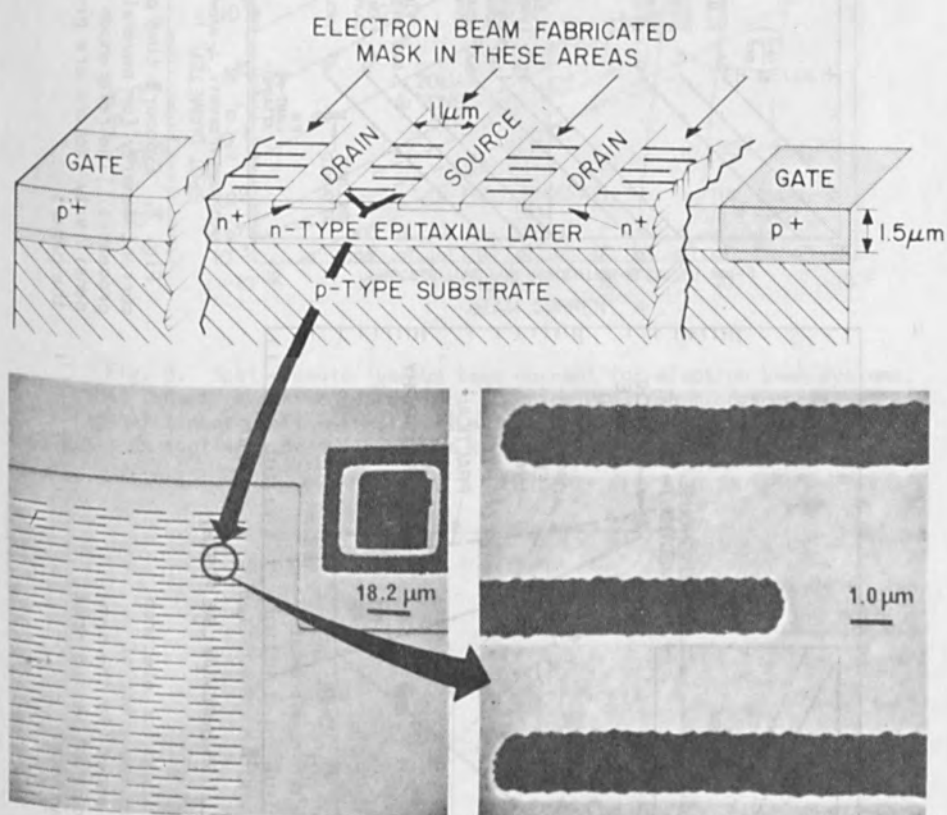


Fig. 5. The exposure of resist by an electron beam to yield windows in a gold layer through which fingers are ion implanted to make the high conductance FET device shown.

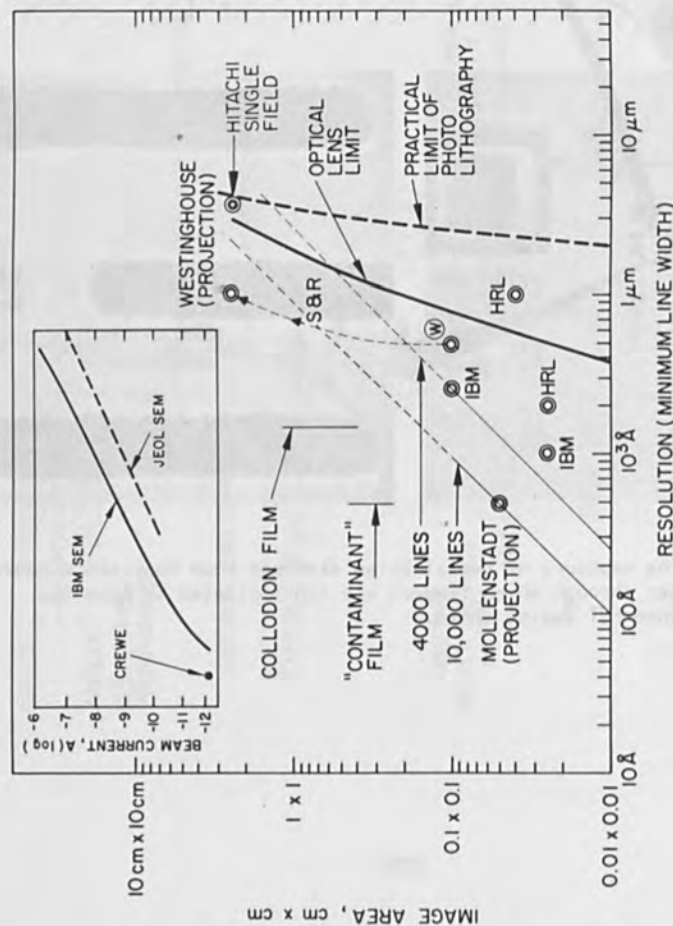


Fig. 6. Image area versus resolution for electron beam exposed patterns, compared with the limit of photography. The upper left graph shows (to the same abscissa scale) the electron beam current versus spot diameter for three electron beam systems.

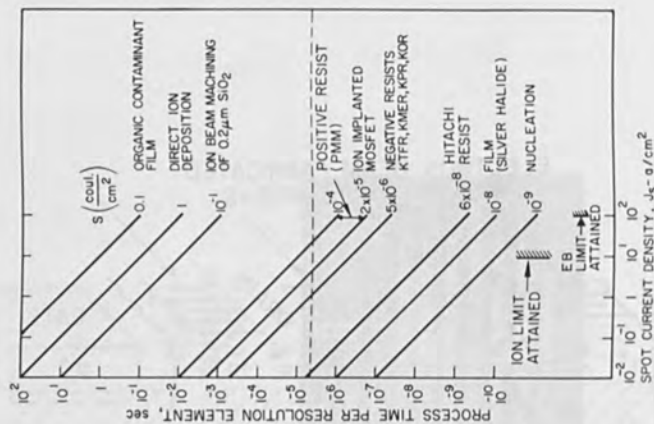


Fig. 7. Exposure time per spot versus current density for several different beam processes. The references for these sensitivity factors are given at the end of the paper.

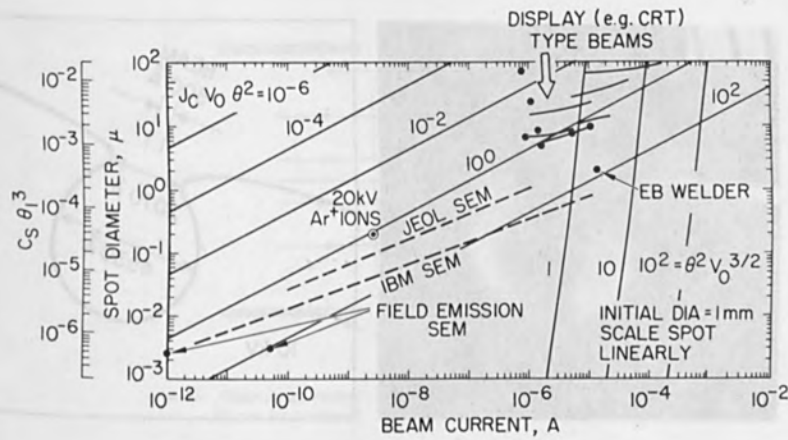


Fig. 8. Spot diameter versus beam current for electron beam systems. Dashed lines: experimental data on scanning electron microscopes. Diagonal lines: left = limit by transverse thermal velocities; right = limit on spot size by space charge effects.

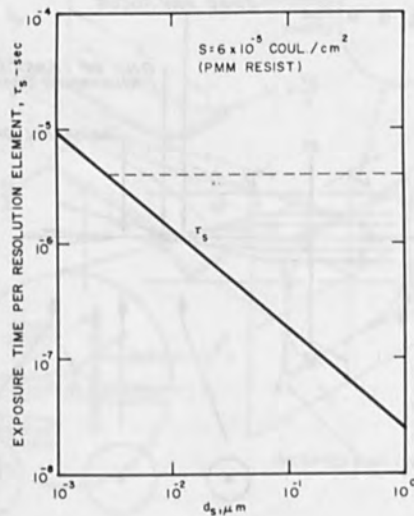


Fig. 9. Tradeoff between exposure time and resolution, from the curves of Figures 7 and 8. See text for details.

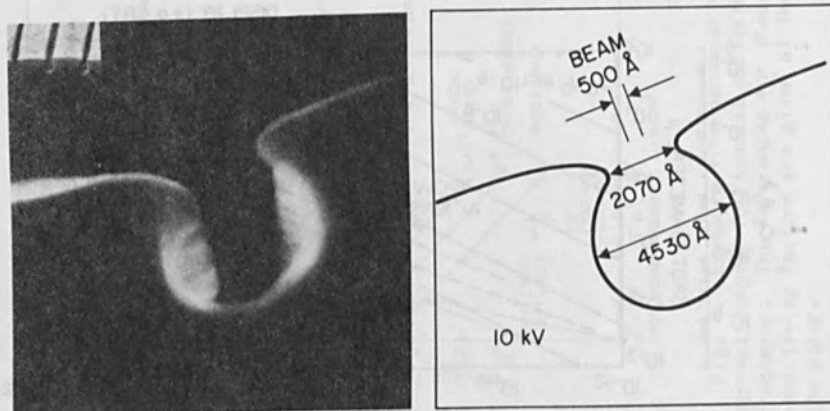


Fig. 10. Electron beam spreading in resist material creates an exposed volume much larger than the beam diameter, as shown here. An electron beam was scanned across a thick resist, as shown in the drawing, to create the cylindrical volume.

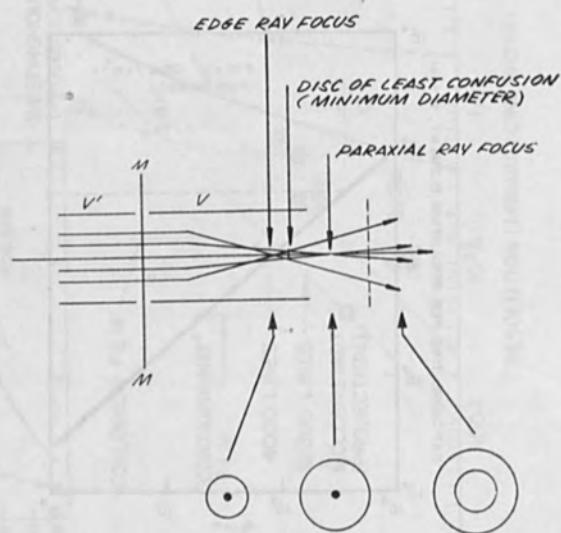


Fig. 11. Illustration of electron/ion rays exhibiting the effects of spherical aberration from a lens.

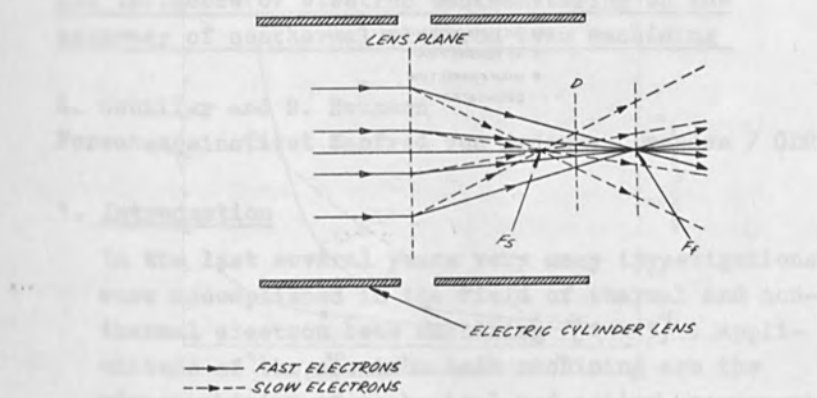


Fig. 12. Illustration of the electron/ion rays exhibiting the effects of chromatic aberration from a lens.

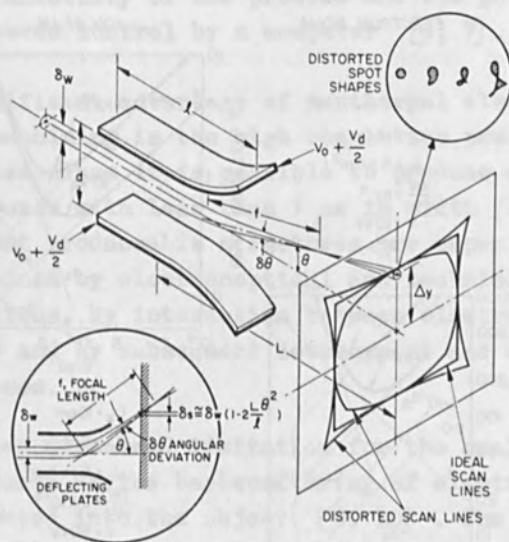


Fig. 13. The several forms of distortion of the spot and the pattern due to beam deflection. The inset shows the change in focal length of the deflection system with deflection angle.

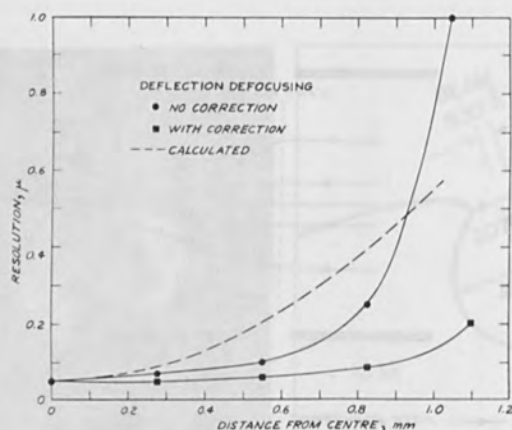


Fig. 14. Measured spot size versus deflection distance in an SEM with and without deflection distortion correction (from Chang and Stewart). The dashed line is calculated from magnetic deflection theory for the parameters used in the experiments.

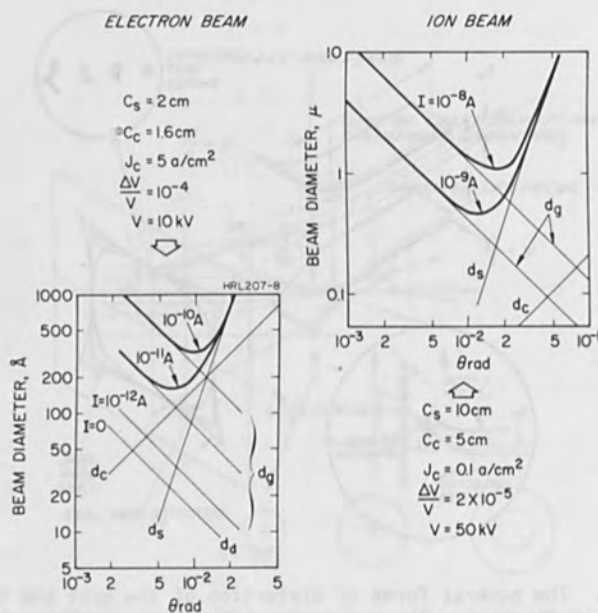


Fig. 15. Calculated spot diameter versus convergence angle for an electron beam in an SEM (left) and an ion beam (right). These curves illustrate the importance of the several types of aberrations present in beams and lenses.

The influence of electron backscattering on the
accuracy of nonthermal electron beam machining

S. Schiller and M. Neumann

Forschungsinstitut Manfred von Ardenne, Dresden / GDR

1. Introduction

In the last several years very many investigations were accomplished in the field of thermal and non-thermal electron beam machining [1 - 5]. Applications of the electron beam machining are the micromachining of mechanical and optical components and the production of microelectronics. Advantages compared to other methods of micromachining such as photolithography and laser beam machining are the flexibility of the process and the possibility of process control by a computer [6, 7].

A significant advantage of nonthermal electron beam machining is the high resolution power. By this technique it is possible to produce microstructures with less than $1\text{ }\mu\text{m}$ in width [8]. The smallest produceable structures are especially determined by electronoptical and technical conditions, by interaction between electrons and object and by subsequent development and etch processes.

An often discussed limitation for the smallest structures is the backscattering of electrons penetrated into the object [9, 10]. The backscattered electrons emerge from the object surface in the environment of the primarily irradiated point and produce a widening of structures.

To estimate the widening of structures produced by the backscattered electrons the local distribution of backscattered electrons at the surface of the irradiated object must be known.

The knowledge of the diameter of scattering circle alone is not sufficient for the determination of structure widening [9, 11]. In the following therefore the distribution of backscattered electrons is calculated by a simple backscattering model [12]. Then the results are verified by experiments and the conditions are formulated, on which a widening of structures by backscattered electrons can be avoided.

2. Calculation of electron backscattering

2.1. Current density distribution of backscattered electrons for irradiation by a point-shaped electron probe

The simplest case of electron irradiation is the irradiation of a point by parallel incident electrons. The current density distribution of backscattered electrons at the surface was calculated according to EVERHART [12] on the following conditions (comp. Fig. 1):

- The incident electrons of energy E_0 are retarded to the energy E penetrating the distance x into the object. The energy E is given by the THOMSON-WHIDDINGTON-law [12] :

$$E^2 = E_0^2 - c_1 \cdot \rho \cdot x \quad (1)$$

where c_1 is a constant and ρ is the density

of substrate material. The maximum penetration depth $x = x_r$ (comp. [13]) is given for a deceleration to the energy $E = 0$:

$$x_r = \frac{1}{c_1} \cdot \frac{E_0^2}{f} \quad (2)$$

-In each penetration depth

$$x = k \cdot x_r; \quad 0 \leq k \leq 1 \quad (3)$$

electrons are scattered at atoms of the object. The probability W of deflection of an electron about the angle Φ is

$$W(x, \Phi) = \frac{c_2}{E^2} \cdot \left(\frac{1}{\sin \Phi/2} \right)^4 \quad (4)$$

according to RUTHERFORD's scattering formula [12], where E is the energy in the depth x and c_2 is a constant.

- The electrons deflected about $\Phi > \frac{\pi}{2}$ emerge from surface of the object without new scattering [12]. This assumption of single scattering is valid for object materials of small atomic numbers only [14]. For higher atomic numbers the probability of scattering increases, so as less electrons are back-scattered than it is calculated assuming single scattering.

Under these presumptions the part of current between $x = k \cdot x_r$ and $x + dx = (k + dk) \cdot x_r$ scattered in the steradian $2\pi \sin \Theta d\Theta$ amounts (comp. Fig. 1) [12]:

$$d^2 I(k, \Theta) = I_0 \frac{n}{4\pi} (1 - k)^{n-1} dk \frac{2\pi \sin \Theta d\Theta}{\cos(\Theta/2)^4} \quad (5)$$

where I_0 is the electron current impinging

on the surface of object. According to ARCHARD [14] for n is to set in :

$$n = 0,041 Z, \quad (5a)$$

where Z is the atomic number of object material.

The current density of backscattered electrons in the distance r from impact point is given by division of the current part $d^2I(k, \Theta)$ by the area segment $dF = 2\pi r \cdot dr$ and by subsequent integration over all substrate depths $x = k \cdot x_r$ [11] :

$$j = \frac{I_0}{x_r^2} \int_{k=0}^{k_g} \frac{n}{\pi} \cdot \frac{(1-k)^{n-1}}{k^2} \cdot \frac{\cos^3 \Theta}{(\cos \Theta + 1)^2} \cdot dk \quad (6)$$

The integration limit k_g is determined thereby, that the total electron path in the object may not be longer than the range x_r of the electrons.

The result of numerical integration is presented in Fig. 2. From this display the current density j of backscattered electrons in the distance r from impact point of a point-shaped electron probe can be drawn, if the probe current I_0 , the atomic number Z of the object material and the range x_r [13] of electrons in the object are known.

By integration over the current densities j presented in Fig. 2 the total backscattered electron current can be obtained

$$I_r = \int_{r=0}^{\infty} j \cdot 2\pi r \cdot dr \quad (7)$$

The backscattered coefficients calculated

on this way $R_k = \frac{I_R}{I_0}$ (7a)

differ less than 10% from the measuring values of several authors [11] up to atomic numbers $Z \leq 40$. At higher atomic numbers the backscattering coefficients obtained by this calculation are too high, because the presumptions of single scattering are not valid [12,14]. In this case it is better to use another backscattering model postulating the complete diffusion of electrons in the object [11,14].

For further estimations the backscattered current densities calculated on the base of single scattering ($Z < 40$) are used. So the most practically interesting substrat materials are included.

2.2. Charge density distribution of backscattered electrons at deflection of a point-shaped electron probe

Practically electron beam machining mostly is realized by deflection of an electron probe along a straight line. The widening of line structures produced thereby is determined by the charge density distribution of backscattered electrons. This charge density distribution was calculated under following presumptions: on a plane object surface with the coordinate directions y and z a point shaped electron probe with the probe current I_0 is deflected.

The deflection takes place along the straight line $y = 0$ with the velocity

$$v = \frac{dz}{dt} . \quad (8)$$

The charge density $q(y)$ backscattered at a point with the distance y from the straight line $y = 0$ is given by integration of the current density j backscattered at this point during deflection of electron probe:

$$q(y) = \int_{-\infty}^{+\infty} j(y, t) \cdot dt = \int_{-\infty}^{+\infty} j(y, z) \cdot \frac{1}{v} \cdot dz \quad (9)$$

By setting in the current density values of Fig. 2 and by numerical integration the charge density distribution presented in Fig. 3 is given [11] .

According to Fig. 3 the widening of a line-shaped machining structure by backscattered electrons can be estimated.

When - q - is the critical charge density of backscattered electrons necessarily for a machining effect,

- I_0 - is the probe current of the point-shaped electron probe,

- v - is the velocity of deflection of the electron probe,

- z - is the atomic number of substrat material and

- x_r - is the range of electrons in the substrate [13] ,

than $2y$ is the width of the machining

structure conditioned on electron backscattering. This estimation also is valid for finite probe diameters, if the probe diameter remains very small compared with the range x_r .

2.3. Charge density ratio of backscattered to impinging electrons

According to Fig. 3a a widening of structures by backscattered electrons can be avoided completely, if the term $q \cdot x_r \cdot v \cdot I_0^{-1}$ is greater than 2. At irradiation by a deflected electron probe of limited size or by an electron image a widening of structures can be avoided under following assumptions:

- The area to be machined is irradiated by a constant charge density q_e , which is greater than the critical charge density q_1 necessarily for obtaining a machining effect.
- At the edge of the machining structure the charge density q_e of impinging electrons decreases abruptly to the value nought.
- The charge density q_r of backscattered electrons at the edge of machining structure must be lower than the critical charge density q_1 , so that no machining results on the non-irradiated area.

So the following postulate is given:

$$q_e > q_1 > q_r; \quad \frac{q_r}{q_e} < 1 \quad (10)$$

The charge density ration $q_r : q_e$ is also a criterion therefore, whether a widening of structures can be avoided under the called conditions.

For the case of irradiation of a (infinitely) long stripe of width b by a constant charge density q_e the charge density ratio $q_r : q_e$ was calculated along the stripe center. For the calculation of backscattered charge density q_r it was assumed, that irradiation of the long stripe takes place by scanning with a very fine electron probe in the direction of stripes. Then the complete backscattered charge density in the stripe center results from superposition of backscattered charge densities for single overspreads presented in Fig. 3. If the ordinate values of Fig. 3 are designated with $f(\frac{y}{x_r}, z)$, then the charge density ratio in the center of a (infinitely) long stripe of width b is given by [1] :

$$\frac{q_r}{q_e} = \int_{-\frac{1}{2} \frac{b}{x_r}}^{\frac{1}{2} \frac{b}{x_r}} f\left(\frac{y}{x_r}, z\right) \cdot d\left(\frac{y}{x_r}\right) \quad (11)$$

The result of numerical integration is displayed in Fig. 4. To it can be noticed the following :

- For stripe widths $b \ll x_r$ the charge density ratio $q_r : q_e$ increases about

linearly with the stripe width b . That is conditioned thereby, that the width of irradiation b increases while the width of backscattering stays nearly constantly for $b \ll x_r$ and is about equally the range x_r (Fig. 3).

- For $b \geq 2x_r$ the charge density ratio $q_r : q_e$ reaches a saturation value, because only those electrons can contribute to the backscattered charge density in stripe center, which impinges in a distance less than x_r to the stripe center. Thus there the same charge density ratio is obtained independently, whether the stripe width b is x_r or infinitely great.
- For uniform irradiation of an infinitely great area the charge density ratio $\frac{q_r}{q_e}$ is according to definition equal the backscattering coefficient R_k (equation (7)). The good conformance of saturation values in Fig. 4 with experimentally obtained backscattering values [14] for $Z \leq 50$ warrents the assumptions of part 2.1. and confirms the correctness of calculations.
- A little charge density ratio $q_r : q_e$ can be obtained according to Fig. 4 for substrate materials with a small atomic number Z , for small widths b of structure and for large ranges x_r , i.e. according to equation (2) for a large electron energy E_0 and for small substrate densities .
- The charge density ratio $\frac{q_r}{q_e}$ is smaller

at the edge of stripe than in the stripe center. For large stripe breadths ($b \geq 2 x_p$) the charge density ratio at the edge is a half of that in the stripe center, because at the edge the electrons are backscattered only from one side and in the center from both sides.

3. Experimental verification of calculations

The charge density of backscattered electrons was measured by coating the substrate surface with an electron sensitive photoresist layer. By irradiation with a minimum charge density q_0 this photoresist is not dissolved during a following development process. This minimum charge density q_0 is a function of electron energy E [11].

At first the charge density distribution of backscattered electrons at deflection of a nearly point-shaped electron probe (Fig. 3) calculated in part 2.2. was verified. For this purpose an electron probe with a diameter of about $1 \mu m$ (probe current $I_0 \approx 5 \cdot 10^{-9} A$) was deflected with different velocities v over a lacquer coated silicon substrate ($Z = 14$). The width d of a lacquer stripe remained after development process is presented in Fig. 5 as a function of deflection velocity v and of acceleration voltage U_B of the impinging electrons:

- For deflection velocities $v > v_0$ no lacquer stripes remain on the silicon substrate after development. Evidently the charge

density of impinging electrons is smaller than the necessary minimum charge density ($q_e < q_0$).

- For $v_1 < v < v_0$ widths of stripes result of the order of probe diameter. In this range of velocity the charge density of impinging electrons is sufficient to laquer cross-linking ($q_e > q_0$), but the charge density of backscattered electrons is too small to produce a cross-linking ($q_r < q_0$).
- For $v < v_1$ the width of stripes becomes essential larger than the probe diameter. At this deflection velocities the charge density of backscattered electrons is large enough to cross-link the laquer ($q_r > q_0$).

From this velocity dependence of stripe width in the $v < v_1$ area the charge density distribution of backscattered electrons can be determined. At the edges of laquer stripes the charge density q of backscattered electrons is equal to minimum charge density q_0 necessarily for cross-linking of laquer. The relation between these charge densities $q = q_0$ at the edges of laquer stripes, the distance $y = \frac{d}{2}$ of the edges from the deflection trace of the electron probe, the probe velocity v , the probe current I_0 and the electron range x_r [13] is presented in Fig. 6 for three accelerating voltages U_B . The continuously drawn curve is the calculated relation for $Z = 14$ (silicon) taken from Fig. 3.

The measured values are independently of accelerating voltage in conformance with the theory. The difference between the measured and the calculated values can be explained by the lower energy and by the inclination of trajectories of backscattered electrons relatively to the direction of impinging electrons.

The necessary minimum charge density q for lacuer cross-linking is proportional to the electron energy E and to the cosinus of angle ϑ of inclination [11] :

$$q \sim E \cdot \cos \vartheta \quad (12)$$

The backscattered electrons have an average energy of 70% of the impinging electrons and an average angle of inclination of 45° [15] . So the minimum charge density q amounts for the backscattered electrons only about 50% of the minimum charge densities q_0 necessarily for the impinging electrons. Using the correlation $q \approx 0,5 q_0$ for the calculation of values in Fig. 6 a good conformance is reached between the measured and the calculated charge density distribution of backscattered electrons.

Besides the charge density distribution of backscattered electrons from Fig. 5 also can be determined the charge density ratio $q_r : q_e$ of backscattered to impinging electrons. It can be shown, that the ratio of the velocity v_1 to the velocity v_2 (see Fig. 5) is about equal to the ratio

of backscattered charge density q_r to the impinging charge density q_e [1] :

$$\frac{v_1}{v_0} \approx \frac{q_r}{q_e} \quad (13)$$

The measured ratios $v_1 : v_0$ from Fig 5 and the calculated ratios $q_r : q_e$ from Fig. 4 differ about by the factor 2. They are presented in table 1 for different acceleration voltages U_B .

T a b l e 1

U_B/kV	20	50	100
$v_1 : v_0$	$2 \cdot 10^{-1}$	$3 \cdot 10^{-2}$	$5 \cdot 10^{-3}$
$q_r : q_e$	$1 \cdot 10^{-1}$	$1 \cdot 10^{-2}$	$2 \cdot 10^{-3}$

The difference between the measured and the calculated ratios also can be explained by the lower energy and by the inclination of trajectories of backscattered electrons.

4. Conclusions

- 4.1. Based on a simple backscattering model the current density distribution of backscattered electrons at the surface of an object irradiated by a point-shaped electron probe was calculated.
- 4.2. By integration over the current density distribution according to 4.1. the charge density distribution of backscattered electrons at deflection of a point-shaped electron probe along a straight line was ascertained.

- 4.3. Using the charge density distribution according to 4.2. the charge density ratio of backscattered to impinging electrons was calculated, which results by irradiation of a stripe-shaped structure.
- 4.4. The charge density distribution and the charge density ratio according to 4.2. and 4.3. could be verified experimentally. Therefore it is possible to apply the results of calculations to several problems, where the local distribution of backscattered electrons is interesting (for instance: scanning electron microscopy and X-ray microanalysis).
- 4.5. At the applications of calculation results to estimate the widening of structure at the nonthermal electron beam machining is to consider, that the charge density of backscattered electrons to obtain the same machining effect must be only a half of the charge density of the impinging electrons. This fact is caused by the lower energy and the inclination of trajectories of the backscattered electrons.
- 4.6. Because of this double efficiency of backscattered electrons a structure widening of sharply limited structures at the nonthermal electron beam machining can be avoided only, if the charge density ratio $q_r : q_0$ of backscattered to impinging electrons is smaller than 0,5.
- 4.7. Because of the tolerances of several parameters of irradiation at the tech-

nical application of the nonthermal electron beam machining it is necessary, that the charge density of backscattered electrons is at least smaller by the factor 2 than the value at most permissibly according to 4.6. Nevertheless this demand ($q_r : q_e < 0,25$) is accomplished for the most interesting substrate materials ($Z < 25$) according to Fig. 4. For substrate materials with a larger atomic number a structure widening can be avoided only, if the width of structure is small against the range of electrons.

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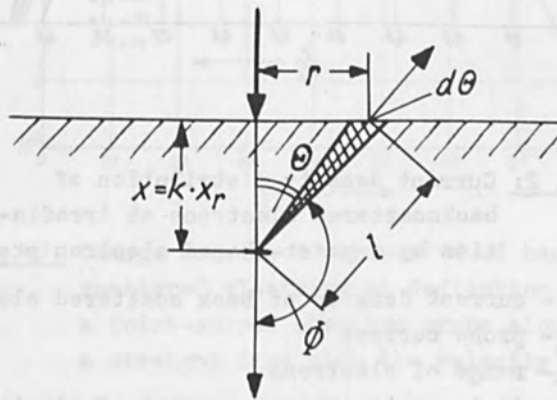


Fig. 1: Schematic graph to the calculation of backscattered current density at the object surface.

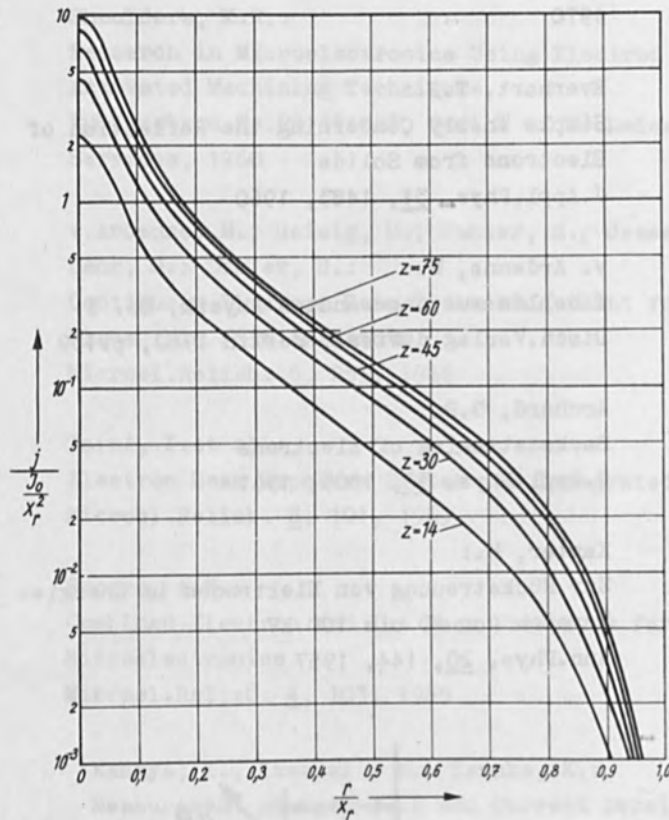


Fig. 2: Current density distribution of backscattered electrons at irradiation by a point-shaped electron probe.

j - current density of back scattered electrons
 I_0 - probe current
 x_r - range of electrons
 r - distance to impinging point of electrons
 Z - atomic number of object material

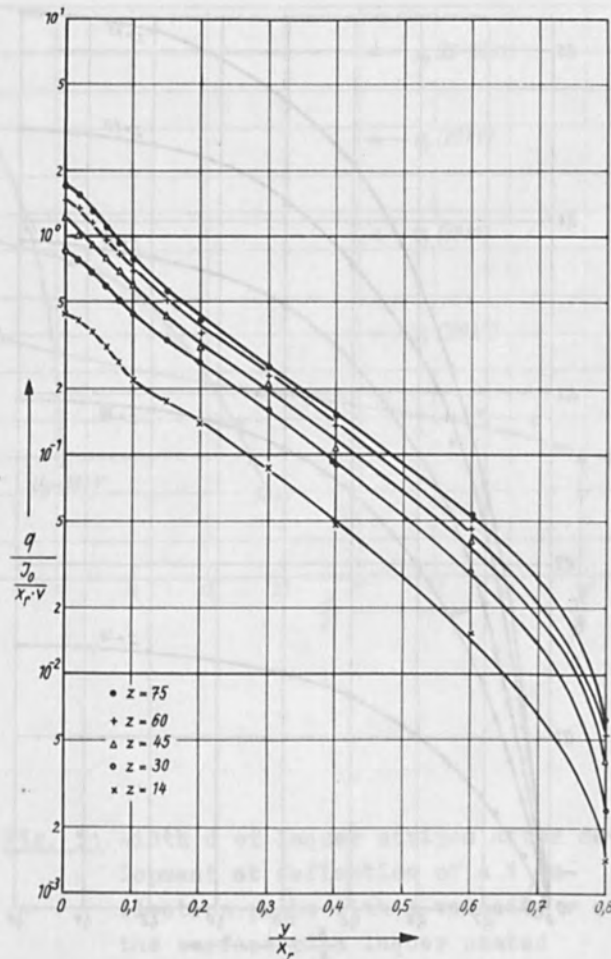


Fig. 3: Charge density distribution of backscattered electrons at deflection of a point-shaped electron probe along a straight line with the velocity v .

q - charge density of backscattered electrons
 y - distance to deflection line

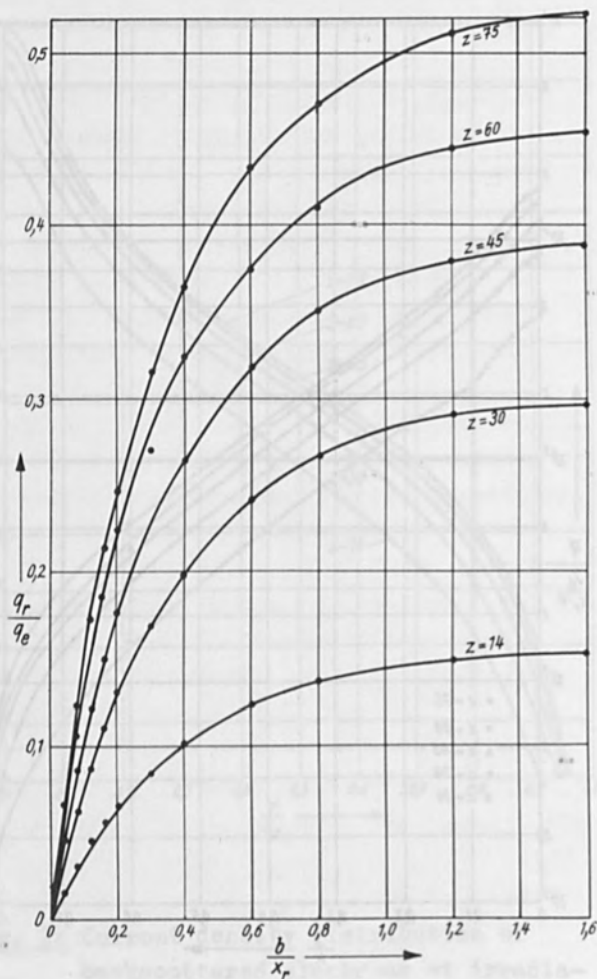


Fig. 4: Charge density ratio $q_r : q_e$ of back-scattered to impinging electrons in dependence on the atomic number Z and the relative width of irradiation $b : x_r$.

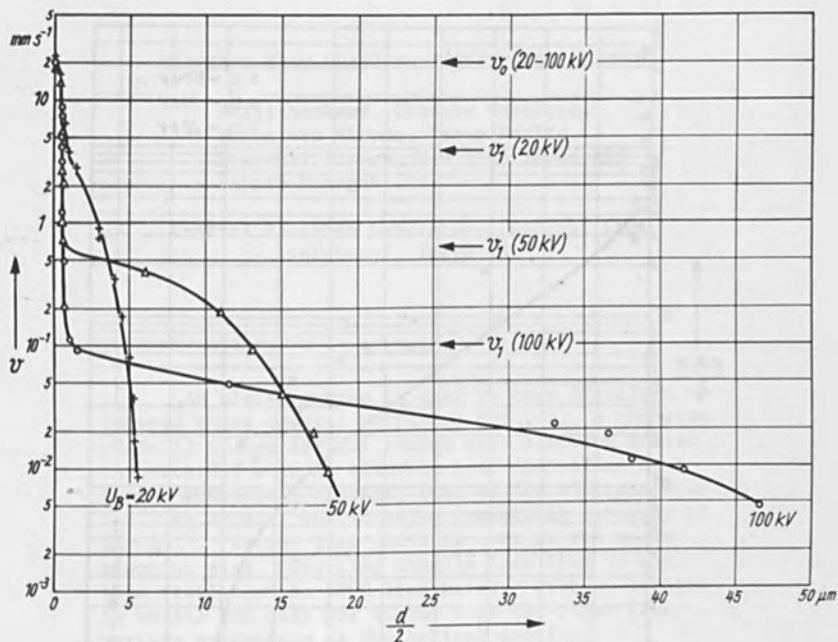


Fig. 5: Width d of laquer stripes after development at deflection of a $1 \mu\text{m}$ -electron probe with a velocity v over the surface of a laquer coated silicon substrate.

- U_B - acceleration voltage
- v_1 - maximum velocity to laquer cross-linking by impinging electrons
- v_2 - maximum velocity to laquer cross-linking by backscattered electrons

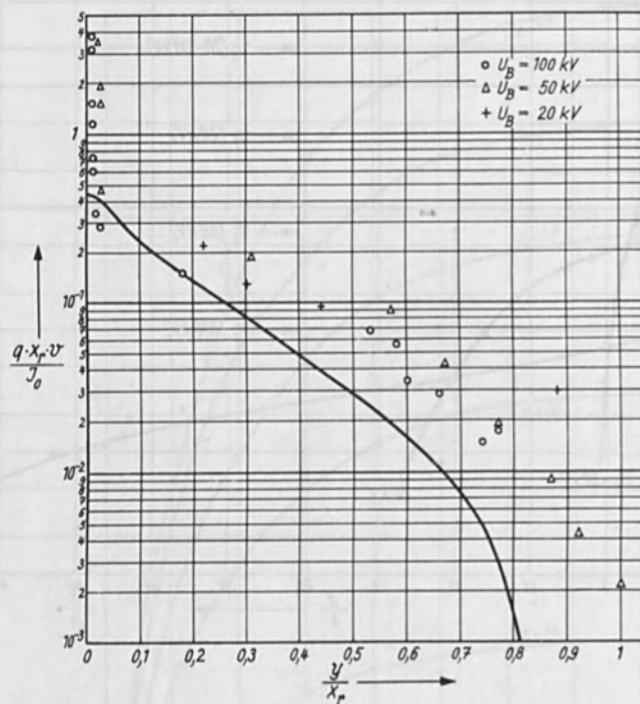


Fig. 6: Comparison of measured and calculated charge density distribution of back-scattered electrons.

Points - charge densities according to values of Fig. 5.

Curve - calculated charge densities according to Fig. 3.

ELECTRON BEAM DRAFTING MACHINE FOR IC MASKS

Akiji Maekawa, Chusuke Munakata,
Chikafusa Hirano, Osamu Morita,
Tokinori Kozawa, Ken'ichi Isoda and
Saburo Nonogaki.

Central Research Laboratory, Hitachi Ltd.
Kokubunji, Tokyo 185

ABSTRACT

An electron beam is used to make firstly-reduced masks (master reticles) for ICs. A chromium plate (2 inches square) coated with electron resist is bombarded with an electron beam controlled by a hybrid computer. Scanning area of the electron beam is 23 mm square, and relative repeatable accuracy is $\pm 1 \times 10^{-4}$. Etched line width is $4 \mu\text{m}$ on the whole scanning area. Drafting time is less than 10 min. Line resolution is 3000 line pairs. Present machine is useful not only for IC but also for other fine pattern generation as the optical grating.

INTRODUCTION

An electron beam has been used to record various informations onto electron sensitive materials since it can be focused into a fine spot and easily controlled.

The direct electron beam exposure is, in fact, applied to making masks for semiconductor integrated circuits (ICs), fabrication of ICs^{1,2,3)}, film recording of TV signal⁴⁾, or computer output display⁵⁾.

Present system aims to make firstly-reduced masks for semiconductor ICs, so called, master reticles.

A chromium plate (2 inches square) is coated with newly-developed electron resist. The plate is bombarded with a controlled

electron beam. Then IC mask pattern is automatically drafted. Whole of this system is simply called as an "Electron Beam Drafting Machine".

Fig. 1 shows a chart of IC pattern process. A conventional process follows the bold line. In the process, it takes more than a few days to make the master reticles, because they mainly depend upon the man power of the artwork. An X-Y plotter and other pattern generators are used to save the man power, or to diminish the time consumed in the pattern generation. Each necessary time is shown in the parentheses. The present machine is advantageous over other apparatuses because of its short time needed for making master reticles.

As for the master mask or the direct exposure of the silicon wafer, several electron beam methods have been already reported^{1,2,3)}. In these methods, it takes comparatively long time of more than 1 h to complete a master mask or a direct exposure of a wafer. In an electron beam apparatus, as is well known, it is not easy to guarantee long-time stability, on which the accuracy of the master mask may depend. The long time stability is unnecessary in the present machine.

An Electron Beam Pattern Generator⁶⁾ similar to the present one has been already reported. It, however, differs from the present one in detail, and the present one might have higher relative accuracy of repeatability compared to the former.

This paper describes the outline of the present system and its some applications.

OUTLINE OF THE MACHINE

Fig. 2 shows a block diagram of the present machine. In Fig. 3 there is shown a photograph of the present electron beam apparatus, and in Fig. 4 that of the hybrid computer system.

The electron gun is a conventional hair-pin type tri-electrode gun⁷⁾. The electron beam is switched by applying pulse voltage to the Wehnelt electrode, focused with two condenser lenses, and scanned with a magnetic deflection coil. A chromium plate, which is bombarded with a computer-controlled electron beam, is set upon the working table which is precisely positioned by the computer.

The control signals are generated by a small hybrid computer (Hitachi CLOAP 2000E), which has 8 kilo-words (18 bits) core memories and the cycle time of 2 μ sec. There are two sets of digital-to-analogue converters, and each one treats 15 bits value. Figure drafting is not raster scanning like the TV scanning mode. A pattern element is painted with the electron beam, and the beam jumps from a pattern element to another following to the computer signals. This mode is defined random drafting in this paper.

Line resolution of etched chromium plate is 3000 line pairs, and this corresponds to 6000 TV lines. New techniques for high precision drafting are as follows:

1. A precision digital-to-analogue converter is developed. (Temperature drift is 2.5~5 ppm/ $^{\circ}$ C, and response time is 1 μ sec for attaining the accuracy of 0.01 %.)
2. Distortion correction of pattern is done with an analogue computer.

3. A deflection amplifier with quick response (several μsec for the accuracy of 0.01 %) is developed.
4. A ferrite-surrounded coil is adapted to reduce the eddy current loss in the iron wall near the deflector in the present magnetic deflection system.
5. Automatic scale calibration is adapted to obtain positional repeatability. Reflected electron signals from a standard plate are treated in the computer.

In addition to the new techniques listed above, smaller aperture angle is necessary to hold a fine beam spot over a wide area.

The time needed to draft over a frame area (23 mm square) is within 10 min even with 12000 lines in a frame. New techniques developed for fast drafting in the present system are as follows:

1. An analogue computer generates function signals.
2. An electron beam scans the chromium plate in both directions of X and Y.
3. When a photo-plate is used, charging up effect is prevented by covering the photo-plate surface with a conductive (aquadag) layer.
4. Highly-sensitive electron resist has been developed ($5 \times 10^{-8} \text{ C/cm}^2$). Usually the electron resist is preferably used, because a photo-plate makes rather broad lines ($7 \mu\text{m}$), while the resist narrower lines ($4 \mu\text{m}$).
5. An electron gun is switched with low pulse voltage (less than 25 V) fed to the Wehnelt electrode.

6. The random drafting mode is used instead of the raster scanning mode.

The newly-developed electron resists are insensitive to visible rays. In Fig. 5 there are shown the threshold charge densities for usual resists and the present newly-developed electron resists. Hitachi electron resists thus show very high sensitivity for electron, and it is proved to be due to Epoxidized cis-1,4-polybutadiene and Epoxidized cis-1,4-polyisoprene.

As for the drafting time, it is easily seen that the present random drafting is advantageous over the raster scanning when the occupied or beam-exposed area is small compared to the unoccupied area in the pattern. With the present system, it takes only a few ten seconds in the case of simple pattern as will be shown in the following examples, while 10 min is always necessary in such a raster scanning case as Bell Telephone Laboratory's new system⁸⁾. Features of the present machine are summarized in Table 1.

EXAMPLES OF DRAFTED PATTERNS

Fig. 6 shows a serial set of IC masks for a TV set part. Drafting times are shown in the parentheses. All drafted patterns shown in Figs. 6~9 correspond to etched chromium plates.

The patterns in Figs. 7~9 are not the IC patterns but the special patterns obtained with the present machine. Fig. 7 shows $4\mu\text{m}$ spot in $12\mu\text{m}$ square. Fig. 8 shows popular game "GO" in Japan. Area of the frame is 23 mm square. Fig. 9 shows parallel lines of

$4\mu\text{m}$ wide. 3000 line pairs in a frame are drawn. Fig. 10 shows the transmitted diffraction pattern of the grating shown in Fig. 9 obtained with a He-Ne laser. In another plate, $4\mu\text{m}$ diameter spots were obtained in $8\mu\text{m}$ pitch (not shown in this paper). This corresponds to 9×10^6 spots in the frame.

CONCLUSION

Outline of the Electron Beam Drafting Machine for IC master reticles is described. Experimental results are summarized as follows; Scanning area is 23 mm square, relative repeatable accuracy $\pm 1 \times 10^{-4}$, line width $4\mu\text{m}$, drafting time less than 10 min and line resolution 3000 line pairs. Examples of IC masks are shown. This machine is useful not only for IC but also for other general fine pattern generation. An optical grating obtained with the present machine has been shown as the example.

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Table 1 Features of E. B. D. M.

Scanning area of electron beam ----- 23 mm square

Positional repeatability ----- $\pm 1 \times 10^{-4}$ in the scanning area

Line width ----- $4 \mu\text{m}$ in the scanning area

Accelerating voltage ----- 20 kV (variable up to 30 kV)

Brightness (computed value) ----- $2 \times 10^4 \text{ A/cm}^2 \cdot \text{ster.}$

Scanning speed ----- 1, 3, 6, 12, 24, 50, 100, 300, 1000 m sec/23 mm

Distance between final lens and sample ----- 250 mm

Working distance ----- 150 mm

Sample current ----- $< 0.5 \text{ nA}$

Half of aperture angle ----- 10^{-3} rad

Drafting time ----- $< 10 \text{ min}$ using Hitachi electron resist

Line resolution ----- 3000 line pairs/23 mm

Point resolution ----- $9 \times 10^6 \text{ points/23 mm square}$

Figure drafting ----- random positioning

Working table movement ----- covering area $80 \times 120 \text{ mm}^2$
 (positional accuracy $\pm 2 \mu\text{m}$)

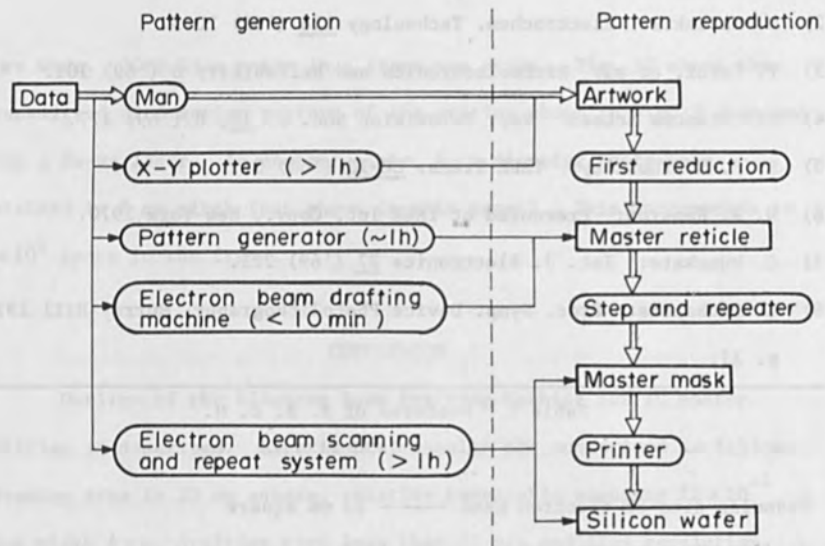


Fig. 1. A chart of IC pattern process.

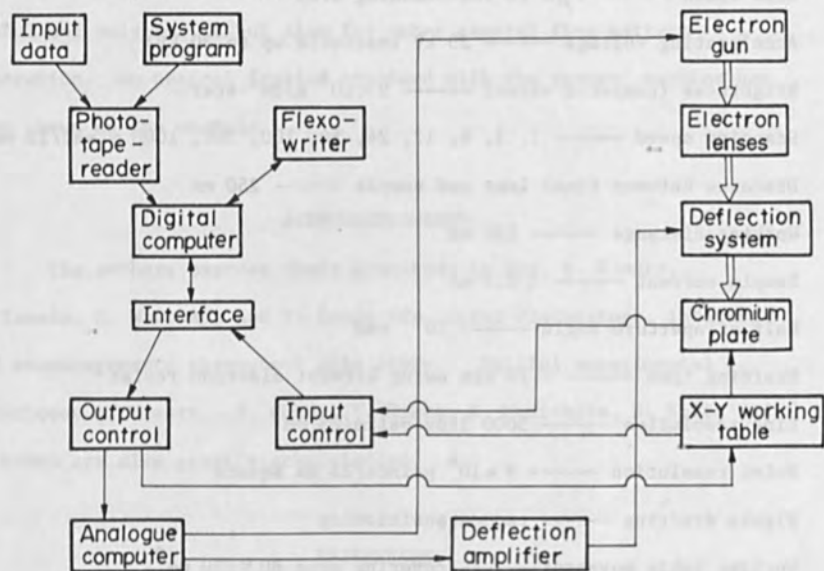


Fig. 2. A block diagram of the present system.

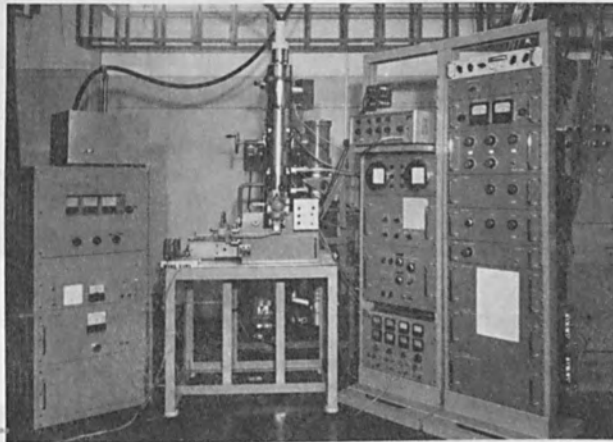


Fig.3. Electron beam apparatus.



Fig. 4. Hybrid computer system.

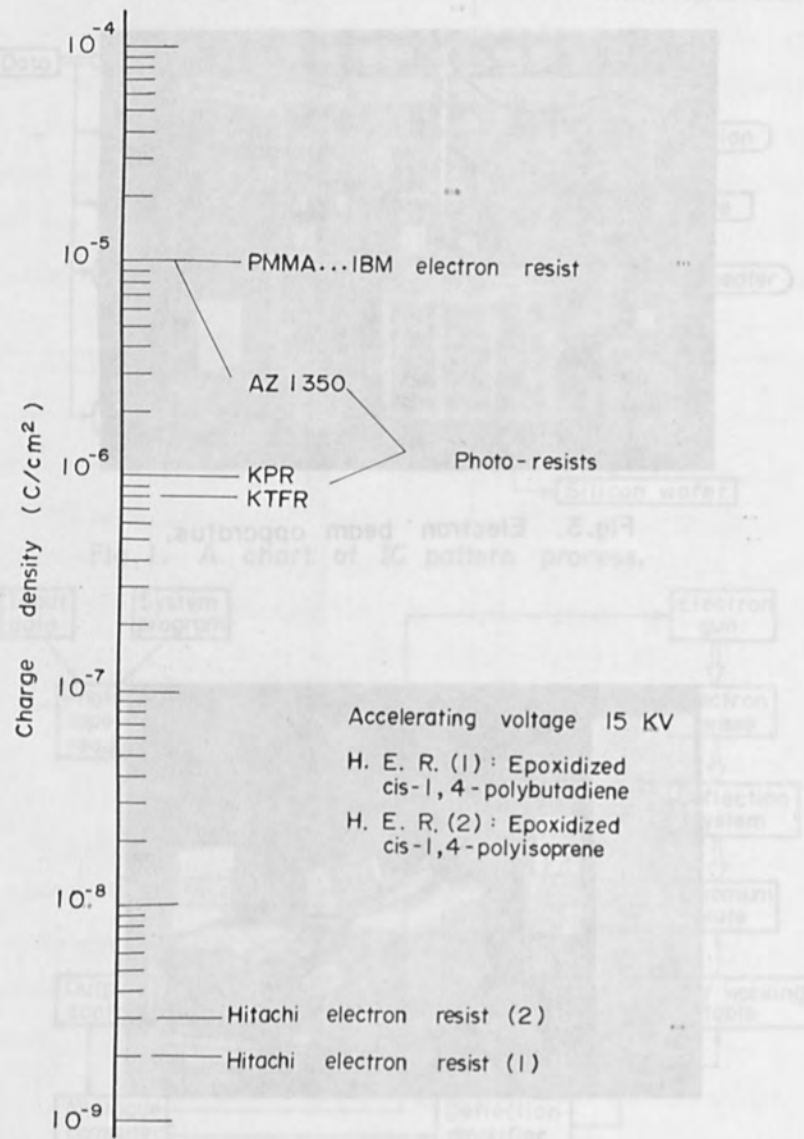
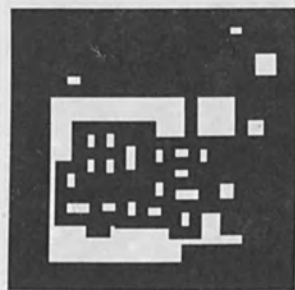
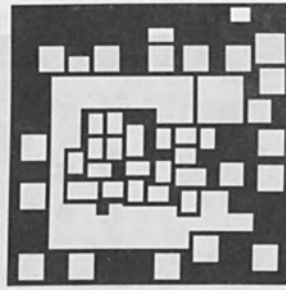


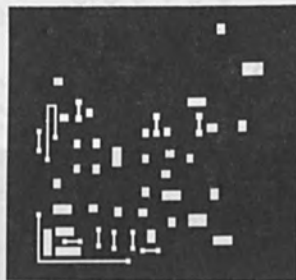
Fig. 5. Threshold charge density for resists.



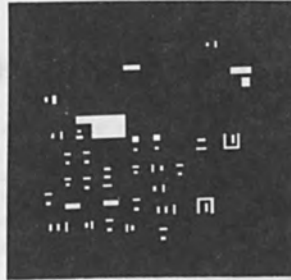
Buried layer (0.9min)



Isolation (2.3min)



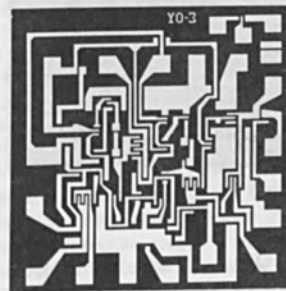
Base (0.4min)



Emitter (0.2min)



Contact (0.4min)



Aluminum (2.1min)

(mm)
0 1 2 3 4 5

Fig.6. Typical chromium masks (master reticles).

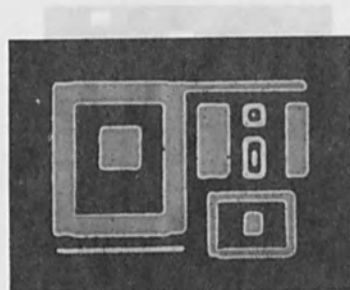


Fig.7. Example of fine pattern.

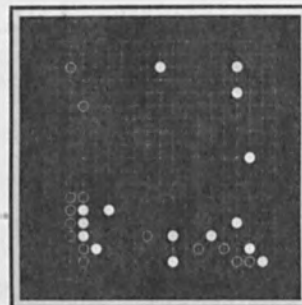


Fig.8. Japanese "GO".

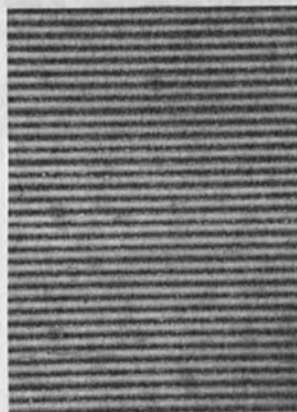


Fig.9. Line resolution
($8\mu\text{m}$ pitch; Etched
chromium plate).



Fig.10. Transmitted
diffraction pattern
obtained with He-Ne
laser.

A COMPUTER CONTROLLED ELECTRON MASK MAKER

J. P. Beasley

Mullard Research Laboratories, Redhill, Surrey, England

In the machine to be described an electron beam is computer controlled to define integrated circuit patterns at their final size. Exposure of resist on chromium on glass is used to produce masks 50 mms square.

The emphasis is on speed and automation. High precision over an unusually large electronic scan is achieved by automatic refocussing and repositioning on predeposited markers so no mechanical movement of the slide is required. Pump down and slide alignment are also automated.

Introduction

There is increasing interest in the application of electron beam processes to the manufacture of microelectronics. This is especially true in regard to pattern generation, both for making integrated circuit masks and for direct exposure of resist on silicon. The outstanding advantage of using electrons rather than photons is that the resolution is no longer limited by the wavelength of light. There are other advantages, however. Electron scanning is simple and fast so the inflexibility of imaging techniques is avoided, backscattered electrons provide convenient feedback for automatic beam control and a wider range of materials are sensitive to high voltage electrons than to photons.

In the machine to be described, a scanning electron microprobe with a 2 micron diameter beam is controlled by a small digital computer and used to make integrated circuit masks at the final size. The masks are made from 50 mm square chromium on glass slides by the following technique. A slide is first coated with a layer of an electron-sensitive material, which could be any photo-resist or a special electron resist. It is then exposed in the machine to the focussed electron beam which traces out the required pattern geometry in response to a paper tape data input to the computer. Afterwards, the exposed areas of the resist layer are removed by immersion in a suitable developer and finally the chromium is etched away through the holes in the resist. The end result is a complete integrated circuit mask. Note that a whole array of I.C. patterns is produced, so that only one stage is required to get from a data description of the circuit to the final chromium mask. This is in contrast to orthodox photolithographic mask manufacture in which the pattern corresponding only to a single integrated circuit is first produced. Its demagnified image then has to be "stepped and repeated" over the surface of the slide to produce the large array of identical

patterns which constitute the mask. One process has replaced two, with a potential saving in time and cost and gain in flexibility.

Apart from making masks we are also interested in exposing resists on the surface of the silicon slice directly and rendering the use of masks unnecessary. Some of this work was described at this conference two years ago by J. Schofield⁽¹⁾ and is still being actively continued. The design of the mask maker has to some extent been influenced by the future application of the system to work on silicon. In particular some attention has been paid to high speed of operation because speed is more important when a slice is being processed than when masks are being made. Every slice has to be processed in the machine, whereas a set of masks will, if contact copied, serve to define the diffusion and interconnection patterns on a very large number of slices. With this speed requirement in mind it would be nice to use an electronic scan covering the whole area of the slide or silicon slice to avoid the mechanical movement between exposures which is necessary if the electronic scanning area is limited. It is difficult of course to obtain the necessary high precision over such a large scan; in television terms it corresponds to trying to produce a picture with something like 20,000 lines, and this is an order better than is normally expected from electronic scanning systems.

In the mask maker we deflect the beam over an area 40 mms square and maintain high accuracy and resolution by repositioning and refocussing the electron beam at frequent intervals throughout the deflection field, using markers predeposited on the surface of the slide. No mechanical movement is required. The markers consist of 100 micron gold squares spaced a few millimetres apart in X and Y all over the slide. The spacing is suited to the size of the integrated circuit patterns being produced. The markers are printed on top of the chromium layer using normal photolithographic methods. The procedure when exposing the slide to the beam consists of alternate integrated circuit pattern drawing and beam correction routines. The effects of d.c. instability and deflection field distortion are rendered insignificant because all the pattern detail is drawn adjacent to one marker or another and no longer than a few seconds after the beam has been refocussed and repositioned.

Such a system is workable even if the markers are not particularly well shaped or accurately positioned, because what is important in a mask is compatibility between the fine detail of one mask and of corresponding detail of another mask which is to be used on the same slice of silicon, i.e. between two masks of the same "set". It does not matter if, say, the transistors near the edge of the slice are very slightly different from those at the centre, but it is vitally important, for example, that the contact windows of individual transistors are placed squarely on top of their emitters and exactly underneath their contact pads. By using the same negative to print the marker arrays on the different slides which will form a mask set this correspondence can be achieved.

The Equipment

Figure 1 shows a photograph of the machine. The computer is a DEC PDP8 with a 4000 word store of 12 bit words.

Figure 2 shows how the electron optical column is interfaced to the computer. Data describing the integrated circuit pattern consists of a list of the co-ordinates of the rectangles and lines which constitute the circuit. The lines may be orthogonal or at 45° . The data is fed to the computer on punched paper tape and stored on a large magnetic disc memory associated with the computer. From there, apart from direct use in specifying the patterns, it can be plotted on an X-Y recorder for checking and also manipulated in various ways, including repetition, scaling, inversion and so on. Thus large and complex circuits can be built up from smaller units already in store. The main components of the electronic interface are the two deflection amplifiers, a beam switching unit and a focus servo unit, the latter being used in the beam correction routine.

Figure 3 shows the main components of the electron optical column. Normal working voltage is 15 kilovolts. Two magnetic lenses bring the beam to a 2 micron focus at the target, the chromium on glass slide. The beam is switched so far as the target is concerned by "blanking" it half way along the column. Two pairs of electrostatic deflection plates are arranged to sweep the beam away from an aperture in the column without moving its point of impact at the target.

A magnetic deflection yoke designed for a precision cathode ray tube produces a total deflection area at the target plane of 40 mms in X and Y. To reduce deflection aberrations and field distortion the overall scanning angle is restricted to less than 20° by using the unusually long working distance of 16 cms. Because the deflection coils are positioned between the final lens and the target this means that the working distance of the lens is about 20 cms. The lens is designed for low spherical aberration, however, and with a 2mm diameter aperture gives a beam current in excess of 2×10^{-7} amps in a 2 micron focus.

Electrons scattered from the surface of the target pass through a cylindrical grid and are accelerated towards four large plastic scintillators placed symmetrically round the work chamber. The scintillations are detected by photomultipliers which provide the video feedback for the focus and position servos and also for an S.E.M. display unit consisting of a modified television receiver. The scintillator geometry has been arranged to provide a large and fairly uniform solid angle for the reception of electrons emitted from all points on the target. It is back-scattered electrons which provide significant information, not true secondary electrons which tend to originate from the top of the resist layer.

The electron optical column is automatically pumped. When the lid of the work chamber is opened a valve automatically closes halfway down the column, isolating the electron gun which does not, therefore, have to be switched off. When the lid is closed again the work chamber is

automatically pumped out, then this valve opens and another slide can be exposed to the beam. The pump-down time is about 20 seconds and a slide can be changed in half a minute.

Figure 4 illustrates certain special features of the deflection amplifiers designed to make the pattern generation faster and more easily programmed.

Because of the large dynamic range between the finest detail and the full scanning area, corresponding to nearly 17 binary stages, two overlapping 12 bit digital to analogue convertors are required to drive each deflection amplifier. The "fine" one handles the pattern information, the "coarse" one provides the electronic step and repeat facility from one pattern to its repeat. The beam could be made to draw a line, say along one of the deflection axes by continuously incrementing the input to one of the D.A.C.'s. Of course, if the increments are too large the line becomes dotted rather than continuous. In our case the smallest increment is $\frac{1}{2}$ micron corresponding to one computer bit and it takes the computer about 12 microseconds to make a single step, and at the same time keep a count. This limits the speed with which a line can be drawn. To avoid this each amplifier includes what we call a "rate" circuit. This has the property of converting a step input to a ramp output, the slope of the ramp being adjustable and under program control. It is then only necessary to output consecutively the two end co-ordinates of a line and the rate value and the beam will draw a uniform line between the two end points. Lines can be drawn in this way much more quickly than by point by point stepping. In addition, a signal is sent back to the PDP8 when a line is completed and the beam can be automatically switched on at the beginning of a line and off at the end. These facilities provide a very flexible control of the beam. Any sequence of linear beam movements can be programmed quite easily. For example, rectangular shapes are filled in by causing the beam to execute a diminishing "picture frame" routine which has some advantages over using a raster pattern.

Beam Correction

Once it has been "calibrated" on a previous set of markers the machine can be programmed to land the beam inside any of the markers in the deflection field. The beam correction system is then used to establish the exact position of the marker relative to the deflection field and to bring the beam to a 2 micron circular focus. We are then in a position to draw sharp, accurately located patterns in the area immediately surrounding the marker.

The correction procedure can be described briefly and consists of two parts:

- a) The positioning routine. The beam is stepped out from its starting position inside the marker to cross the four marker edges in turn. Because gold scatters more of the primary electrons than chromium there is a sharp change in the video signal derived from the backscattered electrons on crossing an edge. This initiates a signal to the computer

denoting that the edge has been crossed. By using multiple traverses across each edge after the beam has been refocussed an accurate estimate of the position of the marker centre is arrived at even if the marker edge is somewhat irregular.

b) The focus routine. The risetime of the video step occurring when the beam crosses an edge depends on the velocity of the beam traverse and on the width of the beam in a direction perpendicular to the edge. Thus, by sweeping the beam at a known rate across two adjacent edges of a gold marker in the form of an L-shaped scan and measuring the two video risetimes an estimate can be made of both the size and shape of the beam. Then the beam can be brought to a sharp circular focus by two stigmators and a fine focus coil situated in the objective lens of the machine. The risetime is measured by starting a timer when the video signal reaches 30% of its new value and stopping it at the 70% level and then outputting to the computer the timing value. Fixed level detectors are employed so the timing value is only meaningful if the video gain has been adjusted to give a standard size of step. The computer, which is the main element in the focus servo loop, first has to normalize the video gain and then to use a hill-climbing routine during the L scan to bring the beam to a focus.

At present the positioning routine is accurate to better than ± 1 micron and the focus is maintained throughout the field to better than three microns.

Results and Discussions

The equipment is only just commissioned, but there are some results to show.

Figure 5 shows a single integrated circuit pattern drawn in polymethylmethacrylate resist by the machine. It is the interconnection pattern for a TTL logic circuit. The total area is about 3×1.6 mm and the width of the lines about 12 microns.

Arrays of patterns similar to the one shown in figure 5 and occupying the full 40 mm square deflection field have been defined in resist and after development have been etched in chromium. Also the beam correction routines have been shown to operate satisfactorily right to the edge of the field. Completely satisfactory masks have not yet been produced because the program which combines the two routines is not quite finished. It does appear, however, that the machine will have the design performance.

The two most important criteria of a mask drawing machine are its accuracy and speed. The accuracy of the existing machine, with its 2 to 3 micron beam is comparable with that obtained with normal photolithographic techniques. An obvious way to improve the accuracy is to scale down the electronic scanning area by decreasing the working distance of the deflection coils. Then the beam diameter can be reduced, still maintaining the same current. Some mechanical movement of the slide

is required to make a full sized mask. The number of mechanical steps is relatively small, however, and because of the electronic marker detection no great mechanical precision is necessary. The present scale was chosen to prove the system. Now that this appears viable the machine is being modified along the lines suggested.

Now as to speed. A complete array of the rather complicated circuit shown in figure 5 takes about 30 minutes to expose. The time was very largely determined by the exposure dosage required for PMMA resist. We have developed several more sensitive resists and we expect the exposure time to be considerably reduced. Then it may be the computing time involved in drawing all the rectangles which will be the limiting factor on the speed. We have already progressed from laying down a pattern as a series of dots to one effectively composed of lines. The next logical step is to develop rectangle generators so the computer only has to output rectangle co-ordinate data.

The most interesting way in which to continue this program is to adapt the machine to expose resist directly on the silicon slice, preferably siliceous resist which constitutes a diffusion barrier or insulating layer directly. Then the marker system will come into its own because undoubtedly the best way to locate a new pattern on top of one already impressed on the silicon is to refer at the most spatially frequent intervals possible to the existing pattern. The outstanding obstacle in this direction is retaining suitable markers throughout the several diffusion processes.

Acknowledgements

The writer wishes to acknowledge the collaboration of M. J. Underhill, who was responsible for the electronic and control aspects of the machine and the extensive help and encouragement of H. N. G. King. He wishes to thank the Directors of Mullard Research Laboratories for permission to publish this paper and to acknowledge the assistance of D. Squire, P. A. Jordan and R. A. Ireland in all the work described. J. Ero of Philips Research Laboratories was responsible for much of the computer programming.

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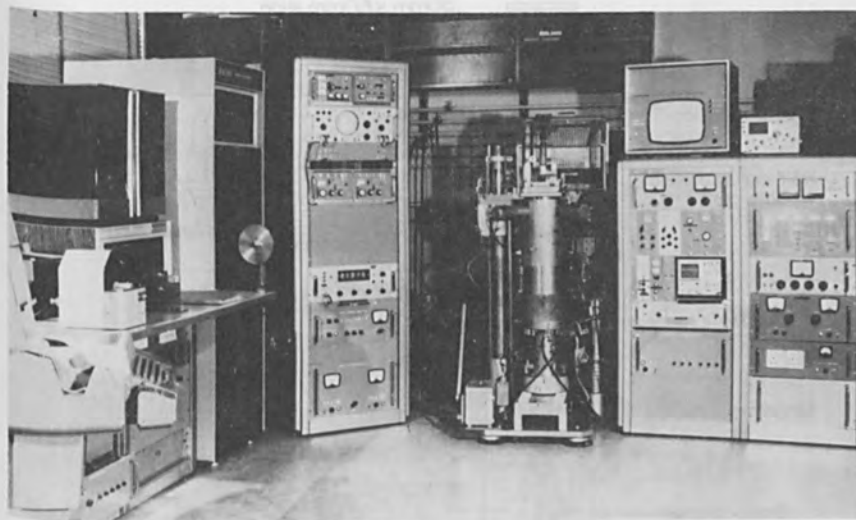


FIG. 1.

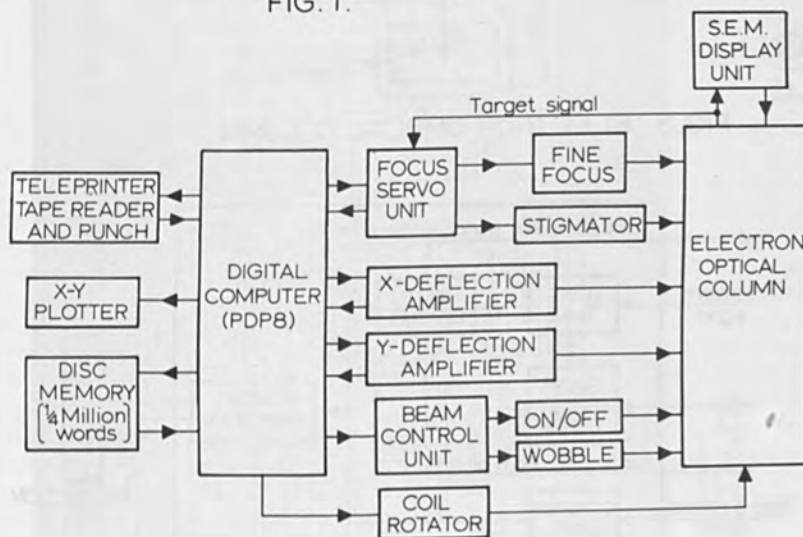


FIG. 2. MACHINE CONTROL SYSTEM

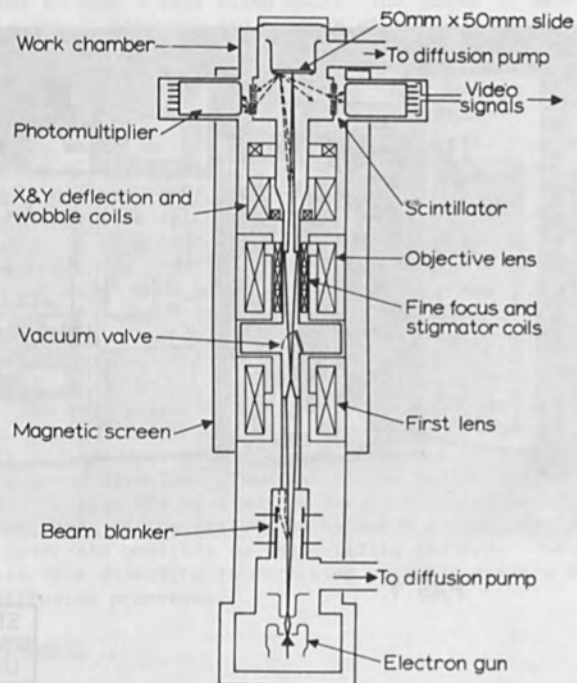


FIG.3. ELECTRON OPTICAL COLUMN

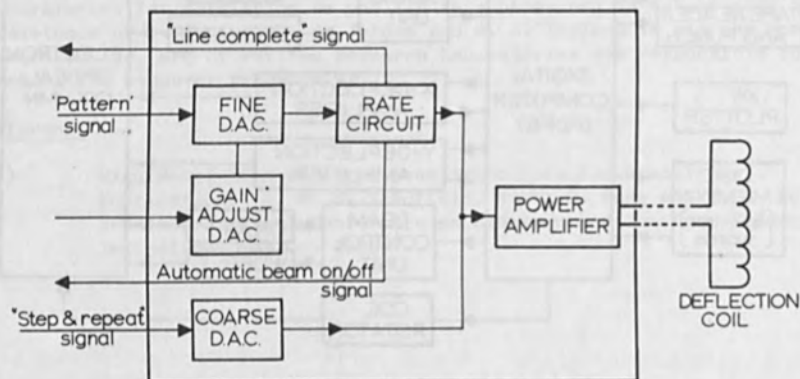
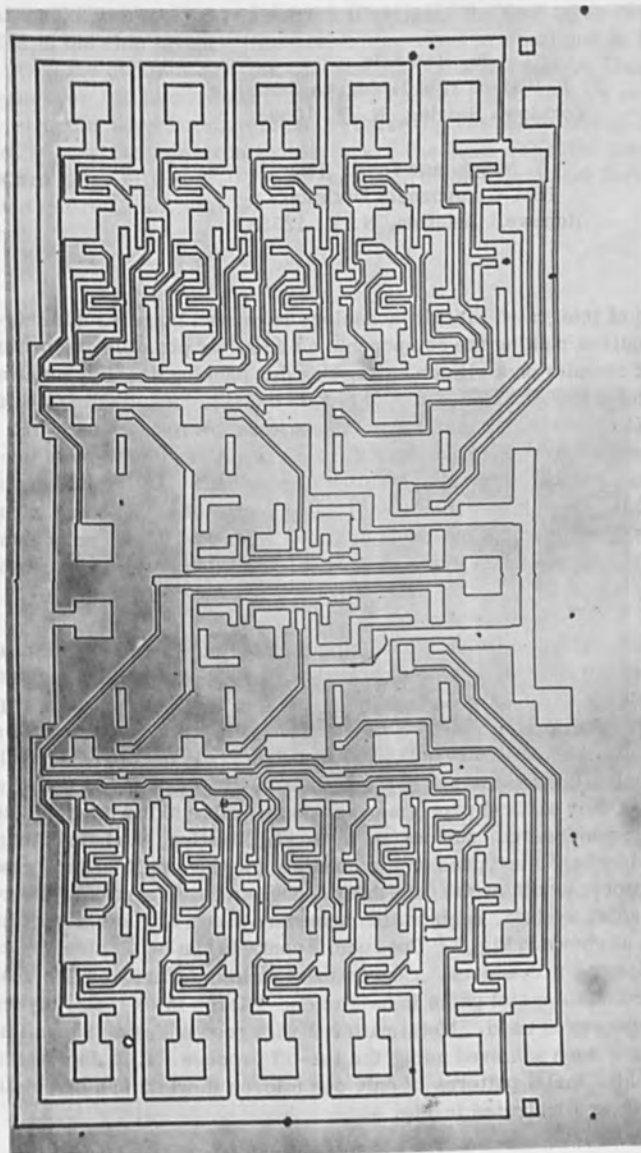


FIG.4. BEAM DEFLECTION AMPLIFIER



250 μm

FIG. 5

ELECTRON BEAM FABRICATION OF MICRON TRANSISTORS

M. Hatzakis

T. J. Watson IBM Research Center
Yorktown Heights, N. Y. 10598

S. Magdo and C. H. Ting
IBM Components Division
Hopewell Junction, N. Y. 12533

The present trend of integrated circuit technology is toward larger-scale integration. This requires making the components as small as possible and packing a large number of circuits on a silicon chip. Besides packing density, smaller components also have the advantages of higher speed and higher yield. One of the fundamental problems in making smaller components is the limitations of the optical masking technology. Electron optical systems represent the most promising fabrication technique in making smaller components in integrated circuits. To illustrate the high resolution capability of the scanning electron beam exposure system, Fig. 1 is an electron micrograph of $1000\text{\AA}$ metal grid lines on one-micron centers. With the electron beam system, the defects associated with masks are also eliminated.

This paper describes the results obtained using the electron-optical system to fabricate small transistors. Planar bipolar transistors with emitter and base contact windows of one micron width have been fabricated using the electron beam exposure system to open all the diffusion windows as well as metallization patterns. An equipment schematic is shown in Fig. 2. A special electron beam resist, polymethyl-methacrylate, is used instead of a standard photoresist for its superior resolution capability. Other critical problems associated with making micron size transistors are also addressed, such as accurate registration, surface damage due to electron beam bombardment, good ohmic contacts for the small contact window, and providing a micron-width metal path thick enough to carry a reasonable current. Using the SEM display system, registration accuracy within a fraction of a micron was achieved, as shown in Fig. 3. Good ohmic contacts can be obtained by careful etching and cleaning procedures. Conventional etching techniques are unable to provide one-micron wide metal paths in half-to one-micron thick metal layers; therefore the lift-off process is used. Metal patterns with good adhesion as well as good ohmic contacts have been achieved using the lift-off process. It is demonstrated that one-micron thick metal patterns of only one micron in width can be obtained with the lift-off process as illustrated in Fig. 4.

The dc and ac characteristics of the micron transistors are presented. These characteristics compare favorably with the best bipolar transistors produced by optical masking processes. Figure 5 illustrates the four types of transistors included in the chip layout. The processing steps are outlined in Figure 6. Figure 7 gives the common emitter characteristics of the micron transistors. Current gain over 100 is obtained after proper annealing to remove surface damage due to electron beam bombardment. Figure 8 gives the values of gain-bandwidth product, f_T , of these micron transistors. The base-collector junction capacitances are given in Fig. 9. These results provide a guide for further refinements in future micron transistor design.

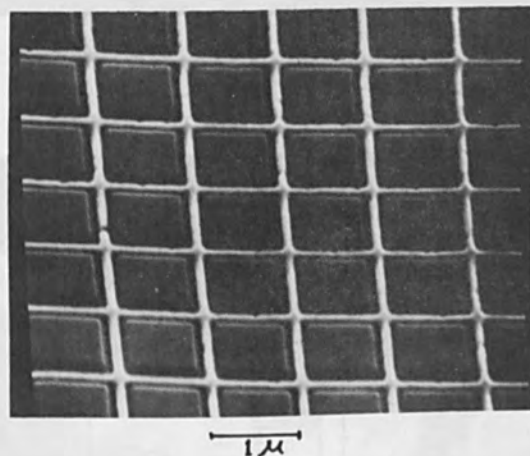


Fig. 1. $1000\text{\AA}$ metal grid lines on $1\text{-}\mu$ centers.

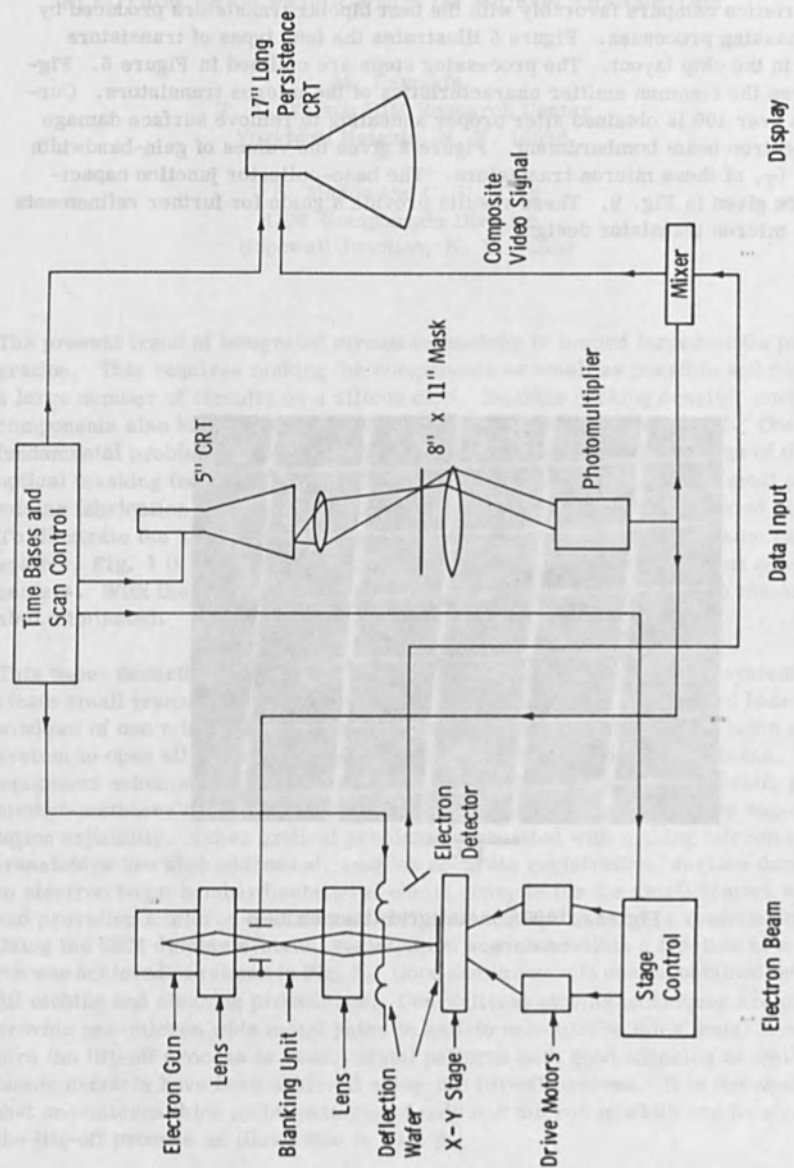


Fig. 2. Electron beam exposure system schematic

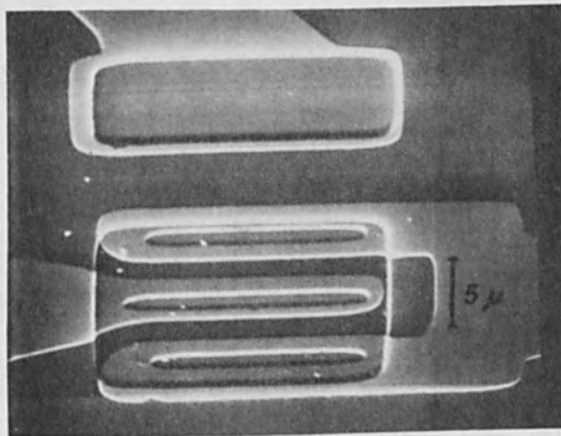


Fig. 3. SEM micrograph of 1μ wide transistors.

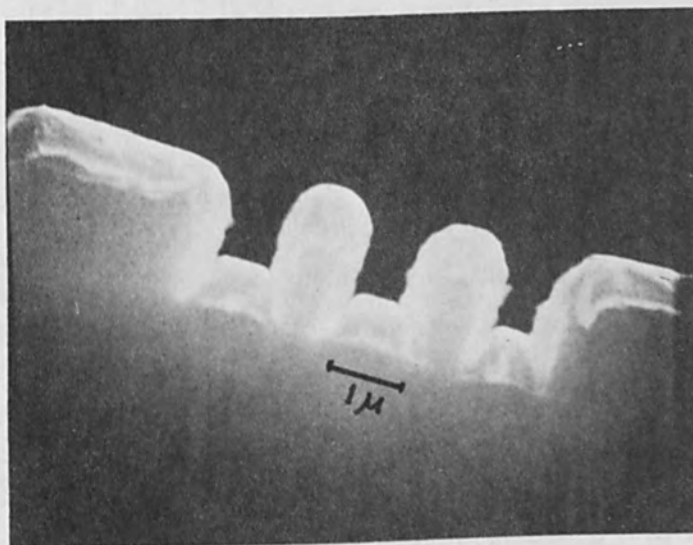


Fig. 4. SEM micrograph of $1\mu \times 1\mu$ metal lines evaporated through resist.

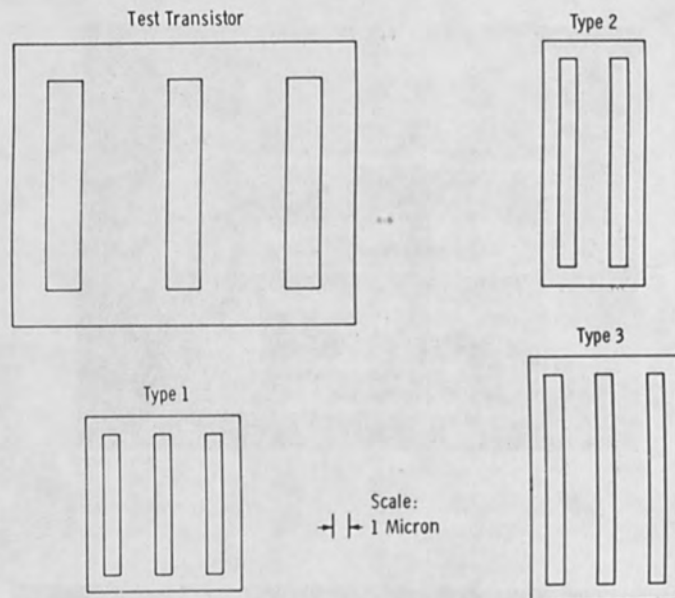
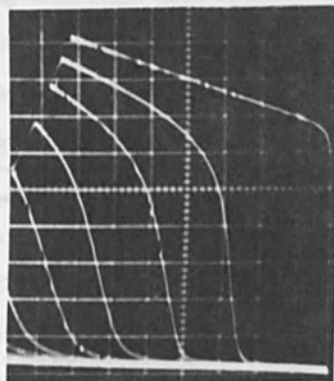


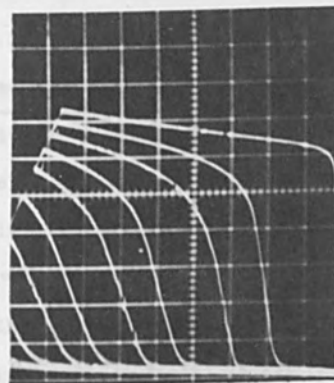
Fig. 5. Horizontal geometry of active areas.

1. p^- Substrate
2. n^+ Subcollector Diffusion
3. n^- Epitaxial Deposition
4. n^+ Collector Reach-Through Diffusion
5. p Base Diffusion
6. Pyrolytic Silicon Dioxide Deposition
7. n^+ Emitter Diffusion
8. Base Contact
9. Metallization

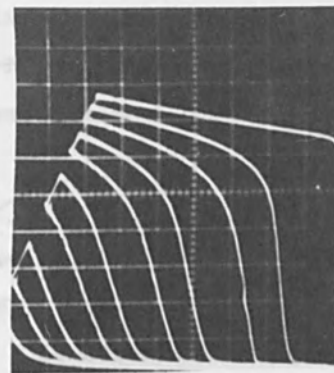
Fig. 6. Processing steps.



Type 1
 $I_C = 1\text{mA/cm}$
 $V_C = 1\text{V/cm}$
 $I_B = 0.5\text{mA/step}$



Type 2
 $I_C = 1\text{mA/cm}$
 $V_C = 1\text{V/cm}$
 $I_B = 0.02\text{mA/step}$



Type 3
 $I_C = 1\text{mA/cm}$
 $V_C = 1\text{V/cm}$
 $I_B = 0.02\text{mA/step}$

Fig. 7. Common emitter current-voltage (I - V) characteristics.

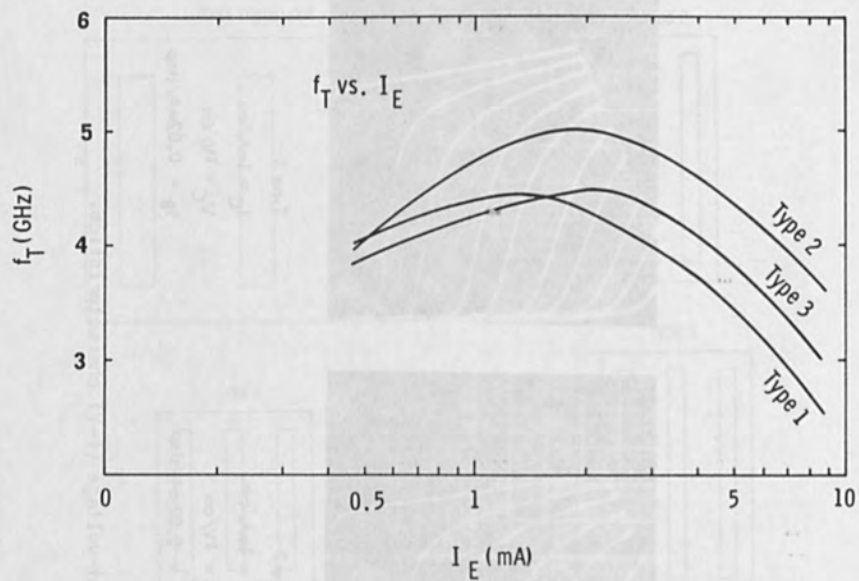


Fig. 8. Gain-bandwidth product (f_T) vs. emitter current.

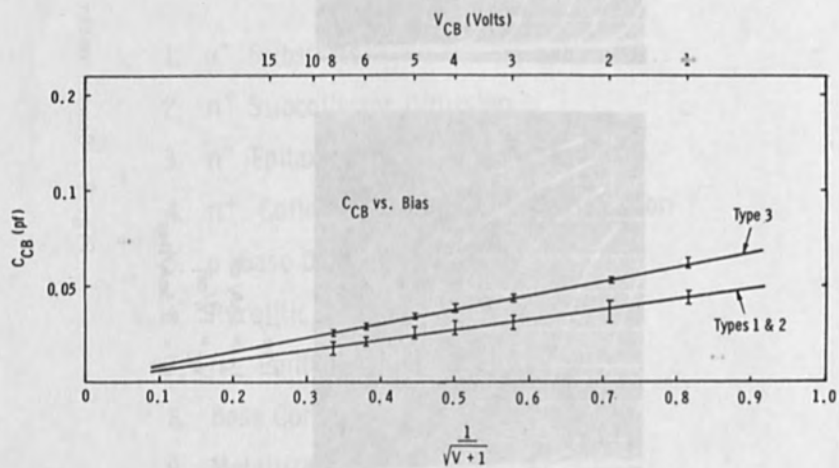


Fig. 9. Base collector junction capacitances (C_{CB}) vs. reverse bias.

ION IMPLANTATION APPARATUS FOR PROCESSING SEMICONDUCTORS

T. Tsuchimoto, T. Tokuyama and T. Ikeda

Central Research Laboratory, Hitachi Ltd.,

Kokubunji, Tokyo, Japan

Abstract:

Ion implantation apparatus for both processing research and small-scale pilot production of semiconductor devices has been constructed. This system is designed to implant boron and phosphorous ions into silicon substrate up to the energy of 200 keV, and is featured by the following specifications; higher output analyzed ion beam density (1 A/cm^2), broad area scanning (100 cm^2) and quick exchange of substrates with heated or cooled tray. The operating characteristics in various conditions are discussed in detail.

1. Introduction.

The ion implantation technique has been used for the purpose of introducing impurities into semiconductor substrates. It has several advantages when compared with conventional thermal diffusion and because of them ion implantation is becoming one of the most important processes in the manufacture of semiconductors.

The main features of ion implantation are itemized below:

1. High purity of doping elements.
2. Possibility to monitor the quantity of introduced impurities during implantation.
3. Impurity densities can be controlled over a wide range.
4. Excellent uniformity of impurity distributions.

5. Good control of junction depth may be easily achieved.
6. Processing temperatures are low.

Several types of ion implantation apparatus are now commercially available and special purpose machines are also constructed in some of the research laboratories^{1,2)}. All of these machines however are for research use and commercial semiconductor device production have not been initiated as yet. We have constructed a new type ion implanter both for processing research and for small scale pilot production of semiconductor devices. The equipment was designed aiming at:

- a. Capability of handling large quantity of substrates, and
- b. Reliable and easy operation.

2. Apparatus.

Fig. 1 shows schematic drawing and photograph of the newly developed ion implanter. Duoplasmatron, high frequency and Nielsen type ion sources (1) are mainly used for generating boron and phosphorus ions. Discharging gases for ion sources are diborane (B_2H_6) and phosphine (PH_3) diluted with helium. The ion source part of the implanter is supported by an insulating support (2) through which high voltage power is supplied. The acceleration tube (3) is of the multi-stage type and the ion beam is analyzed by an analyzing magnet (5) which weights 4.500 kg and is hung and supported by a steel frame (6).

The analyzed ion beam is deflected and scanned by an ion beam scanner (7) into X-Y directions and is implanted uniformly into a large area of substrate at the target chamber (8). The

samples are introduced from the sample chamber (9). The ion beam intensity is measured by an ion beam monitor (10). The monitors are small Faraday cups and are scanned in X-Y directions just upon the target area to measure uniformities of scanned ion beam. After desired quantity of ions are implanted, the ion beam is stopped automatically by a shutter (10) inserted at the target area by the ion beam quantity integrator.

The evaluation systems are two 14 inch oil diffusion pumps (11) for the acceleration tube and the target chamber and a 6 inch pump for the analyzing chamber. Final vacuum is 1×10^{-7} Torr.

Power for the ion source and the acceleration tube are generated at high voltage transformers (13) and are fed by high voltage coaxial cables (14) through the ion source support (2). All the power supplies are controlled directly from the ground side. The stabilities of high voltage source and a magnetic current supply are maintained to 1×10^{-3} and 1×10^{-4} , at the maximum load, respectively. The maximum current of the high voltage power supply is 10 mA. All control systems are contained in a control pannel (15).

Table 1 shows a specification of the ion implanter. The ion energy is 200 keV maximum, and at this energy, H to Ar ions are required. Elements until Se can be implanted at 100 keV, ion energy is reduced in this case.

The intensity of entire ion beam is 1 to 3 mA before entering to the analyzing magnet, and after analyzing, the beam intensity at the target was 0.5 to $1.0 \mu\text{A}/\text{cm}^2$ when scanned into

the area of 10cm x 10 cm. Therefore, the time required for doping of regular device structures generally is very short. Implantations are generally carried out at room temperatures, and after implantation, annealing treatments are carried out at several hundred °C. Not implantations up to 800°C substrate temperature are also possible.

The substrates are inserted in a graphite cassette holder of 10cm x 10 cm size, and the cassette is put into the sample chamber from an atmospheric pressure then transferred to position for implantation in the target chamber by means of turning the table. The cassette contains 9 wafers of 1 inch diameter or 4 wafers of 2 inch diameter, and two cassettes are simultaneously put into the target chamber, one stands by while the other is implanting. The exchange time for the cassette from atmosphere to target chamber is about 10 min. In this apparatus, for ease in the exchange mechanisms and for substrate heating during implantations, the substrates are inserted horizontally.

Fig. 2 shows our small type duoplasmatron ion source. Since it has an insulating permanent ferrite magnet the ion source is made rather small compared with conventional sources. The whole body is air cooled. The maximum magnetic field is 1 to 5 mA.

Fig. 3 shows cassette and wafer holder. Shown in the upper part is a holder for one inch diameter wafers, while the lower part shows a holder for 2 inch diameter wafers. The wafers for implantation are held with this cassette and put into the right position of the target chamber from the atmosphere. After implantation,

the cassette is pulled out to the atmosphere again by the exchange mechanisms.

Fig. 4 shows an upper half of the implanter. From the left to the right windows, the ion source part, the acceleration tube, the vacuum pump and the analyzing magnet are shown. The ion source and the acceleration tube portions generate soft X-rays by the reverse acceleration of secondary electrons, and therefore, shielding is necessary. Fig. 5 shows the lower part of this apparatus, the sample chamber and its accessories are shown.

3. Results of Operation and Application.

Fig. 6 shows an example of mass spectrograph when small type duplasmatron ion source of Fig. 2 is used for generating phosphorus ions. Many side bands of phosphorus hydrogen compounds are seen, but the machine shows excellent mass resolutions during implantation.

Table 2 shows uniformity of phosphorus implantation into silicon substrates when the ion beam is scanned into area of 10 cm x 10 cm.

After room temperature implantation, the substrates are annealed at 800°C, 10 min., and implanted the phosphorus with its variations are measured from sheet resistivity. The results show rather small deviations in and between wafers compared with conventional thermal diffusion approaches.

Table 3 shows the comparisons of transistor characteristics which are made using implantations and diffusions. All the transistors are of the same teardrop type and have 120 micron diameter

emitters. Boron and phosphorus are used for making the base and the emitter. In case 1, base and emitter regions are both fabricated with thermal diffusion, and h_{FE} was 200, f_T was 500 MHz. In case 2, base region is fabricated by diffusion and emitter region is fabricated by implantation, i.e. a hybrid type. The h_{FE} was also 200 but f_T became 710 MHz. In case 3, both base and emitter regions are implanted, i.e. double implantation. The h_{FE} decreased to 80 but f_T increased to 880 MHz. (3).

Noticing the change of f_T s, the introduction of one implantation process in case 2 and two implantation processes in case 3, increased the value of f_T . This means precise junction control is possible by the introduction of the implantation process. When double implantation process is applied to a much fine pattern of 3 strokes, 20 microns wide emitter, f_T was 1.85 GHz. Attempts to fabricate self-aligned MOS field effect transistors also produced good results.

Experience acquired through the construction and operation of the ion implanter permits consideration of the next step, i.e. implantation apparatus for practical industrial uses. This apparatus should have computer control. With provision for selection of desired ion, energy, dose etc., by means of push button or a punched card the computer control implanter will permit to carry out the complete implantation process automatically.

4. Conclusions.

1. A new ion implanter was constructed for both semiconductor processing research and small scale pilot production of semicon-

ductor devices. This machine is a kind of prototype of future systems to be designed for industrial use of the implantation process.

2. Up to 9 wafers can be implanted simultaneously by scanning ion beam. Prompt exchange of wafer holders are also possible.

3. Uniformity of implanted impurity distribution and fine control of junction depth, which are one of the main features of implantation process, were successfully proved.

Acknowledgement.

The authors wish to express their sincere thanks to Drs. T. Kambara and H. Watanabe of Central Research Laboratory, Hitachi Ltd. for their encouragements and valuable discussions. This work was supported by the Research and Development Corporation of Japan.

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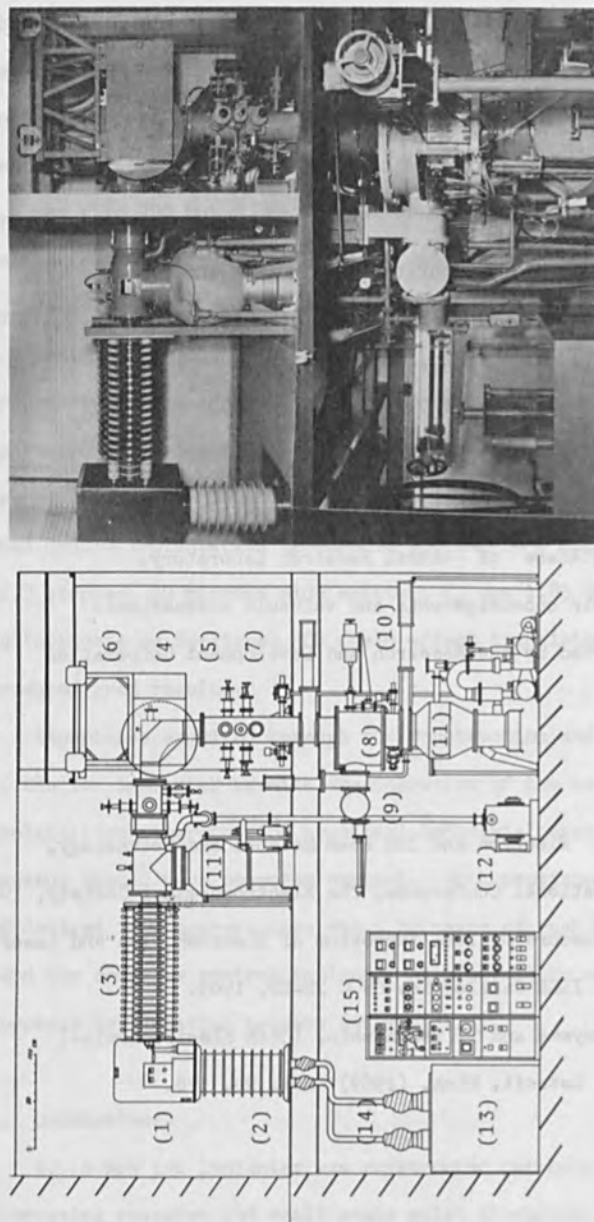


Fig. 1. Schematic Drawing and Photograph of Ion Implantation Apparatus: (1) Ion Source Part, (2) Insulating Supporter, (3) Acceleration Tube, (4) Analyzing Tube, (5) Steel Frame, (6) Ion Beam Scanner, (7) Ion Beam Monitor and Shutter, (8) Sample Chamber, (9) Oil Rotary Pump, (10) High Voltage Transformers, (11) Control Panel, (12) Voltage Coaxial Cable, (13) Oil Diffusion Pump, (14) Target Chamber, (15) Ion Beam Monitor and Shutter.

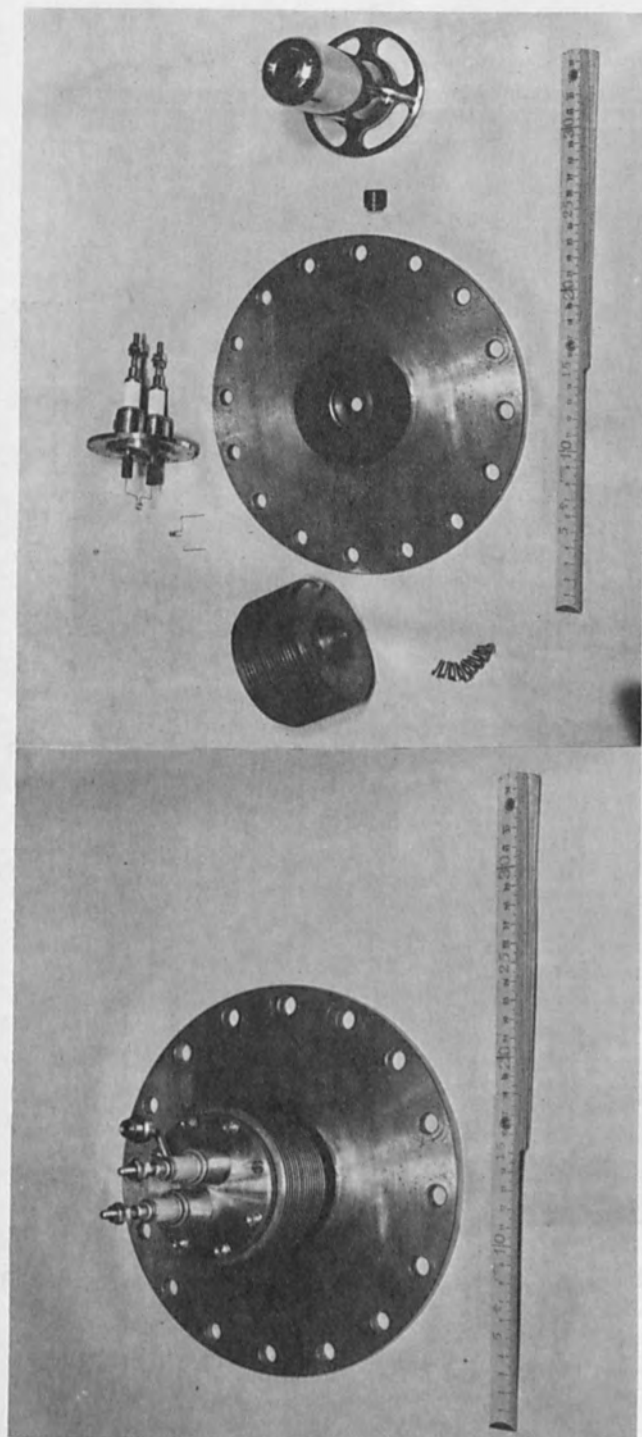
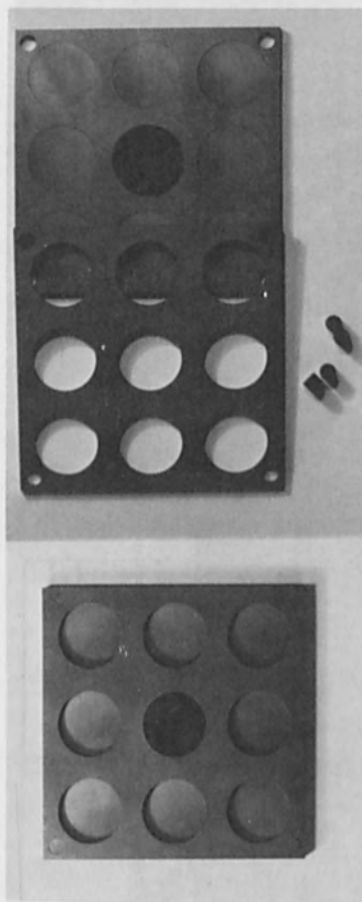


Fig. 2. Small Type Duoplasmatron Ion Source and Its Parts.

9 Wafers
(1 inch diameter)



4 Wafers
(2 inch diameter)

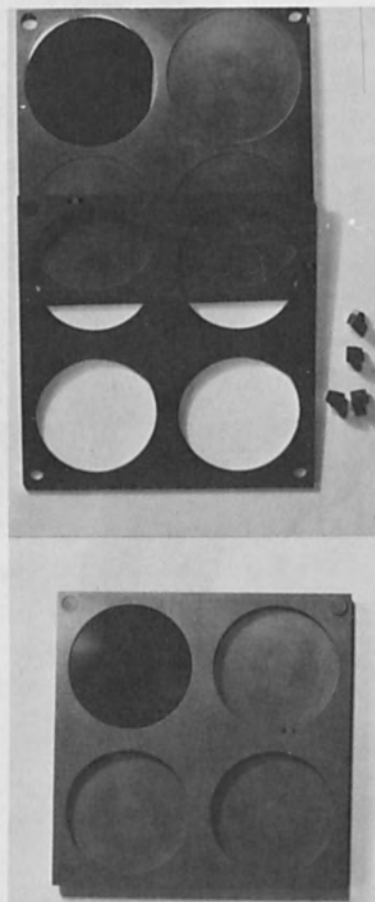


Fig. 3. Cassette of Wafer Holder.

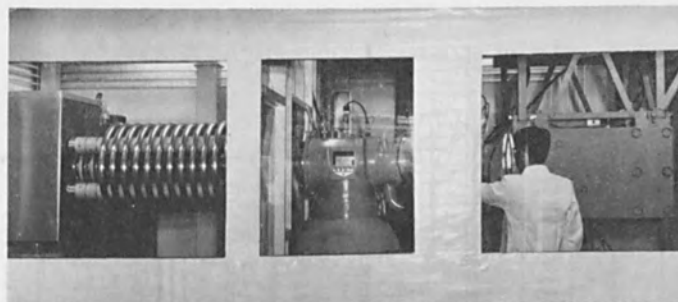


Fig. 4. Photograph of Ion Implantation Apparatus (upper part).



Fig. 5. Photograph of Sample Chamber and Its Accessories (lower part).

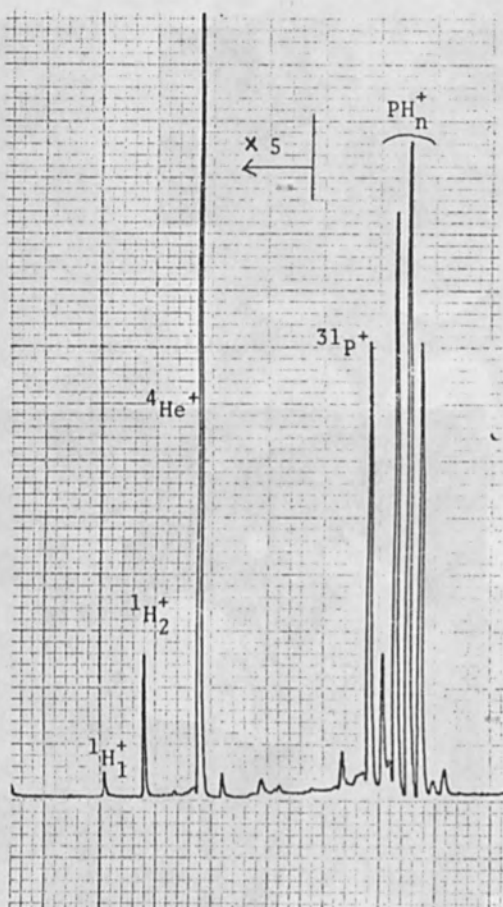


Fig. 6. Mass Spectrograph of Phosphorus. Energy, 200 keV.
Small Type Duoplasmatron Ion Source.

Table 1. Specification of Ion Implantation Apparatus

Acceleration Voltage	200 kV (maximum)
Ion Species	H to Ar (200 kV), H to Se (100 kV)
Substrate	Mainly Si and Ge
Ion Beam Intensity	1.0 $\mu\text{A}/\text{cm}^2$ (scanned to 10 cm x 10cm)
Substrate Temperature	Room Temperature to 800 $^{\circ}\text{C}$
Exchange of Substrate	Quick exchange using turn table and cassette. 9 wafers (1 inch diameter) or 4 wafers (2 inch diameter)
Analyzing Magnet	Radius; 40 cm, Field Intensity; 1×10^4 gauss, Resolving Power; over 100

Table 2. Sheet Resistivity and Its Deviation of Phosphorus Implanted Samples

In a Wafer		
Surface Density (cm^{-3})	Sheet Resistivity ($\text{ohm}/\square$)	Deviation (%)
7×10^{18}	2.85×10^2	± 0.1
7×10^{19}	1.75×10^2	± 0.1
Between Wafers (10 cm x 10 cm Scanned Beam)		
5×10^{18}	4.14×10^2	± 3.1

Table 3 Comparison of Transistor Characteristics

Case 1	Case 2	Case 3
		
Base : Diffusion Emitter : Diffusion Double Diffusion	Base : Diffusion Emitter : Implantation Hybrid	Base : Implantation Emitter : Implantation Double Implantation
$h_{FE} : 200$ $f_T : 500 \text{ MHz}$	$h_{FE} : 200$ $f_T : 710 \text{ MHz}$	$h_{FE} : 80$ $f_T : 880 \text{ MHz}$ ($f_T : 1.85 \text{ GHz}$ with 3 Stripe Pattern)

USE OF AN ELECTRON MICROPROBE TO STUDY ION IMPLANTATION

Dr Ch. LEGRAND and Professeur J.J. TRILLAT

Centre National de la Recherche Scientifique (C.N.R.S.)
Laboratoires de BELLEVUE, 92 (FRANCE)

ABSTRACT: Electron microprobe can be used efficiently to control the implantation of ions in the bombarded targets. In the case of thin films, the sputtering rate can be studied. The authors have been interested in Argon ions energy of which vary between 5 and 30 KV.

INTRODUCTION -

We propose to show in this work how the electron microprobe can be used with efficacy to control ion implantation in targets submitted to ion bombardment of any nature. In the literature relevant to the study of ions paths in solids, we find a large number of references (1) relative to methods based on the use of radioactive ions, or on a later excitation by a radioactive process, but very few on the method of primary excitation, through electrons of X radiation characteristic of implanted ions (2). Our first experiments have show that this last method is very efficient.

1. TYPE OF BOMBARDMENT USED -

We have used an "Eolion" ion device, developped and built by the C.S.F. This device having already been described in ref. (3), we shall only describe its essential characteristics:

- 1) Pulse mode operation: with the modulation we use, the length of pulses is 25 microseconds, which for a 50 cycles frequency current gives a impulsional factor of 800. This exceptionally high factor - adopted for the bombardment of insulators - shields us from temperature rises.
- 2) The mass range is 1 to 40.
- 3) The energy of the ions can vary between 5 and 30 KV.
- 4) The diameter of the beam at the level of the target is important, it can reach 10 cm.
- 5) The maximum opening angle is 2θ with $2\theta \leq 60^\circ$.

- 6) The repartition of the ions intensity in a straight section on the beam must be as uniform as possible.
- 7) The current is constant through the operation.
- 8) The ion source is of duoplasmatron type (4).
- 9) The gaz pressure above the target is 2×10^{-5} Torr.

In this work, we have used exclusively: Argon ions at 20 KV voltage and a current of 8 mA. (Fig. 1) reproduces the profile of the energetic density of the ion beam at the level of the target holder; this profile is obtained directly with the measuring probe and the oscilloscope; we find that, with the exception of the center of the field where the intensity increases, we have a large crown (of a width of about 3 cm) where specimen of a few square centimeters can be bombarded in a uniform manner. These intensity measurements of radial distributions are repeated during the bombardment; they shown a satisfactory reproductibility ($\pm 5\%$).

In the experiments described hereafter, the effective bombardment duration have not been above 60 s. thus giving a little more than 13 hours in experimental lengths.

The device we used allows us, in particular to come to very small length of effective bombardment, 0,1 s. for instance. Also the bombardment of larger surfaces allows us to observe more easily the modification of certain properties: surfaces potentiels, corrosion, passivation, etc ...

2. MEASUREMENTS WITH THE MICROPROBE -

The implantation of ions being always - specially with relatively low voltages such as the ones we used (20 KV) - to depths of a few hundreds of angströms the characteristic radiations of implanted ions are easily excited by electrons; we have also conducted tests with secondary excitation (X Ray Fluorescence); we have indeed detected the presence of ions A, but the sensitivity of measures was too small to allow quantitative measurements, this because in the targets used the path of primary X Rays is much more important than the path of electrons and the efficacy of excitations is thus much raduced. In consequence, only the primary excitation mode must be retained. In this case, the experimental technique is relevant of macroprobe or of microprobe. We have experimented with the microprobe, technique to be used if one wants a local analysis; in the opposite case, where one must integrate the result to a large surface - for instance for a polycrystalline target - a macroprobe would give a larger signal, authorizing thus the use of smaller

electronic densities.

In the first case, it is possible to study a large enough surface by using a sweep technique, thus the local variations are integrated, and the probe current is decreased.

Because the number of ions to be detected is relatively small, one must use a dispersive crystal with a reflecting power as high as possible. The results presented hereafter have been obtained with a curved quartz crystal (1010) (Cameca microprobe) the emergence angle being 18°. Our future work will benefit from a significant increase of the signal obtained with a Pentaerythritol crystal (P.E.T.). The excitation voltage must be selected to obtain the best signal/Noise ratio, 15 KV in our case. The probe current must be regulated; if the necessary current exceeds the intensity compatible with a satisfactory regulation, one must use frequently a stable calibration.

We have verified that, in the case of metallic targets, a succession of countings of 100 s. each gives a regular follow through, which eliminates the fear of an Argon desorption under the electrons impact. It is equally important to define the surface state of the target. One knows that the microprobe technique needs a surface as flat as possible and thus implies a mechanical polish. This operation creates a work hardening to a greater depth than the one attained by implantation phenomena; the kinetic of ion penetration can be influenced by the texture of superficial layers. In consequence one must always define as exactly as possible how the surface to be bombarded has been obtained.

3. RESULTS -

3.1.1. BOMBARDMENT OF MASSIVE POLYCRYSTALLINE TARGETS -

We have used the following elements: Al Ni and Au. Copper has the inconvenience of superposition of the $\text{CuK}\beta$ line of 3rd order to the $\text{K}\alpha$ of argon.

The bombardments have varied between 0,1 s. and 60 s. a shutter mechanism allows us to bombard predetermined spots of the target without breaking vacuum (Fig. 2) shows in logarithmic coordinates, the results of measures for Al and Au. In the limits of fixed ions, it is legitimate to admit the proportionality between the quantity of implanted argon and the number of impulsions/sec. recorded by the counter.

These curves show clearly two phases in the implantation kinetics: After a time of about 20 s. during which the implantation allows

a linear law, we attain a second phase in which saturation occurs. Experiments, which will be related in a future publication, show that the introduction of ions happens first in the crystalline lattice. Then, the ions are fixed with more difficulty thanks to the grain boundaries, and to the lattice defects created by the bombardment itself.

The dispersion of measures for the different lengths of bombardment shows it clearly: For small lengths the measures are very coherent when we vary the sweeps field submitted to the probe on the target. From 40 s. on and mostly for Au and Ni, the dispersion of measures increases translating thus a much less homogeneous ions repartition.

3.1.2. DESORPTION OR DIFFUSION OF IMPLANTED IONS BY THERMAL TREATMENT -

The bombarded targets have been submitted to temperatures of 150, 250, 350, 450 and 550° C. in a vacuum furnace (10^{-6} Torr) during one hour, and there checked with the microprobe. For the first two temperatures, we find no significant variation. Fig. 3, 4 and 5 show how, for temperatures of 350° C. and 450° C. the quantity of ions visible to the probe vary. For Al the ions A fixed during the first 10 s. are not very mobile; it is not the same for Ni: in this metal, the decrease of ions detected by the probe manifests itself even for short bombardment times. Gold seems to fix ions in a more energetic fashion: there is no real difference between the relative curves at 20° C. and 350° C. For 450° C., there is a less in argon, only for the longest bombardment times. These experiments reveal the greater stability of ions A fixed on the target but give no explanations on the nature of the process that removes ions from electron detection. In effect, there can happen a desorption or a diffusion inside the specimen or also the two phenomena simultaneously. More complex experiments done with thin films will be needed to clarify this point.

3.2. BOMBARDMENT OF THIN FILMS -

One of the objectives of researches done on ion implantation is to determine the distribution of ions in depth. With massive targets, one must, to do this, use the attack methods (Sectioning methods) such as the ones described by Davies and Coll. (5) or ion bombardment done with other type of ions. Thin films seem to offer possibilities that have not been exploited up to now.

3.2.1. FOR THE CONTROL OF ION IMPLANTATION -

If we increase the thickness of the film, a relation can be established between the quantity of fixed ions and the thickness of

this film; it is thus possible to apply this differential method to the control of ion distribution.

These thin films must be deposited on substrates on which the implantation is small. This method is illustrated by Fig. 6. Al thin films have been obtained by thermic sputtering on a thick substrate of Platinum very flat and well polished. Bombardment of increasing lengths have been done on each zone of known Al thickness. It is clear that a thickness of 100 Å let ions A go through and that on 800 Å thickness fixes an important number of ions.

To obtain from such experiments precise deductions, films of systematically increasing thickness should be used and controls should be used and controls should be effected on the substrate once the Al film had been removed by chemical etching.

3.2.2. FOR THE STUDY ON SPUTTERING RATE -

In the range of small thickness, the responses of the microprobe follows a linear law. It is thus easy to study the decrease of thickness of such films, either by direct measurement of the emission of the film element, or by absorption in this film of an excited radiation in the substrate. Of course, first the process must be calibrated. In the case of this experiment we have found that the sputtering rate of Al was higher during the first seconds of bombardment. A possible explanation is that, when the bombardment starts, the ions interact essentially with target atoms, then as the bombardment goes on, with a material where are already implanted.

4. CONCLUSION -

The electron microprobe is useful for the study of ion implantation kinetic in various targets: Metals and alloys being the most favorable cases for they allow the use of high enough electron currents to excite notably the A radiation $K\alpha$; in the case of insulating materials, the probe current must be limited. The same technique gives data on the departure of ions under various influences, thermal treatment for instance.

The bombardment on thin film targets can be used to precise the ion repartition and to study the kinetics of sputtering rate.

For a target of a known nature, the results obtained present no interpretation difficulties. But in the case of different targets, the comparison of experimental data calls for the use of various corrections. In particular, account must be taken of the variation of the

absorption coefficient of the target element for the characteristic radiation of the ion, of the slowing law of electrons for the voltage used and lastly of the retrodiffusion coefficient of electrons. The determination of the sputtering rates does not present the same difficulties; it is enough to have the calibration curves obtained at predetermined conditions.

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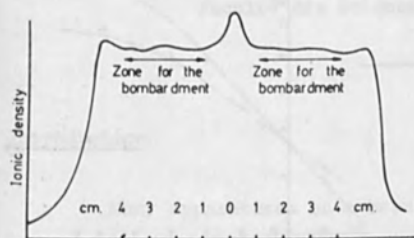


FIG. 1. Repartition of the ionic density on the sample holder.

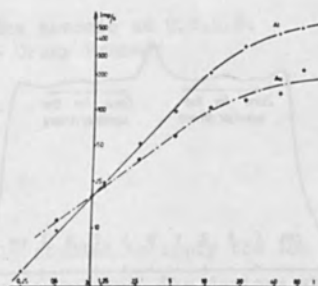


FIG. 2. Ions A implantation in Al and Au.

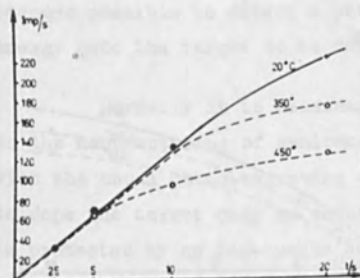


FIG. 3. Decrease of implanted ions in Al by thermal treatment.

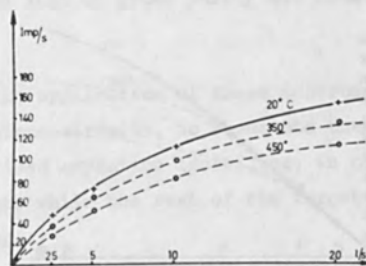


FIG. 4. Decrease of implanted ions in Ni by thermal treatment.

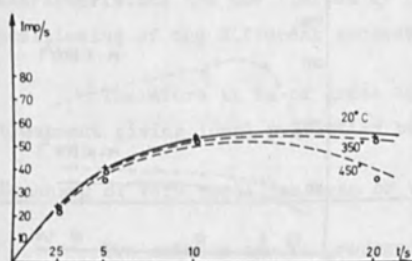


FIG. 5. Decrease of implanted ions in Au by thermal treatment.

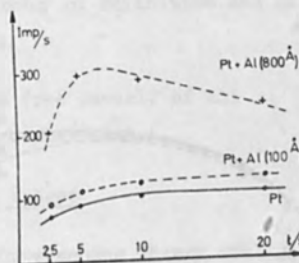


FIG. 6. Implantation of ions A in thin films Al deposited on Platinum.

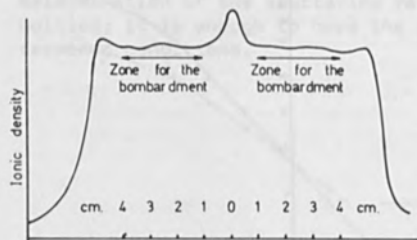


FIG. 1. Repartition of the ionic density on the sample holder.

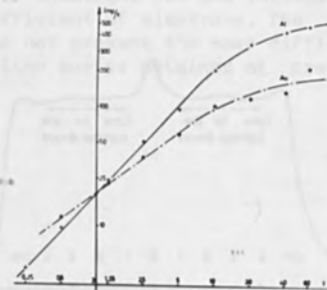


FIG. 2. Ions A implantation in Al and Au.

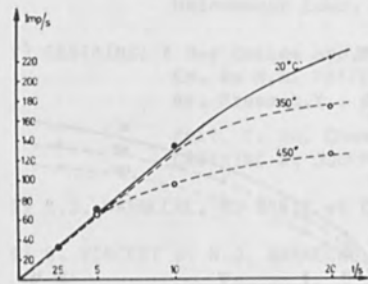


FIG. 3. Decrease of implanted ions in Al by thermal treatment.

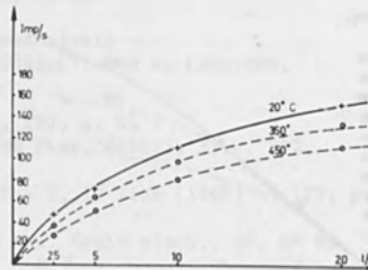


FIG. 4. Decrease of implanted ions in Ni by thermal treatment.

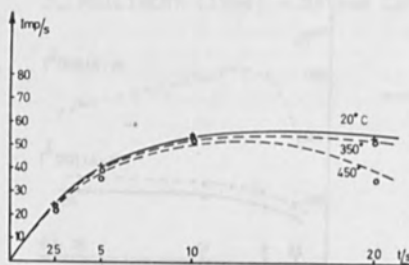


FIG. 5. Decrease of implanted ions in Au by thermal treatment.

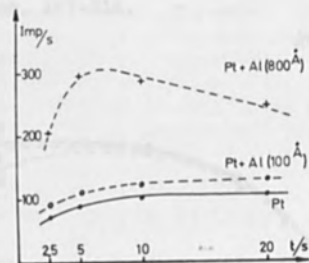


FIG. 6. Implantation of ions A in thin films Al deposited on Platinum.

Ion Implantation Using an Ion Image

M. Bernheim

Laboratoire de Physique des Solides associé au C.N.R.S.
Faculté des Sciences, 91 - Orsay France

Introduction

Many apparatuses have been recently studied to apply the ion implantation technique to the doping of semiconductors. The devices which have been contrived are mainly composed of an ion source followed by simple corpuscular optic elements filtering and accelerating the beam; it thus becomes possible to direct a parallel ion beam of great purity and high energy onto the target to be doped.

Normally it is necessary, for the application of these instruments to the manufacturing of semiconductors micro-circuits, to associate them with the usual photo-engraving and localised oxydation techniques, in order to dope the target only on certain regions while the rest of the target is protected by an ion-opaque oxyde layer.

But photo-engraving operations are one of the most costly in the manufacturing of micro-circuits, and above all the electronic characteristics are now limited by the inaccuracy of definition and of positioning of the different successive masks.

Therefore it is of great interest to free oneself of all treatment giving local protection before ion bombardment.

Scanning of very small ion beam or using ion images

Two methods can be proposed either by scanning a very small ion beam on the semiconducting surface (by electrical deflection of the beam or mechanical shifting of the target), or by focusing an image formed with the doping ions on the semiconductor surface in such a way to direct the ions only on the regions to be doped.

The reason while we use an ion-image apparatus proceeds from working time comparisons. As we show in appendix, an imaging device can use larger ion sources and consequently works a thousand times faster than a scanning instrument.

Apparatus principle

The apparatus we describe here is derived from the one we have previously studied (1, 2, 3) concerning the micromachining by ion sputtering.

It consists essentially of an electrostatic lens which focusses on the target surface an image produced with the doping ions of a model constituted by a perforated structure partially opaque to the ions.

In order to obtain high precision doping and to avoid difficulties in the manufacturing of the model, the ion optics is a reducing lens. Consequently the ion beam density on the model is much smaller than on the target and the same is true for the sputtering rate.

This reducing optics is associated with filtering and focusing elements for illuminating the model with a converging beam of great purity, directing on the target all the ions which have crossed the model.

A double magnetic prism (see figures 1 and 2) makes possible to use two ion sources to dope successively P and N in different regions by changing only the model, the working ion gun and the magnetic field. Therefore many models are fixed on a sliding bar the position of which can be checked within two microns. Such an accuracy is sufficient because of the ion reduction factor (about 20), to localize the doped areas with an accuracy of 0.1 micron.

One of the difficult points is connected with the fine focusing of the ion image of the model just on the target surface. In order to check this focusing, we use an ion - electron image converter (5) as a reducing lens :

- the ion bombardment extracts secondary electrons from the bombarded surface ; those electrons are accelerated and focused by the electrostatic field of the converter, permitting the observation on

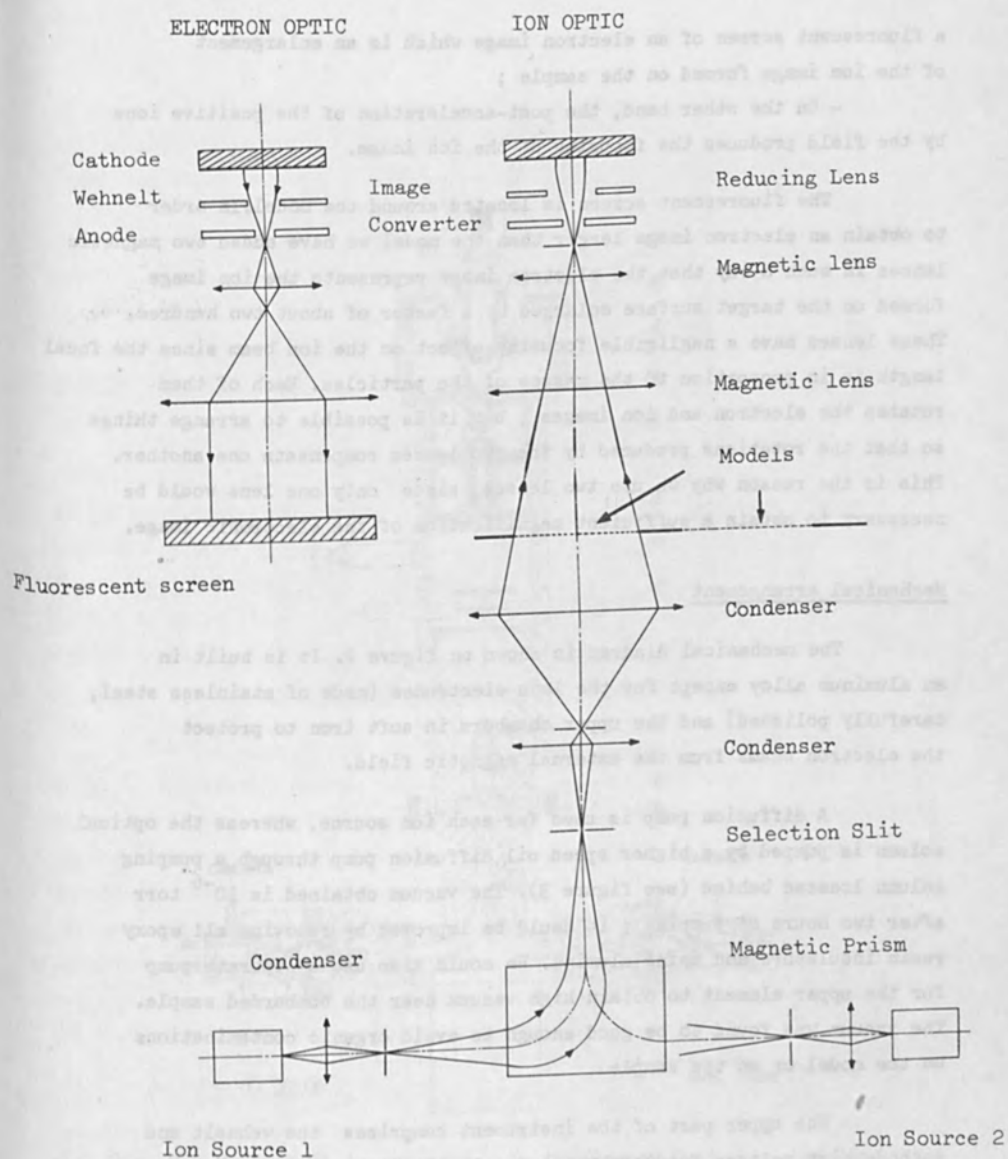


Fig. 1. Optical Diagram. The electron and ion optics are really superposed.

a fluorescent screen of an electron image which is an enlargement of the ion image formed on the sample ;

- On the other hand, the post-acceleration of the positive ions by the field produces the focusing of the ion image.

The fluorescent screen is located around the model. In order to obtain an electron image larger than the model we have added two magnetic lenses in such a way that the electron image represents the ion image formed on the target surface enlarged by a factor of about two hundred. These lenses have a negligible focusing effect on the ion beam since the focal length is in proportion to the masses of the particles. Each of them rotates the electron and ion images ; but it is possible to arrange things so that the rotations produced by the two lenses compensate one another. This is the reason why we use two lenses, since only one lens would be necessary to obtain a sufficient magnification of the electronic image.

Mechanical arrangement

The mechanical diagram is shown on figure 2. It is built in an aluminum alloy except for the lens electrodes (made of stainless steel, carefully polished) and the upper chambers in soft iron to protect the electron beams from the external magnetic field.

A diffusion pump is used for each ion source, whereas the optical column is pumped by a higher speed oil diffusion pump through a pumping column located behind (see figure 3). The vacuum obtained is 10^{-6} torr after two hours of pumping ; it could be improved by removing all epoxy resin insulators and using alumina. We could also use a separate pump for the upper element to obtain high vacuum near the bombarded sample. The vacuum was found to be good enough to avoid organic contaminations on the model or on the sample

The upper part of the instrument comprises the wehnelt and cathode high voltage feedthrough, and a small chamber to remove the bombarded sample without breaking the vacuum.

The two ion sources consist in a low voltage arc with axial magnetic field (4). They provide ion beams whose intensity after filtering

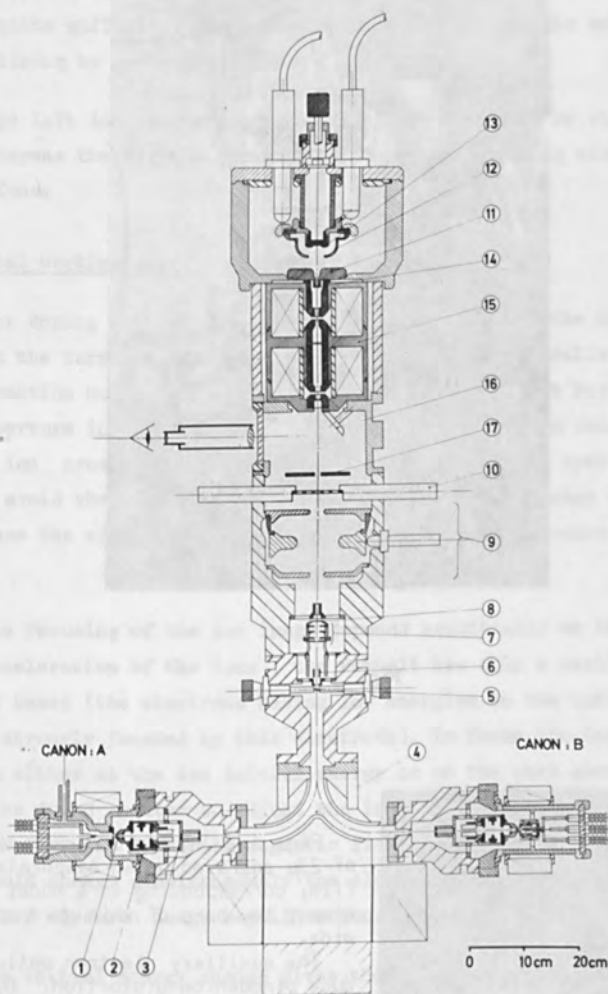


Fig. 2. Mechanical Arrangement: 1--Ion Source Exit Hole, 2--Condenser, 3--Aperture at the Stigmatic Point of the Magnetic Prism, 4--Magnetic Prism, 5--Selection Slit, 6--Faraday Cup, 7--Condenser, 8--Centering Plates for Preliminary Adjustment, 9--Condenser, 10--Sliding Bar Supporting the Models, 11--Anode, 12--Wehnelt, 13--Cathode and Sample Holder, 14. 15--Magnetic Lens, 16--Optical Mirror, 17--Fluorescent Screen.

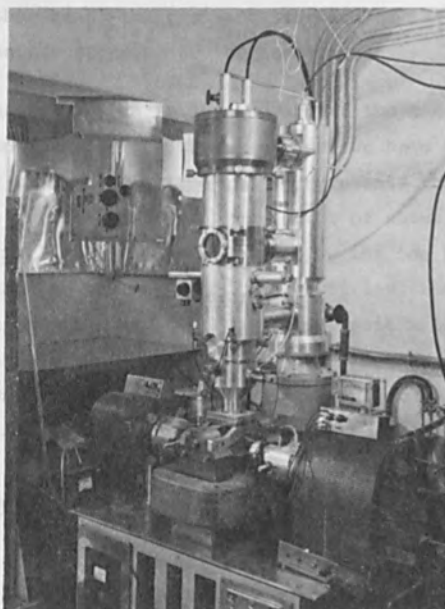


Fig. 3

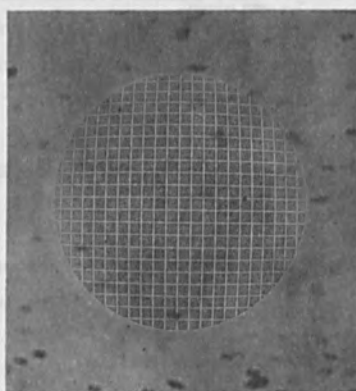


Fig. 4. Light micrograph of a grid of 23μ engraved on an evaporated copper film, corresponding to a model of 400μ steps; the bars of this grid are 2.5μ wide.

The auxiliary electron optics introduce pincushion distortion. By focusing the ion image through the optics, we introduce unfortunately on the first experiments a barrel distortion to compensate this electron optics defect. After a few experiments we were able to suppress all distortion.

is about $0.5 \mu\text{A}$, corresponding to densities of $1 \mu\text{A}/\text{mm}^2$ at the target surface, quite sufficient for semiconductor doping, or thin metallic sheet machining by using the cathodic sputtering.

The left ion source produces ions from any gases or white phosphorous vapour, whereas the right source can be used for producing aluminum or boron ions.

Experimental working conditions and preliminary results

For doping experiments using channeling effects, the image focused on the target surface must be illuminated by a parallel ion beam in the direction normal to the target. For this purpose we have disposed a small aperture in the object focal plane of the reducing lens to focus there the ion cross-over (image of the ion source). This aperture is large enough to avoid the partial occultation of the electron image which could be feared since the electron and the ion cross-over are not located at the same point.

The focusing of the ion image depends essentially on the degree of post acceleration of the ions ; the wehnelt has only a negligible action on the ion beams (the electrons having low energies in the cathode-wehnelt space are strongly focused by this electrode). To focus the ion image we can act either on the ion initial energy or on the post accelerating voltage : we found better to adjust the ion initial energy and correct at the same time the filtering magnetic field. Modifying the post accelerating voltage would change simultaneously the electron energy and consequently the auxiliary electron image magnification.

The initial energy change gives the best sensivity, but this method requires the experimenter's memory. The experimental results are in perfect agreement with the computed ion trajectories.

To appreciate the quality of the ion focusing image we prefer micro-machining to ion implantation experiments.

The ions sputter a metallic polycrystalline target in all regions corresponding to the model's openings and fashion a positive replica of the model. By observing the engraved sample with a classical metallurgical microscope, we measure directly the ion reduction and appreciate the ion image resolution, the distortion defects and the uniformity of illumination.

The figures 4, 5, 6 show some engraved targets photographs.

By post accelerating to 112 keV ions whose initial accelerating voltage is 12 kV, one produces, for the geometry of our converter, an ion image deduced from the model by a reduction factor of 17 ; the ion image diameter can be as high as 600 microns, corresponding to a model of 10 mm and the resolution of this image is about one micron despite the poor quality of the electron image :

Indeed the secondary electrons are emitted by the bombarded surface with energies of the order of a few electron volts, which produces aperture and chromatic aberrations; the resolution of the electron image (attaining only 4 microns) depends on the electrostatic field on the target surface only. If one increases this field, one increases simultaneously the ion reduction factor, reducing at the same time the ion image diameter ; it is impossible to compensate this effect by enlarging the model size without having an apparatus of larger dimensions.

The only possible process is to dispose a small aperture at the converter electron cross-over, to eliminate the electrons corresponding to the higher transverse initial energies. In that way it will be possible to check the ion image focusing with a high accuracy, but the bombarded region will certainly be strongly reduced ; after focusing the ion optics it will be necessary to remove this aperture and control the ion illumination uniformity and the image distortion on the whole field.

This work has been supported by the Délégation Générale à la Recherche Scientifique et Technique.

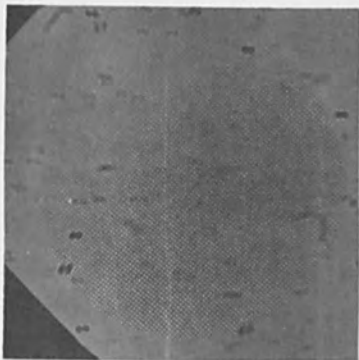


Fig. 5a

Grid of 5.9 microns corresponding to a model of 100 μ

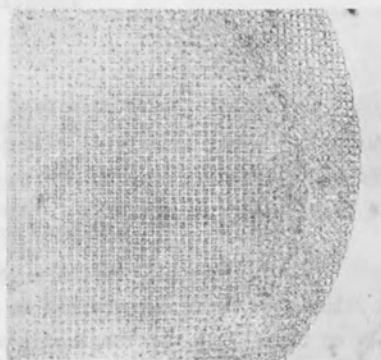


Fig. 5b

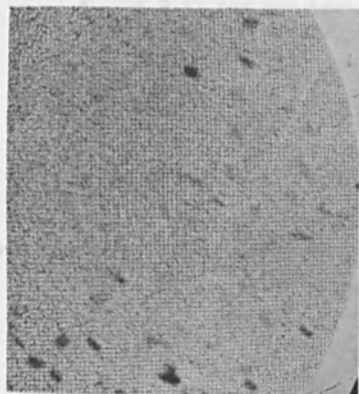


Fig. 6. Grid of 3 microns corresponding to a model of 50 microns.

Appendix

Ion sources have much less emittance and brightness than electron sources and their transverse size is not negligible (6, 7, 8). It is difficult indeed to obtain intense ion beams of small angular divergence and small energy spread.

In our imaging apparatus each point of the source is used to illuminate the model whereas in an ion probe apparatus one focalises only on the target the image of a very small part of the ion source.

Consequently the mean density on the whole image is comparable to the peak density of an ion probe ; the scanning of such a probe would lead to mean densities lower by a factor of about 1,000.

An ion current reaching 10^{-10} A has been obtained on a surface of $4 \mu^2$ (4) ; we have $1 \mu\text{A}/\text{mm}^2$ on a diameter of 600μ ; in spite of the fact that our ion density is 60 times less, the ion imaging apparatus works 1.000 times faster than a scanning device using a 2μ probe, and the resolving power is twice better.

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ELECTRON BEAM INSPECTION OF TANTALUM THIN FILMS

A. E. DUGAN

Western Electric Company, Inc.
Engineering Research Center
Box 900
Princeton, New Jersey 08540

ABSTRACT

The secondary electron and backscattered electron emission properties of sputtered β tantalum and bcc tantalum thin films have been measured for primary electron energies $0.2 \leq E_p \leq 2.0$ keV. The backscattered electron emission coefficient η was found to be similar for the two phases of tantalum. η increased from 0.25 at $E_p = 0.2$ keV to 0.41 at $E_p = 2.0$ keV. However, the secondary electron emission coefficients δ differed appreciably for the two phases. δ was found to be larger for β tantalum over the primary electron energy range investigated. The maxima in the secondary electron emission curves occurred at $E_p = 0.4$ keV for both phases with $\delta_{\max} = 2.67$ for β tantalum and $\delta_{\max} = 2.32$ for bcc tantalum. Measurements of the secondary electron energy distributions revealed maxima at ~ 4 eV for both tantalum phases.

A simple low energy scanning electron beam inspection system has been constructed. It resembles a scanning electron microscope in that it utilizes an electron beam as a probe. The electron beam is produced with a commercial CRT electron gun. Scanning electron micrographs have been obtained of sputtered β tantalum thin films using this inspection system. These micrographs reveal the presence of bcc tantalum as well as other defects.

INTRODUCTION

Sputtered tantalum thin films may have either a tetragonal or a body centered cubic (bcc) crystal structure⁽¹⁾ depending upon the sputtering conditions. The former structure, referred to as tantalum, is preferred in the fabrication of tantalum thin film capacitors.

The secondary electron and backscattered electron emission properties of bcc tantalum and β tantalum have been measured over the primary electron energy range 0.2 - 2.0 keV. It was found that although the backscattered electron emission properties were very similar for both phases of tantalum, the secondary electron emission properties were different. Based upon this difference, a low energy scanning electron beam inspection system has been constructed. This system has successfully been employed to detect the presence of bcc tantalum as well as other defects in sputtered β tantalum thin films.

The results of the electron emission measurements of tantalum thin films are presented and discussed. The scanning electron beam inspection system is described and a micrograph obtained using this system is presented.

EXPERIMENTAL

Tantalum Thin Films

The tantalum thin films used in this study were sputtered on glass substrates under a variety of sputtering conditions. The crystal structure of the films was determined by x-ray analysis. The bcc tantalum thin films had a slight preferred orientation of the (110) axis normal to the substrate surface. All samples were subjected to a detailed cleaning process to remove surface contaminants.

Secondary and Backscattered Electron Measurements

When an electron of energy E_p is incident upon a solid, a number of electrons are emitted from the surface. For purposes of measurement and discussion, the emitted electrons are divided into two groups. Those electrons with energies less than 40 eV are defined as secondary electrons. Electrons with energies greater than 40 eV are referred to as backscattered electrons.

Backscattered electrons originate near the surface. A large portion of these electrons are primary electrons which suffer nearly elastic reflection from the first few atomic layers of the surface. It is generally agreed⁽²⁾ that the number of backscattered electrons

emitted from a solid for a given primary electron energy is largely a function of the atomic number of the target material. However, at low primary electron energies Sternglass⁽³⁾ has found that the backscattered electron yield is also a function of the atomic electron structure.

Secondary electrons are created when the primary electrons penetrate the surface and are scattered in the solid transferring energy to other electrons. Many of these electrons migrate to the surface and are emitted if energetic enough to penetrate the surface potential barrier. The number of secondary electrons emitted is dependent upon such parameters as the average depth of creation of the secondaries from the surface, surface condition, and the work function of the material.⁽⁴⁾

The secondary electron emission coefficient δ is defined as the ratio of the number of secondary electrons emitted to the number of primary electrons incident. Similarly, the backscattered electron coefficient η is defined as the ratio of the number of backscattered electrons emitted to the number of primary electrons incident.

If the target is grounded, a relationship exists between the primary current i_p , the backscattered current i_η , the secondary electron current i_δ , and the current i_t flowing between target and ground.

$$i_p = i_t + i_\delta + i_\eta$$

Measurements of these various currents allow a direct determination of η and δ .

The secondary electron and backscattered electron emission properties were measured with the apparatus schematically shown in Figure 1. The electron collector consists of a solid hemispherical dome. The suppressor grid is concentric with and lies inside the collector dome. Both collector and grid have an apex aperture through which a drift tube projects. The primary electron beam enters through the drift tube and impinges on the target. The target is mounted on one face of a rotatable cube equipped with a Faraday cup and a ZnS viewing screen. The apparatus is mounted in an ion pumped vacuum chamber. The currents to the collector, grid, and target are simultaneously measured with Keithley #602 and #610 electrometers. Typical primary currents were in the 10^{-10} ampere range.

The following procedure was used to measure the secondary electron emission coefficient δ and the backscattered electron emission coefficient η . The primary electron current at a given

voltage was measured by positioning the faraday cup to receive the beam. The dome and grid (switch in position #1) were biased at +120 volts. The currents i_c and i_t were measured and the primary current was then taken to be equal to i_t . The thin film was then positioned to accept the primary electron beam. The current i_c' and i_t' to the collector-grid and to the target respectively were measured. Next the grid was biased -40 volts (switch in position #2) and the currents i_c'' , i_g'' and i_t'' to the collector, grid and target respectively were measured. Finally the value of i_t was again measured with the faraday cup receiving the beam to insure that the primary current had not drifted during the measurement procedure outlined above.

The secondary electron emission coefficient and the back-scattered electron emission coefficient were calculated from the relationships

$$i_\delta = \frac{i_c' - (i_c'' + i_g'')}{i_t}$$

$$i_\eta = \frac{i_c'' + i_g''}{i_t}$$

Using the above procedure the measurements of δ and η were reproducible to better than 1%.

The energy distribution of the secondary electrons was measured using a retarding field technique. The current to the collector was continuously measured and recorded as a time varying negative bias was applied to the grid. Thus at any given time only those electrons with energies large enough to overcome the negative grid bias were able to reach the collector.

ELECTRON BEAM INSPECTION SYSTEM

The electron beam inspection system resembles a scanning electron microscope in that it uses a scanning electron beam as a probe. The secondary electrons and the current flowing from target to ground are monitored and the signals are displayed on an oscilloscope. The system differs from a commercially available scanning electron microscope in that 1) the electron beam is produced and focused using a commercial CRT electron gun, 2) the energy range of the primary electrons is 0.2 - 2.0 keV, considered low for scanning electron microscopy, and 3) the area of beam scan on the target can be $> 4 \text{ cm}^2$, large in comparison to a scanning electron microscope.

Fig. 2 is a schematic diagram of the electron beam inspection system. It consists of two major subsystems: (1) an electron gun and the associated beam control electronics and (2) the detection systems for secondary and backscattered electrons and for target current. The electron beam is incident upon the target at $\sim 30^\circ$ to the normal. The electron gun, target material, and secondary electron detector are housed in a vacuum chamber equipped with a liquid nitrogen trapped oil diffusion pump. The target holder is mounted on an X-Y table to allow positioning of various areas of the target for inspection.

The electron beam is produced by a commercial CRT electron gun modified with a tungsten filament to allow recycling of the gun to atmosphere as target materials are changed. The gun is capable of producing electrons with energies in the range 0.1 to 2.0 keV and beam currents up to 5×10^{-5} amps. The electron beam can be focused to $\sim 50 \mu\text{m}$ spot at the target. The scan of the electron beam is controlled by applying the outputs of Tektronix time bases (2B67) through a magnification control to the deflection plates of the electron gun. The magnification control unit allows control of the area of raster scan on the target. An area of up to 4 cm^2 can be scanned with the present geometric arrangement of gun, target and electron detector.

The measurement of the current flowing from target to ground is made using a Keithley #610 electrometer. The secondary electrons and the backscattered electrons are monitored with a conventional scintillator--light pipe--photomultiplier combination.⁽⁵⁾ This detector was fabricated using a NE-110 (Nuclear Enterprises) scintillation material. The output signals from the electrometer and/or the photomultiplier are applied to the Y axis (amplitude modulation)⁽⁶⁾ or the Z axis (intensity modulation)⁽⁷⁾ of an oscilloscope that synchronously scans with the electron beam on the target.

RESULTS

Secondary and Backscattered Electron Emission Properties

Figure 3a is a plot of the backscattered electron emission coefficients η for β tantalum and bcc tantalum as a function of primary electron energy E_p over the energy range 0.2 to 2.0 keV. The backscattered electron emission coefficients are very similar for both phases of tantalum. The value of η is nearly a linear function of E_p with η increasing from 0.25 at $E_p = 0.2 \text{ keV}$ to 0.41 at $E_p = 2 \text{ keV}$.

Figure 3b is a plot of the secondary electron emission coefficients for β tantalum and bcc tantalum for primary electron energies 0.2 to 2.0 keV. This plot reveals that the secondary electron emission coefficient for β tantalum is greater than for bcc tantalum over most of the energy range investigated. The shape of these curves and the absolute magnitude of δ_{max} varied slightly from sample to sample. The values of δ for both phases of tantalum were found to be very similar for primary electron energies in the range 1.5 to 2.0 keV. The cross-over point (the primary electron energy at which $\delta = 1$) of the secondary electron emission curves for β tantalum and bcc tantalum were found to be ~ 215 eV and ~ 240 eV respectively. The maximum yield of secondary electrons occurred at ~ 0.4 keV for both phases.

Figure 4a is a plot of the collector current as a function of the negative bias applied to the grid (Fig. 1). The primary electrons were incident normal to the surface with an energy of 1.5 keV. As the negative bias was increased the collector current decreased by an amount equal to the current of those secondary electrons retarded by the grid potential. Figure 4a indicates that nearly all secondary electrons have been suppressed with a grid bias of -40 volts.

Figure 4b is a plot of the number of secondary electrons per primary electron per eV as a function of the negative bias applied to the grid. These curves were obtained by differentiation of the curves in Figure 4a. Figure 4b indicates that the energy distribution of the secondary electrons is similar for both phases of tantalum. The maxima in the secondary electron energy distributions occurs at ~ 4 eV.

Figure 5 is a plot of the dependence of the secondary electron emission coefficient upon the angle of incidence of the primary electrons for β tantalum and bcc tantalum. The energy of the primary electrons was 1.5 keV. 0° corresponds to normal incidence of the primary electrons. These curves indicate that the angular dependence of the secondary electron emission coefficient for β tantalum and bcc tantalum are similar. The number of secondary electrons emitted increases $\sim 25\%$ for each phase as the angle of incidence increases from 0° to 35° . Measurements obtained in this study indicate that the shape of the curves in Figure 5 are independent of primary electron energy over the range $0.5 \leq E_p \leq 2.0$ keV.

Scanning Electron Micrographs

Figure 6 is a scanning electron micrograph of a sputtered tantalum thin film. The micrograph was obtained by operating the electron beam inspection system in the amplitude modulation display mode. The primary electron energy was 1.4 keV. The triangular

figure is a scribed reference mark. The predominant feature in the center of the triangle was identified by x-ray diffraction to be bcc tantalum.

DISCUSSION

The backscattered electron measurements revealed that η is nearly a linear function of the primary electron energy over the energy range 0.2 keV to 2.0 keV. The measurements also showed that the value of η is very similar for both β tantalum and bcc tantalum. Sternglass⁽³⁾ measurements of the backscattered electron coefficient for tantalum reveal that η is a function of primary electron energy rising from ~ 0.15 at 0.5 keV to ~ 0.30 at 2 keV. The discrepancy between present results and those of Sternglass may possibly arise from differences in experimental conditions and the type of tantalum material studied. Sternglass' measurements were performed using polished tantalum sheet stock heated to $\sim 400^\circ\text{C}$ in an attempt to maintain a clean surface. The present measurements made no provision for heating the sputtered tantalum thin films. Although heating the tantalum target to 400°C does not remove the surface layer of tantalum oxide, it does produce a change in its conductivity.⁽⁸⁾ This conductivity change may be associated with a material change which might also influence the backscattered electron emission properties.

Although the absolute values of η obtained in the present study do not agree with Sternglass' measurements, the linear dependence of η upon the primary electron energy has been established for a number of other materials. As found by Sternglass, for elements with atomic number greater than 30, η increases with primary electron energy up to ~ 4 keV.

The secondary electron emission yields of β tantalum and bcc tantalum as plotted in Figure 4b exhibit the general shape obtained for all materials. The yield increases with the primary electron energy up to a maximum. The yield then decreases with energy since the average depth of secondary electron creation becomes comparable to the diffusion length of the electrons to the surface.

Warnecke⁽⁹⁾ and Copeland⁽¹⁰⁾ have reported results of secondary electron emission properties of tantalum. Warnecke investigated two polycrystalline samples of tantalum under carefully controlled vacuum annealing conditions. He found that heating the target to $\sim 1900^\circ\text{C}$ for a few hours in a vacuum resulted in a 25% decrease in the secondary electron yield near the maximum, presumably due to a "cleansing" of the surface. This "clean state" condition may have resulted from at least the partial removal of the tantalum oxide since its melting point is 1800°C .⁽¹¹⁾

Warnecke's measurements indicated that in the "clean state" $\delta_{\text{max}} = 1.30$ in the primary electron energy range 0.6 - 0.7 keV. Copeland found $\delta_{\text{max}} = 1.27$ at a primary energy of 0.8 keV. The crossover point (e.g. $\delta = 1$) occurred at ~ 275 eV for Warnecke's "clean state" material.

The discrepancy between the present secondary electron yields and those of Warnecke and Copeland may arise from differences in surface condition and crystalline structure of tantalum targets. In the present study no attempt was made to remove the few monolayers of Ta_2O_5 from the surface of the sputtered films. Consequently the secondary electron emission properties were influenced by this surface oxide layer. Several workers⁽¹²⁾ have established that the surface condition plays a dominant role in the secondary electron emission properties of a solid. Indeed Warnecke's results of heating the tantalum material to 1900°C show this effect conclusively.

It is not possible to discuss the discrepancy between the secondary electron yields of past and present results due to crystalline orientation effects, since Warnecke and Copeland did not report crystalline orientation. However, the discrepancy between past and present results may be partially due to orientation differences.

It should be noted that although surface cleanliness is important in secondary electron yields, it was possible in this study to produce surface conditions of the tantalum thin films such that secondary electron yields were reproducible to within $\pm 2\%$. For practical applications, this implies that secondary electrons can be used as a tool to determine the quality of sputtered thin films as long as the surfaces are consistently exposed to a noncontaminate atmosphere or are cleaned prior to inspection. For sputtered thin films it would be best to inspect the films in the sputtering chamber immediately after sputtering.

The angular dependence of the secondary electron yield for β tantalum and bcc tantalum is similar to that obtained for other materials.⁽¹³⁾ Previous studies of tantalum have not included studies of this angular dependence. The reason for the increase in yield as the angle of incidence (0° corresponds to normal incidence) is increased results from a decrease in the effective average depth of secondary electron production. A number of authors⁽¹⁴⁾ have proposed theories which involve a cosine dependence of the angle of incidence. These theories are adequate for a phenomenological description but lack sufficient detail to explain the experimental results precisely.

From a practical standpoint, an angle of incidence greater than 0° (normal incidence) would increase the signal to noise ratio

in the detection system of the electron beam inspection system. This arises since the signal is proportional to the number of secondary electrons collected. However, the contrast between bcc tantalum and β tantalum is reduced as the incident angle is increased since the ratio of secondary electron yields, upon which contrast depends, decreases.

The measurements of the energy distribution of secondary electrons indicate that the distribution is similar for β tantalum and bcc tantalum. These results imply that the contrast between tantalum and bcc tantalum cannot be improved by energy selection of the secondary electrons.

CONCLUSIONS

The secondary electron and backscattered electron emission properties of β tantalum and bcc tantalum thin films have been measured over the primary electron energy range 0.2 keV - 2.0 keV. The backscattered electron emission properties are similar for both phases of tantalum. However, the secondary electron emission properties of the two phases are appreciably different. Based upon this difference a simple scanning electron beam inspection system has been constructed. The system is capable of detecting the presence of bcc tantalum spots as well as other defects in a sputtered β tantalum thin film.

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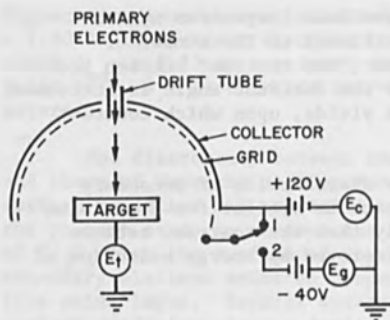


FIG. 1

Schematic diagram of apparatus used to measure secondary electron and backscattered electron emission properties.

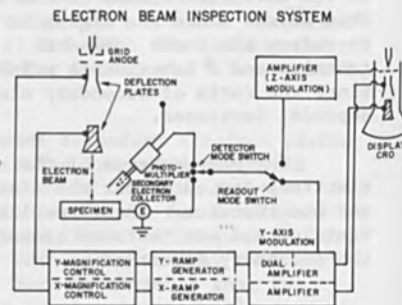


FIG. 2

Schematic diagram of the low energy scanning electron beam inspection system.

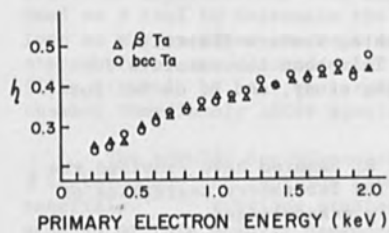


FIG. 3a

Backscattered electron emission coefficients η of sputtered β tantalum and bcc tantalum thin films as a function of the incident primary electron energy.

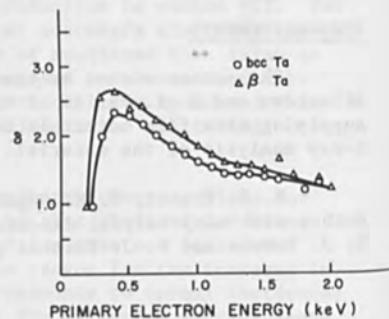


FIG. 3b.

Secondary electron emission coefficients η of sputtered β tantalum and bcc tantalum thin films as a function of the incident primary electron energy.

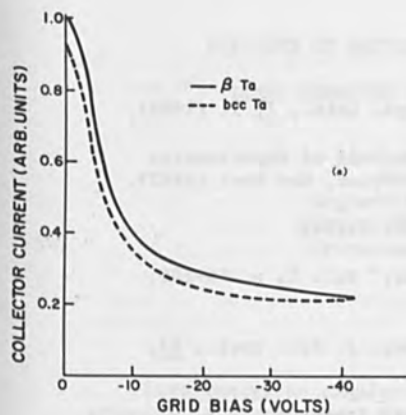


FIG. 4a

Collector currents as a function of retarding grid bias for 1.5 keV primary electrons incident normal to surface of sputtered β tantalum and bcc tantalum thin films.

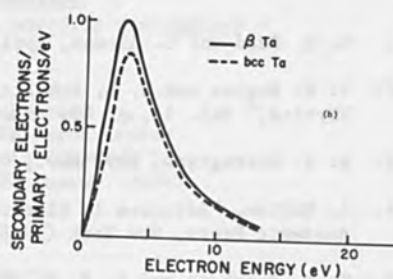


FIG. 4b

Energy distribution of secondary electrons emitted from sputtered β tantalum and bcc tantalum thin films. 1.5 keV primary electrons incident normal to the surface.

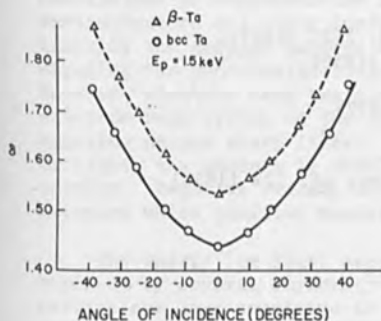


FIG. 5

Variation of secondary electron emission coefficient δ for sputtered β tantalum and bcc tantalum thin films as a function of the incident angle of 1.5 keV primary electrons. (0° corresponds to normal incidence)

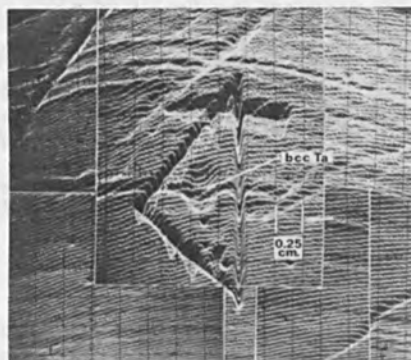


FIG. 6

Scanning electron micrograph of a sputtered β tantalum thin film.

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EXPOSURE OF PHOTORESISTS. II. ELECTRON AND
LIGHT EXPOSURE OF A POSITIVE PHOTORESIST

Barret Broyde
Engineering Research Center
Western Electric Company, Inc.
Princeton, New Jersey 08540

ABSTRACT

Less energy is required to solubilize AZ-1350 when the exposure is accomplished with kilovolt electrons than when light is used to expose the photoresist. In contrast to overexposure with light, overexposure by electrons causes an insoluble cross-linked product to form. The solubilization reaction is enhanced by the addition of small amounts of benzotriazole, indazoles and imidazoles, while the cross-linking reaction is inhibited by these additives.

INTRODUCTION

Photosensitive materials called photoresists, are used during the manufacture of semiconductor devices to protect selected areas of semiconductors and other surfaces against chemical attack. Conventionally the desired pattern in the photoresist is obtained by exposing the photoresist-coated surface to light through a mask. However, electron beam pattern generators, in which a controlled electron beam writes on the photoresist, offer the advantage of exposing narrow sharp lines. After exposure with either electrons or light, the pattern is developed by rinsing the photoresist with a solvent. Negative resists are those that are insolubilized upon exposure while positive resists are solubilized.

The energy (or flux) necessary to expose resists have been reported by several workers,^{1, 2, 3} and it appears that positive resists are less sensitive to both light and electrons than negative resists. In conventional processing this sometimes requires that exposure time be longer than desirable, but more important, the low sensitivity usually leads to very long exposure times when either projection photolithography or writing electron beams are used to pattern the resist.

This work reports that members of the benzotriazole, imidazole, and indazole families decrease the energy required for the light and

electron exposure of a high resolution positive photoresist, AZ-1350, a proprietary product of the Shipley Company, Newton, Massachusetts.

EXPERIMENTAL

Layers of AZ-1350 6000 Å thick were prepared by spinning techniques. The resist was then baked at 60°C for 10 minutes, allowed to cool, exposed and then dip developed for 4 minutes in the Shipley AZ-Developer (full strength). The thickness of the resist before and after development was measured by the techniques described previously⁴. Electron exposure conditions have also been described, while light exposures were carried out with a 150-watt xenon lamp held 1 m from the resist. Glass filters allowed only wavelengths longer than 3000 Å to irradiate the resist. Chromium-coated glass plates were used as the substrates for the electron exposure experiments, while quartz plates were used to study the light exposure.

When the effects of additives were examined, the additive was dissolved in the photoresist solution at least 16 hours before use. Occasionally, it was found that films of the resist with additive required shorter development times.

RESULTS AND DISCUSSION

Electron Exposure of AZ-1350

The electron flux required to expose 6000 Å thick layers of unsensitized AZ-1350 depends on the energy of the exposing electrons as well as the time that the resist has been stored. Typically, 6 μ coul/cm² of 15 keV electrons are required to expose newly purchased solutions while the same resist 4 months later requires about 40 μ coul/cm² for exposure. The loss of sensitivity of the resist on storage may arise from the same cause as the loss in sensitivity of the resist when it is heated.⁵

Varying the accelerating potential between 5 and 20 kV changes the flux required to expose newly purchased AZ-1350 from 3 μ coul/cm² to 9 μ coul/cm². Using the model previously described⁴ to calculate the amount of energy transferred from the beam to thin layers, shows that the amount of absorbed energy required to fully solubilize the film is $1.5 \pm 0.05 \times 10^{21}$ ev/cm³. The range of 5 keV electrons in AZ-1350 has been measured as 6500 Å $\pm$ 200 Å and this value was used for this calculation.

Harwood and Hunter have identified the reactive moiety in AZ-1350 as 3,4-dihydroxybenzophenone-4-(naphthoquinone-(1,2) diazide(2)) sulfonate and have found that it constitutes 23% of the weight of the

solids in the photoresist.⁶ Since the density of dried AZ-1350 films is 0.9, there are 1.6×10^{16} sensitizer molecules in a square centimeter of film 6000 Å thick. If all the sensitizer molecules have reacted when the film is fully solubilized, then the G value for solubilization is about 18. This rather high value implies that energy transfer from the beam to a reactive site in AZ-1350 is much more efficient than in negative resists.

When AZ-1350 is overexposed so that it absorbs about 20×10^{21} ev/cm³ it cannot be removed by AZ-Developer and the exposed spot is insoluble in acetone and benzene. Typical behavior of AZ-1350 after exposure to 15 keV electrons and development is given in Figure 1, curves A and B.

The mechanisms of the reactions involved in the response of AZ-1350 to electron radiation have not been determined yet, but it is likely that a cross-linking reaction of its phenol-formaldehyde polymer⁶ leads to the insoluble product (see below). It is also likely that the solubilization reaction is the same one that occurs with light; quinone diazide groups are converted to carboxylic acids.⁷ Solubilization arising from a scission reaction of the phenol-formaldehyde polymer in which indiscriminate bond breakage occurs and soluble low molecular-weight compounds result cannot be fully excluded, but it is not probable because scission would have to predominate at low exposures and cross-linking at high fluxes.

Exposure of AZ-1350 by Light

Htoo has shown that an incident flux of 3.6×10^{18} ev/cm² is needed to expose a 6000 Å film of the resist.¹ Since the absorptivity of the films has been found to be 0.32, by K. H. Cho of this laboratory, this flux corresponds to an absorbed energy of 1.7×10^{18} ev/cm² and a quantum yield of 0.032 for the solubilization process. Absorbed light energy therefore is only 5 per cent as efficient in causing solubilization as is electron energy. The higher efficiency of solubilization during electron exposure is unexpected, since the primary process of electron energy absorption leads to a large variety of excited bonds and molecules, while during light absorption only the quinone diazide groups are excited.⁸ Overexposure by wavelengths greater than 3100 Å does not lead to an insoluble product, but when the resist is exposed to 2537 Å radiation it forms an undevelopable spot. Since the phenol-formaldehyde polymer absorbs radiation at this wavelength but not at the longer wavelengths, it is probably undergoing a cross-linking reaction.

Sensitization of AZ-1350

When dilute solutions of benzotriazole in AZ-1350 are prepared and spun to films, it is found that a lower flux of both electrons and light is required to solubilize the resist. Typical results are

shown by curve C of Figure 1 and by Figure 2. Benzotriazole also inhibits the cross-linking that occurs with high electron fluxes (Curve D, Figure 1).

Benzotriazole is not the only efficacious additive. All members of the benzotriazole, imidazole, and indazole families that are soluble and that have a hydrogen atom bonded to a nitrogen atom also decrease the flux needed to expose AZ-1350. These additives also sensitize the response of other quinonediazide resists; e.g., AZ-1350-H (a Shipley product) and KAR-3 which is manufactured by Eastman Kodak.

The mechanisms in sensitizing the solubilization reaction and in inhibiting the cross-linking reaction are still being investigated. Since neither the absorption spectrum nor the penetration range of electrons of the resist change significantly when low levels of benzotriazole additives are added to the resist, sensitization of the solubilization probably involves better energy transfer from the site of absorption to the reaction site. Benzotriazole-type additives also cause energy to be transferred away from cross-linking sites, since 2% solutions in KTR cause an increase in the electron energy needed to reach the gel point from 2.5×10^{20} ev/cm³ to 11×10^{20} ev/cm³. Benzophenone and hexaphenyldiiodine which reduce the electron flux needed to reach the gel point of KTR also cause a reduction in the flux that causes overexposure in AZ-1350.

CONCLUSIONS

1. Electron exposure is more efficient in solubilizing AZ-1350 than is light exposure.
2. Overexposure by electrons causes a cross-linking reaction which renders AZ-1350 undevelopable.
3. Benzotriazole-type additives increase the efficiency of the solubilization reaction caused by both electron and light absorption while retarding the cross-linking reaction caused by electrons.

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ELECTRON EXPOSURE OF AZ-1350

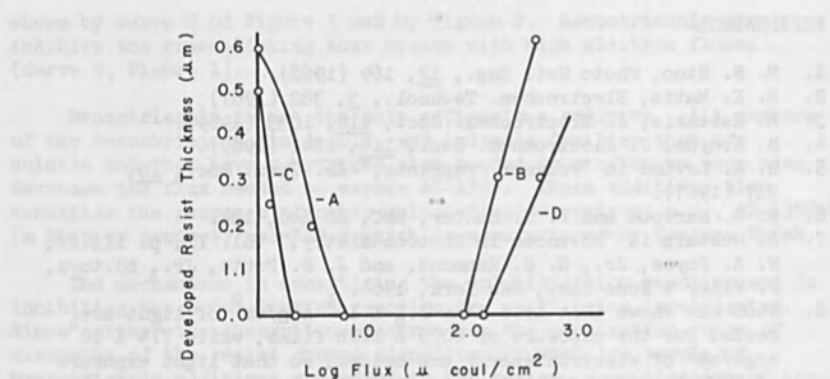


Fig. 1 Exposure of AZ-1350 by 15-keV electrons. Curves A and B newly purchased resist. Curves C and D newly purchased AZ-1350 containing 2% benzotriazole. The initial thickness of resist was 0.6 μm.

LIGHT EXPOSURE OF AZ-1350

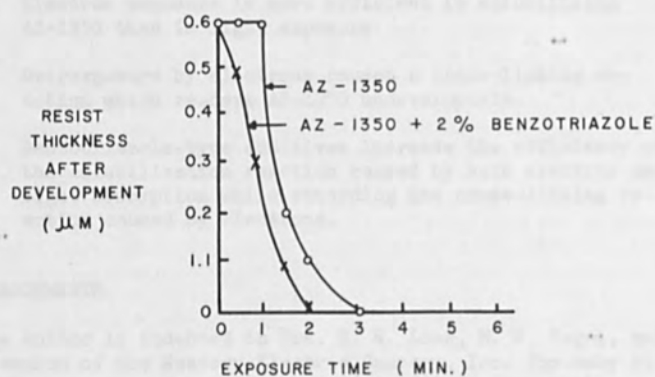


Fig. 2 Effect of benzotriazole on the exposure of AZ-1350 by light. The initial thickness of resist was 0.6 μm. A xenon lamp 100 cm from the sample was used.

HIGH PERFORMANCE STEP AND REPEAT MACHINE USING
AN ELECTRON BEAM AND LASER INTERFERO METERS

O. CAHEN and J. TROTEL

THOMSON-CSF - FRANCE

This talk deals with high accuracy mask fabrication for semiconductor industrial processing. Higher accuracy is needed for better close-tolerance yield and area saving. It allows shorter dimension, smaller capacitance, higher frequency performance and less power dissipation.

Electron beam exposure of photoresist gives a step forward on the high accuracy scale. Figure 1 shows a complete four-gate interconnexion pattern reduced to the size of four mil square. The mean width of lines is .3 micron.

The most difficult problem, as far as such a high accuracy is concerned, consists in maintaining a complete 2-inch array within the same tolerance of .1 micron.

The aim of the talk is to show how this close tolerance can be kept in the periodicity of a large-area array.

The 100-micron square shown can be exposed in one second time with a one nanoamp, ten KV beam, acting on standard Kodak photoresist. The sample is supposed to be moved from each pattern to the next, the needed accuracy lies in perfectly accurate control of the movements of the sample.

Figure 2 is a block diagram of the complete machine. It includes an electron optical system producing a narrow beam focused on the sample, this beam scans over a limited area of the resist coated sample.

The sample is attached to a table which can be moved under the beam to cover a 2 inches by 2 inches area on the sample.

The table is driven by two stepping motors and its movements are measured by two laser interferometers.

The stepping motors are controlled by a logic system which is feeded

by the data representing the desired position and the actual position of the table, as measured by the interferometers.

In this way, the motors are stopped when this sample is within $10\text{ }\mu\text{m}$ at the desired position and then the beam is deflected to compensate the resulting error.

Only two interferometers are used, accidental angular displacements are not measured, but their influence is minimized by making the two laser beams intersect the mean direction of the electron beam. The interferometer measurements are then a very close approximation of the actual coordinates of the pattern being exposed.

Figure 3 shows the CAMECA scanning electron microscope, from which we use the optical column, with some modifications to fit the larger sample chamber.

Figure 4 is a block diagram of the logic system. There are two marks on the sample, these marks must be visible by using the instrument as a scanning electron microscope. The operator has, first, to push the "Mark 1" button, to put this mark within $10\text{ }\mu\text{m}$ under the electron beam, then, by the manual control he has to deflect the beam so as to see the mark at the center of the screen. At this time the interferometer measurements and the beam position are memorized.

The same operation is done for the second Mark.

Then, by pushing the "start button", the complete process goes on automatically, the stepping motor and deflection yokes are fed with instructions computed from the scanning program and the mark positions.

The instructions to draw a given pattern could be taken from a digital memory, but as a first step, we use an analog memory which is a conventional flying spot system.

Figure 5 is a photography of one of the interferometer which has been built for testing.

It is a Michelson Type interferometer with helium neon laser.

There are two detectors for two crossed polarization directions of the light. One direction of polarization is delayed compared to the other one by a birefringent $\lambda/8$ plate. So, when the table is driven at a constant speed, the detectors give two sine waves with ninety degrees phase shift. The sign of the phase shift indicates the direction of the movement.

In order to get sixteen pulses per wavelength translation, the detector signals are electronically processed so as to get two complementary sine waves with a forty five degrees phase shift with the original ones.

The laser is a multimode helium neon laser, it can be used because the displacement to be measured is limited to ± 25 mm.

It gives three modes separated by 385 MHz. In practice, when the different mode appears in the Doppler width, the mean frequency ripple is only about 50 MHz, i. e., a few 10^{-7} of the frequency.

In conclusion, all the main problems have been solved for a step and repeat machine covering an area of 2×2 inches with an accuracy of .1 micron, to be used for integrated circuit pattern generation by scanning electron beams.

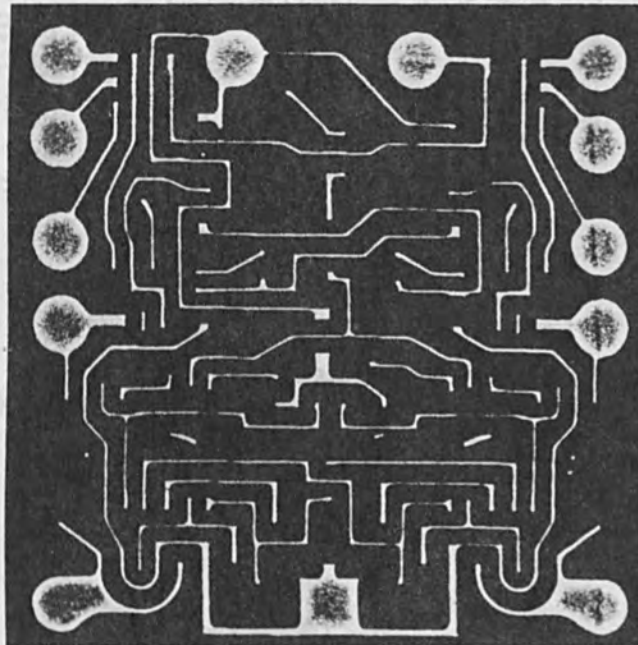


Figure 1

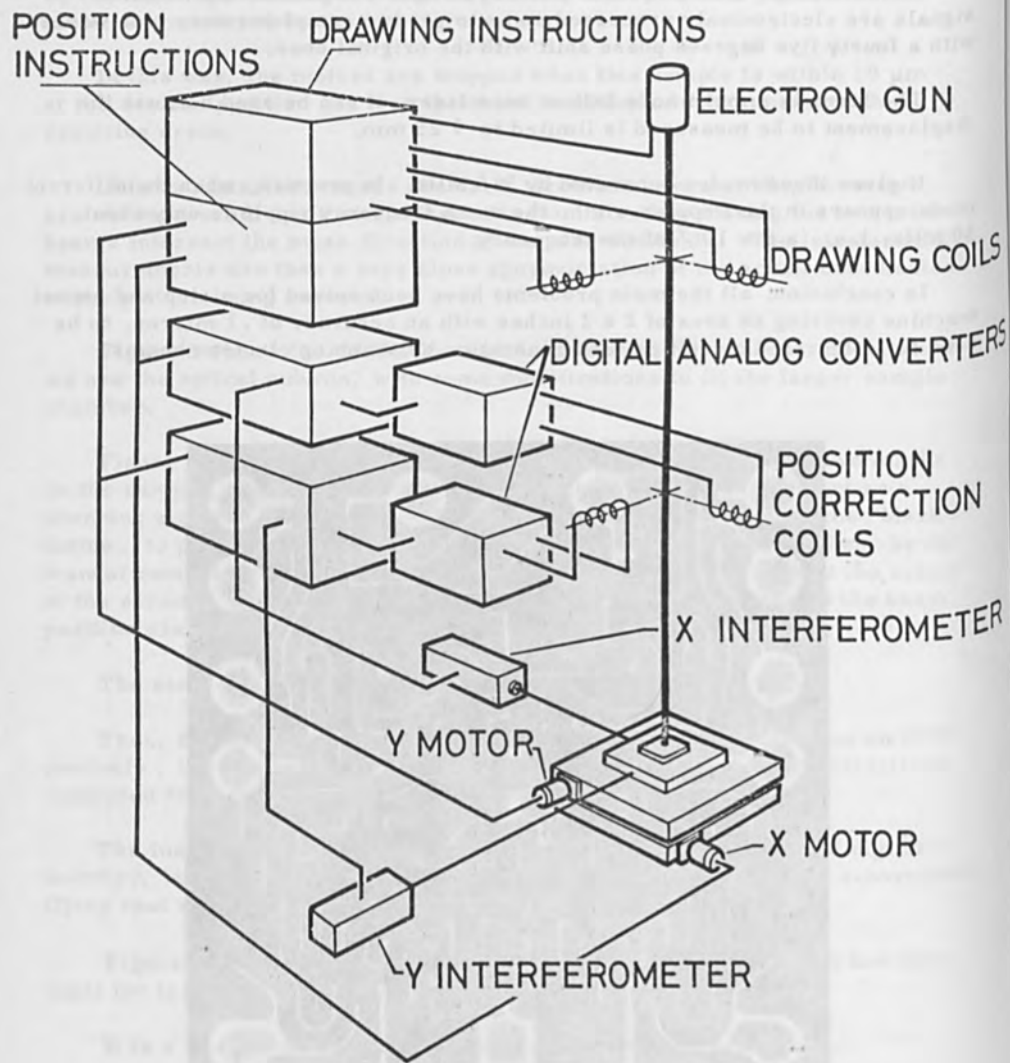


Figure 2

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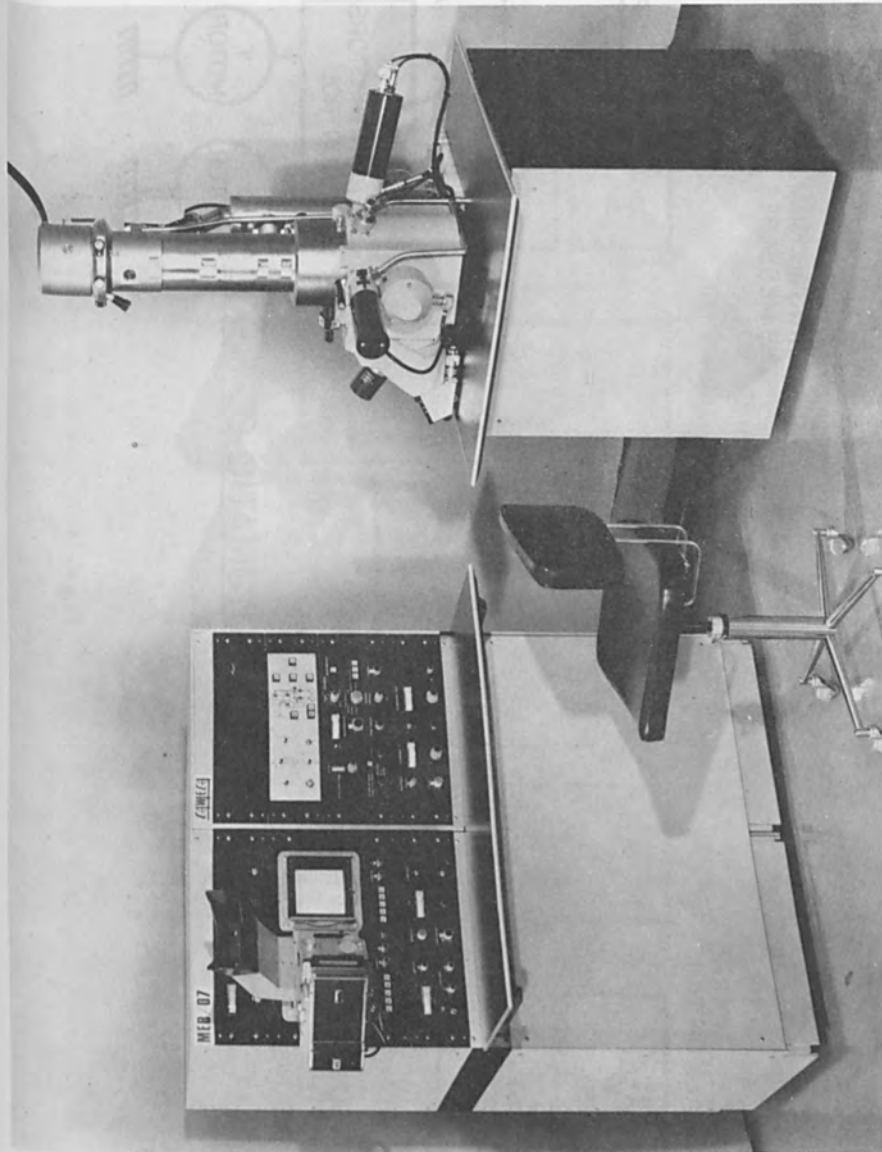


Figure 3

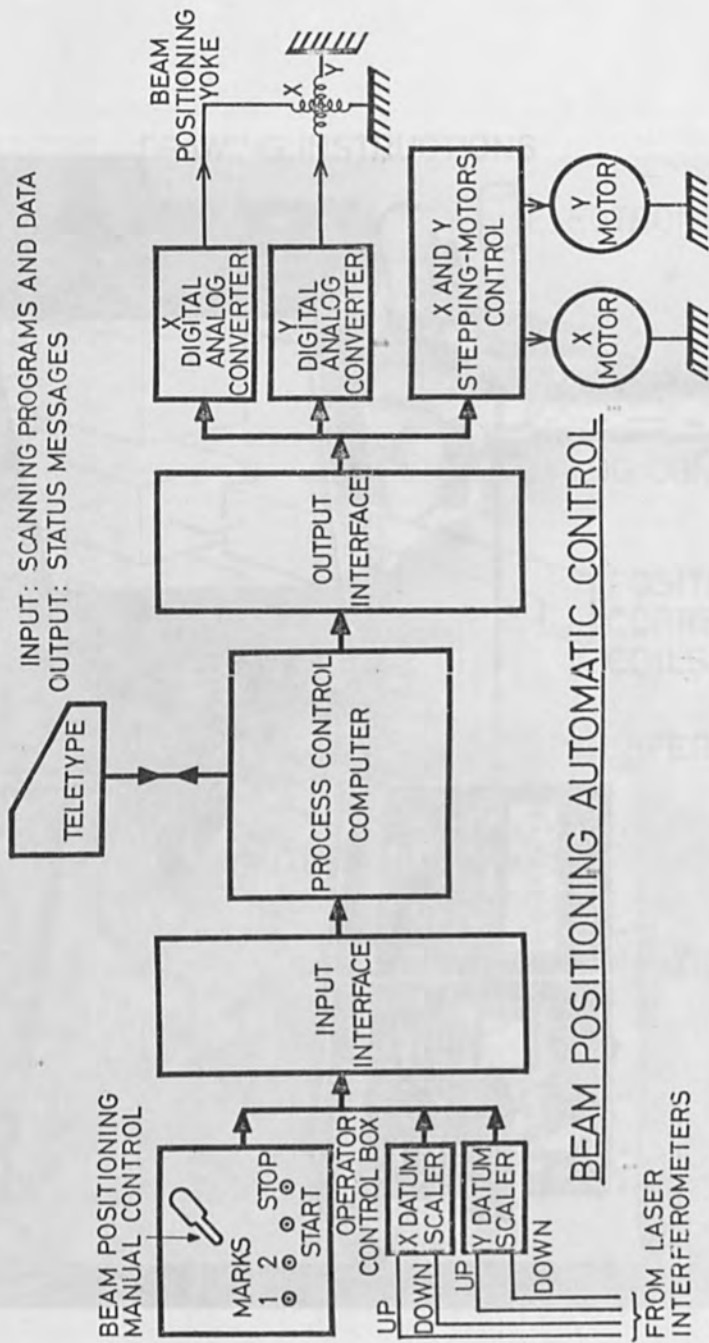


Figure 4

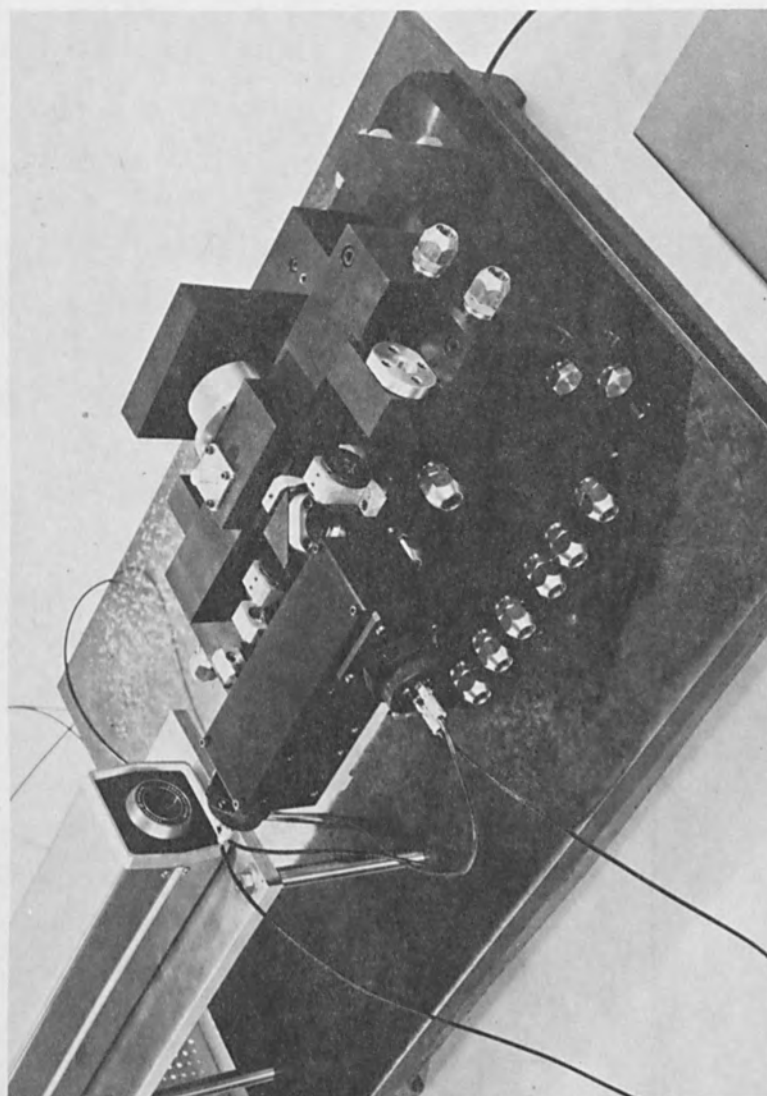
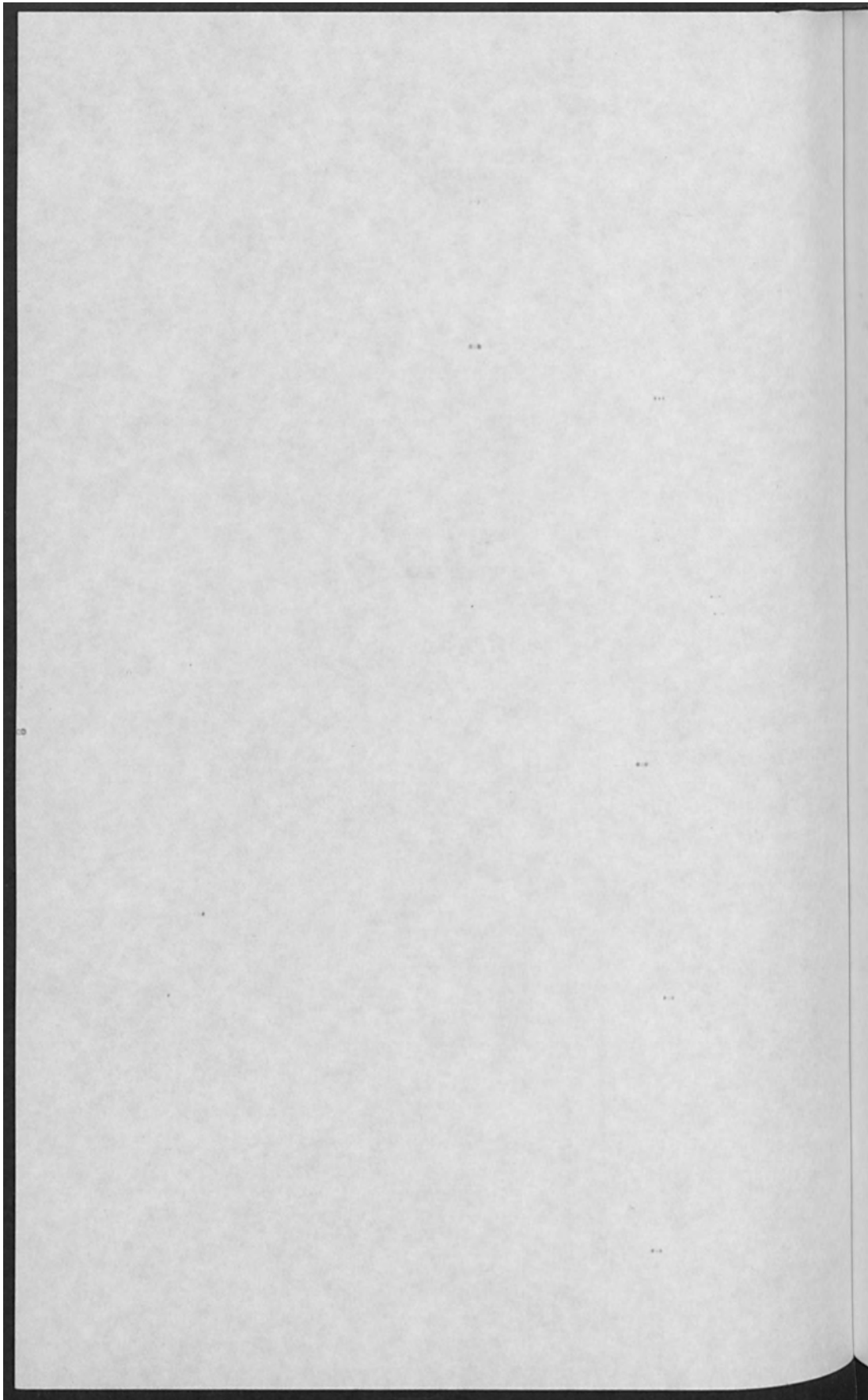


Figure 5



SECTION IV
ELECTRON BEAMS
IN
IRRADIATION PROCESSING



Energy Deposition of Electron Beams in Insulating Materials*

Marvin Rosenstein,¹ William L. McLaughlin,² and Joseph Silverman³

ABSTRACT

Experimental depth-dose curves were obtained for semi-infinite thicknesses of ordinary and conducting polystyrene, irradiated in air by a 1.5-MeV electron beam in the dose range 5 to 20 Mrad. Radiochromic dye films were used as spatial dosimeters. By using a calibration of optical density versus dose, a microdensitometric reading of the irradiated dye films gave continuous depth-dose curves. No difference was observed between the depth-dose curves for the ordinary polystyrene and conducting polystyrene at absorbed dose rates between 2.5 and 10 Mrad/min. Within the limits of uncertainty for the present experiment, the results are in agreement with theoretical and experimental depth-dose data for 1.5-MeV electrons in polystyrene. Under the conditions studied, trapped charge does not measurably perturb the energy deposition of electrons in polystyrene.

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¹Mr. Rosenstein is an engineer with the Bureau of Radiological Health, U.S. Public Health Service, Rockville, Maryland 20852. Currently, he is a graduate student in Nuclear Engineering at the University of Maryland and a guest worker at the National Bureau of Standards.

²Mr. McLaughlin is a physicist in the Dosimetry Section of the Applied Radiation Division, Center for Radiation Research, National Bureau of Standards, Washington, D.C. 20234.

³Dr. Silverman is Professor, Laboratory for Radiation and Polymer Science, Department of Chemical Engineering, University of Maryland, College Park, Maryland 20740.

INTRODUCTION

The purpose of this experiment was to investigate the effects of charge trapping on the depth-dose curve of insulating and conducting materials of about the same composition. The experiment was carried out as a preliminary investigation before obtaining experimental electron energy deposition curves in finite thicknesses of low-Z dielectric materials; e.g., polystyrene. Kniedler and Silverman (1) have used a Monte Carlo technique to compute depth-dose curves in finite slabs of water and tungsten for 0.1, 1 and 10.9 MeV electrons incident normal to the surface of the material. Their calculations show that the energy deposited in finite layers of material (thicknesses less than the electron range) is lower than would be given by energy deposition curves for infinite slabs. Prior to undertaking depth-dose measurements in finite slabs, it was desirable to investigate the possibility that trapped charge in semi-infinite slabs of the test material, polystyrene, would alter the shape of the depth-dose curve, since such an effect would confuse the interpretation of changes observed for finite thicknesses.

Several investigators have studied the effect of charge deposition in certain materials. Lackner, *et al.* (2) concluded that, for 1.5-MeV electrons in polymethyl methacrylate (PMMA), there is a marked decrease in the electron range with increasing charge fluence up to dielectric breakdown. This was determined by means of photographic observation of the depth of the fluorescing region in the sample. They mentioned similar, but less severe, effects for polystyrene at high electron currents (200 mA). Kennedy and Kohlberg (3) made calculations of depth dose using a Monte Carlo technique that included effects of internal electric field buildup from charge trapping. Calculations for

1.5-MeV electrons in PMMA showed the effect of charge trapping on the energy deposition curves with increasing energy fluence (0.25 to 1.8 J/cm²), namely, an increase in the height of the peak of the energy deposition curve and a shift of the peak toward the entrance surface of the material. The maximum electron penetration depth also decreased markedly from 0.54 cm to as low as 0.15 cm. Little, Ottesen and Childers (4) irradiated PMMA with single 0.1 μ s pulses of 2.0-MeV electrons at an energy fluence of 40 J/cm² and measured relative dose with imbedded thermocouples. They stated that the observed energy deposition profile was not greatly perturbed by trapped charge phenomena; however, their results fall somewhere between the curves generated by two theoretical models, one including trapped charge and one not including trapped charge.

In order to investigate the possibility of charge-trapping effects, a series of irradiations with 1.5-MeV electrons were made in ordinary polystyrene and conducting polystyrene samples, over the dose range 5 to 20 Mrad (energy fluence $\sim$ 50 to 200 J/cm²) at the entrance surface of the sample and at the dose rates of 2.5 to 10 Mrad/min. These doses were selected to be in the useful range of response of a dye film dosimeter which will be used in future experiments in finite layers of polystyrene.

EXPERIMENTAL TECHNIQUES

Dosimeter

A radiochromic triphenylmethane dye-cyanide film dosimeter and a recording microdensitometer were used in order to obtain high spatial resolution for the depth-dose measurements. McLaughlin (5, 6, 7) described in detail experimental investigations of this type of dosimeter. In summary, the characteristics of

dye-cyanide dosimeters include: long shelf life, dose-rate independence up to 10^{15} rad/sec, low atomic number constituents (C, H, N, O), energy independence of response, insensitivity to the presence or absence of oxygen, small temperature dependence, image stability, linear dose response to ionizing radiation, high-resolution images, and simple readout.

The specific dye-cyanide used in these experiments is a thin film of colorless pararosaniline nitrile in slightly acidic photographic gelatin, machine-coated on a triacetate base. Upon irradiation in the megarad range, the film takes on a magenta color due to permanent radiolytic formation of pararosaniline dye. The thickness of the sensitive dye layer is about 1.5 mg/cm^2 (~ 0.2 percent of the electron range), and the total thickness of the film about 20 mg/cm^2 . Based on reproducibility studies of dye color intensity at known doses, the variation of the concentration of dye in the sensitive layer varies less than ± 2 percent. The dye film response in terms of optical density is read at its absorption maximum at a wavelength of 560 nm. Response of the microdensitometer was limited to light at this wavelength by means of an interference filter. The precision (one standard deviation) of the relative dose values from photometric measurements is ± 5 percent (6).

Figure 1 is a calibration curve of absorbed dose (in water) versus optical density for the film in the absorbed dose range 1 to 40 Mrad. The plotted points were obtained with ^{60}Co γ -radiation, except for the two points below 4 Mrad. The latter were obtained by irradiating the films with 1.5-MeV electrons and determining the absorbed dose by thin carbon disk calorimetry (6). Table 1 lists the values shown in Figure 1, in terms of net optical density as a function of dose and the net optical density

divided by dose as a function of dose. Since the latter is essentially a constant for all irradiations, the linearity of the curve in Figure 1 is substantiated. Table 2 illustrates the temperature dependence of the film response over the range -78 to 100°C. All of the irradiations in the present experiment were carried out at an initial temperature of 25°C; however, the sample and film dosimeter did increase significantly in temperature with repeated irradiations in the Mrad range. No variation in dosimeter response is expected since the temperature during irradiation did not exceed 100°C.

Dosimeter Response Readout System

The dosimeter readout system consisted of a commercial light source and transmission densitometer adapted to serve as a recording microdensitometer. An aluminum sample holder was constructed to feed through a Bakelite guide placed over the light source aperture. The aperture used in this experiment was 0.133 mm wide and 1 mm long. The film is driven across the narrow dimension of the slit by a d.c. motor connected by gears to a movable arm capable of forward and reverse travel. Variable feeder speed is provided by control of input voltage to the motor. A fast-response chart recorder was added and calibrated in units of optical density vs. translation of the sample. In this way, film response could be determined either visually on a meter as optical density at a specific point or recorded continuously as optical density versus spatial location on the dye film. A counter mounted on the feeder drive recorded the actual distance traveled by the feeder during a tracing run.

Experimental Irradiation Arrangement

Figure 2 is a schematic drawing of the experimental arrangement for irradiation of the polystyrene samples. A 1.57-MeV beam of monoenergetic

electrons from the National Bureau of Standards 4-MeV Van de Graaff accelerator passed through a 13.5 mg/cm^2 aluminum window and then through an aluminum collimator located 15.6 cm from the beam window. The sample was 10.6 cm from the collimator. The total intervening air gap was 29.1 mg/cm^2 . Defocusing and scattering in the window and air gave rise to a broad beam (i.e., beam diameter larger than the target diameter) with a slightly degraded electron spectrum of 1.50-MeV average energy and a dose uniform over the evaluated area (0.5 mm above and below the central axis) within ± 2 percent. Figure 3 shows a typical relative dose profile across the total width of the irradiated film. The dose rate imparted to a layer of carbon of known thickness was calibrated by a thin graphite calorimeter, and the associated current passing through the collimator was monitored on an aluminum shutter.

As shown in Figure 2, the polystyrene samples consisted of 2.22-cm diameter cylinders 2 cm long and cut into two pieces diagonally at 10° to the center line of the cylinder. A dye film dosimeter was placed along the diagonal cut to provide a continuous recorded image of spatial response. The sample and film were placed in a polystyrene sleeve and, except for the front face, was wrapped in aluminum foil which could be either insulated from or connected to ground. These sample assemblies were inserted into a cylindrical hole in a PMMA holder secured perpendicularly to the electron beam.

EXPERIMENTAL RESULTS

Two materials, ordinary polystyrene and conducting polystyrene, were used. Ordinary, unirradiated polystyrene is an excellent insulator with a conductivity of 10^{-17} to $10^{-19} \text{ ohm}^{-1} \text{ cm}^{-1}$ and a density of 1.06 g/cm^3 . The composition of the conducting polystyrene was 50% polystyrene, 27% polyethylene, and 23% carbon. It has a conductivity of the order of $10^{-3} \text{ ohm}^{-1} \text{ cm}^{-1}$ and a

density of 1.11 g/cm^3 . The purpose of the conducting polystyrene was to provide a material essentially equivalent to polystyrene except for its ability to conduct away charge that might build up in the irradiated sample. Twenty samples were irradiated. Duplicate irradiations were made for both polystyrene and conducting polystyrene samples at each of three total doses (5, 10, and 20 Mrad at the entrance surface at a constant dose rate of 5 Mrad/min). Single irradiations were made for both materials at each of three additional dose rates (2.5, 7.5, and 10 Mrad/min at the entrance surface at a total dose of 10 Mrad). These samples were grounded. Two samples, one of each material, were irradiated ungrounded at 10 Mrad (5 Mrad/min).

Figure 4 shows a typical film response plot of optical density versus distance traveled by the film feeder, with appropriate construction lines for determining film background, maximum response, extrapolated range and front edge of film sample. The front part of the response curve ($\sim 1 \text{ mm}$) is perturbed. This is due to some scattering of the electron beam at the entrance edge of the cut sample cylinders and to limitations in the resolution of the instrument across the film edge. Because of this perturbation, the exact shape of the curve near the entrance edge was not obtained. More exacting methods for preparation of sample-dosimeter assemblies and improved film-aperture geometry and optics have been designed for reducing this effect in future experiments. However, the inflection point of the steeply-rising front portion of the response curve represents the front edge of the film. This was established by subsequent runs in which the distance between the front and back edges was known.

All the depth-dose curves had similar shapes. In order to study the curves in more detail, an analysis of variance was performed on the data classified

by material type and total dose (duplicate runs) for each of two parameters that characterize the curves (depth at maximum dose and extrapolated range). Likewise, an analysis of variance was performed on the data classified by material type and dose rate (single runs). Table 3 lists the experimental results of these irradiations in terms of the depth at maximum dose and the extrapolated range. No statistically significant differences were observed for depth at maximum dose or the extrapolated range as a function of type of material, total dose, or dose rate. As a result of the analysis of variance, the experimental results could be treated as a single population of data. The mean values ($\pm$ two standard deviations) for each of the two parameters from twenty runs were as follows:

depth at maximum dose	0.31 ± 0.03	g/cm^2
extrapolated range	0.73 ± 0.05	g/cm^2

A large part of the fluctuation about the mean is attributable to the uncertainty in interpreting the location of the front edge on the microdensitometric trace.

Figure 5 is a comparison of three sets of information for semi-infinite targets of polystyrene. One is a depth-dose curve, typical of our data for both ordinary polystyrene and conducting polystyrene (1.5-MeV electrons). Another consists of the theoretical Monte Carlo histogram of Berger and Seltzer (private communication), scaled from a histogram for 2-MeV electrons in polystyrene. The third consists of absolute values interpolated from ionization measurements by Nakai, *et al.* (8) from curves for 1.4 and 1.6-MeV electrons in polystyrene. The dashed line represents an extrapolation of our curve to the front edge. Our experimental curve was normalized to the

theoretical curve by equalization of areas, which assumes that the fraction of the total available energy deposited in the sample was that given by the theoretical curve (0.99). Within the limits of uncertainty for the present experiment, the three sets of data are in agreement.

CONCLUSION

For the experimental conditions of these irradiations, specifically, 1.5-MeV electrons in air, entrance doses in the range of 5 to 20 Mrad and dose rates of 2.5 to 10 Mrad/min, no difference was observed for energy deposition in semi-infinite thicknesses of ordinary polystyrene and conducting polystyrene. This result indicates that charge trapping does not measurably perturb the depth-dose profiles in ordinary polystyrene and that at the dose rates and dose levels used in our experiment, conditions appear to allow for rapid leakage of electrons.

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Table 1. Response of pararosaniline dye cyanide dosimeter to 1.5-MeV electrons and ^{60}Co γ rays

Radiation Source	Absorbed Dose, D (Mrad)	Net Optical Density, ΔA (at 560 nm)	$\frac{\Delta A}{D}$
1.5 MeV e^-	1.57	0.030	0.019
1.5 MeV e^-	2.43	0.050	0.020
^{60}Co γ rays	3.96	0.080	0.020
^{60}Co γ rays	9.58	0.185	0.019
^{60}Co γ rays	27.6	0.510	0.019
^{60}Co γ rays	36.7	0.680	0.019

Table 2. Temperature dependence of the radiation response of pararosaniline dye film

Temperature $^{\circ}\text{C}$	Optical density relative to value at 22°C
-78	0.52
1	0.95
22	1.00
29	1.00
51	1.05
74	1.00
99	1.00

Table 3. Experimental values for depth at dose maximum and extrapolated range for 1.5 MeV electrons in polystyrene.

Type of sample	Total dose at front surface (Mrad)	Dose rate at front surface (Mrad)	Depth at maximum dose (g/cm ²)	Extrapolated range (g/cm ²)
<u>Grounded samples</u>				
Non-conducting polystyrene	5	5	0.295	0.737
	5	5	0.312	0.761
	10	2.5	0.299	0.728
	10	5	0.325	0.722
	10	5	0.285	0.674
	10	7.5	0.299	0.694
	10	10	0.302	0.709
	20	5	0.321	0.726
	20	5	0.313	0.752
Conducting polystyrene	5	5	0.362	0.759
	5	5	0.312	0.756
	10	2.5	0.296	0.719
	10	5	0.306	0.751
	10	5	0.312	0.750
	10	7.5	0.286	0.728
	10	10	0.305	0.751
	20	5	0.306	0.709
	20	5	0.304	0.748
<u>Ungrounded samples</u>				
Non-conducting polystyrene	10	5	0.302	0.725
Conducting polystyrene	10	5	0.300	0.758
Average			0.31	0.73
2 Standard deviations			0.03	0.05

Pararosaniline Dye Film
560 nm wavelength

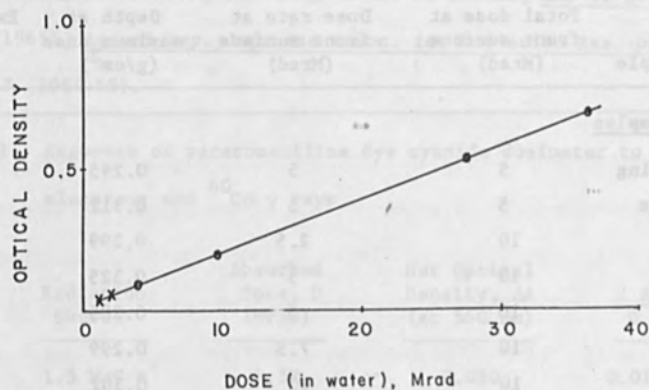


Fig. 1. Calibration curve for pararosaniline dye cyanide film; optical density vs. absorbed dose (in water). O, ^{60}Co γ rays; X, 1.5 MeV electrons.

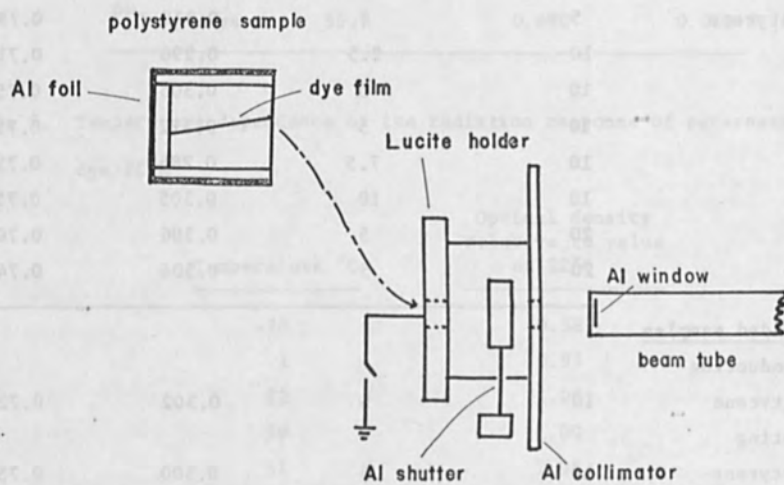


Fig. 2. Experimental arrangement for irradiations of polystyrene samples by 1.5 MeV electron beam.

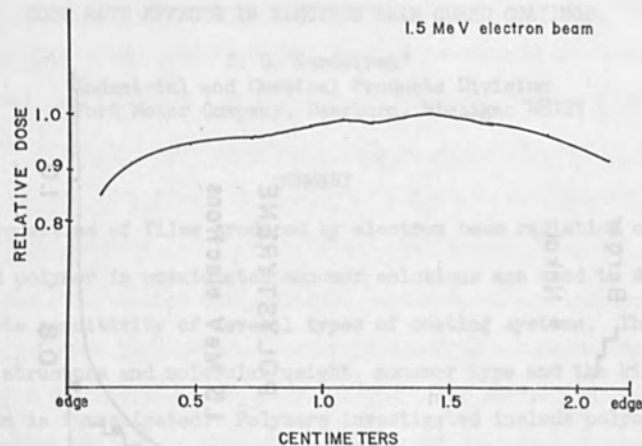


Fig. 3. Typical dose profile of 1.5 MeV electron beam across width of irradiated sample.

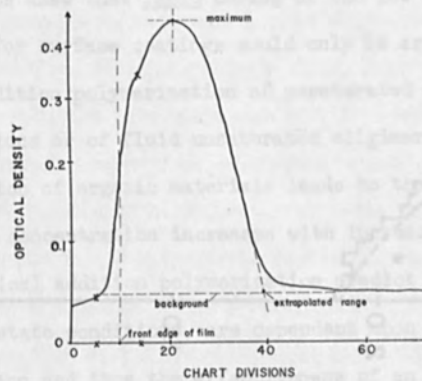


Fig. 4. Typical microdensitometric trace of irradiated dye film showing construction lines for determination of curve parameters; 1 chart division = 0.025 g/cm^2 .

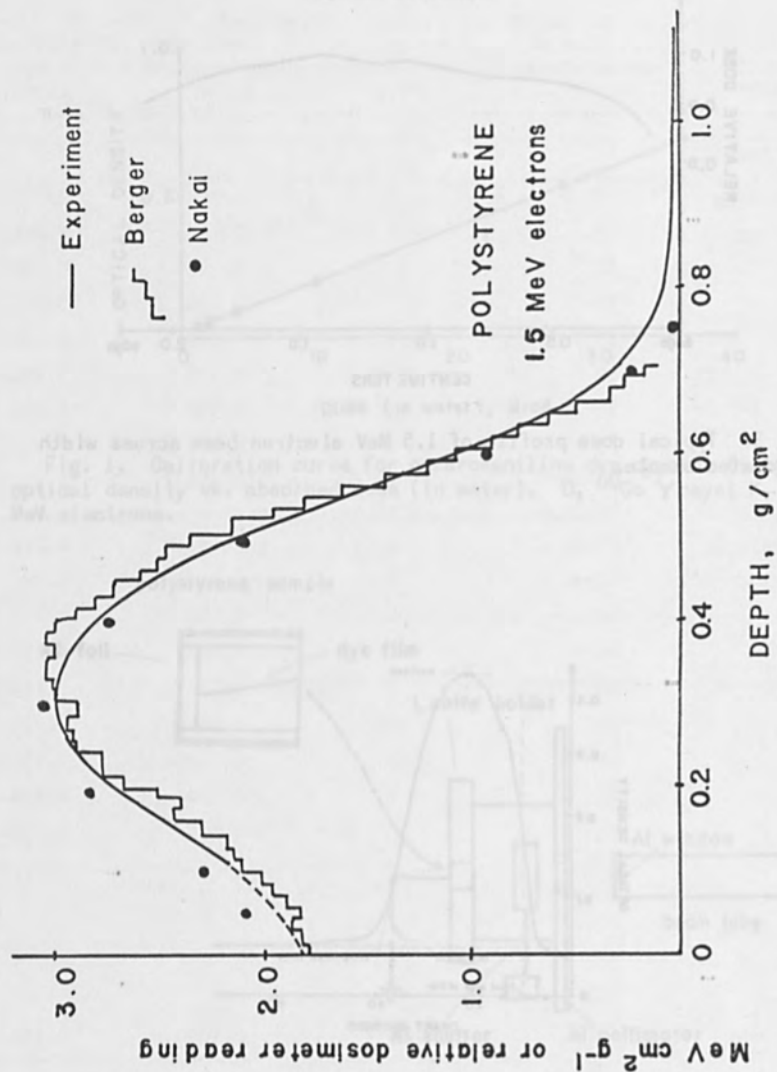


Fig. 5. Comparison of experimental depth-dose results for 1.5 MeV electrons in polystyrene with theoretical calculations of Berger and ionization measurements of Nakai.

DOSE RATE EFFECTS IN ELECTRON BEAM CURED COATINGS

J. D. Nordstrom*

Industrial and Chemical Products Division
Ford Motor Company, Dearborn, Michigan 48121

SUMMARY

The properties of films produced by electron beam radiation of unsaturated polymer in unsaturated monomer solutions are used to determine the dose rate sensitivity of several types of coating systems. The role of polymer structure and molecular weight, monomer type and the kind of unsaturation is investigated. Polymers investigated include polyesters with internal and pendant acrylate and methacrylate unsaturation and acrylic copolymers with pendant acrylate and methacrylate unsaturation.

INTRODUCTION

The potential problems posed by dose rate phenomena were recognized early in the process of designing equipment and materials for electron beam curing of coatings. Chemists knew that rapid curing of the low molecular weight materials necessary for surface coatings could only be accomplished by free radical initiated addition polymerization of unsaturated polymer in unsaturated monomer solutions or of fluid unsaturated oligimeric compositions. Electron beam radiation of organic materials leads to the production of free radicals and radical concentration increases with increasing dose rate. Kinetics of free radical addition polymerization predict that polymerization rates, under steady state conditions, are dependent upon the square root of initiator concentration and thus the effectiveness of an initiating species

*Present Address: Paint Operations
Ford Motor Company
400 Groesbeck Hwy.
Mt. Clemens, Michigan 48043

becomes smaller and smaller as its concentration increases. The primary cause of this limiting phenomenon is that the initiating species has the choice of reacting with a chain propagating entity which leads to the desired polymerization or it can react with another propagator and terminate both species. As the concentration of reactive radicals increase, the greater is the chance of the destructive process in proportion to the constructive process.

Within the boundaries of a real situation one can cite conditions which could cause deviation of the curing process from this theory of radical initiated addition polymerization. Indeed, a steady state may not exist within the system. Burlant and Hinsch have proposed a mechanism wherein reactions occur within cylindrical volume elements swept out by impinging electrons.¹ Davison, from Great Britain, has described several radiation conditions which are not steady state.²

Processes other than addition copolymerization are known to occur under ionizing radiation such as polymer cross-linking and degradation, and these have been shown to be dose rate independent, at least at low dose rate conditions. It is also quite possible that conversion of monomer to polymer, in systems of interest to coatings chemists, does not entirely reflect the degree of cross-linking attained in the system. Hoffman and Smith of High Voltage Engineering³ have shown evidence where gel formation continues to increase with dose after all of the monomer is incorporated into polymer. The local mobility in a gel may determine whether a free radical addition step results in a cross-link or just an extension of a free polymer end.

That which is really of interest to the coatings engineer who is evaluating the electron beam process as a method for curing coatings is - what effect does

dose rate have on the final properties of a film which is on the substrate he wants painted?

The literature presents conflicting evidence regarding dose rate effects on materials of interest in coatings. Several reports indicate that the well known maleate/fumarate type unsaturated polyesters in styrene do exhibit dose rate retardation of gel formation^{1,4,5} and film properties, such as hardness.⁶ An opposite effect is described by Miranda and Huebner⁷ In their work, gel content of terminally unsaturated polymer increased with increasing dose rate until a plateau was reached at about 10 Mrad/sec.

This paper will present information regarding dose rate sensitivity for unsaturated polymer in monomer systems with maleate/fumarate unsaturation, with methacrylate unsaturation and with acrylate unsaturation.

The criteria of sensitivity to dose rate will be two practical tests used by paint technicians to evaluate cured films, solvent resistance and hardness. The tests are a measure of the gross effects of the radiation rather than kinetic data or early stage polymerization data.

EXPERIMENTAL

Film Preparation

Unpigmented films were prepared on phosphated steel panels by drawdowns with a wire wound bar. The conditions were established so that the cured films were 1.0 ± 0.1 mil thick. A controlled time period was established between drawdown and cure so that there would be no differences in composition due to monomer evaporation within a series. Weight loss vs. time studies were made in order to establish the film composition at the time of cure.

The films were cured by attaching the metal panels to a conveyor which brought the panels past the accelerator at a distance of 4 inches from the

window. The accelerator was operated at 275 kilovolts. The current density and beam scan are stated in the following slides. The speed of the conveyor was adjusted in order to maintain a desired dose at different current densities or, in other words, at different dose rate. The atmosphere within the chamber was the effluent of a natural gas burner with an oxygen content less than 0.2%.

Film hardness was rated by the pencil method. The film is considered to have a hardness equal to that of the hardest pencil which does not penetrate the film.

The solvent resistance was rated in terms of MEK (Methyl ethyl ketone) rubs. A soft cloth was soaked in the solvent and rubbed back and forth across the film surface. A rub is one back and forth motion. Failure is at the time the metal panel is exposed. In most cases this was a sharp failure. The MEK rub test was found to be consistent by the same operator and when failure occurred below 60 rubs.

RESULTS

Our first efforts were made to confirm the prior art contentions that the maleate/fumarate type of unsaturated polyesters were dose rate sensitive. Figure 1 indicates that there is a marked decrease in solvent resistance as the dose rate increases with constant dose. The hardness readings paralleled the solvent resistance with ratings from $\leq 2B$ (10 MEK rubs) through F(40-70 MEK rubs) to H(≥ 80 MEK rubs). This polyester was prepared from maleic anhydride, tetrahydrophthalic anhydride and diethylene glycol and contained 2.5 moles of double bonds per thousand grams of polymer.

The composition at the time of cure was 80% polyester/20% styrene. Our work, with equipment designed to be industrially attractive, is rarely used with conditions that give less than 10 Mrad per second. Our experience with this

type of polyester always indicated a very high dose requirement for satisfactory cure. These data indicate why this was observed and points out the limitations of fumarate/maleate type polyester resins.

It is interesting, then, to look at the effects of dose rate changes upon polyester resins in which the unsaturation is present as pendant terminal groups rather than internal. Figure 2 shows, in a general way, how this kind of polyester is prepared. Polyols and dibasic acids are reacted by conventional procedures to yield a carboxyl terminated polyester. It is possible to react the terminal carboxyl groups with unsaturated alcohols or, more conveniently, with unsaturated epoxy monomers. We have prepared polyesters with pendant acrylate and methacrylate groups by this procedure.

Figure 3 shows the effects of dose rate upon film properties of a silicone modified polyester of the type just described. Two resins, similar except for the type of terminal unsaturation, were examined at two levels of total dose.

There is a very marked dose rate sensitivity of the methacrylate terminated polyester. The methacrylate resin did not cure to a dry film at the 3 Mrad range.

The sensitivity of the acrylate terminated material is small at the 6 Mrad dose level and slightly more evident at the 3 Mrad level. There has been previously published evidence that acrylate unsaturation is more reactive than methacrylate type.⁷ The present evidence supports this, at least indicating that ultimate properties are reached at lower radiation dosage.

Figure 3 indicates that further investigation is desirable to establish if the differences between acrylate and methacrylate unsaturation toward dose rate behavior is a general case. Tables 1 and 2 show film properties of acrylic modified silicone resins with high concentrations of double bonds. These

materials are in the 1000 molecular weight range and are completely fluid at room temperature. The unsaturation is terminal (that is, at the end of the polymer chains). Table 1 shows a high degree of sensitivity to dose rate for the methacrylate terminated resin. Only at very high total dose do the film property differences become unmeasurable. The last column indicates why a solvent extraction test, which is much less subjective than the rub test was not used to measure the effects of dose rate. Films which differ in solvent resistance by a factor of two have virtually the same degree of unextractability and there is almost no distinguishability between films after a minimum cure was reached.

In Table 2, the results of a resin which is identical except the methacrylate groups have been replaced with acrylate groups are shown. There is no difference in film properties measured over an eight fold increase in current density. The properties of the 12 Mrad cured film indicates that essentially full cure was achieved at 3.5 Mrad - again indicating the greater reactivity of the acrylate type unsaturation.

The role of the monomeric species in the curing process and their effect on dose rate is of interest. Figure 4 shows the properties of a methacrylate terminated polyester resin dissolved in a monomer of each type. The methacrylate in methacrylate system exhibits the decreased properties at increased current density at two levels of cure, as expected from previous data. The methacrylate polymer in acrylate monomer system appears to be less sensitive. At 10 Mrad it has attained its maximum cure and a zero slope line results. The data of this experiment, along with that from results to be presented later indicate that the polymer portion is much more determinant of dose rate prop-

erties than is the monomer, except as the monomer increases or decreases the total dose requirements of cure.

A very versatile class of unsaturated polymers for electron beam curing is that of unsaturated acrylic resins⁸. The unsaturated acrylics are prepared as shown in Figure 5. A polymer is built up by addition copolymerization of acrylate or methacrylate esters, one of which contains the epoxy functionality. Unsaturation is introduced by reacting the epoxy functionality with acrylic or methacrylic acid. The unsaturation of these polymers, which are much higher molecular weight than those previously discussed (10-15,000 vs. 1-2,000), are pendant and distributed along the chain.

Figure 6 shows the solvent resistance vs. total dose curve for a 0.9 double bond per thousand acrylic with methacrylate type of unsaturation, dissolved in methacrylate monomer. Surprisingly, based on the previous evidence, the points from a 4 fold range of current density fall on a common line. The resistance is relatively low in this case and there is not a wide range of values, but a different study gives the same effect (Figure 7). In this case a 1.6 double bond per thousand acrylic with methacrylate unsaturation is dissolved in butyl acrylate. Again, the points from a 4 fold range of current densities fall on the common line. The spread of points at high dose, indicates the limitation of the test method at high resistance values.

Figure 8 represents data in which acrylic resins of identical backbone content and unsaturation content are cured over an 8 fold range of current density. Every combination of acrylate-methacrylate unsaturation of monomer and polymer are tested at 6 Mrad. As in prior cases of acrylic resins of this type there is no indication of a dose rate effect that would be of concern to

the technologist seeking to operate with electron generating equipment of high output capability.

CONCLUSIONS

This paper provides enough information to identify the kinds of conditions which affect dose rate sensitivity. It shows that dose rate is a factor to be reckoned with in resin design work. The information does indicate that there are resin systems that can be employed which will reduce or even eliminate the dose rate problem. Parameters in need of further study are monomer type and concentration, molecular weight, and the behaviour in very early stages of cure.

There may be two phases in the curing process. The cure may begin by a process which is dose rate sensitive. In this stage there would be a high degree of mobility on a molecular scale and processes very close to the early stages of addition copolymerization may occur.

In the second stage a condition of restricted molecular motion, due to gel formation, may result in dose rate independent interactions in the curing process.

This work does show that coatings which would reach gel formation most rapidly, i.e. the high molecular weight acrylic resin with its high functionality per molecule and the fast reacting acrylic unsaturated materials, do not show dose rate sensitivity. In these cases there may have been a dose rate sensitive region but that appeared at a stage of cure prior to that which we measured.

The resin systems which showed dose rate sensitivity, the maleate-fumarate polyesters and low molecular weight polymers with pendant methacrylate unsaturation, may still be in the first stage of the curing process.

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ABBREVIATIONS USED IN FIGURES AND TABLES

MEK - methyl ethyl ketone rubs

DEPT- double bonds (moles) per thousand grams of polymer

Internally Unsaturated Polyester Resin

2.5 DBPT

80% polyester

20% styrene

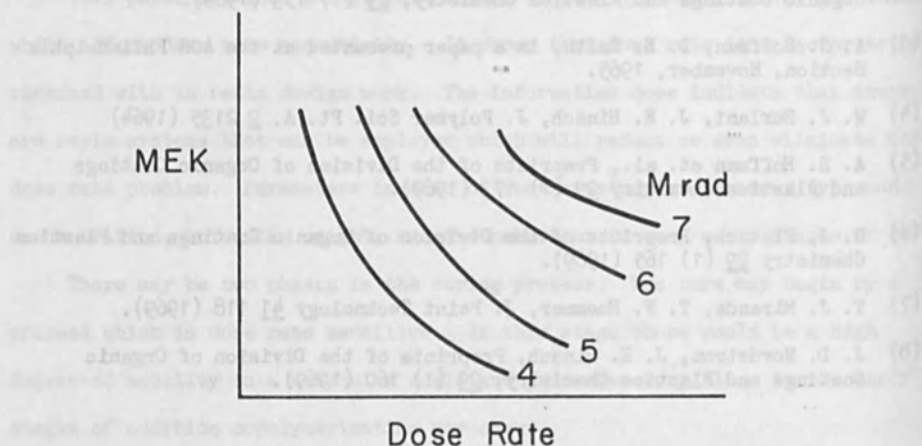


Fig. 1. Effect of Dose Rate Upon Maleate/Fumarate Polyesters.

CHEMISTRY OF VINYL POLYESTERS

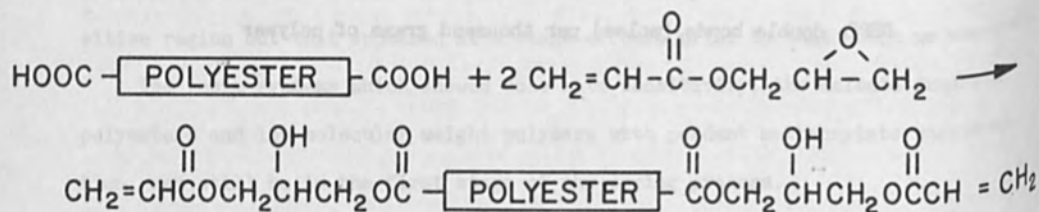


Fig. 2. Chemistry of Vinyl Polyesters.

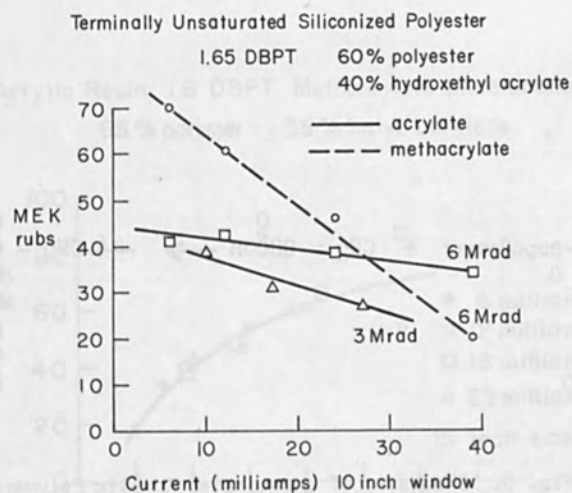


Fig. 3. Effects of Dose Rate Upon Acrylate and Methacrylate Terminated Polyesters.

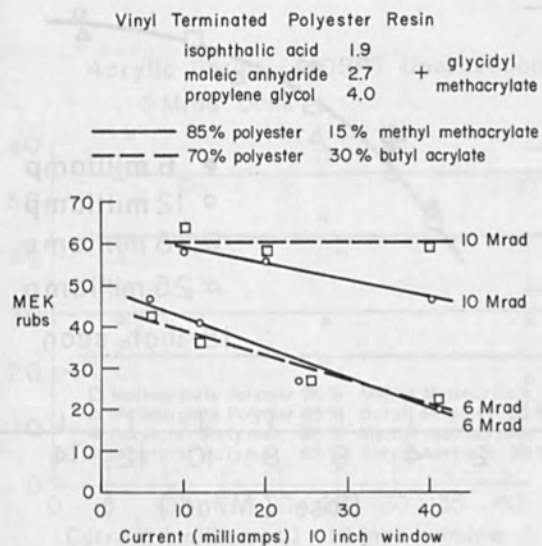


Fig. 4. Effect of Monomer Type on Dose Rate Sensitivity.

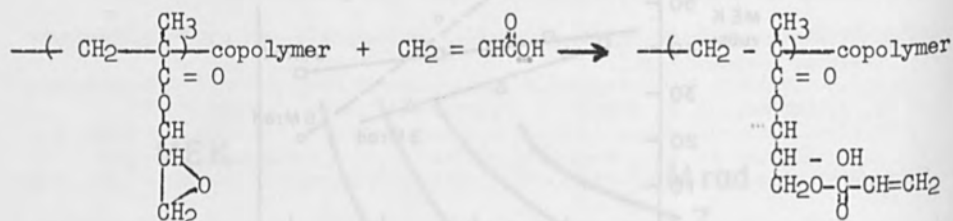


Fig. 5. Chemistry of Unsaturated Acrylic Polymers.

Acrylic Resin 0.9 DBPT Methacrylate Unsaturation
 polymer 85% methyl methacrylate 15%

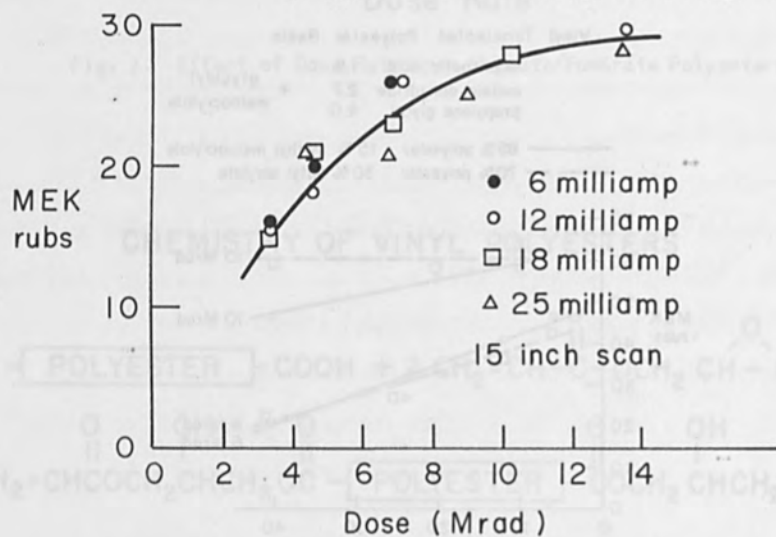


Fig. 6. Dose Rate Effects on Unsaturated Acrylic Polymer.

Acrylic Resin I.6 DBPT Methacrylate Unsaturation
65 % polymer 35 % butyl acrylate

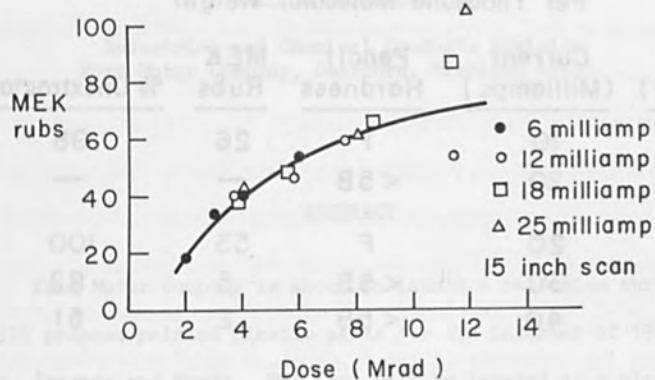


Fig. 7. Dose Rate Effects on Unsaturated Acrylic Polymer.

Acrylic Resins I.3 DBPT Unsaturation
6 Mrad Dose

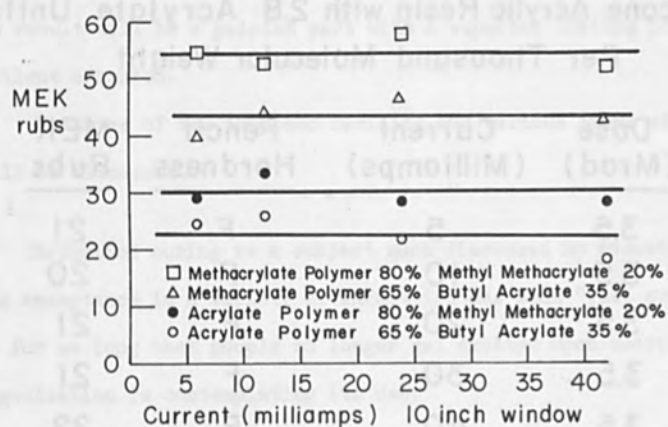


Fig. 8. Dose Rate Effects on Unsaturated Acrylic Polymer.

Table 1. Dose Rate Effects on Methacrylate Terminated Silicone Resin

Silicone Acrylic Resin with 2.7 Methacrylate Units
Per Thousand Molecular Weight

<u>Dose (Mrad)</u>	<u>Current (Milliamps)</u>	<u>Pencil Hardness</u>	<u>MEK Rubs</u>	<u>% Unextractable</u>
7	10	F	26	98
7	20	< 5B	—	—
12	20	F	53	100
12	30	< 5B	5	82
12	40	< 5B	2	51
20	10	F	> 100	
20	40	F	> 100	

Table 2. Dose Rate Effects on Acrylate Terminated Silicone Resin

Silicone Acrylic Resin with 2.8 Acrylate Units
Per Thousand Molecular Weight

<u>Dose (Mrad)</u>	<u>Current (Milliamps)</u>	<u>Pencil Hardness</u>	<u>MEK Rubs</u>
3.5	5	F	21
3.5	10	F	20
3.5	20	F	21
3.5	30	F	21
3.5	40	F	22
12	30	F	25

ELECTROCURE PROCESSING
APPLIED TO PLASTIC AUTOMOTIVE PARTS

Allen H. Turner

Industrial and Chemical Products Division
Ford Motor Company, Dearborn, Michigan 48121

ABSTRACT

Ford Motor Company is about to launch a radiation curing line which will process painted plastic parts for the interior of 1971 Mercurys, Cougars and Fords. The line will be located at a plastics plant in Saline, Michigan.

Special electron sensitive paints will be used with electrons of energies up to 300,000 electron volts. The impingement of these electrons on the coatings will cause instantaneous polymerization and the end result will be a painted part with a superior coating possessed of excellent adhesion.

Features of the proposed facility and various items of equipment will be discussed.

* * *

Radiation curing is a subject much discussed in recent years. Its wide acceptance in a variety of industries has been "just around the corner" for so long that people no longer get excited upon hearing that some organization is contemplating its use.

We at Ford, however, have taken the plunge. We have committed to the curing of paint by radiation on 1971 automotive parts. These are

plastic parts for interior use. This activity will be carried out by our General Parts Division in our Saline, Michigan Plant.

The facility has been constructed, accelerators have been installed, and a number of pre-production runs have been carried out. Full production is scheduled for June, 1970.

This is the calculation of a very extensive research and development program dating back to the mid-50's when the initial coatings work was done. Over the intervening years, various milestones were reached and the program expanded until the Industrial and Chemical Products Division was formed in 1967. The radiation curing activity became part of this Division and we began the work of adapting the process to a variety of uses and industries.

The process which carries the copyrighted name of Electrocare, involves the interaction of energetic electrons with certain specially formulated organic coatings. This interaction leads to the formation of cross-linked, or cured films with a broad range of properties.

The number and distribution of electrons which can be brought to bear on a typical coated part determines the rate at which one can generate a product, so we have spent considerable time developing accelerator concepts which permit increasingly higher current outputs, while maintaining high reliability.

The key device in the process is an electron accelerator. Figure 1 shows an exploded view of a typical unit.

Power for the accelerator comes from a D.C. source which is not shown. This power is introduced by means of the two cables which

also carry A.C. power which is needed to heat a tungsten filament which then emits electrons.

The electrons are accelerated in the evacuated column by means of a series of electrodes which serve to accelerate them in stages rather than all at once. Voltages as high as 300 kilovolts are used and this must be coupled to the column in a manner which is not likely to encourage a sparkover. An insulating gas under pressure coupled with appropriate electrostatic shielding is the normal approach.

The electron beam formed and accelerated in the column passes into a drift zone where it can be scanned at some nominal frequency through an appropriate angle so as to fill a given window aperture.

The "window" is typically a gas tight, thin foil of a light element such as Aluminum or Titanium which will allow high energy electron beams to pass through without giving up excessive energy.

Figure 2 shows an assembled accelerator with the vacuum hardware in place.

It is in the nature of things that when energetic electrons impinge on matter, x-rays are generated, so the accelerators and the part being irradiated must be isolated from personnel by suitable shielding means. Such shielding is normally accomplished through the use of concrete and/or lead with the thicknesses and placement chosen so as to keep the external radiation intensity below levels prescribed by state regulations.

Typically, the equipment for an Electrocure facility will also include an inert gas generator--an item common to heat treating processes. This device burns a mixture of air and natural gas to

produce mainly nitrogen, CO_2 and water vapor. This generated gas is used to purge the curing zone thus minimizing the oxygen content. This is desirable for two reasons. First of all, oxygen can inhibit the curing of some of the coatings we have developed. Its exclusion facilitates a proper cure and reproducible results. Secondly, since the action of the beam on ordinary air produces ozone, it is desirable to purge it for health and pollution reasons as well.

In the new plant the process flows as shown in Fig. 3.

The plastic parts are loaded onto an overhead monorail conveyor and pass through a prep spray in an enclosed booth. This material renders the surface conductive so that they can then be electrostatically painted with Electrocure coatings in one or both of the spray booths shown depending on the needs of specific parts.

The application technique is that of an electrically charged spinning disk. The disk moves up and down to cover the vertical extent of parts on the hooks. Paint is fed to the disk axially and proceeds to the edge because of centrifugal force. There it is charged and atomized.

Following this operation the parts enter the radiation chamber and pass through the electron outputs of the four accelerators which irradiate the parts through slots in the chamber wall.

After curing, the parts pass out of the chamber to a zone where they are removed, assembled and boxed for shipment.

In this facility we expect to coat and cure over eight million parts a year. These parts will be irradiated at up to 300,000 volts using as much as 150 milliamperes of beam current.

As mentioned earlier, we exercise control over the oxygen content of the gas in the curing zone. As you can see in Fig. 3, the conveyor is enclosed in a rather voluminous inner chamber which is approximately 10 ft. high, 8 ft. deep, and 60 ft. in total length. The inner chamber is dead-ended in the vicinity of the accelerators and the output from a 40,000 cubic ft. per hour gas generator is introduced there and sweeps the chamber through a series of part silhouettes in the entrance chamber. Various internal arrangements are provided to achieve proper mixing of the nitrogen and incoming air and to control temperature gradient effects on gas flow. We have been able to maintain oxygen concentrations of less than .1% routinely with the line loaded and moving at over 20 ft. per minute.

For the new plant, the Electrocure process offers a number of specific advantages:

1. Shrinkage and warpage of the plastic parts will be minimized since exposure to the sustained heating of an oven will be eliminated.
2. The cured coatings will provide superior hardness and abrasion resistance. This will allow parts to be handled immediately after curing and it will permit the use of simplified packing techniques.
3. The coatings possess superior adhesion to the substrate. This seems to be a general characteristic of the process where organic substrates are used.
4. The coatings are 100% reactive which means that all constituents interact in the beam, and there is little material lost during the 1 to 2 seconds of beam exposure. Thus, the Electrocure

coatings will provide as much as three times the coverage of conventional coatings for equal dry film thicknesses. The reduction in the amount of pollutants released by the process because of this reactivity is a very significant feature.

5. The liquid coatings are extremely stable. Shelf life is essentially unlimited as a result.

6. Higher curing speed can be obtained in the future without incurring a serious floor space penalty. In this installation, for instance, a second power supply or a higher output single unit is all that is needed to permit doubling of the line speed.

Whereas in the past it has been assumed that this process is particularly suited to the curing of films on essentially flat substrates, in the new plant some rather complicated parts will be painted and cured.

Figure 4 shows several instrument panels--one from a Ford and one from a Mercury automobile. One is 55" long by 19" wide and 10" deep. The other is 62" long by 9" wide and 7" deep.

Figure 5 shows several of the various finish panels which must also be processed along with other items not shown such as glove box doors and ash tray covers.

It should be obvious that special properties are needed in both paint and equipment in order to facilitate the curing of such parts. First of all, the coatings are so designed that they can tolerate exposures to radiation quantities far beyond that required for adequate cure without suffering a serious loss of film properties. Thus, we can cure effectively even though one area of a part passes

quite close to an accelerator window while another area is quite far away. Still we cannot adequately cure all areas of some of the parts with one orientation, so the curing zone provides for the indexing of hooks and the exposure of a given part from two different aspects.

Figure 6 shows two accelerators, one positioned high and one low so as to cover the length of a given part or array of parts. After passing these accelerators, the support hooks are rotated to a new position, and the parts are then exposed to the second pair of accelerators. In this way we are able to cover parts of considerable geometric complexity--depending greatly on air scatter and metal beam deflectors to assist in obtaining proper cure on difficult areas.

This facility is a first of its kind in the automotive industry. We have encountered and solved many problems during its planning and installation. It functions as planned, and we are gaining operational know-how which will be used to facilitate the rapid introduction of Electrocure into other automotive applications and other industries as well.

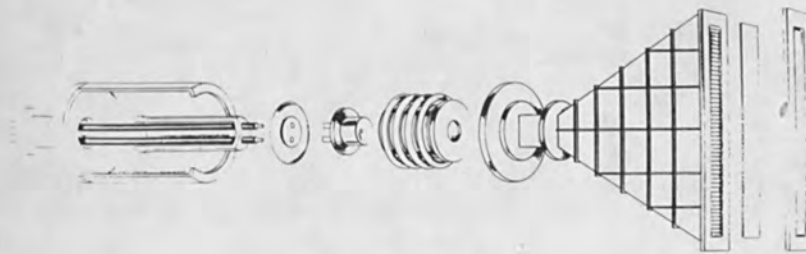


Fig. 1. Exploded View of Typical Accelerator.

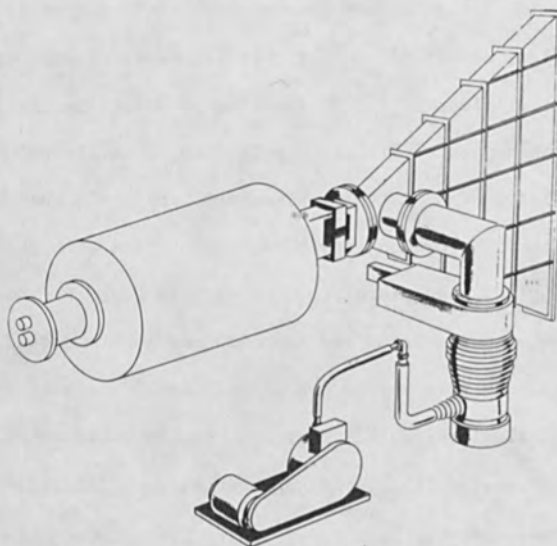


Fig. 2. Assembled Accelerator.

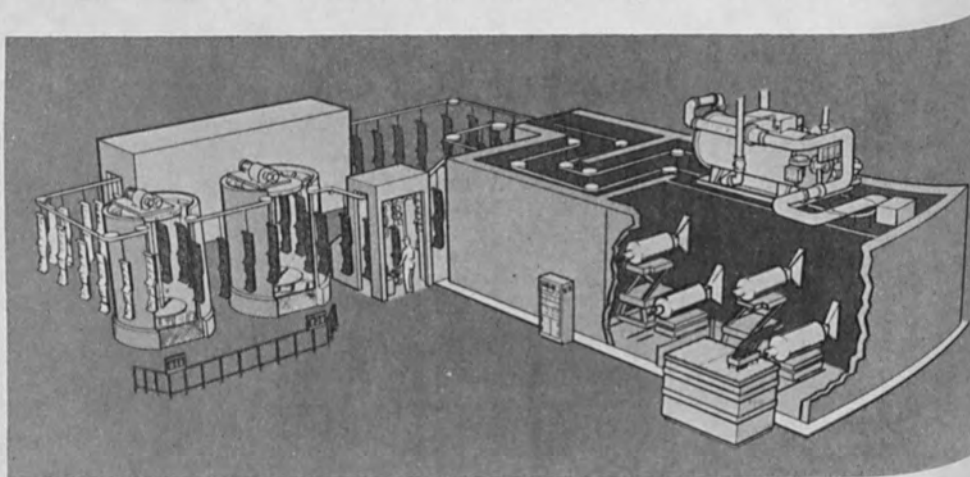


Fig. 3. Saline Electrode System.

STUDY OF ATMOSPHERES FOR USE IN THE
IRRADIATION PROCESSING OF COATINGS

Thomas E. Schuler & James F. Higawell
Research Department - The O'Brien Corporation
South Bend, Indiana 46601

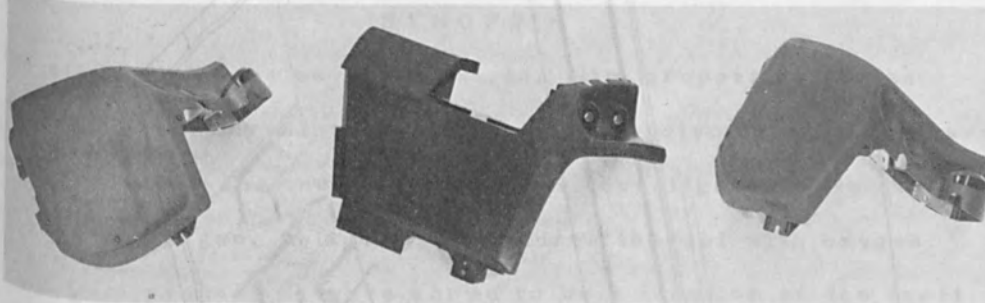


Fig. 4. Instrument Panels

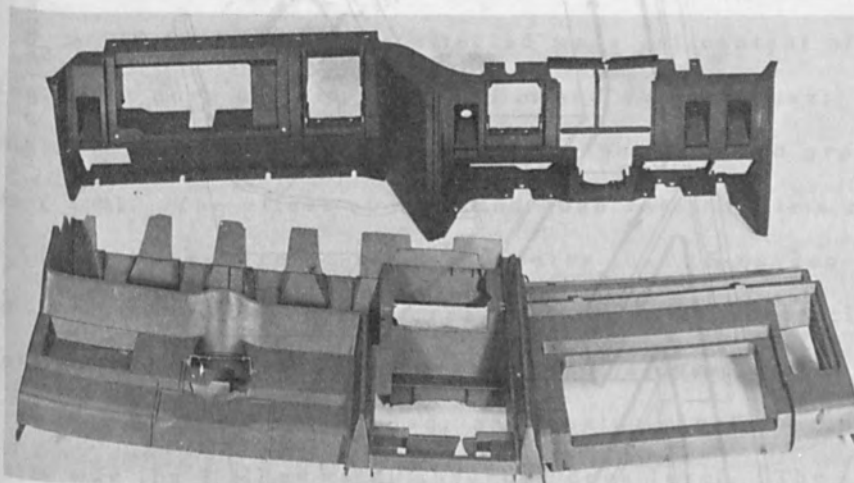


Fig. 5. Finish Panels



Fig. 6. Chamber Overview.

STUDY OF ATMOSPHERES FOR USE IN THE

IRRADIATION PROCESSING OF COATINGS

Thomas F. Huemmer & James R. Ungurait
Research Department - The O'Brien Corporation
South Bend, Indiana 46621

SYNOPSIS

The effects of cure environment, on film properties for an electron beam cured urethane-acrylic prepolymer coating, have been studied. The inert atmospheres investigated were carbon dioxide, nitrogen, helium and mixtures thereof with oxygen. Some film properties were shown to be a function of the inert atmosphere employed when that atmosphere contained 1% oxygen or greater, and in these cases the order of activity was $\text{He} > \text{N}_2 > \text{CO}_2$. Parameters affected were gel content of the coating, post cure activity of the film and surface cure; parameters unaffected were chemical composition and processing losses (2 %). The effect of dose and dose rate on films cured in an inert atmosphere containing greater than 1% oxygen became more pronounced at increasing oxygen concentrations. For atmospheres essentially oxygen free the dose to tack free cure was established at 1 MRad, and maximum gel content of the film was the 2 MRad total absorbed dose level. The inhibited surface layer of films cured in atmospheres containing oxygen appeared to be low molecular weight species, and no unconverted monomer was indicated.

INTRODUCTION

In order to realize many of the advantages of the radiation curing of coatings mentioned by authors (1, 2, 3) on the subject, it is absolutely essential that a cured film of high integrity be developed. A tack free mar resistant surface having good chemical resistance is generally required. The properties of such a coating should at least equal or exceed those of the high performance coatings marketed today. Many of the systems curing in the total dose region defined as acceptable for coating processing are formulated around relatively expensive materials, therefore, these coatings are forced to compete in the area of quality finishes. Once film performance has been developed, then the full potential of radiation curing of coatings can be realized.

The key to the development of the surface described above appears to be principally in the irradiation atmosphere of the processed film. Although paraffin waxes have been successful in the UV processing of coatings (4), their use in electron beams thus far has been disappointing probably because of the very rapid cures employed which do not allow sufficient time for the wax to form a continuous protective film for the surface. Similarly other additives have been found to be equally disappointing for solving the air inhibition problem. On the other hand, the exclusion of oxygen during cure with the electron beam has been successful in the development of good film properties. Recent reports

of electron beam processing using inert atmospheres most often mention nitrogen (5) although reference has been made to helium (6) and carbon dioxide (7) & (8) as well. Nitrogen is available in bulk quantities at high purity. Carbon dioxide is easily generated although nowhere near the purity level of nitrogen. Helium, on the other hand, is expensive, but it is known for its activity (20) with surfaces. Also, helium is lighter than air and for this reason offers some interesting inert chamber designs for radiation processing (8).

In the design of the inert chamber for radiation processing of coatings, it is to be expected that some level of oxygen must be tolerated whichever cure environment is employed. There are possibly three levels to be investigated:

- (1) The region where oxygen is all but excluded, which might be less than about 100 ppm.
- (2) The intermediate region where most of the oxygen is excluded, which might range from about 100 ppm up to perhaps 10% oxygen.
- (3) The region where little oxygen is excluded which would range up to the oxygen content of air.

There is very little mention of atmosphere effect in the radiation curing of coatings in the literature other than the suggestion that inert atmospheres

are a solution to the air inhibition problem.

The system selected for study is a very responsive trivinyl urethane prepolymer dissolved in monomer and this mixture is well adapted to the radiation processing of coatings (10). A divinyl precursor to the system has been studied by Labana (11 & 12). The vinyl groups in the system studied by Labana, however, were methacrylate functions in contrast to the acrylate functions investigated in this work. Acrylate functions have been shown to be far more responsive in the lower dose region (13) which is of current interest to the coatings' industry.

The intent of this work is to evaluate the various inert atmospheres suggested for radiation processing and determining the effect of oxygen content of the gas on coating properties. The system selected for evaluation is one capable of developing the processing advantages envisioned for the radiation curing of coatings.

EXPERIMENTAL

The Prepolymer System

The structure of the prepolymer studied is shown in Fig. 1. This prepolymer was prepared by reacting as received Pluracol 440 Polyether Polyol (1.0 mole) from Wyandotte Chemical Company with TD 80 Toluene Diisocyanate (3.0 mole) from Mobay Chemical Company at $70 \pm 10^\circ \text{C}$. until the isocyanate value reached the theoretical for the intermediate (13.3%).

To this mixture 2-hydroxyethyl acrylate (3.0 mole) was added at $60 \pm 10^{\circ}\text{C}$. until the isocyanate value reached zero. 2-hydroxyethyl acrylate was used as received from the Dow Chemical Company. Finally the reaction product was reduced to 50% by weight with additional acrylate monomer (9.5 mole percent prepolymer). The viscosity of this mixture was 450 ± 100 cps.

The Inert Atmospheres

High purity helium (99.99% pure) was obtained from Linde Division of Union Carbide Corporation; high purity dry nitrogen (99.9% pure) was obtained from Linde Division of Union Carbide Corporation; and commercial grade carbon dioxide (99.5% pure) was obtained from Linde Division of Union Carbide Corporation.

Extra dry grade oxygen (99.7% pure) from Linde Division of Union Carbide Corporation was programmed into the atmosphere under investigation at the desired level and a Westinghouse Oxygen Analyzer, Model 209 (sensitivity 1 ppm) was used to determine the exact concentration. The mixed gases were fed into a chamber (8" x 12" x 1.75") having a 1 mil polyethylene window, after first inserting the film specimen to be evaluated.

Polymerization

The absorbed dose was measured by the blue cellophane method (14). Polymerizations were carried out at room temperature using a 300 kv electron beam. Time average dose rates were determined by a method developed by High Voltage Engineering Corporation (15).

Gel Faction Analyses

A film of the desired thickness was drawn down with a Bird Applicator on tin free steel substrate. After one hour at room temperature, approximately 0.02 gram of this film was removed from the substrate with a razor blade and placed in a 300 Series stainless steel wire mesh thimble obtained from Mectron Industries. This thimble was then placed in a Micro Soxhlet Extractor and extracted with methyl ethyl ketone solvent for a period of 24 hours. The thimble and contents were dried to a constant weight in a vacuum oven at 80^o C. The weight of the polymer remaining was taken as the weight of the coating gel faction. Results were reproducible to $\pm$ 2%. All samples were determined in triplicate and averages were taken for the gel content reported.

Material Balance Analyses

A 3 mil film was drawn down with a Bird Applicator on aluminum substrate. Then a 3.5" x 4" section of this film was cut from the coated substrate, immediately weighed and inserted into the cure chamber. The cure chamber was purged with the appropriate premixed gas at a rate of 100 cfh for approximately 30 seconds. The total elapsed time from drawdown to cure was approximately 6 minutes. After removal from the cure tray the specimen was weighed each minute over a 20 minute increment. The base weight of the metal substrate was obtained by immersing the coated specimen in acetone overnight and stripping the swollen film from the substrate. The base weight of the substrate was determined after drying to constant weight.

Elemental Analysis

Elemental analysis was carried out by Midwest Industries, Inc. Indianapolis, Indiana. Cured films were prepared similar to the method described in the gel fraction analyses above. Duplicate results were obtained and averages were determined for the reported values.

RESULTS AND DISCUSSION

Gel Fraction Measurements

The gel contents of thin films cured in various inert atmospheres containing various levels of oxygen varying from 10,000 to 75,000 (1% to 7.5%) are plotted in Fig. 2. From this data it becomes obvious that significant atmosphere effects began to appear above the 10,000 ppm (1%) oxygen concentration; and, although not large, these effects became more pronounced at increasing oxygen levels. If the inert atmosphere contains less than 10,000 ppm (1%) oxygen the gel contents are well within the accuracy of the experimental technique.

The generally higher gel contents obtained for the carbon dioxide atmosphere is attributed to the stability of this gas to ionizing radiation. Principal radiolyses products of carbon dioxide have been shown to be carbon monoxide, oxygen, and ozone (16). Reported G values are as follows:

Carbon Monoxide	3.5 to 5.0
Oxygen	0 to 0.6
Ozone	0.7

Similar decomposition products have been reported when gaseous

carbon dioxide is exposed to ultraviolet light (17). If appreciable quantities of the radiolyses products are produced, it is reasonable to assume a back reaction of carbon monoxide with either oxygen or ozone will take place thus stabilizing the atmosphere. The back reaction has been shown to proceed almost quantitatively and a chain reaction involving C_2O and C_3O_2 has been proposed in which these molecules act as catalyzers for the reaction of carbon monoxide with either oxygen or ozone (18). If the principal reactions with the coating surface involve either oxygen or ozone, then competitive surface effects might be minimized in carbon dioxide atmospheres... particularly when the system under study is capable of a very fast chain reaction when exposed to ionizing radiation. The addition of oxygen to the carbon dioxide atmosphere results in an increase in the ozone concentration upon irradiation, but has little or no effect on the carbon monoxide concentration (16). Thus only upon the addition of significant quantities of oxygen do some surface reactions become apparent in the data shown in Fig. 2.

Irradiation of mixtures of nitrogen and oxygen yield large amounts of NO_2 which does not readily decompose ($GNO_2 \approx 0.075$) (19). When irradiated, NO_2 can react with N_2 , however, forming both N_2O and NO of which the proportions are largely dependant upon the starting mixtures of N_2 and O_2 . These reaction products can behave as radical scavengers and could tend to inhibit the propagation reaction on the coating surface. In

addition, the lower molecular weight of the inert gas could facilitate these surface reactions with either oxygen, ozone, or the radiolysis products. Thus, in Fig. 2, slightly lower gel contents are shown for nitrogen than for carbon dioxide for the same oxygen concentration in the inert atmosphere.

The greatest surface activity in the presence of oxygen for the gases investigated was observed for helium. Being the lightest of the gases studied, the excited helium atoms are not readily quenched and hence more activity might be predicted. A method of activating polymer surfaces using helium has been reported by several investigators (20 & 21). In these studies attack of the surface with activated helium atoms resulted in the expected oxidation, cross-linking and production of unsaturation through postulated free radical reactions. Since helium is quite unreactive with oxygen, it might be expected that an appreciable amount of free radical telomerization involving oxygen might take place on the surface if the helium atmosphere contains significant quantities of oxygen.

The effect of film thickness on gel content is depicted in Fig. 3. The inhibiting effect of oxygen is clearly demonstrated by the gel content values obtained for specimens cured in air where definite film thickness relationship exists. Lower gel contents are to be expected with thinner films where the surface to volume ratio becomes larger. The independence of gel content as a function of film thickness in pure nitrogen might be also

expected for carbon dioxide and helium atmospheres as well.

Films cured in air failed to cure tack free; the gel data for these tacky films, though indicates that the tacky layer is not deep and is a constant, i. e., if the film thickness is halved the extractibles are doubled. This inhibited layer is calculated to be 2.3×10^{-4} inches in thickness for this resin when cured in air at a 5 MRad total absorbed dose.

In order to evaluate the chemical character of the extractibles obtained from films cured in inert atmospheres containing oxygen, identical extractions were carried out using both ethanol-water and methyl ethyl ketone for the extraction solvents. The data is tabulated in Table I and indicates that the chemical character of the extractibles might be similar since the gel contents remain independent of the extraction solvent. This fact suggests that the soluble fraction could be dimers, trimers, or other low molecular weight species. Ethanol-water is a known poor solvent for high polymers, whereas methyl ethyl ketone is considered a good solvent. The chemical nature of the extractibles becomes important in certain coating applications, for example, those coming in direct contact with foods. Furthermore, it has been suggested that coatings have a molecular weight below about 5,000 are deficient in the development of good coating properties (22).

The effect of total dose on gel content at various dose rates is plotted in Fig. 4 for cures in a typical inert atmosphere. This data points to the responsiveness of the system under study and it becomes apparent

that a total dose in the region of 2 MRads is sufficient to cure this system under the condition cited. This responsiveness is attributed to two factors:

1. The structure of the prepolymer and location of radiation sensitive groups.
2. The utilization of acrylate functions which have been shown to be more responsive to ionizing radiation.

From the data shown in Fig. 5 it is also evident that dose rates of films cured in an inert atmosphere do not effect gel content over the range tested. If a total absorbed dose to cure of 2 MRads is established for this system, a G value can be estimated (23) if the starting molecular weight is calculated. This G value was estimated to be about 670 which is approximately four times greater than that calculated for a polyester-styrene system (23). This very high G value points to the very fast chain type reaction and should be compared with a typical acrylate monomer (G approximately 25) (24), containing no prepolymer type material. The contribution of the prepolymer to cure rate is then demonstrated.

Free radical polymerizations generally tend to terminate by bimolecular reaction, and if certain assumptions are made, then a dose rate dependency should exist according to the dose rate raised to the 0.5 power. The failure to develop this dose rate dependency suggests

non-steady state kinetics. An alteration in the termination rate constant accompanied by the well known Tromsdorff effect is postulated, thus radicals become trapped and buried in the polymer chain.

The data plotted in Fig. 4 indicates a very low dose to gel; in fact, dosimetry at these low total absorbed doses becomes difficult to measure with any degree of accuracy. In Fig. 4 it also becomes apparent that the initial rate of gel formation is very high. This fact, coupled with the relatively low viscosity for the system, is probably a prime factor in the development of very high gel contents at low doses (12).

The effect of gel content as a function of dose rate for films cured in oxygen containing atmospheres at a total absorbed dose of 5 MRads is shown in Fig. 5. Again oxygen effects become apparent above a concentration of 10,000 ppm (1%) in the inert atmosphere. In this plot dose rate effects become evident.

These dose rate effects are attributed to surface reactions with oxygen since it has been shown earlier (Fig. 4) that no dose rate effects are observed for films cured in a non-oxygen containing atmosphere.

In comparing Fig. 5 with Fig. 6 where similar data has been collected only at a lower total absorbed dose, dose rate effects become a little more pronounced at lower levels of oxygen in the inert atmosphere for films cured at the lower total dose. This result might be associated with a slight undercure of the film at the lower total absorbed dose, thus oxygen might more readily diffuse into the film and inhibit the cross-linking

reaction.

The above results clearly demonstrate that the inhibiting effect of oxygen is diminished as the dose rate is increased at any fixed oxygen level in the inert atmosphere. Similar secondary oxygen effects have been mentioned by Davison (25) for systems where the doses to cure are relatively insensitive to dose rate, or where non-steady state kinetics are suggested.

Cure Analysis

Although frequently referred to in coating technology the term "cure" or "dose to cure" as applied to radiation curing needs clarification. Most often the term cure refers to that point where the surface just becomes tack free and this point may or may not coincide with that point where the best balance of properties is achieved.

In the previous dose rate investigations, it has been suggested that surface cure can be promoted by increasing the dose rate at a fixed level of oxygen concentration in the inert atmosphere. Another approach frequently employed in radiation curing for developing tack free surfaces for cross-linking systems is to increase the total dose until the surface becomes tack free. A "dose to cure" curve is shown in Fig. 7 for the system under study up to a total dose of 10 MRads.

The data plotted in Fig. 7 shows that the total dose required for tack free cure is dependant on the cure atmosphere at any fixed oxygen concentration.

The data obtained fits the following straight line relationships over the range evaluated:

$$D_c = 0.86 (O_2) + 1.0 CO_2 \quad (1)$$

$$D_c = 0.92 (O_2) + 1.0 N_2 \quad (2)$$

$$D_c = 1.41 (O_2) + 1.0 He \quad (3)$$

According to the above equation, tack free cures occur in the region of one megarad in an oxygen free atmosphere. It should be emphasized that this point does not coincide with the maximum gel content (Fig. 2), and therefore, maximum film properties, i.e., hardness, chemical resistance, flexibility, would not necessarily be developed at this total dose level.

Some correlation of film properties with surface cure are shown in Table II where chemical resistance as measured by blush of the film after boiling water immersion is tabulated as a function of oxygen content of the inert atmosphere. Blush begins to appear at around the 2-3% oxygen level, or slightly below the tack free point at this total absorbed dose level. The tack free point calculated from equation two would be at 4.3% oxygen at the 5 MRad level, which agrees well with the data tabulated in Table II.

Material Balance

Results obtained from material balance investigations are summarized in Table IV. In general, approximately a 2% processing loss has been established which appears to be independent of total dose

and the cure atmosphere. The slight variations observed do follow the order of gas activity, i.e., the losses for helium greater than nitrogen and nitrogen greater than carbon dioxide. The failure to observe large processing losses in atmospheres containing oxygen point to no residual surface monomer which might be expected to volatilize. Similarly, no total dose relationships might confirm the fact that there is little or no degradation products at least up to 10 MRads. These results might be expected since high gel contents remain down to approximately 2.0 MRads. They also support the fact that the inhibited layer on the surface may be composed of low molecular weight species.

The post cure activities of the irradiated films as measured by weight gains after radiation are shown as a function of total dose in Fig. 8. The total dose effect shown is in the expected order, that is, more weight gains were observed at the higher total dose. This fact suggests that trapped radicals are formed in the immobilized network as suggested earlier. The decline in weight gain with time could be explained by the diffusion of oxygen to the radical sight. The termination of trapped radicals with oxygen might be in competition with terminations from the sol phase with the gel phase as suggested by Labana (12). Unusually high mobilities have been postulated in irradiated organic solids (26) and a polymeric system having a low T_g might be expected to more readily engage in terminations through this route.

The post cure activity of the irradiated films as measured by weight gain after irradiation as a function of cure atmosphere are shown in Fig. 9. Again the post activity follows the expected order of the gas activity, that is, helium greater than nitrogen, and nitrogen greater than carbon dioxide. The much lower values of post cure activity observed for films cured in atmospheres containing oxygen can be attributed to surface reactions taking place with oxygen (or ozone) during the cure process. An inhibited layer is formed which then becomes unreactive with oxygen after cure.

Film hardness as a function of post irradiation time is shown in Fig. 10. The time to achieve maximum hardness correlates with the post cure activity time shown in Fig. 8 & Fig. 9. Evidently additional cross-linking occurs on the surface of the film after irradiation.

The post cure activity of irradiated films is shown to be a function of total dose as well as the cure environment. These facts suggest a surface phenomena involving free radicals. The reduction in post cure activity with time appears to involve oxygen to some degree although the exact mechanism is not postulated, some additional surface cross-linking is suggested.

Elemental Analyses

The results obtained from elemental analyses for both wet and cured films are tabulated in Table IV. In each case duplicate analyses

were obtained and the results averaged. Where no tolerances are given the value was obtained by difference. From the data tabulated in Table I, it is observed that the chemical composition of both the wet and cured films agree very closely with the theoretical and that there are no detectable shifts for the inert atmospheres investigated, nor are there any composition shifts in inert atmospheres containing oxygen. This data agrees with the material balance studies where losses were in the order of 1% to 2% in the respective atmospheres. The failure to obtain any significant increase in the oxygen content in the elemental analysis suggests that the oxygen up-take by the films is extremely small even with films having a significant air inhibited layer. From these elemental analyses, it can also be concluded that there are little or no ozone reaction products and that the attack of the film by oxides of nitrogen is not appreciable. Although the overall effects of oxygen are clearly demonstrated above 10,000 ppm in the inert atmosphere, results of these chemical analyses show that very little of this oxygen becomes an integral part of the final film.

CONCLUSIONS

From this work it can be concluded that film properties of a urethane-acrylic prepolymer coating cured with the electron beam are independent of the inert atmosphere cure environment provided the oxygen content of that atmosphere is 1% or less. Above the 1% oxygen level film properties are sensitive to the type of inert atmosphere employed and the

cure conditions. Other dominant factors in the selection of an inert atmosphere for a system of this type might be economics, chamber design, or possibly purity of the gas under consideration.

Acknowledgment

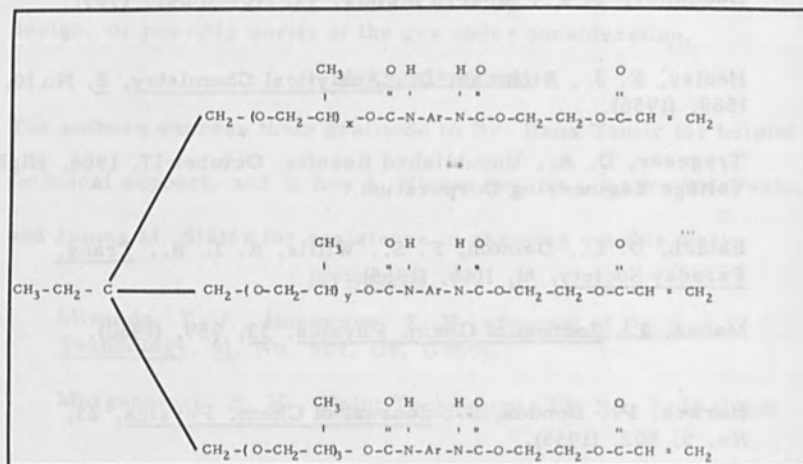
The authors express their gratitude to Dr. Ilana Tamir for helpful technical support, and to Roy S. Nickerson, Sr., Larry Wasowski, and James M. Slater for assistance in carrying out this work.

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Urethane Prepolymer Structure



Average Molecular Weight : 1294 (Calc.)

Figure 1

Gel Content of Coating as a Function of Oxygen Concentration in Various Inert Atmospheres

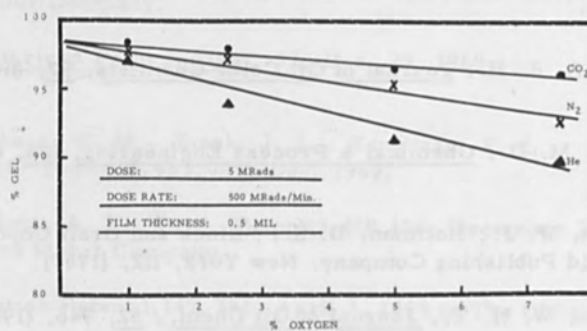


Figure 2

Gel Content as a Function Film Thickness

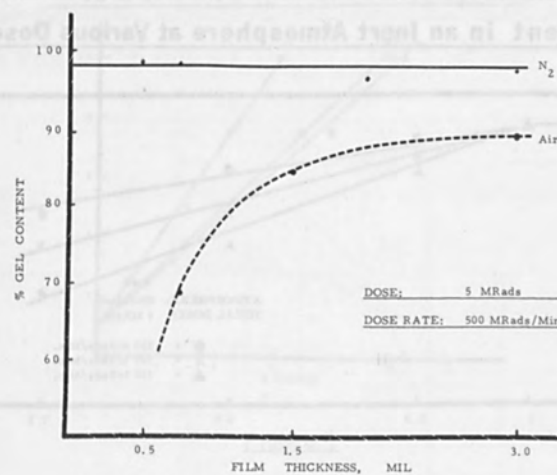


Figure 3

Gel Content as a Function of Total Absorbed Dose at Various Dose Rates

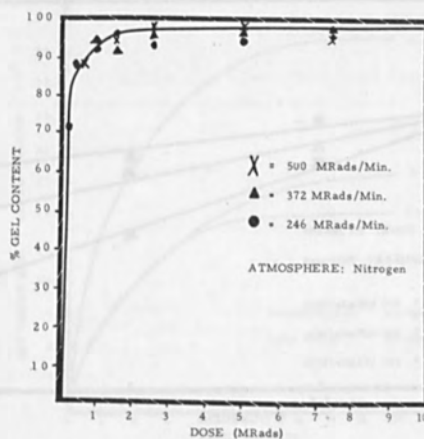


Figure 4

Gel Content as a Function of Oxygen
Content in an Inert Atmosphere at Various Dose Rates

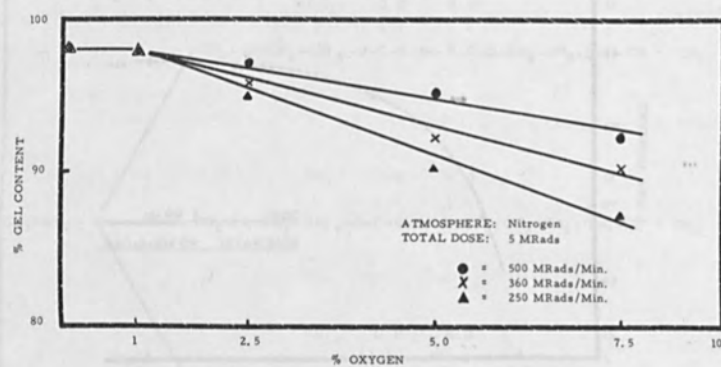


Figure 5

Gel Content as a Function of Oxygen Content
in an Inert Atmosphere at Various Dose Rates

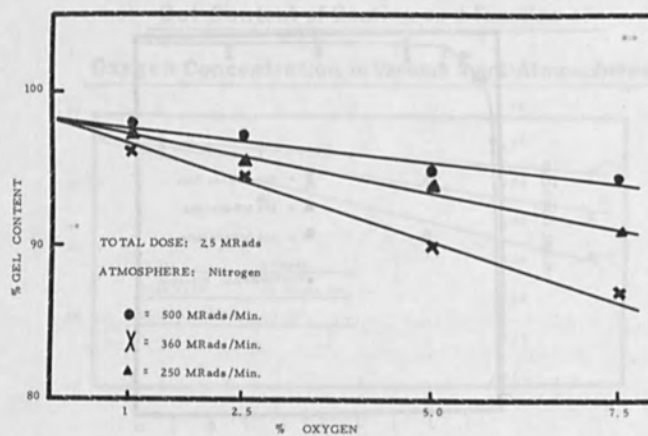


Figure 6

**Dose to Tack Free Cure as a Function of
Oxygen Content of the Inert Atmosphere**

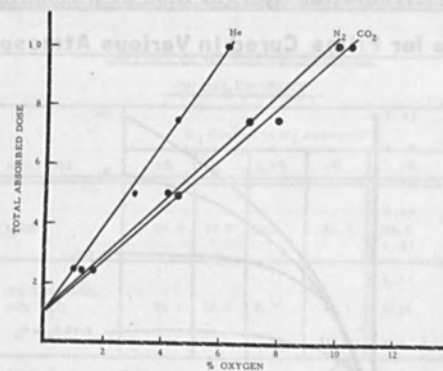


Figure 7

**Post Irradiation Weight Gain as a Function
of Time at Several Total Absorbed Doses**

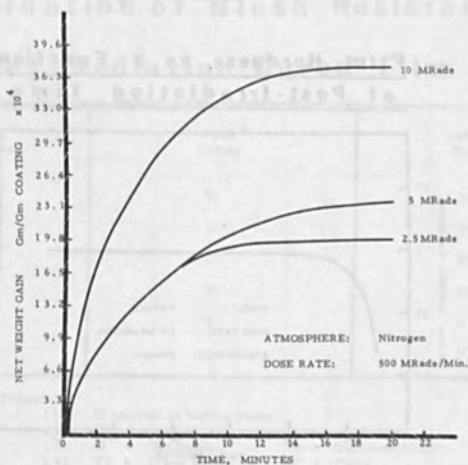


Figure 8

**Post-Irradiation Weight Gain as a Function of
Time for Films Cured in Various Atmospheres**

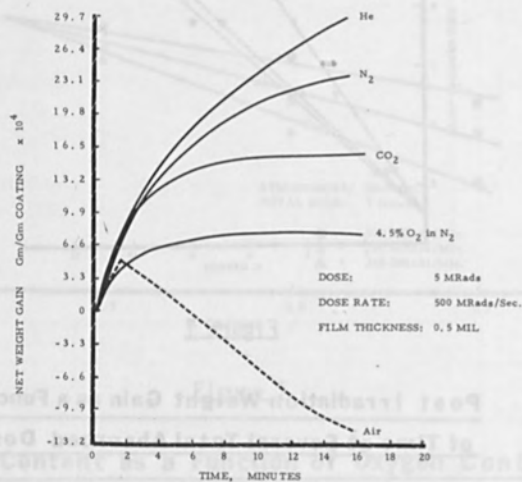


Figure 9

**Film Hardness as a Function
of Post-Irradiation Time**

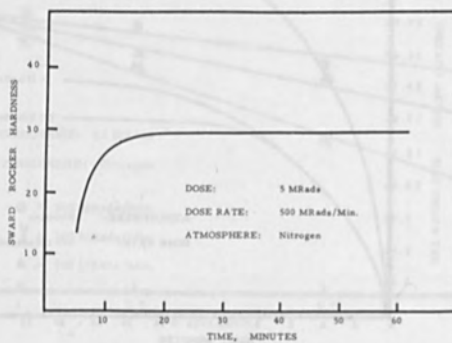


Figure 10

TABLE I

EFFECT OF EXTRACTION SOLVENT ON GEL CONTENT					
SOLVENT	O ₂ Content in N ₂ Atmosphere				
	0%	1%	2.5%	5%	7.5%
MEK	99.5	97.3	91.2	86.7	83.6
10% ETHANOL 90% H ₂ O	99.3	96.0	91.7	86.3	82.2
DOSE: 5 MRads DOSE RATE: 500 MRads/Min. ATMOSPHERE: Nitrogen					

TABLE II

Evaluation of Blush Resistance¹ of Radiation Cured Film

Oxygen Content ² of Inert Atmosphere	Blush ³ Rating	Surface ⁴ Cure
0.1%	10	TF
1%	10	TF
2%	10	TF
3%	9	TF
5%	8	T
7%	6	T
Notes: (1) 10 Minutes in boiling water. (2) Nitrogen. (3) 10 = No Blush 0 = Complete Blush (4) TF = Tack Free T = Tacky		

TABLE III

PROCESSING LOSSES AS A FUNCTION OF ATMOSPHERE AND TOTAL DOSE			
3 Mil Film			
Atmosphere	% Loss @ MRad		
	2.5	5.0	10.0
Nitrogen	1.64 ± 0.3	1.66 ± 0.2	1.59 ± 0.2
Carbon Dioxide	-	1.18 ± 0.1	-
Helium	-	2.62 ± 0.2	-
5% Oxygen in Nitrogen	-	2.41 ± 0.2	-
Air	-	2.07 ± 0.3	-

TABLE IV

ELEMENTAL ANALYSES OF RADIATION CURED FILMS

3 Mil Film Thickness

Element	Theoretical	Wet Film	% Composition for Films Cured in			
			Nitrogen	3% Oxygen in Nitrogen	Air	Carbon Dioxide
Carbon	59.16	58.39	58.64 ± 0.12	58.08 ± 0.10	58.33 ± 0.12	58.88 ± 0.13
Hydrogen	7.52	7.68 ± 0.08	7.83 ± 0.09	7.87 ± 0.11	7.55 ± 0.02	7.61 ± 0.04
Nitrogen	3.23	3.31 ± 0.01	3.39 ± 0.01	3.32 ± 0.09	3.36 ± 0.09	3.15 ± 0.03
Oxygen	29.90	30.43	29.95	30.54	30.57	30.17

ENERGY DEPOSITION IN SURFACE LAYERS OF
SILICON BY MONOENERGETIC ELECTRONS

S.E. Chappell and J.C. Humphreys

National Bureau of Standards

Washington, D.C. 20234

ABSTRACT

The energy absorbed from a normally incident electron beam by a thin silicon layer has been measured. The silicon layer was a transmission, semiconductor detector having a thickness less than the mean range of the incident electrons which had energies of 0.25, 0.50, 0.75, and 1.0 MeV. The fraction of beam energy absorbed in the silicon layer was determined from absorbed-energy distributions which were derived from pulse-height distributions measured with the detector. Measurements were made for two different cases: (1) the energy absorbed in the specific layer alone and (2) the energy absorbed in the same layer as a part of a homogeneous, semi-infinite medium, i.e., a surface layer. In the latter case, a larger fraction of energy is absorbed for each incident energy. The results were compared with Monte Carlo calculations, with the measurements and calculations showing agreement better than 10 percent.

INTRODUCTION

Electron beams with energies in the range from a few tenths to one MeV are often used in radiation processing of materials. The information that is of basic importance in processing is the knowledge of the fraction of the beam energy absorbed in a specific layer of the material. Usually in these applications, the electron beam is directed normally to the surface of the material, and the layer of primary interest is the outermost, or surface layer, with a thickness of the order of tenths of a millimeter. The layer thickness in most cases is less than the range of the incident electrons. Electron transport through such a surface layer is complex, as is consequently the energy loss by the electrons because of the significant probability of electron backscattering from the layer, as well as transmission through it. Therefore, it is desirable to devise a method by which the energy absorbed in surface layers under these conditions can be easily measured. In this paper, a direct measurement of the fraction of electron beam energy absorbed in a thin silicon layer is described for incident beam energies of 0.25, 0.50, 0.75, and 1.0 MeV.

It is possible to calculate the dose deposited in a surface layer if one knows the fraction of beam energy absorbed per incident electron, F_a , in the layer and the cross sectional area, A , in cm^2 , of the beam at the surface of the absorbed material. The absorbed dose per unit time, D_a , in rads per second in a surface layer of thickness, t , in cm, can be calculated from

$$D_a = 10^{11} \frac{IF_a T_0}{\rho A t}$$

where I is the beam current in amperes, T_0 is the energy of the incident electrons in MeV, and ρ is the density of the material in g/cm^3 . This formula assumes that the electron beam is uniform over the area exposed. It further requires the dimensions associated with the exposed area be much greater than t .

The measurements reported here are on the fraction of beam energy absorbed in a specific layer of silicon (a) when it was exposed alone or bare and (b) when it was a part of a semi-infinite medium, that is, a surface layer. There is a significant difference in the amount of energy absorbed in the two cases, a lesser amount of energy being absorbed in the bare layer than in the layer as a part of a semi-infinite medium. These results, therefore, illuminate to some degree the effect of boundaries on energy absorption in a surface layer.

EXPERIMENTAL PROCEDURE

The deposition of energy in a thin surface layer of silicon was investigated by the use of a silicon semiconductor, transmission detector as the silicon layer. The detector was a surface barrier type, and had a thickness of 0.196 mm and an area of 200 mm^2 . The entire volume of the detector was effectively active since negligible layers of about 40 $\mu\text{g/cm}^2$ of gold and aluminum were on its surfaces. The absorption of energy in the surface layer as a result of the interaction of an electron with the detector was recorded as a pulse. The pulse was formed upon the collection of hole-electron pairs produced in the detector, and its magnitude, or height, was assumed to be linearly proportional to the energy absorbed.

A beam of approximately 1000 electrons/sec was incident on the medium. In order to obtain such a low intensity source, a primary electron beam of about 10^{-8} amperes from a d.c. accelerator was scattered from a thin (about $30 \mu\text{g}/\text{cm}^2$) gold foil, and some of the scattered electrons, at 30° with respect to the primary beam, were collimated and directed onto the detector and medium. Figure 1 is a diagram of this arrangement. An angle in the forward direction was selected to minimize the fraction of the inelastically scattered electrons in the beam. For these conditions, about 99 percent of the beam at 30° consists of elastically scattered electrons.^[1] The scattering of the electron beam and its subsequent interaction with the medium occurred under high vacuum in a scattering chamber which has been described elsewhere.^[2]

The pulses from the detector were formed and handled using conventional electronic components, namely a charge-sensitive preamplifier, a main amplifier, and a pulse-height analyzer. A pulser was used to monitor the gain of the system and check its linearity, which was found to be better than one percent. The detector was cooled to -40°C during the measurements. At that temperature, the energy resolution of the detector with its associated electronics was 13.0 keV full width at half maximum, which was an improvement in resolution of about 30 percent over measurements at room temperature. The gains of the amplifiers employed were such that each channel of the pulse-height analyzer corresponded to 2.16 keV. The efficiency of the 0.196-thick detector is nearly 100 percent over the energy range of the measurements.^[1]

A dead time correction was unnecessary, since the beam intensity was relatively low. Therefore, each pulse was assumed to correspond to the interaction of an incident electron.

The measurements were made with the detector bare and with the detector in close contact (a separation of about 0.25 mm) to an aluminum backing, as shown in Figure 2. In the latter case, the silicon detector becomes a surface layer in a semi-infinite homogeneous medium. This assumption is only approximately correct since the amount of energy reflected back into the surface layer depends on the atomic number, Z , of the backing. The difference between using aluminum ($Z = 13$) and silicon ($Z = 14$) is small, and is approximately equal to the ratio of their atomic numbers multiplied by the quantity of energy reflected back into the layer. At most in these measurements, the result obtained employing an aluminum backing is estimated to be about 2 percent lower than that which would have been obtained with a silicon backing.

RESULTS

The absorbed-energy distributions for the 0.196-mm silicon layer are shown in Figures 3a through 3d, respectively, for the incident electron energies of 0.25, 0.50, 0.75, and 1.0 MeV on the layer. These distributions were derived from pulse-height distributions each of which contained approximately 5×10^5 pulses. The curves are plots of the response function $R(T_0, h)$ versus the relative pulse height h/h_0 . The response function is defined as the probability per unit pulse height that an incident electron on the detector with energy T_0 will

give rise to a pulse height between h and $h+dh$, and it is presented in units of number/incident electron-MeV. The abscissa in each plot can be converted into an energy scale by multiplying the relative pulse heights, h/h_0 , by T_0 .

The solid lines in the figures are smooth curves that were drawn through the experimental values (solid points) which are for the surface layer as a part of a semi-infinite homogeneous medium, and the dashed curves show the shape of the absorbed energy distribution obtained for the bare detector. A total absorption peak is evident in the distribution for both $T_0 = 0.25$ and 0.50 MeV, but does not appear in the distribution for either $T_0 = 0.75$ or 1.0 MeV. At low relative pulse heights for each distribution there is a somewhat broad peak which is associated with transmission of electrons through the layer with minimum energy loss. The effect of the addition of the 10-mm-thick aluminum backing to the detector is seen to be a shift of the absorbed-energy distribution to higher relative pulse heights with the exception that the magnitude of the total absorption peak remains unchanged. The distribution shift is accompanied by a reduction in the magnitude of the low relative pulse-height peak, indicating that some of the electrons, which would have been transmitted with minimum energy loss in the bare case, are directed back into the surface layer.

The energy absorbed per incident electron in the silicon layer is acquired by integration of the absorbed energy distributions; this value is then divided by T_0 giving F_a , the fraction of beam energy absorbed per incident electron. The values of F_a for the surface

layer are higher than the values of F_a for the bare layer at each incident electron energy. The experimentally determined values of F_a are given in the following table, along with some corresponding F_a values which were calculated by a Monte Carlo method.^[3]

Fraction of Beam Energy Absorbed per Incident Electron, F_a ,
in a 0.196-mm Silicon Layer

		F_a			
		T_0 (MeV)	0.25	0.50	0.75 1.00
A. Layer bare					
	Calculated		0.840	0.244	0.113 0.073
	Measured		0.760	0.224	0.104 0.068
B. Surface Layer					
	Calculated				0.156 0.101
				(0.294)*	(0.144)* (0.094)*
	Measured		0.772	0.280	0.140 0.090
C. Ratio of F_a for surface layer to F_a for bare layer					
	measured F_a		1.02	1.25	1.35 1.32
	values.				

* These calculated values were for an equivalent surface layer of aluminum. They were obtained from depth-dose distributions in aluminum by just taking into account the difference in densities, $\rho(\text{Al}) = 2.70 \text{ g/cm}^3$ and $\rho(\text{Si}) = 2.33 \text{ g/cm}^3$.

There are differences between the measured and calculated results for F_a . In the case of the bare silicon layer, the measured values are from 7 to 10 percent lower than the calculated ones, and similarly in the case of the surface layer, the measured values are 10 percent lower than the calculated values at 0.75 and 1.0 MeV for a silicon homogeneous medium and from 2 to 5 percent lower than the calculated values at 0.5, 0.75, and 1.0 MeV for an equivalent layer in an aluminum homogeneous medium. The calculated values of F_a for a silicon surface layer are about 7 percent higher than for an equivalent aluminum layer; this difference can be accounted for by the higher electron stopping power (about 3 percent)^[4] and the higher backscatter into the layer (about 2 percent) of silicon over aluminum. Therefore, it is concluded that the difference between the measured and calculated values of F_a are about the same for both the bare layer and the surface layer. There is no clear explanation of why a discrepancy exists between calculations and measurements. These results, however, are consistent with those of response functions for bare silicon detectors previously reported^[1] in which the measurements indicated more transmission of energy than the calculations.

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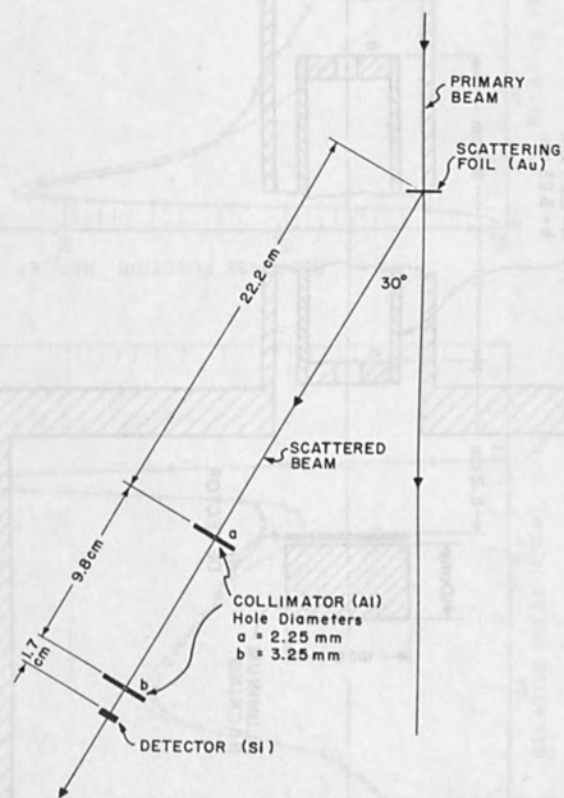


Fig. 1. Experimental arrangement of scattering foil, collimator, and silicon detector.

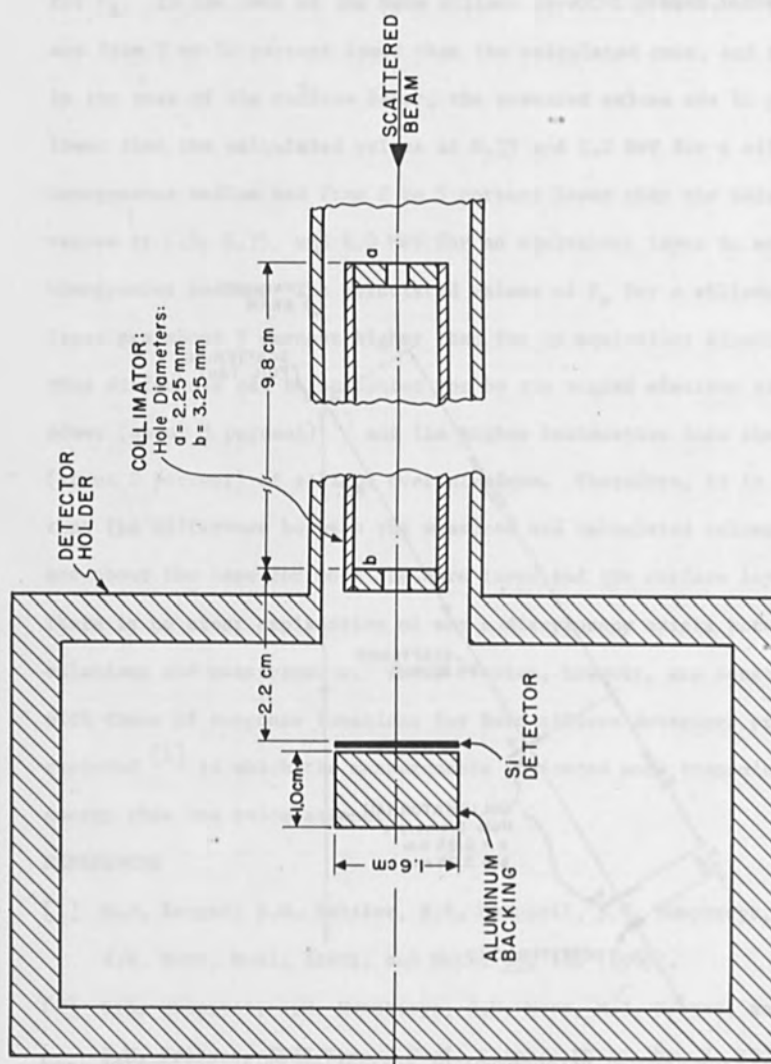
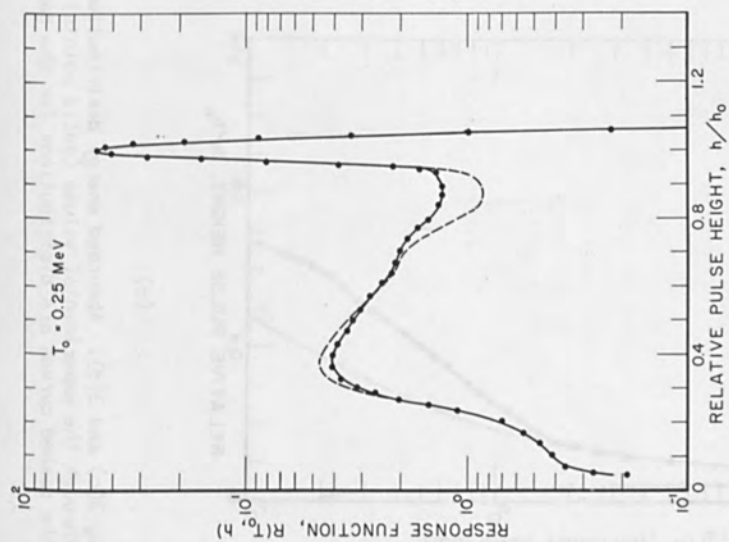
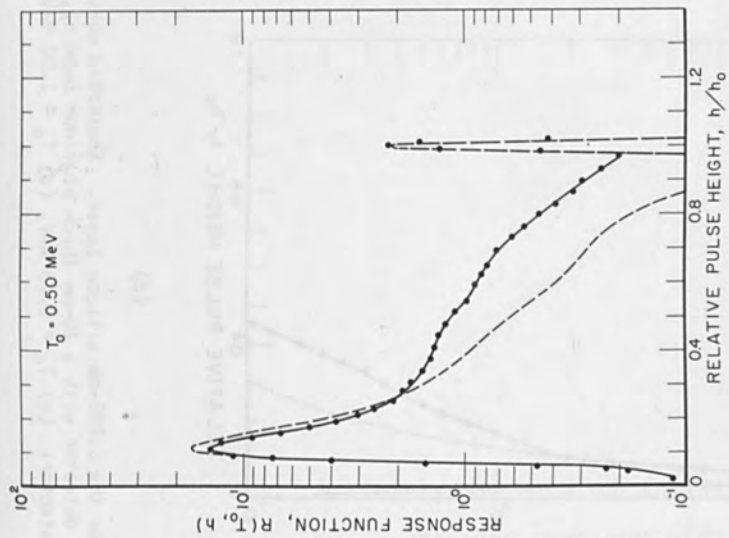


Fig. 2. Silicon detector with a 10-mm thick aluminum backing simulating a surface layer as a part of a homogeneous medium.



(a)



(b)

Fig. 3(a) and 3(b). Absorbed energy distributions for the 0.196-mm silicon layer. The solid curves are through the experimental values (solid points) for detector with a 10-mm thick aluminum backing, and the dashed curves are distributions for the bare detector: (a) $T_0 = 0.25 \text{ MeV}$, (b) $T_0 = 0.50 \text{ MeV}$.

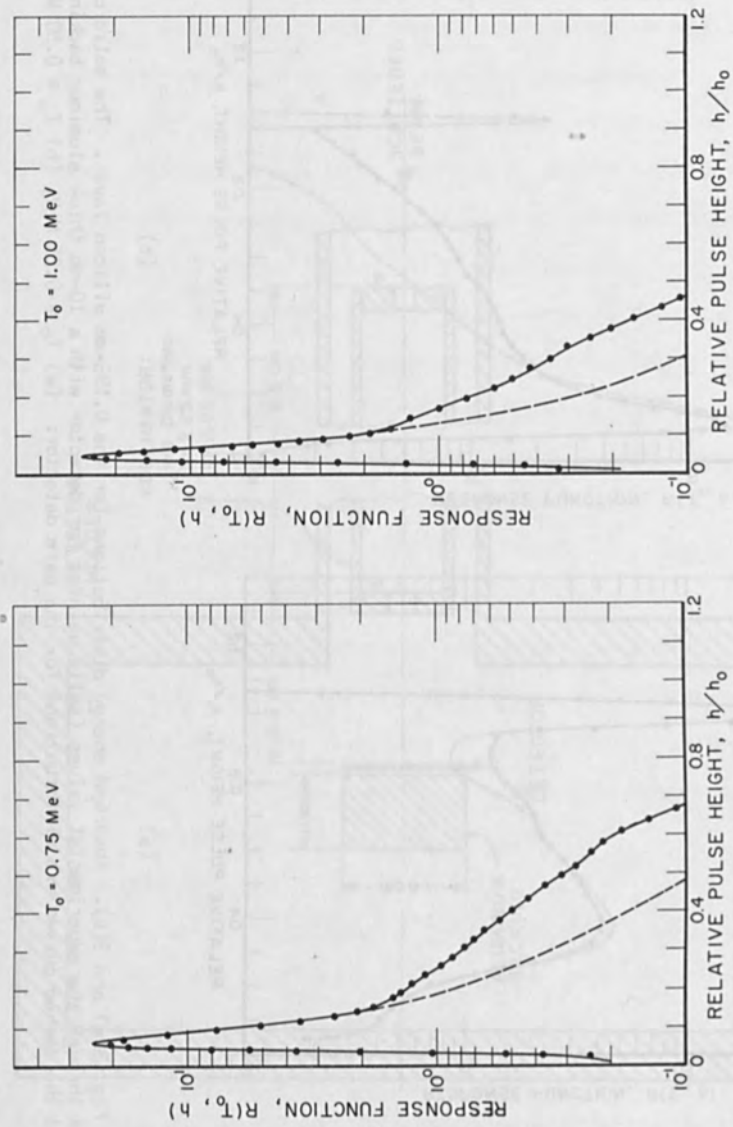


Fig 3(c) and 3(d). Absorbed energy distributions for the 0.196-mm silicon layer. The solid curves are through the experimental values (solid points) for detector with a 10-mm thick aluminum backing, and the dashed curves are distributions for the bare detector: (c) $T_0 = 0.75$ MeV, (d) $T_0 = 1.00$ MeV.

