## Free-standing filaments in thermal nanoimprint induced by pre-filling

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Nanoimprint lithography (NIL) is a simple mechanical low cost technique to manufacture devices with nanometer-sized structures. Although a variety of processes have been successfully developed, some basic questions still remain obscure, as e.g. the process of 3D cavity filling under the combined action of external forces (applied pressure, squeezed flow) and capillary forces (surface tension). At first view high viscosity is associated with high pressure (as is the case with thermal NIL), whereas low viscosity is associated with capillary forces (as is the case with UV-NIL and e.g. used in capillary force lithography).

This simplified view has already been revised by a recent study [1] on the filling of 3D-microcavities [2] with thermal NIL of PMMA at 40-70°C above glass temperature and a pressure as low as 0.05 MPa, the contact pressure being generally used during temperature ramp-up to assure isothermal conditions during imprint. In spite of the high viscosity of the polymer, 'pre-filling' states were observed in partly filled cavities, featuring contact angles towards the cavity sidewalls and pinning at the edges of the stamp protrusions.

Our present study refers to similar structure dimensions as in [1], Fig. 1, but surrounded by a wide recessed area. We used about 1µm thick PMMA (140 kg/mol) and PS (350 kg/mol), imprinted at 250°C and 190°C, respectively, with different tools (Jenoptik HEX 03 and a proprietary lab system [3]) and a pressure of about 0.1 MPa. At this low pressure no full area contact between stamp and polymer surface exists and local gaps remain due to bowing or warping. Results obtained with PS in regions with a relatively high gap between the stamp and the substrate indicate a strong physical self-assembly within the cavity (Fig. 2). The polymer has almost completely de-wetted the substrate, yet making contact with the elevated stamp structures locally. Starting from local contact points the polymer has entered the 3D cavities horizontally along the line-like cavities. Consequently, the polymer forms free-standing filaments, bridging neighboring contact points (Fig. 2).

We will explain the formation of these structures under the action of surface forces and capillary forces, the role of the inner (i) and outer (o) edges as well as their angles, both phenomenologically as well as theoretically [4,5]. Further we will discuss the impact of equipment and imprint material in this respect. Dedicated stamp design for the use of such effects, e.g. with combined micro-and nanostructures, may provide the basis for novel devices.

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Fig 2: Pre-fill situation in a region with a relatively high gap between the stamp and the substrate. Top: Micrograph of the stamp, center: Structures obtained in PS (p = 1 bar, T = 190°C), bottom: Detail.

A strong correlation between stamp structures and polymer structures is found. Free-standing filaments bridge neighboring contact points. Fig. 1: Top: Schematics of the imprint situation with 3D stamp, bottom: Full replication in PMMA showing details of the stamp cavities.

The structures are linear, with an inclined and a steep sidewall each. The cavities are 500 nm in height. Single fields of lines  $(20 \ \mu m \ x \ 20 \ \mu m$ , compare to Fig. 2, top) are arranged side by side, within a wide cavity in the center of a 100 mm wafersize stamp.



Fig.3: Conceptual ideas to explain the course of filling of the 3D stamp cavities. (a,b cross-section; c side view). a) First, physical self assembly (PSA) due to van der Waals interaction results in local contact of the polymer to the stamp (capillary bridges).

b) Then the polymer enters the cavity via the 'quicker' outer edge at the inclined sidewall.

c) When the inner edge is reached, the polymer spreads laterally.

Finally, filaments develop along the 90° inner edges.

Suction of the polymer into the stamp cavities results in a complete dewetting of the substrate so that free-standing filaments are formed.