The Effects of Thin Films and Confinement on Thermal Nanoimprint Lithography Patterning

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Nanoimprint lithography (NIL) is a next generation lithography (NGL) with the potential to realize many emerging technologies with smaller scales compared with conventional lithography. One of the primary differences from conventional lithography is that NIL utilizes a mechanical squeeze-flow mechanism to force the resist material to fill the mold. This requires inducing polymer flow in thin films and into nanoscale spaces. It is well known from the polymer thin film literature that many of the fundamental properties of polymeric materials begin to deviate from their bulk values when the film thickness drops below approximately 50 nm. These are precisely the same length scales that are relevant for NIL. In fact, there are many reports of how viscoelasticity, diffusion, and other transport properties slow down in thin polymer films. These results suggest that NIL mold filling process become more difficult in thin polymer films and nanoscale mold cavities. In this presentation we present evidence in support of this notion.

In the first part of the presentation, we demonstrate that large levels of residual stress can be imparted to patterns created by NIL. These residual stresses originate from the large shear strains and deformation fields that are imparted upon the viscous polymer melts. These deformation fields often do not have time to relax during the imprint, locking residual stress into the pattern. The level of residual stress can be controlled, however, through the rheology of the imprint process. Time, temperature and shear rate variations that reduce the viscosity of the polymer during the imprint process are effective ways to minimize these stresses. However, in the second part of the talk we show that the viscous flow processes can be significantly retarded in thin polymer films, making it more difficult to induce flow into the mold. We show quantitative evidence that polymer flow is impeded when the resist film thickness drops below 50 nm. In the last portion of the presentation we provide evidence, as seen by inelastic neutron scattering, for the reduced molecular mobility in these thin polymer films. We quantify a strong reduction in the thermally induced mean square atomic fluctuations of atoms in the polymer chain and relate this reduced molecular motion to the impeded polymer flow. We will also show, however, under certain circumstances this reduced mobility can have a positive effect in stabilizing nanoscale patterns against residual stresses and mechanical instabilities.



Figure 1. (a) Specular X-ray reflectivity is used to quantify pattern height and residual layer thickness for a series of line-space patterns imprinted into polystyrene films where the initial film thickness is varied. When the initial film thickness is insufficient for complete mold filling, a finite residual layer thickness of approximately 10 nm is still observed. (b) In this figure the evolution of the full pattern heights, taking into account both the incomplete and complete mold filling regimes, are shown as a function of temperature. The pattern heights have been normalized to one at their highest imprint temperatures for each film thickness. In this representation one can clearly see the evidence for reduced polymer flow at low temperatures in the films with the thinner initial thickness; the film must be heated to higher temperature to induce complete filling. (c) This figure shows a strong reduction in the mean-square displacement $\langle u^2 \rangle$, i.e., the thermal fluctuations of the atoms in the polymer, evaluated in inelastic neutron scattering measurement.¹ These reduce polymer dynamics with decreasing film thickness are quantitatively consistent with an increase in the effective viscosity of the thin polymer film.

¹ R. Inoue, T. Kanaya, K. Nishida, I. Tsukushi, M. T. F. Telling, B. J. Gabrys, M. Tyagi, C. Soles, W.-l. Wu, *Phys. Rev. E.* **80**, 031802 (2009)