PERSPECTIVE

Roll in and Roll out: A Path to High-Throughput Nanoimprint Lithography

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ABSTRACT Roller-type nanoimprint lithography is a technique that can potentially increase the throughput of the imprinting process to levels competitive for low-cost, large-area, nanoscale manufacturing. A paper in this issue by Ahn and Guo presents a large-area roll-to-roll and roll-to-plate imprint process that builds on earlier work, increasing the substrate width to 4 in. and transferring patterns of 300 nm width and 600 nm pitch at a rate of 1 m/min.

See the accompanying Article by Ahn and Guo.

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Nanoimprint lithography (NIL) is a patterning technique that is well-positioned to take a key role in manufacturing of nanoscale structures for electronic, optical, biological, and energy applications. It is a process based on mechanical deformation of a material to the shape of a mold and is thus not hindered by optical diffraction limits while retaining the relatively high throughput of a parallel process. However, the fact remains that planar imprint involves a cycle time that may be unacceptable for high-throughput industrial manufacturing, due to the time required for the imprint resist to fill all the cavities of the mold and the subsequent release of the mold from the substrate. To address this issue, a number of groups have taken inspiration from rotogravure—a process wherein an image is rapidly engraved from copper cylinders to paper reels—and have designed tools to pattern substrates using a cylindrical mold. The molding and demolding processes run continuously in such configurations, dramatically increasing the throughput. Moreover, the force required for release is also reduced compared to a full-wafer planar imprint due to the peeling action of the mold rotating away from the substrate.

In this issue, Ahn and Guo report imprinting 4 in. wide flexible and rigid substrates using roll-to-roll (R2RNIL) and roll-to-plate (R2PNIL) imprint processes, respectively.[1] Their apparatus consists of a series of rollers to coat the substrate with resist, to imprint the resist, and to hold the belt under tension, as shown schematically in Figure 1. A high-intensity UV source rapidly cures the resist, enabling impressive belt speeds of 1 m/min. Transfer of grating patterns with 700 nm period and 300 nm line width was demonstrated, and they have previously shown replication of 200 nm period, 70 nm line width gratings on 10 mm wide substrates using a similar process.[2]

Fabrication of the Mold. As with any imprinting process, the mold is of paramount importance. In the case of roller-type imprints, there is an additional challenge introduced by the necessity of fabricating nanoscale patterns on a cylindrical template. (Technically speaking, a flat mold could be exchanged with the substrate in the R2PNIL configuration, but this would make continuous imprinting impractical.) One option is to fabricate the patterns directly on the cylinder, either by machining or by lithographic/plating processes.[3,4] Although such molds are very robust, the fabrication process is more complex and the pattern size is in the micrometer regime. Another option is to wrap a flexible mold around a bare cylinder. In practice, such molds have generally been made from a thin nickel shim or a polymer. Nickel molds have some attractive properties; they are not as brittle as silicon or quartz and can be readily replicated from one another. They can be made with high resolution, but the process becomes more difficult at sub-100 nm dimensions than most polymer casting methods.

For many years, polydimethylsiloxane (PDMS) was used as the material for soft lithography molds due to its low Young's modulus, low surface energy, commercial availability, and ease of replication. However, its flexible nature also makes it impossible to replicate nanopatterns with any useful aspect ratio, due to collapse of the features under imprinting pressure or under their own weight. Some groups have experimented with hybrid molds consisting of a patterned, stiff polymer film on top of a conventional PDMS substrate.[5,6] With the right materials and process, very high resolution patterns can be attained.[7] Depending on the bending radius required, it is possible to employ other polymer compositions that are stiff enough to replicate nanostructures stably but still flexible enough to wrap around the cylinder. Ahn and Guo selected ethylene tetrafluoroethylene (ETFE) for this property, as well as its exceptionally low surface energy (15.6 dyn/cm). The advantage of such polymer molds compared to nickel are their simple replication using NIL or casting methods; however, it will be important to evaluate their long-term durability for use in potential manufacturing processes.

Choosing the Imprint Resist. The imprint resist used in the R2RNIL process is of equal importance to the mold. In order to exploit the higher throughput afforded by the roller setup, the resist should be low-viscosity and fast-curing to enable high-speed imprint processing. At the same time, it must adhere well to the substrate, be mechanically robust, and have good etch resistance for postprocessing. Such resists generally fall into the UV-cross-linkable category, although some fast thermal-curable formulations have been presented.[8] Acrylate-based mixtures are the most commonly used for UV-NIL resists, but due to the free radical polymerization mechanism used, the curing process is impeded by the presence of oxygen. A different curing mechanism is desirable in R2RNIL to avoid costly vacuum or gas-purging. Kim *et al.* reported on the use of various vinyl ethers for use in step-and-flash imprint lithography (S-FIL), which are cross-linked by cationic polymerization and are thus not susceptible to oxygen scavenging.[9] In the same vein, Cheng *et al.* developed epoxysilicone monomers that can be cross-linked by cationic polymerization.[10] This nanoimprint resist was also chosen for use in the R2RNIL process by Ahn and Guo due to its low viscosity, low shrinkage (<3%), sensitivity to UV light, and good etching resistance.

During the imprint process, a residual layer of resist will almost always remain due to the incompressibility of the liquid precursor. The

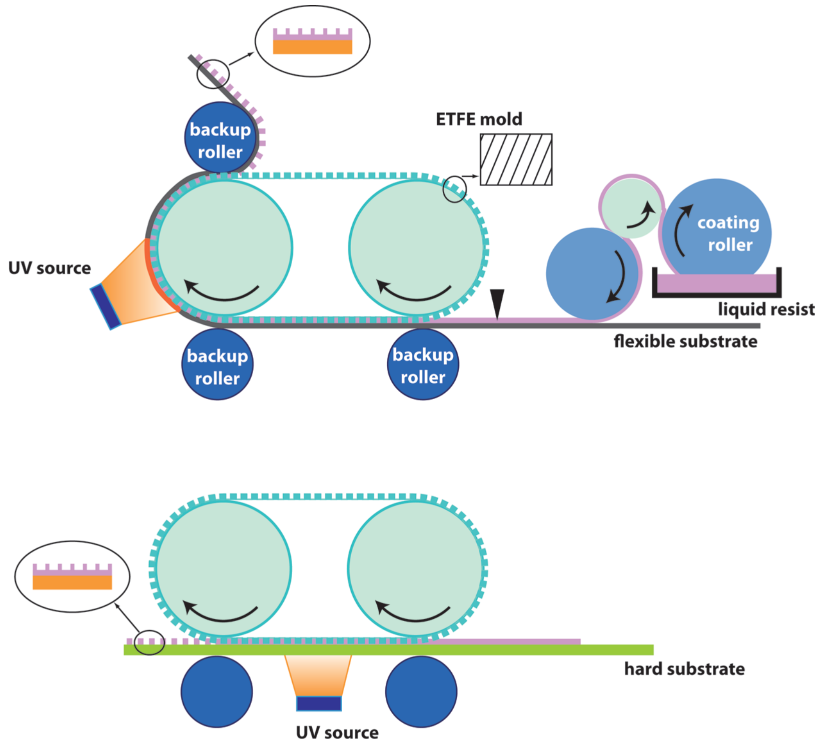

Figure 1. Schematics of (a) R2RNIL and (b) R2PNIL processes. Adapted with permission from ref 1. Copyright 2009 American Chemical Society.

residual layer thickness (RLT) should be minimized if subsequent reactive ion etching (RIE) of the layer will be performed, in order to reduce lateral enlargement of features. Studies of the RLT as a function of mold geometry, initial resist thickness, resist viscosity, and applied force have been performed for standard planar UV-NIL processes;[11] however, roller-type imprinting requires different analyses due to different dynamics, where the rotation of the rollers and the contact length between the mold and the resist comes into play. Ahn and Guo derived three different possible models to predict the RLT for the R2RNIL process and ultimately found the dynamic elastic roller contact model to have the best fit to experimental data. One remaining question is whether the model will remain valid for RLT values that are small compared to the height and width of the mold features, which is a more important scenario for the transfer of the pattern to the substrate *via* RIE.

THE FUTURE OF NANOIMPRINT LITHOGRAPHY

Nanoimprint lithography offers a tantalizing technique for replication of nanoscale structures using cost-effective tools. However, to mature into a true manufacturing process, the throughput must be increased further and the defect density reduced. Roller-type NIL processes such as R2RNIL and R2PNIL offer a way to do both. Ahn and Guo

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have demonstrated a solution that considers all of the aspects of the procedure, including the imprinting apparatus, flexible mold, resist, and RLT modeling. With continuing improvements, one can hope such techniques will become as ubiquitous as their inspiration—the rotogravure.

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