

## Review

### Toward residual-layer-free nanoimprint lithography in large-area fabrication

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(Received December 23, 2013; final revision received January 27, 2014; accepted January 29, 2014)

In the paper, residual-layer-free nanoimprint lithography for large-area fabrication is reviewed. In order to remove the residual layer during the imprint process, polymer resists and mold materials should be designed with the aspects of surface chemistry and mold geometries in mind. Various approaches for residual-layer-free nanoimprint lithography are discussed including incomplete filling by polymer mass, reverse imprint methods, self-removal techniques, and the employment of elastomeric mold deformation. In addition, issues that must be overcome to enable large-area roll-to-roll nanoimprinting without a residual layer are presented.

**Keywords:** nanoimprint, residual layer, polymer, mold, roll-to-roll process

#### 1. Introduction

For decades, polymer processing techniques such as extrusion and injection molding have been used as conventional methods to make macro-scale shapes like pipes, films, trash boxes, bottles, and frames using polymer materials. In the 1990s, nanoimprint lithography was invented and currently shows great potential as a low-cost and high-resolution process for manufacturing devices such as semiconductor devices, optoelectronic devices, and energy devices (Bessonov *et al.*, 2011; Bogdanski *et al.*, 2005; Cheng *et al.*, 2002; Cheng *et al.*, 2004; Cheng *et al.*, 2006; Gates *et al.*, 2005; Hong *et al.*, 2006; Hu *et al.*, 2005; Hwang *et al.*, 2010; Kam *et al.*, 2004; Kang *et al.*, 2010; Kao *et al.*, 2006; Kiyohara *et al.*, 2005; Leising *et al.*, 2006; Liao *et al.*, 2004; Mele *et al.*, 2005; Pisignano *et al.*, 2004; Rogers *et al.*, 1998; Rogers *et al.*, 2009; Rolland *et al.*, 2004; Yoon *et al.*, 2011; Zaumseil *et al.*, 2003). This manufacturing procedure is similar to the macro-scale polymer processing technique, injection molding. A thin polymer film coated onto a substrate is pressed by a nanoscale mold with heat and pressure. After cooling to room temperature, polymeric nanostructures fabricated on substrates such as silicon wafers are obtained (Austin *et al.*, 2002; Bailey *et al.*, 2000; Chou *et al.*, 1996; Chou *et al.*, 1997; Coulburn *et al.*, 1999; Kim *et al.*, 2006). The polymer then acts as a resist for subsequent etching processes of an active layer beneath the resist pattern. Recently, this technique has been applied for fabricating three dimensional and multi-scale structures.

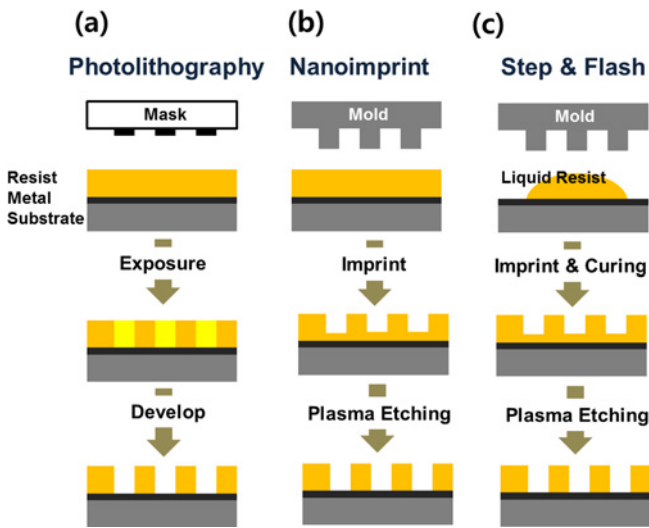
(Almanza-Workman *et al.*, 2011; Bao *et al.*, 2002; Kim *et al.*, 2001; Kim *et al.*, 2002) Rapid development of flat panel displays such as liquid crystal displays (LCDs) and organic light emitting diodes (OLEDs) makes imprint lithography an important technology for large-area fabrication as a potential technique to replace expensive photolithographic technologies. Although nanoimprint lithography has been developed at a commercial level, it still has inherent issues that photolithography does not. Regardless of the types of polymer resists, residual layers remain after the imprint process, which must be removed through a reactive-ion-etch (RIE) process. (Austin *et al.*, 2002; Chou *et al.*, 1996) Unfortunately, RIE is not compatible with large-area fabrication because it requires a vacuum system to generate plasma. Therefore, residual-layer-free imprint lithography is indispensable for large-area fabrication.

In this review, we discuss the fundamental principles for residual-layer-free nanoimprint lithography by examining polymer resists and molds. The conditions for a residual-layer-free process are also presented. By reviewing the various methods for removing residual layers, we provide an outlook for nanoimprint lithography without residual layers for large-area fabrication.

#### 2. Nanoimprint Technology

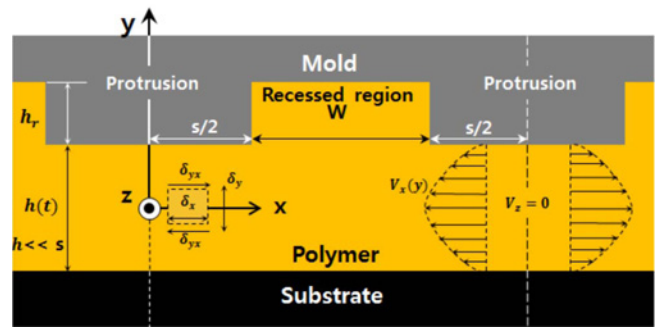
Nanoimprint lithography was invented to fabricate nanoscale patterns, which are difficult to produce using photolithography because of diffraction effects, interference, and scattering of optical energy sources. (Austin *et al.*, 2002; Bailey *et al.*, 2000; Chou *et al.*, 1996; Chou *et al.*, 1997) To maximize memory capacity, the semiconductor industry has attempted to identify alternative

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**Fig. 1.** (Color online) Schematic illustrations of (a) photolithography, (b) nanoimprint lithography, and (c) step-and-flash lithography. (Austin *et al.*, 2002; Bailey *et al.*, 2000; Chou *et al.*, 1996; Chou *et al.*, 1997; Coulburn *et al.*, 1999; Kim *et al.*, 2006).

technologies such as X-ray and e-beam lithography for decreasing the wavelength of the energy source. However, immense equipment cost and low throughput are hurdles preventing replacement of conventional photolithography. In response to this, Chou *et al.* pioneered the field of nanoimprint lithography (Austin *et al.*, 2002; Bailey *et al.*, 2000; Chou *et al.*, 1996; Chou *et al.*, 1997). Instead of writing a pattern by e-beam lithography, they made a very fine pattern using an e-beam and then used a mold to press a polymer resist film coated on a substrate. They were able to realize 25 nm line and dot patterns, which were regarded as impossible using photolithography at that time. Fig. 1 shows the procedure of nanoimprint lithography in comparison to photolithography. For photolithography, the energy from a light source transmitted through a mask cleaves or crosslinks (depending on resist type) a photosensitive polymer followed by a development process to remove the desired polymer pattern. On the other hand, nanoimprint lithography uses two kinds of materials, which are either thermoplastic or photocurable polymers. (Austin *et al.*, 2002; Chou *et al.*, 1996; Costner *et al.*, 2009; Guo *et al.*, 2007; Hong *et al.*, 2006; Hu *et al.*, 2005; Jeans *et al.*, 2010; Jung *et al.*, 2004; Kim *et al.*, 2006) After fabricating a hard master mold typically made of silicon or quartz, a thermoplastic polymer is pressed with heat and pressure and detached from the mold after cooling to room temperature (nanoimprint lithography). A photocurable liquid polymer is then placed in the voids of the mold followed by crosslinking by ultraviolet (UV) light. (Coulburn *et al.*, 1999; Kim *et al.*, 2006)



**Fig. 2.** (Color online) Squeeze Flow Model of Nanoimprint Lithography. (Schulz *et al.*, 2003; Shift *et al.*, 2003).

Nanoimprint technology typically produces a residual layer of resist between the mold and the substrate because it is difficult to bring the mold in direct contact with the substrate. Additionally, when both a mold and a substrate are made of inorganic materials such as silicon, they are susceptible to breakage during direct contact under high pressure (about tens of bar). (Yoon *et al.*, 2004) The residual layer must subsequently be removed by RIE under vacuum conditions, which is not suitable for large-area fabrication, especially for flat panel displays. In addition, it is possible that the edges of the polymer resists are rounded during the RIE process because of the low selectivity of the etchant. Therefore, RIE is not a convenient process to be used.

### 3. Theoretical nanoimprint model: squeeze flow model

Fig. 2 shows a schematic representation of the squeeze flow theory ((Schulz *et al.*, 2003; Shift *et al.*, 2003). In nanoimprint lithography, a mold is placed on the polymer resist coated on the substrate and applied with a constant force. The polymer resist is then heated to a temperature above the glass transition temperature ( $T_g$ ) and maintained. The viscous polymer then moves to the voids of the mold and contacts the recessed region of the mold. Then, it is cooled to room temperature and the external force is released. A simple model for the procedure is a system of a sandwich of two parallel plates with a polymer resist between them. The polymer is thinned between the protrusion regions of the mold (width of  $s$ ) and flows to the cavity (recessed region) of the mold. To solve the squeezing flow problem, it is assumed that the polymer melts are ideally viscous and incompressible fluids, and that the residual polymer thickness,  $h$ , should be negligible compared to the width of the protruding region,  $s$ . From a motion equation, Equation (1) is obtained (Schulz *et al.*, 2003; Shift *et al.*, 2003),

$$-\partial_x p + \nabla \cdot \sigma_{ij} = 0 \quad (1)$$

where  $p$  is the external pressure and  $s$  is a stress. The shear stress in the  $y$ -direction is

$$\sigma_{xy} = \frac{dp(x)}{dx} y. \quad (2)$$

When we consider the pressure as independent from  $y$ , Newton's friction law results in

$$\frac{dv_x}{dy} = \frac{1}{\eta_0} \sigma_{yx} = \frac{1}{\eta_0} \frac{dp}{dx} y. \quad (3)$$

After integration over  $y$ , the velocity field is obtained,

$$v_x(x, y, t) = \frac{1}{2\eta_0} \cdot \left\{ y^2 - \left( \frac{h(t)}{2} \right)^2 \right\} \cdot \frac{dp(x, t)}{dx}. \quad (4)$$

With boundary conditions, the volume flow  $q(x, t)$  is derived by an integration over  $y$ ,

$$\begin{aligned} q(y, t) &= 2L \cdot \int_0^{h(t)/2} v_x(x, y, t) dy = \dots \\ &= -\frac{L}{12\eta_0} \cdot h^3(t) \cdot \frac{dp(x, t)}{dx} = -Lx \cdot \frac{dh(t)}{dt}. \end{aligned} \quad (5)$$

The hydrostatic pressure is given by integrating over  $y$  as a function of polymer thickness  $h(t)$

$$p(x, t) = \frac{6\eta_0}{h^3(t)} \cdot \frac{dh(t)}{dt} \cdot \left\{ x^2 - \left( \frac{s}{2} \right)^2 \right\}. \quad (6)$$

After integration over the width of the protruding region, the exerted force is obtained as,

$$F = L \cdot \int_{-s/2}^{s/2} p(x, t) dx = -L\eta_0 \cdot \left\{ \frac{s}{h(t)} \right\}^3. \quad (7)$$

From Equation (7), the polymer thickness is expressed as a function of time when the force,  $F$ , is constant

$$\frac{1}{h^2(t)} = \frac{2}{h_0^2} + \frac{2F}{\eta_0 L s^3} \cdot t \quad (8)$$

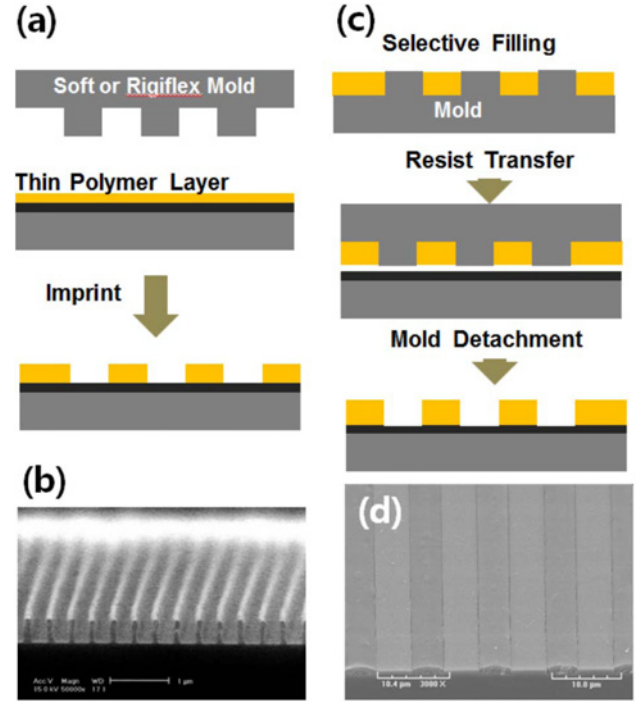
where  $\eta_0$  is the viscosity of the polymer material. Finally, the process time expression is obtained for case where the final polymer thickness is  $h_f$

$$t_f = \frac{\eta_0 s^2}{2F} \left( \frac{1}{h_f^2} - \frac{1}{h_0^2} \right). \quad (9)$$

Equation (9) shows that infinite time is required for achieving an  $h_f$  of zero. Therefore, the squeezing theory may be not suitable for describing residual-layer-free nanoimprint lithography.

#### 4. Residual-layer-free nanoimprint lithography

To remove the residual layer without an RIE process, many techniques have been attempted including UV-curing of polymer resin followed by an additional chemical



**Fig. 3.** (Color online) (a) A schematic showing residual-layer-free nanoimprint with flexible molds. (b) A scanning electron microscopic (SEM) image of polymer patterns without residual layers. (c) A schematic for reversal nanoimprint to remove the residual layer. (d) A SEM image filled selectively in recessed regions of the mold. (Park *et al.*, 2009; Yoon *et al.*, 2004).

development process. (Auner *et al.*, 2009; Auner *et al.*, 2010; Jackman *et al.*, 1998; Jung *et al.*, 2012; Kao *et al.*, 2011; Kim *et al.*, 2003; Kim *et al.*, 2006; Lee *et al.*, 2005; Park *et al.*, 2009; Yang *et al.*, 2009; Yoon *et al.*, 2004; Yoon *et al.*, 2010; Yoon *et al.*, 2011; Zaumseil *et al.*, 2003) In this section, we classify previous attempts into two types of methods as shown in Fig. 3: (1) Using flexible molds to directly contact the substrate in order to eliminate residual layers (Fig. 3 (a, b)), and (2) selective filling into the recessed areas of the mold followed by transferring the polymer patterns to a substrate (Fig. 3 (c, d)). A prerequisite for removing the residual layer is, as shown in Fig. 3 (a), that the polymer thickness should be small enough to fill the voids incompletely. (Yoon *et al.*, 2011) Suh *et al.* defined patterns using capillary forces without external pressure for both thin and thick polymer films. (Suh *et al.*, 2001) After controlling the polymer film thickness, there have been many reports demonstrating polymer patterns without residual layers. Fig. 3 (b) shows an SEM image of high aspect ratio polymer patterns without residues. It should be noted that the polymer patterns may be produced with an elastomeric mold (polydimethylsiloxane (PDMS)) or a rigiflex mold (polyurethane acrylate (PUA), perfluoropolyether (PFPE)), which is a

rigid mold fabricated on a flexible substrate. (Choi *et al.*, 2004; Kim *et al.*, 1995; Kim *et al.*, 1997; Rogers *et al.*, 2005; Suh *et al.*, 2005; Williams *et al.*, 2010; Xia *et al.*, 1995; Xia *et al.*, 1998; Xia *et al.*, 1999) Flexible molds—(PDMS) or rigiflex—have advantages in avoiding breakage during direct contact between the molds and substrates and only require low pressure for conformal contact, even in large-area fabrications. (Yoon *et al.*, 2004) Reversal imprint lithography, which includes selectively filling polymer resins into the recessed regions of a mold, is another technology for nanoimprint lithography without a residual layer. (Kao *et al.*, 2011; Kim *et al.*, 2003; Kim *et al.*, 2007; Lee *et al.*, 2005; Park *et al.*, 2009; Yang *et al.*, 2009) This was attempted by heating the polymers to induce shrinking and dewetting of the resist from the protruded regions of the mold, forcing it into the recessed regions or alternatively by using a doctor blade method to remove the excess polymer from the protruded regions. (Kao *et al.*, 2011; Kim *et al.*, 2003; Kim *et al.*, 2007; Lee *et al.*, 2005; Park *et al.*, 2009; Yang *et al.*, 2009) In addition, reverse-tapered molds were utilized to retain the polymer films in the cavity or to control the surface energy of the molds in order to fill the resin selectively. (Kim *et al.*, 2007) Based on these aspects, we conclude that the surface chemistry of polymers and molds for the dewetting process as well as the geometry of the molds are important for residual-layer-free nanoimprint lithography.

## 5. Design parameters for residual-layer-free nanoimprint

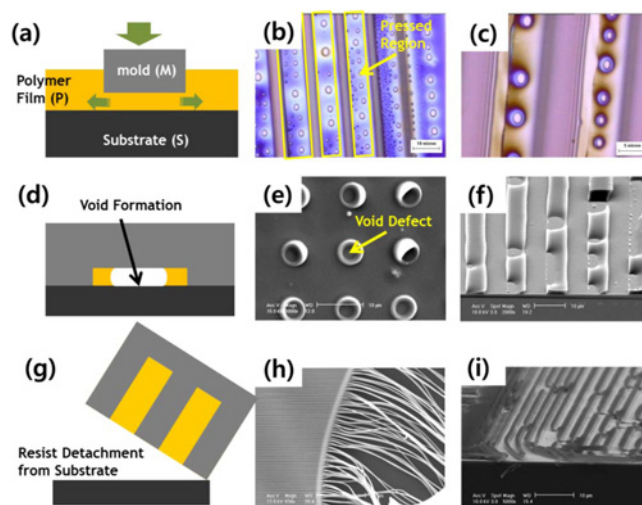
A prerequisite condition for residual-layer-free nanoimprint lithography is that the amount of polymer resin coated on a substrate should be smaller than the void volume of the mold. (Yoon *et al.*, 2010; Yoon *et al.*, 2011) However, even with this condition met, based on the squeezing model, it takes infinite time to decrease the final polymer thickness,  $h_f$ , to zero. One possible solution is dewetting-induced removal of the residual layer. As shown in Fig. 4(a), when a polymer is sandwiched between a mold and a substrate with a thickness,  $h$ , the driving force,  $p$ , for dewetting is (Israelachvili, 1992)

$$p = \frac{A_{\text{eff}}}{6\pi h^3} - \gamma \frac{\partial^2 h}{\partial x^2} \quad (10)$$

where  $A_{\text{eff}}$  is the effective Hamaker constant and  $\gamma$  is the surface tension of the polymer. In a mold/polymer/substrate system,  $A_{\text{eff}}$  is derived as (Israelachvili, 1992)

$$A_{\text{eff}} = (\sqrt{A_{\text{mold}}} - \sqrt{A_{\text{polymer}}})(\sqrt{A_{\text{substrate}}} - \sqrt{A_{\text{polymer}}}). \quad (11)$$

The equation above indicates that  $A_{\text{eff}}$  should be positive for dewetting of polymer films resulting in the removal of the residual polymer layer. Fig. 4(b) and 4(c) show micro-



**Fig. 4.** (Color online) (a) A simplified schematic for the squeezing system. (b) A microscopic image of hole formation in polymer film during squeezing of the film with pressure and heat. (c) A magnified image of (b). (d) A schematic showing unwanted void formation by dewetting of polymer films. Scanning electron microscopy (SEM) images of (e) disk patterns with holes in the center region and (f) line patterns. (g) A schematic of detachment of a polymer resist from a substrate. (h, i) SEM images of detached polymer patterns. (Yoon *et al.*, 2011).

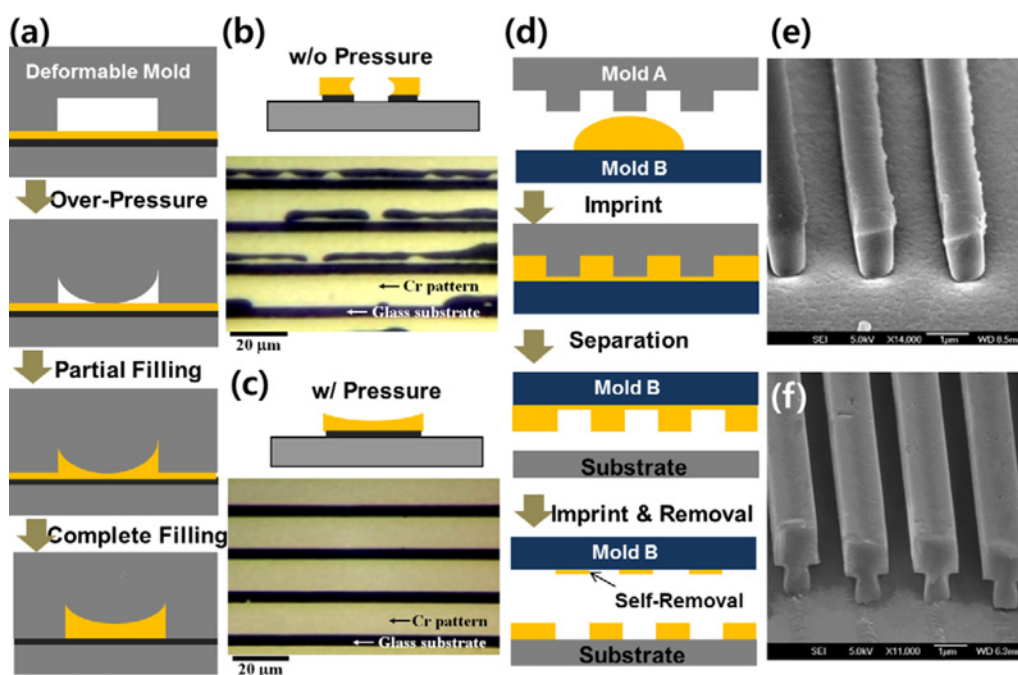
scopic images demonstrating hole formation in the center of the contacted region between a mold and a substrate.

Another effect that must be considered is instability-driven dewetting. (Suh *et al.*, 2002) Satisfying the prerequisite conditions for incomplete filling of mass in the recessed regions of a mold often results in void formation because of the polymer melt meniscus along the void walls, as shown in Fig. 4(d). (Suh *et al.*, 2002) Fig. 4(e) and Fig. 4(f) show defects owing to capillary instability in the center region of disks and fragmented lines, respectively. To avoid defects induced by dewetting of thin polymer films, it was reported that the aspect ratio (pattern height/width) should be high. (Yoon *et al.*, 2011) In the case of high-aspect-ratio patterns, however, weak adhesion between the mold and the polymer pattern should be considered to be beneficial. When there is no residual layer, polymer resins filled in the voids can stick in the mold as shown in Fig. 4(g). Based on a simplified model, we can obtain the conditions required to avoid of the polymer patterns from a substrate as follows, (Yoon *et al.*, 2011)

$$\left(\pi LH + \frac{1}{4}\pi L^2\right)W_{p-m} < \frac{1}{4}\pi L^2 W_{p-s},$$

$$H/L < 0.25(W_{p-s}/W_{p-m} - 1) \quad (12)$$

where  $W_{p-m}$  and  $W_{p-s}$  are the works of adhesion at the



**Fig. 5.** (Color online) (a) A schematic showing the residual-layer-free process using deformable molds. (Yoon *et al.*, 2010) (b) Defect formation without deformation by dewetting in the center region. (c) Defect elimination with the deformation of a PDMS mold. (d) A schematic showing self-removal of residual layer. (e, f) Scanning electron microscopy (SEM) images of transferred polymer patterns on a substrate. (Dumond *et al.*, 2008).

interfaces between the polymer and mold and between the polymer and substrate, respectively. Fig. 4(h) and 4(i) show examples of polymer patterns detached from the substrate when the aspect ratio is too high.

## 6. Advanced methods for residual-layer-free nanoimprint lithography

As discussed in the previous section, the design of polymer and mold materials as well as the geometry of molds for residual-layer-free nanoimprint lithography is required. However, to optimize those properties, the choices of parameters are limited. To realize a residual-layer-free system, some reports have used deformable molds such as PDMS to inhibit incomplete filling of the polymer resin that otherwise induces hole formation in the center regions and other reports have adapted the transfer methods for self-removal of the thin residual layer. (Dumond *et al.*, 2008; Yoon *et al.*, 2010) Fig. 5 (a) shows a schematic illustration of the residual-layer-free imprint method, in which a deformable PDMS mold is placed on a polymer resist film coated on a substrate. (Yoon *et al.*, 2010) The deformable PDMS mold is pressed to induce a roof collapse. When the amount of polymer resin is less than the volume of the cavity (the recessed regions of a mold), dewetting occurs in the form of separated strips or irregular polymer blocks by Rayleigh instability as shown in

Fig. 5 (b). Under a constant pressure of 4 bar, the roof of the mold suppresses the rising polymer film dewetting from the substrate as shown in Fig. 5 (a). Fig. 5 (c) shows a defect-free pattern after etching of metal films using a polymer resist produced by the residue-free imprint method using a deformable PDMS mold.

As another example, a self-removal process was proposed. (Dumond *et al.*, 2008) Fig. 5 (d) shows a schematic illustration for the self-removal process. First, polymer resin was filled into the recessed regions of mold A. After the detachment of mold A from mold B, the polymer pattern on mold B was transferred to a target substrate. During the detachment of mold B from the substrate, the thin polymer resist layers are separated from the polymer patterns adhered to the substrate by the applied compressive and tensile stresses on the resist patterns. Compressive stress is produced when the polymer patterns are pressed on the substrate, and tensile stress is introduced when the mold is detached from the substrate. This causes shear stress in the thin residual polymer layer resulting in separation from the transferred polymer patterns. Fig. 5 (e) shows an SEM image of polymer patterns without a residual layer after self-removal. Furthermore, this technique is relevant for fabricating reverse-tapered or overhang-type structures such as T-bar column structures (Fig. 5 (f)), which could be employed in organic light emitting diode displays or super-oleophobic surfaces.

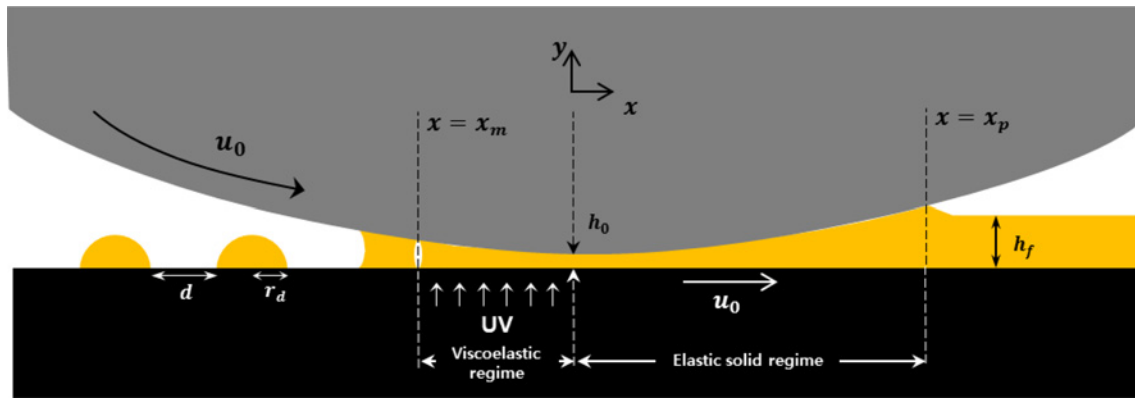


Fig. 6. (Color online) A schematic showing a roll-to-roll nanoimprint process. (Jain *et al.*, 2013).

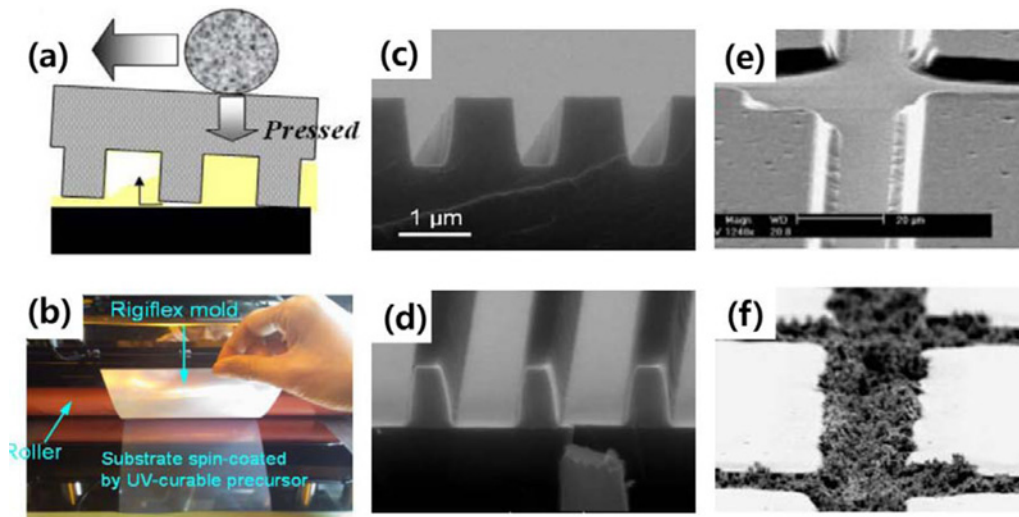


Fig. 7. (Color online) (a) A schematic showing a residual-layer-free roll-to-roll process. (b) A photograph of the experimental setup. (c) A scanning electron microscopy (SEM) image of the mold. (d) An SEM image of the polymer resist. (e) An SEM image of a polymer resist pattern without residual layer. (f) An SEM image of an electroplated pattern. (Choi *et al.*, 2009).

### 7. Roll-to-roll process for large-area fabrication

Roll-to-roll imprint lithography is a process for producing nano- and microstructures by a patterned mold wrapped on a roller. (Ahn *et al.*, 2006; Ahn *et al.*, 2008; Ahn *et al.*, 2009; Choi *et al.*, 2009; Chuang *et al.*, 2011; Dumond *et al.*, 2012; Fagan *et al.*, 2009; Guo *et al.*, 2007; Huang *et al.*, 2009; Inannami *et al.*, 2012; Jackson *et al.*, 2008; Jain *et al.*, 2013; John *et al.*, 2013; Lim *et al.*, 2011; Seo *et al.*, 2007; Stuart *et al.*, 2009; Tan *et al.*, 1998; Vig *et al.*, 2011; Wu *et al.*, 2010; Yeo *et al.*, 2010; Yun *et al.*, 2012) As the demand for large-area fabrication increases, roll-to-roll processes have attracted much attention because of their high throughput and low-cost fabrication, which is essential for commercialization. Since roll-to-roll processes are proven as effective, various application processes have been developed for manufacturing solar cells,

optical films, and OLEDs. Like nanoimprint lithography, UV-curable or thermoplastic polymers may be used in a roll-to-roll process. In UV-assisted roll-to-roll nanoimprint systems, liquid droplets of photocurable prepolymer are dispensed on a substrate as shown on the left side of Fig. 6. (Jain *et al.*, 2013)

A roller with a radius,  $R$ , and substrate move at speed,  $u_0$ . The UV curable polymer has a droplet shape of radius,  $r_d$ , with a drop-to-drop distance,  $d$ . After merging the polymer droplets at  $x_m$ , the polymer film moves to a UV source and is exposed to UV radiation. After pattern transfer, the UV-cured polymer resists are detached from the mold. When the roller radius  $R$  is much larger than the polymer thickness, Equation (13) is derived. (Jain *et al.*, 2013)

$$y = h(x) = h_0 + \frac{x^2}{2R} \quad (13)$$

where  $h_0$  is the film thickness at  $x = 0$ . Based on the mass balance between the liquid droplet volume and the patterned polymer resist, the residual layer thickness is derived as  $h_r = q_v / (2r_d + d)^2$  where  $q_v$  is the droplet volume. Since  $h_r$  cannot be zero based on the equation, we should consider a spontaneous dewetting of polymers from the substrate.

Choi *et al.* demonstrated the concept of residual-layer-free roll-to-roll nanoimprint lithography with a low viscosity polymer resist as shown in Fig. 7 (a) and (b). (Choi *et al.*, 2009) The low viscosity polymer resist also has another advantage: high throughput because of short filling time. When the polymer is sandwiched between a mold and a substrate, the stability can be predicted by the spreading coefficient,  $S$ : (de-Gennes *et al.*, 2004)

$$S = \gamma_{sm} - (\gamma_{sp} + \gamma_{mp}) \quad (14)$$

where  $\gamma$  is the interfacial energy between two materials out of the substrate ( $s$ ), polymer ( $p$ ), and mold ( $d$ ). When the spreading coefficient  $S$  is positive, the polymer film is stable between the substrate and the mold. In the case of negative spreading coefficient, the sandwiched polymer film is too unstable to remain between them. For the residual-layer-free process, the coefficient should be negative in order to remove the polymer layers from the mold and remain on the substrate. As shown in Fig. 4 (b), dewetting starts as a hole-type dry patch and grows with a velocity of  $V_d$  given by (Choi *et al.*, 2009)

$$V_d = \frac{k_1}{\eta} \cdot \frac{|S|^2}{E \cdot h} \quad (15)$$

where  $k_1$  is a prefactor,  $\eta$  is the polymer viscosity,  $E$  is the Young's modulus of the mold, and  $h$  is the film thickness. When we consider roll-to-roll process, the velocity of hole growth should be greater than the roller speed in order to remove the residual layers. From this relationship, we learn that the polymer viscosity should be low enough to obtain a high speed of drainage of the polymer thin film from the mold-substrate interface. Fig. 7 (c) and 7 (d) show SEM images of the rigiflex mold with 500 nm line/space and polymer patterns produced by the mold without residue, respectively. To confirm the absence of residual layers after the process, electrochemical plating of copper on an ITO substrate was carried out. As shown in Fig. 7 (e), a residual layer was not seen between the polymer patterns after measuring with a tilted SEM image. After the ITO surface was patterned by a roll-to-roll nanoimprint process, the polymer patterns were placed in a copper sulfate pentahydrate solution. As shown in the SEM image of Fig. 7 (f), copper was deposited on the interconnected regions, indicating the absence of a residual layer.

## 8. Conclusions

In this review, methods for residual-layer-free nanoimprint

lithography for large-area fabrication were presented. Based on the squeezing theory, it was learned that a dewetting process is required for obtaining high-quality patterns. Also, two types of approaches for removing residues were discussed: incomplete filling and reverse imprint methods. Advanced techniques by self-removal or by using deforming flexible molds were also discussed to suppress unintentional dewetting, which induces defects after processing. It is noted that the choice of polymer and mold materials in addition to mold geometry are important parameters for dewetting based on surface chemistry considerations. It is recommended that roll-to-roll nanoimprint lithography with a low viscosity polymer should be developed for large-area fabrication without residual layers.

## Acknowledgements

This study was supported by Seoul National University of Science and Technology.

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