Scalable Fabrication of Flexible Microstencils by Using Sequentially Induced Dewetting Phenomenon

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ABSTRACT: We present the physics of sequential dewetting phenomenon and continuous fabrication of a polymeric microstencil using dewetting phenomenon with roll-to-roll imprinting equipment. To realize dewetting-assisted residual-free imprinting, mold material, polymer resin, and substrate were selected via interfacial surface energy analysis. In addition, optimal parameters of the continuous process were also studied by experimentally comparing the resultant shape of the microstencil depending on the process speed, aspect ratio of the mold, and applied pressure. As a result, the polymeric microstencil was produced continuously in very high yields, and its maximum resolution reached 20 μm in diameter. For an easy, continuous demolding during the roll-to-roll process, the material chosen for the substrate film was paraffin-coated film, which has the surface energy low enough for dewetting while having a higher adhesion value than polydimethylsiloxane mold. This versatile, high-throughput microstencil fabrication process can be used in many applications requiring flexibility, scalability, and specific material, and high productivity.

1. INTRODUCTION

Inspired by the traditional stenciling, which prints letters or images by applying pigment through a surface having designed cuts, a microscale stencil process has been capitalized in diverse microfabrication works ranging from patterned evaporation or etching to selective cell culturing, mainly by using a thin membrane perforated with desired micropatterns as a shadow mask.1–7 Preparation of such a microstencil has commonly relied on the photolithography on a rigid Si wafer, accompanied by spin-coating of photoresist and later through etching of Si.1,2 However, this conventional wafer-by-wafer process is often unsuitable in broader and more practical applications because of its limitations in area, fabrication speed, and use of flexible substrates.

Alternatively, a soft lithography in which a flexible mold directly imprints a pattern on a UV-curable resin-coated substrate such as polyurethane acrylate (PUA) under a conformal contact can be applied to produce a desired microstencil pattern onto either flexible or rigid substrates at high speed. PUA is the most appropriate UV-curable material because of its small curing-induced shrinkage, mechanical rigidity at high resolutions, flexibility for conformal contact, and high light transmittance for UV curability.8 Although this imprinting-based approach is effective and potentially more scalable, there usually remains a residual layer in the as-imprinted resin pattern, which should be removed by additional etching process to complete a perforated microstencil membrane. For that reason, minimizing the thickness of the residual layer is highly desired. Studies on the efforts to minimize the thickness were attempted by increasing the imprinting pressure, yet this cannot be an eventual solution because of the unwanted crumpling of the soft mold at excessive pressure.9,10

In particular, Kim et al. fabricated a new type of free-standing, yet flexible polymeric membrane as a shadow mask for thermal evaporation using PUA. PUA was drop-dispersed on a patterned polydimethylsiloxane (PDMS) mold and covered with another flat PDMS mold. Pressure was applied from the end of the flat PDMS mold to the other end using a roller that squeezed out PUA between the two PDMS molds. Then, PUA was cured by UV light through transparent PDMS molds for several minutes. The PUA stencil was fabricated by detaching from the two PDMS molds.7 Also, Cho et al. reported a simple replication method to fabricate a flexible free-standing polymeric membrane. They used the fabrication method that involves spontaneous dewetting phenomenon of a...
hydrophilic (hydrophobic), UV-curable resin against a hydrophobic (hydrophilic) mold. Hydrophilic resins (PUA301, PUA311, and NOA73) were used as a lower patterned mold and a flat upper mold to replicate the hydrophobic perfluoropolyether resins. Each material can be exchanged to obtain a free-standing polymeric membrane. Fabrication of a polymeric stencil with UV-curable materials is relatively inexpensive and faster than that utilizing silicon wafer or PDMS as the stencil material. These stencil fabrication methods, however, have a limit on production efficiency because of manual fabrication. Therefore, continuous fabrication studies for stencils made of UV-curable materials are essential to utilize stencils in various fields. In addition, it is required to develop a scalable, high-throughput methodology to create flexible microstencils without applying high pressure and additional etching process.

Addressing these issues in this work, we ingeniously utilized the dewetting phenomenon to change a thin liquid resin layer to a residual layer-free perforated structure upon a conformal contact with a micropatterned mold using the roll-to-roll process, realizing the direct stamping of flexible perforated membranes at a moderate pressure without additional etching procedure. The overall process is integrated in a continuous roll-to-roll imprinting system to further improve the scalability and throughput. Importantly, we analytically confirm that a successful process can be conducted by controlling the substrate surface energy to be low enough to result in good resin dewetting but higher than that of a mold to ease demolding. Parametric effects of the roll-to-roll stamping speed, stamping pressure, and the mold pattern geometry (i.e., pillar diameter and height) are systematically investigated to better understand the principle and optimize the process for many application-specific uses.

2. RESULTS AND DISCUSSION

To derive dewetting phenomenon from specific mold, substrate, and resin materials, a thin layer of resin needs to be created between the mold and the substrate. In this work, PDMS, paraffin-coated film (PPF), and PUA were used as the mold, substrate, and UV-curable resin, respectively. Specifically, for the fabrication of perforated microstencil, the PUA resin between the top of each pillar of PDMS mold and underneath the PPF substrate needs to be thin as shown in Figure 1a. The required thickness of resin can be calculated by

$$t = \left( \frac{A_{\text{eff}} \lambda^2}{2 \pi \gamma} \right)^{1/4}$$

(1)

where $A_{\text{eff}}$ is the effective Hamaker constant for the molecular-level interaction of the thin resin layer with the contacted surfaces, $\gamma$ is the surface energy of the resin, and $\lambda$ is the wavelength of the thin layer. In this case, micropillars of PDMS mold act as a physical barrier to express the half-cycle of waves, and $\lambda$ can be considered to be the pitch of the micropillars in mold. The effective Hamaker constant for the PDMS/PUA/PPF combination is given by the mixing rule combining individual Hamaker constants as follows:

$$A_{\text{eff}} = \left( \sqrt{A_{\text{PPF}}} - \sqrt{A_{\text{PDMS}}} \right) \left( \sqrt{A_{\text{air}}} - \sqrt{A_{\text{PDMS}}} \right)$$

(2)

From the literature, $A_{\text{PPF}} = 0.93 \times 10^{-20} (J)$, $A_{\text{PDMS}} = 5.2 \times 10^{-20} (J)$, and $A_{\text{air}} = 0$, such that $A_{\text{eff}} = 3.01 \times 10^{-20} (J)$. The surface energy of uncured PUA is approximately 40 mJ/m$^2$. Using eq 1 and the acquired variables, the required thickness of the PUA resin for dewetting is estimated by $t (\text{nm}) = 7.4 \times \lambda^{1/2} (\mu m)$ and expressed in Figure 1b in relation to the pitch of the pillar in the mold. For instance, in the case of the 100 $\mu m$
periodic mold shown in **Figure 3c**, the PUA resin layer needs to be thinner than 74 nm for the initiation of dewetting phenomenon. As shown in **Figure 1b,c**, very thin resin layers can be fabricated continuously through sequential contact of pillars by the roll-type patterning system. To fabricate thin resin layers continuously, roll-to-roll patterning system is expected to be a suitable candidate because the curvature of the roll mold leads the line contact between mold and substrate. This gradual contact propagation can produce very thin layers of resin continuously.

**Figure 2** schematically shows the overall process to continuously fabricate the flexible microstencils through the roll-to-roll UV imprint lithography system. Here, a flexible mold with a designed pillar array is fabricated using simple molding (**Figure 2a**) and wrapped around the roll that is used for continuous stamping of the counterprofiled hole pattern on the UV-curable resin-coated flexible substrate as rolling proceeds under a conformal contact (**Figure 2b**). By controlling the interfacial surface energies at the mold–resin–substrate interfaces, the dewetting of a liquid resin coating can be induced at the substrate surface and can locally lead to the

**Figure 2.** Schematic illustration of the roll-to-roll equipment for the fabrication of microstencil. (a) Scheme for PDMS mold fabrication. (b) Scheme for roll-to-roll equipment setup.

**Figure 3.** (a) Photograph of a roll-to-roll fabrication system. (b) Fabricated free-standing microstencil. (c) SEM image of the PDMS pillar mold. (d) SEM image of the fabricated microstencil on PPF by continuous process.
perforation of a resin layer where it becomes particularly thin by contacting to the mold’s pillar tips (Figure 1). In other words, the liquid region engaged by the mold’s pillar tips becomes thin enough to expose the underlying substrate surface by dewetting. Additionally, this perforated resin structure can be smoothly demolded from the mold after the UV curing and can be peeled off the substrate to complete the microstencil fabrication.

Figure 3 demonstrates the exemplary experimental result of the fabrication of the microstencil by following the above-described procedure, where 133 μm-diameter holes are clearly perforated on a 190 μm-thick resin film. The pillar mold successfully creates perforated pillars in the resin layer coated on the flexible substrate in a continuous and high-speed manner in the roll-to-roll imprinting system. Even relatively small size holes (20 μm in diameter) have been fabricated as shown in Figure S1. Notably, the perforated resin layer cured by the UV light remains on the substrate after it was released from the mold contact zone. Here, we used PDMS, PUA, and a plastic PPF (i.e., Parafilm M, Bemis Company, Inc.) for the mold, resin, and substrate materials, respectively. The choice of these materials is essentially based on their surface energies, which is essential to induce proper resin dewetting and demolding at the mold–resin–substrate interfaces.

Such a proper choice of materials can be analytically dictated by calculating the spreading coefficient of a liquid at the mold–resin–substrate interfaces, which can be determined by the interfacial surface energies at mold–resin–substrate interfaces based on the contact angle of the liquid resin for each interface.15

We begin with an equilibrium spreading coefficient $S$, given as

$$S = \gamma_{SM} - (\gamma_{SL} + \gamma_{ML}) \tag{3}$$

where $\gamma_{ij}$ is the interfacial surface energy between $i$ and $j$, and the subscripts $S$, $M$, and $L$ refer to the substrate, mold, and liquid (resin), respectively. If the spreading coefficient $S$ is negative, the liquid will undergo dewetting between the mold and the film.17 The interfacial energy between two surfaces, for instance, $\gamma_{SM}$ in the case of the substrate–mold interface, can be obtained from the geometric mean equation given as18,19

$$\gamma_{SM} = \gamma_s + \gamma_m - 2(\gamma_s^d + \gamma_m^d)^{1/2} + 2(\gamma_s^p + \gamma_m^p)^{1/2} \tag{4}$$

where $\gamma_s$ and $\gamma_m$ are the surface energies of the substrate and the mold, respectively, which are the sum of the dispersion term $\gamma_i^d$ and the polar term $\gamma_i^p$ for each surface. Here, after measuring the contact angles ($\theta$) of deionized water and methylene iodide for each $i$, $\gamma_i^d$ and $\gamma_i^p$ can be obtained from an equilibrium Owens–Wendt term given as20

$$(1 + \cos \theta)\gamma_i = 2(\gamma_i^d \gamma_s^d)^{1/2} + 2(\gamma_i^p \gamma_s^p)^{1/2} \tag{5}$$

Table 1 shows the measured contact angles at each surface. Putting these values to eq 5 and solving the simultaneous equations, we can obtain $\gamma_i^d$, $\gamma_i^p$, and $\gamma_i$ for the micropatterned PDMS mold and PPF substrate as summarized in Table 2 along with the known values reported elsewhere.11,18,19

Table 2. Surface Tension Values of Probing Liquids and Substrates

| probing liquid | $\gamma_s$ (mJ/m²) | $\gamma_m$ (mJ/m²) | $\gamma_{SM}$ (mJ/m²) |
|----------------|-------------------|-------------------|----------------------|
| water          | 21.8              | 51.0              | 72.8                 |
| diiodomethane  | 48.5              | 2.3               | 50.8                 |
| PDMS mold      | 10.8453           | 0.0004            | 10.84                |
| PPF            | 29.18             | 0.32              | 29.5                 |
| PUA (cured)    | 21.6              | 11.2              | 32.8                 |

Putting these obtained surface energy values to eq 4, we finally get the spreading coefficient of $-58.5$ mJ/m², confirming that our material system generates adequate dewetting for perforating the PUA resin layer between the PDMS mold and the PPF substrate.

Another important issue is that the cured PUA stencil should be easily demolded from the PDMS mold but must remain well-attached to the PPF substrate. This can also be analytically verified by comparing the work of adhesion between the PDMS mold and the cured PUA and that of the cured PUA and PPF. A work of adhesion can be calculated as the harmonic mean equation given as18–20

$$W_{12} = 4\left(\frac{\gamma_s^d \gamma_m^d}{\gamma_s^d + \gamma_m^d} + \frac{\gamma_s^p \gamma_m^p}{\gamma_s^p + \gamma_m^p}\right)^{1/2} \tag{6}$$

Putting the above-obtained surface energy values to eq 6, the work of adhesion values for the mold–resin and resin–substrate interfaces are calculated to be 28.8 and 50.9 mJ/m², respectively. These work of adhesion values confirm that the UV-cured PUA microstencil structure could remain on the PPF with stronger adhesion while released from the PDMS mold during the roll-to-roll imprinting process.

Exploring the effects of the imprinting (rolling) speed, imprinting pressure, and mold geometry (pillar size and depth) on the dewetting-assisted microstencil fabrication is crucial to further refine the principle and optimize the process for diverse applications. We first investigate how the rolling speed influences the process by conducting the fabrication at different rolling speeds using the same mold under identical pressure. Figure 4 compares the two experiments carried out at the different film moving speeds of 13.5 cm/min (Figure 4a,b) and 22.5 cm/min (Figure 4c,d) under the imprinting pressure of 15 bar and using the same PDMS pillar mold (50 μm diameter and 20 μm height). The scanning electron microscopy (SEM) images taken from these two cases evidently disclose that a microstencil structure with well-penetrated holes is fabricated at 13.5 cm/min while the holes are not fully perforated at 22.5 cm/min. This demonstrates that a scalable microstencil fabrication can be continuously conducted at a very high speed in a roll-to-roll fashion compared with typical wafer-to-wafer processes, of course, but a sufficient time for contact before curing can allow to better occur the dewetting of viscous liquid during the continuous imprinting.

Understanding the change in imprinted resin layer thickness due to varied imprinting pressures may lead to finding out the appropriate condition for making perforated microstencil structures for the mold used. In addition, we work on the systematic development of microstencils having various hole sizes by applying different molds. Figure 5 shows the dewetting-assisted imprinting results along with the resin layer thickness

| surface | contact angle (°) | contact angle (°) |
|---------|------------------|------------------|
| PDMS mold | 125.0 | 95.5 |
| PPF    | 101.3 | 59.0 |
plot depending on the imprinting pressure and aspect ratios. Here, we use the PDMS micropillar mold with the aspect ratio in the range of 0−2, and the feeding speed of the PPF is fixed to be 13.5 cm/min. The imprinting pressure denotes additionally applied pressure to the ground condition where a mold-wrapped roll and a PPF make a conformal contact by the tensioned film feeding. As the imprinting pressure increases, the imprinted resin thickness reduces; at 10 bar, it reaches 125 μm, which is thinner than the 150 μm pillar height, resulting in successful microstencil fabrication. It is worthy to note that the pressure regime in this experimental space is much lower than that in conventional imprinting processes especially aiming for residual layer-free output, suggesting that our process saves excessive pressure and thus undesirable mold deformation attributed to the dewetting-assisted mechanism. Interestingly, the relation between imprinting pressure and resin thickness is revealed to be nonlinear as indicated in Figure 5; we believe that this is because in the thinner film emerging at increased imprinting pressure, the dewetting effect becomes more dominant to accelerate further thinning of the film. As the pillar diameter decreases, the area of the pillar-contacting, thinned region in the liquid resin layer becomes smaller, which may slightly confine the dewetting phenomenon toward full perforation under identical processing conditions. Also, the increased aspect ratio of pillars for the smaller diameter cases may cause imperfect stamping because of bending, which may be improved by tuning the process conditions and initial pillar height.

3. CONCLUSIONS

In summary, we have presented a continuous process method for fabricating a microscale stencil pattern by a continuous roll-to-roll UV imprint system using dewetting phenomenon. In addition, we analytically presented that a successful process can be conducted by controlling the substrate surface energy. PPF is used in this process as its surface area is low enough to secure good resin dewetting while its adhesion value is higher than that of a mold to ease demolding. Also, we demonstrate parametric effects of the imprinting speed, imprinting pressure, and aspect ratios of the pillar to better understand the principle and optimize the process. Moreover, if a photomask pattern and UV-curable resin are changed, various microstencils can be fabricated by this method. The continuous roll-to-roll UV imprint system using dewetting phenomenon can allow continuous, rapid, and cost-effective fabrication of various microstencils. Therefore, it is expected that this fabrication process can be extensively applied to various fields such as electronic products and cell culturing.

4. EXPERIMENTAL SECTION

4.1. Setup and Operation of the Roll-To-Roll Fabrication System for Microstencil. Aluminum profiles are used in the horizontal and vertical axes for fixing the imprint roll and rails (SMR-18, Namiru optical apparatus) that can adjust the position of the imprint roll. PPF is passed through the lower part of the central imprinting roll with a diameter of 100 mm from the drive roll with motor to a roll covered with PPF. Two idler rolls with a diameter of 60 mm (RORMCS40-
15-L100, Misumi Korea) are fixed to the same height in the coating part for maintaining a tight tension of PPF when PPF is wound by driving roller, and then it makes possible to coat the UV-curable resin on a PPF uniformly. The linear UV LED (SUV-L12S016A, UVSMIT) that is capable of adjusting the intensity at the maximum value of 1600 mW/cm² at a 365 nm wavelength is equipped under the imprint roll. The linear UV LED of this roll-to-roll process has a short time to irradiate the imprint part because the film is moved at a constant speed. The UV intensity is fixed at 480 mW/cm². In the demolding part, the drive roll on the left side is mounted at the stepping motor with a minimum rotation speed of 6 rpm (PKE543AC-TS20, Oriental motor). The decelerator with a 1:20 reduction gear for the drive roll on the left side is mounted at the stepping motor.

**4.2. Fabrication of Master Mold Using Photolithography.** We fabricated the master mold using two methods. First, to apply the SU-8 (2050) that had a thickness of 40 μm on a silicon wafer, a thin layer of photoresist is spin-coated at 4000 rpm for 30 s, followed by soft baking at 65 °C for 3 min and then 95 °C for 6 min on a hot plate because of removal a solvent of photoresist. An FCG (film covered glass) mask of dotted pattern with diameters of 25 and 50 μm is equipped in the UV exposure machine (Mask aligner, CA60TS); a thin layer of photoresist is exposed to an ultraviolet radiation with 155 mJ/cm². To make a strong bond between cross-linking polymer in the photoresist, a post baking was performed at 65 °C for 1 min and 95 °C for 6 min on a hot plate. After the post-exposure bake, the wafer is developed in an SU-8 developer for 5 min, followed by rinsing with isopropyl alcohol (IPA) for approximately 10 s and then drying with nitrogen gas. Second, to apply a photoresist having a thickness of 150 μm on a silicon wafer, we changed the process condition appropriately. Finally, master molds having the thickness of 40 and 150 μm with microholes were fabricated. The patterned master mold is silanized by fluorinated silane to facilitate the demolding of PDMS from the mold. The fluorinated silane is sprayed on the patterned master mold and then vaporized at 120 °C for 40 min. The fluorinated silane involves antiadhesive properties that can control the surface energy between PDMS and the master mold.22

**4.3. Fabrication of Soft Mold Using PDMS.** The soft mold with the micropillar should be equipped on the imprint roll with a curved surface and should not be deformed easily because of the imprint force between imprint roll and substrate. In this study, soft mold is fabricated by PDMS (Sylgard 184 kit, Dow Corning), is chemically stable, durable, and flexible. PDMS prepolymer made of a mixture of a silicone elastomer base and a curing agent at a 10:1 ratio is coated on silicone master mold thinly and uniformly, followed by degassing to remove bubbles generated during the inflow of air while mixing PDMS prepolymer in a vacuum desiccator for 30 min. Then, it is cured by heat at 70 °C for 60 min in an oven, and finally, the cured PDMS mold is released from the master mold.

**4.4. Fabrication of PUA Microstencil.** PUA as a UV-curable resin is composed of oligomer, monomer, and photoinitiator. Oligomer influences the physical property of the resin, and it forms a thin cured membrane through the polymerization reaction. Oligomer is component of a cured membrane and forms polymer bonds by the polymerization reaction in influencing components of the physical properties of the resin. Monomer, the cross-linking agent of reactive oligomer, is a raw material in the formation of a cured film through polymerization. A photoinitiator that absorbs ultraviolet to generate a radical or cation is a raw material to initiate polymerization. When UV curable resin is exposed to UV light, a photo-initiator in resin is excited by UV energy. The excited photoinitiator photopolymerizes an oligomer in a short time; it completes the polymerization reaction that is photopolymerized continuously to monomer and oligomer as the main component of UV-curable resin. Di- or tricarlylated cycloaliphatic urethane oligomer is used as the prepolymer for PUA. In addition, 1-hydroxy-cyclohexyl-phenyl-ketone or 2-hydro-2-methyl-1-phenyl-1-propane is used as the photoinitiator.8,23 In this study, PUA (MINS-311RM, Minuta Tech), which has high curing speed and excellent physical properties, is used as the UV-curable resin for the fabrication of polymeric microstencil. Also, PUA is capable of self-replicating each other and shows a smooth demolding and pattern transfer below a height of scores of micrometers. PUA is coated between PPF (Parafilm M, Bemis Company, Inc.) as a substrate and a patterned PDMS mold and will generate dewetting phenomenon when sufficient pressure is applied. Then, PUA resin between a tip of the PDMS pillar and the flat mold flows to side wall part of pillars by dewetting phenomenon, and a residual layer free area can be induced. As previously stated, the fabrication of a polymeric microstencil with UV-curable resin using dewetting phenomenon is the process in which UV-curable resin is pressurized for approximately 1 min between the patterned mold and a substrate such as a flat PDMS mold. However, these conventional fabrication methods have the practical limitation that the substrate cannot be supplied consecutively. To utilize the polymeric microstencil fabricated by continuous roll process easily, polymeric microstencil must be demolded off the substrate film. PPF can be used in roll-to-roll continuous fabrication process as a substrate film.
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