Patterning of Microstructures on SU-8 Coated Flexible Polymer Substrate Using Roll-to-roll Ultraviolet Nanoimprint Lithography

Nazrin Kooy¹, Khairudin Mohamed^{1*} and Kamarulazizi Ibrahim²

¹School of Mechanical Engineering, Universiti Sains Malaysia, Engineering Campus, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

> ²School of Physics, Universiti Sains Malaysia, 11800 USM Pulau Pinang, Malaysia

*Corresponding author: mekhairudin@eng.usm.my

Abstract: In the present work, 2.2µm-wide line gratings are continuously patterned onto a SU-8 coated flexible polymer substrate using an in-house designed roll-to-roll ultraviolet nanoimprint lithography (R2R-UV-NIL) system. A master mold was first produced by patterning the silicon wafer coated with Microresist's ma-N2410 electron sensitive negative photopolymer with line grating patterns using electron beam lithography (EBL) process. A flexible replica of the master mold was then produced using heat-curable poly-dimethyl-siloxane (PDMS) via cast molding process to be used as the flexible mold in the imprint lithography process. Using a commercially available 50 µm-thick polyethylene terephthalate (PET) film as the flexible substrate and fast curing SU8-2002 UV-curable negative photopolymer as the imprint resist, continuous patterning of the line grating structures has been carried out at the speeds of 50, 100 and 150 mm min⁻¹ using the R2R-UV-NIL imprinting tool. The atomic force microscopy (AFM) measurements of the imprints showed good pattern quality and reproducibility for all three speeds. This work has demonstrated that R2R-UV-NIL is a promising technique and tool for fabricating microstructures on flexible substrate for future applications.

Keywords: Nanoimprint lithography, flexible substrate patterning, SU-8 imprinting, patterning of microstructures, electron beam lithography

1. INTRODUCTION

The increasing demand for lower cost, higher throughput and higher resolution micro/nanofabrication techniques to fabricate the various types of 2D and 3D micro/nanostructures featured in various flexible electronics, microelectro-mechanical systems (MEMS), biochip and optical devices^{1,2} has lead to the introduction of nanoimprint lithography (NIL) by Chou et al.³ in 1995. In the NIL process, a prefabricated mold containing an inverse of the desired patterns is pressed onto a polymer-coated substrate to replicate the patterns via mechanical

[©] Penerbit Universiti Sains Malaysia, 2013

Patterning of Microstructures

deformation, where multiple replications could be produced from a single mold using this technique.

As the NIL process is based on direct mechanical deformation of the resist layer, its resolution is not constrained by the limitations of beam scattering or light diffraction factors as observed in conventional nanolithography methods.¹ The NIL process can be generally divided into two basic variants: Thermal-NIL and UV-NIL. In thermal-NIL, heat is used to soften a thermoplastic polymer for imprinting and followed by cooling of the polymer for solidification. In UV-NIL however, the heating and cooling cycle is not required as a UV-curable liquid photopolymer is used for imprinting, where UV exposure is then utilised to solidify the resist via polymer cross-linking. In many applications however, the UV-NIL process is more preferable due to its simpler process mechanism, lower imprint force requirements and capability to be conducted at room temperature without the need for heating and cooling cycle.⁴

Recent developments on NIL process observed the tendency of adapting roller-based imprint mechanism due to the advantages of lower imprint force requirements and higher process throughput as well as uniformity.^{5,6} Its suitability for large area imprinting has seen the roller-based NIL processes being widely applied in industries for fabrication of flexible electronics as seen in the work of Zang and Liang,⁷ Kim et al.⁸ and Holland et al.⁹ However, most of the UV-based roller nanoimprint system utilises specially formulated solvent-free resist⁹ or low-viscosity resist,⁵ which is much costlier as compared to commercially available solvent-based resists such as MicroChem SU-8 permanent epoxy negative photoresist. Nevertheless, the imprint capability of the in-house developed prototype roll-to-roll ultraviolet nanoimprint lithography (R2R-UV-NIL) system using solvent-based SU-8 2002 epoxy photoresist from MicroChem for sub-2.2µm structures are demonstrated and evaluated in the present work.

2. EXPERIMENTAL

2.1 R2R-UV-NIL System Setup

The imprinting system used in the present work is an in-house developed lab-scale prototype of a roll-to-roll ultraviolet nanoimprint lithography system designed for imprinting onto a 200 mm-wide flexible substrate. An image of the prototype nanoimprint system as well as its operating concept is as shown in Figure 1.



Figure 1: Illustration of (a) photograph image and (b) operational concept illustration of the prototype R2R-NIL system.

The operation of the prototype NIL system consists of four stages which includes resist-coating stage, soft-bake, imprint and curing. In the first stage (resist-coating), a flexible polymer substrate of 200 mm in width was first fed into the system, where it was coated with a layer of UV-curable negative photopolymer using a coating roller. A doctor blade is used to meter the coating layer to improve the coating uniformity and thickness. In the present work, a 50 μ m-thick commercially available polyethylene terephthalate (PET) film (Lumirror T60, Toray Industries) was used as the flexible substrate due to its high transparency, resistance to solvent content and its relatively high surface energy of 40 dynes cm⁻¹⁰, which will promote adhesion of resist coating onto the film. As for the imprint resist, UV-curable solvent-based SU-8 2002 epoxy negative photoresist from MicroChem was selected due to its relatively low cost and availability as compared to specialty resists as well as its low UV-dosage requirements (below 200 mJ cm⁻²) for cross-linking,¹¹ which allows high-speed curing of the polymer during imprinting.

In the following soft-bake stage, the SU8-coated PET film was then baked at an elevated temperature to remove the solvent content in the resist, which would improve its curing properties as well as reducing the tendency to stick onto the mold and roller during imprinting. The introduction of the softbake stage in the process was aimed to allow solvent-based and higher viscosity resists such as MicroChem SU-8 to be used as the imprint resist without the need for specially formulated low-viscosity or solvent-free resist.

After the baking process, the SU-8 coated PET film was fed into the imprinting unit, where a prefabricated flexible mold containing the negative of the desired pattern was pressed against the SU-8 coating between the imprint rollers under a preconfigured force of 100 N. The imprint pressure would cause the SU-8 coating to fill in the mold cavity to form the desired structures, before

Patterning of Microstructures

being cured via cross-linking due to UV-A exposure (wavelength 320–420 nm, peak 365 nm) at the curing stage. The cured SU-8-coated PET film was then separated from the mold at the other end of imprint roller and dispensed at the process output.

2.2 Imprint Mold Fabrication

A master mold was first fabricated as the template for cast molding of the flexible mold. A P-type silicon wafer was first cut into 10×10 mm samples, where it was then cleaned using acetone, methanol and isopropyl alcohol (IPA) in ultrasonic bath for 5 min in each solvent. The sample was then spin-coated with Microresist ma-N2410 electron-sensitive negative photoresist at a spinning speed of 1500 rpm for 30 s, followed by soft-bake at temperature of 90°C for 30 min in a convection oven. Line gratings of sub-2.2 μ m in width were then patterned onto the sample using electron-beam lithography (EBL). The patterned sample was then developed using Microresist ma-D525 developer for 80 s before being rinsed with deionised water and blow-dried with compressed air.

To produce the flexible mold using cast molding technique, a heatcurable polydimethyl-siloxane (PDMS) solution (Sylgard 184 Silicone Elastomer, Dow Corning) was first prepared by thoroughly mixing the base elastomer and the curing agent at a mass ratio of 10:1 and put in vacuum for 10 min for the removal of air bubbles. PDMS was selected in this work due to its low surface energy of 24 dynes cm⁻¹⁰, which reduced the sticking tendency of the resist during imprinting. Using a spin-coater, the PDMS solution was spin-coated onto the EBL-patterned wafer at a spinning speed of 800 rpm for 30 s to achieve thickness of approximately 150 μ m. Using a convection oven, the PDMS coating was cured at temperature of 120°C for 1 h, before being peeled off using a sharptipped tweezers after it cooled down to room temperature.

2.3 Imprint Demonstration

Using the R2R-UV-NIL setup and PDMS mold elaborated previously, the imprinting process was carried out at three different speeds of 50, 100 and 150 mm min⁻¹. The mold was positioned at the centre of the imprint roller for imprinting and secured using cellulose tape from Loytape. 10 consecutive imprints were produced at each speed, where the imprinted profiles were then measured using atomic force microscope (AFM) for evaluation.

3. **RESULTS AND DISCUSSIONS**

The AFM evaluation of the imprints shows good replication quality for all produced imprints at all three imprint speeds without a significant mold sticking issue. Figure 2(a) shows an AFM image of the flexible mold used, whereas Figure 2(b) shows the AFM image of one of its associated imprints. However, a comparison between the measurement profile of the imprinted line gratings and the original gratings on the mold as shown in Figure 3 shows that the height of the imprinted structure is actually higher than the mold cavity depth.



Figure 2: AFM image of line grating on (a) PDMS mold and (b) the 10th imprint produced at 100 mm min⁻¹.

However, this anomaly is a known problem caused by the deformation of the PDMS soft mold during imprinting. The low modulus of the mold, combined with the applied pressure caused the resist in the mold cavity to slightly stretch the mold cavity in the height-direction as illustrated in Figure 4, resulting in an increased feature height and a slightly reduced width profile of line gratings as observed in Figure 3 (although peak-to-peak of cavity remained unchanged). Nevertheless, materials with higher modulus such as "hardened PDMS" or h-PDMS¹² and ETFE⁵ may be utilised as the mold material to minimise mold deformation during imprinting while maintaining low resist sticking tendency.

As the mold cavity was stretched during imprinting stage, the decompression of the stretched mold cavity after the imprint stage (during curing stage) should result in a forced resist reflow which theoretically would cause the imprint height to be reduced to the equal depth of the mold cavity, or lower. However, the low UV dosage requirements of the SU-8 resist results in rapid curing of the resist layer (approximately 7 s under measured intensity of 30 mW cm⁻²), which results in insufficient time for resist reflow and thus, maintaining the height of the structure produced at imprint stage. Nevertheless, the low curing dosage requirements of the SU-8 resist remains an important property of the R2R-NIL resist to ensure complete pattern replication and high speed imprinting.



Figure 3: AFM profile comparison of the PDMS mold and its imprint shown in Figure 2.



Figure 4: Illustration of PDMS mold stretching during imprinting due to its low modulus.



Figure 5: Photograph image of resist layer peeling due to sticking on adhesive tape during imprinting.

In addition, another issue observed in the imprint process was the resistpeeling issue. While no mold-sticking issue observed, the direct irradiation from the UV lamp without filter also included heat-producing infrared irradiation, which caused an increase in roller temperature during imprinting. The temperature increase caused the property of the cellulose tape, used to secure the PDMS mold, to change, resulting in an increased sticking tendency of the tape to the resist layer.

Sticking of the resist layer onto the adhesive tape may result in peeling of resist layer from the PET film as shown in Figure 5, should the tear resistance of the resist layer be sufficient to withstand the demolding force. While no significant resist peeling was observed with the thin coating thickness of approximately $5.5 \mu m$ obtained in the this work, the utilisation of a thicker resist coating for imprinting of larger structures may result in peeling of the imprint region from the PET film due to the increased tear resistance, even though the imprinted region did not stick to the mold. Nevertheless, the use of adhesion promoter may be required for imprinting using thicker coating of SU-8 resist to improve the resist adhesion to the substrate to minimise resist peeling. Additionally, the usage of infrared filters to minimise the infrared irradiation as well as high temperature tape (i.e., aluminum foil tape) should also reduce the roller-sticking tendency of the resist during imprinting.

4. CONCLUSION

Continuous patterning onto flexible substrate using commercially available SU-8 epoxy photoresist has been carried out using the in-house designed prototype nanoimprint lithography system, where AFM evaluations have observed good replication quality for sub-2.2µm line gratings imprinted using a cast-molded PDMS mold. While minor issues such as PDMS mold deformation as well as resist-peeling were observed in the present work, further efforts in characterising and optimising the process as well as improvements on the properties of the polymers used in the imprinting process should allow better imprint quality to be achieved at higher imprint speeds. Nevertheless, the findings obtained from this work provide a potential solution towards low-cost and high-throughput micro/nanopatterning technique using commercially available solvent-based photoresists.

5. ACKNOWLEDGEMENT

Financial support from Universiti Sains Malaysia via its Short Term Grant (Grant number 304/PMEKANIK/60311043) is gratefully acknowledged by the authors.

6. **REFERENCES**

- 1. Guo, L. J. (2007). Nanoimprint lithography: Methods and material requirements. *Adv. Mater.*, 19(4), 495–513.
- 2. Maury, P. et al. (2011). Roll-to-roll UV imprint lithography for flexible electronics. *Microelectron. Eng.*, 88, 2052–2055.
- 3. Chou, S. Y., Krauss, P. R. & Renstro, P. J. (1995). Imprint of sub-25nm vias and trenches in polymers. *Appl. Phys. Lett.*, 67(21), 3114–3116.
- 4. Lee, J. J. et al. (2008). Nano-scale patterning using the roll typed UVnanoimprint lithography tool. *Microelectron. Eng.*, 85, 861–865.
- 5. Ahn, S. H. & Guo, L. J. (2008). High-speed roll-to-roll nanoimprint lithography on flexible plastic substrates. *Adv. Mater.*, 20, 2044–2049.
- 6. Lan, S. et al. (2010). Continuous roll-to-flat thermal imprinting process for large area micro-pattern replication on polymer substrate. *Microelectron. Eng.*, 87, 2596–2601.
- 7. Zang, H. M. & Liang, R. C. (2003). Microcup® electronic paper by rollto-roll manufacturing processes. *The Spectrum*, 16(2), 16–21.
- 8. Kim, J. G. et al. (2009). Large area pattern replication by nanoimprint lithography for LCD-TFT application. *Microelectron. Eng.*, 86, 2427–2431.
- 9. Holland, E. R. et al. (2011). An enhanced flexible color filter via imprint lithography and inkjet deposition methods. *J. Display Technol.*, 7(6), 311–317.
- 10. Flexcon. (2011). Adhesive solutions to the challenges of bonding to low surface energy surfaces. Online white paper. Available from: http://www.flexcon.com/ Resource-Center/White-Papers.aspx.
- MicroChem. (2012). SU-8 2002 Permanent epoxy negative photoresist: Processing guidelines for SU-8 2000.5, SU-8 2002, SU-8 2005, SU-8 2007, SU-8 2010 and SU-8 2015. Version 4.0. Newton (MA): MicroChem. Available from: http://www.microchem.com/pdf/SU-82000DataSheet2000_5thru2015Ver4.pdf.
- 12. Schmid, H. & Michel, B. (2000). Siloxane polymers for high resolution, high accuracy soft lithography. *Macromol.*, 33, 3042–3049.