Room-Temperature Nanoimprint Lithography Using Photosensitive Dry Film

Ken-ichiro Nakamatsu and Shinji Matsui

University of Hyogo, Graduate School of Science, LASTI, 3-1-2, Koto, Kamigori, Ako, Hyogo 678-1205, Japan

Keywords: Photosensitive dry film, room temperature, nanoimprint, lithography

Nanoimprint lithography (NIL) has been attracting much interest from many industrial fields because of its many capabilities of providing various micro- and nano-structure applications such as optical devices. NIL, in which resist patterns are fabricated by deforming the physical shape of a resist by embossing it with a mold, promises the high-throughput fabrication of various nanostructure devices including quantized magnetic disks and polarization plates. The advantage of NIL is that patterns less than 10 nm in size can be realized over a large area at a high throughput and low cost, meaning that highly precise pattern transfer can be performed.

Thermal-NIL requires a resist thermal cycle to enable the deformation of a resist with a mold. However, the resist thermal cycle yields serious problems such as reductions in throughput and pattern accuracy. To address these problems, we previously developed room-temperature nanoimprint lithography (RT NIL) using hydrogen silsesquioxane (HSQ; Dow Corning Co. FOX-16) as the replicated material. The HSQ used in our experiments is an inorganic polymer and consists of repeating units of HSiO$_2$. The RT-NIL we developed can achieve nanostructure replication by imprinting without the resist thermal cycle, and has demonstrated 50-nm-linewidth HSQ patterns.

In addition, UV-NIL is better suited for the replication technique because the resist thermal cycle can be eliminated, as in RT-NIL. Moreover, although thermal-NIL requires a pressure of approximately 5 MPa in the UV-NIL, a low pressure of around 1 MPa can be used because UV-curable monomer liquids with a low viscosity are used as the replication material. However, UV NIL is sensitive to the flatness of the stage and mold owing to the low viscosity of the UV-curable liquids. The low viscosity leads to the nonuniformity of the thickness of the residual layer in the recessed area after imprinting. Also RT-NIL requires a higher imprinting pressure within a range from about 5 to 40 MPa in nanostructure replication. To overcome this problem, a commercially available photosensitive dry film (Photec RY-3300 series, Hitachi Chemical.) was used as the replication material. Either the dispensing of droplets or spin coating is commonly applied to resist formation in conventional NIL. NIL based on dispensing resists causes problems such as air bubble formation and absorption. In our experiment, lamination was applied to UV-curable resin formation, as an alternative to the dispensing of droplets and spin coating. Lamination is capable of realizing homogeneous dry resin for uniform imprinting. As another benefit, it can be applied to
various substrates such as Si, glass and polymer substrates with various shapes.

In this study, we demonstrate dry film patterns replicated by RT-NIL based on UV curing.

We applied RT-NIL to the photosensitive dry film. Figure 1 shows a manufactured dry film produced by Hitachi Chemical, which was used in our experiment. This winding product was composed of trilayer structures in which a UV-curable dry layer was sandwiched between two polymer films. The photosensitive layer was formed on the poly(ethylene terephthalate) (PET) used as the base sheet by lamination using a lamination tool. The pressure and temperature for the lamination were 0.4 MPa and 110 °C, respectively. A rapid speed of 1.5 m/min was used in the lamination. The applied thickness after the lamination of the dry film was 10 μm. The polyethylene (PE) film with sufficient tolerance to various solvents covered the dry film, which prevented a deterioration of the photosensitive layer.

After developing, the resist patterns were completed on the Si substrate. Subsequently, the Si layer was etched away by CHF₃ reactive ion etching (RIE) and the resist patterns were used as the etching mask. The gas flow, RF power and pressure for RIE were 50 sccm, 100 W, and 2.0 Pa, respectively. The etching time for the RIE process was 10 min. Finally, the residual resist layer on the Si surface was removed by O₂ RIE. Figure 2 shows a schematic of RT-NIL using a UV-curable dry film; the steps are as follows: (1) The dry film on the PET base film was set to the nanoimprint apparatus (NANOIMPRINTER, Meisyo Co.). The PE cover film was removed beforehand to expose the surface of the photosensitive layer. (2) The Si mold 20 mm by 20 mm in size was pressed onto the dry layer at room-temperature. The Si master mold had protrusions of 100 nm and various patterns with linewidths ranging from about 30 nm to 1 μm. The release agent (Duikin Industries Ltd. Optool DSX : Demnansolvent =1: 1000 by weight.) was pre coated onto the Si mold to avoid the adhesion of the dry film during imprinting. A pressure of 2.5 MPa was used in the imprinting. (3) For curing, the imprinted dry layer was UV-exposed

(1) Initial

[Diagram of Initial Step]

(2) Mold pressing

[Diagram of Mold Pressing]

(3) UV exposure

[Diagram of UV Exposure]

(4) Mold removal

[Diagram of Mold Removal]

(a)

(b)

Fig. 2 Schematic of nanoimprint process using the photosensitive dry film.

We fabricated a Si mold prior to imprinting using electron-beam (EB) lithography and dry etching. The fabrication is as follows. First, patterns were written by EB lithography on the commercial negative tone EB resist (NEB-22A2, Sumitomo Chemical).
while the press pressure was kept constant. The exposure dose and time for the UV curing were 1 J and 20 s, respectively. (4) The Si master mold was separated from the dry layer, and then the patterns of the dry film were replicated. By the way, another imprint process shown in Fig. 2(b) can be also applied when UV exposure is performed immediately after mold separation. An advantage in application of the process of Fig. 2(b) is that UV exposure can be performed at outside of a nanoimprint apparatus.

Figure 3 shows a scanning electron microscopy (SEM) image of the dry film pattern with a 150 nm linewidth and a 300 nm pitch replicated by RT-NIL and UV-curing. As shown in Fig. 3(b), the RT-NIL also provided a nanoscale pattern of 30 nm linewidth.

Figure 4(a) shows an optical photograph of a semicircular Si mold used to provide a dry film pattern shown in Fig. 4(b). This dry film pattern was successfully fabricated by a one-step RT-NIL.

Thus, we can use a UV-curable dry film in RT NIL, and expect it to be widely used in nanostructure replications.

In conclusion, we propose the use of a photosensitive dry film as the replication material for RT-NIL for the first time. Lamination was used for the formation of the dry film on a base sheet. RT-NIL was able to use the dry film as the replication material. Dry film patterns with a 30 nm linewidth were successfully obtained. Moreover, dry film patterns 60 mm × 100 mm in area were obtained. These results demonstrate that a photosensitive dry film is likely to be useful and effective as the replication material for RT-NIL. In addition, we believe that the dry film we used in RT-NIL is suitable for a Rool-to-Rool imprinting because it has winding structures.

Acknowledgements

The authors are particularly grateful to Ms. Miyoshi of Hitachi Chemical Ltd. for offering the UV-curable dry film, and helpful comments and suggestions.

References