Fabrication of Replica Mold by Room Temperature Nanoimprinting using Organic Spin-on-glass

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Keywords: room temperature nanoimprinting, organic spin-on-glass, replica mold

1. Introduction

Nanoimprint lithography (NIL) can transfer the nano-patterns on the master mold into nanoimprint resin by physical contact. [1-3] It can be used effectively to fabricate nano-patterns with simple process and low cost and is therefore considered to be one of the most promising next-generation nano patterning technologies.

A mold replication process with easy and low-cost is required to prevent the damage to a master mold. So far, the duplication of polymer molds from the master mold has been reported. [4] The polymer mold is also required a high mechanical strength because the pattern deformation of the polymer mold with a low mechanical strength is induced by a NIL with a high pressure.

Hydrogen silsesquioxane (HSQ) is a type of spin-on-glass material. As a technique to fabricate the nanostructure of the HSQ, room temperature (RT) NIL is reported. [5-8] The resin-thermal cycle and UV exposure is not needed in the RT-NIL, as shown in Fig. 1. However, we must apply a high pressure to the HSQ in this process. To reduce the imprinting pressure, the organic SOG (O-SOG) is produced from Tokyo Ohka Kogyo Co.. Although the O-SOG has the organofunctional groups, the main chain consisted of -Si-O- bond. We therefore assume that the O-SOG has a higher mechanical strength than the polymer.

![Mold](image1)

![SOG Substrate](image2)

![Pressing](image3)

![Demolding](image4)

Fig. 1 Schematic of room temperature nanoimprint process.

In this study, we fabricated the replica mold of the O-SOG by RT-NIL and demonstrated the thermal nanoimprinting using the replica mold.
2. Replica mold fabrication using O-SOG

A SiO$_2$/Si mold was fabricated by electron beam (EB) lithography and reactive ion etching (RIE). Figure 2 shows the scanning electron microscopy (SEM) image of the line and space (L&S) pattern on the mold. The line- and space-width and height were 190, 300, and 150 nm, respectively. The L&S pattern area was 3.6 mm square on the mold.

This mold was coated an antisticking layer by a dip coating. We used a silane-coupling agent with fluoropolymer (OPTOOL DSX; Daikin Industries) as the antisticking layer. The dip coating process is as follows; (1) The mold is dipped into the 0.1 weight% OPTOOL DSX diluted by HD-TH (Daikin Industries) for 1 min at room temperature in air atmosphere. (2) After dipping into the OPTOOL DSX, the mold is left in the high humidity atmosphere for 1 h. (3) Then, to remove the multiple layers, the mold is rinsed by HD-TH for 5 min.

We used OCNL540 (Tokyo Ohka Kogyo Co.) as the O-SOG. Figure 3 shows the chemical structure of O-SOG. This O-SOG has a methyl group in the polymer structure instead of the H group in the SOG. The curing speed of the OCNL540 is reduced by controlling the sol-gel reaction. The low curing speed induce to the low imprinting pressure.

We fabricate the replica mold for thermal nanoimprinting by RT-NIL using O-SOG. The O-SOG was spin-coated with 2000 rpm on the 15 mm square SiO$_2$/Si substrate. The O-SOG film thickness was about 200 nm in this case. We used the NM-0901SQ (MEISYO KIKO Co.) as a nanoimprinting system. The imprinting pressure and time were 15 MPa and 2 min, respectively. Figure 4(a) shows the SEM image of the L&S pattern on the O-SOG. The pattern was clearly imprinted.

To demonstrate thermal nanoimprinting using the O-SOG replica mold, the replica mold was coated with the antisticking layer by above dip coating process. However, the imprinted O-SOG pattern was disappeared after leaving in the high humidity atmosphere. We assumed from this result that the imprinted O-SOG pattern was removed by the water because the O-SOG was not perfectly cured by RT-NIL. We therefore tried to dip the imprinted O-SOG pattern into the water.

![Fig. 2 SEM image of L&S pattern on mold.](image)

![Fig. 3 Chemical structure of OCNL540.](image)

![Fig. 4 SEM image of (a) imprinted O-SOG pattern. (b) post-baked O-SOG pattern.](image)
we carried out post-baking at 120 °C for 2 min. The pattern shrinkage was not occurred by this baking process, as shown in Fig. 4(b). We confirmed that the post-baked O-SOG pattern was maintained after dipping into the water. This result indicates that the O-SOG was cured by post-baking. In fact, we were able to carry out the dip coating process of the antisticking layer for the post-baked O-SOG replica mold.

3. Thermal nanoimprinting using O-SOG replica mold

We next demonstrated the thermal nanoimprinting using the O-SOG replica mold. NEB-22 (Sumitomo Chemical Co.) was used as a thermal nanoimprint resin. The NEB-22 was spin-coated on the Si substrate and, then, this substrate was prebaked at 110 °C for 2 min. The mold and substrate temperatures increased by 150 °C during thermal nanoimprinting. The imprinting pressure and time were 20 MPa and 60 sec, respectively. After cooling process, the mold was demolded from the NEB-22/Si substrate. The L&S pattern was successfully transferred onto the NEB-22, as shown in Fig. 5(a).

In the nanoimprinting process, the adhesion force between the O-SOG and SiO₂/Si substrate is an important factor because the replica mold must be in direct contact with the resin. If this force is small, the O-SOG pattern was separated from the substrate during nanoimprinting process. To evaluate the adhesion force between the O-SOG and SiO₂/Si substrate, we demonstrated step and repeat thermal nanoimprinting using the O-SOG replica mold. The imprinting pressure was 20 MPa. The mold and substrate temperatures were 180 and 90 °C, respectively. Figure 5(b) shows the SEM image of the 40th imprinted pattern. The pattern was clearly imprinted on the NEB-22. Furthermore, after 40 times of repeated nanoimprinting, the O-SOG pattern was not separated from the SiO₂/Si substrate. We confirmed from this result that the adhesion force between the O-SOG and SiO₂/Si substrate is sufficient to carry out the thermal nanoimprinting.

4. Conclusion

We proposed the replica mold fabrication using the O-SOG. The pattern was imprinted on the O-SOG with RT-NIL. We demonstrated thermal nanoimprinting using the O-SOG replica mold with the antisticking layer. The pattern was successfully imprinted on the NEB-22. In addition, we were able to carry out 40 times of repeated thermal nanoimprinting using the O-SOG replica mold.

References