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Pattern replication of 100 nm to millimeter-scale features by thermal nanoimprint lithography

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Abstract

The aim of this work is to demonstrate the ability of nanoimprint lithography (NIL) to replicate patterns having feature sizes ranging from nanoscale to millimeter scale. The pattern replication process includes NIL on PMMA, PMMA RIE, metal liftoff and then silicon RIE for final pattern transfer. A tri-layer resist scheme was employed to facilitate the liftoff. We studied systematically the dependence of the maximum duplicable feature size on imprint temperature, pressure and time, which shows good agreement with a simple squeeze flow model. The maximum duplicable feature size also depends on PMMA molecular weight and the amount of PMMA RIE. For example, with NIL at 200 °C and 20 bar for 20 min and PMMA etching of 180 nm, we duplicated 1.3 mm square pattern without defects using 12 kg/mol PMMA. Such amount of PMMA RIE leads to the nanoscale grating line-width increase of 18 nm.

Keywords: Thermal nanoimprint lithography; Hot embossing; Pattern replication; Liftoff; Polymer viscosity

1. Introduction

NIL has attracted more and more academic and industrial attentions in recent years. It can be divided into two categories: thermal NIL (or hot embossing lithography) and UV-curing NIL based on photo-polymerization of monomers. While UV-NIL has advantages including absence of thermal expansion that impedes precise alignment, low imprint pressure and low viscosity of the uncured resist, thermal NIL is now more widely employed for nano- and micro-pattering, largely because it is more straightforward and works with a broad range of polymer materials.

Thermal NIL has demonstrated high resolution of 5 nm and molecular scale replication of single-walled carbon nanotube [1,2]. On the other side, the replication by NIL of very large features such as bonding or welding pads is much more challenging since more polymer has to be displaced over longer distances. One way to circumvent this issue is to design the mould layout with additional cavities as polymer sinks within large protruded pads. Other approach to replicate patterns having very different sizes involves a mix-and-match method, a two-step process where small features are patterned by NIL while large features are created by photolithography with alignment to the underlying pattern. More recently, a simplified mix-and-match method, termed as combined nanoimprint-and-photolithography, was demonstrated capable of patterning features having various sizes by using a hybrid mask-mould and SU-8 as both photoresist and thermal NIL resist [3]. Nevertheless, a process able to replicate over multiple length scales using NIL alone will take advantage of its low cost and high throughput nature. Previously, pattern size ranging from 250 nm to 100 μm has been imprinted by thermal NIL [4]. In this paper, we will present a systematic study of using NIL for replicating up to millimeter-scale features while retaining reasonable pattern duplication fidelity for nanoscale features.

2. Experimental

The mould used in this work was fabricated by the mix-and-match method. It consists of isolated square and line

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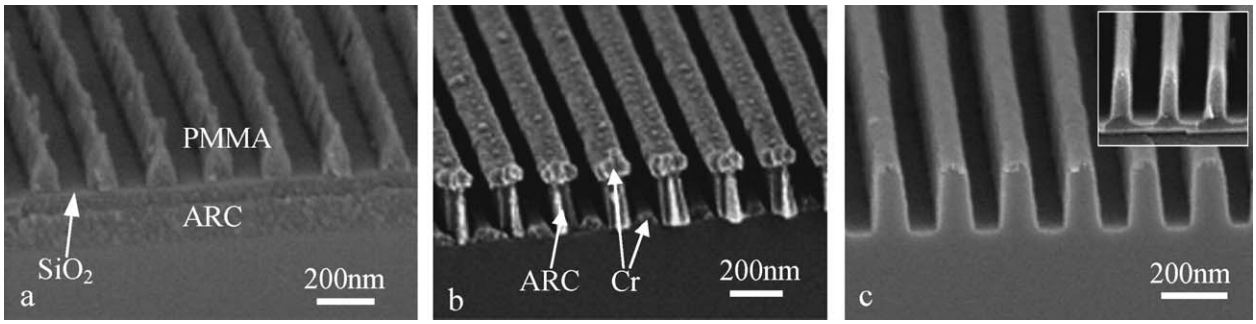

Fig. 1. (a) 200 nm period grating in PMMA resist after 180 nm PMMA RIE; (b) after etching into ARC and evaporating 30 nm Cr at normal incidence angle; and (c) after RIE pattern transfer into silicon substrate. The insert shows the grating of the mould having line-width 80 nm, about 18 nm narrower than the duplicated grating in (c).

patterns with size ranging from 50 μm to 2 mm and a 200 nm period grating covering the remaining area of a 4″ wafer. The height of the patterns on the mould is 250 nm.

Using this mould, pattern replication was realized by NIL, metal liftoff and RIE. To facilitate the subsequent metal liftoff process, the substrate for imprint consists of three layers [5]: a 163 nm crossed-linked polymer ARC (antireflection coating XHRiC-16, from Brewer Science), a 7 nm evaporated SiO_2, and a 250 nm PMMA resist. After NIL, PMMA was etched for 120–240 nm by oxygen RIE (Fig. 1(a)). The thin SiO_2 layer was then etched using CHF_3 gas that also etched another ~8 nm PMMA, and the pattern was transferred into ARC with over etching to create an undercut profile for easy liftoff. Next, 30 nm Cr was evaporated at normal incidence (Fig. 1(b)) and lifted off by dissolving ARC. Finally, the pattern was etched into silicon using CF_4/O_2 gas, followed by Cr removal using Cr-4S etchant. The completed grating duplicated into silicon is shown in Fig. 1(c).

3. Results

Our results show that for the nanoscale grating pattern, the mould feature was faithfully duplicated into the PMMA resist regardless of the NIL parameters. So the CD (critical dimension) change, or the line-width increase for the present case, depends only on the amount of PMMA RIE. The CD change is caused by lateral etch, and as shown in Fig. 2, it increases with the amount of PMMA RIE. For instance, 180 nm PMMA etching will lead to CD change of about 18 nm. In practice, this predicable CD change could be compensated by adjusting accordingly the line-width of the mould structure.

The pattern replication for large features strongly depends on the NIL parameters, the molecular weight of the PMMA resist and the amount of PMMA RIE (see Section 4). Fig. 3 shows the result for NIL with 12 kg/mol PMMA at 200 °C and 20 bar for 20 min, followed by 180 nm PMMA etching. We found that square patterns with size up to 1.3 mm were duplicated without void defects. For the line patterns undergoing the same process, the lines have been duplicated faithfully only up to 0.7 mm

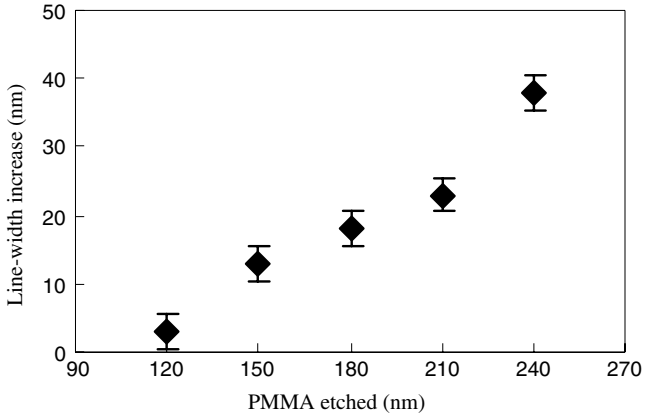

Fig. 2. Line-width increase of the duplicated grating as a function of the amount of PMMA RIE.

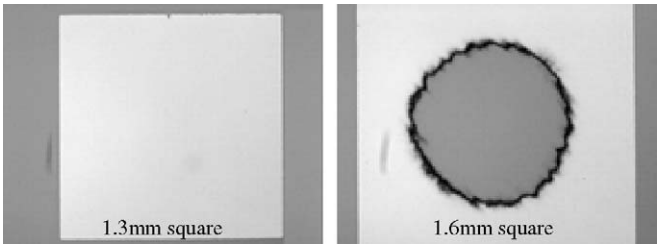

Fig. 3. Squares duplicated by NIL with 180 nm PMMA RIE, showing that the 1.3 mm square was faithfully duplicated, while the 1.6 mm square (the next size) was not.

(not shown), which represents about half that of the duplicable square size. This result qualitatively agrees with the fact that for lines the polymer can be squeezed out from only two sides.

4. Discussion

For a periodic grating structure, the maximum duplicable feature size, L, achieved when the resist etching is equal to its residual thickness, can be evaluated using a simple squeeze flow model assuming the polymer as a viscous and incompressible Newtonian fluid. The following relation can be obtained [6–8]:

$$L \propto \left(\frac{1}{h_f^2} - \frac{1}{h_0^2}\right)^{-1/2} \sqrt{\frac{pt}{\eta}} \approx h_f \sqrt{\frac{pt}{\eta}} \quad \text{if } \frac{h_f^2}{2h_0^2} \ll 1, \tag{1}$$

where h_f and h_0 are the residual and original resist thickness, respectively, p is the applied pressure, t is the imprint time and η is the viscosity of the polymer. In this work, the pattern to be duplicated is not a periodic grating, so deviation from the model can be expected and the equation only serves for an approximation.

In order to compare our results with values predicted by Eq. (1), we conducted a series of trials with fixed h_0 of 250 nm. First, we measured the residual layer thickness at the center of the squares as a function of square size for both a "favorable" (200 °C) and an "unfavorable" (150 °C) imprint condition using 12 kg/mol PMMA and 20 bar for 20 min. As shown in Fig. 4, for both cases, L is roughly proportional to h_f when h_f is less than roughly 180 nm. For higher residual layer thickness, wafer bending becomes more significant, leading to h_f even greater than h_0 for large squares.

Second, the effect of temperature and molecular weight is reflected in the resist viscosity, which can be roughly estimated by the following equation [6,9,10]:

$$\log \eta = n \log M_w - \frac{12.21(T - T_g)}{70.1 + (T - T_g)} + \text{const}, \tag{2}$$

with $n = 1$ for $M_w < M_C$ (critical molecular weight, 30 kg/mol for PMMA) and $n = 3.4$ otherwise. The viscosity can be changed by over two orders when varying the temperature between 150 °C and 200 °C, or increasing the molecular weight to 120 kg/mol. The L vs. $1/\sqrt{\eta_r}$ curve, where η_r is the relative viscosity calculated from Eq. (2) and L is the duplicable square size derived from the corresponding L vs. h_f curves (two of them shown in Fig. 4) at $h_f = 180$ nm, is shown in Fig. 5. These results indicate a fair agreement with the model represented in Eq. (1). The deviation can be partly explained by the inaccuracy of Eq. (2) [10].

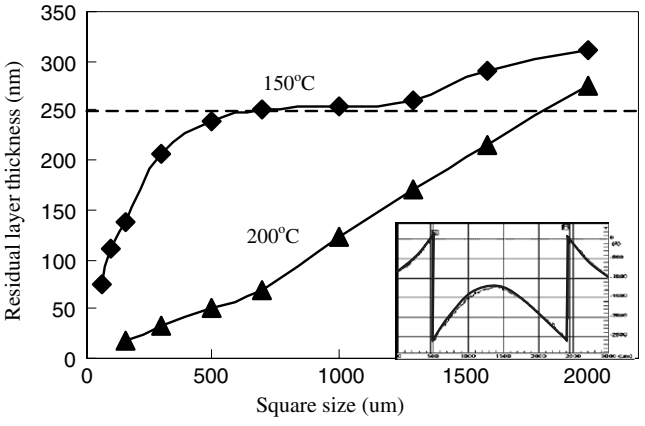

Fig. 4. Residual layer thickness, h_f, as a function of square size for two imprints at 20 bar for 20 min using 12 kg/mol PMMA. h_f is measured by Dektak with a mechanical scratch at or near the square center. Insert shows a typical surface profile across a square (here 2 mm with NIL at 200 °C).

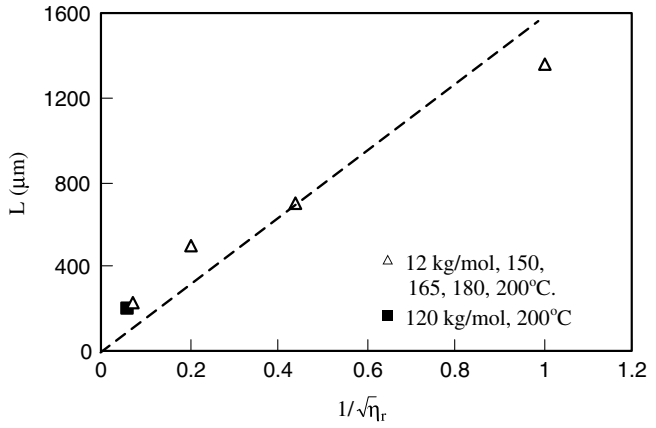

Fig. 5. Dependence of duplicable square size L on $1/\sqrt{\eta_r}$, where η_r is relative viscosity normalized to the viscosity for 12 kg/mol PMMA at 200 °C and varied by changing the imprint temperature or resist molecular weight. NIL 20 bar for 20 min.

Finally, we changed the imprint pressure and time using 12 kg/mol PMMA while keeping the imprint temperature at 165 °C. For residual layer thickness equal to 180 nm, the L vs. $\sqrt{p}$ and L vs. $\sqrt{t}$ curve are shown in Figs. 6 and 7, respectively, which both show a very good agreement with Eq. (1).

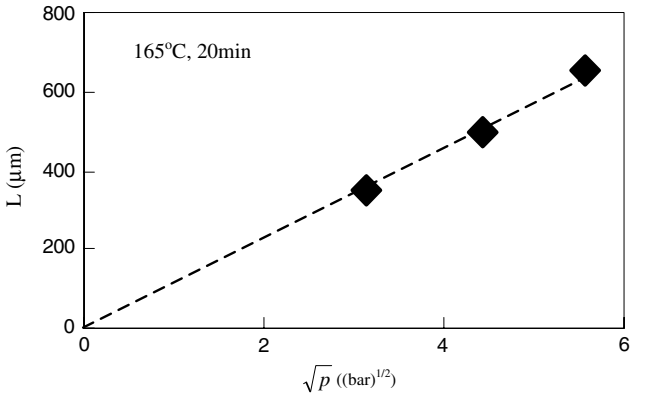

Fig. 6. The duplicable square size L as a function of imprint pressure p, showing $L \propto \sqrt{p}$. Imprint at 165 °C for 20 min using 12 kg/mol PMMA.

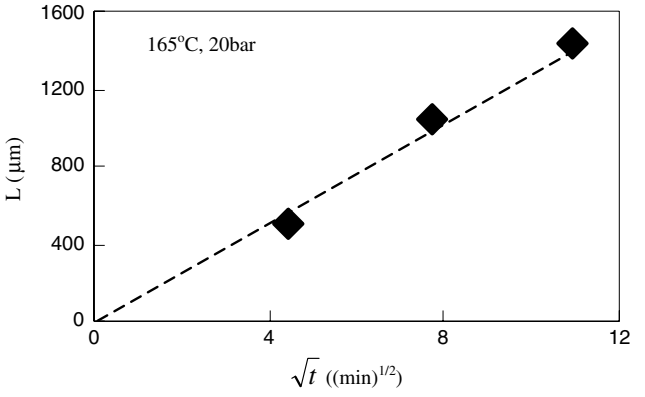

Fig. 7. The duplicable square size L as a function of imprint time t, showing $L \propto \sqrt{t}$. Imprint at 165 °C and 20 bar using 12 kg/mol PMMA.

5. Summary

In this work we investigated the potential of duplicating patterns having feature sizes ranging from 100 nm to 2 mm by thermal NIL. The pattern replication process includes NIL with PMMA as resist, PMMA RIE, metal liftoff and silicon RIE. Using 12 kg/mol PMMA with NIL at 200 °C and 20 bar for 20 min and PMMA etching of 180 nm, we demonstrated simultaneous replication of 1.3 mm squares and 80 nm lines with a line-width increase of 18 nm. We studied systematically the dependence of the maximum duplicable feature size on imprint temperature, pressure and time, which shows good agreement with a simple squeeze flow model.

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[10] In Ref. [6], T_g is taken as 120 °C, 3 °C higher than experimental value, for best fit to the measured viscosity data. It used PMMA with molecular weight 25, 75 and 350 kg/mol. In the present work, T_g is taken as 105 °C for 12 kg/mol PMMA, while keeping other parameters the same as in Ref. [6].