Investigation on the neighbouring effect of filling micro- and nanoscale cavities in ultraviolet nanoimprint lithography

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Micro-/nanoscale patterns are popular in many applications, and several techniques to transfer multiscale patterns have been developed. Ultraviolet nanoimprint lithography (UV-NIL) is the most promising method to quickly produce complex structures, and there is a need to understand the resist-flowing and cavity-filling processes because partial cavity filling may cause defects. One crucial element of a stamp is the layout of the multiscale cavities. They can induce some difficulties during the filling process. Five different stamps with various micro-/nanoscale features (one nano feature, one micro feature, one nano feature, and one micro feature, two nano features and one micro feature) were employed in this simulation study. The imprint results show that the neighbouring effect has major issues in the microscale cavity filling and minor issues in the nanoscale. The layout of complex cavities results in poor filling proportions, and longer imprint times and a symmetrical layout were needed to improve the imprint quality. The contact pressure between the stamp and the resist generally declined when the location approached the centre from both sides of the stamp. The overall physical performance of neighbouring micro-/nanoscale cavities was obtained, and this is essential for further understanding and improvements in the UV-NIL process.

1. Introduction: Conventional optical lithography suffers from light diffraction and high cost. Nanoimprint lithography (NIL) is now a focus of some large integrated circuit companies and research centres because it offers convenient and efficient manufacturing. It has two main variants – thermal NIL and the ultraviolet NIL (UV-NIL). Both have good prospects to produce advanced devices of high quality that might replace optical lithography.

NIL has been widely applied in various fields, including optical and biological products. A nanoscale convex lens array was fabricated with the nanoimprint [1], and this plays an important role in light-emitting diodes and microelectromechanical systems. The efficiency of solar cells can also be improved by a layer of nanoimprinted texture [2]. Plasmonic nanostructures are also very popular in biological sensing, and UV-NIL makes this easily accessible [3].

This cost-effective technique is different from focused ion beam lithography [4] or electron beam lithography [5]. A special combination of UV-transparent stamps and low-viscosity polymers is common in the UV-NIL process. The setting allows this method to quickly replicate micro-/nanoscale patterns at high resolution.

Complex micro-/nanoscale structures have been replicated by processes related to NIL [6, 7]. The stamp design is a crucial element in NIL because the variation in stamp material, stamp thickness, and layout of multiscale cavities can make a difference in imprint quality. Other than the traditional rigid quartz stamp, flexible materials such as polydimethylsiloxane (PDMS) and Teflon have been studied in UV-NIL [8, 9]. Despite the various choices of stamps, the flow behaviour of the resist in UV-NIL has some common rules that should be studied. A group from Toshiba Corporation studied the filling process with a fluid simulation and found that the filling process consists of two phases – the capillary phase and the gas-dissolution phase [10]. Resist deformation modes and filling times were also investigated by Rowland *et al.* These were closely related to the location and the shear rate of the resist. They also proposed an accurate way to predict the deformation and the filling time using three process parameters. They studied the ratio of cavity width to resist thickness, resist filling ratio, and capillary number [11]. This revealed the underlying physics of resist flow as well as designs of stamps and process parameters that can be efficiently optimised. Although many efforts have been made to study the filling process, there is still a lack of investigation on resist flowing in neighbouring micro-/nanoscale cavities. This study compares the filling behaviours of five different conditions to quantify the neighbouring effects. The influences of residual layer thickness (RLT) and contact pressure are discussed for further understanding of improved stamp designs.

2. Modelling and simulation: Unlike conventional molecular dynamics simulation or finite element method, the simulation platform Simprint Core provided by Simprint Nanotechnologies Ltd is based on contact mechanics, which requires a series of stamp and process parameters to perform calculations using the MatLab core. Taylor *et al.* have proposed a fast simulation method using an analytical function to describe the resist deformation [12–15]. A series of experiments have validated its reliability on simulating both feature-scale and chip-scale NIL. Five stamps with mixed-scale cavities are listed in Fig. 1, and all of these are 3.6 µm wide and 1 µm thick. Several 360×360 matrices were created to describe the surface topography of the stamps. These were made of quartz (Young's modulus: 7.1×10^{10} Pa, Poisson's ratio: 0.17) but with different layouts. Figs. 1*a* and *b* show the cross-sectional profiles of single micro-/nanoscale cavities, where the microscale cavity is 1 µm wide and the nanoscale is 300 nm wide. Fig. 1*c* presents a combination of adjacent micro-/nanoscale features with the same width as in Figs. 1*a* and *b*. The stamp in Fig. 1*d* has an unsymmetrical layout of one 1 µm-wide cavity and two 300 nm-wide cavities while the one in Fig. 1*e* is symmetrical. The height of all cavities is 500 nm.

The process environment setting needs other detailed information on external pressure and capillary force to accurately simulate experiments of UV-NIL. Thus, the selection of resist and substrate material can also make a difference in the simulation results. Here, a UV curable resist PAK-01 (surface tension: 0.0391 N/m) from Toyo Gosei Co., Ltd and a common silicon substrate (Young's modulus: 1.6×10^{11} Pa, Poisson's ratio: 0.27) were used. A layer of resist with an initial thickness of 300 nm was spun on the substrate. The contact angle between the resist and the substrate was

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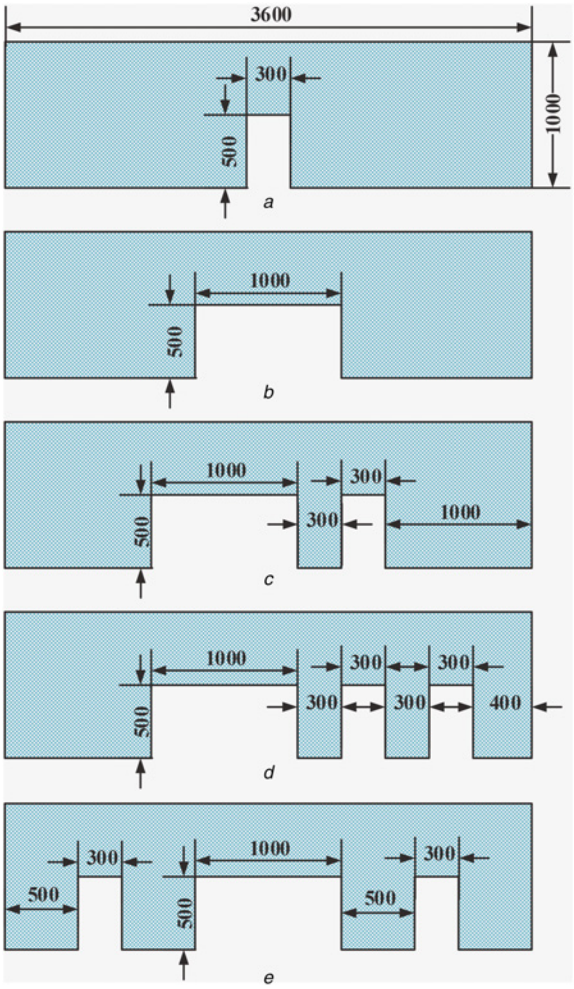

Fig. 1 *Five stamps with different micro-/nanoscale cavities (unit: nm)*
a Single 300 nm-wide cavity
b Single 1 μm-wide cavity
c One 1 μm-wide cavity and one 300 nm-wide cavity
d Unsymmetrical layout of one 1 μm-wide cavity and two 300 nm-wide cavities
e Symmetrical layout of one 1 μm-wide cavity and two 300 nm-wide cavities

30° and that between the stamp and the resist was 85°. The entire process was completed in 3 s under no external pressure. However, the weight of the stamp itself, about 10 Pa, was considered. The final results of cavity filling proportion and RLT distribution are important factors used to measure imprint quality. The contact pressure analysis enables us to develop a deep understanding of the resist's flowing behaviour.

3. Results and discussion: The deformation modes of micro-/nanoscale cavities are illustrated in Fig. 2. Both filling profiles were recorded at 0.3 and 3 s, respectively. Fig. 2 shows that the nanoscale filling process leads to a 'single peak' deformation of resist while the microscale one causes a 'dual peak' deformation. The deformation height of the nanoscale cavity is always larger than the microscale one before achieving the maximum (i.e. 500 nm). This means that the filling process for the nanoscale cavity is relatively faster than the microscale one. The transition of deformation mode had been observed in previous experiments [16–18], which are consistent with our simulation outcomes. Rowland *et al.* [11] proved that the polymer deformation is mainly influenced by the deviatoric stress and the parameter 'directional flow ratio' defined as W/h_i (W stands for half width of the cavity and h_i for initial resist thickness) can accurately predict the deformation mode. Though the threshold value of W/h_i

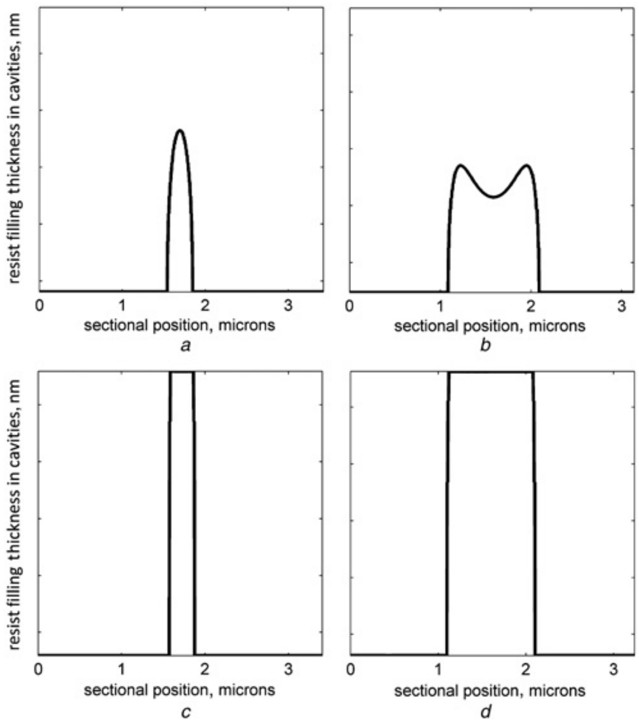

Fig. 2 *Different resist filling modes for microcavities/nanocavities*
a Nanoscale cavity at 0.3 s
b Nanoscale cavity at 3 s
c Microscale cavity at 0.3 s
d Microscale cavity at 3 s

between 'single peak' and 'deal peak' can be different due to the stress relaxation after elastic deformation. Generally, nanoscale cavities have a low ratio of W/h_i that results in a single peak; the high ratio of W/h_i in the microscale system contributes to dual peak deformation.

The cavity filling profiles at four different moments for stamps (c), (d) and (e) in Fig. 1 are presented in Figs. 3–5, respectively. Fig. 3*a* shows that nanoscale filling was faster than microscale filling even when they were adjacent. The left peak of the microscale deformation is about 114 nm higher than the right peak, and this asymmetrical geometry means that the microscale cavity filling was greatly influenced by the neighbouring effect. In contrast, the nanoscale filling was only slightly affected and was slightly asymmetrical. At 0.6 s, the left microscale peak and the nanoscale peak reached the top of the cavities. Meanwhile, the asymmetrical defect was mildly relieved. While there was still a large amount of space filled in the microscale cavity, over 95% of the nanoscale cavity was filled. Figs. 3*c* and *d* show that the neighbouring cavities could be fully filled, and the asymmetrical filling caused by the neighbouring effect was reduced by increasing the process duration.

The complex structure induces some changes in the filling processes in neighbouring cavities. Fig. 4*a* shows that the peak in the central nanoscale cavity was the lowest of the three cavities and slightly inclined to the right side. The left microscale peak was the second highest, but the right one was 154 nm lower. In contrast, the right nanoscale peak reached the top of the cavity. This means that this filling process was the fastest. It also showed that the right nanoscale cavity was the least influenced.

Fig. 4*b* shows that the left microscale peak and the central nanoscale peak also reached the top part of the cavities. The rise in the right microscale peak between 0.3 and 0.6 s was about 38 nm, and this rising trend was slower than the left microscale peak. This proves that the neighbouring effect delays the filling process in cavities. At 1.2 s in Fig. 4*c*, the deficiency was mostly located in

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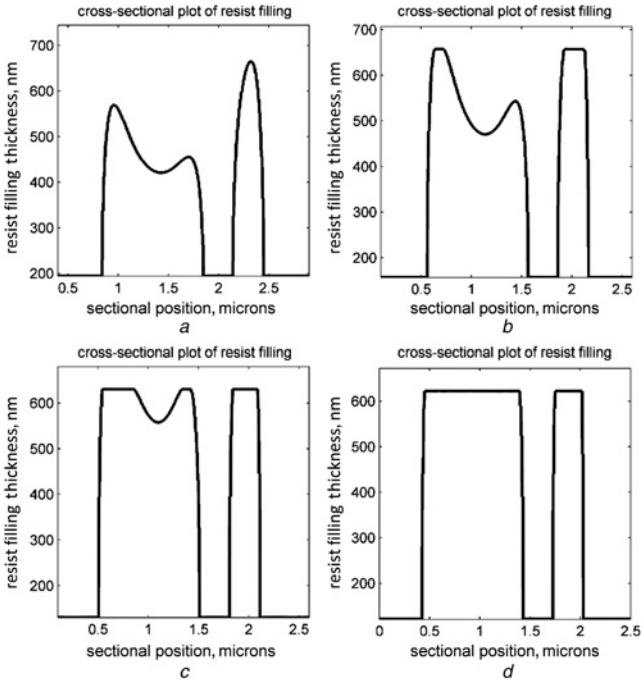

Fig. 3 *Cross-sectional profiles of cavity filling in one microscale and one nanoscale cavities at four moments*
a At 0.3 s
b At 0.6 s
c At 1.2 s
d At 3.0 s

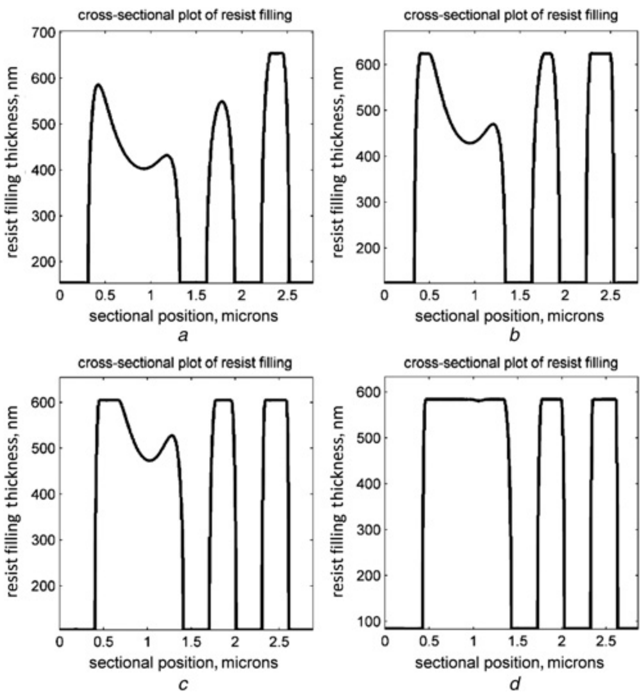

Fig. 4 *Cross-sectional profiles of cavity filling in one microscale and two nanoscale cavities (unsymmetrical layout) at four moments*
a At 0.3 s
b At 0.6 s
c At 1.2 s
d At 3.0 s

the right half of the microscale cavity. The filling proportion of the central nanoscale cavity is moderately smaller than that on the right side. Fig. 4*d* shows that all cavities were roughly filled despite the

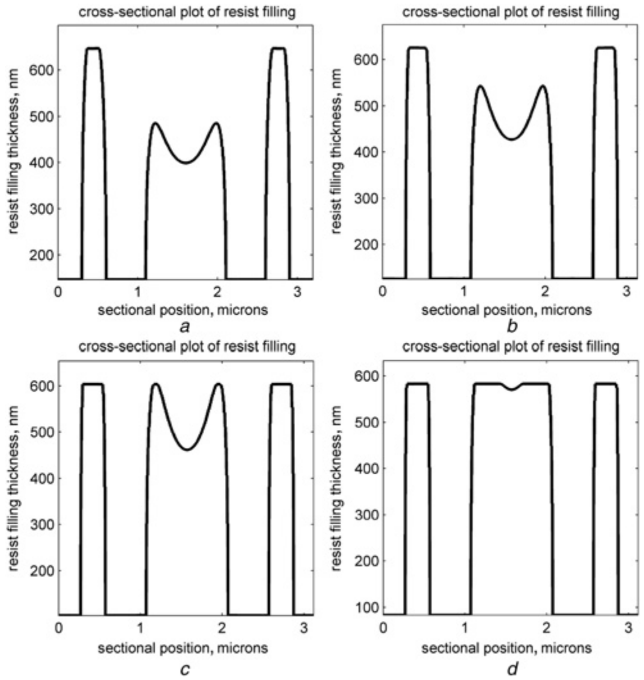

Fig. 5 *Cross-sectional profiles of cavity filling in one microscale and two nanoscale cavities (symmetrical layout) at four moments*
a At 0.3 s
b At 0.6 s
c At 1.2 s
d At 3.0 s

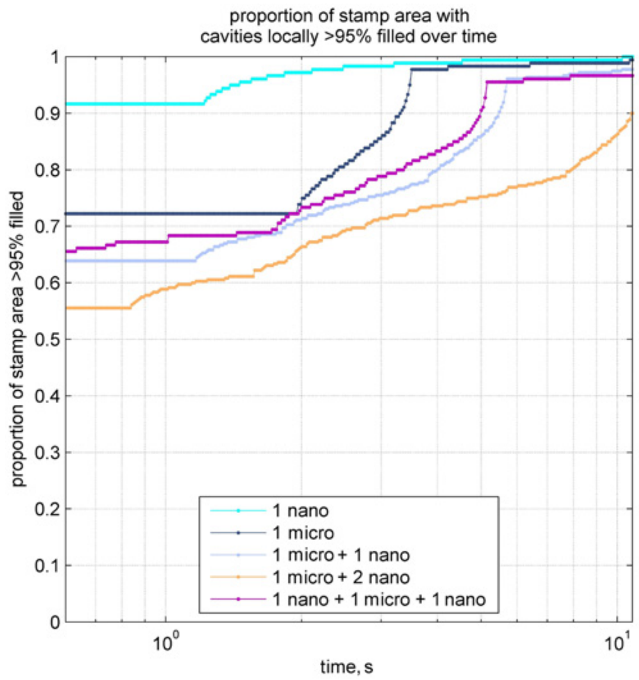

Fig. 6 *Evolution of cavity-filling proportion for the five stamps*

fact that some defects occurred on the adjacent sides of these cavities.

Filling profiles for a symmetrical arrangement of cavities are revealed in Fig. 5. It is illustrated that filling of nanoscale cavities had nearly completed at 0.6 s, however, dual peaks of the microscale cavity were still 68.2 nm away from the top of the cavity. At 1.2 s, microscale filling maintained ‘dual peak’, but only

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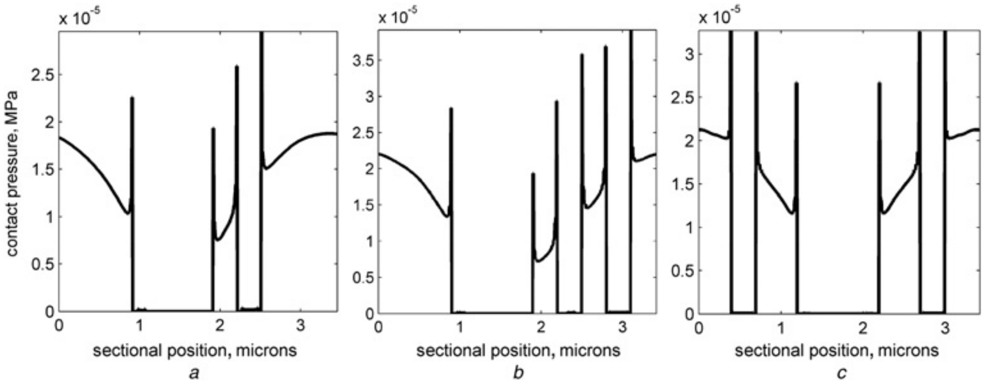

Fig. 7 *Cross-sectional profiles of contact pressure during the filling process for stamps*
a With one micro and one nanocavity
b With one micro and two nanocavities (unsymmetrical)
c With one micro and two nanocavities (symmetrical)

peaks reached the top. Then at 3.0 s, only a small proportion of the microscale cavity remained unfilled at the centre. It is obvious that resist filling keeps symmetrical throughout the whole process.

Figs. 3 and 4 show that the addition of a new nanoscale cavity induces larger differences between dual peaks' heights (from 114 nm in Fig. 3 to 154 nm in Fig. 4) and delays the entire filling process. The nanoscale cavity in the centre was the most strongly influenced by the neighbouring effect. The one on the right was the least affected. This is because neighbouring cavities have to share the resist between them, and the microscale cavities have smaller capillary forces than the nanoscale cavities. Consequently, the major deficiencies occur in the microscale cavities; nanoscale cavities are the least affected. Resist from other places will quickly flow to fill the cavity, and the defects can be relieved. In addition, the contrast between Figs. 4 and 5 demonstrate that the symmetrical arrangement can lead to a symmetrical filling behaviour. Meanwhile, the adverse influence of the neighbouring effect was reduced by placing cavities symmetrically.

Data about the proportion of cavity filling for the five stamps in Fig. 1 is detailed in Fig. 6. There is a stable period before the proportions continuously climb to the final value – only the area over 95% filled is counted. The curve of a single nanocavity is convex, and that of a microcavity is concave. This is due to different filling rates in the filling modes. The final values for the five stamps in Fig. 6 are 100, 98.9, 97.8, 90 and 96.7%, respectively. Though a long duration can improve the filling condition, the addition of neighbouring cavities makes it much more difficult to fully fill all cavities. It is also noted that the symmetrical layout efficiently increased the filling ratio.

Fig. 7 shows the contact pressure distribution of stamps (c), (d) and (e) of Fig. 1 during imprinting. In general, the contact pressure gradually declines from both sides to the centre. This is because the resist at both sides needs a higher pressure to flow into cavities than at the centre. Even between the two cavities, the pressure distribution of the protrusions has a concave shape, and the minimum point is closer to the centre of the stamp. The maximum and minimum values over the entire stamp (namely 0–3.6 μm) in Fig. 7*a* are 7.49 and 29.5 Pa; in Fig. 7*b* they are 7.19 and 39.2 Pa; in Fig. 7*c* they are 11.5 and 32.7 Pa, respectively. This proved that the pressure differences can be enlarged by adding complex neighbouring cavities. However, a symmetrical layout of cavities can bring a symmetrical pressure distribution and reduce the pressure difference, which can extend the life of the stamp.

4. Conclusion: This investigation used simulations with five feature-scale stamps (one nano feature, one micro feature, one nano feature, and one micro feature, and two nano features and one micro feature) to study the flow behaviour of resist, the cavity-filling conditions, and the contact-pressure distribution. The features of 'single peak' for a nano-cavity and 'dual peak' for a micro-cavity were seen while filling the single cavity. The width of the cavity is responsible for the filling behaviour, and its increase can lead to a transfer from 'single peak' to 'dual peak' when keeping the resist thickness constant. In cases of neighbouring cavities, the microscale and nanoscale cavities had interactions on both. Thus, this filling became asymmetrical.

The neighbouring effect had a major negative influence on microscale filling but only a minor influence on the nanoscale filling. The addition of another nanoscale cavity also delayed the filling process and intensified the asymmetrical filling. The conditions of cavity-filling proportions indicate that some filling defects will exist, and a long process duration is necessary if stamps have neighbouring complex structures. The filling processes of single micro-/nanoscale cavities had a convex and a concave curve for filling proportions. Observations of the contact-pressure distribution show that the contact pressure generally fell from both sides to the centre. The addition of cavities resulted in a larger contact pressure difference. However, it was found a symmetrical layout of cavities can be helpful to improve the quality of the imprinting process, namely making it faster and more evenly. This detailed flow behaviour of the resist is essential for stamp and process designs. These findings can lead to some important guidelines that avoid defects caused by neighbouring effects and can consequently offer high-quality imprints.

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