Analysis of von Mises Stress and Deformation of Resists during Demolding Process of Nanoimprinting

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Imprinting quality in thermal imprint lithography is influenced to a large extent by the defects caused by demolding between mold and resists. It is necessary to investigate the demolding behavior of resists in contact detaching process. In this paper, von Mises stress and deformation of poly(methyl methacrylate) resists during demolding process were simulated and analyzed. A model considering adhesion and friction forces was adopted in the simulation of finite element method. The results indicated that stress concentration occurred at two places in the resists. Then stress concentration and deformation in resists were explained by means of simulating the evolution of demolding contact forces. Furthermore, contact forces of key locations were compared and related regularities were proposed. Depending on these regularities, targeted measures can be applied to protect different locations of resists.

Keywords: Nanoimprint, von Mises stress, deformation, contact force, finite element method

1. Introduction

Thermal nanoimprint lithography is an emerging technology with great potential to fabricate micro- and nanoscale patterns in polymer resist with high throughput and at low cost [1]. In thermal nanoimprint lithography, a thermoplastic polymer is heated up above its softening temperature, i.e. glass transition temperature. The soften polymer is pressed into the microstructured cavities. After mold filling, the polymer is cooled down below the glass transition temperature, while maintaining the applied force in order to avoid shrinkage and sinking marks. Finally, the mold is released from the polymer during demolding process [2]. Since the successful demonstration of fabricating various three-dimensional (3D) patterns, nanoimprint lithography has intensively been explored for use in many applications such as electronics, photonic devices, biosensors, and micro-/nano-electromechanical systems [3-4]. Moreover, buildings and automobiles have recently been manufactured using 3D imprint technology, which is based on the nanoimprint lithography.

In spite of the rapid progress, there are still challenges for further development of the thermal nanoimprint lithography [1]. One important challenge is contact detaching in demolding process, where fatal pattern defects are induced by adhesion of the polymer to the mold. For reducing these defects, in-depth understanding of deformation behavior and stress characteristic of imprinted polymer in demolding process is necessary. Furthermore, with the expanding application of thermal nanoimprint lithography, it is imperative to improve imprinting quality through optimizing demolding process.

The deformation and stress distribution of the imprinted polymer can be obtained by numerical simulation, which is an effective way to analyze the demolding process in thermal nanoimprint lithography. There have been several numerical studies on demolding by applying the finite element method (FEM). Song et al. simulated the demolding by using viscoelastic model of FEM [1]. They found that von Mises stress in the polymer layer is highly localized at root and top edge of the polymer microstructure, where deformation and even damage are often found. Worgull et al. studied the moment of occurrence of the stress concentration and deformation [2]. Tang et al. analyzed the demolding stress...
and proposed changing rules for maximum stress of the stress concentration [5]. Guo et al. investigated the forming mechanism of pattern defects caused by demolding [6]. They considered that surface friction force of the nickel (Ni) molds is dramatically reduced by coating anti-adhesion film. He et al. investigated the influence of friction factor on demolding [7-8]. They confirmed the anti-adhesion film contributes to higher-quality patterns. Wang et al. showed that adhesion between mold and polymer is reduced by cooling temperature, with few numbers of pattern defects [9].

Despite much work investigated by aforementioned researchers in revealing the forming of pattern defects, effects of adhesion and friction forces on detaching interface have not been fully studied in these finite element models. Without setting up adhesion and friction forces on the contact interface, the complicated stress state of the polymer in demolding process cannot be comprehensively reflected. Consequently, it is rather difficult to simulate deformation behavior of the polymer realistically.

The purpose of this study is to explore the contact detaching of demolding process by employing an improved FEM model incorporating the adhesion and friction forces. The stress state and deformation behavior of the poly (methyl methacrylate) resists (PMMA) were analyzed in all stages of demolding process. In addition, the developing curves of contact force with demolding displacement were plotted. Based on analysis of these curves, stress concentration and representative deformation were essentially explained in terms of demolding contact force. Eventually, related regularities were proposed from the comparisons of contact forces on different positions in polymer.

2. Method
2.1. Geometry Model

Fig. 1 shows a two-dimensional (2D) FEM model with Ni mold/PMMA resists structure and boundary conditions adopted for simulation. The height (H) as well as the width (W) of polymer microstructures is 200 nm and the aspect ratio of microstructure (H/W) is 1. The interface between the Ni mold and the PMMA resists was defined to be slip-allowed, indicating the contact detaching during demolding is allowed. For accomplishing the detachment, displacement of 200 nm normal to the top surface of the Ni mold was applied. The bottom surface of the PMMA resists is fixed. Horizontal displacement on left and right edges of the whole model is confined, where nodes can only move vertically.

2.2. Material Properties

The polymer resist (PMMA) is much easier to deform than the mold (Ni), as a result, an accurate presentation of mechanical properties of polymer resist is crucial for the demolding simulation. Due to the assumption of incompressibility and isotropy made for PMMA, a Mooney-Rivlin model was used to accommodate the properties of PMMA. In this simulation, the Mooney-Rivlin model parameters, $C_{10}$ and $C_{01}$, were included as PMMA properties considering the actual temperature-dependent behavior.

According to the Mooney-Rivlin model [10-11], stress is given by:

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i}$$

(1)

Where $\lambda$ is the expansion rate and $W$ is a strain density function which is expressed as:

$$W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3)$$

$$I_1 = \lambda_1^3 + \lambda_2^3 + \lambda_3^3$$

$$I_2 = \lambda_1^2 \lambda_2^2 + \lambda_2^2 \lambda_3^2 + \lambda_1^2 \lambda_3^2$$

(2)

Where the $C_{10}$ and $C_{01}$ are Mooney constants, they are derived from the following approximate relations:

$$6(C_{01} + C_{10}) \approx E \cdot C_{01} = 0.25C_{10}$$

$$C_{10} = \frac{4}{30} E \cdot C_{01} = \frac{1}{30} E$$

(3)

(4)

where $E$ refers to an elastic coefficient of the polymer, of
which is assumed to be 3.3 GPa. Then by inserting the value of $E$ into above formulas, $C_{10}$ and $C_{01}$ can be calculated to be 440 MPa and 110 MPa, respectively. However, Ni mold is assumed to be linear elastic material with constant Young’s modulus of 207 GPa and Poisson’s ratio of 0.31.

2.3. Finite Element Model

A finite element analysis software, ANSYS 10.0, was used to simulate the demolding behavior. The plane stress assumption was adopted to simplify the simulation as the perpendicular deformation of polymer resist to the mold surface during demolding is negligible. A 2D four-nodal linear structural element PLANE 42 was used to represent the Ni mold, and a 2D four-nodal hyperelastic structural element PLANE 182 was employed to represent the PMMA resists. A contact element of CONTA 172 was applied for the interface between the Ni mold and the PMMA resists, representing the slip-allowed boundary. The meshing of the finite element model is shown in Fig. 2.

Pattern defects are mainly caused by friction and adhesion generating on the interface between the mold and the polymer. In this simulation, friction and adhesion were respectively applied on the contact interface by means of setting up the friction coefficient and the contact cohesion, in ANSYS program.

3. Results and Discussion

3.1. Analysis of Concentration Stress

From Fig. 3, it can be seen that stress concentrates at two typical locations in PMMA. At the beginning of demolding, the first location is the root of the polymer microstructure—the transition corner zone between the Replicated patterns and the residual polymer. Afterwards,
the second location is the contact region with the releasing mold edge.

3.2. Explanation of Deformation

During the contact detaching process, the polymer resist at the mold/resists interface experiences contact forces (acting along and perpendicular to the interface), which account for stress concentration and structure deformation. Therefore, the contact forces of typical positions should be studied.

As shown in Fig. 4 a), in the initial demolding, the horizontal contact force on Node 3, $H_3$, changes little. The vertical contact force on Node 3, $V_3$, reaches a peak value of 0.7 nN when the demolding displacement is 18 nm. At this moment, $H_3$ approximates to zero. Resultant force $F_3$ has an oblique direction, as indicated in Fig. 4 b).

Simultaneously, the horizontal and vertical contact forces on Node 4, $H_4$ and $V_4$, have respectively peak values of 0.8 nN and 1.6 nN. The direction of resultant force $F_4$ is displaced in Fig. 4 b). These striking contact forces on polymer, $F_3$ and $F_4$, will trigger local high stress and obvious deformation, as shown in Fig. 4 c) and Fig. 4 d).

In the late demolding, the horizontal and vertical contact forces on Node 1, $H_1$ and $V_1$, reach peak values while the demolding displacement is 163 nm, as shown in Fig. 5 a). $H_1$ is 1.1 nN and $V_1$ is 1.3 nN. Resultant force $F_1$ has a declivitous direction, as illustrated by Fig. 5 b). Meanwhile, $H_2$ and $V_2$, also have peak values of 0.9 nN and 1.3 nN, respectively. And their resultant force $F_2$ is represented as well in Fig. 5 b). The appearance of $F_1$ and $F_2$ brings about stress concentration at Node 1 and Node 2. Fig. 5 c) shows the stress concentration in the polymer towards to the end of demolding. As a result, the upper surface of the polymer microstructure has a distinct deformation, which can be seen in Fig. 5 d).

3.3. Regularities of Contact Force

The objective of this section is to summarize regularities of contact force. To this end, the vertical contact forces on four feature nodes of PMMA were compared. Fig. 6 a) shows four feature nodes of PMMA besides position of the demolding centerline. Fig. 6 b) shows contact force curves of these feature nodes. From these curves it can be seen that contact force on Node 3 and Node 4 is noticeably larger than on Node 1.
Fig. 5. Explanation of deformation in the top of PMMA: a) Contact force curves of top nodes, b) Schematic map of contact forces on top nodes, c) Stress concentration in the top of PMMA, d) Deformation in the top of PMMA.

Fig. 6. Regularities of contact force: a) Four feature nodes of PMMA and position of the demolding centerline, b) Contact force curves of four feature nodes.

and Node 2. Therefore, deformation in the root region is more likely to appear than in the top edge. Comparing the contact force values at four different locations from the demolding centerline, the contact force on Node 2 and Node 4 is obviously larger than on Node 1 and Node 3, namely, the closer microstructure approximates to the demolding centerline, the larger its stress and deformation are likely to be.

In actual operation, with the coating of anti-adhesion film such as polytetrafluoroethylene (PTFE) on the mold surface, friction between the mold and the polymer is considerably reduced. In this way, polymer microstructures will be effectively protected. Based on the obtained regularities, the anti-adhesion film PTFE on the
mold in contact with the vulnerable area should be thickened. This method has potential in improving imprinting quality and optimizing the demolding process. In this paper, we only discuss the simple single symmetric structure. However, the FEM simulation for complicated structures remains to be determined. A more practical model with 3D patterns will be reported in near future.

4. Conclusion

The contact detaching of demolding process in thermal nanoimprint lithography was studied in detail by the FEM simulation. The simulation was improved by adopting a model incorporating the adhesion and the friction. Two locations of stress concentration and deformation in the polymer layer during demolding were observed: the root and the upper edge of the polymer microstructure. To explain the typical stress concentration and deformation, developing curves of contact force were plotted. We compared the contact force curves of different locations to get more applicable protection measures.

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