Analysis of Debonding Problem during Demolding Process of Nanoimprinting

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Template release process during nanoimprint lithography was investigated, and the detailed steps of this process were described in order to reduce defects caused by demolding process. The debonding problem was discussed by using optimization of model, numerical simulation and theoretical analysis. The rules and mechanism of debonding were obtained by analyzing the relations between the bonding strength and the vertical stresses of the cementing surface linking the residual layer and the substrate in terms of stress balance theory in fracture mechanics. The location and demolding process which is prone to debond were analyzed. It is concluded that the debonding phenomenon took place at the intermediate position during the second demolding stage, when the adhesion force between the resist and the mold was 0.8 times of other interfaces between the mold and the resist. The debonding phenomenon took place at the angular position during the fourth demolding stage, when the adhesion force between the resist and the mold was 1.2 times bigger than the other interfaces between the mold and the resist.

Keywords: Hot embossing, Demolding, Contact properties, Deformation, Finite element method

1. Introduction

In nanoimprint lithography, one of the most important issues is defects elimination during the template release process [1]. A vast body of research has focus on mechanical characteristics such as the template release load. The stress mechanism of the resist and template and the reasons for defects were analyzed [2-4]. To suppress the release load and eliminate defects, using novel mold with low surface energy materials[5], increased performance of the resist [6], structure modification or novel template release methods have been proposed [7-10].

For embossing process, the deflection of the mold is easily caused by the large embossing force [11]. While the mold cavity will be filled incompletely if the pressure is not large enough [12]. During demolding process, friction force and adhesion force work at the interfaces between resist and mold [13]. And under the function of the demolding force, the resist is stretched and has elastic deformation. Due to the difference of the thermal elastic deformation, there will be lateral strain applied to the polymer pattern during the cooling and demolding process [13-16]. In addition, when the mold is removed with an inclined angle normal to the substrate, as is the case for the peeling and roll-to-roll processes, the resist suffers a lateral force from the template and bending strain is induced [17].

Demolding-related defects can include necking, inclining, overstretched, undercut [18], as shown in Fig. 1. Other defects, reported in the literature, consist of broken mold structures/features, such as fracture, delamination [19, 20] and debond [1]. Adhesion and friction forces significantly affect the pattern transfer. Considering the structure shape, the factors influencing the demolding

Fig. 1. Defects during the demolding process.
process include high-aspect-ratio, duty ratio and residual layer thickness [21].

Many researches mainly focused on the defects except the debonding problem about the mechanical mechanism and optimized method. In this paper, the characteristics of the vertical stress for the interface between the resist and substrate were investigated. The process and location which is prone to debond were studied. And we discussed the influence of the adhesion force between the residual layer and substrate on debonding.

2. Simulation method

The demolding process was numerically simulated based on conventional continuum mechanics and the finite element method to study the stress characteristics of polymer resist and substrate. A two-dimensional model with nickel (Ni) mold/poly (methyl methacrylate) resists (PMMA) structure and boundary conditions adopted for simulation were illustrated schematically in Fig. 2. The substrate was defined as a rigid material. The structural period was 400 nm as well as the height (H) of the embossed microstructure and the width (W) of microstructure was 200 nm. The thickness of the substrate and residual layer was 150 nm and 200 nm respectively. Since the mold had the repetitive structure, only the half-width of the line and space was taken into the analysis. The PMMA resist was fixed on the substrate and a fixed boundary was applied on the substrate. Horizontal displacement on the broadsides was confined, where nodes can only move vertically. Displacement of 200 nm normal to the mold was applied on the top surface of the model.

The polymer resist (PMMA) is much easier to deform than the mold (Ni). Due to the assumption of incompressibility and isotropy made for PMMA, a Mooney-Rivlin model was used to accommodate the properties of PMMA. The mold cavity was assumed to be filled completely. And the basic properties of two materials are shown in Table 1.

3. Results and discussion

3.1. Mechanism analysis about interfacial debonding

First, the debonding will appear when the maximum stress of the interface reaches the critical stress. In the simulation, the critical stress of the interfaces between the mold and the resist is assumed as 110 MPa. The interface bond between the mold and the residual layer was set as 0.8 times of the other three interfaces between the mold and the resist to calculate vertical stress of the interface between substrate and the residual layer in the process of debonding through the numerical simulation software. To facilitate the analysis of the problems, the demolding process was divided into five stages according to the characteristics of stress variation in the embossment of polymer and the sequence of interface crack propagation between mold and the polymer resist from our previous research [22-25]. At the first stage, the load increased linearly with increasing displacement of the mold due to elastic deformation of the residual layer and there was no initial crack on the contact interface between mold and polymer. The process of crack propagation between the mold and residual layer was defined as the second demolding stage. At the third stage, the adhesion force of the vertical contact lost efficacy. And the crack propagated upward along the sidewalls. The process of the third demolding stage being completed to the horizontal interface crack propagation between the embossed structure and mold to be finished was defined as the fourth demolding stage. Then the last stage of the demolding process was the fifth stage that the mold and the embossed structure slid with each other.

| Structure | Material | Young’s modulus (MPa) | Poisson’s ratio | Mooney-Rivlin constants (MPa) | Friction coefficient | Debonding criterion (MPa) |
|-----------|----------|-----------------------|----------------|-------------------------------|---------------------|--------------------------|
| Mold      | Ni       | 207000                | 0.31           | ——                            | 0.45                | 110                      |
| Resist    | PMMA     | 3300                  | 0.35           | C_{II} = 440/C_{II} = 110     |                     |                          |
3.1.1. The stress distribution characteristics and mechanical principles in the first and second demolding stage

The maximum vertical stress distribution curves of the left half interface between the residual layer and substrate in the first and second demolding stage are shown in Fig. 3 respectively. It can be seen from the simulation that each node of the cementing surface achieves the maximum vertical stress of the node at the same time within the first and second demolding stage. From Fig. 3, the stress distribution features can be concluded when the vertical stress of the cementing surface reaches the maximum within the first or second demolding stage. The vertical stress value decreases from the angular node location to the intermediate node during the first demolding stage. When proceed to the second stage, the vertical stress value of each node increases generally, and the decreasing trend of the vertical stress from the angular node to the intermediate node becomes more obvious.

Because the elastic modulus of the embossed structure is small, the stress transferred by the embossed structure to the residual layer is less than by the mold. According to the vertical stress distribution of the structure Fig. 4 corresponded with Fig. 3, it can be concluded that the effect of demolding force on the top of template during the first demolding stage is small with little demolding displacement. Influenced by the embossed structure and the residual layer, the vertical stress distribution curve of the cementing surface is relatively flat. Along with the increase of demolding displacement, the effect of demolding force on the top of mold increases gradually. Because the elastic modulus of embossed structure is smaller than the mold, influenced by the embossed structure, the vertical stress of cementing surface at the bottom of the residual layer gradually decreases from both sides towards the middle area.
3.1.2. The stress distribution characteristics and mechanical principles about the third, fourth and fifth demolding stage

Figure 5 shows the vertical stress distribution curves of the cementing surface when the vertical stress of each node reaches the maximum within the third, fourth and fifth demolding stage. It can be seen from Fig. 5 in each stage of demolding process that the distribution of vertical stress at the cementing surface are presented gradually increases from the angular node to the node in the middle. In the three stages of demolding process, the vertical stress value of the cementing surface for the fourth demolding stage is the largest, and the stress distribution shows more obvious increasing trend from the angular node to the intermediate node of the cementing surface.

Figure 6 shows the vertical stress distribution of each demolding stage when the cementing surface vertical stress reaches the maximum value associated with each curve in Fig. 5. It can be seen from the figure, with the increase of demolding displacement, the demolding force acting on the mold plays a role on the cementing surface by the embossed structure after the separation of the interface between the mold and the residual layer. Because the cross section area of the embossing structure is smaller than the cementing surface area, the distribution of vertical stress on cementing surface shows gradually increasing trend from the angular point position to the middle point as shown in Fig. 5. With increasing the demolding displacement, the internal vertical tensile stress of embossed structure increases gradually until the moment represented by Fig. 6(b). Then, with decreasing the contact interface area between the embossed structure and the mold, the internal vertical tensile stress of embossed structure becomes smaller, and the embossed structure springbacks. The vertical stress of the interface between the substrate and the residual layer has less effect on the cementing surface through the residual layer.

3.1.3. Determination of location and time for debonding

Figure 7 shows the vertical stress distribution curves for the cementing surface when the angular node and the intermediate node reach one’s maximum value. It can be

Fig. 5. The maximum vertical stress distribution curves of the cementing surface within the third, fourth and fifth demolding stage.

Fig. 6. The vertical stress distribution of the structure when vertical stress of the cementing surface reaches the maximum value. (a) The third demolding stage; (b) The fourth demolding stage; (c) The fifth demolding stage.
determined to the fourth stage, the most prone to occur debonding phenomenon, the intermediate node as the debonding position comparing the vertical stress distribution characteristics of the cementing surface for the second and fourth demolding stage. However, if the tensile strength of cementing surface is smaller than the maximum vertical stress value of the angular node for the second demolding process, the demolding phenomenon may occur in the second stage at the angular node of the cementing surface.

3.2. The effect of adhesion force between the residual layer and mold on debonding position

Figure 8(a) shows the maximum vertical stress distribution curves of the cementing surface within each demolding stage when the adhesion force of the interfaces between the resist and the mold is equal with each other. It can be seen that the vertical stress of angular node at the cementing surface reaches the maximum value of all nodes in the whole process of releasing when the demolding process proceeds to the second stage. So the debonding phenomenon will take place at the angular node during the second demolding stage.

Figure 8(b) shows the maximum vertical stress distribution curves of the cementing surface within each demolding stage when adhesion force between the residual layer and the mold is 1.2 times bigger than other interfaces between the resist and the mold. From Fig. 8 and Fig. 9, it can be seen that debonding problem occurs in the second demolding stage at the corner position of the cementing surface in both cases. The vertical stress of the angular node is bigger than that of the intermediate node at the cementing surface, leading to the debonding occurring at the second demolding stage. While when the adhesion force of the interface is equal to the other three interfaces between the resist and the mold, debonding phenomenon will take place at the angular point of the cementing surface between the residual layer and the substrate in the second demolding stage. This is because only the upper horizontal interface of the embossing structure is under the function of external force when proceed to the fourth demolding stage.

Due to the elasticity modulus of the resist is small, the stress distribution trends to be more uniform affected by the embossed structure when the applied force acting on the upper face of the residual layer. Moreover, part of the interface between the residual layer and the mold close to the embossed structure has been cracked when proceeding
to the second demolding stage, leading the stress distribution for the interface being uneven. Compared with the stress of the angular node, the stress of the intermediate node affected by the interface between the mold and residual layer is smaller. So when the adhesion force of the interface is equal to each other for three interfaces between the resist and mold, the maximum vertical stress of the angular node during the second demolding stage is bigger than that of the intermediate node during the fourth stage and easier to become debonding.

4. Conclusion
The impact of the adhesion force between the resist and the mold was investigated computationally. It is concluded that the vertical stress of every node at the cementing surface will reach the maximum at the same time in each demolding stage. This is because the embossed structure design cycle is narrow relatively. Affected by the residual layer, stress of each node at the cement surface in the process of demolding has the same change trend, and reaches the maximum at the same time.

It is found that the location the most prone to debond is the angular or the intermediate node of the cementing surface, the moment the most prone to debond is the time when the crack of the horizontal interface between the mold and the residual layer is propagating. It can be concluded that under the function of the embossed structure the debonding phenomenon occurs first at the cementing surface at the bottom of the embossed mold when the adhesion force between the resist and the mold is equal. While, when the adhesion force between the resist and the mold is 0.8 times of other interfaces, the debonding phenomenon takes place at the intermediate node during the second demolding stage. When the adhesion force between the resist and the mold is 1.2 times bigger than the other interfaces, the debonding phenomenon takes place at the angular node during the fourth demolding stage.

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