

[Review Paper]

Tribology issues in nanoimprint lithography[†]

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Abstract

Nanoimprint lithography (NIL) is one of the most promising technologies for nanofabrication because it can create nano- and micro-scale structures and devices in a cost-effective manner. In the NIL process, a mold with patterns on its surface comes in contact with a polymer film on a substrate. The patterns are transferred to the polymer film and then the mold is separated from the film. Mechanical contact between the mold and the polymer film, and between the film and the substrate, is inevitable. In some cases, during the separation process, adhesion and friction forces at the interfaces can deform and fracture the transferred patterns and detach the polymer film from the substrate. Thus, controlling the adhesion and friction between the materials in contact is very important in achieving a successful pattern transfer and making the NIL process a robust nanofabrication technique. Many theoretical and experimental research efforts have been made to clarify the tribological phenomena in NIL and to reduce defects due to adhesion and friction. This article describes the tribological problems encountered and reviews the related research.

Keywords: Adhesion; Deformation; Fracture; Friction; Nanoimprint lithography

1. Introduction

The manufacture of many types of extremely small devices and structures, such as microelectromechanical systems (MEMS) and nanoelectromechanical systems (NEMS), is possible due to recent developments in micro- and nanofabrication technology. Photolithography is widely used to transfer mask patterns onto silicon substrates and is an important process that determines the size of devices and structures. However, the pattern size in photolithography is limited to the sub-micron range, due to the diffraction and scattering of light. Other techniques, such as electron-beam lithography or scanning-probe-based lithography, can be used for smaller patterns. These methods can be used to transfer patterns less than a few tens of nanometers in width, but they are time-consuming and expensive, due to their sequential processes. Various alternative techniques have been proposed [1, 2] to overcome the shortcomings, such as the limited pattern resolution, low throughput, and high cost of conventional techniques based on MEMS and of semiconductor processes.

Nanoimprint lithography (NIL) is one such nanofabrication technique [3, 4] that operates on a very simple principle. A rigid mold with nanopatterns on its surface is pressed into a

deformable material, typically polymer film, on a rigid substrate. The polymer film deforms according to the mold patterns and retains a negative image of the patterns after the mold is removed. The transferred pattern on the film is then used directly as a functional structure and etch barrier during the subsequent transfer of the pattern onto the underlying substrate.

When it was first proposed in the mid-1990s, NIL techniques were roughly classified as thermal NIL [5] or ultraviolet (UV) NIL [6, 7], depending on the polymer material used in the process. Fig. 1 shows schematic diagrams of both thermal and UV NIL processes. In thermal NIL, the polymer film is usually a thermoplastic polymer, such as poly(methyl-methacrylate) (PMMA), polycarbonate (PC), or polystyrene (PS). The film is heated above its glass-transition temperature T_g before the imprinting process, and the film makes contact with the mold for several minutes under a pressure of several megapascals. The film is then cooled to below T_g and separated from the mold. In UV NIL, a liquid UV curable polymer is dispensed onto a substrate before the imprinting process. The mold then makes contact with the polymer under a comparatively low pressure of less than 1 MPa at room temperature, followed by UV exposure. The mold is separated from the cured polymer after the UV exposure. Patterning with NIL is relatively easy, fast, and inexpensive, due to the simple process. High-resolution patterns with resolu-

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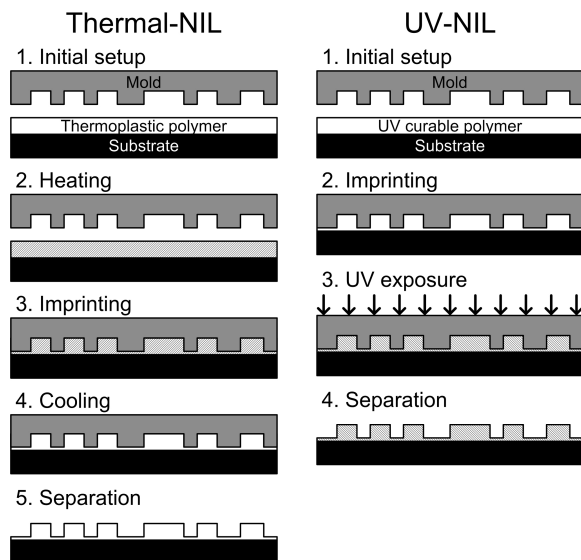


Fig. 1. Schematic diagrams of thermal and UV NIL.

tions of less than 10 nm can be transferred using this technique [8] and numerous electrical and optical devices have been fabricated [9–13]. Refinements of the basic NIL procedure have resulted in a number of different process variants [4].

Adhesion and friction are inevitable when the mold and the polymer film come in contact and are then separated from each other. Adhesion and friction forces significantly affect the pattern transfer. In the separation process, in particular, in which the mold is separated from the polymer, friction is applied to the side wall surfaces of the polymer, and adhesion is applied to the top and bottom surfaces of the pattern. This friction and adhesion can cause deformation and breakdown of the transferred patterns on the polymer. Fig. 2 illustrates the generation of defects during the separation process and shows some examples of defects that can occur. In some cases, the film detaches from the substrate, and the mold pattern is damaged [14, 15]. The fractured polymer pattern left on the mold after separation leads to a serious deficiency in the formation of the pattern on the substrate, and thus requires cleaning the mold before its next use. This cleaning process reduces the throughput of the NIL process and is a significant obstacle to the commercialization of NIL.

Many attempts have been made to reduce the defects caused by adhesion and friction forces. However, tribological problems still exist and threaten the productivity and robustness of the NIL technique. This paper introduces the tribological issues in the traditional thermal and UV NIL processes and reviews efforts to reduce the defects. Section 2 introduces tribological phenomena in NIL and clarifies the origins of the defects. Sections 3 and 4 review various methods for controlling the adhesion energy between contacting materials and the characterization of the interfacial adhesion property, respectively. Section 5 discusses the friction and wear phenomena related to NIL and Section 6 is the summary.

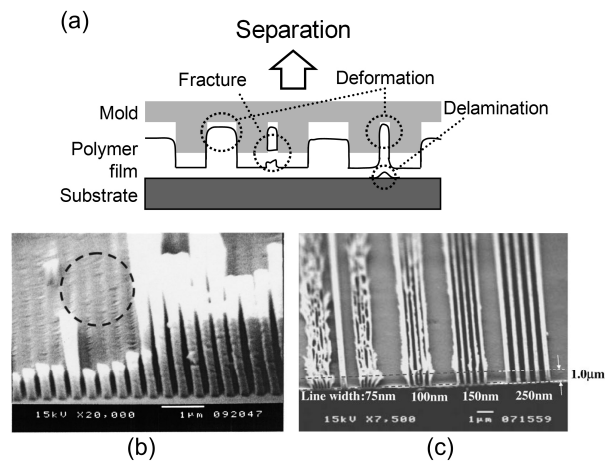


Fig. 2. Defects during the separation process: (a) a schematic diagram of the defect generation, (b) example of fracture defect, and (c) example of deformation defect. Figures (b) and (c) are adapted from [14] and [15], respectively.

2. Origin of defects due to tribological phenomena in NIL

There are three main elements in NIL: the mold, the polymer film onto which patterns are transferred, and the substrate. Fabricating the patterns on the surface of the mold should be simple, but the mold must be sufficiently hard to deform the polymer film during the imprinting process. Mold materials such as Si, SiO₂/Si, quartz, diamond, and Ni meet these requirements and are widely used. Various materials can be used for the substrate, depending on the application and the subsequent process. These include Si, GaAs, SiO₂/Si, and glass because of their well-established compatibility with the micromachining process. Polymer films, such as PMMA, PS, and PC, are widely used for thermal NIL, and laboratory-synthesized curable polymers are used for UV NIL. Many specialized commercial polymers exist that have been designed with various etch resistances and thicknesses.

Two main interfaces are formed in NIL: the interface between the mold and the polymer film and the interface between the polymer film and the substrate. Because the materials for the mold and the substrate have high surface energy, the adhesion strength of the interfaces is sufficiently high that defects occur readily.

Various types of defects are generated, according to forces acting on the pattern. Fig. 3 shows forces acting on a single line pattern during the separation process. The friction force and adhesion force due to the mold act on the pattern concurrently. Additionally, the adhesion force due to the substrate that acts on the bottom of the pattern must be greater than the forces due to the mold to avoid detachment of the polymer film from the substrate. In most cases, the mold has fine micro- and nanopatterns on its surface, so the contact area between the mold and polymer film is significantly greater than that between the polymer film and the substrate, and the friction forces contribute significantly to the increase of the net

be coated on the mold patterns, and nanoscale patterns can be fabricated directly on the DLC surface. A fluorinated DLC mold has low surface energy and is suitable for successful transfer of sub-50-nm patterns [31].

Much research has been conducted on the development of new materials for molds and polymer films that can reduce their mutual adhesion. Polymeric molds with low surface energy and high mechanical strength have been proposed [32–34] and no anti-sticking layer is required in these cases. A polymer film with low surface energy at the surface where it makes contact with the mold has also been developed [35].

Controlling the adhesion is also possible by controlling process conditions, such as the separation velocity of the mold [36] and the process temperature [37]. The adhesion force increases with increasing separation velocity when the viscoelastic dissipation is dominant in the adhesion mechanism of the NIL process. The velocity dependence of the adhesion can be transformed into temperature dependence due to the time-temperature superposition of polymeric materials [38]. Retracting the mold slowly after imprinting is desirable to prevent the detachment of the polymer film from the substrate. A slow retraction decreases the adhesion between the mold and the polymer film, but can also reduce the production throughput of the overall NIL process. The process conditions should be determined carefully, based on the pattern fidelity and process throughput.

4. Characterization of interfacial adhesion

Because adhesion is the main cause of imperfections during pattern formation, the characterization of the adhesion behavior between materials is essential for a successful NIL process. Theory, simulation, and testing to measure the interfacial adhesion have been used to better understand NIL adhesion and to improve the NIL process.

4.1 Theory and simulation

The work of adhesion (W_a) at an interface composed of two materials is equal to the surface generation energy S , and can be expressed in terms of surface energies as

$$W_a = S = \gamma_1 + \gamma_2 - 2\gamma_{12} \quad (1)$$

where γ_i is the surface energy of material i , and γ_{12} is the interfacial surface energy of the interface [39]. This equation is valid for a reversible process, i.e., there is no dissipation of energy during bonding or debonding. In the actual process, energy dissipation always occurs during debonding due to plastic, viscoelastic, and/or viscoplastic deformation. In this case, the work of adhesion is expressed by

$$W_a = S + W_d \quad (2)$$

where W_d is the dissipation energy. For a ductile metal and a polymer, the dissipation energy W_d is much larger than the

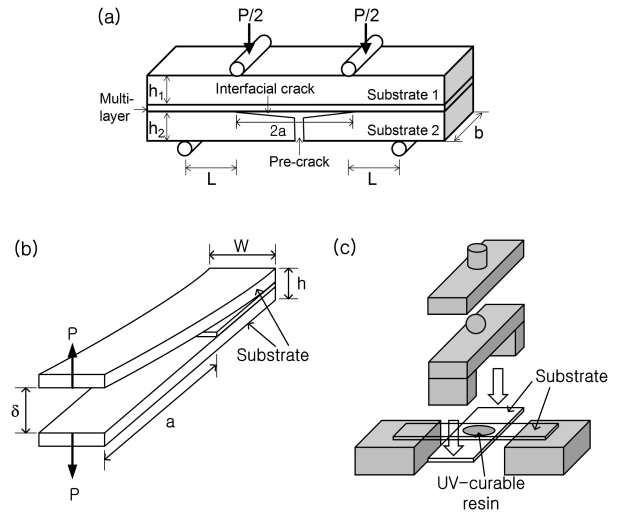


Fig. 5. Examples of testing methods for interfacial adhesion: (a) four-point bending test, (b) double-beam cantilever test, and (c) cross-lap test.

surface generation energy S . The interfacial adhesion can be conveniently quantified using the concept of fracture mechanics in which an interfacial crack is introduced along the interface. The interfacial crack extends when the strain energy release rate G is equal to the work of adhesion. The critical energy release rate for crack extension is referred to as the fracture toughness G_c and is expressed by

$$G_c = W_a \quad (3)$$

The strain energy release rate can be easily estimated for several types of specimens, and some expressions for strain energy release rate are listed in [40]. The adhesion can be readily quantified by evaluating the strain energy release rate at the moment of the crack extension.

The work of adhesion for a given interface can be predicted by a molecular-scale simulation or a quantum mechanical simulation. The latter is the strictest form of adhesion estimation, but is accompanied by a high computational cost. Molecular simulation [41] can be cost-effective for simulating the adhesion, but the potential parameters for the interface are not accurate in many cases and should be determined experimentally or by quantum simulation. The adhesion can be due to electrostatic force, covalent bonding, or van der Waals interactions. Molecular or quantum mechanical simulation can be used to estimate each contribution to provide a deeper understanding of the adhesion. These simulation methods can be useful for estimating the surface generation energy, but have limited capability to estimate the dissipation energy because the time and length scales of the simulation are far different from those of the actual experiment.

The finite element method (FEM) can be used for cost-effective simulation of debonding. Several methods exist for modeling the debonding behavior of the interface. The simulation parameters for the debonding behavior are determined

from the adhesion or fracture test. The NIL process conditions can be simulated by using FEM [14, 42], and the simulation results can provide a good guide to achieving a successful NIL process.

4.2 Test method

Many methods are available for testing adhesion [43]. Some are easy and simple to perform but lead to qualitative results that are only suitable for relative comparisons. Other methods require sophisticated test apparatus and complex sample preparation, but can be used to obtain quantitative adhesion data that can be correlated with the characteristic properties of the interface.

The selection of testing method depends on the information required for the specific NIL application. Measurement of contact angle provides a useful insight into the adhesion property of the interface and is the method that is the most widely used [19–21], although it is no substitute for the direct measurement of the adhesion strength. The interfacial fracture mechanics concept [40] is frequently used for the quantitative measurement of adhesion. Four-point bending test shown in Fig. 5(a) and the double-beam cantilever test shown in Fig. 5(b) are typical tests based on fracture mechanics. The results from these tests give the interfacial toughness, which is the fracture energy per unit area for generation of the delaminated surface. The fracture energy is a characteristic property of the interface that is a function of loading phase angle [40]. The interfacial adhesion energy between polymer resin for UV NIL and mold material coated with various anti-sticking layers has been measured by using these methods [24, 39, 44, 45]. The fracture energy measured with different test methods should be consistent for similar loading phase angles. Simulating interfacial delamination based on finite element analysis and cohesive zone modeling is possible using the interfacial toughness.

Fig. 5(c) illustrates the cross-lap test [23, 35, 44], a qualitative measure. Adhesive force or strength can be measured using this type of test, and the results may be used for relative comparisons. The relative magnitude of the adhesion strength for several types of NIL resin can be obtained with this type of test, although the results cannot be compared with those obtained by other test methods. The stress state around the interface is very complex when delamination occurs, and the stress state is dependent on the testing types and specimen geometry as well as the material combination.

Attempts have been made to measure the adhesion forces between NIL materials directly under NIL process conditions. Atomic force microscopy (AFM) and a microtribometer can be used to investigate the adhesion property of NIL materials under various process conditions. The adhesion force between various anti-sticking layers and UV-curable polymer pairs was measured [46, 47] and the effects of the process conditions, such as temperature, on the adhesion properties were examined [48–50]. An NIL machine can also be used to investigate

the adhesion property under actual NIL conditions [7, 17, 51]. This kind of test provides useful qualitative information for developing the NIL process, but is not cost effective and has the risk of mold contamination. An adhesion tester that could mimic the actual NIL process has been used to measure the adhesion force of the actual NIL environment [52].

5. Friction and wear of NIL materials

The friction and wear of NIL materials are also important. When the mold is separated from the polymer film, the friction force and adhesion force are applied at the interface between the mold and the polymer film, and they can deform and fracture patterns. The friction force can significantly affect the generation of defects in the transfer of high aspect-ratio patterns because the friction force increases in proportion to the pattern sidewall area. Wear of the anti-sticking layer and mold may also occur. The anti-sticking layer degrades gradually through repeated use, so the release properties of the layer become worse, and the adhesion and friction forces between the mold and polymer film significantly increase. Finally, many defects occur in the patterns transferred onto the polymer film, and in severe cases, even the mold patterns may be damaged.

The tribological characteristics of the materials used in the mold, anti-sticking layer, polymer film, and substrate have been examined in detail. However, these results are of little assistance in the NIL process, because they did not consider NIL-specific conditions, such as low contact pressure, low sliding speed, high temperature and heating cycle in thermal NIL, UV exposure in UV NIL, and contacting materials. The friction and wear characteristics of materials are strongly dependent on the test conditions. Thus, the NIL process conditions must be considered to obtain more useful results when testing the tribological characteristics between the contacting materials in NIL.

Some efforts have been made to investigate the tribological characteristics of NIL materials. Friction forces against thermoplastic polymer films [49] and anti-sticking layers coated on mold [51] have been measured using AFM. The tribological behavior of thermoplastic polymer film for thermal NIL and the effect of temperature on the behavior have been investigated [54]. Additionally, an attempt was made to examine the effect of the anti-sticking layer on the damage to the polymer film in a test simulating a thermal NIL process [51, 52].

Characterizing the friction and wear characteristics between the mold and polymer film in NIL presents some obstacles. The contact pressure may vary during the NIL process. In thermal NIL, a thermal mismatch between the mold and the polymer film usually occurs due to the difference in the thermal expansion coefficients of the materials. In UV NIL, most UV curable polymers shrink when cured by UV exposure. Thus, the contact pressure during the separation process depends on the process conditions, and accurate estimation of the contact pressure is impossible. The ambiguous contact

pressure makes the conduct of the friction test and estimation of the tribological characteristics difficult. The inclination and roughness of the sidewall cause another difficulty. The shape and surface roughness of the mold pattern change during the fabrication process and affect the tribological phenomena. If the mold pattern has negative slopes of its cavity sidewall, the polymer film sticks to the mold due to mechanical interlocking. On the other hand, if the mold pattern has a large positive slope, sliding can be eliminated, leaving only the adhesion phenomenon.

Thus, it is clear that friction is one of the causes of defects and contributes significantly to the net adhesion force during the separation process. Additionally, the wear of contacting materials, especially the anti-sticking coating material, is a crucial factor in the life expectancy of the mold under repeated use. The friction and wear characteristics of NIL materials should be examined under NIL conditions. Although some obscure conditions may be required to perform experimental tests, comparing the results relative to one another is possible, and the comparison is useful in developing an optimal process and materials and estimating the life expectancy of the materials.

6. Summary

NIL is a fascinating method of nanofabrication. However, it suffers from a tribological drawback in that the mold must make physical contact with the polymer film to transfer patterns. The adhesion and friction forces generated between the mold and the polymer film can generate defects, such as the deformation, fracture, and delamination of transferred patterns, and the fracture of mold patterns. These defects reduce the chance of successful pattern transfer.

Controlling the adhesion energy between the contacting materials is necessary to avoid generating defects. An anti-sticking layer is commonly used to reduce the surface energy of the mold. Although the use of a SAM as the anti-sticking layer is currently the most popular method, the degradation of the SAM under repeated use remains a problem. DLC coating has been proposed as an alternative anti-sticking layer with high durability and mechanical strength, and low surface energy. Additionally, investigations continue to find new materials with low surface energy for the mold and polymer film. The adhesion energy between the mold and the polymer film must be reduced as much as possible to achieve defect-free pattern transfer.

Understanding the interfacial adhesion is essential for successful NIL, but this is not an easy task because the NIL process includes nanoscale phenomena related to contact mechanics and tribology. Continuum and molecular-based simulations have been applied to the NIL process. The effect of material properties and process conditions on the NIL process can be simulated on the computer, and this can reduce the time and cost required for process optimization. The adhesion characteristics between the mold and the polymer film, or

between the polymer film and the substrate, need to be measured experimentally. The measurement of adhesion improves the understanding of the NIL process and provides an alternative method for controlling it.

Consideration of the friction and wear of NIL materials is also important in achieving reliable pattern transfer. The problem with defects due to the tribological phenomena in NIL still exists even as new materials and process conditions for new applications are developed and tested. The tribological characteristics of the contacting materials in NIL should be investigated more closely through testing that considers actual NIL process conditions. An appropriate test apparatus and test procedure should also be developed. Adequate test results should lead to the design of an optimal process and choice of appropriate materials to achieve successful low-cost, high-speed, and large-area pattern transfer.

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