**In-situ Monitoring of Cavity Filling in Nanoimprints by Capacitance**

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This report deals with the feasibility of carrying out in-situ monitoring of cavity filling in nanoimprints using capacitance measurements with a capacitance circuit built-in on the mold body. A finite element model valid for the numerical description of a parallel-plate capacitor has been developed, and simulations were carried out to predict the influence of cavity filling on capacitance values. On the other hand, in order to measure the continuous variations in capacitance of a parallel-plate capacitor during the course of imprinting, a series of experiments have been performed isothermally, and the capacitance values have been measured at various imprinting stages. The correlation between capacitance values and replicated polymer heights was studied. The preliminary results presented indicate that capacitance measurement is a feasible tool for the monitoring of cavity filling in nanoimprints. [DOI: 10.1143/JJAP.45.5590]

KEYWORDS: nanoimprint, cavity filling, finite element method, capacitance, monitoring

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1. Introduction

With the advancement of nano-electronics in the semiconductor industry, effort has been devoted for more than ten years to the development of reliable non-lithographic techniques with a potential to fabricate nanometer scale patterns. As a consequence, during the past several years, a number of imprint-based alternative lithographic techniques have been investigated and demonstrated on a laboratory scale, namely nanoimprint lithography (NIL), mold-assisted lithography, and microcontact printing.\(^{1-8}\) NIL, first presented by Chou et al.,\(^{7-8}\) has been recognized as one among the promising non-lithographic methods for nano-scale device manufacturing, because of its characteristics such as flexibility, low cost, high resolution, and a wide range of applicability.\(^{6-8}\) However, several bottlenecks still exist, and some fundamental problems remain unsolved.\(^{9}\) Up to now, the nanoimprinting technique has been capable of patterning only fairly small samples, which severely limits the industrial applications of the technique. How to improve the throughput to meet the requirements of large-scale industrial use is the most challenging issue for the research community working on the nanoimprint technique. In addition, since the pattern transfer quality strongly affects the throughput of the NIL process, determining how to ascertain the pattern transfer quality is becoming a priority while trying to find appropriate ways to improve the throughput of the NIL process. The process by which polymer fills the cavities plays a key role in determining the final residual thickness distribution of the polymer layer for imprinting. The slight variations in residual layer thickness are sufficient to cause an inhomogeneous pattern transfer; hence the cavity filling process is vital in pattern transfer and governs the pattern transfer quality as well as the throughput of the NIL process. For these reasons, as efforts are directed towards improving the nanoimprint technique as a reliable and high throughput process for large-scale industrial use, cavity filling must be a particular concern, and in-situ process monitoring in a timely fashion to determine the status of cavity filling is needed to reduce variations in cavity filling as well to enhance the quality of pattern transferring.

Various qualitative investigations have yielded models of the behavior of polymers and mold filling mechanisms in recent years. For instance, Scheer and Schulz\(^{10}\) indicated that two major problems arising with respect to pattern transfer are sticking and mass transport of viscous flow. Heyderman et al.\(^{11}\) reported that imprinting conditions (time, temperature, and pressure) required for replication at the micro-scale also result in good replication at the nano-scale. The key factor for a good and fast molding process is the viscosity of the material, which is largely dependent on the imprinting temperature. An investigation conducted by Scheer and Schulz\(^{12}\) has proven that the flow behavior of the polymer is the limiting factor in the replication of micrometer sized features, but does not adversely affect nanoimprint structure. Schiff et al.\(^{13}\) have explored principles of pattern formation when imprinting a thin molten polymer film and indicated there are two different filling mechanisms: simple flow of polymers from the borders, and formation of polymer mounds. In contrast to qualitative investigations of pattern formation, few studies can be found in the quantitative characterization of cavity filling, while Yu et al.\(^{14,15}\) presented a method that attempted to monitor the nanoimprinting process in real-time and in-situ. The method proposed by Yu et al. can be used for in-situ real-time NIL process characterization and control as well as for studying the effects of different mold features on NIL by time-resolved diffractive scatterometry (TRDS). However, the method is still in elementary development and also suffers because TRDS requires establishing an optical system with a laser beam, making the design of the monitoring system very equipment-dependent and limiting the method to an imprinting system with a transparent imprinting mold. Considering the shortcomings of the real-time in-situ monitoring approach and increasing nanoimprinting throughput by improved pattern transfer quality, a strong need exists to explore quantitative information on cavity filling variations and developing a more promising in-
situ monitoring technique for the nanoimprinting process, which is able to overcome the limitations of the use of a transparent mold for an imprint.

An attempt has been made in this research to extend the recent study\cite{16-17} to take a further step towards exploring the feasibility of implementing an in-situ cavity filling monitoring system that is based on approved and pending patents\cite{18-20} and the capacitance measurement technique for nanoimprinting. Such a technique is used to detect the continuous variations of capacitance of the parallel-plate capacitor formed by the top plate electrode made on an imprint mold and the ground plate electrode on a polymer substrate during the course of an imprint. The variations in capacitance values can be correlated to the status of the patterned polymer between the imprint mold and the substrate at different imprinting stages, and eventually the method will be employed as a timing reference for stopping the imprinting process.

The subsequent sections of this paper present the details of this study and are organized as follows. Section 2 describes the finite element method (FEM) modeling and simulations of the parallel-plate capacitor for varied states of cavity filling. Section 3 describes the specific details about the implementation of the monitoring system for capacitance measurement in-situ to detect cavity filling stages for imprinting operations associated with the experimental setup. Section 4 gives the preliminary results obtained with the experiments and the discussion. Finally, §5 presents the conclusions drawn from this research.

2. Modeling and Analysis of a Parallel-Plate Capacitor

In order to find the appropriate geometry and layout for a detecting capacitor, it is a very important task to predict the effect of the state of cavity filling on the capacitance level and the interference of the electric field between neighboring electrodes. Meanwhile, to reduce the repetitive calculations, the modeling and simulation for a parallel-plate capacitor were done by commercial finite element software ANSYS. The details of the numerical examples are presented as follows.

The dimensions and geometry of the imprint mold and the sensing array electrodes are illustrated respectively in Figs. 1(a) and 1(b) (not drawn to scale). In the model, since all four disc-shaped electrodes on the imprint mold have the same geometry, only one parallel-plate capacitor is considered. The parallel-plate capacitor with disc-shaped top and bottom electrodes as well as a polymer can be approximated with a cylindrical configuration and simplified as a two-dimensional and axially symmetric model. Because the top electrode is made by metal deposition, the metallic film is very thin and can be neglected. As shown in Fig. 1(b), the real area of each top electrode includes the disc-shaped electrode and the connecting metal line; thus, the equivalent diameter for each top electrode can be calculated and equals 6.8 mm. Furthermore, the trench cavities under the top electrode have been assumed to be an equivalent circular cavity with a diameter such that the circle area is equal to the sum of total area of each covered trench. On the other hand, to estimate the electric field coverage of a sensing electrode, the model was constructed by enlarging the top disc-shaped electrode outward in the radial direction. Figure 2(a) shows the cylindrical configuration as well as the two-dimensional and axially symmetric model.

The two-dimensional FEM model of a parallel-plate capacitor, which has a top disc-shaped electrode with an equivalent diameter of 6.8 mm, was generated by ANSYS software considering the axial symmetry shown in Fig. 2(b). \(A_1\), \(A_2\), and \(A_3\) are under the top electrode of the capacitor, while \(A_4\), \(A_5\), and \(A_6\) are the enlarged part out of the coverage of the top electrode. Moreover, \(A_2\) and \(A_3\) are the silicon imprint mold, \(A_4\) and \(A_5\) are the silicon substrate, and \(A_2\) and \(A_3\) represent the polymer. Figure 2(c) shows the meshed finite element model, and the voltages applied to the top and the bottom electrodes are \(V_1 = 5\) V and \(V_0 = 0\) V, respectively.

With the FEM model constructed and the specified relative electric permittivities of silicon for both the imprint mold and the substrate as well as for polymer of \(\varepsilon_{\text{silicon}} = 10\) and \(\varepsilon_{\text{polymer}} = 3.6\), respectively, we have conducted a
Fig. 2. FEM modeling (a) Cylindrical configuration as well as the two-dimensional and axially symmetric model for a parallel-plate capacitor with disc-shaped electrodes. (b) Two-dimensional FEM model for the right-hand side of the parallel-plate capacitor. (c) Meshed finite element model.

Fig. 3. Electric field distribution of the top sensing electrode.

Table 1. Equivalent diameter of the trench pattern covered by the top sensing electrode.

<table>
<thead>
<tr>
<th>Pattern</th>
<th>Equivalent Diameter (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1</td>
<td>4.48</td>
</tr>
<tr>
<td>P2</td>
<td>4.00</td>
</tr>
<tr>
<td>P3</td>
<td>3.20</td>
</tr>
<tr>
<td>P4</td>
<td>2.88</td>
</tr>
</tbody>
</table>

The analysis shows that the effective range of an electric field for the top sensing electrode stretches out about 1 mm from the rim of the sensing electrode. Thus, it is reasonable to draw a preliminary conclusion that the distance separating the rims of two neighboring top disc-shaped sensing electrodes should be more than 1 mm to avoid the interference of the electric field established by the two neighboring top disc-shaped sensing electrodes. According to the simulated electric field distribution, the capacitance sensing array was designed with a 2 mm separation between the rims of two neighboring top disc-shaped sensing electrodes. Moreover, Fig. 4 presents the quantitative variations of the capacitance value against varied cavity fillings for the four different patterns on the imprint mold. From the data shown on the curves in Fig. 4, we found that the variation in cavity filling has significant effects on the value of capacitance. The results obtained also point out that the parallel-plate capacitor is a potential alternative for developing an in-situ monitoring technique for nanoimprinting.

3. Experimental Setup

A setup for performing capacitance measurements has been built. A schematic drawing is shown in Fig. 5, which consists of an imprinting system, an in-situ capacitance detection unit, described in §3.3, to monitor the cavity filling during imprinting, a PLC scanner to switch the detection capacitors, a Keithley 590 C-V analyzer to measure capacitance, and a computer with Windows-based Metrics Technology ICS software to control the measuring sequence and to provide an environment for the collection of data. To prevent the capacitance detection unit from short-circuiting, tapes coated with a Teflon layer were placed on both the backside of the imprinting mold and the surface of the bottom heater and then attached beneath the sample substrate.

3.1 Imprinting system

The NIL experiments have been carried out using the embossing equipment developed by MIRL/ITRI. The maximum size of the wafers and molds imprinted on this equipment is 4-in. in diameter. The imprint force applied by the piston can reach 60 kN with a variance of less than 0.01%. The top and bottom heaters can be regulated at temperatures up to 250 °C with a variance of less than 1% and both the heating and cooling rates range from 60 to 100 °C/min. In this study, the printing process was performed at atmospheric pressure, and the variable parameters...
in all the imprinting experiments were imprint forces ranging from 300 to 1500 N and imprinting temperatures ranging from 60 to 130°C.

3.2 Specimen and imprint mold

The imprint substrate is cut from standard 500-μm-thick, 6-in. silicon wafers. Prior to spin-coating, the substrate is cleaned to remove organic residues. The substrate is first soaked in NH₄OH : H₂O₂ : H₂O (1 : 1 : 5) solution at 75°C for 15 min and then transferred and rinsed in excess deionized water for 5 min to remove the solution. Following the decontamination, the substrate was blown dry using filtered nitrogen. After being blown dry, the substrate was then soft-baked at 170°C for 12 min. In the current work, we used a commercial thermoplastic polymer, mr-1 7030 from Micro Resist Technology GmbH, as the resist material. Its glass transition temperature $T_g$ is 60°C. To produce the silicon substrate samples, the mr-1 7030 solution was spin-coated on silicon substrates 500 nm thick and soft-baked at 140°C for 3 min.

The silicon molds for imprinting experiments were also sliced from standard 500-μm-thick, 6-in. silicon wafers, and the patterns of the imprint molds were fabricated by standard e-beam lithography and reactive ion etching. Meanwhile, in order to investigate the cavity filling situations for four different pattern geometries during the course of nanoimprinting, the imprint mold was designed with four 6.5 × 6.5 mm² square fields of different pattern geometries. The pattern on each of the four fields of an imprint mold consists of trenches with a constant depth of 300 nm and a width of 500 nm. More specifically, one field is the pattern geometry with non-periodic trenches separated by spaces varying by an increment of 500 nm as well as spaces ranging from 500 to 5000 nm, and the other three fields are periodic trenches with spacings of 500, 1000, and 1500 nm. Furthermore, to reduce the sticking problem between the resist and the mold, the imprint molds are coated with a thin anti-sticking layer of 1H,1H,2H,2H-perfluorodecyltrichlorosilane (FDTS), which is commonly used in NIL processes.

3.3 Fabrication of capacitance-detecting electrode

As shown in Fig. 1(b), the top sensing electrodes for parallel-plate capacitors designed for our experiments were fabricated on the backside of the mold surface, and each top sensing electrode was deposited on a location corresponding to one array of the patterns on an imprint mold surface. To reduce the oxidation rate, platinum was selected as the main material for both top sensing electrodes and connecting lines. The process for fabricating the top sensing electrodes is preceded by plasma enhanced chemical vapor deposition (PECVD) to deposit an insulating layer of silicon oxide ($SiO₂$) 1 μm thick on the backside of the mold surface. Then the top sensing electrodes were fabricated by sequentially depositing films of photorecast, titanium, and platinum and
performing the lift-off technique to pattern electrodes on the imprint mold. A 1-μm-thick layer of photoresist was formed by spin-coating and used to protect the silicon oxide layer. A mask with patterns for electrodes 6.8 μm in diameter and connecting lines 1 mm wide has been used to define the shape of Ti and Pt films. Prior to Pt deposition, a 500-Å-thick Ti layer was deposited over the photoresist by sputtering in order to enhance the adhesion of Pt films to the mold. Following Ti deposition, a 3000-Å-thick Pt layer was then deposited using the same procedure as for the Ti layer. Finally, the photoresist was removed by the lift-off process, and the top sensing electrodes and connecting lines were patterned on the backside of mold surface. In this study, the common bottom electrode for the capacitance detection unit was formed with a metal plate, which was placed under the imprint sample (a silicon substrate coated with a layer of polymer). After fabricating all sensing electrodes, shielded signal cables were then attached to the connecting lines of the sensing electrodes at the perimeter of the silicon mold using a special epoxy resin (solidified at high temperature).

3.4 Capacitance Measurements

Before performing the capacitance measurements, the authors conducted a series of imprint tests to confirm that the imprint mold cavities would be completely filled within a reasonably short time, such as 10 s, after starting the imprint under the conditions described. Consequently, a capacitance measurement cycle for one sensing electrode can also be finished at this time.

While doing the capacitance measurements for any one of the four sensing electrodes, the capacitance value of the sensing electrode at imprint starting point is taken as a reference value. A capacitance measurement cycle for one sensing electrode during the 10 s of imprinting consists of a sequence of capacitance measurements, and capacitance values are collected at a frequency of 0.5 Hz; thus, during each sequence, six capacitance values are collected (one every 2 s) and stored. After six capacitance measurements altogether, the cycle is finished for one sensing electrode. The sensing electrode is then shifted via the programmable logic controller (PLC) switch, and the capacitance measurement cycle is repeated until all four sensing electrodes have been run through.

4. Results and Discussion

The quality of the imprint mold is characterized by a series of scanning electron microscopy (SEM) inspections. Figures 6(a)–6(d) show the SEM images of the imprint mold with patterns of the periodic trenches with spacings of 500, 1000, and 1500 nm, and non-periodic trenches, respectively. In studying the monitoring technique for cavity filling during nanoimprinting, it is necessary to design an imprinting process in which acceptable imprinted pattern quality is achieved with specific controllable variables before adopting it as the baseline process for the monitoring experiment. In this study, quite a few imprint test runs were performed by employing the imprint mold and various imprinting conditions of time, temperature, and imprinting pressure. All polymer replicates for each imprinted sample were measured by atomic force microscopy (AFM). Among the series of imprinting tests, imprinting with 20 bar (2 MPa) pressure, 10 s time, and 90 °C temperature provides a very uniform profiles of imprinted patterns and complete cavity filling. Hence, these imprinting conditions have been adopted as the baseline process for the investigation of monitoring feasibility. The average replicated pattern heights have been measured by AFM for all different imprint patterns at various times, and the corresponding cavity filling percentage for all different imprint patterns at various times have been calculated and are listed in Table II. These AFM measurements clearly reveal that the height of the imprinted polymer increases with time and gradually reaches a steady-state height.

The authors have performed capacitance measurements for the four sensing electrodes, denoted S1–S4 with respect to the periodic trenches with spacings of 500, 1000, and 1500 nm, and the non-periodic trenches separated by spaces varying at increments of 500 nm, as well as spaces ranging from 500 to 5000 nm. For each sensing electrode, capacitance measurements were performed at imprinting times of 0, 2, 4, 6, 8, and 10 s. Thus, in total, six capacitance values were obtained for each sensing electrode. In Table III, the capacitance values of a measured sequence for each sensing electrode are shown, and in total 24 capacitance values were collected. Figure 7 shows the experimental results of a change in capacitance values of the sensing electrode with a variation of cavity filling. The measured values are corrected by a uniform difference of 4 pF to accommodate the system-generated noise level. The difference arises from the parasitic capacitance established among the connecting lines, connecting points, silicon imprint mold, and silicon substrate and the specified relative electric permeability of silicon and the polymer. The trend of capacitance change in both the experimental results and simulated results is identical in that the capacitance value changes slightly at the beginning of cavity filling, then increases beyond 60% filling of the cavity up to the full cavity for each sensing electrode. The major stage of cavity filling near the end can be monitored. The results show that the capacitance measurements indeed provide information in-situ that can feasibly tell the cavity filling status during nanoimprinting.

5. Conclusions

This research was focused on exploring the feasibility of implementing capacitance measurements for in-situ monitoring of cavity filling in nanoimprinting. The study included a broad range of areas including numerical modeling as well as simulations of the parallel-plate capacitor for various stages of cavity filling, tuning the imprint process in which acceptable replicated polymer quality is achieved with specific controllable variables, designing a reliable capacitive sensor that can be used in the imprinting environment, finding out both the right materials for sensing electrodes and a suitable surface micromachining process to fabricate the suggested capacitive sensor, and data analysis. The parallel-plate capacitor with a disc-shaped electrode was chosen to detect cavity filling for its simplicity as well as the convenience of manufacturing the plate electrodes on an imprinting mold. In order to predict the effect of the stage of cavity filling on the capacitance range and the interference.
of the electric field of two adjacent capacitors on the optimal design and layout of the detecting capacitors, a finite element model valid for the numerical description of a parallel-plate capacitor has been developed, and simulations were carried out at various stages of cavity filling with ANSYS software. Simulated results show that the change in capacitance is large enough to measure. At the same time, by interpreting the numerical simulations, it has been shown that the parallel-plate capacitor is a potential alternative for developing in-situ monitoring techniques for nanoimprinting.

The authors have proposed a capacitance detection unit and developed the required fabrication method, designed an imprint process as the baseline process for this study, and constructed the experimental setup and performed capacitance measurements at various imprint stages. These preliminary results have demonstrated the feasibility of employing the capacitance measurement technique for monitoring cavity filling in nanoimprinting. We continue to seek a higher signal-to-noise ratio in the system.

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Fig. 7. Experimental capacitance vs cavity filling for various patterns.