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A review of monitoring for nanoimprinting

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ABSTRACT

Purpose: This article provides an overview of the monitoring technique for nanoimprint. Optical and electrical monitoring approaches that have the potential for detecting and monitoring mold deformation and cavity filling in nanoimprint will be reviewed. The major part is devoted to the review of an in-situ mold filling monitoring system based on capacitance measurement for nanoimprinting operations.
Design/methodology/approach: In carrying out the capacitive monitoring method, a broad range of areas including tuning the imprint process, designing a reliable capacitive sensor, finding out the right materials and a suitable surface micromachining process for sensing electrodes, and data analysis have been covered. In addition, to measure the continuous variations in capacitance, a series of imprinting experiments have been performed isothermally, and the capacitance values have also been measured at various imprinting stages.
Findings: The final stage of mold filling near the end, which is of particular interest, can be monitored and the experimental results have demonstrated that the capacitance measurements indeed provide the in-situ information that can tell the mold filling status during nanoimprinting.
Research limitations/implications: The use of neural networks to model the functional relationship between the capacitance value and the rising height of mold filling is recommended as another potential area for future research to realize the ultimate objective of providing online real-time mold filling information for imprinting process control.
Practical implications: Throughout the study, the authors conclude the proposed capacitance measurement technique is a promising approach for monitoring mold filling in nanoimprinting process, and the practical application of the conducted research is feasible with certain improvement of the robustness of the monitoring technique in harsh environment of higher imprinting temperature.
Originality/value: The reviewed monitoring methods are based on the authors' approved and pending patents [22,32,33]. They are considered pioneering works for the research community of nanoimprint techniques and as the basis of making nanoimprint process automated.
Keywords: Nanofabrication; Nanoimprint; Mold deformation; Cavity filling; Capacitance measurement; Monitoring

1. Introduction

1.1. Need for nanoimprint process

With the advancement of the nano-electronics in the semiconductor industry leading to the feature miniaturization, the demand for sub-100-nm feature size in lithography becomes indispensable. This critical need has made photolithography face serious obstacles due to the limitation of wavelength. The alternative nonphotolithographic techniques not limited by the effects of wave diffraction, scattering, and interference in a resist, and backscattering from a substrate have become more prominent in the semiconductor industry. The effort has been devoted to the development of reliable non-photolithographic techniques potential for fabricating nanometer scale patterns since more than ten years. As a consequence, during the past several years, a number of imprint-based alternative lithography techniques have been investigated and demonstrated, namely nanoimprint lithography, mold-assisted lithography, and microcontact printing [1]. Among them nanoimprint, firstly presented by Chou *et al*. [2, 3, 4, 5], has been recognized as one among the promising non-photolithographic methods for nano-scale devices manufacturing, because of its attractive characteristics such as flexible, low cost, high resolution, and a wide range of interesting potential applications [6, 7, 8]. However, several bottlenecks still exist and some fundamental problems remain unsolved [9]. Up to now, the nanoimprint 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 requirement for a large-scale industrial use is the most challenging issue for the research community of the nanoimprint technique. In addition, since the pattern transfer quality strongly affects the throughput of the nanoimprint process, how to ascertain the pattern transfer quality is becoming a priority while trying to find the appropriate way for improving the throughput of the nanoimprint process.

1.2. Process principle of nanoimprint

Three basic components are required for nanoimprint. These are: (1) a predefined rigid stamp with a nano-sized surface relief fabricated by state-of-the-art silicon technology, for example, electron beam lithography and dry etching, if features below 200 nm are needed or, by optical lithography for larger features; (2) the material to be printed, usually a thin film of thermoplastic polymer with suitable glass transition temperature (T_g) and molecular weight, spun onto a substrate; and (3) equipment for printing with adequate control of temperature, pressure and control of parallelism of the stamp and substrate.

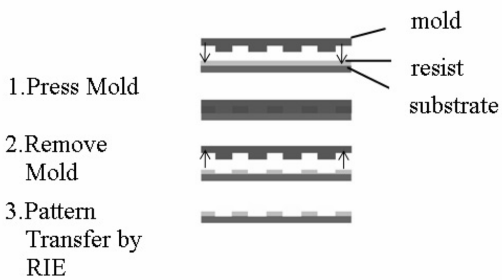


Fig. 1. Schematic diagram of the nanoimprint lithography process

Nanoimprint involves an imprint step with physical deformation of a thin film of thermoplastic polymer cast on a substrate by using a rigid stamp, followed by a pattern transfer step involving the complete removal of the residual polymer in the recessed areas of the pattern. The former imprint step consists of three sub-steps. First, a stamp and a polymer substrate are pre heated some 90-100□ above glass transition temperature (T_g) of the polymer. Then, the stamp is pressed onto the polymer substrate under an applied pressure in the range of about 50-100 bar and hold for minutes, during which the polymer can flow into and fill up the cavities in the stamp. Finally, the stamp and polymer substrate are cooled below the glass transition temperature (T_g) and the stamp is detached from the substrate thereby producing a recessed structure in the polymer layer. The latter pattern transfer step is to use an anisotropic etching process, such as reactive ion etching (RIE), to remove the residual resist layer in the compressed areas to open the windows to the substrate. Fig. 1 illustrates a typical nanoimprint process.

1.3. Achievements and challenges

Nanoimprint has been studied and investigated by several research groups and some significant achievements have been reported since the technique was proposed in 1995. The significant achievements are as follows: (1) Chou and co-workers demonstrated that sub-10 nm features and 40 nm pitch on silicon as well as on gold substrates can be generated over remarkably large areas in 1997 [6,10]; (2) Sun *et al.* devised multilayer resist methods for nanoimprint lithography on nonflat surfaces in 1998 [11]; (3) Heidari *et al.* reported on replication of sub-100 nm resolution patterns over a 6 inch wafer area by nanoimprint lithography in 2000 [12]; (4) In 2002, Bao *et al.* developed a new nanoimprinting technique that allows patterning over a nonflat substrate without the need for planarization and also successfully fabricated multilayer three-dimensional polymer structures using this technique [13]; (5) White and Wood II proposed a novel alignment system for nanoimprint lithography in 2000 [14]. This alignment system possesses alignment marks on an imprint mold and can measure misalignment at a number of sites in the mold/wafer system and bring all of them into registration simultaneously. In addition, Zhang and Chou demonstrated multi-level alignments on four-inch wafers in 2001. The average alignment accuracy is 1µm with a standard deviation 0.4µm in both X and Y directions [15]; (6) Step and flash imprint lithography (SFIL) was demonstrated by Bailey et al. in 2000 [16]; (7) Low temperature nanoimprint using silicon nitride molds was proposed by Alkaisi in 2001 [17]; (8) The pattern transfer fidelity of nanoimprint lithography was investigated by Li *et al.* and reported in 2003, this study showed, compared with conventional photolithography, that nanoimprint lithography has higher resolution and better pattern transfer fidelity with critical dimension controls about four times smaller [18]; and (9) In 2000, a range of thermoplastic and thermosetting polymers were investigated by Schulz *et al.* to optimize the imprinting and subsequent etching steps [19, 20].

While various imprinting processes and applications have been developed to demonstrate the extensibility and application capabilities of nanoimprint, several bottlenecks yet exist [21]. The first of the challenging issues for nanoimprint is the multilevel capability. Zhang and Chou [15] indicated that there are a number of issues that can affect the alignment accuracy. These are relative mechanical shift between the mask and the substrate, relative thermal expansion between the mask and wafer, wafer bending during imprinting, and imprint temperature. At present, the average alignment accuracy is 1µm with a standard deviation 0.4µm in both X and Y [15]. The second issue is throughput. Although a working definition of throughput in nanoimprint as understood in conventional lithography is still lacking, taking the practical situation into consideration, a rough approximation would consider the actual printing time, loading, alignment and separation times, which currently add up to 10-15 min. Factors contributing to throughput include: the stamp size, high density of features, absence of an anti-adhesive layer, polymer curing time, printing temperature and pressure and stamp lifetime. One figure of merit used when discussing nanofabrication techniques is the ‘‘exposure rate’’, which for nanoimprinting is about 0.152 cm^2/s to print the 10 nm features over a 150 mm wafer in 20 min including heating and cooling cycles. The third problem facing nanoimprint is the common standards unavailable at this stage. As any other technology coming under what is generically known as printing, nanoimprint must face up to validation and standards. Validation, or the quality control issue, needs agreement as to what counts as tolerances for a good print, whereas standards depend strongly on design rules and it is too early to expect definitive statements.

1.4. Need for monitoring technique

For nanoimprinting process, there are two-fold aspects important to be monitored. First is about the large-scale production when large mold is to be applied. Since the nanoimprint involves physical contact between the mold and the polymer layer on the substrate surface under applied pressure, the deformation of the mold needs to be measured and controlled in order to avoid the inaccurate pattern transform across the mold area. The mold is of a plate-shape structure thus easily gets deformed subject to imprinting pressure. In the nanoimprint process, it requires a high imprinting speed to achieve a high yield rate. At such a high imprinting speed, the uniformity and precision of imprinted micro and nano scale features and components will be lost when the mold is deformed. If the deformation of the mold is not caught by the production operators in time, a lot of defected products will be produced, and the yield rate suffers.

Secondly, the polymer filling up the cavities is a very important step in the pattern transfer for nanoimprinting. In practice, this mold filling process governs the quality of the final imprint product and plays a key role in determining the productivity of nanoimprinting process. As a consequence, the control of pattern formation requires high precision for nanoimprinting process. The incomplete mold filling has a detrimental effect on the final imprinted pattern dimensions. Once the incomplete mold filling exceeds an acceptable level, it can result in the final transferred pattern dimension going off the specifications and losing the fidelity as well as the uniformity, leading to poor quality of the final imprint products. The pattern formation/mold filling will be particularly concerned when nanoimprint process become an automatically imprint technique for large scale production. Thus, the status of the pattern formation must be monitored to ascertain imprinted patterns with high quality.

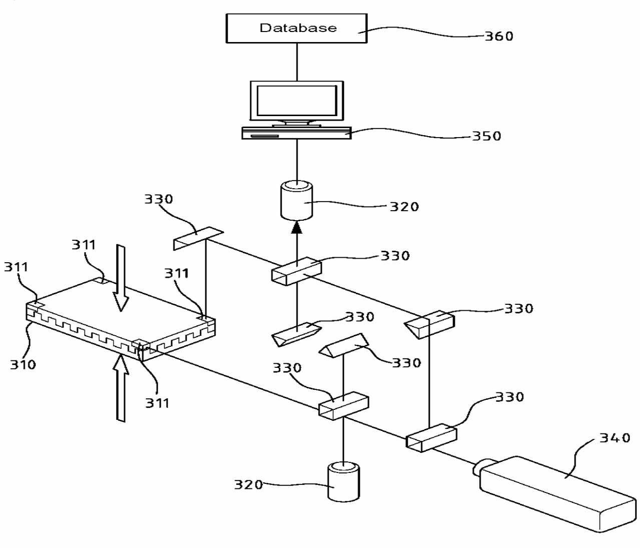
Also, the process of polymer filling up the cavities plays a key role in determining the final residual thickness distribution of the polymer layer for imprinting process. The slight variations of the residual layer thickness are sufficient to cause an inhomogeneous pattern transfer, hence the mold filling process is vital in the pattern transfer and governs the pattern transfer quality as well as the throughput of the nanoimprint process. For these reasons, as the efforts are directed to improve the nanoimprint technique as a reliable and high throughput process for a large-scale industrial use, while the mold filling must be particularly concerned and *in-situ* process monitoring being able to offer timely mold filling process status information is thus needed for reducing mold filling variations as well as for enhanced quality in pattern transferring.

2. Monitoring approaches

2.1. Monitoring for mold deformation

The purpose is to provide real-time accurate measurement of the mold deformation in nanoimprint in order to ensure the high-yield rate automatic manufacturing process. In the nanoimprint process, the mold is deformed due to the subsequent imprinting pressure, repetitive uses, or other external factors. The deformation of the mold also causes the deformation of the marks on the mold body.

a)



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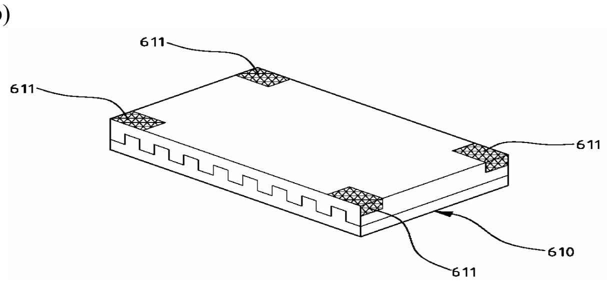

Fig. 2. Optical method for monitoring the mold deformation [22] a-system setup, b-mold with markings

A method is therefore developed to provide in-situ monitoring of mold deformation by application of the following steps: (a) providing a mark on the mold body that is easy to observe in order to monitor the mold deformation, (b) installing a signal source and a monitor device for monitoring the deformation quantity on the mold, (c) transforming the above deformation quantity into computer signals for storing in the database and (d) issuing controlling or warning signals to the imprinting machine based on the processing results of the stored information in the database [22].

Fig.2 illustrates the system setup and the mold with markings. There is a signal source and at least one monitoring device around the mold recording a reference pattern in the database before imprinting. The reference pattern is obtained by using the signal source to emit signals to the monitor marks, and using the monitoring device to receive the interference pattern reflected from the monitoring marks, and the interference pattern is recorded as the reference pattern. By detecting and comparing the interference patterns during the imprinting, one can measure the mold deformation and stop or issue warning if necessary.

2.2. Monitoring for filling mold cavity

Optical method

As to the concerns of mold filling for the nanoimprint process, various qualitative investigations have reported the models and explorations on the behavior of polymers and mold filling mechanism in recent years. Scheer and Schulz [23] indicated that two major problems arising with respect to the pattern transfer are the problem of sticking and the problem of mass transport and viscous flow. Heyderman *et al*. [24] reported that the imprinting conditions (time, temperature and pressure) required for replication at the micro-scale also result in a 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. The investigation conducted by Scheer and Schulz [25] has proven that the flow behavior of the polymer will be the limiting factor in the replication of micrometer sized features, but does not adversely affect nanometer structuring. Schift *et al.* [26] have explored principle of pattern formation during imprint a thin molten polymer film and indicated there are two different types of filling mechanisms: simple flow of polymers from the borders and formation of polymer mounds.

Yu *et al.* [27, 28] presented the method attempted to real-time *in-situ* monitor the nanoimprint process. The method proposed by Yu *et al.* can be used for *in-situ* real-time nanoimprint process characterization and control as well as for studying the effect of different mold features on nanoimprint by the use of a time-resolved diffractive scatterometry (TRDS). In TRDS, a surface relief diffraction grating is used as the imprint mold, and the diffracted light intensity is monitored continuously during the imprint process. Fig. 3 shows a schematic of the TDRS setup used to detect the nanoimprint process. The method requires the establishment of optical system to provide a laser beam, making the design of the monitoring system very equipment-dependent.

The fluorescence microscopy method proposed by Finder *et al.* [30] has potential for integrating with nanoimprint process as an automatic process. It is a quick and easy tool to evaluate the stamp and imprint quality when a fluorencence labelled polymer is used.

However, the fluorescence microscopy method using light image as the judging criteria and the method proposed by Yu *et al.* both require an imprinting system with a transparent imprinting mold, which is often not the optimal for the process.

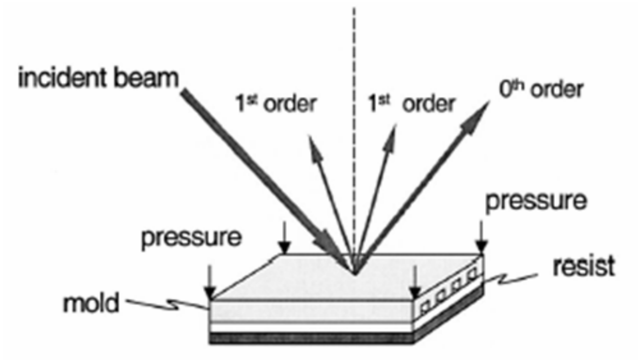

Fig. 3. Schematic of NIL characterization using TRDS [29]

Considering the shortcomings of the above monitoring approaches, a strong need still exists for exploring the quantitative information on mold filling and developing a more promising real-time *in-situ* monitoring technique for a nanoimprint process to overcome the limitation of the use of a transparent imprint mold for an imprint system.

Electrical method

An attempt has been made in this research to continue on the recent study [31] to take a further step towards understanding the mold filling variations at any nanoimprinting stage in constructing a finite element model for simulating the nanoimprint process and to explore the feasibility of implementing an *in-situ* mold filling monitoring system that is based on the concept synthesized by several approved and pending patents [22, 32, 33] and the capacitance measurement technique for a nanoimprint process. Such a capacitance measurement technique would be used to detect the continuous variations of capacitance of the parallel-plate capacitor formed by the top plate electrode made on imprint mold and ground plate electrode on polymer substrate during the course of imprinting. The variations of capacitance values can be correlated to the status of patterned polymer between imprint mold and substrate under different imprint stages, and eventually it will be employed as timing references for stopping the imprint process.

3. Principle of monitoring by capacitance

The parallel-plate capacitor was chosen for mold cavity filling sensing for its simplicity and the convenience of manufacturing the plate electrodes on the imprint mold. The parallel-plate capacitor for this investigation is formed by a top electrode plate, which is one of a plurality of independent metal thin films deposited on the backside surfaces corresponding to the patterned regions of imprint mold, and the bottom electrode plate, which is usually a metal plate used to carry the polymer-coated substrate for a nanoimprint process. The schematic representation of proposed capacitance sensing array for monitoring the mold cavity filling of a nanoimprint process, consisting of four identically disc-shaped electrodes, is shown in Fig. 4.

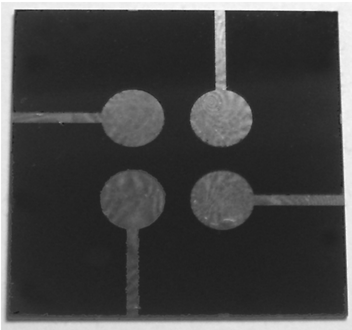

Fig. 4. Photograph of sensing electrodes on the imprint mold unit

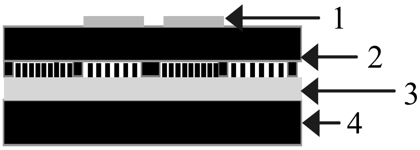

Fig. 5. Schematic of capacitive sensing device, where (1) is the platinum top sensing electrodes; (2) the silicon imprint mold; (3) the polymer layer; and (4) the silicon substrate

The basic principle of using parallel-plate capacitor as a monitoring sensor for cavity filling lies in that the capacitance measurements varied continuously while the gap distance of parallel-plate electrodes and the combinations of different dielectrics which fill the space between the plates of a parallel-plate capacitor were changing during the course of a nanoimprint process. The parallel-plate capacitor for this investigation is formed by a top electrode plate, which is one of a plurality of independent metal thin films deposited on the backside surfaces corresponding to the patterned regions of imprint mold, and the bottom electrode plate, which is usually a metal plate used to carry the polymer-coated substrate for a nanoimprint process. Moreover, the proposed capacitance detection unit can be regarded as three capacitors that are stacked in series. One capacitor corresponds to the silicon substrate, the second to the polymer film, and the third to the silicon imprint mold. Fig. 5 shows cross section of the capacitance.

The expression of theoretical value of capacitance regarding to a parallel-plate capacitor consisting of two conductive plates separated by a dielectric is

$$C = \frac{\varepsilon_0 \varepsilon_r A}{d} \tag{1}$$

where C is the capacitance in Farads, ε_r is relative electric permittivity of dielectric material, ε_0 is the permittivity of vacuum equal to 8.854×10^{-12} F/m, A is the overlapping area of electrode plates, and d is the distance between electrode plates.

In this equation it is assumed that the electric field between the electrodes is homogeneous, and that therefore purely the overlapping area of the electrodes contributes to the capacitance. According to Eq. (1), any change of ε_r, A, and d will cause the capacitance variation of the parallel-plate capacitor. As a result, we can apply the parallel-plate capacitor as a detecting sensor for a nanoimprint process monitoring.

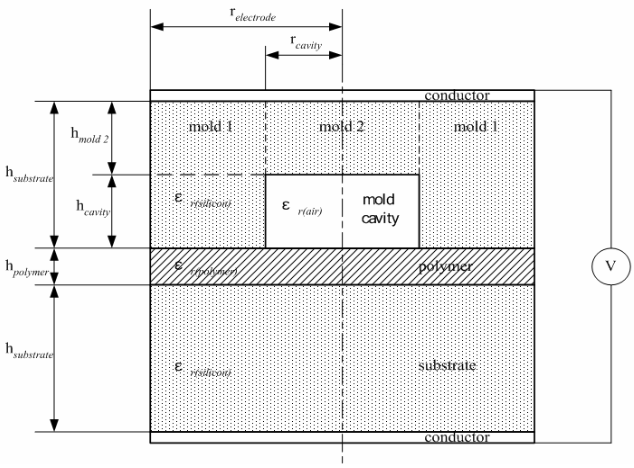

Fig. 6. Simplified equivalent model for the proposed capacitance detection unit

The capacitance for each capacitor in the simplified equivalent model can be expressed as follows.

$$C_{substrate} = \frac{\varepsilon_0 \varepsilon_{r_{silicon}} \pi (r_{electrode})^2}{h_{substrate}} \tag{2}$$

where $\varepsilon_{r_{silicon}}$ is the relative electric permittivity of silicon dielectric, $r_{electrode}$ is the radius of sensing electrode, and $h_{substrate}$ is the thickness of the silicon substrate.

Secondly, the capacitance for the capacitor of polymer film is

$$C_{polymer} = \frac{\varepsilon_0 \varepsilon_{r_{polymer}} \pi (r_{electrode})^2}{h_{polymer} - \Delta h_{polymer}} \tag{3}$$

where $\varepsilon_{r_{polymer}}$ is the relative electric permittivity of polymer dielectric, $r_{electrode}$ is the radius of sensing electrode, $h_{polymer}$ is the original thickness of polymer film, and $\Delta h_{polymer}$ is the corresponding decreased thickness of polymer film resulting from polymer being displaced into mold cavity during imprinting process.

Thirdly, as referred to Fig. 6, the capacitor of imprint mold can be further tackled by considering it to be equivalent to the capacitor of the hollow cylinder of mold 1 and the capacitor of the series combination of mold 2 and mold cavity connected in parallel. The capacitance of mold 1 is

$$C_{mold1} = \frac{\varepsilon_0 \varepsilon_{r_{silicon}} \pi \left[(r_{electrode})^2 - (r_{cavity})^2 \right]}{h_{substrate}} \tag{4}$$

where r_{cavity} is the radius of mold cavity.

The capacitance of mold 2 is

$$C_{mold2} = \frac{\varepsilon_0 \varepsilon_{r_{silicon}} \pi (r_{cavity})^2}{h_{substrate} - h_{cavity}} \tag{5}$$

where h_{cavity} is the depth of the mold cavity.

The capacitor of cavity can also be described as two capacitors stacked in series. One capacitor corresponds to the part of the air in the mold cavity and the second to the polymer filled into the mold cavity. The capacitance of air is

$$C_{air} = \frac{\varepsilon_0 \varepsilon_{r_{air}} \pi (r_{cavity})^2}{h_{cavity} - \Delta h_{cavity}} \tag{6}$$

where Δh_{cavity} is the decreased depth of the mold cavity due to filling polymer into mold cavity.

The capacitance of the polymer filled into the mold cavity is

$$C_{filled-polymer} = \frac{\varepsilon_0 \varepsilon_{r_{air}} \pi (r_{cavity})^2}{\Delta h_{cavity}} \tag{7}$$

Since the capacitor of mold 2, the capacitor of air, and the capacitor of the filled polymer are connected in series, the capacitance of the capacitor of the series combination of mold 2 and mold cavity can be expressed in terms of C_{mold2}, C_{air}, and $C_{filled-polymer}$. The capacitance of the capacitor of the series combination of mold 2 and mold cavity is

$$C_{mold2-polymer-air} = \frac{C_{mold2} C_{filled-polymer} C_{air}}{C_{filled-polymer} C_{air} + C_{mold2} C_{filled-polymer} + C_{mold2} C_{air}} \tag{8}$$

Therefore, the capacitance of the imprint mold is

$$C_{mold-polymer-air} = C_{mold1} + C_{mold2-plymer-air} \tag{9}$$

Finally, the capacitance of the simplified equivalent model for the proposed capacitance detection unit is:

$$C = \frac{C_{substrate} C_{polymer} C_{mold-polymer-air}}{C_{polymer} C_{mold-polymer-air} + C_{substrate} C_{mold-polymer-air} + C_{substrate} C_{polymer}} \tag{10}$$

Additionally, for calculating $C_{polymer}$ with ease, the $\Delta h_{polymer}$ in Eq. (3) can be related to Δh_{cavity} as the following expression:

$$\Delta h_{polymer} = \frac{\Delta h_{cavity} (r_{cavity})^2}{(r_{electrode})^2} \tag{11}$$

The calculation of the theoretical capacitance value for the proposed capacitance detection unit in this study is based on Eq. (10) and the specified relative electric permittivities of air, silicon for both the imprint mold and the substrate, and for polymer of $\varepsilon_{r_{air}} = 1$, $\varepsilon_{r_{silicon}} = 12$, and $\varepsilon_{r_{polymer}} = 3.6$, respectively. Moreover, for the reason of reducing the repetitive calculations work, the calculation has been done by MATLAB for various imprint cavity geometries. Fig. 7 shows the theoretical capacitance value against varied cavity fillings for the four patterns of different spacing period on the imprint mold. The results show that the capacitance of the proposed capacitance detection unit is effectively determined by both the imprint cavity geometry and mold filling status. The theoretical analysis has provided a consolidated rationale for using parallel-plate capacitor as a detecting sensor for a nanoimprint process monitoring.

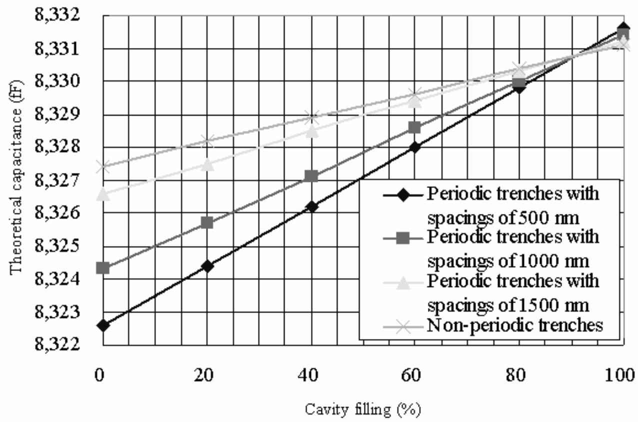

Fig. 7. Correlation between theoretical capacitance and cavity filling for various patterns

In evaluating the feasibility of carrying out *in-situ* monitoring of mold filling in nanoimprints using a built-in capacitance circuit on the mold body, a broad range of areas including tuning the imprint process for acceptable replicated polymer quality, designing a reliable capacitive sensor, finding out the right materials for sensing electrodes, a suitable surface micromachining process for the capacitive sensor, and data analysis have been covered. In addition, in order to measure the continuous variations in capacitance, a series of experiments have been performed isothermally, and the capacitance values have also been measured at various imprinting stages.

A proportional relationship between the experimental capacitance value and the cavity filling percentage has been observed for each sensing electrode. Apparently, the stage of cavity filling near the end can be monitored and, simultaneously, the results have demonstrated that the capacitance measurements indeed provide information *in-situ* that can feasibly tell the cavity filling status during nanoimprinting.

4. Conclusions and future development

In developing a monitoring technique for a specific manufacturing process, the first important step is to decide what quantity is of interest in monitoring. Ideally, this quantity should be the final product quality of interest. However, this may not always be possible. In the nanoimprint process, the final quality of interest is the imprinted pattern dimensions, which does not begin to form until the mold is detached. The authors have found that in addition to the controllable variables including temperature, pressure, and time, the mold deformation and cavity filling are also the consequential variables with significant effect on the pattern formation and transferred pattern quality of imprinting process. Therefore, to produce the imprinted patterns to the required quality, it is essential to monitor the consequential variable of mold filling in imprinting process.

The mold deformation during imprint can be measured by optical signals emitted and reflected from the markings on the mold body. The cavity filling can be monitored by optical and electrical signals. The former requires a transparent mold thus imposes a limitation of this approach.

Since the capacitance varies continuously with the gap distance of parallel-plate electrodes and the dielectric constant of imprinted polymer during the course of a nanoimprint process, the parallel-plate capacitor was proposed as a monitoring sensor for mold cavity filling. In the capacitance measurements, a proportional relationship between the capacitance value and the cavity filling percentage has been observed for each sensing electrode. The proposed capacitance measurement technique is considered a pioneering and promising technique for monitoring the mold filling in nanoimprinting process.

Since all the *in-situ* monitoring methods of mold deformation or cavity filling in nanoimprinting process are still in the early stages, it remains a big challenge to improve the reliability of the monitoring technique. The recommendations for further research are two-fold. The first is to improve the system hardware such as electrical materials and joints, wiring, and signal processing unit. Secondly, the use of neural networks to model the functional relationship between the measured value and the status of mold deformation or cavity filling is feasible. To build an effective empirical neural network model is an interesting area for future research.

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