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# Development of Composite Micro-Patterns on Polymer Film using Repetitive Ultrasonic Imprinting

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*Ultrasonic imprinting is a novel process in which micro-patterns can be replicated on thermoplastic polymer with short processing time and low energy consumption. Ultrasonic imprinting uses ultrasonic vibration energy to soften thermoplastic polymer, and to replicate micro-patterns on the softened polymer surface from a patterned horn or mold. In this study, a new patterning method based on ultrasonic imprinting is proposed to develop composite micro-patterns using a simply-patterned mold. The proposed patterning technology uses repetitive ultrasonic imprinting in which a patterned replica is imprinted repeatedly on a rotated position. To implement this process for the development of composite micro-patterns, two-step repetitive imprinting with 90º rotation was performed using a prism-patterned mold, from which pyramid patterns can be developed. This repetitive imprinting was then further applied to fabricate composite micro-patterns that contain prism and pyramid micropatterns on a single polymer film.*

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## 1. Introduction

These days, the demand for components with micro-features is driving the development of micro-fabrication techniques for polymer products. The micro-molding of thermoplastic polymers is a promising fabrication method for micro/nano devices because several thousand parts can be produced after a mold insert has been fabricated.[1] The most popular micro-molding techniques for thermoplastic polymers are hot embossing and micro-injection molding. The hot embossing process uses a pre-heated mold to form micro-patterns on a thermoplastic polymer substrate.[2] Despite its effectiveness in replicating micro/nano features, hot embossing has a drawback in its long cycle time.[3] Injection molding is advantageous in its high productivity and flexibility, and has recently been applied in the fabrication of micro-features.[4] The challenge in micro-injection molding is to overcome difficulty of filling micro-channels. To this end, rapid mold heating has been applied in order to raise the mold temperature without significantly increasing the cycle time.[5]

Recently, ultrasonic imprinting (or embossing) was developed to replicate micro/nano patterns on thermoplastic polymer using ultrasonic vibration energy.[6-8] Ultrasonic imprinting is an alternative micro-molding process in which micro-patterns can be replicated on thermoplastic polymer with short cycle time and low energy consumption. Ultrasonic imprinting uses ultrasonic vibration energy to soften a thermoplastic polymer, and the micro-scale vibrations cause heat generation due to repetitive deformation and friction on the polymer surface. This process was also applied to the fabrication of superhydrophobic surfaces by forming micro/nano hierarchical structures on thermoplastic polymer substrate.[9,10]

In general, these micro-molding technologies replicate micro-features on polymer surfaces from patterned molds. Therefore, a different mold insert has to be used in order to develop another pattern, because the conventional techniques globally soften the polymer. In this study, a new patterning method based on ultrasonic imprinting is proposed to develop composite micro-patterns out of a simply patterned mold. The proposed patterning method uses repetitive ultrasonic imprinting in which a patterned replica is imprinted repeatedly on a rotated position so that a local softening can be obtained. The feasibility of the proposed approach for fabricating pyramid patterns using a prism-patterned mold is investigated by experimental observation. The effect of the imprinting conditions on the replication quality is also discussed.

## 2. Process Overview

### 2.1 Ultrasonic Imprinting for Micropattern Replication

Ultrasonic imprinting is a micro/nanoscale patterning technology applied to thermoplastic polymer. Ultrasonic vibration energy is used to soften the surface of thermoplastic polymer enough replication to occur. Fig. 1 illustrates a typical ultrasonic imprinting process. A number of micro-patterns (e.g., prism patterns) are engraved on the mold surface. A polymer film is placed between the patterned mold and an ultrasonic horn. Ultrasonic vibration is then transferred from the horn to the polymer film, which causes repetitive deformation and frictional heat on the polymer surfaces. After the polymer surface is sufficiently softened, the micro-patterns can be replicated on the polymer surface. Thus, if the mold patterns are prism-shaped, the negative prism patterns are replicated on the polymer surface, as illustrated in Fig. 1.

### 2.2 Repetitive Ultrasonic Imprinting for Composite Patterning

In this study, a method for repetitive micro-pattern replication based on the ultrasonic imprinting is proposed. Fig. 2 illustrates a configuration of the repetitive imprinting process. In this process, ultrasonic imprinting is performed on a polymer replica that has already been patterned, by giving it a certain amount of rotation angle. For example, a horizontally oriented prism replica can be used for the second imprinting with a 90° rotation angle. Thus, the vertically oriented prism patterns on the rotated polymer replica are repeatedly contacted by the mold surface where the prism patterns are oriented horizontally. This repetitive imprinting results in localized softening of the pattern tips, from which pyramid patterns can be obtained as shown in Fig. 2.

## 3. Results and Discussion

### 3.1 Comparison of the Pattern Replication Characteristics

Ultrasonic imprinting was performed using a prism-patterned mold. The pitch and depth of the prism patterns were 48.6 and 16.5 mm, respectively. Amorphous polyethylene terephthalate (APET) films with 200 $\mu$m thickness were used for the imprinting material. Ultrasonic excitation was induced at a frequency of 19.8 kHz. A rectangular horn was fabricated using AA1050, by applying the optimal design to ensure uniform vibration.[11] A prism-patterned mold was fabricated using a micro-grooving machine. The micro-prism patterns were fabricated to have 48.6 $\mu$m pitch and 16.5 $\mu$m depth.[12] The mold was preheated by circulating water at 45°C, which facilitate the imprinting process by reducing the vibration time to heat the polymer substrate. Imprinting experiments were performed using several vibration times (1.5, 2.5, and 3.5 s). After vibration, 0.6 MPa holding pressure was imposed for a duration of 3.0 s.

Figs. 3(a) through 3(c) show the measured surface profiles of the replicated micro-patterns for 1.5, 2.5, and 3.5 s vibration times. It can be seen that the heights of the replicated patterns increased with increasing vibration time, showing that most prism patterns are well developed along the horizontal direction after 3.5 s imprinting. These replicated samples were then used for the secondary imprinting.

The secondary imprinting was performed using the same mold, just installing the horizontally-oriented replicas with a rotation angle of 90°. Thus, the pattern directions on the first replica changed to the vertical direction, which was perpendicular to the direction of the mold patterns.

Figs. 4(a) through 4(d) show the measured surface profiles of the developed micro-patterns with variations in the secondary imprinting time (0.1, 0.3, 0.5 and 0.7 s). In the case of 0.1 s vibration, the developed micro-patterns show pyramid shapes, of which size in the horizontal direction is larger than that in the vertical direction, as shown in Fig. 4(a). The generation of these horizontal pyramids can be

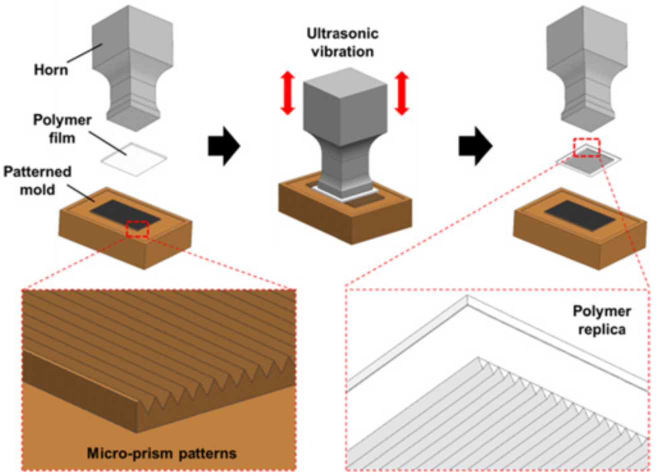

Fig. 1 Configuration of a conventional ultrasonic imprinting process

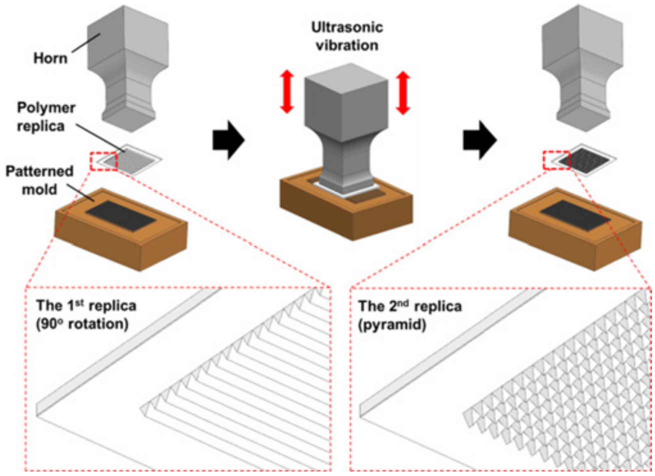

Fig. 2 Configuration of repetitive ultrasonic imprinting on a patterned replica (pyramid pattern development out of prism patterns)

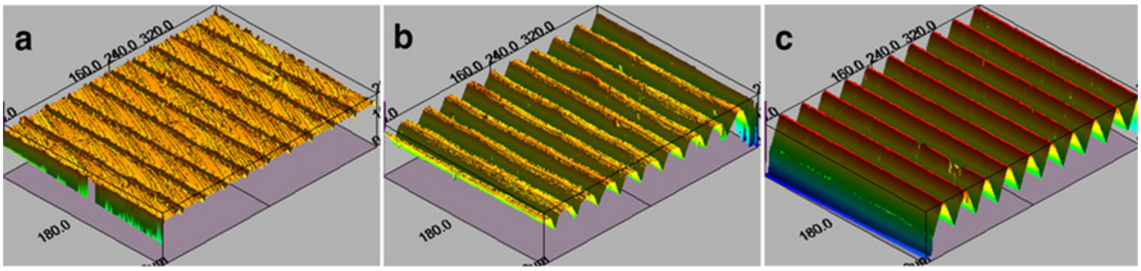

Fig. 3 Comparison of replicated micro-patterns for the 1st imprinting at imprinting times of: (a) 1.5 s, (b) 2.5 s, and (c) 3.5 s

explained by the fact that the secondary imprinting is insufficiently performed; the effect of the second imprinting was not sufficient to fully change the prism patterns into pyramid patterns. In contrast, it can be seen that pyramid patterns were fully developed (Fig. 4(b)) when the vibration time was set to 0.3 s. As shown in Fig. 4(c), however, the 0.5 s imprinting resulted in the generation of vertical pyramids of which vertical size was larger than its horizontal size. This indicates that excessive imprinting time increases the effect of the second imprinting that becomes more dominant than the first imprinting. This excessive imprinting effect became even more severe when the secondary imprinting time was further increased to 0.7 s. In this case, the developed patterns were changed into vertical prism shape, as shown in Fig. 4(d).

For a quantitative comparison, the replication ratios along the horizontal and vertical direction are plotted in Fig. 5, with an increase of the secondary vibration time. It can be seen that the vertical replication ratio improved as the secondary vibration time increased, showing values higher than 90% after 0.4 s imprinting. In contrast, the horizontal replication ratio decreased due to the second imprinting effect, showing that the horizontal patterns had almost disappeared after 0.6 s imprinting. From these results, it can be concluded that moderate vibration time for the second imprinting is required, not only to ensure enough development of the secondary patterns, but also to prevent crushing the first-imprinted patterns. The secondary vibration time was then selected to be 0.3 s, at which both horizontal and vertical replication ratios were higher than 80%.

### 3.2 Functional Evaluations of the Composite Micro-Patterns

To fabricate functional surfaces containing composite micro-patterns, the proposed repetitive imprinting was performed for different patterning areas. APET films with a size of 30 × 30 × 0.2 mm$^3$ were used for ultrasonic imprinting. The primary imprinting was performed on an area of 16 × 16 mm$^2$, using the prism-patterned mold described in Section 3.1. The imprinting conditions were set to be the same as in the previous experiments: 3.5 s vibration time, 0.6 MPa holding pressure, 3.0 s holding time, and 45°C mold temperature. Fig. 6(a) shows an SEM image of the replicated prism pattern, indicating that the prism pattern was successfully replicated on the polymer film.

This first-replica was then rotated 90°, and the secondary imprinting was performed using the same mold with a vibration time of 0.3 s. To develop a composite pattern on a single film, the imprinting area was reduced to 8 × 8 mm$^2$ in the secondary imprinting. Thus, prism and pyramid patterns are able to coexist on a single polymer film. The replicated patterns are illustrated in Fig. 6(b), showing that pyramid patterns were well developed in that region.

To evaluate the functionality of the composite micro-patterns, optical diffusion characteristics were investigated at various locations. Fig. 7 compares images by laser light, observed from behind the patterned film at various locations: pure polymer regions without micro-pattern (A and E); singly patterned regions with horizontal prisms (B and D); and a doubly-patterned region with pyramids (C). In the case of the singly-patterned regions (B and D), light diffusion occurred in the vertical direction due to the horizontal prism patterns. On the other hand, cross-shaped diffusion was observed in the doubly-patterned region (C). These versatile diffusion characteristics are useful in fabricating functional optical components in which different micropatterns can be developed according to their functional requirements.

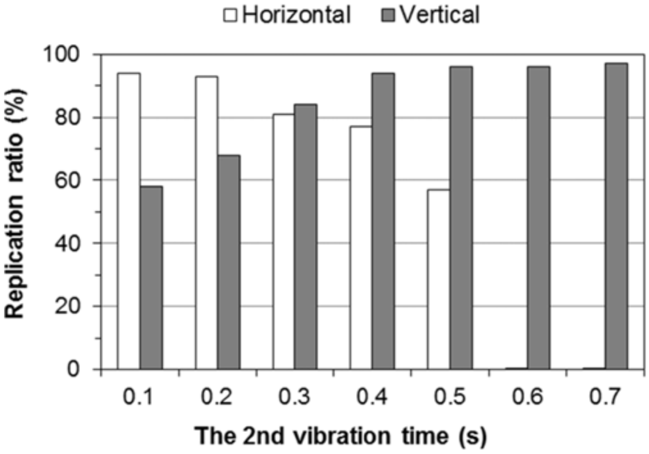

Fig. 5 Comparison of the replication ratios along the horizontal and vertical directions with increasing the secondary vibration time

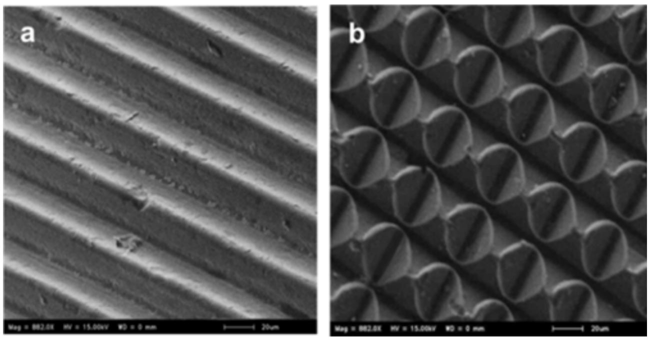

Fig. 6 SEM photographs for: (a) imprints of primary prism-patterns and (b) imprints of secondary pyramid-patterns

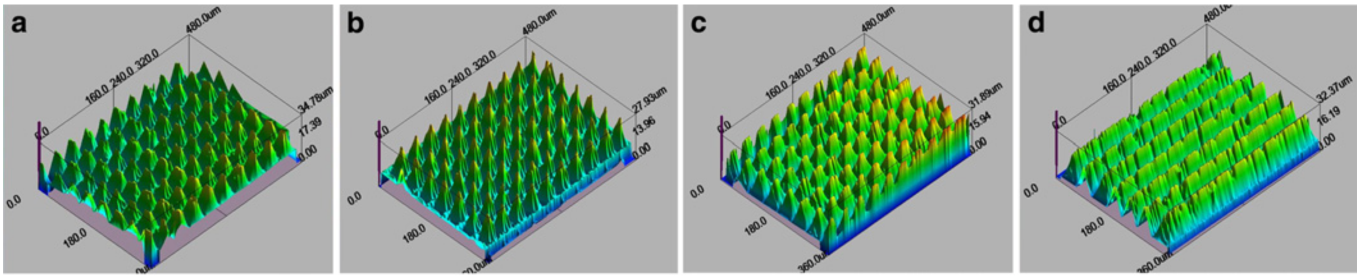

Fig. 4 Comparison of replicated micro-patterns for the 2nd imprinting at imprinting times of: (a) 0.1 s, (b) 0.3 s, (c) 0.5 s and (d) 0.7 s

To investigate the change in surface energy of the different patterns, water contact angle (CA) was measured at various locations. Fig. 8 shows a photograph of water drops at the five different measurement locations (parts A to E in Fig. 7). It can be seen that the water drops in the primary imprinted region show horizontally-oriented oval shapes unlike the other regions, due to the horizontally-developed prism pattern on that region. The CA measurements were performed using a contact angle goniometer (KSV CAM-200). The CA for the pure APET film was 70.9 ± 5.1°, showing a hydrophilic property. In the singly-patterned regions, the CA was 112.2 ± 6.8° from the *x*-direction and 80.7 ± 3.1° from the *y*-direction. This indicates that the surface property of this region was changed into hydrophobic only in the horizontal direction (*x*-direction) due to the horizontally-oriented prism pattern. On the other hand, in the doubly-patterned region, the CA was 94.2 ± 2.8° from the *x*-direction and 102.7 ± 5.1° from the *y*-direction. This indicates that this region was changed into hydrophobic in both directions due to the imprinted pyramid pattern.

## 4. Conclusions

In this study, repetitive ultrasonic imprinting was proposed to develop composite micro-patterns using a simply patterned mold. The proposed repetitive imprinting used a unique mechanism of ultrasonic imprinting: localized softening of the polymer surface. To develop pyramid patterns using a prism-patterned mold, two-step repetitive imprinting was performed. The appropriate vibration time for the secondary imprinting was determined to 0.3 s, not only to ensure sufficient development of the secondary pattern, but also to prevent crushing of the pattern initially imprinted.

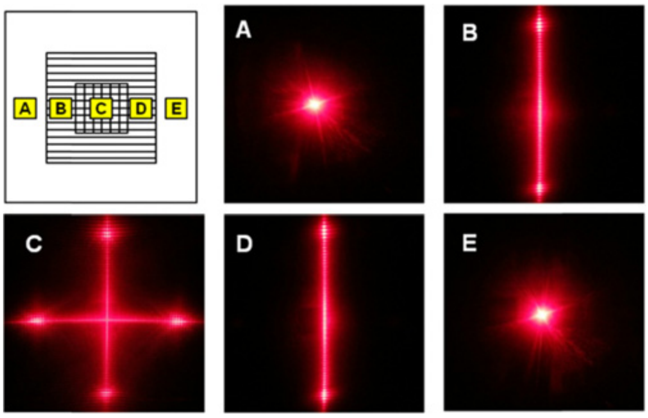

Fig. 7 Images of laser light observed behind the patterned film at various locations

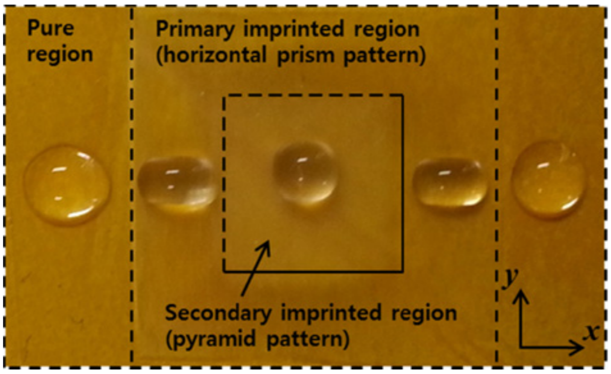

Fig. 8 Comparison of water-drop shapes at various locations on the composite pattern

The proposed repetitive imprinting process was further applied to fabricate composite micro-patterns that contained both prism and pyramid patterns on a single polymer film. For the composite micro-patterns, the coexistence of the versatile patterns could be used to differentiate areas of optical diffusivity and surface energy on a single polymer film, which should be very useful in developing functional surfaces in the future.

## ACKNOWLEDGEMENT

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