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# Microstructure formation via roll-to-roll UV embossing using a flexible mould made from a laminated polymer–copper film

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**Abstract**
Roll-to-roll large format UV embossing processes aim to revolutionize the manufacturing of functional films, with the ability to process a large area at one time, resulting in high throughput and cost reduction. In this paper, we present the experimental results obtained during the process development for roll-to-roll large format UV embossing. Flexible moulds were fabricated from a hybrid film substrate made of a liquid crystal polymer with clad copper foils laminated on both sides of it. The effective pattern area of the fabricated flexible mould was 400 mm × 300 mm with a minimal feature size of 50 $\mu$m. The results show that the roll-to-roll embossing processes are capable of producing micro-scale structures and functional devices over a large area at one time. Large-area roll-to-roll embossing was demonstrated by using the hybrid flexible mould, and micro-features and structures such as micro-channels and dot arrays were replicated on thermoplastic substrates. In addition to its ease and low cost in fabrication, the hybrid flexible moulds demonstrated to have acceptable fidelity and durability. The hybrid flexible mould is a novel solution for large-area embossing.

(Some figures may appear in colour only in the online journal)

## 1. Introduction

To manufacture microscale and nanoscale features, continuous development of photolithography could result in significant cost increases, while scanning beam lithography requires many hours to pattern a small area [1]. Using conventional fabrication techniques would eventually result in massive cost increases and low throughput levels [2]. Therefore, new techniques based on moulding and embossing have emerged as promising alternatives, and nanoimprint lithography is such a technique [3–9].

Micro-embossing is a promising method for forming micro-patterns on thermoplastic substrates [10–12]. To improve the fidelity of embossed features, increasing the process temperature or pressure is one solution. However, this may result in cracks or even breakage of the mould if the mould is made of a brittle material. The embossed thin polymer layer may suffer from the breakage or warpage when it is peeled off from a rigid mould during mould release. A flexible and thin mould can prevent the embossed thin polymer layer from being deformed or warped during mould release.

However, utilizing flat moulds is inappropriate for large-area patterning because the cost of the moulds is very high and there are uniformity and releasing problems in large-scale flat moulding processes [13]. In contrast, roll-to-

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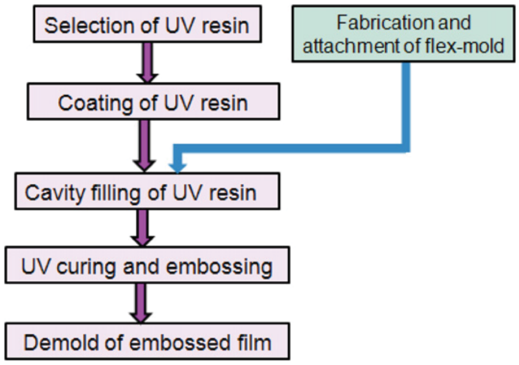


**Figure 1.** The process flow chart for roll-to-roll UV embossing.

roll embossing applies the continuous roll-to-roll process to drastically increase the patterning speed. There is on-going research to develop roll-to-roll embossing and increase its range of applications [6, 7, 14–20]. Roll-to-roll embossing is productive and cost-effective for high-volume fabrication once moulds are fabricated [15, 17]. Roll-to-roll embossing and roller embossing have been used in fabricating components for microfluidic devices [11, 12, 15, 21], textile fibres [22] and micro-optical devices [23].

There are three approaches for mould configuration: (1) patterns are formed directly on an embossing roller surface via precision machining, plating or chemical etching [24, 25], which is a high-cost approach; (2) patterns are fabricated on a thin film by plating or etching, which is overlapped atop a plastic substrate and fed by embossing rollers [26], and (3) patterns are fabricated on a flexible and thin film to form a mould, which is wrapped around an embossing roller [6]. Flexible moulds are frequently used in approaches (2) and (3), and were fabricated via photolithography and Ni plating on thin Ni foils, and are essential for large-area roller embossing [27] and roll-to-roll embossing [6, 17]. One challenge for large-area embossing is how to fabricate a mould with high fidelity at a low cost.

Recently, flexible moulds have gained popularity for large-area imprinting [28–31]. Flexible moulds made up of polycarbonate, polydimethylsiloxane (PDMS) and polyethylene terephthalate films can be fabricated using Ni masters with micro-patterns and thermal nanoimprint equipment [32]. They have found many new applications for life science, biomedicine, solar cells and optoelectronics. For new applications such as e-paper, flexible solar cells, flexible displays and polymeric microfluidic devices, the product sizes become larger and larger. Continuous roller-pressing processes have been currently applied in industrial fields such as flexography printing and gravure printing [33]. A flexible PDMS magnetic mould and an electromagnetic disk-controlled magnetic force can replicate the microstructures onto large-area, curved surface glass, which finds applications in micro-sensory and optical facilities [34]. An approach for soft nanoimprint lithography on nonstandard sample sizes has been proposed, which uses PDMS as a flexible mould material [35]. UV nanoimprint using flexible moulds also enables the fabrication of bio-devices, micro-optics, MEMS and a wide range of nanoelectronic components [36].

A flexible mould fabrication process called pressure-assisted moulding has been reported for high-resolution soft UV nanoimprint lithography [37]. PDMS is used as the flexible mould to duplicate the pattern of a hard mould, and an imprinting material is sprayed onto the PDMS mould to build *SU*-8 microstructures on LED surfaces by UV-assisted roller imprinting [38]. Nanoscale patterning of magnetic islands by imprint lithography using a flexible mould has also been demonstrated for the increasing magnetic recording density towards 100 Gbit cm$^{-2}$ [39].

This work focused on the investigations of the microstructure formation via a roll-to-roll UV embossing process using a flexible mould as a master template. Different from those discussed above, the flexible moulds used in this work were fabricated from a hybrid film substrate made of a liquid crystal polymer with clad copper foils laminated on both sides of it. Micro-patterns on the moulds were formed by etching the copper foils. Characterizations of micro-features on the flexible moulds were performed. Various experiments were carried out to investigate the embossing process using a prototyped roll-to-roll UV embossing machine.

## 2. Experiments

The process flow of roll-to-roll UV embossing is shown in figure 1. After the establishment of a roll-to-roll UV embossing machine, proper flexible films and liquid resins were selected. A flexible mould was attached on the embossing roller, and

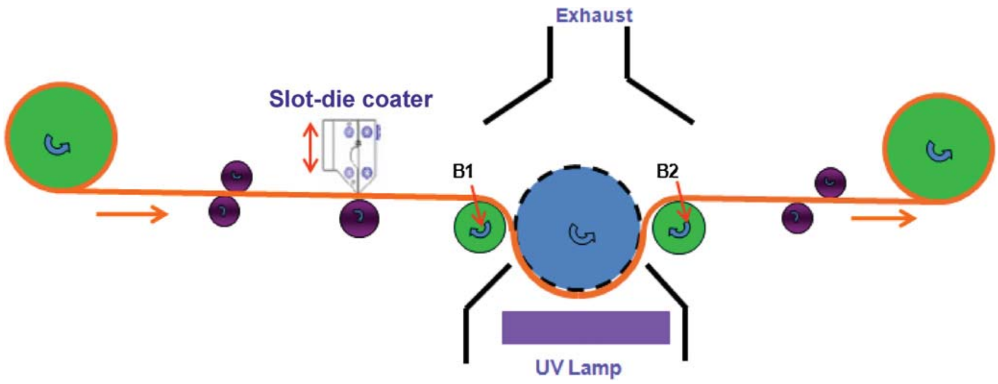


**Figure 2.** Schematic diagram of the UV embossing system.

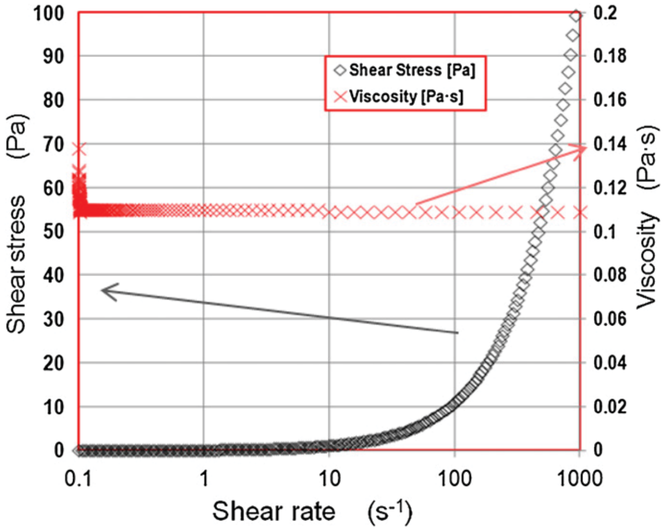


**Figure 3.** The resin viscosity and the shear stress versus shear rate.

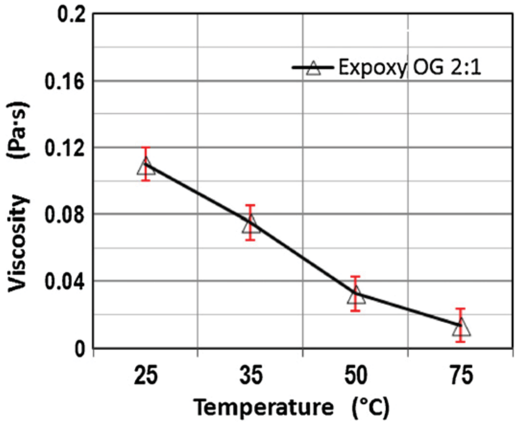
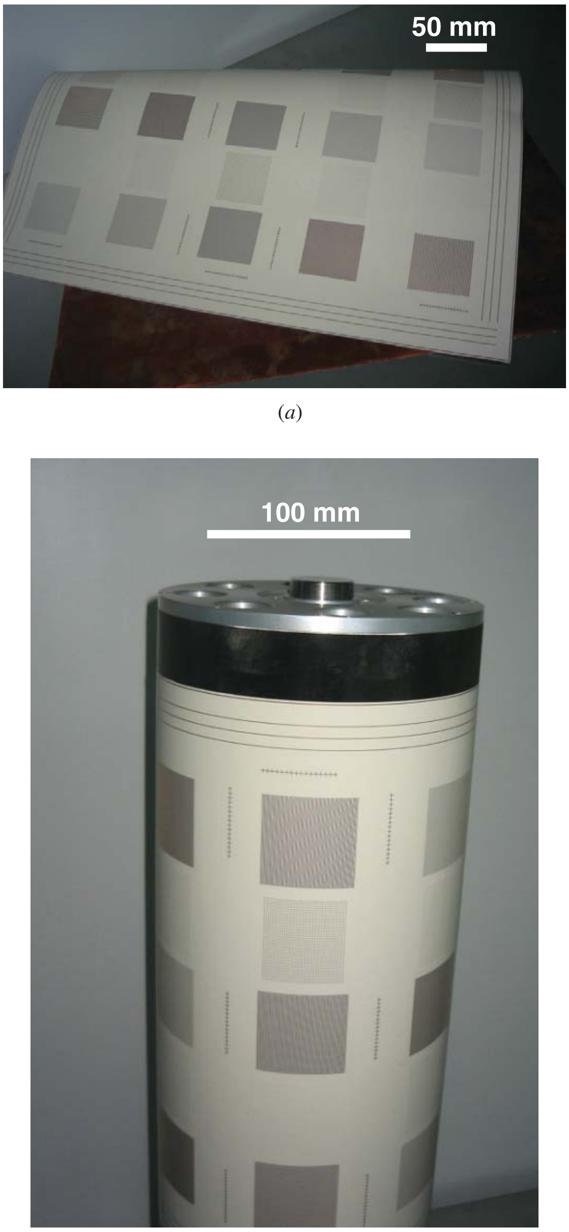


**Figure 4.** The resin viscosity versus the temperature.

roll-to-roll coating of liquid UV curable resin was performed. The pressure between the film and mould, which resulted from the web tension, enabled mould-filling of the resin. Embossing was conducted by UV exposure. The embossed film was then separated from the embossing roller by means of the web tension applied by a rewinding motor. The profiles of the embossed structures were evaluated via off-line measurements by using an optical microscope and a stylus profilometer.

We target to develop roll-to-roll large-area ultraviolet (UV) embossing processes for fabricating micro-features on flexible plastic films. The prototyped roll-to-roll embossing machine used in this work is shown in figure 2. A coating and embossing module is located at the centre of the machine and contains all the components required for the photosensitive resin coating on the flexible film substrate, the embossing and the curing through UV light exposure. The key components in this model include a slot die coater, an embossing roller and a UV lamp. By using different flexible moulds, various structures can be fabricated by roll-to-roll UV embossing. A web handling module is located at the two ends of the machine, drives the flexible film substrate, controls its web-speed and provides tension to the film substrate in the coating and embossing processes. It consists of a de-reeler station and a reeler station. A raw film roll is installed on a motorized roller at the de-reeler station, which feeds the raw film substrate into the coating and embossing module. After the coating, embossing and curing processes are completed; another motorized roller at the reeler station receives the film substrate containing the embossed features, and rolls it back into a roll so that it can be removed later by the machine operator. The web-speed of the film substrate is determined by the rotational speed of the reeler roller.

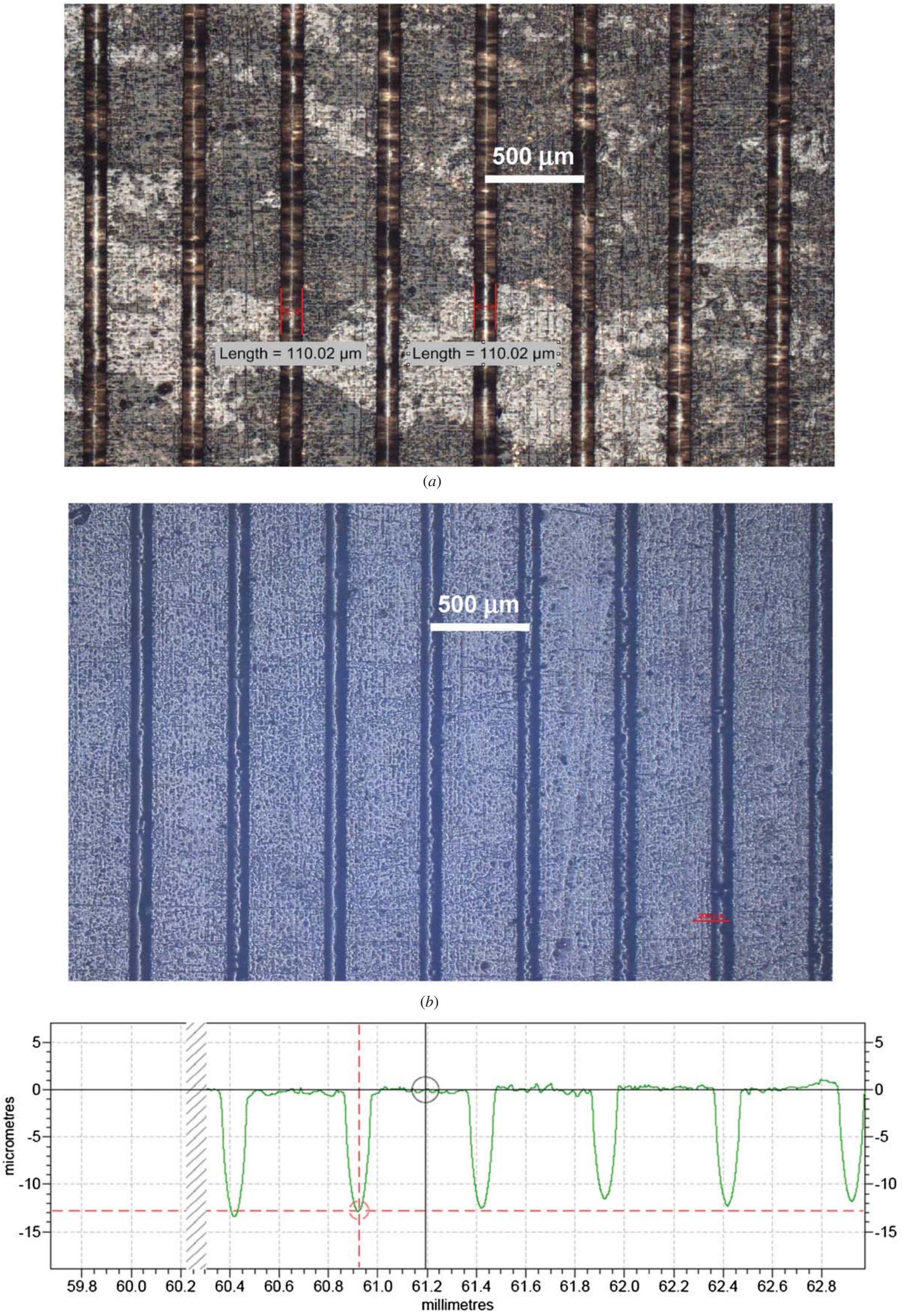


(*a*)

(*b*)

**Figure 5.** A flexible polymer mould mounted on a metal roller. (*a*) Patterned microstructures on a LCP–Cu film via copper etching. (*b*) The film was attached on a metal roller.

The film substrate used was PET (polyethylene terephthalate) film. This PET is a semi-crystalline polymer, and it has a weak glass transition phenomenon at a temperature

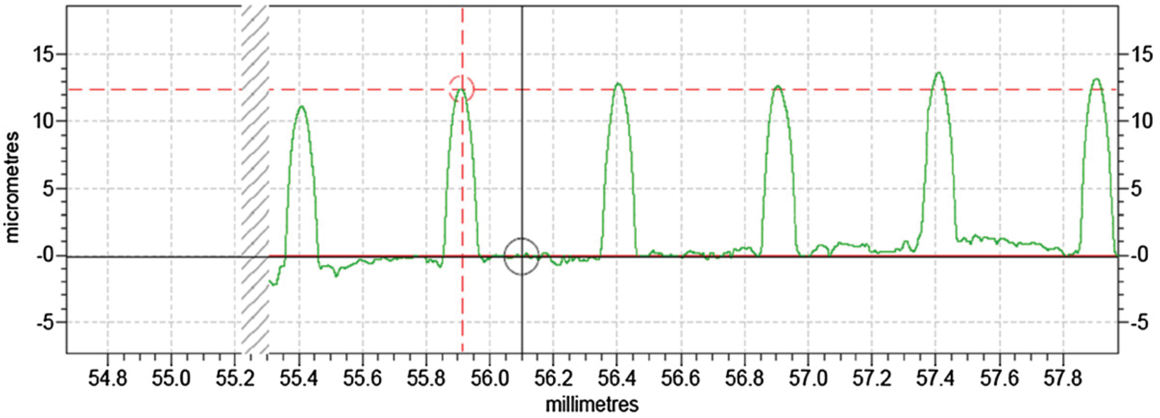


(*a*)

(*b*)

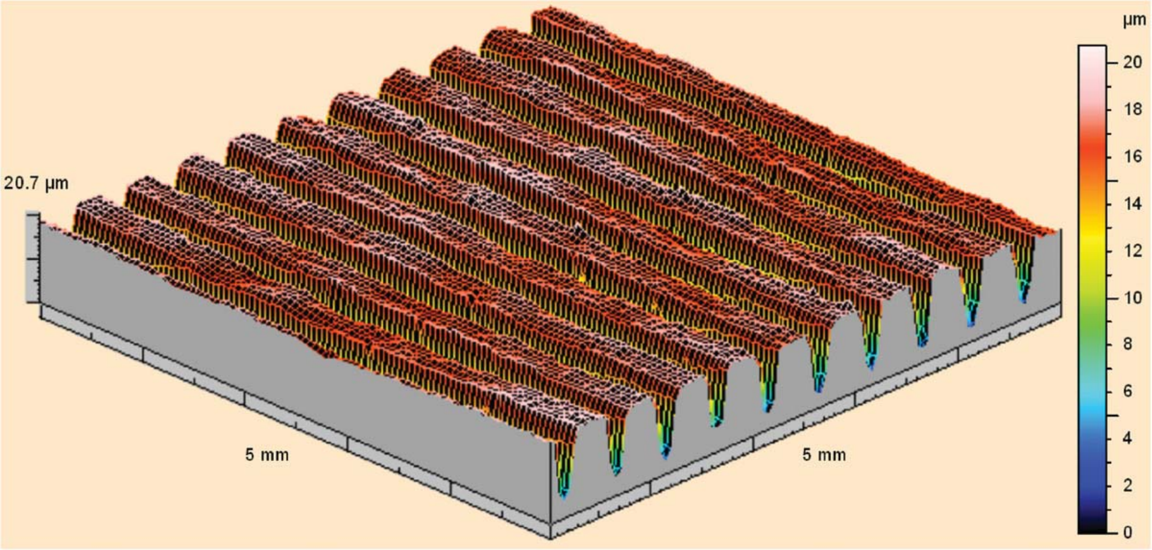
(*c*)

**Figure 6.** Channels of a flexible LCP–Cu mould with the width and pitch of 100 and 500 $\mu$m, respectively, and the embossed protrusive lines on the PET film obtained via roll-to-roll UV embossing using the flexible mould. (*a*) and (*b*) are two optical microscope images of the micro-channels on the mould and the protrusive lines on the PET film, respectively; (*c*) and (*d*) are two profiles of the channels on the mould and the lines on the PET film, respectively, measured using the stylus profilometer.



(*d*)

**Figure 6.** (Continued.)



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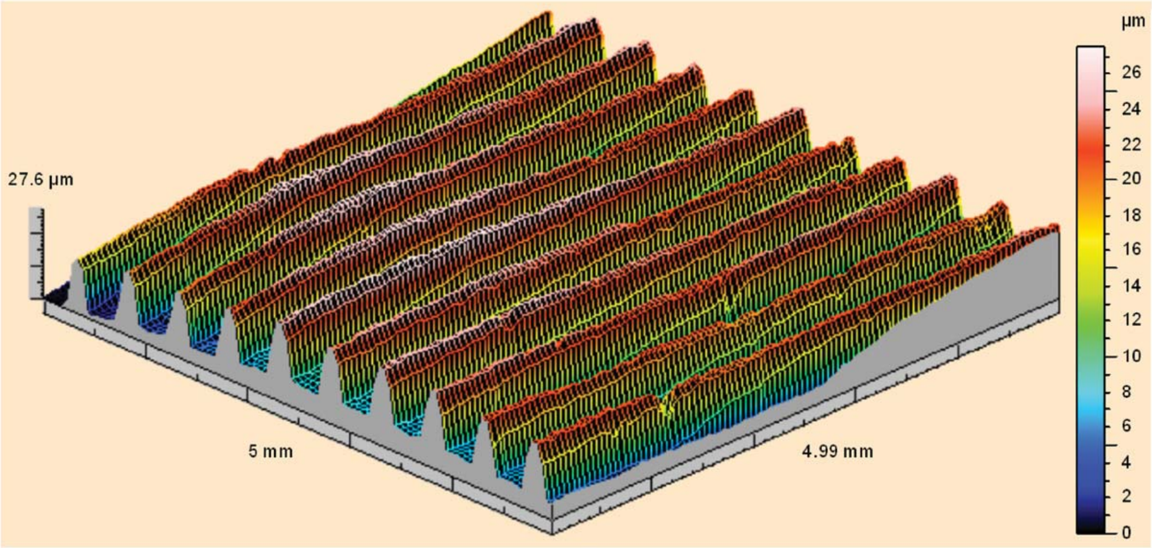
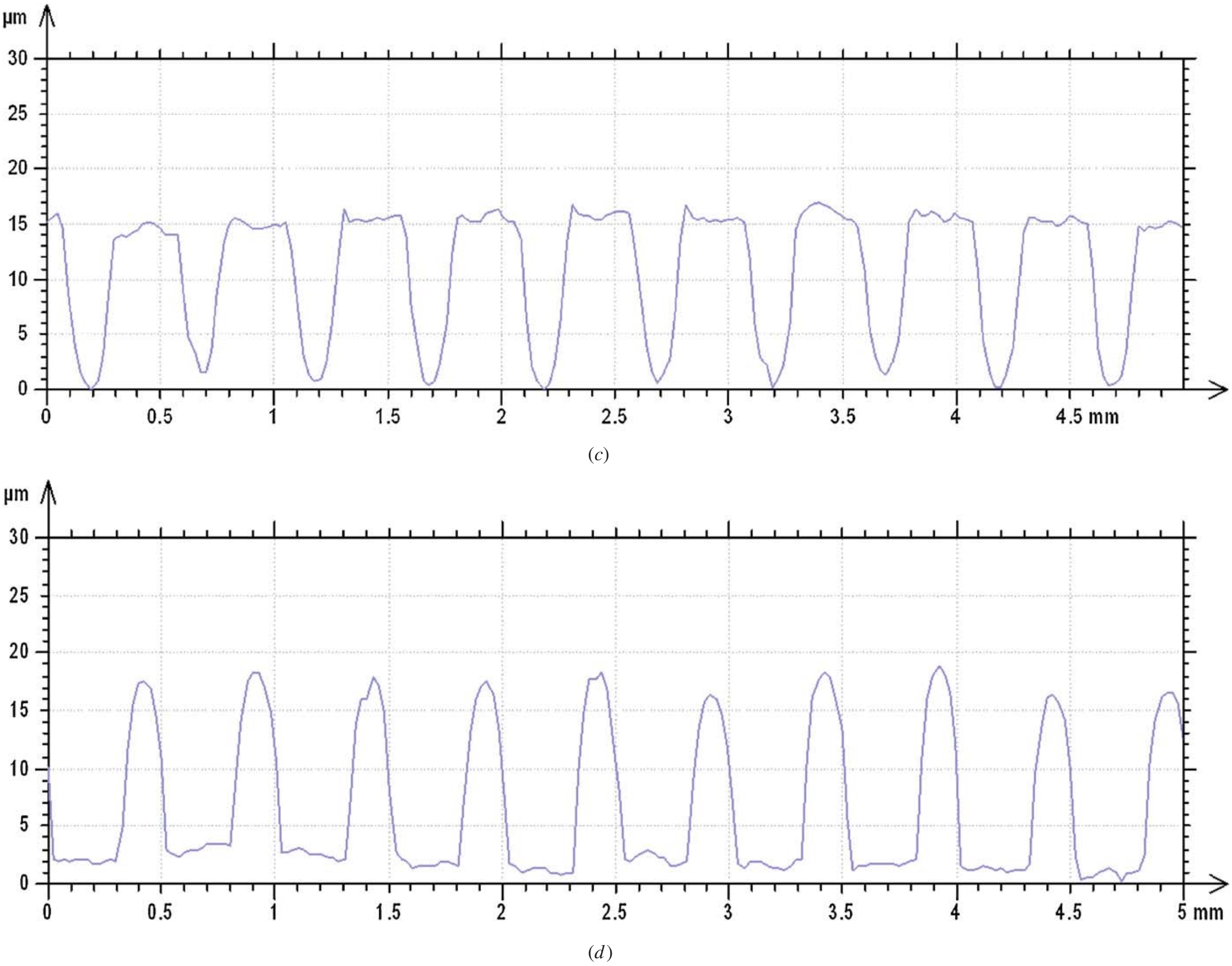


(*b*)

**Figure 7.** Channels of a flexible LCP–Cu mould with the width and pitch of 200 and 500 $\mu$m, respectively, and the embossed protrusive lines on the PET film obtained via roll-to-roll UV embossing using the flexible mould. (*a*) and (*b*) are two 3D images of the micro-channels on the mould and the protrusive lines on the PET film, respectively; (*c*) and (*d*) are two profiles of the channels on the mould and the lines on the PET film, respectively, measured using the stylus profilometer.



(*c*)

(*d*)

**Figure 7.** (Continued.)

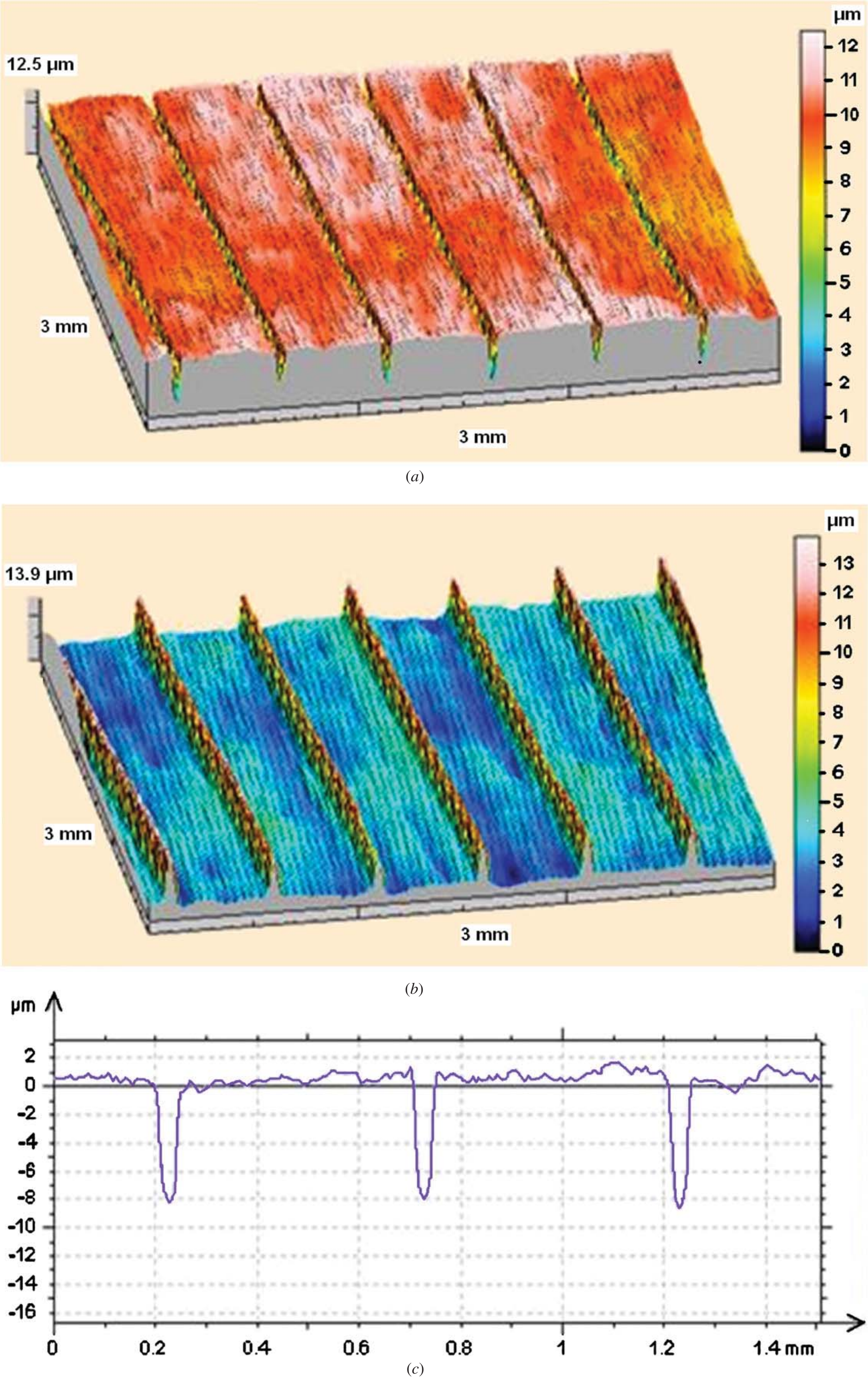
($T_g$) of 81.5 °C and a melting point at 254 °C. The PET film used is HK-31 from Higashiyama New Technology. The PET film can be heated up to 120 °C without any obvious deformation or wrinkle. The PET film thickness selected is 125 $\mu$m. The PET film is pre-treated on its both sides with a primer treatment for promoting adhesion of coated layers. Thus, this film has a structure of primer, base film and primer. The film is highly transparent with good ink adhesion properties.

The UV-curable liquid resins (Epoxy Technology, EPO-TEK® OG134 and OG172) were mixed with a ratio of 2:1 and used for forming microstructures. Then, its viscosity and the shear stress versus the shear rate at the room temperature were studied and are shown in figure 3. The UV curable resin is close to a Newtonian fluid. This means that its viscosity keeps constant as the shear rate varies, while its shear stress shows a linear proportion versus the shear rate in a certain range. Its viscosity, however, decreases with the increasing temperature, and the obtained investigation result is shown in figure 4. A rotational cylinder-type rheometer (Physica MCR 301 from Anton Paar GmbH) was used for measuring the viscosity and shear stress of the UV curable liquid resins.

The roll-to-roll UV embossing process used a flexible polymer–metal mould. The mould was made from a hybrid film of a liquid crystal polymer (LCP) laminated with copper foils on both sides of it. The LCP has a high glass transition temperature $T_g$, which is around 280 °C. Therefore, the flexible mould made from the LCP is suitable for both UV embossing and hot embossing of microstructures.

This hybrid LCP–Cu film was made via laminating two copper foils simultaneously with a LCP foil sandwiched in-between. The LCP foil is 50 $\mu$m thick, and the copper foils on both sides of the LCP are 36 $\mu$m thick. Both sides of the LCP–Cu film are covered with dry photoresist films. The dry film on the top surface was patterned by photolithography, and the copper foil on this side was etched by wet etching. The dry film on the bottom surface of the LCP–Cu film was used to protect the copper foil from being etched.

The height of the microstructures on the mould is defined by the etching depth of the copper foil, which can be determined by the etch rate and the etching period. In addition to etchant-related parameters such as the density and temperature of etchant solution, the etching rate is also related to the pattern size and pattern density. For a certain micro-



(*a*)

(*b*)

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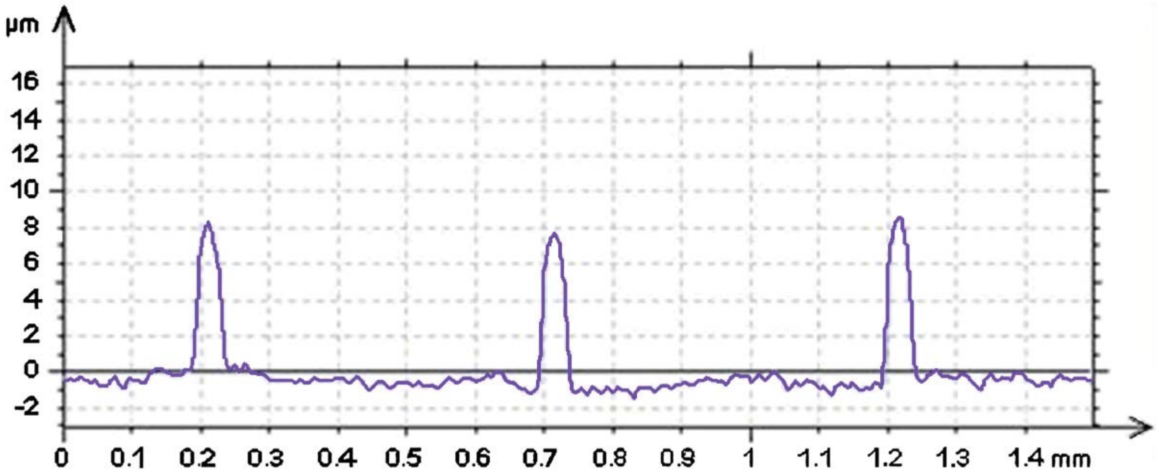
**Figure 8.** Channels of a flexible LCP–Cu mould with the width and pitch of 50 and 500 $\mu$m, respectively, and the embossed protrusive lines on the PET film obtained via roll-to-roll UV embossing using the flexible mould. (*a*) and (*b*) are two 3D images of the micro-channels on the mould and the protrusive lines on the PET film, respectively; (*c*) and (*d*) are two profiles of the channels on the mould and the lines on the PET film, respectively, measured using the stylus profilometer.



(*d*)

**Figure 8.** (Continued.)

pattern, the etching depth can be controlled by the etching period.

Figure 5(*a*) demonstrates a fabricated flexible mould. The copper foil was etched to the bottom and the LCP foil was exposed, so that microstructures with a vertical height of 36 $\mu$m could be obtained. In our actual UV embossing, the Cu etching was stopped when the etching depth reached about 15 $\mu$m, leaving a residual copper layer of about 20 $\mu$m in order to increase the strength of the flexible mould. The size of the LCP–Cu mould was 400 mm × 300 mm. Hence, two pieces of such patterned films were tiled and wrapped around the embossing roller to form the flexible mould. The LCP–Cu mould was attached on a metal roller as shown in figure 5(*b*), via a double-sided adhesive tape, which is a method often used in flexo-printing. The mould contained micro-scale patterns to be embossed onto the resin coating on the film substrate.

The diameter of the embossing roll is 160 mm. If the thickness (0.5 mm) of a double-sided tape is considered, the circumference of the embossing roll is 505.8 mm. The size of a fabricated flexible mould was 400 mm (roll axis direction) × 300 mm (roll circumference direction). Hence, two pieces of the fabricated flexible mould need to be cut and tiled precisely in order to attach them around the roll with minimum seam. Because the lateral and vertical dimensions of each micro-pattern on the mould are sub-millimeter scales, the changes in the dimensions and shapes of the patterns on the flexible mould before and after it is attached to the roll can be neglected.

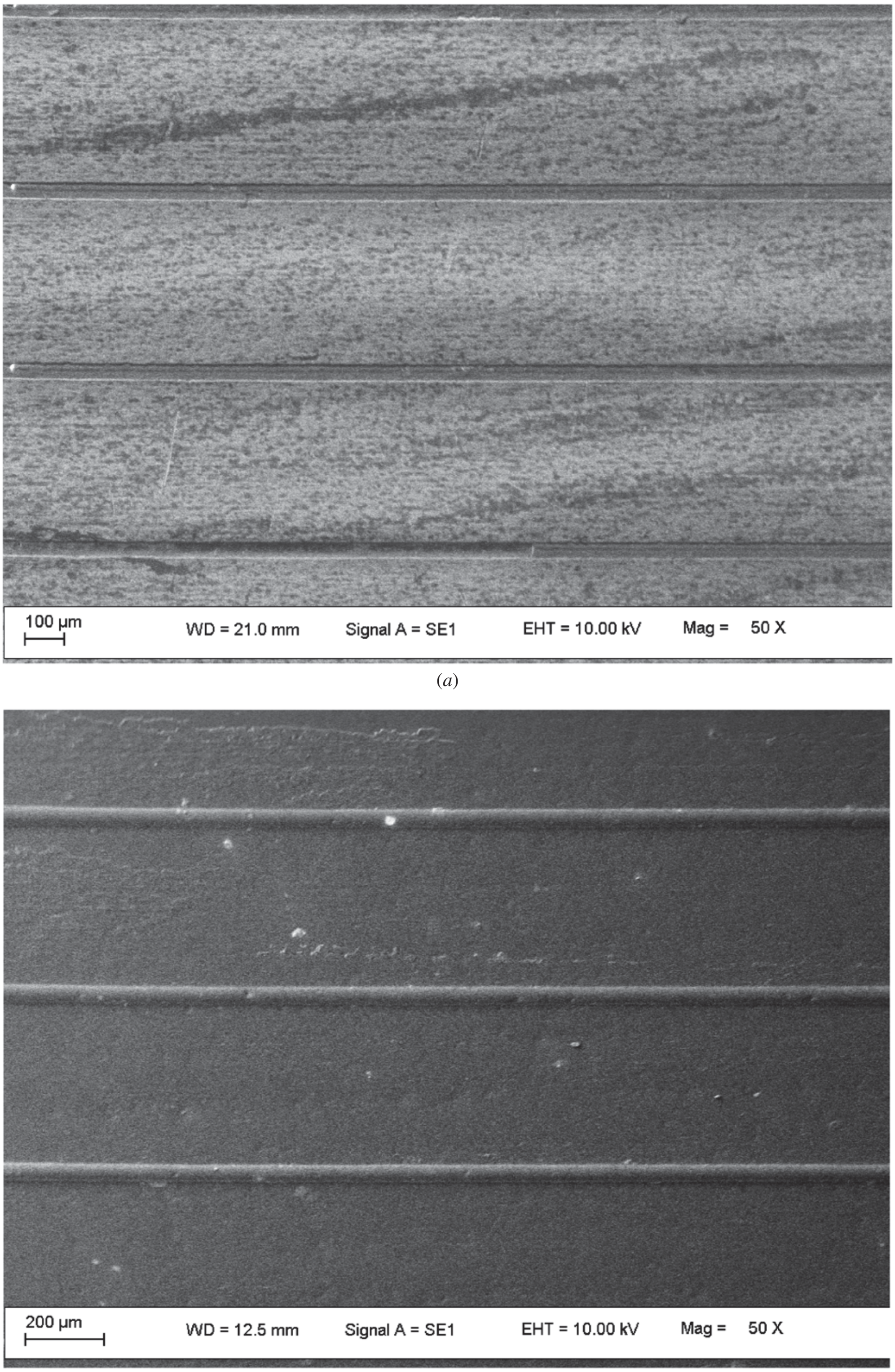
The UV lamp was located directly under the embossing roller. This configuration allows simultaneous embossing and curing of the liquid resin, increasing the production speed. Profiles including 3D surfaces of the embossed patterns and their corresponding mould patterns were evaluated using a stylus profilometer. These 3D surfaces were then compared to determine the quality and fidelity of the embossed patterns, which are reported in the following section. The stylus profilometer used is Form Talysurf Series 2 from Taylor Hobson, which can be used for 3D and 2D measurements of embossed profiles.

## 3. Mould characterization and embossing results

Figures 6(*a*) and (*c*) show the micro-channel array patterned on a LCP–Cu flexible mould. The width and pitch of the micro-channels are 100 and 500 $\mu$m, respectively. It can be seen from figure 6(*c*) that the average depth of the micro-channels is 12 $\mu$m measured using the stylus profilometer. Figures 6(*b*) and (*d*) show the embossed protrusive lines on a PET film obtained via roll-to-roll UV embossing using the LCP–Cu mould. The embossed protrusive lines replicated the profiles of the micro-channels precisely, with the width and pitch of 100 and 500 $\mu$m, respectively. The height of the embossed lines ranged from 11.5 to 12 $\mu$m, with a fidelity error $< -4.2\%$ in the vertical direction.

Figures 7(*a*) and (*c*) show the micro-channels of another flexible LCP–Cu mould. The width and pitch of the micro-channels are 200 and 500 $\mu$m, respectively. It can be seen from figure 7(*c*) that the average depth of the micro-channels is 15 $\mu$m measured using the stylus profilometer. Figures 7(*b*) and (*d*) show the embossed protrusive lines on the PET film obtained via roll-to-roll UV embossing using this LCP–Cu mould. The embossed line heights agreed well with the line depths of the patterns on the mould with the width and pitch of 200 and 500 $\mu$m, respectively. The height of the embossed lines ranged from 14.5 to 15 $\mu$m, with a fidelity error $< -3.4\%$ in the vertical direction.

Figures 8 (*a*) and (*c*) show the channels of another flexible LCP–Cu mould, and figures 8(*b*) and (*d*) show the embossed protrusive lines on the PET film obtained via roll-to-roll UV embossing using this LCP–Cu mould. Figures 9(*a*) and (*b*) show the SEM images of the channels of the flexible mould and the embossed protrusive lines on the film obtained via embossing using the mould, respectively. Both the embossed line heights and channel widths coincided with the channel depths and widths on the mould. The embossed protrusive lines replicated the profiles of the channels precisely, with the width and pitch of 50 and 500 $\mu$m, respectively. The height of the embossed lines ranged from 8.5 to 9 $\mu$m, with a fidelity error $< -5.6\%$ in the vertical direction.

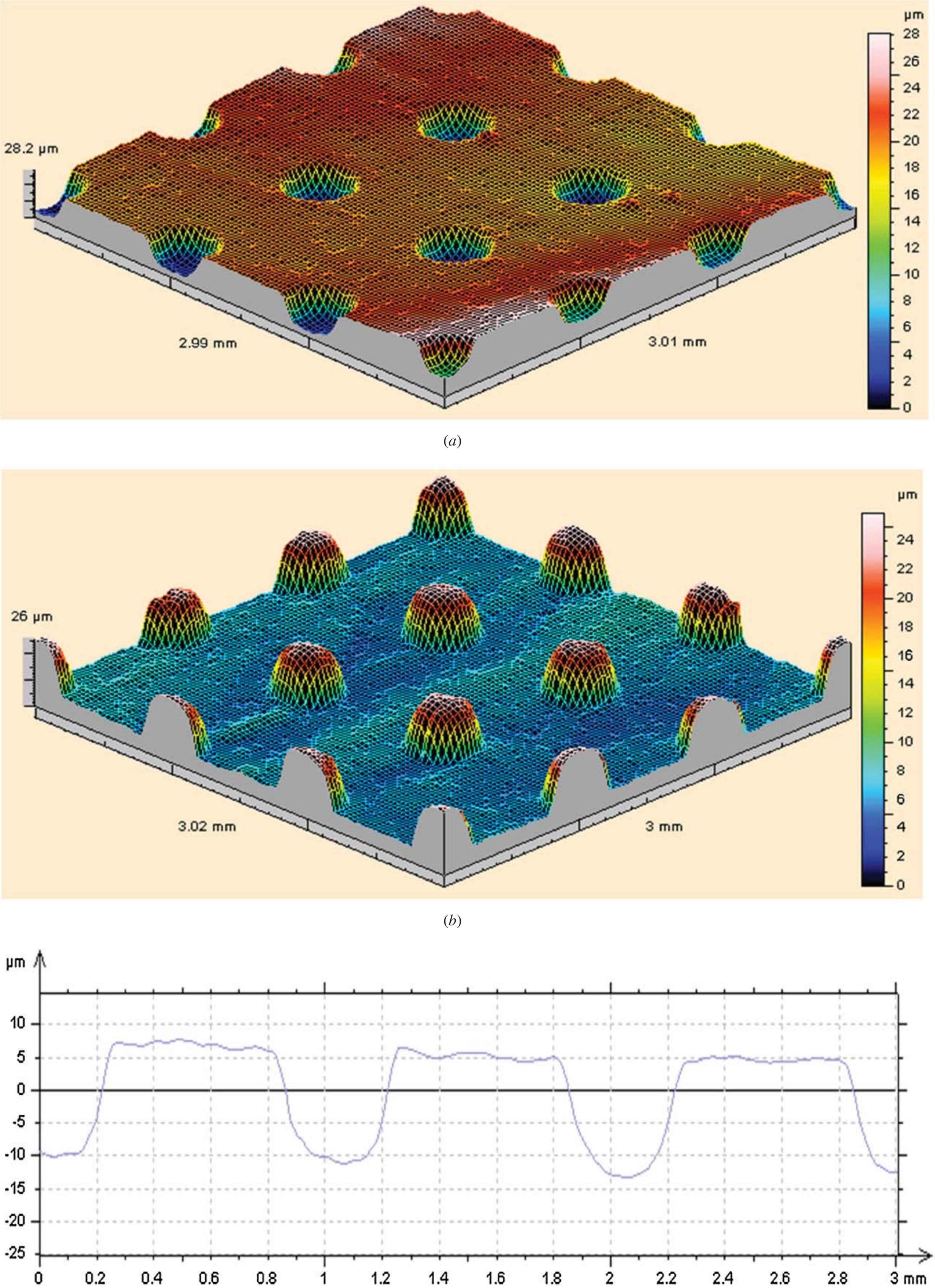


(*a*)

(*b*)

**Figure 9.** SEM images of the channels of (*a*) the flexible LCP–Cu mould with the width and pitch of 50 and 500 $\mu$m, respectively, and (*b*) the embossed protrusive lines on the PET film obtained via roll-to-roll UV embossing using the flexible mould.

Figures 10(*a*) and (*c*) show the holes on a flexible LCP–Cu mould, and figures 10(*b*) and (*d*) show the embossed dot pillars on the PET film obtained via roll-to-roll UV embossing using this LCP–Cu mould. The embossed dot pillars replicated the profiles of the holes precisely, with the diameter and pitch of 400 and 1000 $\mu$m, respectively. The height of the embossed dot pillars ranged from 16 to 16.5 $\mu$m, with an average fidelity error in depth $< -3.1\%$.

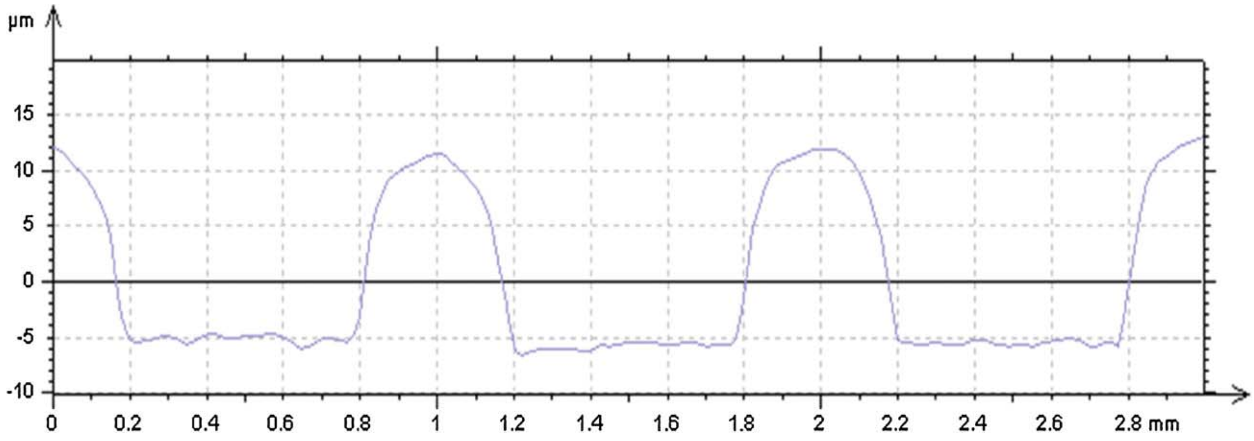


(*a*)

(*b*)

(*c*)

**Figure 10.** Holes on a flexible LCP–Cu mould with the diameter and pitch of 400 and 1000 $\mu$m, respectively, and the embossed dot pillars on the PET film obtained via roll-to-roll UV embossing using the flexible mould. (*a*) and (*b*) are two 3D images of the micro-holes on the mould and the dot pillars on the PET film, respectively; (*c*) and (*d*) are two profiles of the micro-holes and the pillars on the mould and the PET film, respectively, measured using the stylus profilometer.



(*d*)

**Figure 10.** (Continued.)

## 4. Conclusions

Flexible moulds were fabricated from a hybrid film substrate made of a liquid crystal polymer with double clad copper foils. The effective pattern area of the fabricated flexible mould was 400 mm × 300 mm with a minimal feature size of 50 $\mu$m. Large-area roller embossing using the hybrid flexible mould was demonstrated, and micro-features and structures such as micro-channels and dot arrays were replicated on thermoplastic substrates. In addition to its ease and low cost in fabrication, the hybrid flexible moulds demonstrated to have acceptable fidelity and durability. The hybrid flexible mould is a novel solution for large-area roller embossing. The roll-to-roll embossing processes are capable of producing micro-scale structures and functional devices over a large area at one time.

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