Development of Roll-to-Flat Thermal Imprinting Equipment and Experimental Study of Large Area Pattern Replication on Polymer Substrate

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열방식 Roll to Flat 임프린트 공정 장비의 개발과 이를 이용한 대면적 고분자 기판의 패턴 성형에 관한 실험적 연구

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Abstract

Large area micro pattern replication has promising application potential in many areas. Rolling imprint process has been demonstrated as one of the most competitive processes for such micro pattern replication, because it has advantages in low cost, high throughput and high efficiency. In this paper, we developed a prototype of roll-to-flat(R2F) thermal imprint system for large area micro pattern replication process, which is one of the key processes in the fabrication of flexible displays. Experimental tests were conducted to evaluate the feasibility of system and the parameters' effect on the process, such as flat mold temperature, loading pressure and rolling speed. 100mm × 100mm stainless steel flat mold and commercially available polycarbonate sheets were used for the tests. The experimental results showed that the developed R2F system is suitable for fabrication of various micro devices with micro pattern over large area.

Key Words : Large area(대면적), Pattern replication(패턴 성형), Roll-to-Flat Micro thermal imprinting(열방식 롤투플랫 마이크로 임트린팅), Polymer substrate(고분자 기판)

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1. Introduction

In recent years, the polymer based micro- and nano-fabrication using rolling imprint process has been gaining more attention since it has the advantage of better uniformity, less imprint pressure, simple structure, and high efficiency and low cost.

This is because it is basically continuous fabrication process while the traditional flat-press type imprint process is discontinuous. Especially for the large area micro pattern replication, rolling imprint process has been demonstrated as one of the most competitive processes. Many studies have been performed for various applications using such rolling process, such as flexible display, e-paper, flexible solar cell and polymeric microfluidic devices and so on [1~3].

Continuous rolling process in manufacturing, i.e. roll-to-roll (R2R), has been currently applied in many industrial fields, such as gravure printing or flexography printing (or flexo). It is used to imprint intended patten on flexible thin films that is consistent with the pattern on the roller, for example, printing newspapers, magazines and packages, etc. In the field of micro-/nano- fabrication field, S. Y. Chou group firstly proposed the well-known nanoimprint lithography (NIL) process. Such imprinting process is a promising technique for the pattern replication of polymer substrate[4]. Up to date, many literatures have been reported including thermal and ultra-violet (UV)-based imprinting process[5,6]. It is also S. Y. Chou group that firstly proposed the combination of imprinting process and rolling process and then developed roller nanoimprint lithography (RNIL) process for rapid pattern replication on a large area substrate[7]. Such RNIL process provides some distinctive advantages such as compact-sized system configuration, lower imprint force, and better replication uniformity because only a linear area is in contact during imprinting. Especially in the field of manufacturing for flexible electronics, the fabrication method via R2R processes for flexible displays have been demonstrated by some researchers. For example, Wang Xiaojia et al. have developed a novel full color electrophoretic film manufacturing process using Roller-type method[8]. S. C. Liang group developed a continuous process using R2F method combining all process steps including: polymer coating, pattern embossing, filling/sealing, and lamination for manufacturing of flexible display or electronic paper at a low-cost and high speed[9~12].

However, the rolling process for the fabrication of flexible display has not been investigated adequately and most of them are limited to using UV-based techniques. In this study we aim to develop a prototype of a roll-to-flat (R2F) micro thermal imprint system and to evaluate its feasibility for the large area replication as an potential application of flexible display. Furthermore, series of tests were conducted to investigate the effects of process control parameters, i.e. rolling speed, loading pressure and imprinting temperature, on the replication of polycarbonate (PC) substrate.

2. Development of R2F thermal imprint system

In this study, we developed a prototype of lab-scale roll-to-flat (R2F) micro thermal imprint system. A photograph of the system is shown in Fig. 1. It consists of several major sub-units, including roller imprint unit, polymer holding and auto-releasing unit, control unit for process parameters input (temperature, speed, force, and etc.) and result data acquisition unit.

Figure 2 shows the R2F micro thermal imprint system consisting of major components such as the pressing roller

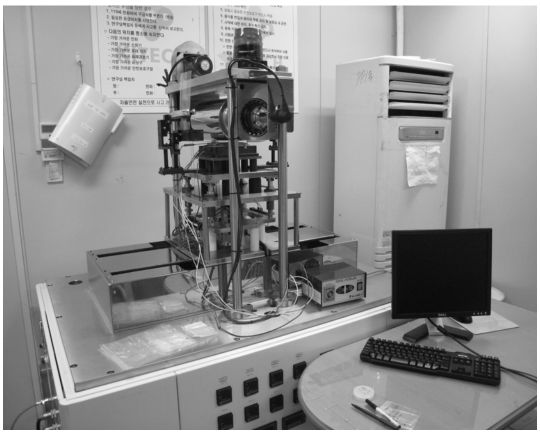


Fig. 1 Overview of the roll-to-flat (R2F) system

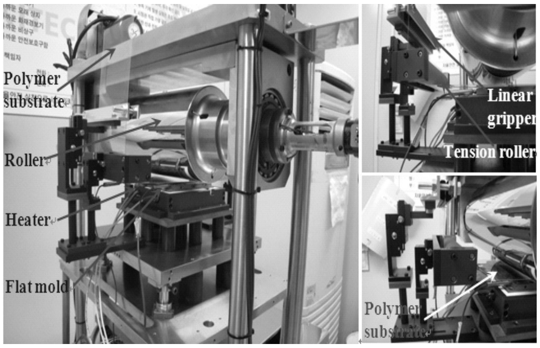


Fig. 2 Photographs of major components

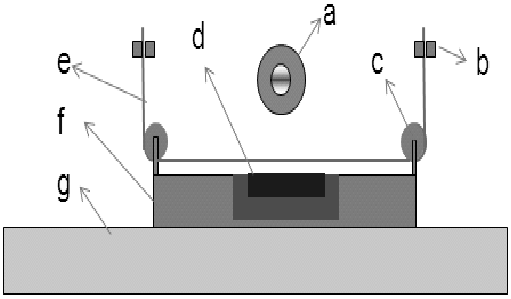


Fig. 3 schematic diagram of R2F system: (a) roller (b) linear gripper (c) tension roller (d) flat mold (e) polymer substrate (f) movable stage (g) system base

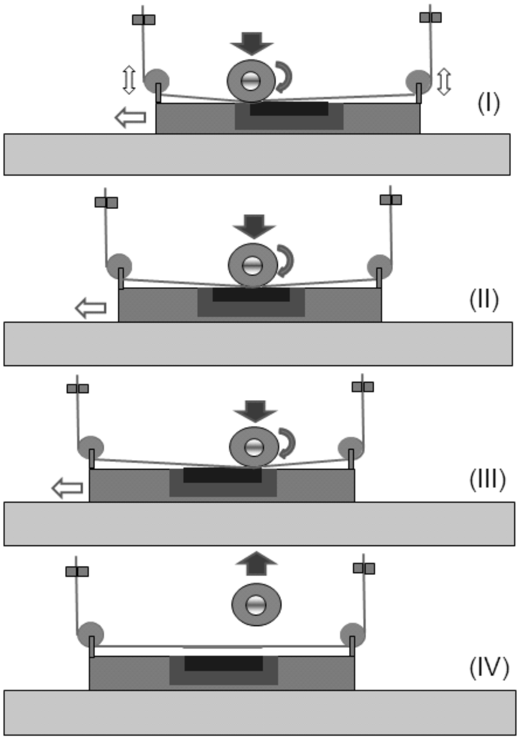


Fig. 4 Basic procedure of the R2F process

with heaters in the core, which is actuated by AC-servo motor, the movable stage, the flat mold combined with a heater, and the polymer substrate holding and auto-releasing device. The engineers from display industry usually do not want to use the two heaters simultaneously to enhance throughput process and to save energy. The polymer substrate supply and holding device has a pair of linear grippers and a pair of tension rollers. This device can adjust the small gap between the polymer substrate and flat mold. When the pattern replication is being performed in the semi-linear contact area between the substrate and mold, other area of polymer will not contact with the mold. After rolling and the mold moving forward, the replicated substrate will be automatically separated from the mold. Such concept is introduced because the imprinted pattern can be released vertically without damage from the flat mold by the continuous rolling movement.

The movable stage, supporting the flat mold, can move parallelly to the sample surface in one direction, at the speed range from 0.1 mm/s to 10 mm/s. The stroke of the movable stage is about 700 mm. The maximum pressing force of the rolling device is 500 kgf and the force is controlled with a precision of less than 1 N. The detailed specification of the system is summarized in Table 1.

The schematic diagram of the R2F thermal imprint system is illustrated in Fig. 3. The basic procedure of the R2F process are shown in Fig. 4. Firstly the polymer substrate is loaded on the holding and auto-releasing device, and then the gap between substrate and flat mold is adjusted by turning the height of tension rollers. Simultaneously, the flat mold is heated above the glass transition temperature (T_g) of polymer substrate whereas the roller temperature is set at room temperature since the

Table 1 Specifications of R2F micro thermal imprint system

Item		Content
System size (mm)		1500×1100×1800
Stage stroke (mm)		700
Roller	Diameter (mm)	160
	Width (mm)	290
	Maximum temperature (℃)	200
	Maximum loading pressure (kgf)	500
	Scanning speed (mm/s)	0.1~10
	Material	Stainless steel
Flat mold	Maximum mold area (mm^2)	150×150
	Maximum temperature (℃)	300
	Moving speed (mm/s)	0.1~10

substrate is basically heated by the flat mold. Then, the roller moves down to press the polymer substrate onto the surface of flat mold with a specific pressure (Fig. 4 (I)). The flat mold supported by the movable stage moves in one direction at a constant speed. So the roller is rotated over the polymer substrate and flat mold (Fig. 4 (II) and (III)). After the rolling over the flat mold area is completed, the roller moves up vertically then the substrate is separated from the flat mold (Fig. 4 (IV)). The step I ~ IV is one cycle of imprinting and rolling.

3. Flat mold and substrate preparation

A stainless steel flat mold with size of 100 mm × 100 mm was used in the experiments, which was fabricated by a dicing process. The entire surface of the mold is composed of micro square array with a height of 95 µm, width of 200 µm and 110 µm spacing. Fig. 5 shows a photograph of the flat mold that is embedded on the movable stage. Fig. 6 illustrates the micro pattern on the mold. It is measured using a typical 3D white-light scannig interferometry (SNU Precision, Korea). Commercially polycarbonate (PC) substrates were imprinted in the tests. Under the process condition that flat mold temperature is above PC's glass transition temperature (T_g = 150℃) and low rolling speed, the micro patterns on the flat mold can be transferred to the PC substrate.

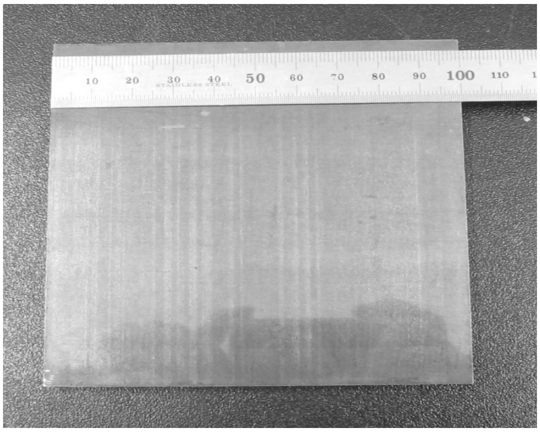


Fig. 5 Photograph of flat mold

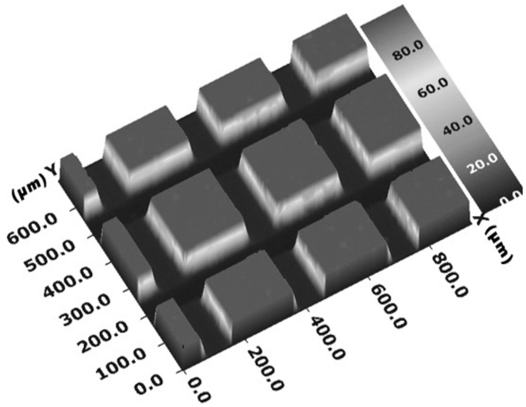


Fig. 6 Scanned 3D image of mold sub-feature: The pattern height is about 90㎛

4. Test and evaluation

4.1 Experiment results

In order to evaluate the performance and feasibility of the developed R2F micro thermal imprint system and to study the rolling process parameters, including imprinting temperature, loading pressure and rolling speed, series of experiments were conducted.

Using the white-light scanning interferomety, the appearance and dimensions of the micro pattern on the flat mold and the imprinted pattern on the substrate surface were observed and measured. Based on these measured results, the evaluation of process formability, defined as replication ratio R_r, can be calculated as: $R_r = D_c/H_m$, where D_c and H_m are the depth of replicated micro cup and the height of pattern on the flat mold, respectively.

Initial trial tests were conducted using a rigid and thick PC substrate with thickness of 2 mm. Fig. 7 shows an example of replication result from flat mold to PC substrate. The process was conducted under the condition of 200 kgf of press force, 1 mm/s rolling speed (also the moving speed of movable stage) and 160℃ of temperature of flat mold. The roller was not heated and it was in room temperature. From the figure we can see that good replication results of uniform micro cup arrays were fabricated over the whole surface area. Some defects on the beginning part (on the bottom of the figure) due to the change of rolling speed from 0 to the constant speed 1 mm/s at the beginning of process. In the test, the polymer holding and releasing device was not used because the substrate is rigid enough.

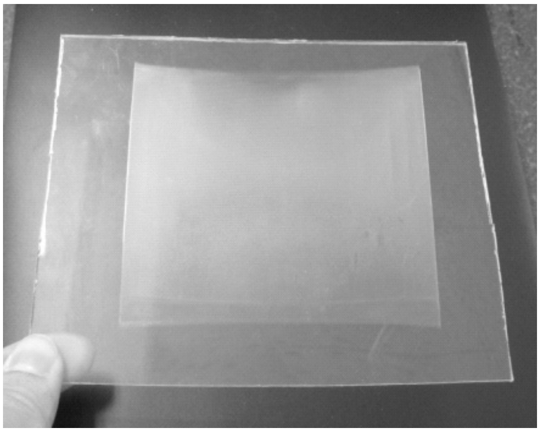
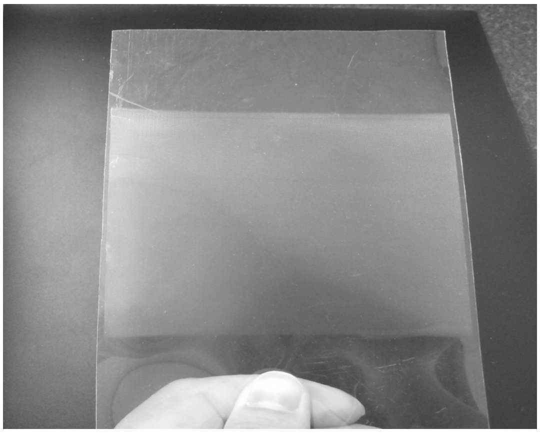
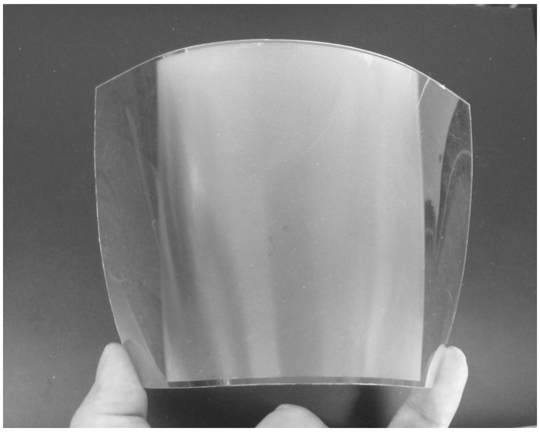


Fig. 7 Photograph of imprinted 2 mm-thick substrate

Tests for thin and flexible PC sheets with thickness of 0.5 mm were conducted using the polymer holding and auto-releasing device. The tests were performed under different process conditions. The substrate was firstly loaded through the tension rollers and clamped by the linear rubber grippers. By adjusting the height of the gripper and tension roller, the small gap between the substrate and flat mold can be adjusted. Considering the roller diameter and rolling speed, the gap would decide the



(a)



(b)

Fig. 8 (a) Photograph of imprinted 0.5 mm -thick substrate, (b) Photograph of imprinted 0.5 mm-thick substrate

releasing speed between substrate and flat mold when the roller rolls forward. Qualitatively speaking, larger gap results in the quicker releasing. In our experiments, the gap was set to about 10 mm.

One of the replicated results for the flexible substrate is shown in Fig. 8 and Fig. 9. It can be seen from Fig. 8 that the pattern was well replicated to the flexible substrate without thermal distortion of substrate. Thermal distortion is generally a serous problem when imprinting and rolling is adopted. The temperature of the flat mold was set at 160℃ and the pressing roller was set at room temperature (without heating). Loading pressure and rolling speed were set at 180 kgf and 0.5 mm/s, respectively. However, one can also see from the figure that there are some defects or insufficient filling at the edge area (beginning and ending part of rolling) due to the change of rolling speed, as described previously. Such defect could be regarded as an "edge effect" in the rolling process. In the practical manufacturing, the change of rolling speed could be avoided by designing a dummy area without pattern near by the edge so as to reduce the edge effect. Except the edge effect, all the measurements data from the several representative points over 100 mm × 100 mm substrate showed the small variation of the replication ratio (the variation is less than 10%). This means the equipment and process has potential to be applied to large area patterning.

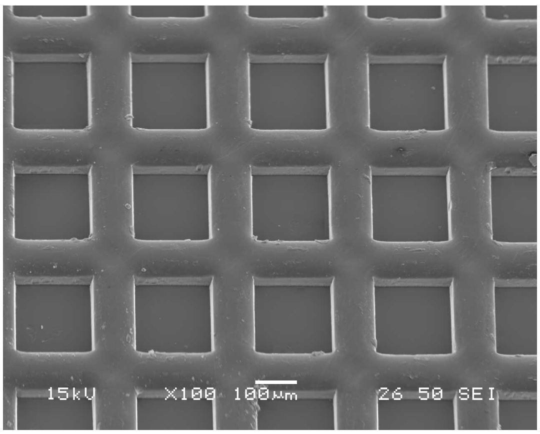


Fig. 9 SEM image of imprinted patterns

In order to evaluate the effect of rolling speed, the speed was varied from 0.2 mm/s to 2 mm/s whereas temperature and loading pressure were kept constant. Then imprinted PC substrates were measured to calculate the replication ratio. Relationship between rolling speed and replication ratio is shown in Fig. 10. The replication ratio decreases as the rolling speed increase from 0.2 to 2 mm/s. This is because lower speed results in longer contact time between mold and substrate, which induces more material flow to transfer. An empirical model based on the experimental data was obtained. The model para-

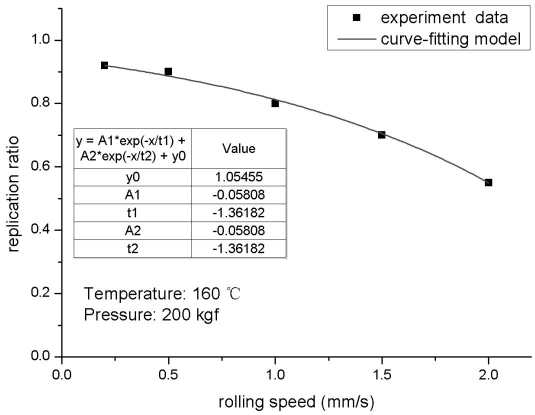


Fig. 10 Relationship between rolling speed and replication ratio

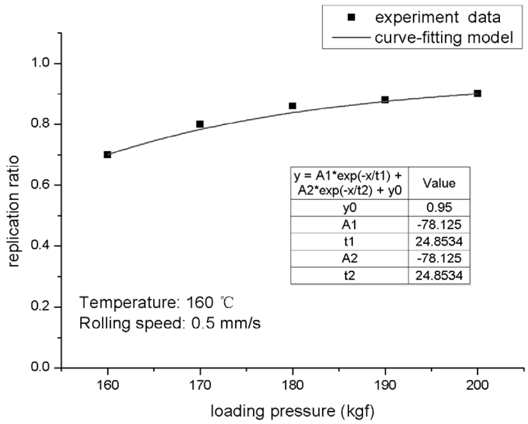


Fig. 11 Relationship between loading pressure and replication ratio

meters are shown in the figure. In addition, to evaluate the effect of loading pressure, the pressure was varied from 160 kgf to 200 kgf whereas temperature and rolling speed were kept constantly. The result is shown in Fig. 11. The replication ratio increases as the loading pressure increase from 160 kgf to 200 kgf, since higher pressing force would result in sufficient polymer flow. An empirical model based on the experimental data was also obtained as shown in the figure. As for the analysis of pattern precision of this manufacturing method, higher replication ratio means higher replication fidelity and higher aspect ratio of the ridge of the cup. Based on the analysis of relationship between replication ratio and process control parameter, i.e. rolling speed and loading pressure, a brief conclusion could be achieved as: higher replication ratio or better pattern precision could be achieved under the condition of lower rolling speed and higher loading pressure. Within the parameter range of rolling speed loading pressure at 0.2 mm/s and 200 kgf in this study, an increase of 63.6% in replication ratio was obtained when rolling speed was decreased from 2 mm/s to 0.2 mm/s (loading pressure isconstant). And an increase of 28.7% in replication ratio was obtained when loading pressure was increased from 160 kgf to 200 kgf. For the process, the temperature was constant.

4.2 Discussion

It should be noted it is convenient to change flat mold to the one with various micro patterns even though only a flat mold composed of square pattern array was used in this study. It should also be noted that the pattern aspect ratio can also affect the final replication result. Higher aspect ratio needs longer filling time in the cavity and then lower rolling speed or larger loading pressure is necessary. However, rolling speed and loading pressure could not be decreased and increased without limitation, considering the manufacturing efficiency and equipment damage. So a tradeoff would be necessary in the practical fabrication. On the other hand, it is also possible to use thin film mold which can be enwrapped around the roller surface. In this case the roller could be heated to the temperature higher than T_g of polymer substrate, whereas the flat stage could be set to a lower temperature. Thus, the developed R2F imprinting system is flexible to be applied to various process and pattern.

5. Conclusion

We have developed a prototype of lab-scale roll-to-flat (R2F) thermal imprint system and also demonstrated a micro pattern replication process over a large area. Series of tests were carried out using a stainless steel flat mold at size of 100 mm × 100 mm and commercially available polycarbonate (PC) substrates to evaluate the system feasibility and process control parameters' effect, such as flat mold temperature, loading pressure and rolling speed. The test results show that the developed R2F system is suitable for fabrication of various micro devices with micro pattern replication over a large area. For the thin and flexible PC substrate with thickness of 0.5 mm, using the polymer holding and auto-releasing device can achieve high replication ratio and uniformity over a large surface area when the rolling speed is lower (0.2 mm/s). And the thermal distortion of the substrate could be avoided. On the other hand, higher pressing force results in better replication ratio since it provides more sufficient polymer flow. The test results show that the developed R2F system is suitable for fabrication of various micro devices with micro pattern replication over a large area.

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