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PAPER

Push-pull process for contact defect-free patterning in reverse offset printing

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Abstract

Reverse offset printing is one of the most promising printing techniques for printed electronics because it enables us to generate fine patterns with high fidelity. However, in the pattern-generation process, where some of the ink applied on a blanket is removed by a cliché upon contact, there may be contact-defect formations due to unwanted contact between the bottom regions of the cliché and the blanket. This may occur when the pattern size is wide or when the depth of the cliché is shallow. To solve this problem, we develop a modified printing protocol that adopts a ‘negative’ printing pressure condition called a ‘push-pull’ process. In this process, the blanket first comes into contact with the cliché by pushing a blanket roller, and the roller is then pulled back to prevent the blanket indentation into the grooves of the cliché. By incorporating the push-pull process in reverse offset printing, we demonstrate defect-free formations of 4.0 mm × 5.0 mm large patterns using a shallow cliché with a depth of 2.6 μm, which is unattainable with the conventional push-only process. Further, we show that the adhesion between the ink-coated blanket and the cliché contributes to the maintenance of the contact through the printing process, even in the pull situation.

1. Introduction

The pattern-generation capability of printing methods is expected to be a low-cost alternative to photolithography and other vacuum processes used in electronic-device fabrication [1]. For devices in which either a coarse resolution is sufficient, screen/stencil printing [2–4], inkjet printing [5–7], gravure printing [8] and flexography [9, 10], all of which are widely used in graphic art printing, are employed to form electronically functioning layers. For example, typical applications of screen printing are solder paste printing in printed circuit boards, solid-oxide fuel-cell electrodes [11] and electrodes in multilayer ceramic capacitors [12]. However, with the increased demand for higher patterning resolution, more precise printing methods have been developed. Examples of methods that enable finer patterning are gravure offset printing [13–16] and screen offset printing [17, 18], in which ink is not patterned directly on a substrate, but on a silicone blanket. Moreover, reverse offset printing [19–23] and microcontact printing [24–28], where ink is applied to a silicone blanket or a silicone stamp, are being developed as single-micrometre-order patterning methods. In those methods, as opposed to conventional printing methods where a pattern is printed directly onto a target substrate, silicone materials are used in order to keep the structural integrity of the ink resulting from rapid solidification through solvent uptake by the silicone [27].

Although the above-mentioned printing methods are regarded as potential candidates for next-generation patterning processes, because silicone materials are soft and deformable, printing methods which utilise silicone suffer from defects due to an unwanted bottom contact (called ‘contact defect’ hereafter). For example, in gravure offset printing and reverse offset printing, the blanket may accidentally come into contact with the clichés’ grooves which correspond to the image area when a strong printing load is applied [29], resulting in the formation of an ‘open-type’ contact defect. As another example, microcontact printing may produce a ‘short-type’ contact defect (roof collapse) because of undesired contact between the
bottom of a stamp (non-image area) and a substrate under a strong printing load condition [30, 31]. In general, both of the defect formations may also occur if the aspect ratios of the cliché’s image area and the stamp’s non-image area are [32] low. To address this problem, in this paper, we propose a push-pull process for reverse offset printing which enables printing above the kiss-touch condition, and accordingly provides contact defect-free patterning. We examined the push-pull process in terms of contact-defect prevention by comparing it with a conventional pattern-generation process. We also propose a simple experimental method for the estimation of blanket deformation using a spherical silica-contaminated substrate. Then, we discuss key factors validating the developed push-pull process based on the adhesion characteristics between the ink-coated blanket and the cliché.

2. Experimental

2.1. Fabrication of a cliché

We made clichés by carrying out the photolithographic procedure. We spin-coated an epoxy-based negative photoresist SU-8 5 (MicroChem, US) on a UV–ozone cleaned 4 inch silicon wafer at 3000 rpm or 4000 rpm for 30 s. After pre-annealing at 65 °C for 2 min and 95 °C for 1 min, a UV light was exposed though a mask for 5 s. After post-annealing at 65 °C for 2 min and at 95 °C for 1 min, we developed the resist using propylene glycol monomethyl ether acetate (PGMEA) to form a relief structure on the wafer. We tested two types of relief patterns: rectilinear regions with widths of 0.8, 1, 2, 3 and 4 mm and each length of 5 mm for a wide feature, and interdigitated patterns with L/S of 10, 20 and 50 μm for fine structures. The resist layer thicknesses corresponding to the depths of the clichés were 2.6 and 1.6 μm for the wide and fine patterns, respectively. A schematic of the wide-feature pattern design is shown in figure 1(a).

2.2. Push-pull process in reverse offset printing

In this section, we first describe the conventional procedure employed for reverse offset printing, after which we provide details regarding the push-pull process. In reverse offset printing: (1) ink is coated on a blanket made of poly(dimethylsiloxane) (PDMS) with a capillary-driven slit coater at 20 mm s⁻¹, and dried for 30 s in ambient, (2) the ink-coated blanket comes into contact with the cliché, (3) parts of the ink layer are removed by transferring them to the raised areas of the cliché and (4) the ink remnant on the blanket is transferred onto a glass substrate to complete the printing. Details of the printing substrate have been reported in literature [19]. In this study, we performed the above printing process in a roll-to-sheet manner. In other words, we set the blanket on a ϕ200 mm roll, and we also set the cliché and a target glass substrate on precision granite tables. In steps (3) and (4), we carried out the ink transfers in such a way that the rotation of the blanket and translation of the table were operated synchronously. The mechanical flatness of the table was within ±1.0 μm, and the vertical parallelism errors between the blanket roll and the tables were less than 5.0 μm.

In step (2), we examined a conventional ‘push-only’ process and the proposed ‘push-pull’ process. In the push-only process, we applied positive printing pressure by pushing the blanket roller at a certain length so as to ensure that there is contact between the cliché and the blanket (note that even in the push-only process, the contact area experiences a negative pressure at the tailing side of the roller [32]). We set the indentation lengths Δp in the range of 0 to −70 μm from the kiss touch position. Here, we determine the kiss touch to be the position at which the load cells first
detect a negative (pushing) force by the stepwise descent of the blanket roller in 5 μm steps. Note that those indentations are in the same order when compared with typical values obtained in reverse offset printing that are reported elsewhere [20]. Hereafter, we express the indentation lengths using the origin of the kiss touch position (positive and negative indentation values correspond to pulling and pushing positions, respectively). On the other hand, in the push-pull process, the blanket roller was first pushed into the cliché at \( \Delta_p = -30 \mu m \), and the roller was subsequently pulled back before proceeding to step (3). The blanket roller positions after the pulling were varied between \(-20 \) and \( 20 \mu m \). The temporal changes in the roller positions from step (2) through the pattern-generation process of step (3) are illustrated in figure 1(b).

To evaluate the printing pressures and to determine the kiss touch position, we placed three load cells (TC-SR(T)G with hysteresis values less than 1% R.O. and maximum load capacity of 20 N, TEAC, Japan) on the cliché-mounted supporting glass substrate (160 mm × 230 mm × 13.8 mm), which was elevated by a 1 mm thick stainless spacer. We established the contacts between the load cells and the supporting glass using micrometre adjusters (figure 1(c)), and we monitored the total load during the aforementioned printing process. For the ink, we used silver nanoparticles (Ag NP) suspended in ethanol (RAGT-19, DIC, Japan). For the blanket, we used a 0.20 mm thick PDMS (Young modulus of 3 MPa) sheet supported by a 0.58 mm thick polyurethane foam, with inserted base polyethylene terephthalate (PET) films which were 0.5 mm thick (Kinyosha, Japan). The Young modulus of the polyurethane cushion estimated by a rheometer (MCR302, Anton Paar, US) was \( 3.5 \times 10^2 \) kPa. We observed the printed patterns using microscopes (VHX-1000, Keyence, Japan and MX51, Olympus, Japan). All of the printing experiments were carried out at 25°C and a humidity of 40 ± 5 RH%.

2.3. Characterisation of blanket deformability with silica-contaminated surface

We evaluated the deformation of the blanket during the reverse offset printing process using a silica-loaded glass substrate as follows: in the silica-loaded glass preparation, an SU-8 5 solution, which was diluted 1:1 by weight with PGMEA, was first spin-coated at 3000 rpm on a glass substrate. Before proceeding to pre-annealing, the SU-8 surface was exposed to silica-contaminated air to scatter the silica particles onto the corresponding surfaces. Here, we used poly-dispersed silica powders with diameters ranging from 5 to 40 μm (MSV-25, Tatsumori, Japan). After the particle deposition, the sample was pre-annealed, exposed by UV and post-annealed to adhere the particles. The thickness of the SU-8 layer was 0.7 μm. In the deformability tests, the Ag NP ink was first uniformly coated onto the blanket, and then transferred to the above silica-contaminated glass substrate using the same setup described in section 2.2. The indentation lengths were differentiated from \(-20, -80 \) and \(-200 \mu m \), and the printing speed was set to 5 mm s\(^{-1} \). We measured the diameters of silica particles and defects encompassing the centric silica particles using an optical microscope (MX51, Olympus, Japan). We also obtained a scanning electron microscope (SEM) image.

3. Results and discussion

3.1. Estimation of blanket deformation from particle defects

Figure 2(a) shows the SEM image of a defect formed around a silica particle after ink transfer (section 2.3).
From the figure, we see that the Ag NP ink was transferred to the top of the silica particle and a region outside the defect. This observation strongly indicates that the deformability of the blanket is not sufficient to complianlty contact with the peripheral of the silica particle, and there was room between the blanket surface and circumference of the silica particle under the transfer process (figure 2(b)). To estimate the degree of the blanket deformation, we evaluated the dependence of the defect size formed around a silica particle on its silica size using 30 samples (figure 2(c)). It is clear that the defect size increased almost linearly with particle size. Further, the defect size for a given particle size decreased as the indentation length increased. The estimated slopes of the defect size/particle size were 3.8, 3.5 and 2.8 for indentations of \(-20\), \(-80\) and \(-200\) \(\mu\)m, respectively (inset of figure 2(c)). This tendency is again in line with the contact mechanics prediction, where an increase of the indentation lengths results in a greater deformation of the blanket. Therefore, the contact area of the blanket increases (the defect size becomes smaller).

### 3.2. Effect of push-pull process on pattern generation

Figure 3 shows Ag NP patterns of the wide feature printed by the push-only process with indentation lengths \(\Delta_p\) of \(-20\), \(-10\) and \(0\) \(\mu\)m and by the push-pull process with final blanket positions \(\Delta_p\) of \(-20\), \(-10\), 0, 10 and 20 \(\mu\)m. We tested patterns that were printed in mutually opposite directions. The average thickness of the Ag NP patterns was \(90 \pm 10\) nm. For the cases with \(\Delta_p = -20\) and \(-10\) \(\mu\)m (pushing conditions), both the push-only and push-pull processes resulted in a severe contact-defect formation in the areas with widths of 3 and 4 mm. Those results are reasonable because the printing pressures applied during the patterning process were identical for those methods, and the depth of the cliché was only 2.6 \(\mu\)m. However, considering the previous results on particle defects where the critical defect/particle-size ratio was about 3.8 for an indentation length of \(-20\) \(\mu\)m, it was unexpected that 1 and 2 mm patterns can be successfully generated as the ratio of the width/depth is 385 (1 mm/2.6 \(\mu\)m). Although it is not possible to make a straightforward interpretation as the three-dimensional deformation of the blanket in motion is significant, we speculate that the finite thickness effect of the incompressible elastic PDMS in deformation characteristics [34], size effects of the raised surface of the cliché, a stress-strain hysteresis of the blanket and/or local shear stresses exerted at the interface between the blanket and the cliché during the patterning process may have been involved.

In the case of \(\Delta_p = 0\) \(\mu\)m (the kiss touch position), we observed contact defects at the edges of the pattern in the both cases. Because the kiss touch position defined here is a position for which the pushing force was first detected with a precision of 5 \(\mu\)m (see section 2.2), and therefore a finite indentation may have already occurred, it is possible that it resulted in the presence of contact defects. For the cases with \(\Delta_p = 10\) \(\mu\)m (pulling condition) in the push-pull process, the pattern was successfully generated without any defect formations. We also demonstrated successful patterning even when the patterns were in the opposite direction. When printing in the opposite direction, the engraved area in the cliché gradually narrowed from 4 to 0.8 mm as the patterning proceeded. Therefore, we can deduce that in the push-pull process, the blanket can come into contact with a raised area of the cliché, even if it had previously passed through an engraved, non-contacting area of the cliché. This result indicates that the push-pull
process may be feasible, even for more complicated shapes. In the last case, i.e. $\Delta_p = 20 \, \mu m$, the blanket and the cliché were separated during the patterning process, and the pattern generation failed. Therefore, we determined that the upper limit of the pulling window in the push-pull process is within the range of 10–20 $\mu m$.

Figure 4 shows the temporal change of the detected load divided by the cliché’s width of 100 mm during the push-pull process and subsequent patterning process with the condition of $\Delta_p = 10 \, \mu m$, as shown in figure 3. In the push-pull process (B–D), negative (pushing) pressure is exerted on the cliché, and the pressure was then inverted to a positive value as the pull-back process continued (note that the load cells were placed upside down, see figure 1(c)). This pulling force is due to adhesion, and was maintained after the pulling process had finished (E). Further, we detected the positive adhesion forces throughout the patterning process, where the stage and the blanket roll moved together (F). This result strongly supports the fact that the adhesion is the underlying mechanism of the ink-layer transfer in the push-pull process. Note that the gradual change of the applied forces under the constant indentation length found in C and E, for example, in figure 4, is caused by the stress relaxation of the blanket’s polyurethane cushion.

To demonstrate the validity of the push-pull process for the patterning of a fine and complicated design, printability of the interdigitated patterns were also investigated. Here, printing speed was set to 20 mm s$^{-1}$ and other conditions are same as the previous pattern. Figure 5(a) shows Ag NP patterns with $L/S = 50 \, \mu m$ printed by the push-only process and the push-pull process. In the case of the push-pull process with a final blanket position $\Delta_p = 5 \, \mu m$, a perfect pattern structure was generated with no apparent contact defects (we confirmed that the blanket did not touch over the entire area of the cliché when the push-only process was conducted with $\Delta_p = 5 \, \mu m$). This result again proved that the design of the cliché amenable to the push-pull process is not regulated by its shape complexity. On the other hand, contact defects at T-type corners as well as pads were found in the cases of the push-only process with the indentation lengths $\Delta_p$ of 0, −10 and −70 $\mu m$. Complementary height images of the corresponding areas obtained by an optical surface profiler (NewView 6000, Zygo) were also shown (the height scale was calibrated with absolute height information taken by a stylus profilometer). As can be seen, highly uniform Ag NP layers were formed by virtue of reverse offset printing. To quantitatively evaluate the validity of the push-pull process, the number of the defects appeared at the T-type corners of the interdigitated patterns (total inspection points of 200 for each experimental condition) was counted (figure 5(b)). As can be seen, rates of defect formation increased with the indentation lengths (58%, 87% and 100% for $\Delta_p$ of −0, −10 and −70 $\mu m$, respectively), while no defects were observed in the push-pull process. We add that the interdigitated patterns with $L/S = 10 \, \mu m$ and 20 $\mu m$ were also successfully printed without any defects by the push-pull process (images not shown). From those results, we conclude that the proposed push-pull process is valid in the pattern generations of both fine and wide features.

![Figure 4](image-url)
3.3. Estimation of blanket-ink layer adhesion

In this section, we estimated the adhesion forces between the SU-8 surface, pristine blanket surface and Ag NP ink-coated blanket using the same machine setup described in the previous sections. In this test, we pushed the blanket roller down from the position of \( \Delta_p = 10 \mu m \) to \(-30 \mu m \), and then pulled back until the contact interface is separated. As a result, in the cases of both the pristine blanket and the ink-coated blanket, the detected forces were almost identical to the pushing regions down to the position of \( \Delta_p = -30 \mu m \) and the pulling regions up to the position of \( \Delta_p = 10 \mu m \) (figure 6). However, in the case of the ink-coated blanket, at \( \Delta_p = 20 \mu m \), the adhesion force decreased sharply to zero, and accordingly, the blanket was separated from the SU-8 surface. This behaviour is consistent with the patterning results shown in section 3.2, as patterning had failed at the pulling condition of \( \Delta_p = 20 \mu m \). On the other hand, in the case of the pristine blanket surface, the adhesion was retained up to \( \Delta_p = 50 \mu m \).

Note that in the ink-coated blanket case, the size of the ink layer (nip) which was transferred to the SU-8 surface after experiencing the maximum pushing force of \(-1.97 N \) at the indentation of \( \Delta_p = -30 \mu m \) (figure 6) was 100 mm \( \times \) 1.2 mm. The average printing pressure calculated from those values is 16 kPa. This experimental value agrees with the predicted value of the printing pressure at \( \Delta_p = -30 \mu m \), i.e. 17 kPa, which was calculated from the Young modulus of the 0.58 mm thick cushion, i.e. \( 3.5 \times 10^2 kPa \). Therefore, we confirmed that the load cell system prepared in this study adequately monitored the forces experienced by the blanket.

4. Conclusions

In the present study, we demonstrated the validity of the push-pull process for contact defect-free formations of wide patterns with a size of up to 4.0 mm \( \times \) 5.0 mm and of finer patterns with size of 10–50 \( \mu m \) width lines using the cliché with 2.6 \( \mu m \) and 1.6 \( \mu m \) shallow depths, respectively, which was inaccessible using the conventional push-only process of reverse offset printing. We also proved that the push-pull process is valid even for such patterns where the engraved area in the cliché gradually narrows from the starting point to the ending point and even with an interdigitated structure during the pattern-generation process. This aspect is important, as the push-pull process is potentially amenable to more complicated shapes. Further, the pulling margin was up to around 10 \( \mu m \), and this value was found to be controlled by the maximum adhesion force between the ink-coated blanket and the substrate. We believe that the proposed method may be especially beneficial for a wet-etched cliché for which isotropic etching is used for...
engraving because the resulting minimum dimension of the pattern unavoidably increases with the depth, thus limiting the pattern-design flexibility. Further, we anticipate that our method may be useful for applications that require a high overlay accuracy, as a finite indention of the blanket in the conventional push-only process often causes a pattern deformation because of a mismatch in the rotational and translational motions of the blanket roller and the stage. This is due to the reduced superficial speed of the compressed blanket roller [35], as well as a pattern-width reduction due to undesired contact at the edge of the cliché’s grooves [29]. A roll-to-sheet printing was adopted in the present study and thus a further investigation is required to apply for a roll-to-roll printing. Regarding reverse offset printing, it is not problematic to add the push-pull motion in the sequence because the intermittent stop of a cliché roller for the push-pull step does not affect the motion of a target roller as the two rollers are separated by a blanket roller. However, the application of the push-pull process to the microcontact printing in which a stamp roller directly comes into contact with a target roller, badly influences the continuous film conveyance. For such a case, a modified roller consisting of a raised launching strip (10 μm height, for example) at the starting position of contact might be used to imitate the push-pull process without the vertical motion of the rollers; namely, a contact onto the launching strip corresponds to the process B and C in figure 4 and the subsequent rotating motion corresponds to the pattern generation process with negative indentations (pull process D to F in figure 4).

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