Fabrication of Roll-Printed Organic Thin-Film Transistors using Patterned Polymer Stamp

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Abstract

Roll-printed organic thin-film transistors (OTFTs) were fabricated by gravure or flexography printing using patterned PDMS stamp with various channel lengths, silver pastes, coated polyvinylphenol dielectric, and jetted bis(triisopropyl-silylethynyl) pentacene semiconductor on plastic substrates. The roll-printed OTFT parameters were obtained: field-effect mobility of 0.1 cm^2/Vs, an on/off current ratio of 10^4 and a subthreshold slope of 2.53 V/decade.

1. Introduction

Since the early 2000s, scientists and engineers have succeeded in applying printing-related techno1ogies to create low-resolution organic electronics devices with micron-sized features [1]. The graphic arts printing equipment and printing processes must also be improved and adapted to meet the rather strict design rules for electronic circuits. Typical resolution capability of standard offset, gravure, screen, or inkjet printing is of the order of 50 ~ 100μm. Clearly, these are not necessarily fundamental limitations. This field of application requires pattern and overlay accuracies down to 20 μm for high-quality reproduction. In particular, roll printing is the collective name for traditional printing, which has been developed as alternative to conventional methods and as fabrication technology for microfabrication [2].

In this paper, we report in more detail a related study in which a roll-printed OTFT was used for the fabrication of printed electrodes and soluble organic semiconductors on various plastic substrates. Gate, source, and drain electrodes of roll-printed OTFT were fabricated by a high-resolution printing technique based on transferring a pattern from a patterned poly(dimethylsiloxane) (PDMS) stamp to a plastic substrate by physical contact using high-conductivity Ag pastes. A roll printed OTFT with a polyvinylphenol (PVP) as polymeric dielectric layer was formed using a spin-coating [3,4], and a bis(triisopropyl-silylethynyl) pentacene (TIPS-pentacene) as an organic semiconductor layer was used in the ink-jet printing [5]. This is an attempt to enhance the accuracy of traditional printing to a precision comparable with optical photolithography, creating a low-cost, large-area, and high-resolution patterning process.

2. Fabrication of roll-printed OTFT

To fabricate a high-resolution and large-area printed OTFT, the following steps were performed: the design and manufacture of a engrave plate, the fabrication of a PDMS stamp, and a roll printing process.

In this work, we fabricated master patterns by electron-beam lithography onto a quartz substrate. The channel lengths (L) of the unit elements were split between 10 and 80 μm, and line width (W) between 200 μm and 2 mm on the engraved plate with the pattern designed for 40 elements of different channel lengths and line widths to be placed.

The PDMS stamp was fabricated by mixing Sylgard 184A (silicone elastomer base) with Sylgard 184B (silicone elastomer curing agent) at a ratio of 10:1. It was poured onto the mask and air bubbles were removed by a vacuum pump. It was cured in a thermal dryer at 80 °C for 1hour. Then, the composite stamp was carefully peeled off from the master surface.

Figs. 1 and 2 show the schematic fabrication process of roll-printed OTFTs by gravure or flexography printing using patterned PDMS stamp with various channel lengths and line widths, and with low-resistance Ag paste. The electrodes of roll-printed OTFT were fabricated by changing the print heads with patterned PDMS stamp using both gravure and flexography printing. As shown in Fig. 1, the roll-printed electrodes were manufactured using gravure printing by the following process: a flexible plastic substrate, whose surface was hydrophilic and treated with plasma, was mounted on a flat plate; Ag paste was then poured into the patterned PDMS stamp surface; the paste was inked into the patterned PDMS stamp using the doctor blade; finally, the gate electrode was formed by forwarding and rotating the rubber roller wrapped patterned PDMS stamp. A polymeric dielectric of 10 wt% PVP having a thickness between 4000 Å and 7000 Å was formed on the fabricated gate electrodes using spin coating; it was then cured at 80 °C for 10 min. The rubber roller was replaced with a source and drain electrode patterned PDMS stamp. The substrate formed with the gate electrode and polymeric dielectric layer was then mounted on the flat plate again. We fabricated source and drain electrodes using the patterned PDMS stamp of dimensions of 150 × 150 × 3 (±0.5) mm^3 between with 10 and 80 μm channel lengths and differing channel widths and pattern shapes, and then it was replaced on roller wrapped PDMS stamp physically. The source and drain electrodes were formed by inking Ag paste onto the patterned PDMS stamp, doctor blading, and then roll printing. Finally, to form the organic semiconductor layer on the fabricated contact electrodes, we used ink-jet printing from a 1 wt% solution of TIPS-pentacene in chlorobenzene. The thin-films were formed by annealing at 60 °C for 5 min in air.

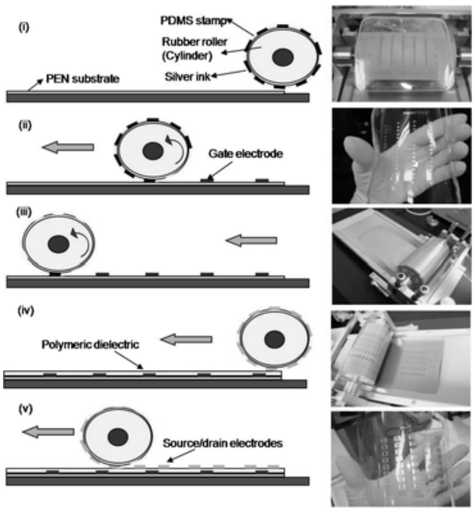


Fig. 1. Fabrication of roll-printed OTFTs using gravure printing process.

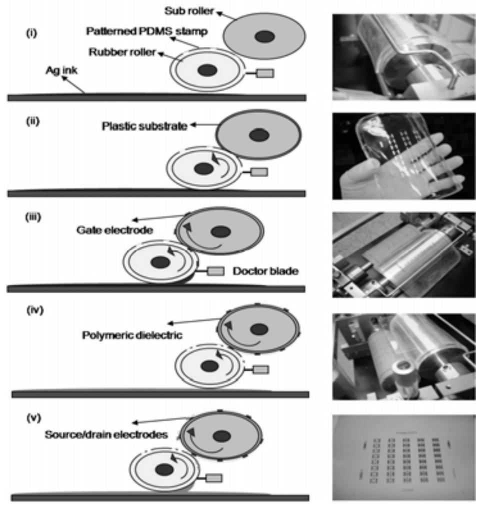


Fig. 2. Fabrication of roll-printed OTFTs using flexography printing process.

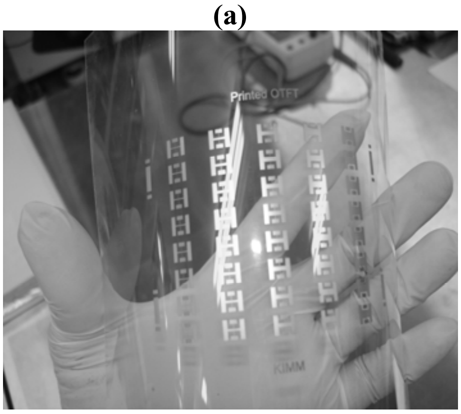
Fig. 2 shows the fabricating process of the printed OTFTs using flexography printing, where the pattern is transferred by contact printing to the sub roller with wrapping a plastic substrate from the rubber roller with patterned PDMS stamp. The conditions for forming the electrode are a doctoring speed of 16 m/min and a patterning speed of 24 m/min. The roll-printed OTFT was fabricated using the same method as the gravure printing process.

3. Experimental and results

Figs. 3(a) and 3(b) show the respective actual rubber roller wrapped patterned PDMS stamp and the roll-printed OTFT devices with various channel lengths between 12.6 (13.4) and 76.7 (72.1) μm, with various channel widths and spaces between patterns. Fig. 3(a) shows that the patterned PDMS stamp has a replicated pattern exactly corresponding to the master pattern and has a high accuracy of filling and releasing. Also, as a surface characteristic, by measuring the wettability, it was demonstrated that the patterned PDMS stamp was hydrophobic with a contact angle of 110 °, and a surface energy of 22 dyne/cm. The adhesion force was 12 nN, which was very low. Fig. 3(a) shows the result for a patterned PDMS stamp of gate electrodes (see the inset). Fig. 3(b) shows an image of a printed OTFT array using TIPS-pentacene as the semiconductor, PVP as the gate dielectric layer, and Ag paste gate, source and drain electrodes.

The pattern transfer experiments were performed using DG-1502 or TEC-I010 as roll printing Ag pastes; these pastes were applied onto a surface treated plastic substrate using the rubber roller. With regard to the electrical characteristics, measurements of the conductivity, confirmed that the roll-printed electrode had a higher conductivity with a sheet resistance of 0.052~0.0211 Ω/□ and line resistance of 0.6~0.3 Ω/cm, which was very low. The surface properties were also enhanced, as revealed by a measurement of the surface roughness (Ra) that changed from ~1 μm to less than 500 nm (not shown here). The measurements revealed that the surface properties and performance characteristics of roll-printed OTFTs, such as the roughness, sheet resistance, line resistance, and density, were improved.

(a)

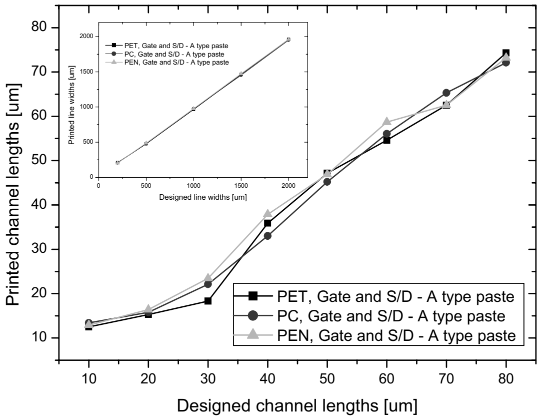


(b)

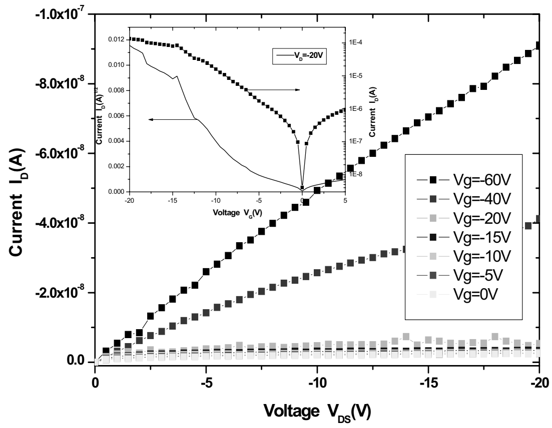
Fig. 3. Images of fabrication of (a) patterned PDMS stamp and (b) roll-printed TIPS-pentacene OTFTs.

Fig. 4 shows the a graph of the measurement results of variation characteristics including the designed channel lengths versus the printed channel lengths plot for roll-printed OTFTs. Figs. 4(a) and 4(b) show the respective results of the variation between the reproducibility of printed patterns obtained by gravure printing and by flexography printing; these results were obtained by analyzing cases in which systems have both line widths and channel lengths. Fig. 4(a) shows the gravure printed OTFTs with channel lengths between 12.6 and 76.7 μm (designed L = 10 to 80 μm) and channel widths between 204.3 and 1965.6 μm (designed W = 200 to 2000 μm). Figure 3(b) shows to flexography printed OTFTs with channel lengths between 13.4 to 72.1 μm and channel widths between 211.2 to 1995.2 μm for Ag source and drain contact electrodes on various gate electrode patterns. Fig. 4(b) also shows that the roll-printed electrodes have a transferred pattern exactly corresponding to the engraved plate and they have a higher accuracy of transferring onto plastic substrate as compared to the electrodes shown Fig. 4(a). The roll-printed electrodes were showed high fidelity and good reproducibility above 40 μm. The channel length deviations for 40 to 80 μm patterns were less than -10 %. However, the channel lengths for 10 to 30 μm patterns increased by -20 ~ -30 % due to Ag paste diffusion and shrinkage, which occurred during transferring, printing, and curing between the Ag paste electrode and polymeric dielectric layer or plastic substrate interface. As compared to Fig. 4(a), the distribution of channel length variation within each pattern in Fig. 4(b) is a little bit different, which known to be because of the ink transfer mechanism, coefficient of thermal expansion (CTE), and surface roughness of the polymeric dielectrics used.

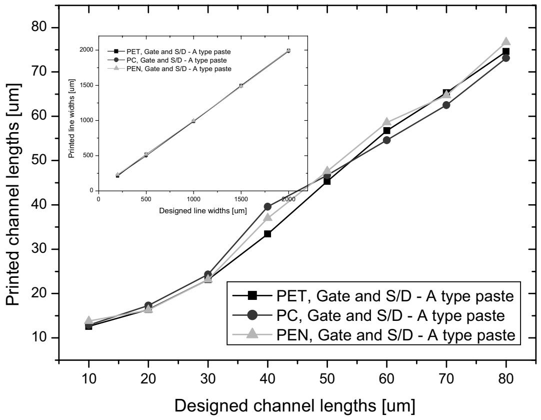
Roll-printed OTFTs were fabricated by a near room temperature process and characterized in air. Figs. 5(a) and 5(b) show the measurement results for the transfer and output characteristics, respectively. Fig. 5(a) shows a typical plot of the output characteristics at various gate voltages V_{GS} for drain current I_{DS} versus drain voltage V_{DS}, as well as a graph of the transfer characteristics; these characteristics include an I_{DS} versus V_{GS} plot and an $|I_{DS}|^{1/2}$ versus V_{GS} plot (shown in the inset), and correspond to a roll-printed OTFT using gravure printing with a channel length of L = 16 μm (designed L = 20 μm), width of W = 477 μm (designed W = 500 μm), 1 wt% TIPS-pentacene as the semiconductor, 5600 Å spin-casted PVP as the gate dielectric, an Ag paste gate, and source and drain electrodes. The field-effect mobility was 0.08 cm^2/Vs, while the threshold voltage was about –3.54 V. The on/off current ratio was above 10^4 when V_{GS} was scanned from –20 to +5 V. Fig. 5(b), which corresponds to a roll-printed OTFT using flexography printing, as shown in Fig. 5(a), the following parameters were obtained: field-effect mobility as large as 0.1 (±0.02) cm^2/Vs, on/off current ratio of 10^4, a subthreshold slope of 2.53 V/decade, and a threshold voltage of –3.54 V.



(a)

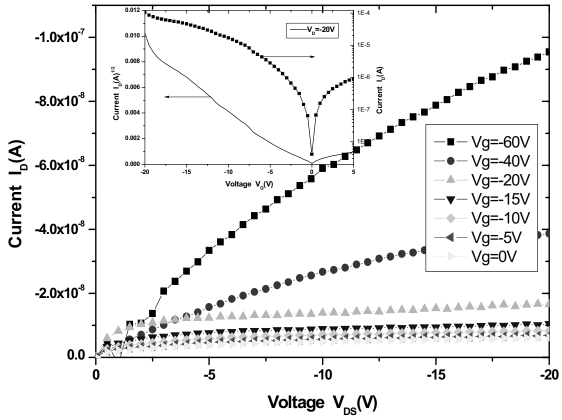


(a)



(b)

Fig. 4. Variation characteristics of designed channel lengths versus printed channel lengths for source and drain electrodes of (a) gravure and (b) flexography printed OTFTs.



(b)

Fig. 5. The drain current I_{DS} versus drain voltage V_{DS} output characteristics of roll printed TIPS-pentacene OTFT on PEN substrate in the (a) gravure printing and (b) flexography printing.

4. Conclusions

The roll-printing using patterned PDMS stamp for Ag paste transfer patterning made it was possible to fabricate printed OTFT with a channel length as small as 12 μm on plastic substrates, which had been hardly patterning in the previous traditional printing. The number of steps in the proposed fabrication process was reduced by 20 steps as compared to that in conventional fabrication techniques. Further, it was established that the roll-printed OTFT was completely fabricated due to the effect of the contact between the Ag paste electrode and the plastic substrate and polymeric dielectric layer. In our proposed method, it is necessary to optimize the surface treatment process, accurately control the printing conditions, and the fabrication processes including the curing temperature and the curing time.

5. References

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