40.2: Super-High Resolution Transfer Printing for Full-Color OLED Display Patterning

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Abstract
We describe a transfer printing method for patterning of thin polymer layers. A hard stamp with raised feature is brought into contact with a spin coated organic film under elevated pressure and temperature to break the films. The patterned film is then transfer printed onto devices. We use this method to print red/green/blue sub-pixel arrays with pattern size as small as 12 μm at a resolution of 530 ppi to demonstrate its ability for full-color organic light-emitting display fabrication.

1. Introduction
An important challenge for Organic Light-Emitting Diodes (OLED) manufacturing is to find a simple and low-cost method for patterning the organic materials used for different colors. Conventional photolithography is usually not capable of patterning organic materials since the solvent used in the process may degrade the materials to be patterned. Ink-jet printing (1; 2; 3; 4) has been widely used but it suffers from potential drawbacks such as poor resolution, low throughput and non uniform thickness. Other alternative patterning methods demonstrated includes laser-induced thermal imaging (5; 6) and laser-induced sublimation transfer. (7; 8) With laser-induced thermal imaging sharp edge patterns are difficult to achieve for polymer layers, and laser-induced sublimation can only be used to pattern small molecules. In this paper we demonstrated a transfer printing technique over a large area to pattern active polymer layers and OLEDs with the transferred layers are fabricated and tested.

2. Transfer Printing
Figure 1 presents the procedures of our transfer printing patterning technique. The process begins with the fabrication of a hard stamp [Figure 1a], where the raised features correspond to the final regions without the polymer on the final device plate. A silicon wafer is used for the hard stamp and conventional lithography and etching is used to make it. In this work, square patterns of two dimensions (12 μm × 40 μm, 24 μm × 80 μm) are etched into the stamp to a depth of ~700 nm. A second flat plate, also silicon for convenience, is coated with a 1.2-μm poly(dimethylsiloxane) (PDMS) layer and used as a backing layer for the active polymer layer to be patterned.

Control of surface energies for transfer printing is critical. The PDMS film is oxygen plasma treated for 5-10 seconds in order to make the surface hydrophilic so that spin coating is possible. (9) Longer plasma treatment, however, will make the PDMS surface too sticky and polymer layer would not detach easily from PDMS layer during printing steps.

For the active light-emitting organic layer, a mixture of polymer and small molecules is used. Poly(9-vinylcarbazole) (PVK) (Mw=1.1M, Tg=200ºC) is used as the host polymer. It is doped with 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) for electron transport and the dyes nile red, coumarin-6 or coumarin-47 for red, green and blue color tuning, respectively. (10) The entire blend is spin-coated onto the PDMS-coated silicon substrate, using chlorobenzene as the solvent to yield a thickness of 70-90 nm.

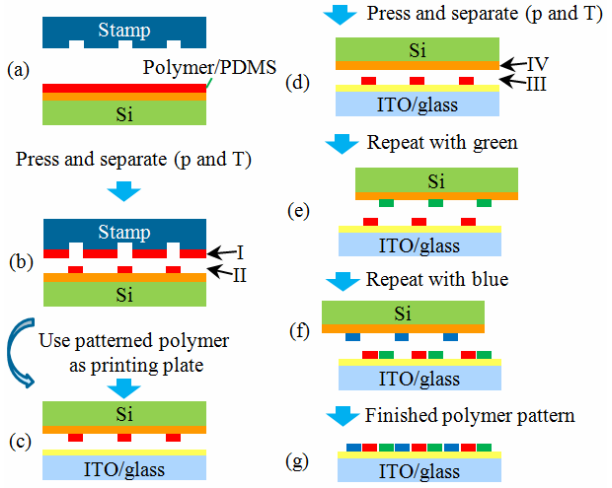

Figure 1. Schematic illustration of transfer printing for full-color display patterning.

The stamp is then pressed against the polymer film under raised temperature of 80-150ºC for 2-6 minutes at a pressure of 50-400 psi [Figure 1b]. As a result the polymer film breaks at the stamp edges. Upon separation, polymer film in contact with the raised regions of the silicon stamp is transferred onto the stamp and a complementary pattern is left on the PDMS side.

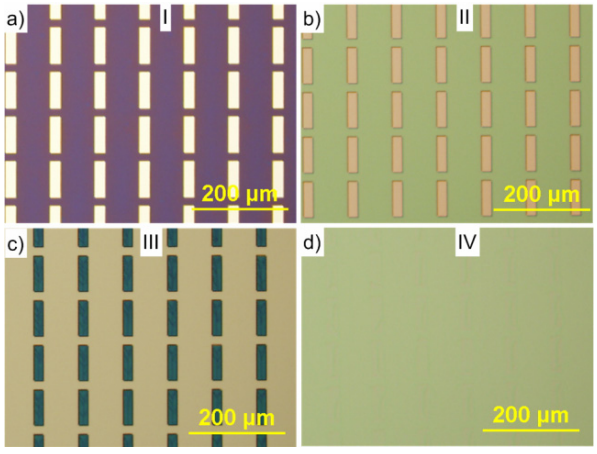

Figure 2. Optical micrographs of polymer patterns at different steps.

Figure 2a gives a typical polymer pattern formed on the hard stamp surface at this step (label "I" in Figure 1). The squares shown, with a size of 24 μm × 80 μm, are regions with exposed silicon and the rest is covered with polymer film. Figure 2b is the polymer pattern (squares) left on PDMS/Si substrate (label "II").

Next, by a similar press and separate step, the patterned polymer film is transferred from the PDMS onto a device plate [Figure 1c-d]. In our work the same temperature and pressure conditions are used for the two transfer printing steps for simplicity [Figure1 b and d]. Figure 2c is a micrograph of polymer squares arrays after they have been transfer printed onto a device plate (silicon wafer used as a demo) (label "III"). After the second transfer step, the PDMS/Si substrate is free of polymer films, as shown in Figure 2d (label "IV"). Finally for full-color OLED display patterning, we only need to repeat the previous steps twice so that three (red, green and blue-emitting) polymer blends are created [Figure 1e-g].

The edges of the patterned polymer film are very sharp (Figure 2c), in contrast to those laser induced thermal imaging, which shows ruggedness on the order of 10 μm for a 80 μm stripe. (5) Both the stamp (after stripping of polymer film) and the PDMS/Si plate are reusable, making the process a potentially low-cost technique.

A similar two-step method has been demonstrated to pattern passive polymer, such as polymethyl methacrylate (PMMA) and polyvinyl acetate (PVAc). (9) However, in their work the electrical properties of the transferred layers were not measured and no sequential printing of different layers onto a single substrate was demonstrated.

3. Printing Results

Figure 3 demonstrates the ability of the transfer printing method to pattern active organic layers required for full-color display by repeating the transfer printing steps with multilevel registration.

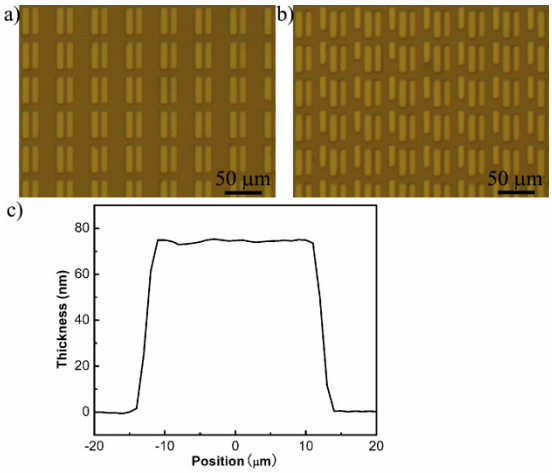

Figure 3. Optical micrographs of printed polymer patterns (a-b) and (c) surface profile of a typical printed polymer film.

Figure 3a is an optical micrograph after green and red light-emitting sub-pixel arrays have been deposited by transfer printing method. Figure 3b is an image after green, red and blue sub-pixel arrays (12 μm × 40 μm polymer pattern) have been transferred. The misalignment of the third layer was caused by poor manual alignment of the plates due to an equipment limitation. The RGB combined pixel has a pitch of 48 μm × 48 μm or a display resolution of 530 ppi. These results show that the method could be used to fabricate displays with a very high resolution. Results are similar for printing onto silicon (Figure 2), glass (Figure 3) or ITO-coated glass (for device performance test).

Figure 3c is a typical surface profile scan of a 75-nm transfer printed polymer film. The root-mean-square (rms) roughness is calculated to be ~1.2 nm from multiple measurements. This thickness uniformity is critical for a uniform light emission. In contrast, solution-based printing techniques usually give non-uniform layers which require further process steps for improved planarity. (2; 3; 4)

4. Device Performance

Since active materials are patterned in this work, it is essential to study the electronic properties of the organic layers after processing and the interface between the printed material and the underlying layers. We have fabricated OLED devices where the polymer blend film was transfer printed onto ITO-coated glass substrate followed by thermal evaporation of Mg:Ag (10:1) as cathode.

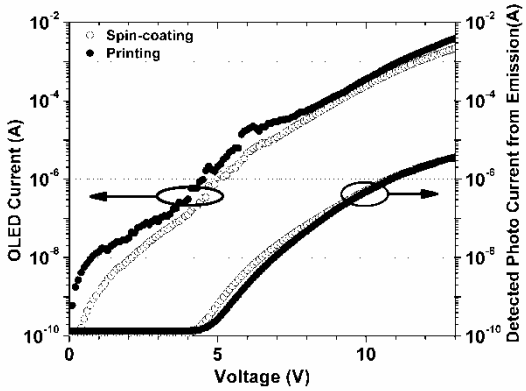

Figure 4. Comparison of I-V characteristics of OLEDs with polymer film deposited by transfer printing and spin-coating.

Both the current-voltage and light emission (converted to photocurrent by a silicon photodiode) -voltage characteristics of the printed OLEDs are similar to those of spin-coated devices for a printing condition of 90 ºC and 50 psi [Figure 4]. This is a reverse test of the electrical properties of ITO/printed organic interface, as the interface directly controls the hole injection into the polymer blend. Thus we conclude that the printing leaves the polymer layer and the ITO in intimate contact on an atomic scale.

5. Summary

In summary, a transfer printing method to pattern active polymer layers used for OLED application is demonstrated. Red/green/blue light-emitting sub-pixel arrays have been patterned by this method by three successive steps with a three-color pixel resolution of 530 ppi. Films deposited by this method are uniform in thickness. OLEDs with printed polymer layer show similar performance compared to spin-coated control devices, showing that hole transport occurs easily across the printed-organic-layer/ITO interface.

6. Acknowledgement

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7. References

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