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Multicolor organic LEDs processed by integration of screen printing and thermal transfer printing

Hsin-hua Chang ^a, Chung-chih Wu ^b, Cheng-chung Yang ^b, Chieh-wei Chen ^b, and Cheng-chung Lee ^a

^a Institute of Optical Sciences, National Central University, Chung-Li, Taiwan, 32054

^b Graduate Institute of Electro-optical Engineering and Department of Electrical Engineering,
National Taiwan University, Taipei, Taiwan 10617

ABSTRACT

Color integration in organic LEDs (OLEDs) on a substrate has always been a challenge due to the incompatibility of OLED materials with the conventional photolithography. In this paper, we report a process for the fabrication of large-area multicolor OLEDs of arbitrary patterns by combination of thermal-transfer printing and screen-printing. Thermal transfer printing is used to introduce color-tuning dyes into a thermally stable OLED polymer layer from a dye-dispersed polymer layer on the donor plate. Such a process permits controllable and uniform doping of a polymer layer over large areas. By using a patterned color donor plate, color integration in OLEDs could be accomplished with a single thermal transfer step. In this work, the source plate containing multicolor patterns is fabricated by screen-printing. The RGB color patterns were printed sequentially by using RGB inks prepared by dispersing Nile red, C6 and perylene into a commercial screen-printing paste. Based on these printing approaches, we have successfully fabricated multicolor single-layer and heterostructure OLEDs.

Keywords: Color integration, screen -printing, thermal-transfer printing

1. INTRODUCTION

In last decade, organic light emitting devices (OLEDs) based on small molecule organic materials or on polymer materials have been extensively studied because of their various advantages for flat panel display applications [1-2]. The capability of solution processing of polymers leads to manufacturing advantage for large-area coating. However, it is found difficult to integrate multicolor polymer materials or devices for full-color displays by conventional coating and sequential photolithography steps [3-4]. To resolve this issue, printing approaches have been aggressively pursued recently. For instance, ink-jet printing has been used to deposit directly patterned polymers or to introduce color-tuning dye patterns into/onto continuous buffer polymer layers [5-8]. However, ink-jet printing is in general a sequential process and might have a limit for substrate throughput [9]. Alternatively, a large-area thermal transfer process was recently proposed for color patterning of polymer layers in OLEDs [9]. Owing to a transfer mask inserted between the donor and the receiver films, this process involves several mechanisms: the sublimation of dye molecules from the heavily dispersed polymer film, travel of dye molecules to the surface of the receiver film, and the diffusion of dye molecules into the receiver film. Interaction of all these mechanisms has led to difficulty in controlling profiles of dye concentration and to complication of processing steps. For instance, long periods of annealing may be required to redistribute dye dopants in the receiver film after the transfer process.

In our previous paper [10], we report an effective transfer process for performing controllable doping of polymer films in OLEDs. In this process, the polymer receiver film is placed in direct contact with the dye-dispersed polymer donor film to permit direct dye-diffusion thermal transfer. We performed theoretical and experimental studies of this doping process and show that it can be modeled by Fick's diffusion theory under impermeable film-substrate boundary conditions. We have therefore named this process as finite-source dye-diffusion thermal transfer (FS-D2T2). We demonstrated doped-polymer OLEDs with device characteristics same as those made from the conventional blending process.

Using a source plate containing multicolor dye-dispersed polymer patterns, FS-D2T2 may be used to make multicolor OLEDs or color pixels in OLED displays. The color source plates may be fabricated with any patterning technique, ranging

from photolithography to printing, as long as the fabrication processing does not degrade the emission characteristics of dyes to be transferred. In this paper, we report the screen-printing process for the fabrication of color source plates for FS-D2T2. Screen-printing method is an additive and high-throughput patterning technique suitable for large areas. Using screen-printed source plates, we have been able to produce line width of $\sim 150\ \mu\text{m}$ in the receiver film and have successfully fabricated multicolor single-layer and heterostructure OLEDs based on FS-D2T2.

2. EXPERIMENTAL RESULTS AND DISCUSSIONS

2.1 Thermal-Transfer Printing – Finite-Source Dye-Diffusion Thermal Transfer (FS-D2T2)

The arrangement for performing FS-D2T2 is shown in Fig. 1. A pressure is applied on top of the receiver plate to ensure intimate contact with the source film. At an elevated temperature, the dye dopants transfer thermally from source polymer layer to receiver polymer layer. In our previous paper [10], we showed that FS-D2T2 is a reliable and effective method of performing controllable doping of polymer films in OLEDs. The characteristics of FS-D2T2 include: (1) it is a self-limiting process. There would be no concern of over-doping or consequent concentration quenching even for elongated diffusion time as long as concentration of dye in the source polymer is chosen to match the desired final concentration in the receiver. (2) Doping level is controllable by concentration of dye in the source polymer. (4) Devices using the FS-D2T2 films exhibited characteristics same as those using conventional dye-blended films.

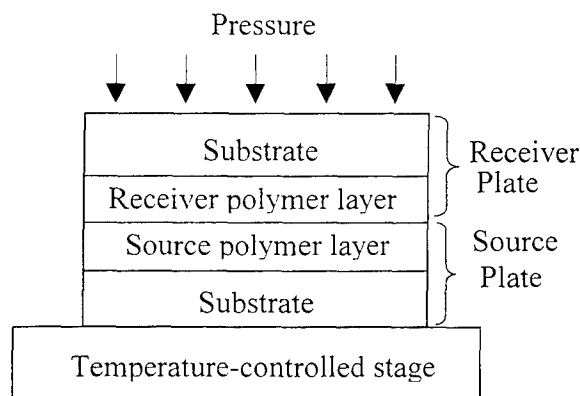


Fig.1. Arrangement for finite-source dye-diffusion thermal transfer.

2.2 Color Integration

Fig. 2 shows color integration process for OLEDs by FS-D2T2. Multicolor patterns are designed on the source plate. The source plate containing multicolor dye-dispersed polymer patterns may be fabricated with any patterning technique, from photolithography to screen-printing, as long as the fabrication processing does not degrade the emission characteristics of dyes to be transferred. By using a source plate containing color patterns, FS-D2T2 could be used to accomplish color integration for OLED devices or displays on a substrate in a single thermal transfer step.

One concern in this color integration process is the lateral diffusion of dyes. The edge definition of patterns will be influenced by the lateral diffusion effect. Since the diffusion length to get complete dye transfer is of the order of receiver

film thickness ($\sim 1000 \text{ \AA}$) in the FS-D2T2 process for OLEDs, the lateral diffusion would not hinder the accomplishment of $\sim \mu\text{m}$ resolution eventually by reducing feature sizes on the source plates.

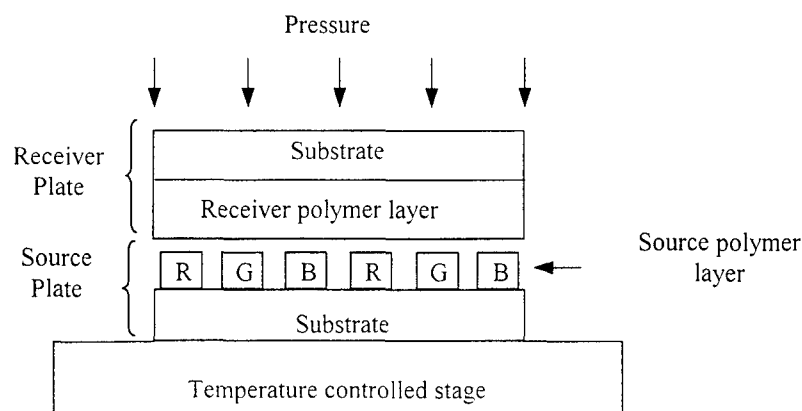


Fig. 2. Color integration of OLED by FS-D2T2

Another concern is the difference in the diffusion speed of different dyes used. For instance, Fig. 3 shows the diffusion coefficients of dyes C6 and perylene vs. temperature in a bipolar polymer matrix PVK:PBD (100:40 wt%), where PVK is a hole transport polymer and PBD is an electron-transport molecule. The diffusion coefficient of perylene is larger than C6 at the same temperature, indicating that perylene diffuses into PVK:PBD film more easily than C6. Since the FS-D2T2 is a self-limiting process and the final dopant concentration in the receiver films is controlled by the initial concentration in the source films, the difference in the diffusion speeds of different dyes would not impose difficulties in controlling the concentrations of different dyes.

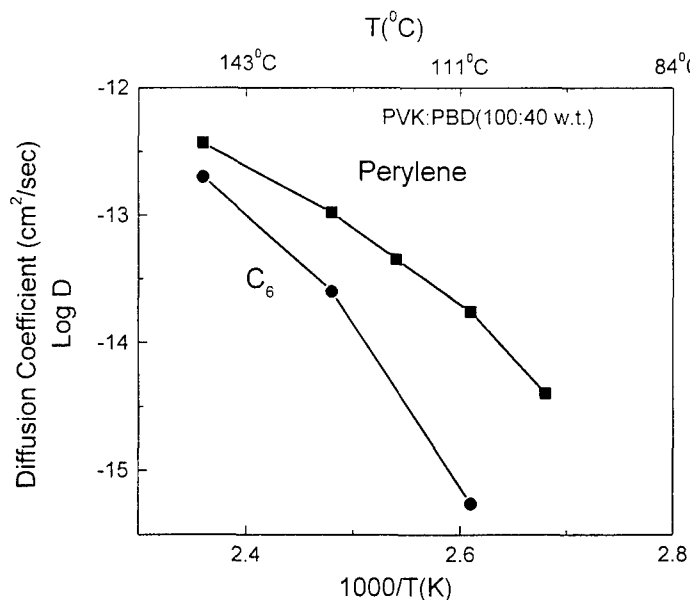


Fig. 3. Diffusion coefficients of C₆ and Perylene in bipolar polymer film PVK:PBD at different diffusion temperatures.

2.3 Multicolor Source Plate Prepared by Screen-Printing

Color source plate was fabricated by screen printing technique in this work. Screen-printing is a basic technology for thick film microcircuitry and inorganic electroluminescent devices. It is suitable for arbitrary large-area patterning. We applied this technology for the preparation of multicolor source plate needed in the FS-D2T2. The screen-printing process is illustrated in Fig. 4. Stainless-steel screen was chosen for better resolution and longer lifetimes than the polyester screen. The screen printer used in this work is manually aligned between sequential printings. Patterns on the stainless-steel screen are defined by photolithography of photoreactive emulsion over the screen surface. The screen printer applies the screen-printing paste evenly to the screen and rubbed it with a squeegee, which then pushes the paste onto the source plate substrate through the patterned openings in the screen. The screen printing paste is considered for a printable carrier of dye and is not thermally interactive with the receiver polyester film.

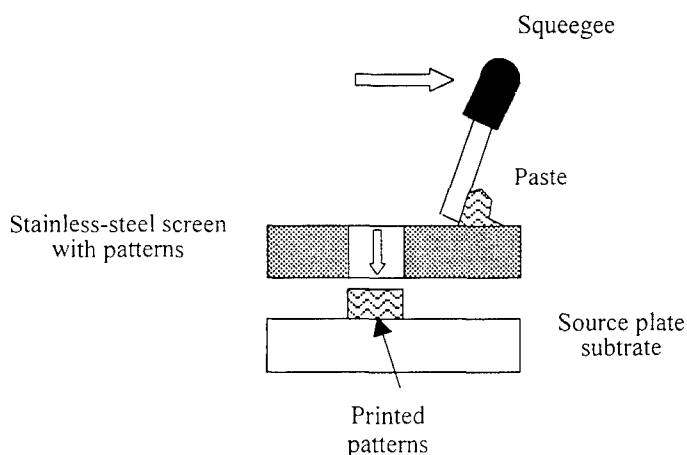
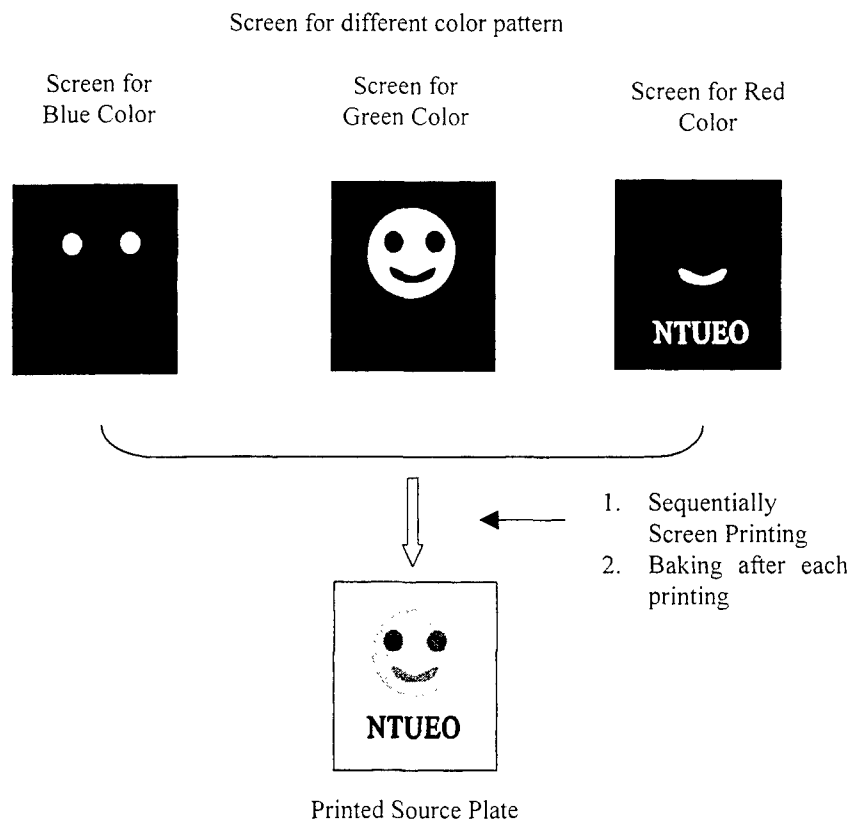


Fig. 4. Schematic representation of the screen printing process

Fig 5. shows how to prepare multicolor pattern source plates by sequentially screen sprinting. RGB inks were prepared by dispersing nile red ($\sim 0.006\%$ wt.), Coumarine6 ($\sim 0.01\%$ wt.) and perylene ($\sim 0.02\%$ wt.) into a commercial screen-printing paste (DuPont 7155), respectively. Patterns of different colors are separated onto different screens, and are sequentially printed onto the substrate. The alignment of different colors was achieved by designed alignment marks on each screen. After each printing, the substrate was baked at the temperature-controlled stage ($110\text{ }^{\circ}\text{C}$ 6 minutes) to harden and fix the pastes. With the manually aligned screen printer, a linewidth of $\sim 150\text{ }\mu\text{m}$ was achieved. However, a linewidth of tens of μm should be easily achieved by state-of-the-art semiautomatic or automatic screen printers..



Step 5 : Preparation of Multicolor Pattern Source Plate
by Sequentially Screen Printing

2.4 Multicolor Organic LEDs

By using a source plate containing color patterns, FS-D2T2 could be used to prepare multicolor organic polymer films for OLED devices or displays on a substrate in a single thermal transfer step. This multicolor layer may be the only active emissive layer in a single-layer OLED device structure, or it may be the emissive layer in a multi-layer OLED structure. In this paper, we demonstrated both types of multicolor OLED devices using the FS-D2T2 films.

The cross section of the device structure for the single-layer multicolor OLEDs is shown in Fig. 6. For this type of devices to work, the organic films must be able to conduct both holes and electrons. That is, the organic layer needs to be a bipolar layer. The hole-transport polymer PVK dispersed with ~25 wt.% of electron-transport molecules PBD has been used as the bipolar organic layer in this work. In addition, for regions of different colors in a same device to emit at the same time with comparable brightness, they all must have similar operation voltages and emission efficiency. Fortunately, for the RGB dyes, nile red, coumarin 6 and perylene used in this work, near their optimum concentrations, they all show similar electrical characteristics, permitting the realization of multicolor OLEDs. Such a multicolor OLED made from an FS-D2T2 film is shown in Fig. 7.

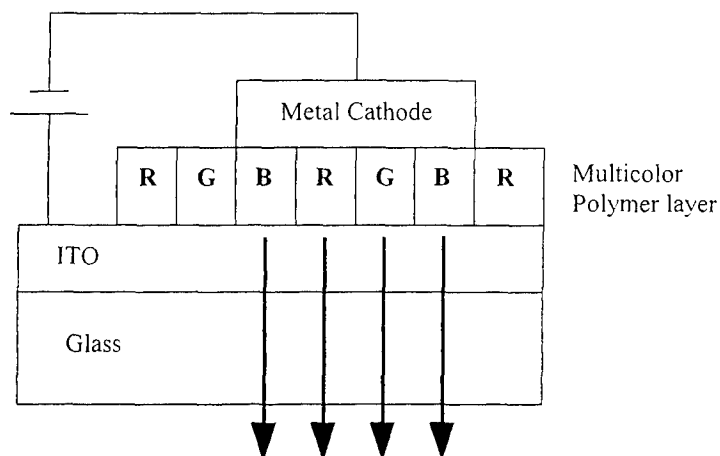


Fig. 6. Schematic illustration of operation of multicolor OLED device.

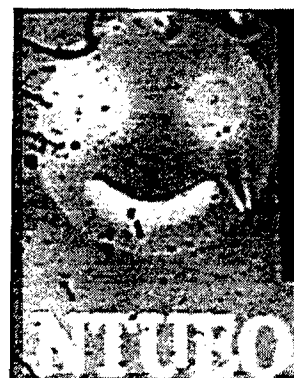


Fig. 7. EL Photograph of a Multicolor OLED from the FS-D2T2 film with single layer device structure.

The Cross section of a multicolor heterostructure device is shown in Fig. 8. The electron transport layer, Alq, and the metal cathode are vacuum evaporated after the FS-D2T2 process of the hole-transport polymer PVK. Fig. 9 shows EL spectra of the heterostructure OLED. Only emission from Alq₃ is observed in the PVK/Alq₃ structure, indicating that without dopants in the PVK excitons are formed and confined in the Alq layer. Compared with the PVK/ Alq₃ structure, we observed contribution from dopants in the doped-PVK/ Alq₃ structure. Since excitons are initially formed in the Alq layer, the emission contributions from dopants are through the interlayer energy transfer between Alq and dopants in PVK. This interlayer energy transfer is allowed since the Förster energy transfer mechanism is a long-range process. Making use of this interlayer energy transfer mechanism, a multicolor heterostructure OLED is thus achieved.

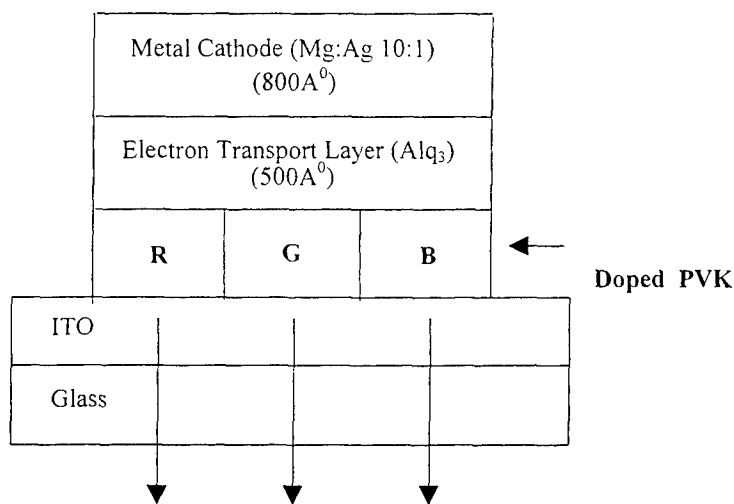


Fig. 8. Schematic cross section of multicolor heterostructure device.

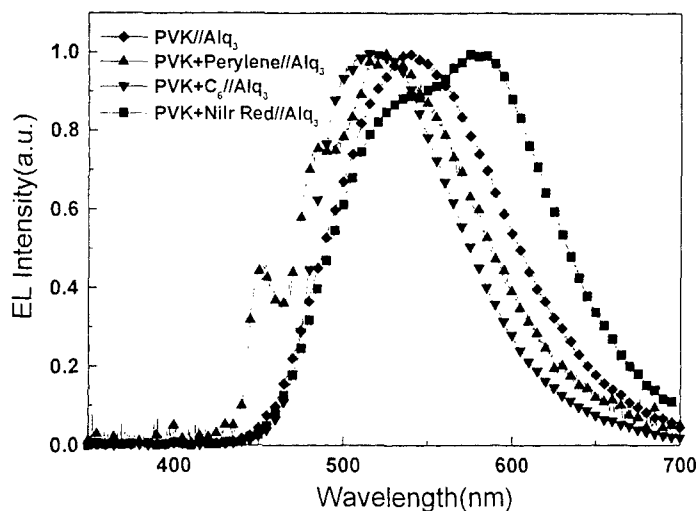


Fig. 9. EL spectra of multicolor heterostructure OLED from a FS-D2T2 film.

Although multicolors can be obtained from the above structure, the energy transfer is, however, incomplete through the interlayer energy transfer. In the future, a structure of introducing a hole-blocking and electron-transport layer between the electron-transport layer and the hole-transport layer may confine the formation of excitons within the PVK layer, ensuring complete energy transfer to the dopants and obtaining improved color purity.

3. CONCLUSION

We used FS-D2T2 to accomplish color integration for OLED devices or displays on a substrate in a single thermal transfer by using a source plate containing color patterns. Color source plate is prepared by screen-printing in this work. We have successfully fabricated multicolor single-layer and heterostructure OLEDs based on FS-D2T2 with screen-printing approaches.

4. ACKNOWLEDGE

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