All-Printed Flexible OLEDs

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Abstract

We have investigated printing techniques for processing organic light-emitting diodes (OLEDs). We succeeded to gravure print uniform organic thin films as well as screen print low work function cathode for OLED structure. Furthermore, by using roll-to-roll manufacturing methods, we have been able to fabricate all-printed flexible OLED demonstrator.

1. Introduction

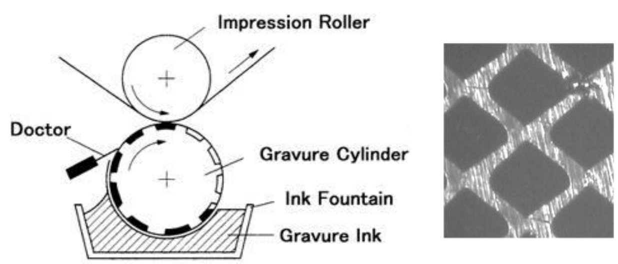
Organic electronics has grown into a promising candidate to replace some low end silicon applications. In detail, printed and organic electronics are expected to grow a remarkable business of 250 – 300 billion USD in 20 years.[1] In general, the fabrication is carried out in traditional and expensive batch processes like vacuum evaporation technique. However, a new interesting way is to apply roll-to-roll printing methods for the manufacturing of flexible organic devices.[2] Therefore, the processes with high through-put capability would decrease considerably the fabrication costs.

The printing techniques are currently under investigation for the roll-to-roll production of OLEDs in the 6th framework EU funded project entitled ROLLED - "Roll-to-roll manufacturing technology for flexible OLED devices and arbitrary size and shape displays".[3] The objective in the project is to fabricate an entire OLED structure by using roll-to-roll manufacturing methods and to examine, how the commercial production could be set up and integrated into an existing printing process. In order to attain a roll-to-roll compatibility, all the materials, inks and device structures need to be suitable for printing.

2. Experimental

Printing process

Fig 1(a) presents the scheme of gravure printing process. A gravure cylinder picks an ink from an ink fountain below and the ink fills the engraved cells (shown in Fig 1(b)). A doctoring blade takes off the excess ink and the gravure cylinder transfers the ink from the cells onto the surface of flexible substrate. Printing parameters in terms of doctoring blade loading, ink properties, gravure cylinder characteristics, the pressure between cylinder and impression roll, and printing speed define the amount of ink that is transferred from cells onto the surface of the moving substrate and thus, have a great impact on the thickness and quality of the printed film.



(a) (b)

Fig. 1. (a) Scheme of gravure printing process, and (b) image of engraved cells in the gravure cylinder.

Processing of OLEDs

In this process, the OLED structure consisted of a transparent anode electrode, a hole injection layer, a light-emitting layer, and a reflective cathode electrode. Commercial ITO coated PET (A=1”x1”) substrates with sheet resistance of 50 Ω/sq, were pre-cleaned in an ultrasonic bath of detergent, acetone, and isopropyl alcohol, respectively. Any patterning of anode as well as hole injection and emitting layers were not needed thus printed cathode layer defined active area of fabricated OLED pixels. On the top of ITO, PEDOT:PSS was gravure printed using optimized parameters of ink and printing process. PEDOT:PSS ink was modified with Tween®80, and isopropyl alcohol in order to have perfect wetting on the ITO substrate.[4] The printed film was subsequently dried in N_2 oven at 75°C for 2 hours. Yellow light-emitting layer, PFBT, was gravure printed on the top of PEDOT:PSS using 2.5 wt-% *p*-xylene solution. The optimum layer thicknesses of 40 nm and 70 nm for an efficient device were achieved for printed PEDOT:PSS and PFBT, respectively. Both layers were gravure printed using table-top IGT printability tester and engraved printing cylinder.

The preparation of aluminium (Al) cathode ink comprises ball milling of Al particles, and the addition of polystyrene in toluene solution.[5] The ink was screen printed on the top of the device in inert atmosphere to avoid oxidation of Al particles. 10 µm thick metal layer was achieved using 325 mesh count metal screen. Finally, printed OLED layers and pixels with the active area of 9 mm^2 were characterized.

Flexible all-printed OLED demonstrator was encapsulated using gas barrier coated flexible front and backside substrates in order to maintain its performance in air. The substrates were laminated under UV light using epoxy as glue.

3. Results and discussion

Hole injection layer, PEDOT:PSS

The printability of standard PEDOT:PSS was examined with various printing parameters and ink formulations. The printability trials showed visually that aqueous PEDOT:PSS solution is necessary to be modified in order to achieve appropriate ink for gravure printing process.

According to our printability tests, a surfactant, Tween®80, was found to be a good additive to enhance the wetting of the ink. Also, the solvent affects the fluid properties, the fluid flow and the way of drying, and thus, it has also a great influence on the morphology of the printed structure. Addition of isopropyl alcohol as a solvent improved significantly the wetting of the ink during printing process. 40 nm thick, smooth and pinhole-free film was achieved with 70 l/cm gravure cylinder containing the cell depth of 35 µm using modified ink containing 73.8 wt-% of PEDOT:PSS, 1.2 wt-% Tween®80, and 25 wt-% isopropyl alcohol.[4] The white light interferometer (WLI) 3D surface profile and an image of the film are presented in Fig 2(a) and (b), respectively.

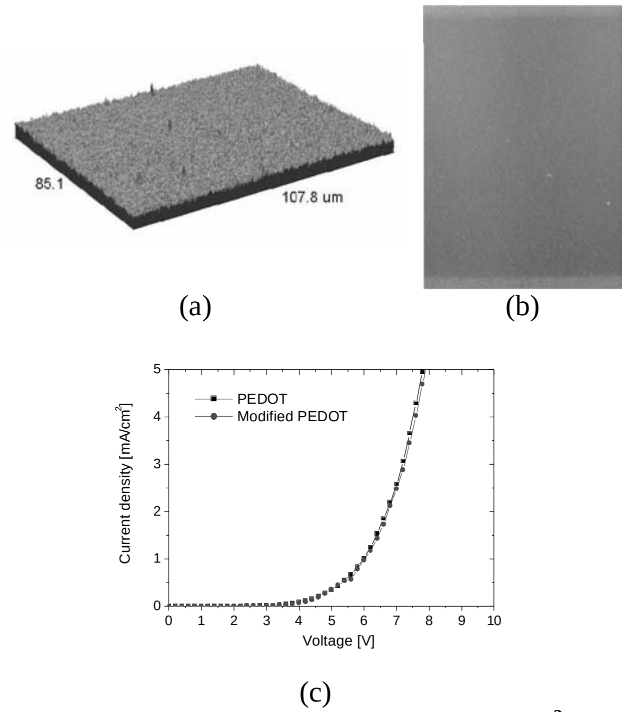
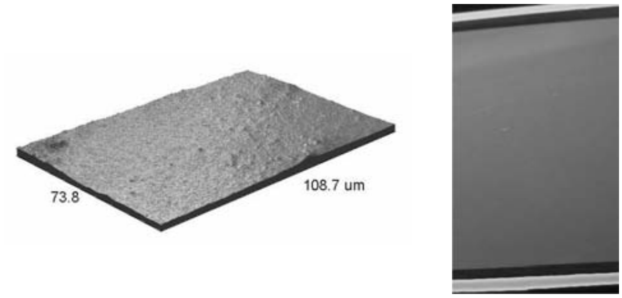


Fig. 2. (a) 3D surface profile, and (b) 3x2 cm^2 size image of modified PEDOT:PSS film. The comparison of (c) I-V characteristics of standard (■) and modified (●) PEDOT:PSS based OLEDs.

Any effect of modified PEDOT:PSS ink on device electrical characteristic was not obtained compared to standard PEDOT:PSS as plotted in Fig. 1(c). In both devices, the current density started to increase after 3.5 V. At the forward bias of 7V, the current densities of 2.6 mA/cm^2 and 2.5 mA/cm^2 were obtained for standard and modified PEDOT:PSS based OLEDs, respectively. Slight difference in current density might occur from thickness variation of PEDOT:PSS layers. In conclusion, the difference in the device operation between the standard and modified PEDOT:PSS was not significant. Lifetime measurements are currently on going.

Light-emitting layer, PFBT

The printability of yellow light-emitting material, PFBT, was examined on the top of printed PEDOT:PSS. 70 nm thick, smooth and pinhole-free film was achieved using the mixture of 2.5 wt-% of PFBT in *p*-xylene without additives. Any waviness or interfacial mixing was not observed after printing and drying. Fig 3(a) and (b) present WLI 3D surface profile from printed PFBT film on the structure of PEDOT:PSS/ITO-PET and an image of the film, respectively.

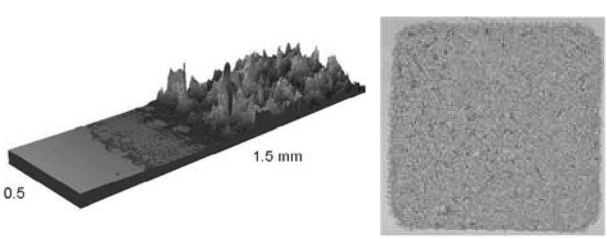


(a) (b)

Fig. 3. (a) 3D surface profile, and (b) 3x2 cm^2 size image of the printed PFBT on PEDOT:PSS.

Cathode, Al-ink

Usually in OLED structure the cathode layer consists of vacuum evaporated low and high work function metal such as Ca and Ag, respectively. Our target is to process cathode from printable metal ink.[5] So far, Al metal has selected for the ink preparation because of lower work function (4.2 eV) than Ag (4.6 eV), which is commonly-used metal in conductive pastes for other electronic applications. After preparation of printable Al-ink, screen printing method was used in order to achieve thick layer on top of OLED structure. Mesh count of 325 l/cm resulted the film thickness of 10 µm for Al cathode pixel as shown in Fig 4(a) WLI 3D surface profile and Fig 4(b) an image of Al pixel. Further optimization of screen printing process will significantly smooth Al layer.[5]



(a) (b)

Fig. 4. (a) 3D surface profile from the edge of screen printed Al cathode, and (b) image of Al cathode pixel (size of 3x3 mm^2).

Device characteristics

The substrate with four OLED pixels was characterized in inert atmosphere. The devices operated at the voltage of 5 V, and the average brightness of 79 cd/m^2 was achieved at the voltage of 16 V, as shown in Fig 5. Current density versus voltage characteristics illustrated no leakage current at low operating voltage indicating good compatibility of printed organic and metal structures. Device performance can be significantly enhanced by adding small amount of low work function metal in Al ink. The work of tuneable work function metal ink is currently under investigation.

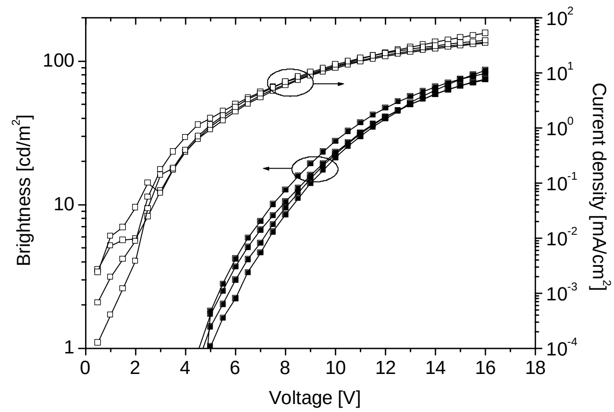
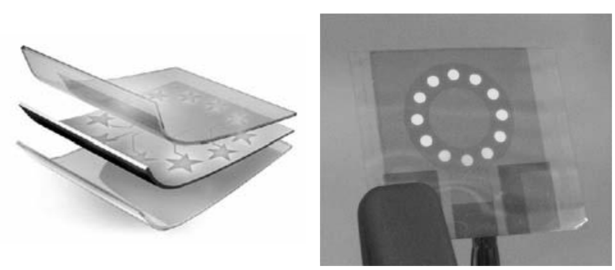


Fig. 5. Brightness and current density versus voltage characteristics of flexible all-printed OLEDs.

Flexible all-printed OLED demonstrator

The aim was to demonstrate flexible all-printed OLED in the ROLLED project's business card model. In the design, the OLED pixels operated as blinking EU stars as illustrated in Fig 6(a). The OLED demonstrator was printed on flexible plastic substrate with gas barriers as described above. Fig 6(b) presents the photograph of operating flexible all-printed OLED pixels before integration to business card. At the voltage of 20 V, clear and uniform yellow emission from the device is achieved.



(a) (b)

Fig. 6. Images of (a) ROLLED EU stars, and (b) operating flexible all-printed OLED pixels.

4. Summary

In summary, the fabrication of flexible all-printed

OLEDs, in which entire OLED stack, including PEDOT:PSS, yellow light-emitting PFBT polymer, and Al cathode were printed on flexible plastic substrate was introduced in this work. We have shown that gravure printing technique can be used for processing smooth and pinhole-free organic thin film layers.

Commercial production of OLEDs was demonstrated with batch to batch printing process, which can be transferred to roll-to-roll manufacturing using same process parameters as above in text. In the future, we will further demonstrate OLEDs, which are processed using roll-to-roll pilot printing machinery.

5. Acknowledgement

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6. References

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