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Multilayer slot-die coating of large-area organic light-emitting diodes

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A B S T R A C T

We investigate the slot-die coating process for the fabrication of large-area OLED lighting panels. Of many OLED layers, aqueous polymer-based hole injection layer (HIL) and small molecule-based hole transport layer (HTL) are formed using large-area slot-die coating. We are faced with three technical issues related with slot-die coating such as the flow down of an aqueous polymer solution near the inner perimeter of an insulator bank, pinhole-like surface in solution-processed small-molecule films, and the dissolution between two stacked layers with different solvents. We have suppressed those phenomena to a great extent and demonstrated that OLEDs with slot-die coated multiple layers show almost the same device performance as a reference OLED device with spin-coated HIL and vacuum-evaporated HTL. The peak-to-peak roughness of the slot-die coated bilayer (HIL/HTL) films is observed to be less than 12.5 nm. The OLED device with the slot-die coated bilayer film exhibits the power efficiency of 27.2 lm/W at 1000 cd/m^2, which is even higher than that (25.5 lm/W) of the reference device.

1. Introduction

As a flat-panel light source, organic light-emitting devices (OLEDs) draw much attention due to their superior features such as large-area surface emission, flexibility, transparency, etc. [1–4]. In spite of many advantages of OLED lightings, however, a successful market entry has not yet been made. One of the reasons lies in high fabrication costs. Currently, most of commercial OLED lighting panels have been fabricated using vapor deposition under vacuum. It facilitates the fabrication of OLEDs having functional multiple layers for enhanced device performance. However, the thermal evaporation process entails high fabrication costs (i.e., high material loss) and initial investment in equipment. To reduce it and thus improve the cost competitiveness of OLED lighting panels, it is desired to fabricate them using solution process. It includes various printing technologies (e.g., inkjet printing, nozzle printing, and gravure offset printing) and coating methods (e.g., spin coating, blade coating, slot-die coating, and spray coating) [5–7]. Those printing methods are suitable for the fabrication of OLED displays rather than large-area surface-emitting lighting panels. A spin coating method provides the most uniform films, yet its scalability is limited. A blade coating method has been used to fabricate multilayer OLEDs and solar cells under top and bottom drying condition [8–10]. This technique is suitable for processing of wide areas without any pattern. Spray coating has a high roll-to-roll compatibility, but requires a mask and thus shows low edge resolution. In [11], various large-area coating methods (knife coating, slot-die coating, and spray coating) were compared in terms of wet film thickness, homogeneity, and process stability and their dependence on process parameters. Slot-die coating is the promising fabrication scheme for large-area OLED lighting panels because it provides large and uniform films and the simultaneous coating of multiple layers of different solutions [12,13]. It is capable of coating a wide range of process materials (low and high viscosity fluids) and depositing a wide range of thickness (20 nm–150 μm). It involves many process variables such as coating speed, flow rate, coating gap, surface tension, viscosity, plate and drying temperatures, etc. This technique was used to fabricate polymer solar cells [14] and OLEDs [15]. Small-molecule OLED layers were also fabricated using a slot-die coating process and gap-to-film thickness ratio of up to 50 was feasible [16]. It was also demonstrated that OLED devices could be fabricated using slot-die roll coating under ambient air [15]. However, not much information on multilayer slot-die coating, especially for large-area OLED lighting panels, is available in the open literature.

In this paper, we investigate three technical issues related with large-area slot-die coating for OLED lightings. One issue is the flow down problem of an aqueous polymer solution, which occurs near the inner perimeter of an insulator bank. Another issue is that other than polymers, no chain entanglement occurs in dilute solution for small molecules. Since only the packing density is changed,

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Table 1
Fixed values of coating process variables.

Variables	Value
Die shim thickness	0.05 mm
Coating gap	200 μm
Flow rate	0.2 ml/min
Plate temperature	82 °C
Ambient temperature	22 °C

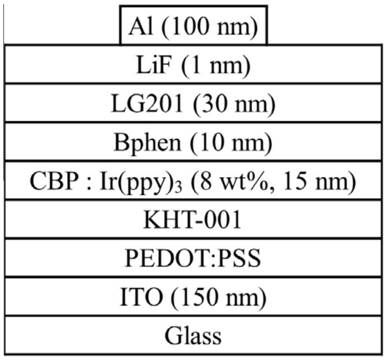
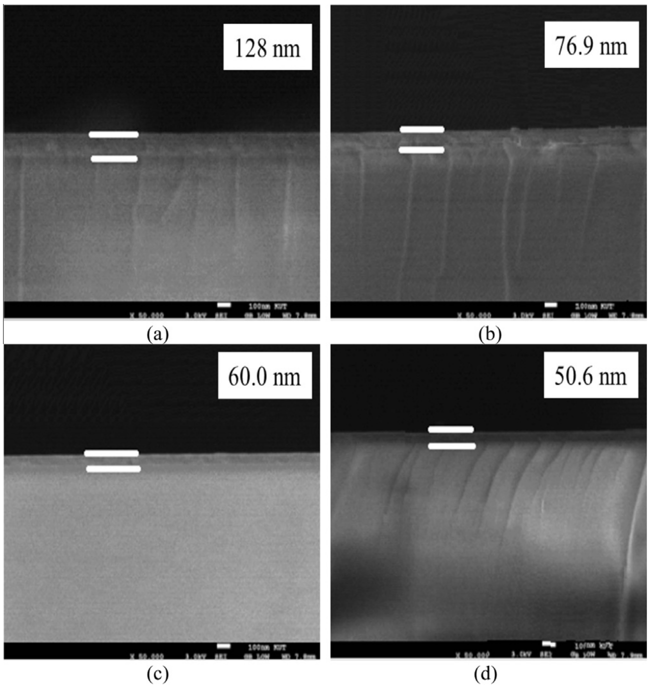


Fig. 1. Layer structure of green phosphorescent OLED device.

pinhole-like surface appears in solution-processed small-molecule films [17]. The other issue is the dissolution problem that occurs between two stacked layers with different solvents during slot-die coating. We have suppressed those phenomena to a great extent by changing the process parameters and solvents and demonstrated that OLEDs with slot-die coated multiple layers show almost the same device performance as reference OLED devices.

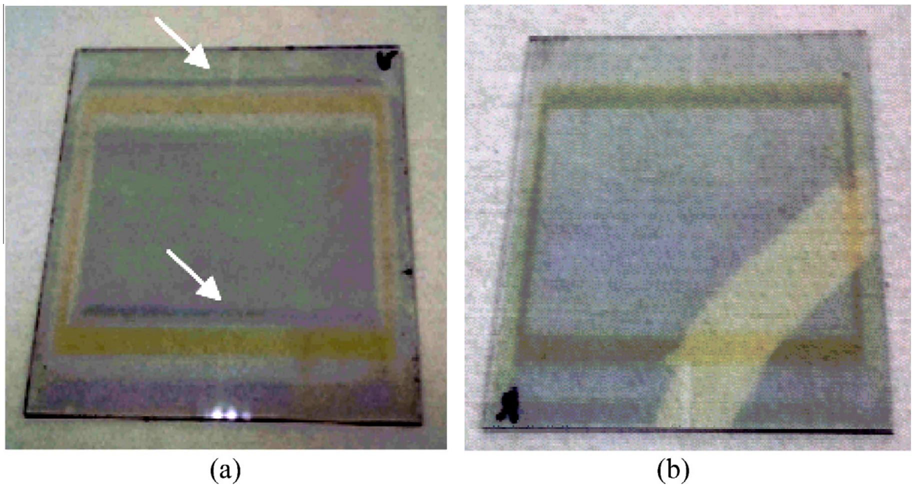
2. Experiment



(a) (b) (c) (d)

Fig. 3. Measured SEM images showing the thickness of PEDOT:PSS films coated at the velocity of (a) 1 mm/s, (b) 3 mm/s, (c) 5 mm/s, and (d) 7 mm/s.

Slot-die coating was done with a commercial table slot coater (TSDC-KTEU, DCN) under ambient air. It consists of a moving plate (210 mm × 300 mm) with plate heater (up to 150 °C), dry unit (NIR lamp up to 400 °C), syringe pump system (flow rate 0.1–10 ml/min), and slot head module (head size: 150 mm × 30 mm × 56.5 mm, surface flatness: ±3 μm, coating gap: 3–2000 μm with resolution 1 μm). Since slot-die coating involves many process variables, it is indispensable to fix some process variables to carry out experiments efficiently. Summarized in Table 1 are typical values of coating process variables. Unless otherwise specified, those process variables are kept unchanged. In this study, we have varied the coating speed (1–9 mm/s) and investigated its effect on the film thickness, roughness, and thickness uniformity. The film property is further evaluated by fabricating and measuring a green phosphorescent OLED device (Fig. 1) that consists of a 150-nm-thick ITO pre-coated on a glass substrate (purchased from Geomatech Co., Ltd), poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonate) (PEDOT:PSS) for a hole injection layer (HIL), KHT-001 (Duksan Neolux Co., Ltd.) for a hole transport layer (HTL), 15-nm-thick 4,4′-bis(*N*-carbazolyl)-1,1′-biphenyl (CBP) for an emission layer (EML), 10-nm-thick 4,7-diphenyl-1,10-phenanthroline (Bphen) for a hole/exciton blocking layer (HBL), 30-nm-thick LG201(LG



(a) (b)

Fig. 2. Image of PEDOT:PSS films coated on ITO glass (a) without and (b) with a fluorosurfactant.