

Effects of hole injection layer thickness on the luminescent properties of white organic light-emitting diodes

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Abstract

This work investigates how the thickness of the hole injection layer (HIL) influences the luminescent characteristics of white organic light-emitting diodes (WOLED). Experimental results indicate that inserting a thin HIL ($<200 \text{ \AA}$) into a WOLED without an HIL reduces the brightness and clearly changes the chromaticity because the surface of the 4,4',4''-tris{*N*-(3-methylphenyl)-*N*-phenylamino}-triphenylamine (m-MTDATA) film is extremely rough. In contrast, a dense film structure and the fine surface morphology of m-MTDATA of moderate thickness (350–650 \AA) provides a uniform conducting path on which holes cross the indium tin oxide (ITO)/HIL interface, improving luminescent performance, associated with the relatively stable purity of the color of the emission, with Commission Internationale l'Eclairage (CIE) coordinates of ($x = 0.40$, $y = 0.40$). However, inserting a thick HIL ($>650 \text{ \AA}$) reduces the luminescent performance and causes red-shift, because the holes and electrons in the effective emissive confinement region become less optimally balanced. Moreover, optimizing the device structure enables a bright WOLED with CIE coordinates of ($x = 0.34$, $y = 0.33$) to reach a luminance of 7685 cd/m^2 at a current density of 100 mA/cm^2 , with a maximum luminous efficiency of 1.72 lm/W at 5.5 V .

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1. Introduction

White organic light-emitting diodes (WOLEDs) are useful in full-color displays when combined with color filters, as backlights behind liquid crystal displays (LCDs) and solid-state lighting [1–3]. White emission is obtained by mixing two complementary colors such as blue and orange or three primary colors (red, green and blue) from differently emitting molecules. Various WOLED device structures with a variety of added fluorescent dopants in their emission layers have extensively reported to emit white light [4–6]. Of these, regardless of the device structure, a WOLED should be bright, efficient and provide high color purity. The reports on efficiently the luminescent performance of WOLEDs have mostly focused on devices with various configurations and

fluorescent dyes [7,8]. A hole injection layer (HIL) of suitable thickness inserted between the hole transport layer (HTL) and the transparent anode (typically an indium tin oxide (ITO)) typically reduces the operating voltage and improves the luminescent characteristics of the device [9–11]. Unfortunately, to the authors' knowledge, no clear mention has been made of the effects of the HIL thickness on the luminescent performance of WOLEDs.

2. Experimental

This work contributes to the understanding of the origins of the effect of HIL thickness on the luminescent performance of devices and chromaticity deviation in WOLEDs. The WOLED structure (Fig. 1) includes a 4,4',4''-tris{*N*-(3-methylphenyl)-*N*-phenylamino}-triphenylamine (m-MTDATA) HIL with various thicknesses of between 0 and 1250 \AA , an *N,N'*-diphenyl-*N,N'*-bis(1-naphthyl-phenyl)-(1,1'-biphenyl)-4,4'-diamine (NPB) HTL (600 \AA), a 0.4% 4-dicyanomethylene-2-

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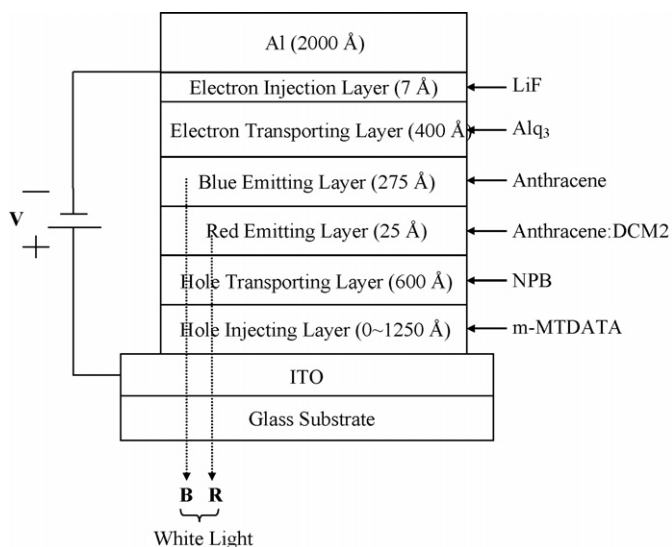


Fig. 1. Schematic configuration of WOLED and selected materials.

methyl-6-[2-(2,3,6,7-tetra-hydro-1H, 5H-benzo[*ij*]-quinolizin-8-yl)vinyl]-4H-pyran (DCM2)-doped Anthracene red-emitting layer (25 Å), undoped Anthracene, which acts as the blue-emitting layer (275 Å), a tris-(8-hydroxyquinoline) aluminum (Alq_3) electron-transporting layer (400 Å), an electron injection layer (EIL) of LiF (7 Å) and an Al top layer (2000 Å). The active emission area of these devices, defined by the overlap between the ITO and Al electrodes, was $0.4 \text{ cm} \times 0.6 \text{ cm}$. All of the organic layers were evaporated at a pressure of under 5×10^{-6} Torr, and the evaporation rate was 0.2–0.3 nm/s. All devices were encapsulated in a dry nitrogen glove box.

The EL spectra and Commission Internationale l'Eclairage (CIE) coordinates of the WOLEDs were measured using a TOPCON SR-1 spectroscan spectrometer. The luminance–current density (L – J) characteristics were recorded at the same time as the EL spectra, by combining the spectrometer with a Keithley 2400 programmable voltage–current source. Atomic force microscopy (AFM) measurements [12] were made to examine the surface roughness.

3. Results and discussion

Fig. 2 shows the EL emission intensity of WOLEDs with various m-MTDATA thicknesses between 0 and 1250 Å. The EL spectra cover a wide range of wavelengths, from 400 to 700 nm. Two prominent emission peaks at 450 (Anthracene) and 575 nm (DCM2) were consistently observed. Fig. 2 clearly indicates that, adding an HIL had been added to the WOLED, degraded the balance of the luminescent white emission. The luminance of WOLEDs with various m-MTDATA thicknesses was measured, and presented in Table 1. Table 1 indicates that the decrease in brightness is accompanied with an obvious red-shift in the chromaticity when a thin HIL (<200 Å) is inserted, but that the brightness is enhanced, with the stable emission of a pure color at CIE coordinates of ($x = 0.40$, $y = 0.40$) when a moderately thick HIL (350–650 Å) is inserted. The brightness examine is degraded as the thickness of the inserted HIL increases further.

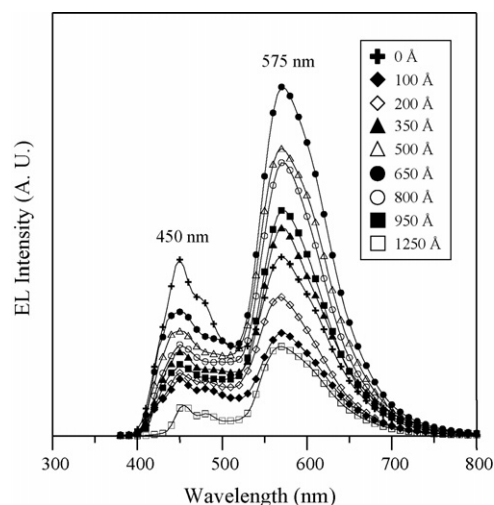


Fig. 2. Variation in EL spectra of WOLEDs with thickness of m-MTDATA between 0 and 1250 Å.

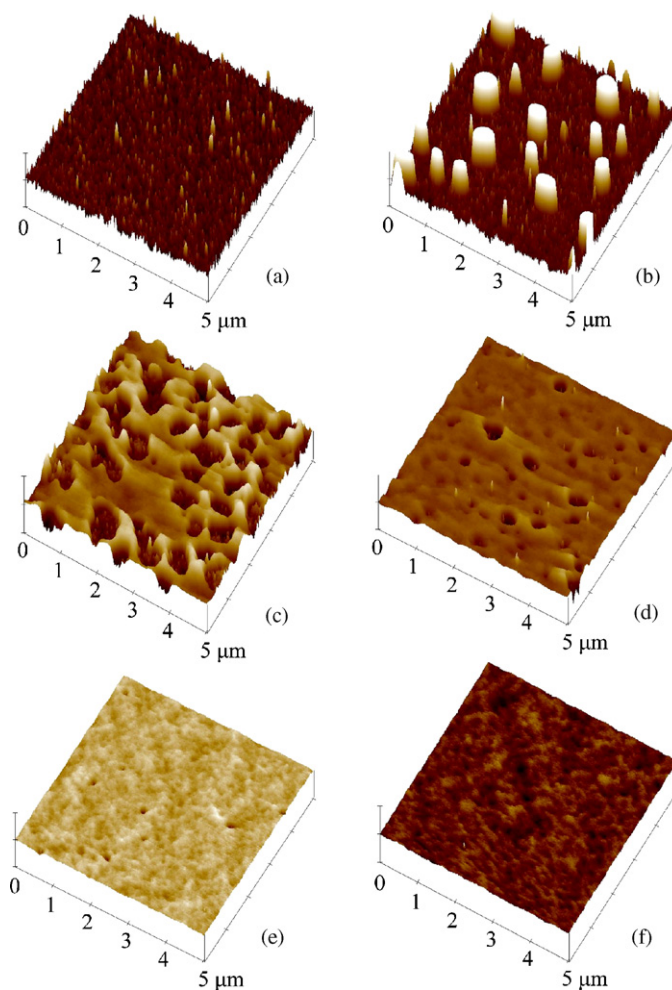


Fig. 3. AFM images of (a) ITO glass and m-MTDATA of various thicknesses – (b) 100 Å, (c) 200 Å, (d) 350 Å, (e) 650 Å, and (f) 950 Å – deposited on top of ITO glass.

Table 1
 Brightness, CIE coordinates of chromaticity, and measured average area roughness, R_a , of WOLEDs with m-MTDATA of various thicknesses between 0 and 1250 Å

Thickness of m-MTDATA (Å)	Brightness (cd/m ²) (at 100 mA/cm ²)	CIE coordinates (x, y) (at 100 mA/cm ²)	Average area roughness, R_a (Å)
0	5876	(x = 0.33, y = 0.35)	22.19
100	4734	(x = 0.36, y = 0.38)	78.65
200	5315	(x = 0.38, y = 0.40)	45.84
350	6227	(x = 0.40, y = 0.40)	12.13
500	7574	(x = 0.40, y = 0.40)	5.32
650	9014	(x = 0.40, y = 0.40)	5.18
800	7217	(x = 0.41, y = 0.40)	5.11
950	6478	(x = 0.43, y = 0.41)	5.05
1250	4456	(x = 0.45, y = 0.42)	5.01

AFM measurements were made to examine the surface roughness of HIL and thus elucidate the causes of the various effects of HIL on device luminescent performance and chromaticity deviation in WOLEDs. Fig. 3 displays the variation in the AFM images of m-MTDATA with various thicknesses. Table 1 presents the average area roughness (R_a) values measured by AFM at various different m-MTDATA thicknesses. The evidence shows a thin m-MTDATA (<200 Å) has a much rougher surface morphology than ITO glass. At greater thicknesses (>350 Å), the film structure becomes dense and the surface morphology becomes fine, indicating that the insertion of the thin m-MTDATA (<200 Å), with an extremely rough film surface, between the ITO anode and NPB, significantly reduces the hole injection efficiency and moves the electron-hole-recombination emission zones toward the anode side, causing undesirable luminance degradation, which is associated with a red-shift in the EL spectrum from the WOLED (Table 1). Conversely, the observed extraordinary flat film surface of the m-MTDATA with moderate thickness (350–650 Å) on top of the ITO glass may provide a uniform conducting path along which the holes cross the ITO/HIL interface. Such a mechanism may dominate, causing more holes to accumulate at the HTL/red-emitting layer interface, improving luminescent performance, which is associated with relatively stable emission of a pure color with CIE coordinates of (x = 0.40, y = 0.40). However, inserting a thick HIL (>650 Å) degrades luminescent performance and causes red-shift phenomenon, even though the surface morphology of m-MTDATA remained fine. This result is attributable to the degradation of the balancing of holes and electrons in the effective emissive confinement region.

The structure ITO/m-MTDATA (650 Å)/NPB (600 Å)/Anthracene:0.4% DCM2 (20 Å)/Anthracene (280 Å)/Alq₃ (400 Å)/LiF/Al improved the luminescent performance of the WOLED, which performance was associated with high color purity. A bright WOLED with CIE coordinates of (x = 0.34, y = 0.33) reaches a luminescence of 7685 cd/m² at a current density of 100 mA/cm² and a maximum luminous efficiency of 1.72 lm/W at 5.5 V.

4. Conclusions

In summary, the effects of the HIL thickness on the luminescent performance of WOLEDs were investigated.

Inserting a thin HIL into a WOLED without HIL reduced the brightness, and clearly changed the chromaticity (<200 Å). This change was associated primarily with the extremely rough film surface of the m-MTDATA. In contrast, the dense film structure and the fine surface morphology of a moderately thick m-MTDATA (350–650 Å) yield a uniform conducting path through which holes cross the ITO/HIL interface, improving superior luminescent performance, associated with the relatively stable emission of pure color with CIE coordinates of (x = 0.40, y = 0.40). However, inserting a thick HIL (>650 Å) degrades luminescent performance and causes red-shift, because the balancing of the holes and the electrons in the effective emissive confinement region is degraded. Furthermore, optimizing the device structure improves the luminescent performance of the WOLED, which improvement is associated with high color purity.

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