



Unlocking the potential of *p*-doped hole transport layers in inverted organic light emitting diodes

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ABSTRACT

The MoO₃ doped N,N'-bis-(1-naphthyl)-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB:MoO₃ in 2:1 mass ratio) and 4,4'-N,N'-dicarbazole-biphenyl (CBP:MoO₃ in 2:1 mass ratio) as *p*-doped hole transport layers have been used in inverted organic light emitting diodes (IOLEDs). Compared to the NPB/20 nm NPB:MoO₃ structure, the NPB/10 nm CBP:MoO₃/10 nm NPB:MoO₃ structure showed increased device performance, mostly because the hole transport barrier from CBP:MoO₃ to NPB was smaller than that from NPB:MoO₃ to NPB; it also presented improved device performance than the NPB/20 nm CBP:MoO₃ structure, ascribed to the higher conductivity of NPB:MoO₃ than that of CBP:MoO₃. We provide a manageable way to unlock the merits of *p*-doped hole transport layers for markedly increasing the performance of IOLEDs.

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1. Introduction

Inverted organic light emitting diodes (IOLEDs) have attracted much attention, due to their compatibility with *n*-channel amorphous silicon field-effect transistor, favoring the low-cost production of active-matrix flat-panel display [1–10]. However, in order to compete with the conventional liquid crystal display, the active-matrix OLED display needs to be more efficient, stable, and cheaper. Thus, it is of an urgent need for OLEDs to reduce the power loss as much as possible, which has been considered as a fundamental issue for LEDs.

The improvement of hole injection can contribute to the reduction in the power loss of OLEDs. During the past decade, *p*-doped hole transport layers (HTLs) have been widely used to enhance hole injection of OLEDs [11–13], since (1) the *p*-doped HTL can offer ohmic contacts with metal even with medium work function, e.g., Al, as a result of the thin depletion zone formed between *p*-doped HTL and metal, and (2) the *p*-doped HTL can lead to the significant decrease in ohmic loss during hole conduction relative to the undoped HTL. The MoO₃ doped N,N'-bis-(1-naphthyl)-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB:MoO₃) and 4,4'-N,N'-dicarbazole-biphenyl (CBP:MoO₃) are among the most frequently employed *p*-doped HTLs. As shown in Fig. 1, the highest occupied molecular orbital (HOMO) levels for NPB:MoO₃ and CBP:MoO₃

are –5.35 and –6.25 eV; the work functions for NPB:MoO₃ and CBP:MoO₃ are 4.9 and 5.6 eV [14,15], respectively.

In conventional *p-i-n* OLEDs, a single *p*-doped HTL has been used. Despite achieving the remarkable device performance, the usage of a single *p*-doped HTL is currently stuck in a dilemma that the hole conduction advantage of *p*-doped HTL is unavoidably counteracted by the hole transport barrier (ϕ_B) across the interface with the undoped HTL [16–18]. As of now, how to unlock the potential of *p*-doped HTLs remains still challenging in IOLEDs. In this paper, for the first time, the combination of two *p*-doped HTLs has been applied to improve the performance of IOLEDs, and the underlying mechanism was discussed.

2. Experimental

100 nm-thick ITO thin film coated glass substrates were commercially bought with a sheet resistance of 10 Ω per square. After being carefully cleaned in acetone, alcohol, and de-ionized water sequentially by an ultrasonic horn, the pre-patterned ITO substrates were blown dry by a nitrogen gun. The NPB, CBP, bathocuproine (BCP), and tris(8-quinolinolato) aluminum (Alq3) were obtained from Jilin OLED material Company; the MoO₃ and Li₂CO₃ were purchased from Sigma-Aldrich Company, and applied as *p*- and *n*-typed dopants, respectively. The ratios of dopant to host were recorded in mass. All the materials were used as received.

The base pressure of the device fabrication was 4×10^{-4} Pa. The five IOLEDs were fabricated as follows:

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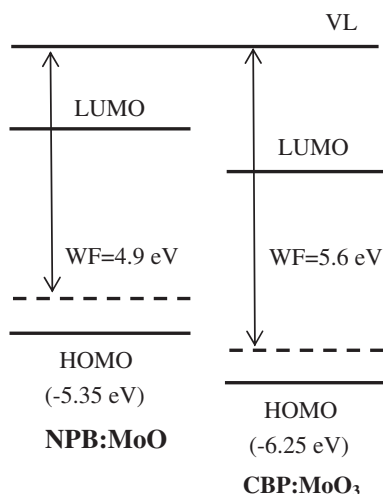


Fig. 1. The energetic parameters for NPB:MoO₃ and CBP:MoO₃. The WF, LUMO, and VL stand for work function, lowest unoccupied molecular orbital, and vacuum level, respectively. The horizontal dashed lines represent the Fermi levels.

- IOLED 1: ITO/4:1 BCP:Li₂CO₃ 10 nm/Alq₃ 40 nm/NPB 60 nm/2:1 NPB:MoO₃ 20 nm/Al;
 IOLED 2: ITO/4:1 BCP:Li₂CO₃ 10 nm/Alq₃ 40 nm/NPB 60 nm/2:1 CBP:MoO₃ 20 nm/Al;
 IOLED 3: ITO/4:1 BCP:Li₂CO₃ 10 nm/Alq₃ 40 nm/NPB 60 nm/2:1 CBP:MoO₃ 10 nm/2:1 NPB:MoO₃ 10 nm/Al;
 IOLED 4: ITO/4:1 BCP:Li₂CO₃ 10 nm/Alq₃ 40 nm/NPB 60 nm/2:1 CBP:MoO₃ 5 nm/2:1 NPB:MoO₃ 15 nm/Al;
 IOLED 5: ITO/4:1 BCP:Li₂CO₃ 10 nm/Alq₃ 40 nm/NPB 60 nm/2:1 CBP:MoO₃ 15 nm/2:1 NPB:MoO₃ 5 nm/Al.

Note that, The 2:1 doping ratio of organic material to MoO₃ was an optimized value [19,20]. It should be stressed that some part of MoO₃ degraded into lower valance oxides in the vacuum deposition, but for simplifying the device expression, MoO₃ was used as the nominal notation.

The current versus voltage (*I*-*V*) characteristics of the OLEDs were measured by the programmable Keithley 2400 sourcemeter, and the luminance was recorded by an ST-86LA Spot Photometer under the ambient condition.

3. Results and discussion

3.1. The performance comparisons of the IOLEDs 1–3

Fig. 2(a) shows the *I*-*V* characteristics for these three devices. At a given driving voltage ≥ 4 V, there was a decrease of current density in the order of IOLED 3 > IOLED 2 > IOLED 1. At a driving voltage of 9 V, the current densities of the IOLEDs 1–3 were 47.5, 140.0, and 384.3 mA/cm², respectively. It demonstrates that the combination of 10 nm 2:1 CBP:MoO₃ and 10 nm 2:1 NPB:MoO₃ outperformed the individual 20 nm 2:1 NPB:MoO₃ and CBP:MoO₃ in terms of generating hole current in to the NPB. As shown in Fig. 2(b), at a given driving voltage ≥ 4 V there was a decrease of luminance in the order of IOLED 3 > IOLED 2 > IOLED 1. At a driving voltage of 9 V, the luminance of the IOLEDs 1–3 was 778, 2493, and 6798 cd/m². Fig. 2(c) shows that at a given current density, the current efficiency of the IOLED 1 was slightly lower than that of the IOLED 2; The IOLED 3 gave higher current efficiency than the IOLEDs 1 and 2. This indicates that the hole–electron balance in the IOLED 3 was improved compared to those in the IOLEDs 1 and 2. The maximum current efficiencies for the IOLEDs 1–3 were 1.7, 1.8, and 2.0 cd/A, respectively. Hence, it is conclusive that the

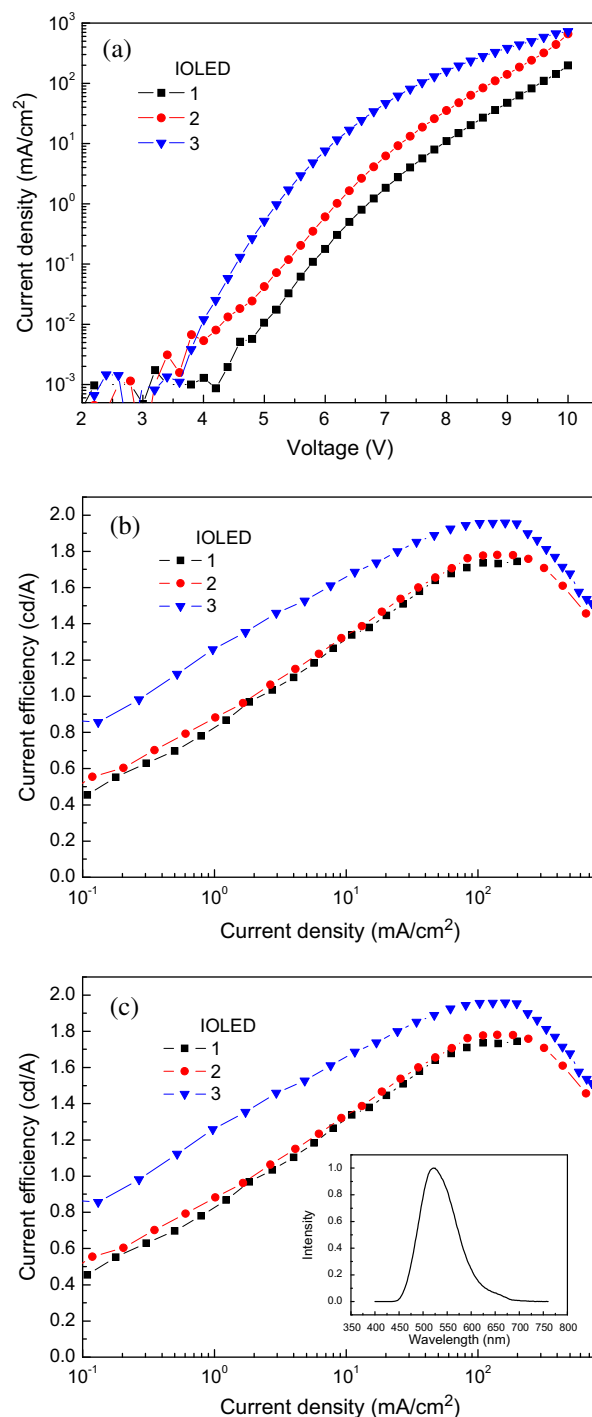


Fig. 2. The *I*-*V* (a), luminance versus voltage (b), and current efficiency versus current density (c) characteristics of IOLEDs 1–3. Note that, all of IOLEDs 1–5 gave nearly same electroluminescent spectra, as shown in the inset of figure (c).

combinatorial CBP:MoO₃ and NPB:MoO₃ provided increased IOLED performance than the single used NPB:MoO₃ and CBP:MoO₃.

3.2. Understanding the hole current comparisons of the IOLEDs 1–3

The hole current in the IOLEDs 1–3 is up to not only the conductivity of *p*-doped HTL, but also the ϕ_B across the interface with undoped NPB. It is worth mentioning that the ϕ_B often deviates from the Schottky–Mott limit [21], due to the formation of the interfacial dipole (Δ) expressed as

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