



# Chromatic-stability white organic light emitting diodes based on phosphorescence doped electron transport layer



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## ABSTRACT

The influence of *fac*-tris(2-phenylpyridine)iridium [Ir(ppy)<sub>3</sub>] doped into electron transport layer was investigated by a series of electron-only devices, and the mechanism of the reduced field-dependent electron-transporting properties was also discussed. Utilizing the method of optimized doping concentration, a hybrid white organic light emitting diode with high efficiency, low efficiency roll-off and stable spectra was fabricated. Organic layer doped Ir(ppy)<sub>3</sub> serves two functions: emit supernumerary green light with excess holes when the applied bias are low; weaken electron-transport ability when the bias increase. Both the two functions can improve the carrier balance and confine the exciton recombination region. For Device B, the maximum current efficiency and power efficiency reach 37.4 cd/A and 28.6 lm/W, respectively. The Commission Internationale de l'Éclairage (CIE) coordinates vary slightly from (0.48, 0.43) at 1.1 cd/m<sup>2</sup> to (0.46, 0.43) at 18,600 cd/m<sup>2</sup>.

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## 1. Introduction

White organic light emitting diodes (WOLEDs) have been gained considerable concern in the last decade, driven by the tempt of their potential commercial application in flat panel displays and large-area solid-state lighting source [1–4]. They have many advantages over other displays or light sources such as flexibility, design freedom, thin, distributed light emission, and low cost in manufacturing [5–9].

The quality of the light generated by WOLEDs depends on the color temperature and the Color Rendition Index (CRI). In addition, making sure that these quantities remain stable, as the brightness of the WOLED is changed, is of great importance [5,10]. In fact, Most WOLEDs reported in literatures show brightness-dependent color-shifts that significantly exceed the tolerable margin. This margin is typically defined as the CIE (Commission Internationale de l'Éclairage) coordinate change of 0.005–0.01 or less for both the *x*- and the *y*-value over orders of magnitude in brightness and over the entire operational lifetime [11]. Although different explanations for the notorious color-shift observed in most devices have been proposed there is still no consistent quantitative explanation [12]. The color-shift observed in WOLEDs based on a single

emissive layer of a white-emitting copolymer originates from the competition between electron trapping on red-emitting sites and unperturbed charge transport through the organic layer [11]. WOLEDs were based on multilayer, however, this situation is more complicated. Because people have to consider the field-dependence of holes and electrons transfer rates across each interface, which may be caused by a shift of the recombination/emission zone with increasing brightness.

In order to improve the power efficiency and concomitantly lower the operation voltage, the concept of electrical doping, which is adopted from OLEDs, is widely employed [13]. It has been shown that OLEDs with p-doped hole transport layer and/or n-doped electron transport layer can provide efficient carrier injection and transport [14–16], which can be used to regulate the position of recombination/emission zone. Recently, we reported the hole transport layer respectively doped with three different Ir-emitters that greatly promotes device efficiency due to improved hole transport ability [17].

In this paper, we focus on the development of color-stability WOLEDs with stacked multi-emissive layers structure. *fac*-tris(2-phenylpyridine)iridium (Ir(ppy)<sub>3</sub>) was doped into organic layer, thus electron-transport properties and possible mechanisms were investigated. Because we realized that the device design not only could improve the carriers balance in the emission region, but also play an important role in improving the color-stability and efficiency of the devices, a new WOLED was achieved. The WOLED

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utilizing the optimal doping concentration of 6% exhibits better power efficiency of 28.7 lm/W at 4.0 cd/m<sup>2</sup>, 28.6 lm/W at 10.0 cd/m<sup>2</sup>, 25.5 lm/W at 100 cd/m<sup>2</sup>, and 17.2 lm/W at 1000 cd/m<sup>2</sup>. It is worth mentioning that the low efficiency roll-off and little variation of CIE coordinates varies from (0.48, 0.43) to (0.46, 0.43) as the brightness increases from 1.0 cd/m<sup>2</sup> to 18,600 cd/m<sup>2</sup>.

## 2. Experimental

The devices were fabricated by conventional vacuum deposition of the organic layers and cathode onto an ITO (15 Ω/sheet, 150 nm) coated glass substrate under a base pressure lower than  $2 \times 10^{-4}$  Torr. The substrates were successively cleaned in detergent, de-ionized water, acetone, and isopropanol. Immediately prior to loading into a custom-made high vacuum thermal evaporation chamber, the substrates were exposed to a UV-ozone environment for 15 min. The typical deposition rates were 0.6 Å/s, 0.1 Å/s and 5.0 Å/s for organic materials, lithium fluoride (LiF), silver (Ag) and aluminum (Al), respectively.

In this paper, to investigate the electron-transporting ability of the varied Ir(ppy)<sub>3</sub> doping concentration, we fabricated electron-only devices with a structure of ITO/TPBi (20 nm)/CBP: *x* wt% Ir(ppy)<sub>3</sub> (20 nm)/TPBi (20 nm)/Al (100 nm) were fabricated, in which *x* was 0, 5, 10, and 15, respectively.

To further study the performance of white OLEDs with doped transport layer, we designed Device A with the structure of: ITO/2-TNATA (40 nm)/NPB (5 nm)/CBP:15 wt% bt<sub>2</sub>Ir(acac) (10 nm)/CBP (10 nm)/ADN:1 wt% BmPAC (30 nm)/CBP (5 nm)/CBP (5 nm)/Bphen (25 nm)/LiF (0.3 nm)/Al (200 nm). To elucidate the reason of high efficiency, reduced efficiency roll-off and stable spectrum of the proposed hybrid WOLED, we designed the Device B, which has the same thickness of CBP with the structure of: ITO/2-TNATA (40 nm)/NPB (5 nm)/CBP:15 wt% bt<sub>2</sub>Ir(acac) (10 nm)/CBP (10 nm)/ADN:1 wt% BmPAC (30 nm)/CBP (5 nm)/CBP:6 wt% Ir(ppy)<sub>3</sub> (5 nm)/Bphen (25 nm)/LiF (0.3 nm)/Al (200 nm). Device C is used to tune the CIE coordinate of the structure: ITO/2-TNATA (40 nm)/NPB (5 nm)/CBP:5 wt% bt<sub>2</sub>Ir(acac) (10 nm)/CBP (10 nm)/ADN:1 wt% BmPAC (30 nm)/CBP (5 nm)/CBP:6 wt% Ir(ppy)<sub>3</sub> (5 nm)/Bphen (25 nm)/LiF (0.3 nm)/Al (200 nm). Compare with Device B, which is just reduced the concentration of PYD01. Obviously, 4,4,4-[N-(2-naphthyl)-N-phenylamino]-triphenylamine (2T-NATA) was used as the hole-injection layer (HIL) onto the ITO, TCTA as the hole-transport layer (HTL) and 4,7-diphenyl-1,10-phenanthroline (Bphen) as the electron-transport layer (ETL). 4,4'-bis(N-carbazolyl)biphenyl (CBP) layer was employed as the space layer confining the exciton formation area effectively within the emitting layer (EML). The bis(2-phenylbenzothiazolato)(acetylacetonate)iridium bt<sub>2</sub>Ir(acac) doped in 15 wt% CBP, 6,12-bis([N-(3,4-dimethylphenyl)-N-(2,4,5-trimethylphenyl)] amino) chrysene (BmPAC) doped in 1 wt% 1,4-bis[N-(1-naphthyl)-N'-phenylamino]-4,4'-diamine/9,10-di(2-naphthyl)anthracene (ADN) and Ir(ppy)<sub>3</sub> doped in 6 wt% CBP were used as orange, blue and green emitting layer, respectively.

To prove two functions of the Ir doped layer, we also fabricated the hole-only device with the structure of ITO/2-TNATA (40 nm)/NPB (5 nm)/Ag(100 nm), the HTL-EML only (HE-only) device: ITO/2-TNATA (40 nm)/NPB (5 nm)/CBP:15 wt% bt<sub>2</sub>Ir(acac) (10 nm)/CBP (10 nm)/ADN:1 wt% BmPAC (30 nm)/CBP (5 nm)/CBP (5 nm)/Ag (100 nm), while it was used as HIL and HTL, EML respectively, in both Devices A and B. Equally, from the fabricated electron-only device (ITO/Bphen (30 nm)/LiF (0.3 nm)/Al (100 nm)), we used (Bphen (30 nm)/LiF (0.3 nm)/Al (100 nm)) in the Devices A and B as ETL, EIL and cathode, respectively.

The active area of the devices defined by the overlap between the electrodes was 4 mm<sup>2</sup> in all cases. The current

density–voltage–luminance (*J–V–L*) characteristics, electroluminescence (EL) spectra and CIE coordinates were measured and recorded by a testing setup comprising of a Keithley 2400 source-meter and a Minolta PR-6500 spectrometer. The measurements were carried out in the dark at room temperature and only light emitting from the front face of the OLED was collected.

## 3. Results and discussion

### 3.1. Electron transporting properties

An effective hole/electron injection is required in high efficiency OLEDs. To improve the transmission of electrons, we fabricated a series of electron-only devices with the following structure: ITO/TPBi (20 nm)/CBP: *x* wt% Ir(ppy)<sub>3</sub> (20 nm)/TPBi (20 nm)/Al(100 nm) while *x* was 0, 5, 10, and 15, respectively. The *J–V* characteristics and structure of these electron-only devices are shown in Fig. 1(a and b) respectively. The doping layer was made by co-evaporating the electron transporting materials (CBP) with Ir(ppy)<sub>3</sub>.

We observed that the electron current reduced rapidly when the concentrations of Ir(ppy)<sub>3</sub> increased. As the concentrations increased from 0% to 15%, the highest current density at 10 V reached 345, 138, 21, 0.3 mA/cm<sup>2</sup>, respectively. What interesting is that the reduced current densities also presented classical *J–V* properties of the diode.

This dramatic diminution may be attributed to the electron-accepting ability of 2-phen-ylpyridine (ppy) ligand of Ir(ppy)<sub>3</sub> by which part of electrons are trapped while transport in CBP, we conclude that Ir(ppy)<sub>3</sub> serves as the electron-transporting channel which prevent hole injection. Accepted electrons play a significant role on the transport transfer from dopants to host, because mobility of Ir(ppy)<sub>3</sub> is far lower than that of host. Though much higher concentration of Ir complexes trapped too much electron, transporting properties were poor. Therefore, less electrons were injected into CBP layer, resulting in lower electron current densities.

To further study the performance of the carrier transporting in OLEDs, we designed the hole-only, HE-only and electron-only devices both in Devices A and B. For the electron mobility measurement of Bphen, lithium fluoride (LiF) is used as a buffer layer between the Bphen layer and the Al electrode. Therefore, we conclude that the electron conduction is not limited by the injection [18]. Fig. 2 shows the current density–voltage characteristics of holes and electrons: as the driving voltage is low, electron mobility is about three orders of magnitude less than that of holes. However, electron mobility is just one magnitude less than that of holes as the driving voltage tends to 8 V. The reason for this phenomenon is that the charge mobility is electric-field dependent, as is shown in Eq. (1) [19].

$$\mu = \mu_0 \exp(\gamma\sqrt{E}) \quad (1)$$

Here,  $\mu_0$  is the zero-field mobility,  $E$  is the electric-field coefficient to the mobility and  $\gamma$  describing the field activation of the mobility. Absolutely,  $\gamma$  of electrons for Bphen [20] is much higher than that of holes for 2-TNATA or NPB [21]. In other words, electron mobility of Bphen grows much more than that of holes for 2-TNATA or NPB with the increscent applied bias.

On the other hand, for the HE-only device, the current density of holes is much lower than that of hole only device, which is caused by the relatively low hole mobility of CBP and traps of emitting materials. As the driving voltage is low, hole mobility is about 2 orders of magnitude higher than that of electrons. However, electron mobility is just at the same magnitude with that of holes as the driving voltage grows.

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