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RESEARCH ARTICLE

Properties of non-doped organic light-emitting devices based on an ultrathin iridium complex phosphor layer

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Abstract Organic light-emitting devices (OLEDs) were constructed with a structure of indium tin oxide (ITO)/*N*,*N*′-bis(naphthalen-1-yl)-*N*,*N*′-bis(phenyl)-benzidine (NPB) ($50-x$ nm)/bis[2-(4-tertbutylphenyl)benzothiazolato-N,$C^{2\prime}$] iridium (acetylacetonate) [$(t\text{-bt})_2Ir(acac)$] (d nm)/NPB (x nm)/2,2′,2′′(1,3,5-benzenetriyl)tris-(1-phenyl-1H-benzimidazole) (TPBI) (30 nm)/Mg:Ag (200 nm). A thin blue emission material of NPB was used as a separating layer, and the $(t\text{-bt})_2Ir(acac)$ yellow phosphorescent dye was acted as an ultrathin light-emitting layer. TPBI acted as both hole-blocking and electron-transporting layer. By changing the location (x) and the thickness (d) of the phosphor dye, the variation of device performance were investigated. The results showed that all the devices had a turn-on voltage of 2.8 V. In the case of $d=0.2$ nm and $x=5$ nm, the OLED had a maximum luminance of 18367 cd/m^2 and a maximum power efficiency of 5.3 lm/W. The high performance is attributed to both direct charge carrier trapping of iridium phosphor dye and the thin NPB separation layer, which effectively confines the recombination zone of charge carriers.

Keywords organic light-emitting devices (OLEDs), iridium complex phosphor dye, ultrathin layer, separation layer

1 Introduction

There has been a growing interest in organic light-emitting devices (OLEDs) due to their light weight, low cost, high brightness, flexible availability, and high power efficiency. Recently, a power efficiency as high as 90 lm/W at 1000 cd/m^2 was reported by Sebastian Reineke’s group, using efficient phosphorescent emitter layers with high-refractive-index substrate and a periodic out-coupling structure [1]. To date, more work for the light emitters of OLEDs are focusing on phosphorescent dyes, especially for transition metal complexes with heavy atom effect, such as Ir, Pt, Eu, and Os, which can greatly improve device efficiency, owing to both singlet and triplet excitons emission through efficient mixing of the singlet and triplet excited states. Therefore, the internal quantum efficiency can approach 100% [2]. However, most attempts to achieve high efficiency are primarily focused on iridium complexes, which contribute to comparably shorter triplet lifetime and higher emission efficiency [3]. Meanwhile, they are easy to fabricate, and both emissive wavelength and efficiency can be regulated by modifying functional groups on ligand. In addition, there is also high efficiency achieved due to the improvement of device structures [4–6]. Among them, the non-doped ultrathin structure, which can show some information about the dye’s nature property, such as charge trapping and charge transfer, is more effective than the traditional doping structure for energy transferring according to Förster and Dexter energy transfer theory [7–9]. It can also reduce the difficulty in adjusting the co-evaporation of organic materials and improve device stability and reproducibility. On the other hand, the imbalance of charge carrier injection will decrease the device luminous efficiency. So far, there are some measures to improve electrons and holes injection by introducing exciton blocking layer and interface modification [10,11], double emission layers by confining the exciton formation region [12,13], appropriate energy-level alignment between each layers [14], and so on.

In this work, we studied the performance of non-doped OLEDs by employing a novel yellow iridium complex of bis[2-(4-tertbutylphenyl)benzothiazolato-N,$C^{2\prime}$] iridium (acetylacetonate) [$(t\text{-bt})_2Ir(acac)$] as an ultrathin layer, *N*,*N*′-bis(naphthalen-1-yl)-*N*,*N*′-bis(phenyl)-benzidine (NPB) as hole transporting layer, and separator layer, 2,2′,2′′(1,3,5-benzenetriyl)tris-(1-phenyl-1H-benzimidazole) (TPBI) as hole-blocking layer and electron-transporting layer. The

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optoelectronic characteristics of the devices were systematically investigated by optimizing the thickness and the position of the phosphorescent layer.

2 Experiment

ITO-coated substrate was ultrasonically cleaned with detergent, deionized water, acetone, and ethanol step by step and then dried by nitrogen blow. It was treated by oxygen plasma for 5 min prior to the deposition of organic layers to increase the wok function. Organic and metallic layers were subsequently deposited at a rate of 0.1–0.3 nm/s and 1–2 nm/s, respectively, while keeping the pressure on the order of magnitude 10^{-4} and 10^{-3} Pa, respectively, without breaking the vacuum. Film thickness and deposition rate were monitored *in situ* by oscillating quartz thickness monitors. Two kinds of device structure were constructed as follows:

Group I: ITO/NPB ($50-x$ nm)/$(\text{t-tb})_2$Ir(acac) (0.2 nm)/NPB (x nm)/TPBI (30 nm)/Mg:Ag (200 nm), $x = 5, 6, 7, 8, 10$, corresponding to devices A1, B1, C1, D1, and E1.

Group II: ITO/NPB ($50-x$ nm)/$(\text{t-tb})_2$Ir(acac) (0.15 nm)/NPB (x nm)/TPBI (30 nm)/Mg:Ag (200 nm), $x = 5, 6, 7, 8, 10$, corresponding to devices A2, B2, C2, D2, and E2.

Figure 1 shows the schematic of energy level for the OLEDs. The electroluminescent (EL) spectra of the unencapsulated devices were measured with an OPT-2000 spectrometer, and the luminance-bias voltage-current density ($L-V-J$) characteristics were recorded with a Keithley 4200 source. All measurements were performed at room temperature under ambient conditions.

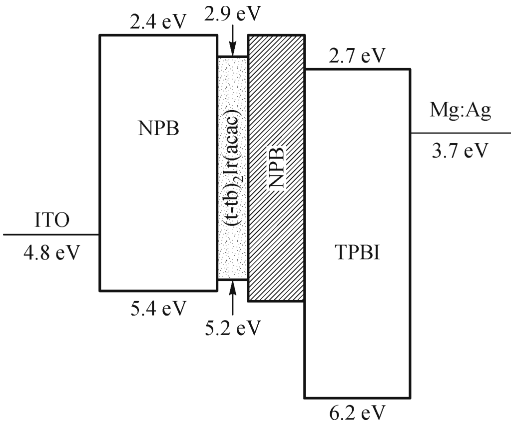


Fig. 1 Energy level structure of OLEDs

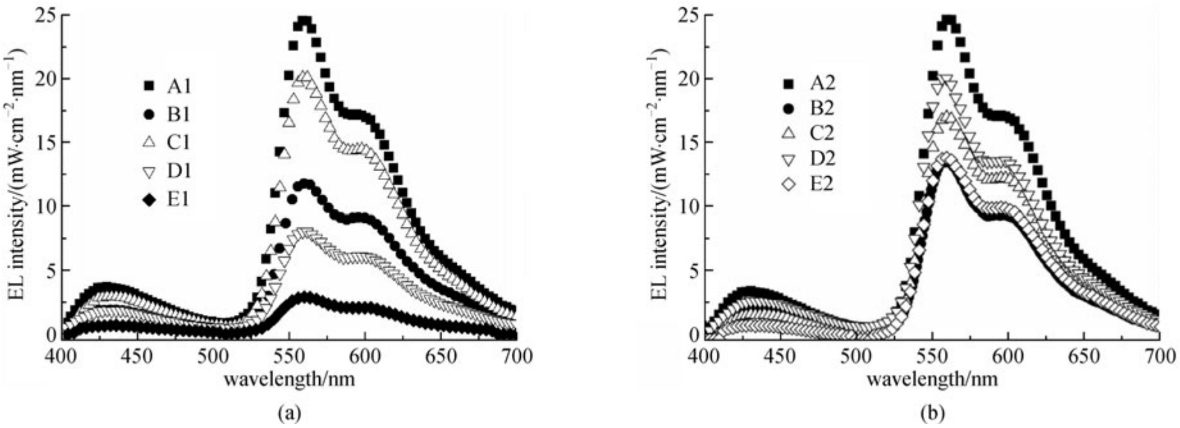
3 Results and discussion

Figure 2 shows the EL spectra of devices A1–E1 and A2–E2 at a bias voltage of 3 V. There is a peak at 432 nm caused by NPB blue light emission, which acts as both separator layer and emission layer. A peak at 560 nm with a shoulder at 598 nm due to $(\text{t-bt})_2$Ir(acac) yellow light emission are presented from these two group devices. The NPB emission is induced by energy transfer from excitons at NPB/TPBI interface to the thin NPB layer. We can also see that the yellow emission intensity is higher than that of blue emission, which is due to efficient direct charge carrier trapping by $(\text{t-bt})_2$Ir(acac) dye. Moreover, as NPB exciton has high triplet energy of 2.4 eV [15], and $(\text{t-bt})_2$Ir(acac) triplet energy is about 2.2 eV that can be deduced from its emission peak at 560 nm using the formula $E_T = hc/\lambda$ (where E_T is the triplet energy, h is Planck constant, c is light speed, and λ is the wavelength), so there will be an additional energy transfer channel from NPB triplets to the yellow phosphor. A maximum emission intensity of both devices A1 and A2 is realized when the film thickness of ultrathin NPB layer x equals to 5 nm. Compared to the light intensity of NPB, the intensity of $(\text{t-bt})_2$Ir(acac) light decreased with x increasing. The reasons are that thicker NPB separator layer with only hole transporting capability is not favorable for electrons transferring from TPBI to phosphor layer. Taking the electric field inside the device into consideration, the charge carrier tunneling rate is given as [16]

$$R_J \propto E\exp\left[-\frac{\pi}{h}(2mE_g)^{1/2}\left(\frac{E_g}{qE}\right)\right], \tag{1}$$

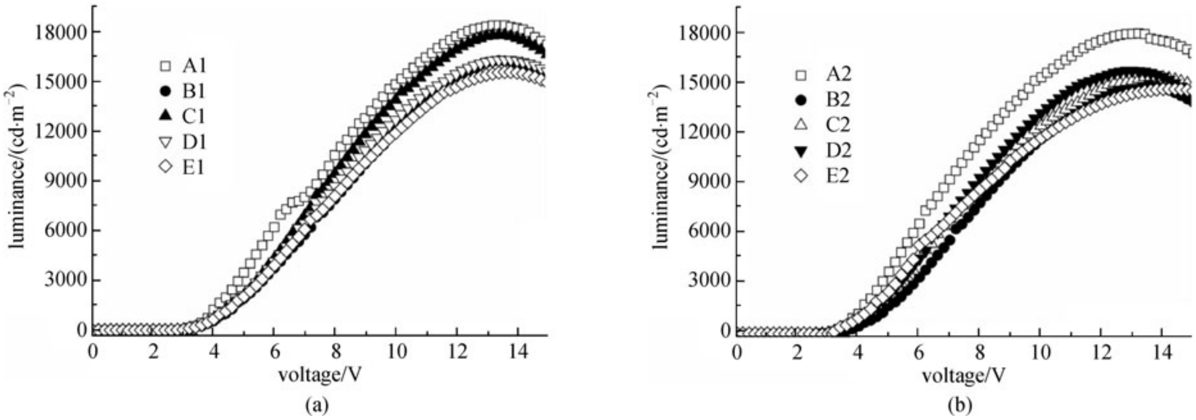
where $E = V/d$ is electric field intensity, E_g is energy gap, V is bias voltage, and d is the thickness of organic films. Therefore, it can be clearly derived that R_J is dependent on d. Although the total electric field is kept as a constant for the total film thickness is fixed, the internal eclectic field applied on each layer does change. This leads to the movement of charge carrier recombination region.

Figure 3 shows the luminance-bias voltage curves of devices A1–E1 and A2–E2. All the devices have almost the same turn-on voltage as low as 2.8 V. The luminance intensity of the OLEDs first increases while the voltage keeps on increasing, and then, it decreases gradually before breaking. In Figs. 3(a) and 3(b), a maximum luminance of 18367 cd/m^2 for device A1 is higher than that of 17014 cd/m^2 for device A2, when x is 5 nm. For devices A1 to E1, the brightness reduces when the thickness of NPB separator layer increases from 5 nm, while at the same time, the thickness of NPB hole transporting layer decreases, which is the same for devices A2 to E2. This can be explained from two aspects. First, there are more holes trapped on $(\text{t-bt})_2$Ir(acac) molecular, resulting in serious hole-triplet or triplet-triplet annihilation [17]. Second, the hole mobility of NPB is much larger than that of electron reported in most papers, and there is also 0.3 eV LUMO (the lowest unoccupied molecular orbital) energy barrier between NPB and TPBI; both are not facile for electron injection to the thin NPB separator layer and then to the $(\text{t-bt})_2$Ir(acac) layer. These two cases cause less effective electrons and



(a) (b)

Fig. 2 Electroluminescent spectra. (a) Devices A1–E1; (b) devices A2–E2



(a) (b)

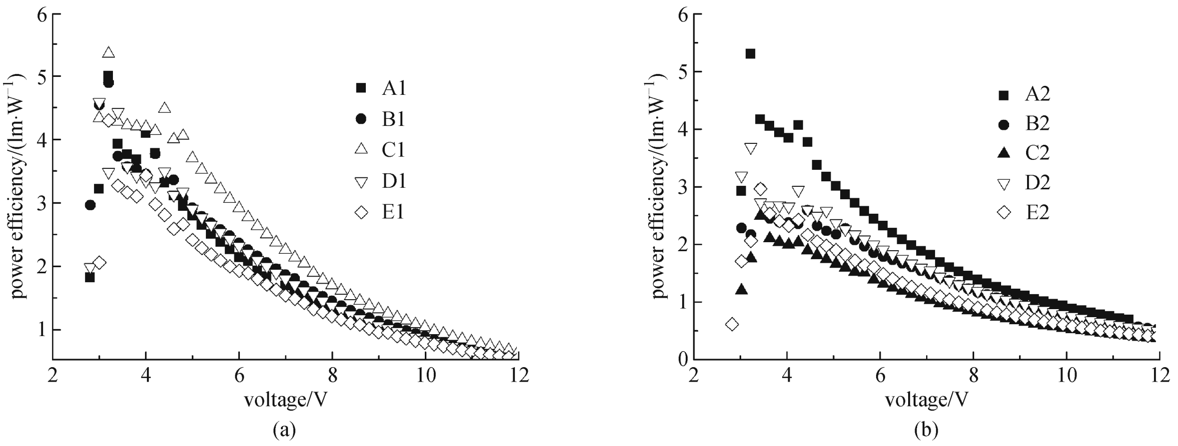
Fig. 3 Luminance-bias voltage curves. (a) Devices A1–E1; (b) devices A2–E2

holes, then lower charge carrier recombination rate, and lower brightness accordingly. In contrast, thinner NPB separator layer, which is more favorable both for direct carrier trapping of the phosphorescent dye and for charge carrier transport to the emission layers than thicker NPB layer, will yield higher luminance.

The power efficiency-bias voltage curves of A1–E1 and A2–E2 are shown in Fig. 4. Device C1 with $x = 7$ nm achieves a maximum power efficiency as high as 5.36 lm/W, whereas the device without thin NPB separator layer reported before achieved only 4.6 lm/W [9], i.e., enhanced by 16.5%. The energy level structure in Fig. 1 indicates that the LUMO and HOMO (the highest occupied molecular orbital) energy level of (t-bt)$_2$Ir(acac) is located in the middle of that of NPB, and the ultrathin phosphor layer sandwiched between two NPB layers is similar to doping in NPB host [18], forming deep electron traps and shallow hole traps in the phosphor layer. More excitons can be efficiently trapped in the (t-bt)$_2$Ir(acac) layer by direct charge carrier trapping mechanism, which has higher efficiency compared to energy transfer mechanism [19]. In the first group, device C1 with 7 nm NPB separator layer has the highest power efficiency dues to the best balance of charge carrier injection in light emission layers. However, for the second group, device A2 with only 5 nm NPB separator layer has the highest power efficiency of 5.3 lm/W. Compared to the first group devices, the thickness of (t-bt)$_2$Ir(acac) emission layer in the second group devices is thinner, so the film thickness of NPB emission layer also need to be thinner in order to achieve a same balance of charge carrier injection for two emission layers as the first group. An ultrathin (t-bt)$_2$Ir(acac) layer can be treated as doping in the NPB host, which is similar to the effect of concentration variance on device performance. It indicates the importance of controlling the dopant concentration in order to obtain high device performance. The thin separator layer confining the emission region also plays an important role on the device performance.

4 Conclusion

Non-doped ultrathin organic light-emitting devices based on a novel iridium phosphorescent dye are fabricated. By



(a)
(b)

Fig. 4 Power efficiency-bias voltage curves. (a) Devices A1–E1; (b) devices A2–E2

changing the film thickness of thin NPB layer, the relationship between the location of the phosphor dye and device performance is studied. The results showed that all the devices have a low turn-on voltage of 2.8 V. When the thickness of the phosphor dye and the NPB emission layer were 0.2 and 5 nm, respectively, a maximum brightness of 18367 cd/m^2 and a maximum efficiency of 5.3 lm/W are achieved. High device performance results from the balance of charge carrier injection with a thin NPB separator layer by confining light emission region, appropriate energy-level alignment, as well as effective charge transporting capability in the device. More work still needed to optimize the device structure so that the device performance can be improved much better.

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