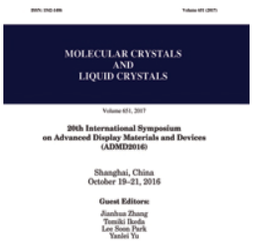

Molecular Crystals and Liquid Crystals

ISSN: 1542-1406 (Print) 1563-5287 (Online) Journal homepage: http://www.tandfonline.com/loi/gmcl20

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To cite this article: Xiang Wei, Hao-Jian Yu, Jin Cao, Chulgyu Jhun & Chao-Ping Chen (2017) Efficient blue and white phosphorescent organic light-emitting diodes with a mixed host in emission layer, Molecular Crystals and Liquid Crystals, 651:1, 108-117, DOI: 10.1080/09273948.2017.1338908

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2017, VOL. 651, 108–117
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Efficient blue and white phosphorescent organic light-emitting diodes with a mixed host in emission layer

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ABSTRACT

Efficient blue and white phosphorescent organic light-emitting diodes (PhOLEDs) with low turn-on voltage were fabricated by using mixed hole and electron transporting layer materials as the co-host in the emissive layer. The optimized performance of blue PhOLEDs was achieved with the current efficiency, external quantum efficiency and power efficiency are 45.11 cd/A, 19.21% and 45.67 lm/W, respectively. Based on the blue-PhOLEDs, efficient white PhOLEDs were achieved with ultrathin green and red emitters were respectively inserted into different locations of the emitting layer. The influences of the locations on the emissive spectra of white PHOLED were investigated. The optimized white PHOLED with the maximum power efficiency of 45.22 lm/W and the peak external quantum efficiency of 16.39% were achieved.

KEYWORDS

Blue PhOLEDs; mixed host; ultrathin emissive layers; white PhOLEDs

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1. Introduction

In recent years, white organic light-emitting diodes (OLEDs) have attracted increasingly attention with a great promise for display or solid-state lighting [1-4]. The phosphorescent OLEDs (PhOLEDs) have great potential due to achieving 100% internal quantum efficiency (IQE) via harvesting both singlet and triplet excitons as light [5–7]. High efficiency, low cost white OLEDs are required to meet widespread commercial applications [8]. To fabricate highly efficient white PhOLEDs, the component of blue emitters is inevitable. The blue phosphorescent materials require that the triplet energy levels of host materials must greater than the dopant (e.g., ≥2.7 eV for Firpic) to ensure the energy can be transported from host to dopant effectively [9–14]. There are some papers reporting that PhOLEDs properties can be improved by bipolar host materials [15–18], which facilitate carrier injection and control the balance of holes and electrons in the emissive layer. However, it's hard to synthesis high-triplet-energy bipolar host materials to satisfy all the conditions for the requirement of blue PhOLEDs [19]. Some papers reported to blend hole-transporting materials and electron-transporting materials as the co-host, which have been demonstrated to help the injection of both holes and electrons [19–22].

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There are many methods to fabricate white OLEDs, such as the doping method [23], which can effectively reduce the concentration quenching effect, leading to high efficiency. However, the doping method also causes some problems in the commercialization, for example, the doping concentration must be controlled very accurately, which increases the cost of devices fabrication. Introducing non-doped emissive layers (EMLs) in white OLEDs is a feasible method to overcome the problem of doping concentration. The non-doped EMLs reduce the cost of white OLEDs [24].

Enlighten from the previous works, blue and white PhOLEDs with co-host in emissive layer were fabricated. Low operating voltage is necessary to achieve a high power efficiency [25], thus MoO_3 doped 4,40,400-tri(N-carbazolyl)triphenylamine (TCTA) as the role of p-type hole transport layer was employed to the devices to reduce the operating voltage and realize high power efficiency [26,27]. On the basis of the efficient blue PhOLEDs, we further investigate the white PhOLEDs. The white PhOLEDs consist of an ultrathin non-doped red EML, an ultrathin non-doped green EML and host-guest blue EMLs. Compared to conventional multiple EMLs white OLEDs, the introduction of ultrathin reduced the thickness of EMLs and save materials. With the adjustment of the devices structures, efficient and low-voltage white PhOLEDs were realized. The optimized white PhOLEDs with the maximum power efficiency of 45.22 lm/W and the peak EQE of 16.39% were achieved.

2. Experimental

2.1. Materials

All materials were purchased from YuRui Chemical Company in Shanghai, China. The chemical structures and detailed energy level diagram of the materials proposed are shown in Fig. 1.

2.2. Device fabrication

The various layers of PhOLEDs in this work were fabricated by a vacuum evaporation technique, and the thickness values and the deposition rates of the materials were monitored in situ by an oscillating quartz thickness monitor. All the layers were fabricated on a glass substrate with a patterned indium tin oxide (ITO), which is 150 nm thickness, ~10 Ω/sq. The ITO glasses were cleaned for 15 min in sequential ultrasonic baths of detergent, de-ionized water, acetone and isopropanol, and then cleaned in a UV-ozone chamber for 15 min. The hole injection layer (HIL), hole transporting layer (HTL), emissive layer (EML), electron transporting layer (ETL), electron injection layer (EIL) and the cathode of aluminum (Al) were deposition sequentially in a high vacuum ($\sim 10^{-4}$ Pa) evaporation device. The 35 nm of MoO_3 doped TCTA was co-evaporated onto the ITO substrate as HIL at a speed of 1 Å/s. On top of HIL layer, a 10 nm HTL (TCTA) was evaporated. Then, an EML with the hosts and guest was co-evaporated. After depositing EML, a 40 nm Tm3PyPB was deposited on top of EML as ETL at a rate of 1 Å/s. Upon the ETL, a 1 nm LiF was deposited and following 120 nm Al cathode, and the deposition rates of LiF and Al were 0.1 Å/s and 5.0 Å/s, respectively. The devices active areas by the overlap of the electrodes were 3×3 mm^2 in all cases.

2.3. Measurements

The voltage-current density, luminance and EL spectra were measured with a programmable source meter (Keithley 2400), luminance meter (LS110, Konica Minolta) and a spectrophotometer (Spectrascan PR670, Photo Research). The time-resolved PL spectra was measured by FLSP 920 spectrometer series.

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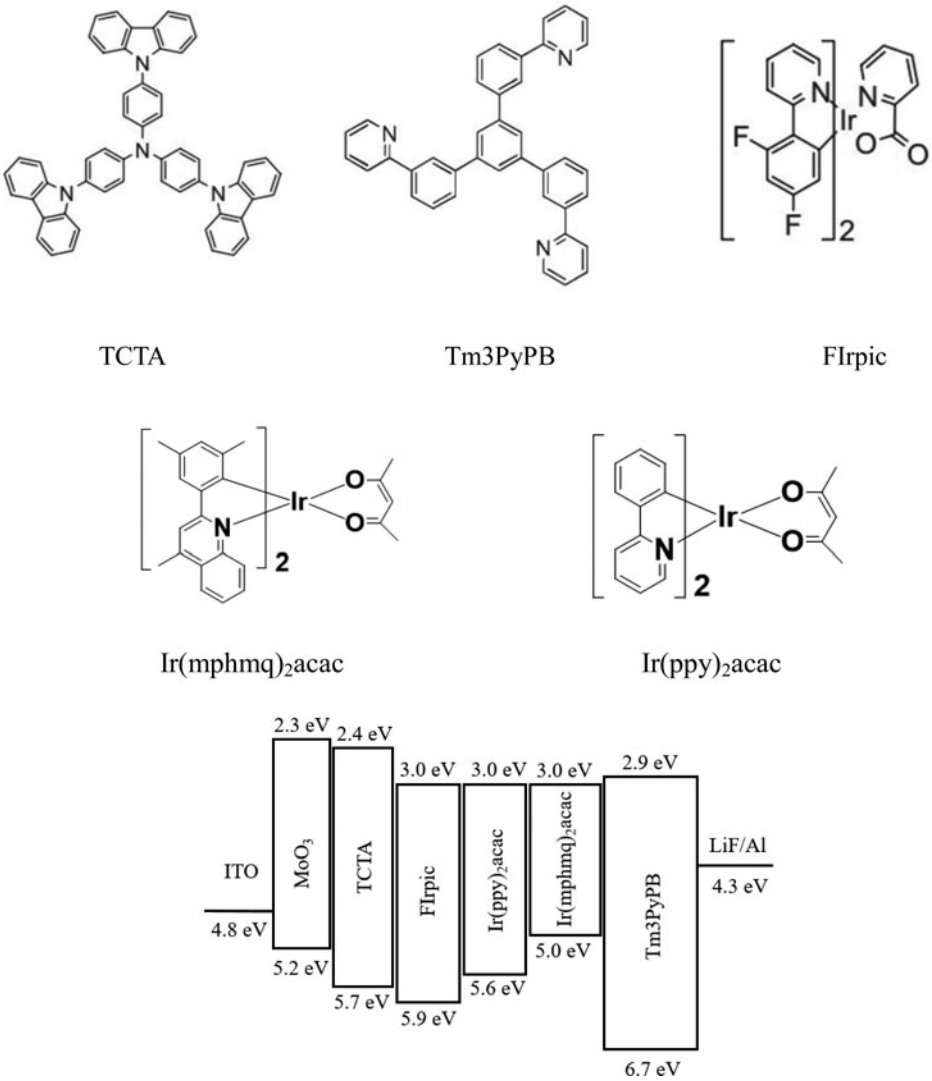

Figure 1. Chemical structures and detailed energy level diagram utilized in this study.

3. Result and discussion

3.1. Hole only and electron only devices

To investigate current characteristics of the mixed host, a series of only- devices were fabricated. The structures of hole only devices and electron only devices were shown below:

Hole only devices: ITO/MoO_3 (5 nm)/TCTA doped Tm3PyPB (X:1, 20 nm)/MoO_3 (5 nm)/Al (120 nm); X = 3, 3.5, 4, 5.

Electron only devices: ITO/TPBi (20 nm)/TCTA doped Tm3PyPB (X:1, 20 nm)/LiF (1 nm)/Al (120 nm); X = 3, 3.5, 4, 5, the proportion in here represents the weight ratio.

Fig. 2 shows the hole current characteristics and electron current characteristics of the devices. For convenience, we use A1, A2, A3 and A4 instead of the devices with TCTA: Tm3PyPB = 5:1, TCTA: Tm3PyPB = 4:1, TCTA: Tm3PyPB = 3.5:1, TCTA: Tm3PyPB = 3:1, respectively.

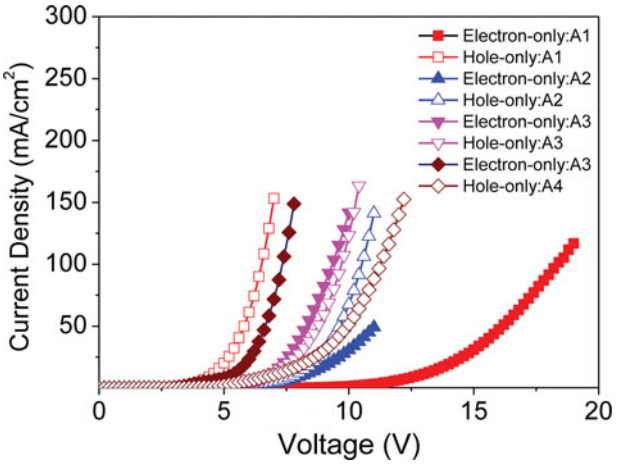

Figure 2. J-V characteristics of hole only devices and electron only devices, the values of proportion from the figure above represent the result using weight of TCTA to divide by the weight of Tm3PyPB.

When the TCTA: Tm3PyPB = 5:1, the current density of the hole-only device was much more than that of electron-only device. With the increase of proportion of Tm3PyPB in the mixed host, the gaps of the current density between hole-only devices and electron-only devices were narrowed. The current density of hole-only device and electron-only device got close to equilibrium when TCTA: Tm3PyPB = 3.5:1. After that, further increasing the proportion of Tm3PyPB in the co-host, the current density of electron only devices would exceed the hole only devices. The optimized ratio of TCTA and Tm3PyPB = 3.5:1 was achieved, the co-host showed the performance same as the synthetic bipolar host materials.

3.2. *Blue PhOLEDs research*

To optimize the performance of the blue PhOLED, the influence of dopant concentration on the electroluminescence was investigated. Devices with three different dopant concentrations (6%, 9% and 12%) were researched. The device structures were depicted in the Fig. 3.

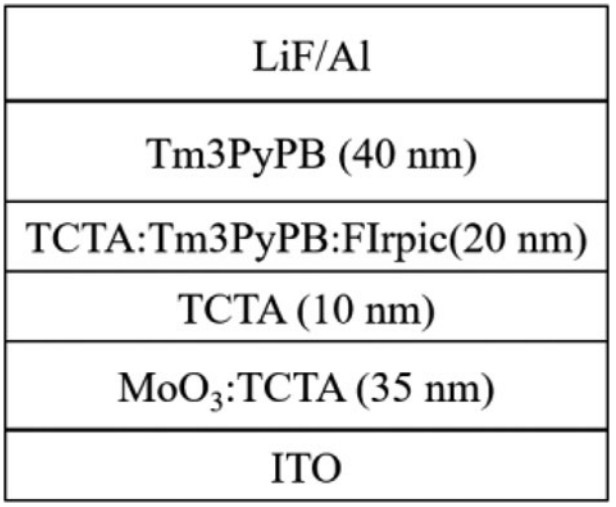

Figure 3. The device structure.

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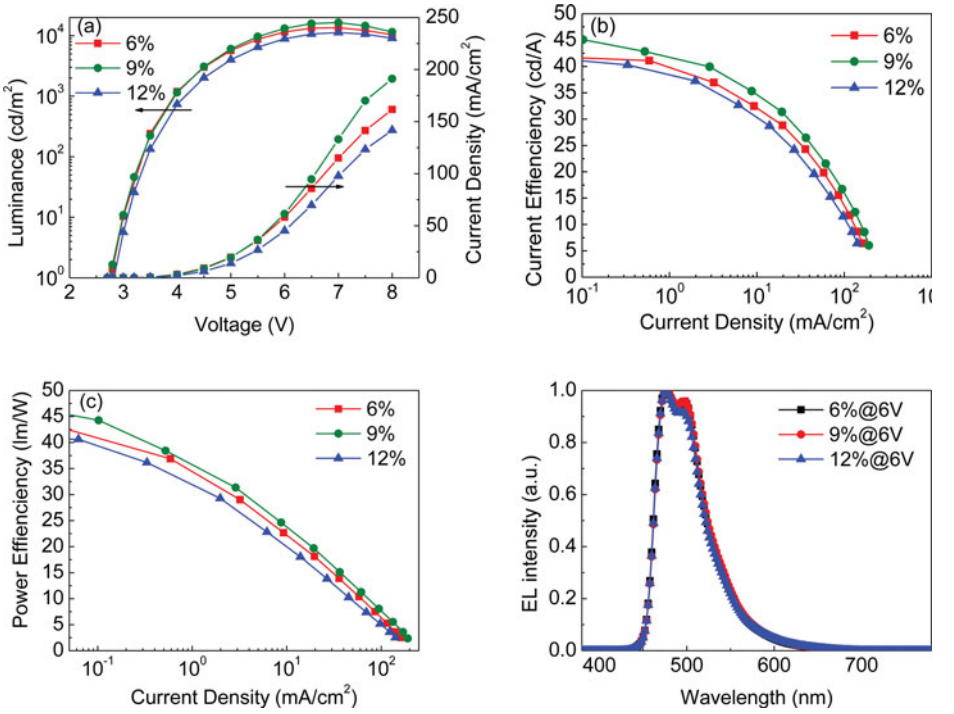

Figure 4. (a) Current density-Voltage-Luminescence (*J-V-L*) characteristic of devices; (b) Current efficiencies versus current density of mixed host blue PhOLEDs with different dopant concentrations; (c) Power efficiencies versus current density of mixed host blue PhOLEDs with different dopant concentration and (d) Normalized EL spectra of devices at 6 V.

The proportion of TCTA: Tm3PyPB is 3.5:1, which is the optimized proportion of hole- and/or electron-only devices. The MoO3 doped TCTA with a ratio of 2:3 as a p-type HIL was designed, which improved the injection of holes and reduced the drive voltage. The thickness of the HIL, HTL, EML, ETL, EIL and Cathode is 35 nm, 10 nm, 20 nm, 40 nm, 1 nm and 120 nm, respectively.

As the current density-voltage-luminance (J-V-L) curves shown in Fig. 4(a), the values of both luminance and current density increased in the order of device 1 (12%)>device 2 (6%)>device 3 (9%). The turn-on voltage of the device with 12% dopant in host: dopant is apparently greater than 6% and 9% dopant in host, which can be ascribed that the high concentration of dopant induced to the concentration quenching [28], and the injection of electrons into the emissive layer may exceed holes, inducing the unbalance of carrier. The decrease in electroluminescent intensity in these devices at high current densities is due to triplet-triplet annihilation [29]. The performance of the device with 6 wt% dopant in host is inferior to the device with a 9 wt% dopant concentration because the energy transferred from host material to the dopant is incomplete. The devices achieves optimal performance when the dopant concentration is 9 wt%, with maximum current efficiency of 45.11 cd/A, power efficiency of 45.67 lm/W. Fig. 4(d) shows the EL spectra of the three devices at 6 V, all the devices shows a peak at 476 nm, which correspond to the blue emission of FIrpic.

To achieve insight into the blue PhOLEDs based on mixed host, a series of blue PhOLEDs devices were fabricated. The doping concentration of FIrpic was fixed at 9%, and structures of these devices were ITO/MoO_3:TCTA (35 nm)/TCTA:Tm3PyPB:FIrpic or TCTA:FIrpic or Tm3PyPB:FIrpic (20 nm)/ Tm3PyPB (40 nm)/LiF (1 nm)/Al(120 nm). To explore the

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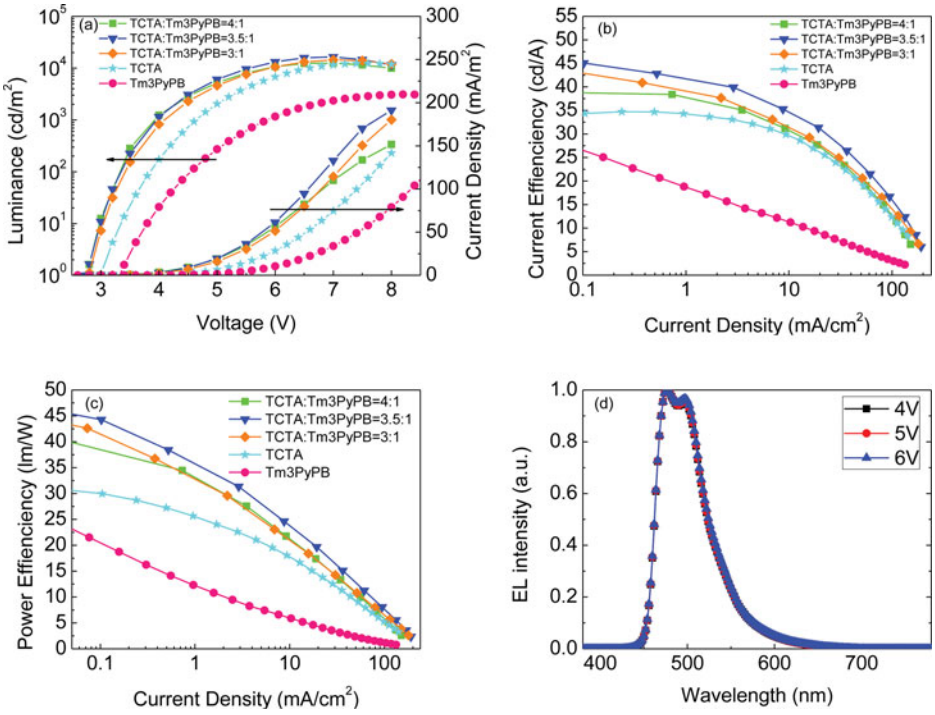

Figure 5. Electroluminescent characteristics of blue PhOLEDs with different structures of host. (a) *J-V-L* characteristics; (b) current efficiency-current density characteristics; (c) power efficiency-current density characteristics and (d) EL spectra of the device B2 with the optimized concentration at the operating voltages of 4 V, 5 V and 6 V, respectively.

influence of the proportion of TCTA to Tm3PyPB on the blue PhOLEDs devices, three different doping ratio with TCTA:Tm3PyPB = 4:1, 3.5:1, 3:1 were fabricated, respectively. The devices of TCTA host and Tm3PyPB were also fabricated.

Figure 5(a) shows the J-V-L characteristics of the blue devices. We use devices B1, B2, B3, B4 and B5 instead of the devices based on the host of TCTA:Tm3PyPB = 4:1, TCTA:Tm3PyPB = 3.5:1, TCTA:Tm3PyPB = 3:1, TCTA and Tm3PyPB, respectively. The luminance and current density increases in order of the B5, B4, B3, B1 and B2, respectively. The turn-on voltage of the device B2 is the minimum among these devices with different host materials. The devices based on co-host with different doping ratio were fabricated to confirm the result of only devices and it turned out that the blue PhOLEDs in accord with the result of the hole- and/or electron-only devices. Fig. 5(b) and Fig. 5(c) showed the current efficiency-current density and power efficiency-current density properties. The device B2 has the optimal electroluminescent performance due to the optimized mixed host facilitating the injection of holes and electrons. The balance of the carriers in the emission layer is also improved by the mixed host. FIrpic is a well-known electron-transport triplet emitter, electrons can be injected and electron hopping can be achieved among FIrpic molecules, which indicates that the electrons in the emissive layer may exceed holes in the device based on the host material of Tm3PyPB, which unbalances the carrier, leading to the reduction of efficiencies. Fig. 5(d) showed the electroluminescent spectra of the device B2, the CIE coordinates of the device are the same at (0.16, 0.38) at the operating voltage of 4 V, 5 V and 6 V, respectively. The maximum efficiencies with current efficiency of 45.11 cd/A, power efficiency of 45.67 lm/W and external quantum efficiency of 19.21% were realized by the device B2.

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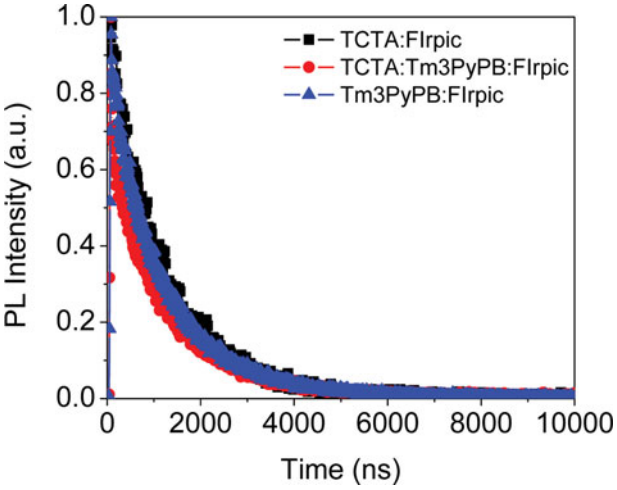

Figure 6. Time-resolved PL decays of emission films.

To further explore the characteristics of the mixed host system, the photoluminescence (PL) transient characteristics with different emission films were fabricated. Fig. 6 shows the time-resolved PL decays in different cases of emission films (film 1: TCTA: FIrpic, film 2: TCTA:Tm3PyPB:FIrpic, film3: Tm3PyPB:FIrpic). The PL spectra of films were monitored at $\lambda = 470$nm to investigate the emission of FIrpic in different host. The lifetime of the films 1-3 is almost unanimous, the value of the lifetime of film 2 is imperceptibly inferior to the film1 and film 3, which can be explained by the undesired energy transfer from TCTA to Tm3PyPB [30,31]. It implicated that the improvement of efficiency of device based on mixed host was mainly attributed to the bipolar transport characteristic, which both facilitated the injection of carriers and balanced the holes and electrons in EML.

The detailed EL data were listed in the table 1. The maximum power efficiency of this structure is 45.67 lm/W, which is 49.64% more than that 30.52 lm/W of the device with the host of TCTA and 97.28% more than 23.15 lm/W of the device with the host of Tm3PyPB. The maximum EQE arrives at 19.21%.

Table 1. The detailed EL performances of devices B1-B5.

		η_c (cd/A)[b]		η_p (lm/W)[c]		η_{ext} (%)[d]	
Device	V_{on} (V)[a]	Max[e]	100[f]	Max[g]	100[h]	Max[i]	100[j]
B1	2.74	38.94	38.65	40.01	36.65	17.38	16.69
B2	2.72	45.11	43.95	45.67	41.48	19.21	18.72
B3	2.80	42.90	41.67	43.48	38.39	18.27	17.46
B4	2.99	34.80	30.71	30.52	28.44	14.59	14.50
B5	3.33	26.61	21.28	23.15	14.75	11.92	9.25

[a]At 1 cd/m^2.
[b]Current efficiency.
[c]Power efficiency.
[d]External quantum efficiency.
[e]Maximum current efficiency.
[f]Current efficiency at the luminance of 100 cd/m^2.
[g]Maximum power efficiency.
[h]Power efficiency at the luminance of 100 cd/m^2.
[i]Maximum external quantum efficiency.
[j]External quantum efficiency at the luminance of 100 cd/m^2.

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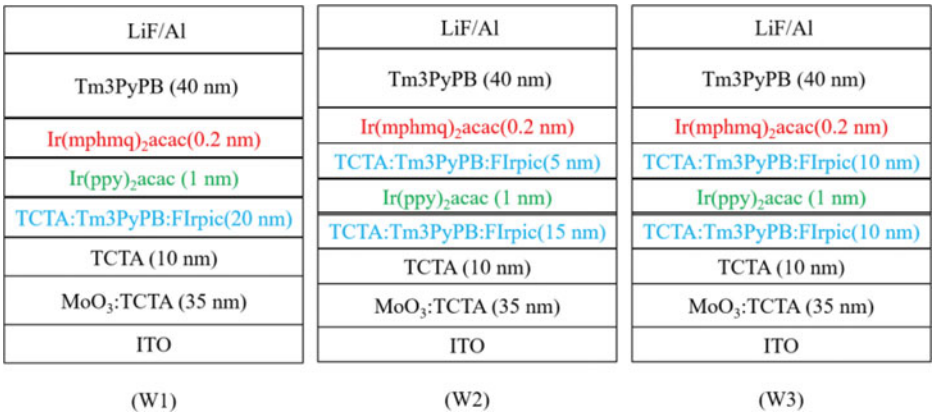

Figure 7. The structures of devices W1-W3.

3.3. White PhOLEDs research

On the basis of blue PhOLEDs, highly efficient white PhOLEDs with mixed-host blue EMLs, an ultrathin non-doped green EML, and an ultrathin non-doped red EML were fabricated. To realize efficient white PhOLEDs, we investigate the location of the ultrathin EMLs to make the best of the excitons in the excitons recombination zone. Three devices as shown in Fig. 7 were fabricated. The green EML was placed in three different situations to explore the excitons recombination zone. The structure of device is ITO/MoO_3:TCTA (35 nm)/TCTA (10 nm)/TCTA:Tm3PyPB:FIrpic (3.5:1, 9%, X nm)/Ir(ppy)$_2$acac (1 nm)/ TCTA:Tm3PyPB:FIrpic (3.5:1, 9%, 20-X nm)/Ir(mphmq)$_2$acac (0.2 nm)/Tm3PyPB (40 nm)/LiF (1 nm)/Al (120 nm), the X was chosen as 20, 15 and 10 for devices W1, W2 and W3, respectively. The J-V-L, J-CE, J-PE and the EL spectra were shown in Fig. 8, the detailed performances of these devices are summarized in Table 2. The peak current efficiencies of the devices W1, W2 and W3 are 18.19 cd/A, 43.14 cd/A and 35.11 cd/A, respectively. From the Fig. 8(d), it can be seen that the device W1 does not contain green light, which indicates that the excitons quenching in this device is very serious, thus excitons confinement are not effective [32]. The J-V-L properties of device W2 and device W3 were shown in Fig. 8(a), indicating that the ultrathin non-doped EMLs hardly have influence on the carrier transporting. As shown in Fig. 8(d), it implies that the white OLEDs can be fabricated and the colors can

Table 2. The detailed EL performances of devices W1-W3.

Device	V_{on}(V)[a]	η_c (cd/A)[b]		η_p (lm/W)[c]		η_{ext} (%)[d]	
		Max[e]	100[f]	Max[g]	100[h]	Max[i]	CIE@7V
W1	2.79	18.19	18.02	18.75	16.32	12.03	(0.41,0.36)
W2	2.79	43.14	41.38	45.22	37.64	16.39	(0.38,0.44)
W3	2.76	35.11	33.77	37.56	31.02	15.74	(0.43,0.41)

[a]At 1 cd/m^2.
[b]Current efficiency.
[c]Power efficiency.
[d]External quantum efficiency.
[e]Maximum current efficiency.
[f]Current efficiency at the luminance of 100 cd/m^2.
[g]Maximum power efficiency.
[h]Power efficiency at the luminance of 100 cd/m^2.
[i]Maximum external quantum efficiency

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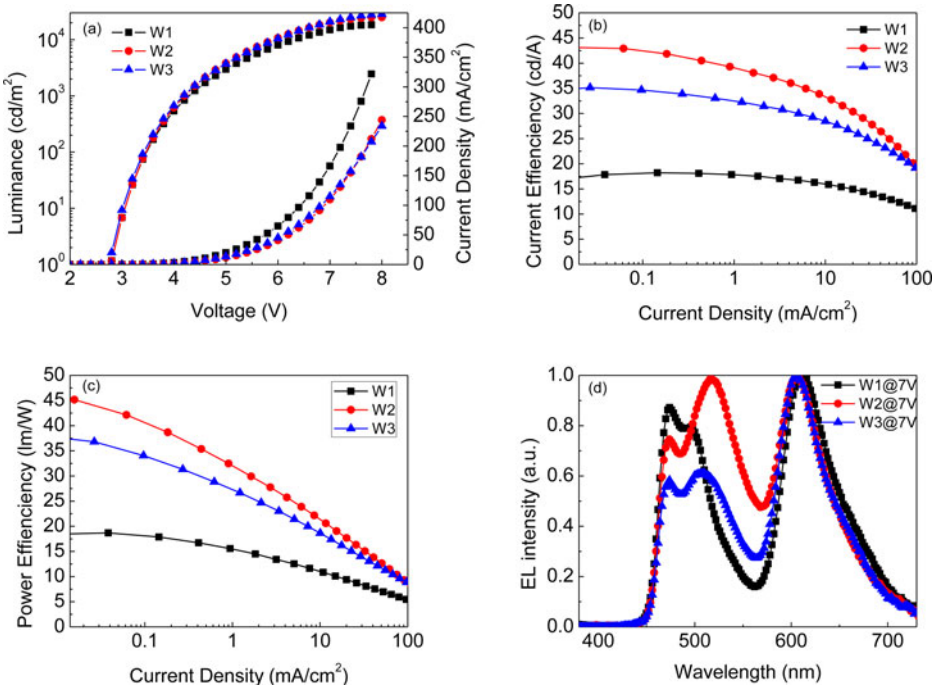

Figure 8. (a) *J-V-L* characteristics of devices W1-W3; (b) J-CE characteristics of devices W1-W3; (c) J-PE characteristics of devices W1-W3 and (d) Normalized EL spectra of the three devices at the voltage of 7 V.

be tuned by changing the location of green layer. The blue EML between the ultrathin green EML and red EML takes effect not only for blue lighting but also suppress the energy transfer between green EML and red EML, which is necessary to get three-band white emission. When the thickness of the blue EML between two ultrathin EMLs is 5 nm in device W2, the white OLED spectra have three peaks of 476 nm, 520 nm and 610 nm, corresponding to the blue emission of FIrpic, green emission of $Ir(ppy)_2acac$ and red emission of $Ir(mphmq)_2acac$, respectively. However, when the thickness of the interlayer increased to 10 nm in device W3, the green emission dramatically reduced. Simultaneously, the current efficiency and power efficiency decreased dramatically. Though the exciton recombination zone is slightly extended toward HTL with the increase of driving voltage, the green EML was placed far away from the excitons recombination zone, which led to the weak green emission. In all, optimized white PhOLEDs with the maximum power efficiency of 45.22 lm/W and the peak EQE of 16.39% were fabricated. The CIE coordinates changed from (0.49, 0.44) to (0.47, 0.40), corresponding to the luminance changed from 100cd/m^2 to 10000cd/m^2.

4. Conclusion

In summary, we reported a mixed-host system to realize efficient and low-voltage blue and white PhOLEDs. The ultrathin green EML and red EML were introduced to achieve white emission without increasing driving voltage. We investigated the influence of the EMLs locations on the emissive spectra and efficiency of white PhOLED. The optimized white PhOLED with maximum power efficiency of 45.22 lm/W and peak EQE of 16.39% were achieved.

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