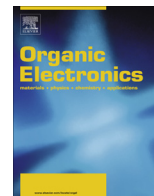




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Extremely high chromatic-stability white organic light-emitting device with symmetrical cascade emissive layer

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ABSTRACT

We have employed four adequate complementary phosphor emitters (blue, green, yellow, and red) and two host and transport materials (CBP: 4,4'-bis(carbazol-9-yl)biphenyl; TPBi: 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole)) to configure a symmetrical cascade emissive layer white organic light-emitting device (WOLED), in which four emitters are in turn incorporated in CBP and TPBi from high to low energy level from CBP/TPBi interface. The symmetrical cascade emissive layer WOLED shows extremely high chromatic-stability with a slight CIE coordinates shift at wide range of driving voltage. As the voltage increases from 5 V to 9 V, the symmetrical cascade emissive layer WOLED maintains CIE coordinates of (0.384, 0.467)–(0.387, 0.469) and high color rendering index (CRI) of 85–86 with ideal correlated color temperature (CCT) of 4407–4292 K. Furthermore, this WOLED also exhibits a peak current efficiency of 43.6 cd/A and a maximum power efficiency of 31.1 lm/W, which are much higher (by 1.43 and 1.50 times) than those of the reference device with four emitters only being doped in single CBP host. The improvement of color stability is due to the fact that symmetrical cascade emissive layer suppresses the shift of carrier recombination zone as the voltage increases; the improvement of efficiency is attributed to the effective utilization of excitons.

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1. Introduction

White organic light-emitting devices (WOLEDs) have attracted considerable attention in recent years owing to their potential applications in next-generation full-color flatpanel displays and solid-state lighting sources [1–6]. The research in WOLEDs has mainly focused on improving the device efficiency, color rendering index (CRI), and color stability. To improve these performances, much work has been carried out either through better materials and more efficient device architecture or a combination of both. From materials aspect, the use of phosphorescent emitters in WOLEDs has been demonstrated to be the most effective method because phosphors can harvest both singlet and triplet excitons and realize a theoretical internal quantum efficiency of 100%

[7–9]. In term of device architecture, a variety of methods have been reported to realize high performance WOLEDs, including single emissive layer WOLEDs [10,11], hybrid WOLEDs [12–15], tandem WOLEDs [16–18], and multiple emissive layer (M-EML) WOLEDs [19–24]. Among them, M-EML WOLEDs technology, which has good reproducibility and simplified device preparation process, is considered to be one of the most cost-effective methods to achieve WOLEDs aiming for mass production [25]. However, especially in three or more color M-EML WOLEDs, the color stability is a major challenge because of carrier recombination zone alteration in different EMLs under different voltages. Obviously, the color stability is a key factor as important as other parameters such as high efficiency. Without high color stability, the appearance of items in a room illuminated via WOLEDs would be dependent on the operating voltage of WOLEDs, which is unacceptable for general use. In addition, in M-EML WOLEDs, in order to confine charge carriers in intended recombination zone, more charge blocking layer and interval layer are introduced to WOLEDs, leading to an increased heterojunction interfaces. Though the increased

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heterojunction interfaces are favored for controlling the emission intensity of different emitters and obtaining wide spectrum white device [26,27], they also lead to a high driving voltage and a low device efficiency and lifetime, because the energy barriers at the heterojunction interfaces lead to difficulty for carrier injection/transport and accumulations of space charges at interfaces [28–30]. However, these heterojunction interfaces are inevitable for traditional M-EML WOLEDs. So, rational design of the device structure with reduced heterojunction interfaces to develop high color stability WOLEDs has been taken into account nowadays.

In 2013, Chang et al. [20] demonstrated all-phosphor, four-color (i.e., blue, green, yellow, and red emissions) WOLED with only one organic/organic heterojunction interface (CBP: 4,4'-bis(carbazol-9-yl)biphenyl/TPBi: 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole)), in which four complementary emitters were incorporated in CBP from high to low energy level from excitons-generating interface (CBP/TPBi) without interlayer. This WOLED obtained a high efficiency and CRI by triplet exciton conversion (TEC) process. However, this WOLED also suffered from a poor color stability because of the shift of the exciton generation towards the yellow and red doped regions at higher driving voltages.

In this work, firstly, we chose CBP as triplet host and hole transport material and TPBi as triplet host and electron transport material to fabricate a reference WOLED (see W0 in Fig. 1(b)), in which red, yellow, green and blue four emitters were only doped in CBP as the case of the paper by Chang et al. [20]. Then, the reference device was modified by doping different emitters into TPBi, and finally developed a symmetrical cascade emissive layer WOLED (see W4 in Fig. 1(b)), in which four complementary emitters were in turn incorporated symmetrically in the host CBP and TPBi from high to low energy level without any spacers from CBP/TPBi interface. Compared with reference device, the symmetrical cascade emissive layer WOLED shows extremely high color stability with a slight CIE coordinates shift at wide range of driving voltage. As the voltage increases from 5 V to 9 V, the symmetrical cascade emissive layer WOLED maintains CIE coordinates of (0.384, 0.467)–(0.387, 0.469) and high CRI of 85–86 with ideal correlated color temperature (CCT) of 4407–4292 K. In addition, the symmetrical cascade emissive layer WOLED also exhibits a maximum current efficiency (CE) of 43.6 cd/A and a maximum power efficiency (PE) of 31.1 lm/W, which are larger by 1.43 and 1.50 times, respectively, those of unilateral non-symmetrical cascade emissive layer WOLED (see W0 in Fig. 1(b)).

2. Experimental

The devices were fabricated on the pre-patterned indium tin oxide (ITO) glass substrate with sheet resistance of $15 \Omega/\square$ by vacuum deposition. ITO substrates as the anode were scrubbed and sonicated consecutively with detergent water, deionized water, and acetone, dried in drying cabinet, and then exposed to a UV-ozone environment for 10 min. After these processes, the substrates were transferred into a vacuum chamber for sequential deposition of all organic functional layers by thermal evaporation below a vacuum of 5×10^{-4} Pa. The deposition rates for organic materials, molybdenum trioxide (MoO_3), lithium fluoride (LiF), and aluminum (Al), were about 1, 0.3, 0.1 and 3 Å/s, respectively. The thickness and deposition rates of the films are controlled by a quartz thickness monitor. The overlap between ITO anode and Al cathode is $3 \text{ mm} \times 3 \text{ mm}$, as the active emissive area of the devices. The proposed structure of the white devices with different emissive layer profiles is shown in Fig. 1. These WOLEDs are named Devices W0, W1, W2, W3, and W4, in which Device W0 was used as a reference, Devices W1, W2, W3, and W4 were optimized by in-

turn doping different emitters into TPBi layer from CBP/TPBi interface.

In this work, bis(2-(3-trifluoromethyl-4-fluorophenyl)-4-methylquinolyl) (acetylacetonate)iridium(III) (**Ir(ffpmq)₂(acac)**) was synthesized in our laboratory. Other materials used for device fabrication were obtained through commercial sources and used without further purification. In these devices, bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl) iridium(III) (**Flrpic**), tris(2-phenylpyridine)iridium(III) (**Ir(ppy)₃**), $\text{Ir}(\text{ffpmq})_2(\text{acac})$, and bis(1-phenylisoquinoline) (acetylacetonate)iridium(III) (**Ir(piq)₂(acac)**) were used for blue, green, yellow, and red emission, respectively. CBP served as hole transport layer (HTL) and triplet host, and TPBi served as electron transport layer (ETL) and triplet host. MoO_3 , LiF and Al were used as hole injection layer (HIL), electron injection layer (EIL) and cathode, respectively.

The current–voltage–forward luminance characteristics of devices were measured with a ST-900M luminance meter and a Keithley 2400 programmable voltage current source. CIE coordinates, EL spectra, CRI and CCT of devices were measured with a computer controlled PR655 Spectra scan spectrometer. All the samples were measured directly after fabrication without encapsulation in ambient atmosphere at room temperature in the darkroom.

3. Results and discussion

The M-EML WOLEDs technology has many advantages for mass production, but more heterojunction interfaces lead to a high driving voltage and a low device efficiency and short lifetime, which limit the device reliability. So, it is necessary to reduce the number of heterojunction interfaces of M-EML WOLEDs for improving performance of WOLEDs. Based on this idea, the broadband gap CBP and TPBi were selected as HTL and ETL, respectively, to structure WOLEDs with only one organic/organic heterojunction interface. Electrons and holes should combine at CBP/TPBi interface to form excitons, which can diffuse freely from CBP/TPBi interface to deeper CBP and TPBi layers. In order to validate this conjecture, firstly, two blue devices were fabricated with the structures of ITO/ MoO_3 (3 nm)/CBP (47 nm)/CBP: 8 wt% Flrpic (8 nm)/TPBi (55 nm)/LiF (1 nm)/Al (200 nm) and ITO/ MoO_3 (3 nm)/CBP (55 nm)/TPBi: 8 wt% Flrpic (8 nm)/TPBi (47 nm)/LiF (1 nm)/Al (200 nm), as named Device B1 and Device B2, respectively. Except for the difference in doping position of blue emitter, Devices B1 and B2 have the same device structures and total film thickness.

Fig. 2(a) shows the luminance–voltage characteristics of Devices B1 and B2. Devices B1 and B2 exhibit the maximum luminance of 9196 cd/m^2 and 8581 cd/m^2 , respectively. These results indicate that excitons can be exactly formed on both sides of CBP/TPBi interface, and if CBP or TPBi side is doped with Flrpic, efficient energy transfer occurs from host CBP or TPBi to Flrpic. The EL spectra of Devices B1 and B2 (Fig. 2(b)) further confirm this point, in which the maximum emission peaks of two devices are all located at 472 nm with a shoulder peak at 498 nm, exclusively originating from the triplet excited state emission of the phosphor Flrpic [5]. On the other hand, the small emission peaks at 404 nm in Device B1 and at 396 nm in Device B2 can be observed, which are corresponding to the emission of CBP and TPBi, respectively; no emission peak occurs at wavelengths longer than 500 nm in these devices, indicating no exciplex formation at the interface between CBP and TPBi. These weak emission peaks are mainly attributable to the <100% energy transfer from CBP and TPBi to Flrpic in Devices B1 and B2.

In order to testify whether the excitons formed at CBP/TPBi interface can diffuse into deeper CBP and TPBi layers, further, two red

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