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Highly efficient and high colour rendering index white organic light-emitting devices using bis(2-(2-fluorphenyl)-1,3-benzothiozolato-N,C^{2'}) iridium (acetylacetonate) as yellow emitter

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Abstract

Multiple emissive layer (MEML) white organic light-emitting devices (OLEDs) are fabricated. In the MEML, the yellow phosphorescent emitter bis(2-(2-fluorphenyl)-1,3-benzothiozolato-N, $C^{2'}$) iridium(acetylacetonate) [(F-BT)₂Ir(acac)] is introduced to codope with the red one bis(2,4-diphenyl-quinoline) iridium acetylacetonate [Ir(piq)₂acac] into 4,4'-N,N'-dicarbazole-biphenyl (CBP), fac tris(2-phenylpyridine) iridium [Ir(ppy)₃]-doped CBP is used for green emitting and N,N'-di(naphthalene-1-yl)-N,N'-diphenyl-benzidine (NPB) is used for both hole transporting and blue emitting. By adjusting the devices' structure, we get the OLED which has the maximum luminance of 50 030 cd m^{-2} , maximum efficiencies of 21.6 cd A^{-1} (at 7 V), 10.3 lm W^{-1} (at 5 V) and 8.4% (at 7 V) and colour rendering index (CRI) of 85. In addition, its Commission International de L'Eclairage (CIE) coordinates slightly change from (0.3867, 0.4075) to (0.3638, 0.4070) when the drive voltage shifts from 7 V (1862 cd m^{-2}) to 13 V $(39\ 270\ cd\ m^{-2}).$

Research on the organic light-emitting device (OLED) has developed extraordinarily rapidly since the report of the first high-efficiency and luminance OLED by Tang and coworkers in 1987 [1]. Nowadays, OLED products have been commercially manufactured for small area displays which are used in cell phone, MP3, digital camera, etc. Moreover, the potential application of OLED in the field of lighting is also becoming a realization. White OLED (WOLED) is a hotspot of the field since both full colour displays and solid-state lighting mentioned above can be realized based on WOLED [2, 3]. Many groups are devoted to exploiting the high-efficiency and high-CRI WOLEDs [2, 4–8]. As compared with WOLEDs based on fluorescent materials, phosphorescent-based WOLEDs are more attractive to researchers because both singlet and triplet excitons can be harvested in the phosphorescent materials [9–13] and that may make the phosphorescent OLED efficiencies quadruple those of fluorescent ones. In this paper, we will present high-



Figure 1. The device architecture of device I, device II and device III and the chemical structure of (F-BT)₂Ir(acac).

efficiency and high-CRI MEML [4–6, 14–16] WOLEDs which use the phosphorescent iridium complex $(F-BT)_2Ir(acac)$ as the yellow emitter.

We have fabricated WOLEDs in three different configurations named as device I, device II and device III. The device architecture of device I, device II and device III and the chemical structure of (F-BT)₂Ir(acac) are shown in figure 1. In the devices, 30 nm 4,4',4''-tris(3methylphenylamino)-triphenylamine (m-MTDATA) and 40 nm bathophenanthroline (Bphen) are used as the hole-injecting and electron-transporting/hole-blocking layers (EML/HBL), respectively. The 20 nm NPB layer is for hole transporting and blue emitting; Ir(ppy)₃ and Ir(piq)₂acac are used as green and red emitters, respectively. We designed and synthesized the yellow emitter (F-BT)₂Ir(acac) ourselves. The synthetic procedure and analysis of the (F-BT)₂Ir(acac)-based device performance will be published in another paper. In all three devices, the doping concentrations of (F-BT)₂Ir(acac) and Ir(piq)₂acac in CBP are fixed at 5 wt% and 3 wt%, and the doping concentration of Ir(ppy)₃ in CBP is fixed at 8 wt%. In order to keep both devices working in the same overall thickness, the thicknesses of the two doping layers are both 10 nm in device I and 8 nm in device II and device III. The EL spectra and CIE coordinates of these devices were measured by a PR650 spectrascan spectrometer. The luminance-current density-voltage characteristics were recorded simultaneously with the measurement of the EL spectra by combining the spectrometer with a Keithley

model 2400 programmable voltage–current source. All measurements described above were carried out at room temperature under ambient conditions.

Device performances (including current density versus voltage, luminance versus voltage and efficiencies versus voltage) of device I are shown in figure 2. Maximum luminance and maximum efficiencies (current and power efficiency) of the device are 43 450 \pm 1700 cd m⁻² (at 15 V), 16.1 \pm 0.6 cd A^{-1} (at 7 V, 1530 \pm 60 cd $m^{-2})$ and 7.3 \pm 0.3 lm W⁻¹ (at 5 V, 290 \pm 10 cd m⁻²), respectively; the maximum external quantum efficiency (EQE) of the device is 6.8 \pm 0.3% (at 7 V). The normalized EL spectrum of device I at a drive voltage of 7 V has been shown in figure 3(a). For ease of analysis, the spectra are normalized at a wavelength of 616 nm which is the location of the emission peak from Ir(piq)₂acac. The CIE coordinates of the device at 7 V are (0.3789, 0.4092) and the CRI is 82. We find that even when the doping concentration of $Ir(ppy)_3$ reaches 8 wt%, the green emission from it is very weak. Apparently, this can be attributed to the triplet energy transfer from green to yellow and red iridium emitters because Ir(ppy)₃ has a relatively higher triplet energy level compared with (F-BT)₂Ir(acac) and Ir(piq)₂acac. It is clear that, as far as our devices are concerned, to fabricate a high-efficiency WOLED the presence of Ir(ppy)₃ emission is necessary. In order to eliminate that transfer, we add a 4 nm undoped CBP layer between the two EMLs (device II). The triplet energy transfer (Dexter transfer) requires spatial overlap of the donor/accepter molecular



Figure 2. Performances of devices. (*a*) Open circle, open triangle and open square are for luminances of device I, device II and device III, respectively; closed circle, closed triangle and closed square are for current densities of device I, device II and device III, respectively. (*b*) Open triangle, open circle and open square are for current efficiencies of device I, device II and device III, respectively; closed triangle, closed circle and closed square are for power efficiencies of device I, device II and device III, respectively.

orbitals and it can only happen significantly within a range of 1–2 nm. So, the 4 nm interlayer can effectively prevent the transfer mentioned above. From device II's EL spectra (at 7 V), which are shown in figure 3(*b*), we find that the green emission is enhanced. Performances of device II are also shown in figure 2. Maximum luminance and efficiencies of device II are 46 530 \pm 1800 cd m⁻² (at 15 V), 17.9 \pm 0.7 cd A⁻¹ (at 7 V, 1608 \pm 60 cd m⁻²), 8.2 \pm 0.3 lm W⁻¹ (at 6 V, 770 \pm 30 cd m⁻²), 7.4 \pm 0.3% (at 7 V), (0.4046, 0.4140) (at 7 V) and a CRI of 85, respectively. However, as shown in figure 3, although the Ir(ppy)₃ emission is enhanced in device II, the enhancement is limited. It seems that other process(es) may also affect the triplet green emission undesirably.

In order to improve the performance of the devices, we fabricated device III. In device III, we exchange the locations of the two phosphorescent EMLs in device II. We observe the EL spectrum of device III under drive voltage of 7 V (which is shown in figure 4(a)) and find that green emission is enhanced obviously. The phenomenon may be attributed as follows: as we all know, in OLEDs there are always more excitons formed at the interface between the EML and ETL than those at the EML and HTL interface because of the imbalanced injection



Figure 3. EL spectra of (a) device I and (b) device II.

of holes and electrons. In device III, the Ir(ppy)₃-doped layer is adjacent to ETL so more excitons can be harvested by Ir(ppy)₃ to enhance green emission. Device III's performance is also shown in figure 2: the maximum luminance reaches $50\ 030 \pm 2000\ \text{cd}\ \text{m}^{-2}$ at 15 V, the maximum current efficiency, maximum power efficiency and EQE are $21.6 \pm 0.9\ \text{cd}\ \text{A}^{-1}$, $10.3 \pm 0.4\ \text{Im}\ \text{W}^{-1}\ \text{and}\ 8.4 \pm 0.3\%$, respectively, they are obtained at 7 V ($1862 \pm 70\ \text{cd}\ \text{m}^{-2}$), 6 V ($790 \pm 30\ \text{cd}\ \text{m}^{-2}$) and 7 V, respectively. Device III's power efficiency versus luminance plot is shown in figure 4(*b*), the efficiency is about $10\ \text{Im}\ \text{W}^{-1}\ \text{at}\ 1000\ \text{cd}\ \text{m}^{-2}$, and even at $10\ 000\ \text{cd}\ \text{m}^{-2}$ the efficiency is still above 6 Im W⁻¹.

Figure 5 shows the normalized EL spectra of device III with the drive voltage of 4 V, 5 V, 6 V, 7 V and then 12 V, the spectra are also normalized at the peak wavelength of the red light for simplicity of analysis. In the beginning (4 V), the emission of $Ir(piq)_2acac$ is much stronger than others, this may be attributed to two aspects: (i) the stronger trapping ability of $Ir(piq)_2acac$ compared with (F-BT)₂Ir(acac) and Ir(ppy)₃ and (ii) energy transfer from (F-BT)₂Ir(acac) to the red dopant. With increasing drive voltage, more carriers inject into MEML and more excitons



Figure 4. EL spectrum and power efficiency versus luminance plot of device III.



Figure 5. EL spectra of device III under different drive voltages.

are formed/harvested in $(F-BT)_2Ir(acac)$ and $Ir(ppy)_3$, thus green and yellow emissions are obviously enhanced over the

red one. Moreover, we find that when the voltage increases from 7 V to 13 V, the normalized EL spectra only experience a little change. This makes the CIE coordinates only slightly vary from (0.3867, 0.4075) to (0.3638, 0.4070) with the luminance ranging from 1862 \pm 70 cd m⁻² to 39 270 \pm 1600 cd m⁻² and the CRI increasing from 85 to 86.

In conclusion, we have fabricated WOLEDs utilizing three iridium–ligand complexes as the green, yellow and red phosphorescent emitters and NPB as the blue fluorescent emitter. By adjusting device structure, we get the optimal WOLED with a CRI of 85 at CIE coordinates of (0.3867, 0.4075) and maximum efficiencies of 21.6 ± 0.9 cd A⁻¹, $10.3 \pm 0.4 \text{ Im W}^{-1}$ and $8.4 \pm 0.3\%$, at 7 V ($1862 \pm 70 \text{ cd m}^{-2}$), 6 V ($790 \pm 30 \text{ cd m}^{-2}$) and 7 V. At luminance of 1000 cd m^{-2} and 10 000 cd m⁻², the power efficiency is about 10 Im W⁻¹ and above 6 Im W⁻¹, respectively. And the CIE coordinates only slightly vary from (0.3867, 0.4075) to (0.3638, 0.4070) when the drive voltage shifts from 7 V ($1862 \pm 70 \text{ cd m}^{-2}$) to 13 V ($39 \text{ } 270 \pm 1600 \text{ cd m}^{-2}$).

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