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Efficient white organic light-emitting diodes based on iridium complex sensitized copper complex

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Abstract

Efficient double emission-layer white organic light-emitting diodes comprising a yellow emission from bis[(4,6-difluorophenyl)-pyridinato-N,C²](picolinato)Ir(III) (FIrpic) sensitized [Cu(bis[2-(diphenylphosphino)phenyl]ether) (6,7-Dicyanodipyrido[2,2-*d*:2',3'-*f*]quinoxaline)]BF₄ (Cu^I complex) and a blue emission from 4,4'-bis(2,2'-diphenylvinyl)-1,1'-biphenyl (DPVBi) were demonstrated. The emission spectrum can be fine tuned by effectively controlling the thicknesses of the two emission layers. The optimized device with 18 nm FIrpic and the Cu^I complex codoped 4,4'-N,N'-dicarbazole-biphenyl layer and 12 nm DPVBi layer shows a maximum current efficiency of 8.5 cd A⁻¹, a maximum power efficiency of 5.3 lm W⁻¹ and a maximum luminance of 3290 cd m⁻². Moreover, the device exhibits a CIE coordinate of (0.345, 0.357) at a bias of 8 V and a slight colour variation with increased voltage from 6 to 16 V.

1. Introduction

White organic light-emitting diodes (WOLEDs) attract particular attention due to their potential applications as full colour displays and backlights, as well as next generation lighting sources [1–8]. According to whether fluorescent or phosphorescent materials are used, WOLEDs can be broadly classified into two types. Compared with fluorescent devices, phosphorescent devices are more efficient because they can harvest both singlet and triplet excitons and hence have the potential of reaching a maximum internal quantum efficiency of 100% [9]. Due to the high efficient electrophosphorescence of iridium [10–14] and platinum [15, 16] complexes, phosphorescent WOLEDs were mainly focused on these two type complexes.

Recently, efficient OLEDs based on inexpensive and non-toxic Cu^I complexes were realized [17–20]. In a previous work [21], we demonstrated white light emission based on

yellow emitting [Cu(bis[2-(diphenylphosphino)phenyl]ether) (6,7-Dicyanodipyrido[2,2-*d*:2',3'-*f*]quinoxaline)]BF₄ (Cu^I complex) in combination with a blue emitter. The efficiency of these devices, however, was still inferior due to the low efficient Cu^I complex and the blue emitter adopted. To increase the efficiency of the WOLEDs, both the efficiency of the Cu^I complex should be improved and an efficient blue emitter should be used. Recently, we demonstrated that the electroluminescent (EL) performance of the Cu^I complex can be significantly improved by codoping bis[(4,6-difluorophenyl)-pyridinato-N,C²](picolinato)Ir(III) (FIrpic) into the emitting layer (EML) [22]. This improvement proved the efficient energy transfer from FIrpic to the Cu^I complex. In this paper, we report an efficient WOLED in which the yellow and blue emissions come from FIrpic sensitized Cu^I complex and 4,4'-bis(2,2'-diphenylvinyl)-1,1'-biphenyl (DPVBi), respectively. The device with 18 nm FIrpic and the Cu^I complex codoped 4,4'-N,N'-dicarbazole-biphenyl (CBP) layer and 12 nm DPVBi layer shows a maximum current efficiency of

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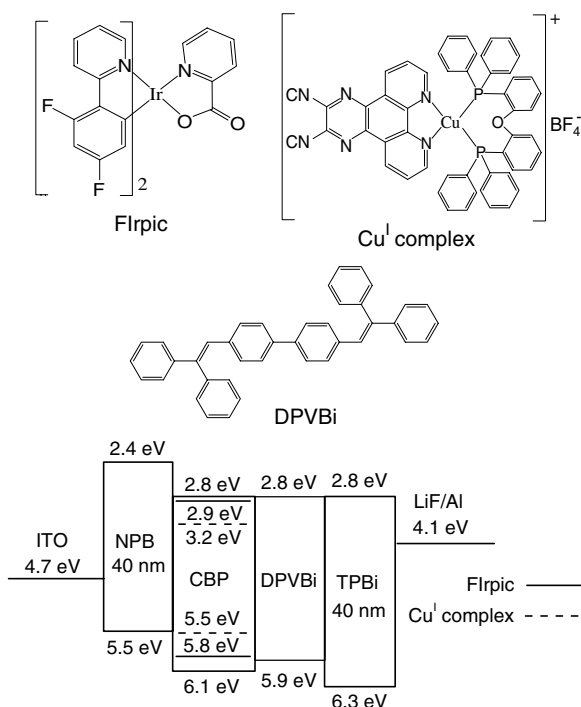


Figure 1. Molecular structures of the materials used and device structure of the WOLEDs.

8.5 cd A⁻¹, a maximum power efficiency of 5.3 lm W⁻¹ and a maximum luminance of 3290 cd m⁻².

2. Experimental

The WOLEDs have a structure of ITO/NPB (40 nm)/CBP:Flrpic:Cu^I complex (8 wt%, 2 wt%, 30-*x* nm)/DPVBi (*x* nm)/TPBi (40 nm)/LiF (0.5 nm)/Al (100 nm), as shown in figure 1. Here, N,N'-diphenyl-N,N'-bis(1-naphthyl)-(1,1'-benzidine)-4,4'-diamine (NPB), 2,2',2''-(1,3,5-benzenetriyl)tris-[1-phenyl-1H-benzimidazole] (TPBi) acted as a hole-transporting layer and an exciton-blocking layer/electron-transporting layer, respectively. The CBP:Flrpic:Cu^I complex and DPVBi layers acted as yellow and blue EMLs, respectively, and the thickness of DPVBi layer was varied from 0 to 15 nm while fixing the whole thickness of the two EMLs at 30 nm. Due to the ambipolar conduction and predominantly electron-transporting properties of CBP and DPVBi, respectively, DPVBi layer was deposited behind the CBP layer. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital levels of DPVBi, TPBi and the Cu^I complex are cited from [3, 12, 19], respectively, while the others from [23]. Organic layers were deposited onto a pre-cleaned indium tin oxide (ITO) glass substrate with a sheet resistance of 10 Ω sq⁻¹ by thermal evaporation in vacuum chamber at 3 × 10⁻⁴ Pa, followed by a LiF buffer layer and an Al cathode in the same vacuum run. Deposition rates and thicknesses of the layers were monitored *in situ* using oscillating quartz monitors. The evaporating rates were kept at 0.5–1 Å s⁻¹ for organic layers and LiF layer, and 10 Å s⁻¹ for Al cathode, respectively. EL spectra and Commission Internationale de l'Eclairage (CIE) coordinates of

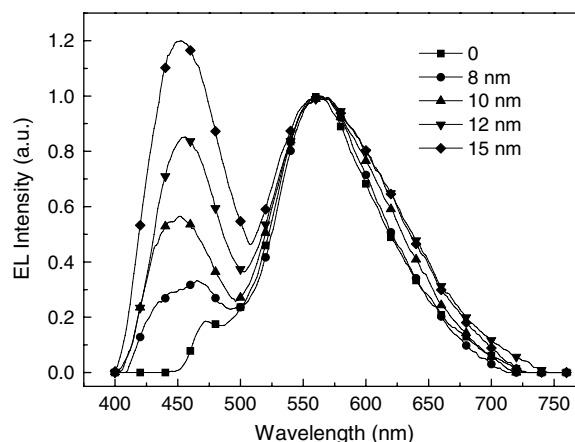


Figure 2. Normalized EL spectra of the devices with different layer thicknesses of DPVBi at a bias of 8 V.

the devices were measured with a Hitachi MPF-4 fluorescence spectrophotometer. The luminance–current–voltage (*L–I–V*) characteristics were measured with a 3645 dc power supply combined with a spot photometer and recorded simultaneously with measurements. All the measurements were carried out at room temperature under ambient conditions.

3. Results and discussions

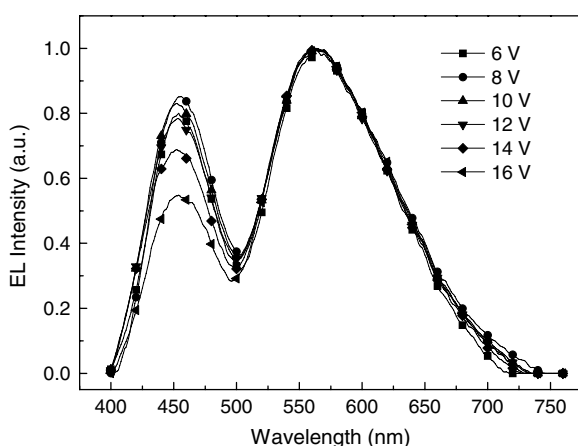
Figure 2 shows the normalized EL spectra of the devices with different layer thicknesses of DPVBi at a bias of 8 V. It can be found that the EL spectra of the double EMLs devices consist of two primary emission bands with peaks at about 455 and 562 nm. The 455 nm emission is attributed to the comprised emission of DPVBi and a low emission of Flrpic with the peak at about 472 nm, while the 562 nm emission is attributed to emission of the Cu^I complex. With the increased layer thickness of DPVBi from 0 to 15 nm, the emission intensity of DPVBi increases relatively to that of the Cu^I complex. Correspondingly, the CIE coordinates shift from (0.427, 0.516) of the device without DPVBi layer to (0.314, 0.314) of the device with a 15 nm DPVBi layer, as shown in table 1. In the double EMLs devices, the relative EL intensity of the two emitters is in proportion to the recombination zone distributed in the two EMLs. The blue shifted CIE coordinates indicate that the proportion of the recombination zone located in DPVBi layer increases with its layer thickness.

Figure 3 shows the normalized EL spectra of the device with 18 nm CBP:Flrpic:Cu^I complex and 12 nm DPVBi layers at different biases. With increased bias from 6 to 8 V, the emission of DPVBi slightly increases compared with that of the Cu^I complex, and then decreases with further increased bias. The CIE coordinates of the device at 6, 8, 10, 12, 14 and 16 V are (0.349, 0.361), (0.345, 0.357), (0.344, 0.356), (0.346, 0.361), (0.354, 0.373) and (0.372, 0.399), respectively. Although the EL spectrum has a slight change in the applied bias, all the CIE coordinates are in the white region. Such a stable emission colour is important for applications of the WOLEDs.

It is well known that the excitons formation probability is in proportion to the product of the densities of holes and

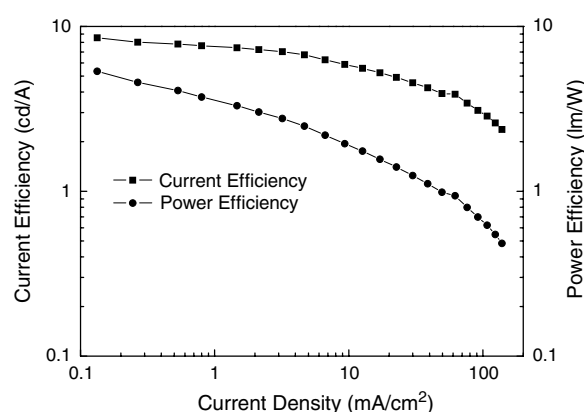
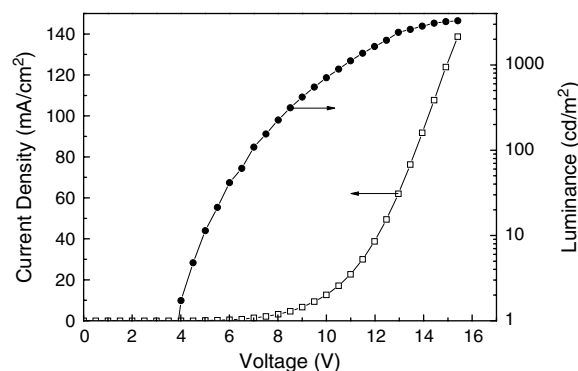
Table 1. EL performance of the devices with different layer thicknesses of DPVBi.

Thickness of yellow EML (nm)	Thickness of blue EML (nm)	Maximum η_c (cd A^{-1})	Maximum η_p (lm W^{-1})	Maximum luminance (cd m^{-2})	CIE coordinate at 8 V
30	0	26.6	16.7	3880	(0.427, 0.516)
22	8	10.1	5.7	3059	(0.368, 0.451)
20	10	9.7	5.5	3720	(0.366, 0.393)
18	12	8.5	5.3	3290	(0.345, 0.357)
15	15	7.2	4.1	3400	(0.314, 0.314)

**Figure 3.** Normalized EL spectra of the device with 18 nm CBP:Flrpic:Cu^I complex and 12 nm DPVBi layers at different biases.

electrons in the recombination zone [2]. Due to the high densities of holes in CBP and electrons in DPVBi layers, the excitons formation probability in CBP and DPVBi layers are dominantly determined by the densities of electrons and holes, respectively. As shown in figure 1, the higher HOMO levels of the Cu^I complex (5.5 eV) and Flrpic (5.8 eV) compared with that of CBP (6.1 eV) indicate that holes can be trapped in their HOMO levels when transportation is in the CBP layer. At low drive voltage, most holes would be trapped in the molecular sites of the Cu^I complex and Flrpic, which leads to low hole density in DPVBi layer. With increased drive bias, the traps will be filled up by holes, and then the hole density in DPVBi will dramatically increase. Although the density of electrons in CBP layer simultaneously increases with drive voltage, the increased DPVBi emission compared with the Cu^I complex at low drive voltage indicates that the increase in hole density in DPVBi layer is faster than that of the electron density in the CBP layer, which may be because the mobility of holes is much higher than that of electrons. In contrast, the trap effect can be ignored due to the large number of holes injected at high drive voltage, a slight shift of the recombination zone towards the CBP layer was observed, leading to increased emission intensity of the Cu^I complex compared with that of DPVBi.

Table 1 lists the EL performance of the devices with different layer thicknesses of DPVBi. With the increased layer thickness of DPVBi from 0 to 15 nm, the current efficiency and power efficiency of the devices decrease from 26.6 cd A^{-1} and 16.7 lm W^{-1} to 7.2 cd A^{-1} and 4.1 lm W^{-1} , respectively. Among the two emitters, the Cu^I complex is

**Figure 4.** Current efficiency and power efficiency versus current density of the device with 18 nm CBP:Flrpic:Cu^I complex and 12 nm DPVBi layers.**Figure 5.** Current density–voltage–luminance characteristic of the device with 18 nm CBP:Flrpic:Cu^I complex and 12 nm DPVBi layers.

more efficient than DPVBi, the decreased proportion of the Cu^I complex with increasing layer thickness of DPVBi leads to increased emission of DPVBi and decreased current and power efficiencies of the devices. The optimized device with 18 nm CBP:Flrpic:Cu^I complex and 12 nm DPVBi layers shows a maximum current efficiency of 8.5 cd A^{-1} and a maximum power efficiency of 5.3 lm W^{-1} at current density of 0.13 mA cm^{-2} , as shown in figure 4. The device has a turn on voltage of 4 V and a maximum luminance of 3290 cd m^{-2} at 15.5 V, as shown in figure 5. The performance of this WOLED is significant improved compared with the previous report [21], and the improvement is ascribed to the increased efficiency of the Cu^I complex and the high efficient blue emitter DPVBi used in this paper.

4. Summary

In summary, high efficient WOLEDs in which the yellow and blue emissions come from FIrpic sensitized Cu^I complex and DPVBi, respectively, were demonstrated. The optimized device exhibits a maximum current efficiency of 8.5 cd A⁻¹, a maximum power efficiency of 5.3 lm W⁻¹ and a maximum luminance of 3290 cd m⁻². This performance is significantly improved in comparison with our previous report. Moreover, the stable emission colour with drive voltage is more important for white light applications.

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