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Efficient white organic light-emitting devices based on blue, orange, red phosphorescent dyes

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Abstract

We demonstrate efficient white organic light-emitting devices (WOLEDs) based on an orange phosphorescent iridium complex *bis*(2-(2-fluorphenyl)-1,3-benzothiozolato-N, $C^{2'}$)iridium(acetylacetonate) in combination with blue phosphorescent dye bis[(4, 6-difluorophenyl)-pyridinato-N,C^2)](picolinato) Ir(III) and red phosphorescent dye bis[1-(phenyl)isoquinoline] iridium (III) acetylanetonate. By introducing a thin layer of 4, 7-diphenyl-1,10-phenanthroline between blue and red emission layers, the diffusion of excitons is confined and white light can be obtained. WOLEDs with the interlayer all have a higher colour rendering index (>82) than the device without it (76). One device has the maximum current efficiency of $17.6\,\mathrm{cd\,A^{-1}}$ and a maximum luminance of $39\,050\,\mathrm{cd\,m^{-2}}$. The power efficiency is $8.7\,\mathrm{lm\,W^{-1}}$ at $100\,\mathrm{cd\,m^{-2}}$. Furthermore, the device has good colour stability and the CIE coordinates just change from (0.394, 0.425) to (0.390, 0.426) with the luminance increasing from 630 to $4200\,\mathrm{cd\,m^{-2}}$.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

White organic light-emitting devices (WOLEDs) have been gaining interest owing to their potential use in full-colour flat-panel displays or as solid-state lighting sources [1] and backlights for liquid crystal displays. Several approaches have been used to generate WOLEDs [2–14]. Among these approaches, WOLEDs employing phosphorescent materials are most effective because the phosphorescent materials can harvest both singlet and triplet excitons leading to the potential for achieving 100% internal emission efficiency [15, 16]. WOLEDs with multiple emission layers in which each layer emits a different colour light to generate white-light emission are usually reported. Compared with other architectures, WOLEDs with multiple emission layers can obtain white light just by varying the doped layer thickness, adjusting dopant concentrations and introducing charge blocking layers [1]. Thus WOLEDs with multiple all-phosphor doped emission layers can easily generate efficient white-light emission. Because wide band-gap host materials for blue phosphorescent dyes are rare, so in all-phosphor WOLEDs, blue dye and other colour dyes are often separately doped into the same host 4, 4′-N, N'-dicarbazole-biphenyl (CBP) because of its good ambipolar conductivity [17, 18]. But energy transfer from the blue emission layer to other colour emission layers can quench the blue light [17]. In order to obtain a balanced output light, the blue emission layer needs an increase in thickness which increases the turn-on voltage and reduces the power efficiency. In fact, the diffusion of excitons can be confined by introducing an interlayer between blue and other colour emission layers [4, 18].

In this paper, we demonstrate WOLEDs based on an iridium complex *bis*(2-(2-fluorphenyl)-1, 3-benzothiozolato-N, $C^{2'}$)iridium(acetylacetonate) [$(\mathrm{F\text{-}BT})_2\mathrm{Ir(acac)}$] [19]. By introducing a thin 4, 7-diphenyl-1,10-phenanthroline (Bphen) layer between blue and red phosphorescent emission layers, the diffusion of charge and triplets excitons is well confined. Moreover, the efficiency and the colour rendering index of the devices with the interlayer of Bphen are both higher than those of the device without it. One device has good EL performance and good colour stability, especially when the luminance varies from 630 to 4200 cd m^{-2}.

2. Experiment

Indium tin oxide (ITO) on glass substrates with a sheet resistance of 10 Ω sq^{-1} was used as a transparent anode. The ITO-coated glass substrates were pre-cleaned by scrubbing using absorbent cotton to degrease, soaking in ultrasonic acetone, ethanol and deionized water in sequence and baking for 20 min and were then exposed to an oxygen plasma followed by UV ozone for 5 min before loading into a high vacuum thermal system. $4, 4', 4''$-tris(3-methylphenylphenylamino)-triphenylamine (m-MTDATA) and N, N'-bis-(1-naphthyl)-N, N'-diphenyl-1, $1'$-biphenyl-4, $4'$-diamine (NPB) were used as a hole-injection layer and a hole-transportation layer. bis[(4, 6-difluorophenyl)-pyridinato-N,C^2)](picolinato) Ir(III) (FIrpic), bis[1-(phenyl)isoquinoline] iridium (III) acetylanetonate [$Ir(piq)_2(acac)$] and $(F\text{-}BT)_2Ir(acac)$ were doped into the same host, CBP, as blue, red and orange emission layers, respectively. Bphen acted as the hole-blocking and electron-transporting layer. Finally, 0.8 nm LiF covered by Al was used as the cathode which was defined by a shadow mask with a 2 × 2 mm^2 opening. A thin Bphen layer was introduced between the blue and red emission layers to confine the diffusion of charge and excitons. All organic layers were grown in succession by high vacuum (10^{-6} Torr) thermal evaporation at a rate of 0.1–0.2 nm s^{-1}, using resistively heated quartz boats. The layer thickness and the deposition rate of materials were monitored *in situ* by an oscillating quartz thickness monitor. Electroluminescence (EL) spectra and Commission International de L'Eclairage (CIE) coordinates of the devices were measured by using a PR650 spectroscan spectrometer. The luminance–voltage and current–voltage characteristics were measured simultaneously with the measurement of the EL spectra by combining the spectrometer with a programmable Keithley 2400 voltage–current source. All measurements were carried out at room temperature under ambient conditions.

3. Results and discussion

The device structure, the electronic level diagrams and the molecular structure of the organic materials are shown in figure 1.

To illuminate the excitons confinement of the thin Bphen layer, we fabricated four devices first. The configurations of the devices are as follows:

ITO/m-MTDATA (30 nm)/NPB (20 nm)/6 wt% FIrpic : CBP (10 nm)/Bphen (X nm)/8 wt% $Ir(prq)_2(acac)$: CBP (4 nm)/ 8 wt% $(F\text{-}BT)_2Ir(acac)$: CBP (8 nm)/Bphen (30 nm)/LiF (0.8 nm)/Al. $X = 0$ nm, 2 nm, 3 nm, 4 nm for devices A, B, C and D, respectively.

Figure 2 shows the normalized EL spectra at 548 nm of devices A–D at 10 V. The spectra contain three primary peaks at 468, 548 and 616 nm ascribed to light emission from FIrpic, $(F\text{-}BT)_2Ir(acac)$ and $Ir(prq)_2(acac)$, which can be seen from the inset of the individual EL spectra of the conventional homochromous devices based on FIrpic, $(F\text{-}BT)_2Ir(acac)$ and $Ir(piq)_2(acac)$. As we can see, the intensity of FIrpic emission is much weaker relative to the $Ir(prq)_2(acac)$ emission for device A. The following may be the reason. In the system of FIrpic : CBP, the triplet energy of (2.56 ± 0.1) eV for CBP is lower than that of (2.65 ± 0.1) eV for FIrpic [20], suggesting that triplet excitons are primarily on the host [21]. The triplet excitons have longer diffusion lengths than the singlets because of their longer lifetimes, so the triplets generated in the blue phosphor-doped layer will easily diffuse out of the region before being captured by FIrpic and are then captured by the adjacent red dye, which leads to the quenched blue emission and the unbalanced output colour. When a thin Bphen layer is introduced in devices B–D, FIrpic emission greatly increases relative to the red emission. As seen from figure 1, the HOMO energy of Bphen at 6.4 eV is 0.4 eV above that of CBP at 6.0 eV, which acts as an effective barrier for holes to cross. The flow of holes to the adjacent layer is blocked by the thin Bphen layer, so more holes will reside in the blue emission layer and more excitons will be generated. Furthermore, Dexter energy transfer from the triplets of FIrpic to the lower energy triplets of $Ir(prq)_2(acac)$ is also prevented because it can only occur within 1–2 nm [22]. The effects lead to the FIrpic emission increasing relative to the red and the orange emission. The relative intensity of FIrpic emission continues to increase as the thickness of the interlayer increases from 2 to 4 nm. This may be because fewer holes and triplets can transport through the thin Bphen layer as its thickness increases. Note that the red emission decreases relative to the orange emission as X varies from 0 to 4 nm due to the energy transfer from FIrpic getting more difficult with increasing X.

Performances of devices A–D are summarized in table 1. Devices B–D all have a higher efficiency than device A, which may be attributed to eliminating the energy transfer between blue and red emission layers. Moreover, the colour rendering index of devices B–D is also higher than that of device A due to the increasing emission of blue at 468 nm. Devices B–D all emit white light.

$(F\text{-}BT)_2Ir(acac)$ is a highly efficient orange phosphorescent dye and more efficient white-light emission can be obtained by adjusting the thickness of the orange emission layer. Here we fix the thickness of the interlayer of Bphen at 3 nm and adjust the thickness of the $Ir(prq)_2(acac)$ doped CBP layer. WOLEDs have the structure of ITO/m-MTDATA (30 nm)/NPB (20 nm)/6 wt% FIrpic : CBP (10 nm)/Bphen (3 nm)/8 wt% $Ir(prq)_2(acac)$: CBP (4 nm)/8 wt% $(F\text{-}BT)_2Ir(acac)$: CBP (Y nm)/Bphen (30 nm)/ LiF (0.8 nm)/Al. $Y = 6$ nm, 8 nm, 10 nm, 12 nm for devices E, F, G and H, respectively. Figure 3 shows the normalized EL spectra of devices E–H at 10 V. The orange emission from $(F\text{-}BT)_2Ir(acac)$ increases relative to blue emission because

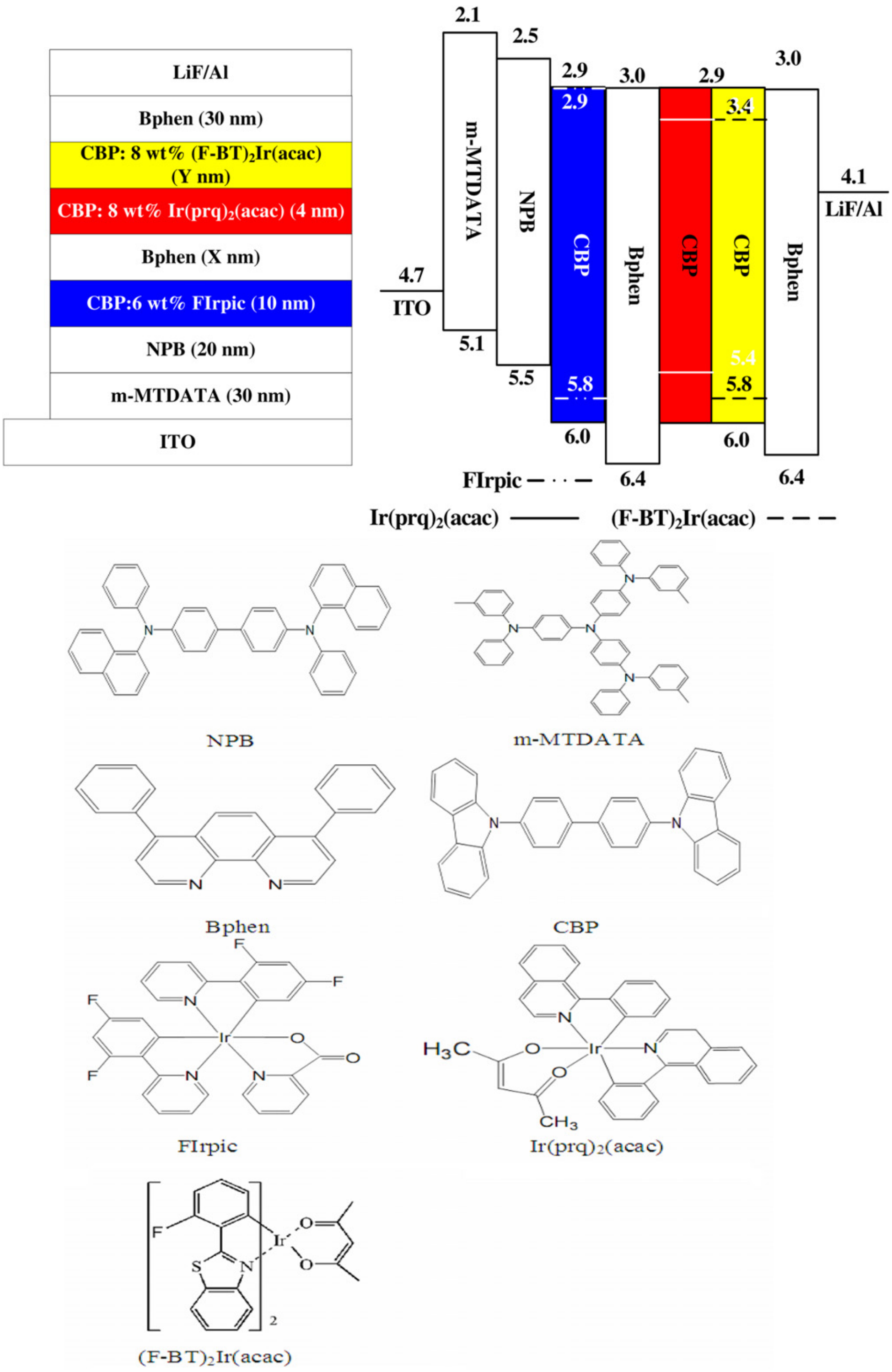

Figure 1. The device structure, the electronic level diagrams and the molecular structure of the organic materials.

more excitons will be recombined in the (F-BT)$_2$Ir(acac) : CBP layer with increasing thickness of it. The intensity of red emission also increases relative to that of blue emission because more energy will transfer from the orange emission layer to the red emission layer as Y changes from 6 to 12 nm. The performances of devices E–F are summarized in table 2. The current efficiency increases from 11.5 to 17.6 cd A^{-1} as the thickness of the orange emission layer increases from 6 to 12 nm, as well as the maximum luminance increases from 32 980 to 39 050 cd m^{-2}. The increasing current efficiency and luminance can be attributed to the increasing emission of (F-BT)$_2$Ir(acac) which has a higher efficiency and luminance than the blue and red dyes. As can be seen in table 2, devices E–H all emit white light at warm white CIE coordinates and have a colour rendering index higher than 84.

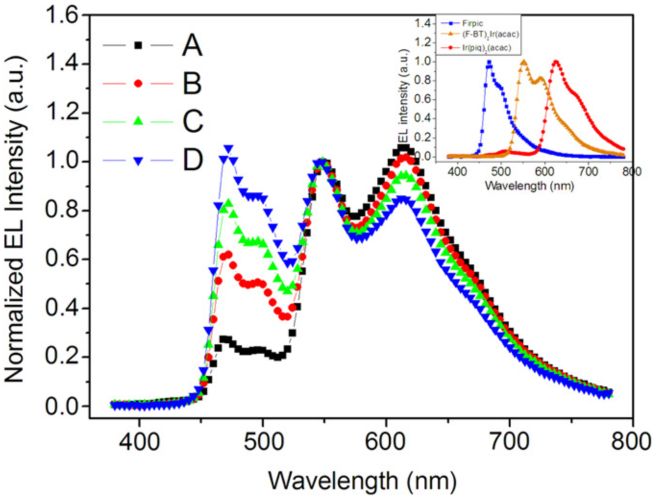

Figure 2. Normalized EL spectra at 548 nm of devices A–D at 10 V. The inset shows the individual EL spectra of the conventional homochromous devices based on FIrpic, $(F\text{-}BT)_2Ir(acac)$ and $Ir(piq)_2(acac)$.

Table 1. Performances of devices A–D.

Devices (L_{Bphen})	Maximum efficiency (cd A^{-1})	Maximum luminance (cd m^{-2})	CIE coordinates at 1000 cd m^{-2}	The colour rendering index
A (0 nm)	11.7	26 450	(0.478, 0.427)	76
B (2 nm)	13.8	30 220	(0.409, 0.410)	84
C (3 nm)	14.4	35 040	(0.382, 0.408)	88
D (4 nm)	14.7	27 810	(0.341, 0.391)	83

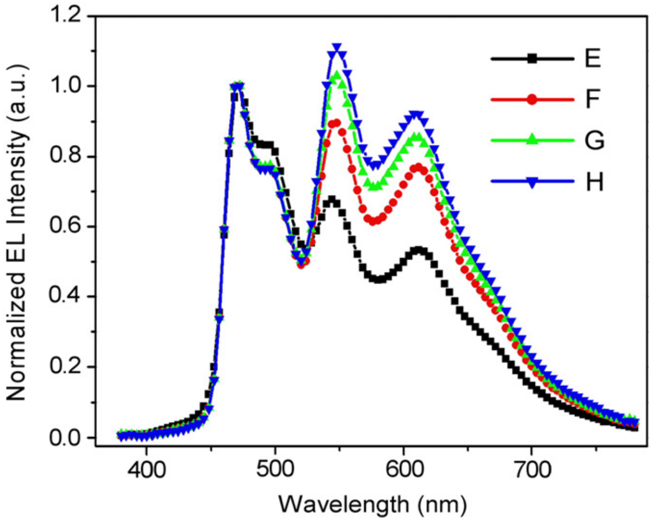

Figure 3. Normalized EL spectra of the devices E–H at 10 V.

Table 2. Performances of devices E–H.

Devices (L_{CBP})	Maximum efficiency (cd A^{-1})	Maximum luminance (cd m^{-2})	CIE coordinates at 1000 cd m^{-2}	The colour rendering index
E (6 nm)	11.5	32 980	(0.388, 0.406)	87
F (8 nm)	13.9	36 886	(0.383, 0.409)	85
G (10 nm)	15.4	38 600	(0.392, 0.422)	86
H (12 nm)	17.6	39 050	(0.392, 0.420)	84

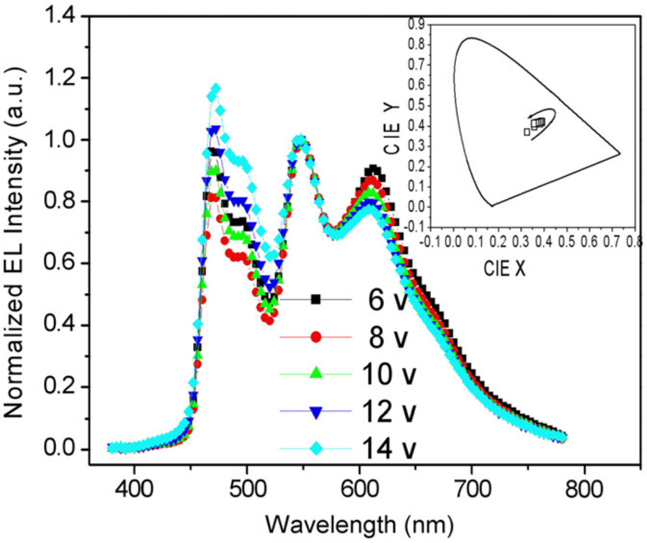

Figure 4. Normalized EL spectra of device H at various voltages. The inset shows the CIE coordinates of device H at different luminances. The arrow indicates increasing luminance from 6 to 39 050 cd m^{-2}.

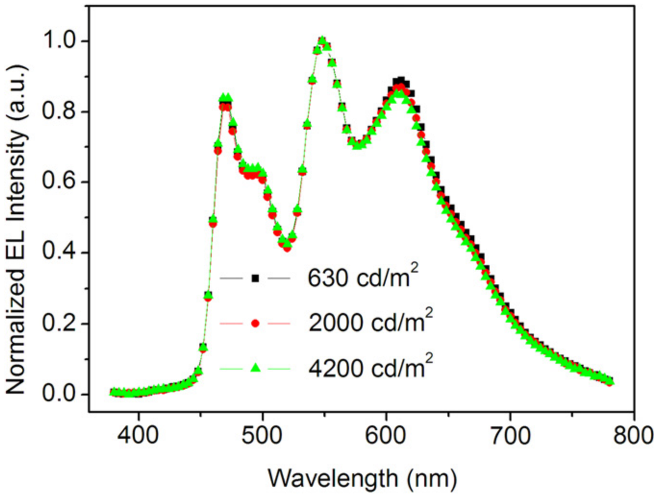

Figure 5. Normalized EL spectra of device H at different luminances.

Figure 4 shows normalized EL spectra of device H at different applied voltages. The spectra show only a little change when the voltage ranges from 6 to 14 V. The inset of figure 4 shows the CIE coordinates of device H at the luminance changing from 6 to 39 050 cd m^{-2}. We can see that the CIE coordinates are fairly stable over a large range of luminances. As is shown in figure 5, our device shows good colour stability of the emission, especially when the luminance varies from 630 to 4200 cd m^{-2}, corresponding to the CIE coordinates changing from (0.394, 0.425) to (0.390, 0.426). Figure 6 shows the power efficiency–luminance–voltage characteristics of device H. Device H obtains a maximum power efficiency of 8.9 lm W^{-1} and a maximum luminance of 39 050 cd m^{-2}. When luminance is

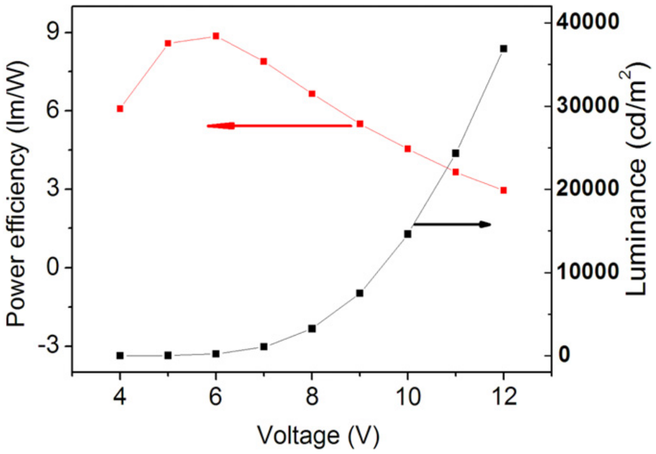

Figure 6. Power efficiency–luminance–voltage characteristics of the device H.

100 cd m^{-2}, 1000 cd m^{-2}, 10000 cd m^{-2}, the power efficiency is 8.7 lm W^{-1}, 8.0 lm W^{-1} and 5.2 lm W^{-1}, respectively.

4. Conclusion

We have demonstrated efficient WOLEDs with triple phosphor-doped emissive layers. Balanced emission colour can be obtained by introducing an interlayer between the blue and red emission layers. One device has the maximum current efficiency of 17.6 cd A^{-1} and the maximum luminance of 39 050 cd m^{-2}. The power efficiency is 8.7 lm W^{-1} at the luminance of 100 cd m^{-2}.

Acknowledgments

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