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## Color Stable White Organic Light-Emitting Diodes Having High Color Rendering Index Utilized by Simple-Hybrid Structures

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**ABSTRACT**

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This article reports the highly color stable white organic light-emitting diodes (WOLEDs) having a high color rendering index (CRI) utilized by simple-hybrid structures involving emitting layers (EMLs) of phosphorescent yellow polymer and a thermally deposited fluorescent blue. To derive the yellow EML, a highly soluble red dopant is developed and used with a highly soluble green co-dopant. Using a simple-hybrid structure, the emission tuned WOLEDs are successfully demonstrated with a high CRI of 93 and a highly color stable CIE 1931 coordinate over the full driving voltages from (0.33±0.00, 0.35±0.01) to (0.42±0.01, 0.44±0.01) depending on the blue EML host.

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**Keywords**

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## Color Stable White Organic Light-Emitting Diodes Having High Color Rendering Index Utilized by Simple-Hybrid Structures

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This article reports the highly color stable white organic light-emitting diodes (WOLEDs) having a high color rendering index (CRI) utilized by simple-hybrid structures involving emitting layers (EMLs) of phosphorescent yellow polymer and a thermally deposited fluorescent blue. To derive the yellow EML, a highly soluble red dopant is developed and used with a highly soluble green co-dopant. Using a simple-hybrid structure, the emission tuned WOLEDs are successfully demonstrated with a high CRI of 93 and a highly color stable CIE 1931 coordinate over the full driving voltages from (0.33 ± 0.00, 0.35 ± 0.01) to (0.42 ± 0.01, 0.44 ± 0.01) depending on the blue EML host.

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Phosphorescent organic light-emitting diodes (OLEDs) are being widely investigated because of their potential ability to realize 100% internal quantum efficiencies for developing the next generation of displays and light sources.<sup>1-3</sup> Following these efforts, and on the basis of considerable interest in developing white light-emitting diodes (WOLEDs),<sup>4-6</sup> one advance of the highly efficient WOLEDs has been reported,<sup>7</sup> but this achievement in realizing the initial WOLEDs still requires further improvement in many aspects; these include improving the color rendering index (CRI) for the full color gamut, simplifying the device structure, and improving the color stabilities sufficient for use as the next generation of light sources. However, the emission tuning of WOLEDs requires the control of emissive dopant concentrations, and this becomes an obstacle for the thermally deposited WOLEDs, which require greater time for the doping procedure than the soluble process-based polymer OLEDs.

The relatively low solubility of the phosphorescent dye dopant has been a major obstacle in using a soluble-based fabrication process for OLEDs.<sup>8,9</sup> To address this issue, there has been much interest in developing highly soluble organic materials, such as iridium(III) *fac*-tris[2-(4-octyl-phenyl)],<sup>8</sup> tris(2-(4-tolyl)phenylpyridine) iridium,<sup>9</sup> *fac*-tris(2-(3-*p*-xylyl)phenyl)pyridine iridium(III),<sup>10</sup> iridium(III) tris(9-(6-phenyl-pyridine-3-ylmethyl)-9*H*-carbazole) [Ir(Czppy)<sub>3</sub>],<sup>11</sup> iridium(III) tris(2-(2'-benzo[*b*]-thienyl)-5-(4'-triphenylsilylphenyl)pyridinato-*N*,C3'),<sup>12</sup> and bis(2-phenylpyridinato-*N*,C2')iridium(2-phenylpyridine-acetylacetonate).<sup>13</sup> These materials show improved solubility capabilities that could be sufficient for use in future desired soluble fabrication processes.

This work demonstrates the highly stable color WOLEDs using a simple-hybrid structure fabricated as follows: To tune the emission of the WOLEDs, a highly soluble phosphorescent polymer emitting layer (EML), wherein the dye-doping concentration is comparably easier to control than in the thermally deposited EML, was used as the yellow EML, comprising the green-red visible light emission, and for the blue emission, the fluorescent blue EML was thermally deposited. By controlling the doping concentration of the highly soluble yellow EML and controlling the blue EML host, the emission tuning of WOLEDs was easily carried out. For the phosphorescent polymer yellow EML, highly soluble phosphorescent red emitting dopant iridium(III) bis(1-(4-((9*H*-carbazole-9-yl)methyl)phenyl)isoquinoline)(acetylacetonate) [Ir(Czpiq)<sub>2</sub>(acac)] was developed and used with the highly soluble green emitting Ir(Czppy)<sub>3</sub> co-dopant.

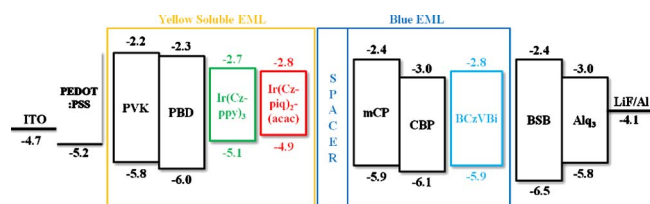
The structure and energy band diagram of the fabricated

WOLEDs are shown in Fig. 1 and Table I, where indium tin oxide (ITO) is used as the transparent anode, 30 nm poly(ethylenedioxythiophene):poly(styrenesulfonate) as the hole injection layer, 30 nm poly(*N*-vinyl-carbazole) (PVK) and 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) mixed host:Ir(Czppy)<sub>3</sub>:Ir(Czpiq)<sub>2</sub>(acac) as the highly soluble phosphorescent yellow EML, 4 nm 4,4'-bis(carbazole-9-yl)biphenyl (CBP) or 1,3-bis(carbazole-9-yl)benzene (mCP) as the spacer for the singlet/triplet exciton management, 16 nm CBP or mCP doped with 4,4'-bis(9-ethyl-3-carbazovinylene)-1,1'-biphenyl (BCzVBi) as the fluorescent blue EML, 10 nm 4,4'-di(triphenylsilyl)-biphenyl (BSB) as the exciton and the hole blocking layer, 30 nm tris(8-hydroxyquinolinato)aluminum as the electron transport layer, lithium fluoride (LiF) as the electron injection layer, and aluminum as the cathode. The WOLEDs were fabricated in two categories, the first class had the CBP blue EML host material (devices C-1.5 and C-2.9) and the second class had the mCP blue EML host (devices M-1.5 and M-2.9). Between the yellow EML and the blue EML, a thin layer (4 nm) of blue EML host was inserted to control the exciton confinement.<sup>7,14</sup> The PVK and PBD were mixed by 7:3 wt %, and the doping ratios of Ir(Czppy)<sub>3</sub>, Ir(Czpiq)<sub>2</sub>(acac), and BCzVBi were 10 wt %, (1.5, 2.9 wt %), and 4.2 mol %, respectively.

The electroluminescence (EL) characteristics of the fabricated WOLEDs are shown in Fig. 2 and 3, and a summary of the device performance of the WOLEDs is given in Table II. Comparing the CBP blue EML host devices with the mCP blue EML host devices, the CBP blue EML host devices show relatively high EL characteristics (especially over the lower driving voltages). These higher EL characteristics of the C-series devices, having the CBP blue EML host material, are explained by the carrier injection barrier EML interfaces and carrier mobility of EML. Because the CBP has a much lower lowest unoccupied p-molecular orbital (-3.0 eV) than BSB hole blocking layer (HBL) (-2.4 eV), the electrons from the BSB HBL can easily go into the CBP blue EML host without any obstacles. Also, the CBP has a much lower highest occupied molecular orbital (-6.1 eV) than that of mCP (-5.9 eV). These injection barrier differences result in the driving voltage differences. Moreover, the CBP has a higher carrier mobility than the mCP; the electron and hole mobilities of the CBP are known to be ~10<sup>-3</sup> cm<sup>2</sup>/V s, whereas those of the mCP are known to be ~10<sup>-4</sup> cm<sup>2</sup>/V s.<sup>15,16</sup>

Although the CBP host devices have shown better EL characteristics (lower driving voltages, higher currents, and power efficiencies), the CRI of mCP shows better values (CRI at 5000 cd/m<sup>2</sup>; device C-2.9 = 84 and device M-2.9 = 91). The increased blue emission in the mCP blue EML host devices comes from the host

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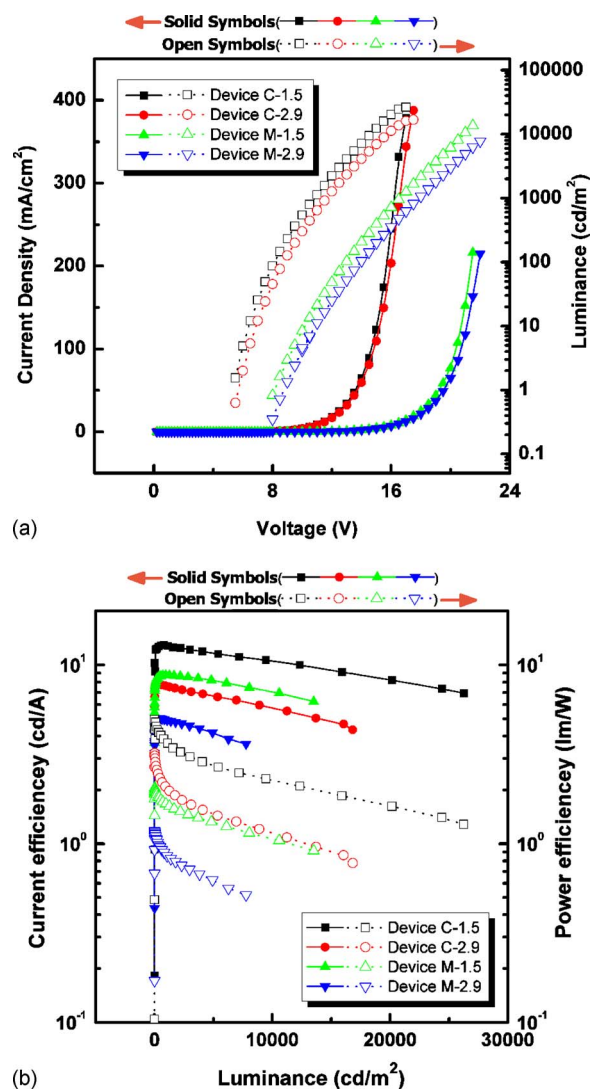


**Figure 1.** (Color online) Energy level diagram and device structure of fabricated WOLEDs.

property of the mCP and the carrier injection status of this device. The mCP is well known for the blue EML host, more so than the CBP, due to its wide bandgap, and so is more suitable for use as the blue host. Due to the carrier injection limitation by the electron injection barrier at the blue EML/HBL, the emission zone shifts to the blue EML in the mCP blue EML host devices and results in a stronger blue emission induced emission tuning; the CIE 1931 color coordinate at 5000 cd/m<sup>2</sup> changes from (0.42, 0.44) of device C-2.9 to (0.36, 0.39) of device M-2.9.

Figure 3 shows the external quantum efficiency (EQE) characteristics as a function of the applied current density and emission spectrum of devices C-2.9 and M-2.9, which show the EQE in the order of CBP > mCP blue EML host, 1.5 > 2.9 wt % Ir(Czpiq)<sub>2</sub>(acac) doping concentration. The unradiative concentration quenching becomes higher following the increased doping concentration and, according to a recent article, the ideal doping concentration of the red Ir complex is ~1%.<sup>17</sup> It is expected that if more enhanced materials are applied (using the deep-red soluble emissive dopants and the highly conductive blue EML host material, which has higher PL efficiencies than mCP), the EL characteristics would be further improved. The WOLEDs are highly color stable so that the CIE 1931 color coordinate shows a small change of ( $\pm 0.02$ ,  $\pm 0.02$ ) in the overall driving current density from 1 to 100 mA/cm<sup>2</sup>. This high color stability comes from the structure of the device as the simple bilayer structure improves the color stability more than the conventional three-layer structure, which has much more dependence of color on current density (driving voltage). Controlling the doping concentration of the red Ir complex, which has a deep trap, enables the modulation of the carrier distribution and results in the tuning of the CRI and the stability of the color. Also, the inserted spacer efficiently suppresses the singlet-triplet exciton quenching at the yellow EML/blue EML interface and blue emission is continuously obtained. In addition, the spacer improves the performances of the device compared to the unused devices, which are not presented here. However, the device performances of the fabricated WOLEDs are comparably low, and because there is no exciton blocking layer between the hole transport layer/yellow EML, exciton quenching would take place. These device performances could be improved further because these results have been derived without using any additional device performance improving techniques, such as ITO surface treatment, p-i-n type carrier doping, out-coupling enhancement systems, employing exciton blocking layer, etc.

Optimized devices for a high CRI have been fabricated; for the CBP blue EML host (device C-2.8), the 2.8% red Ir(III) complex was doped into highly soluble yellow EML, whereas that of the mCP blue EML host device (device M-3.2) was 3.2%. (The current density–voltage–luminance (*J-V-L*) characteristics are similar to

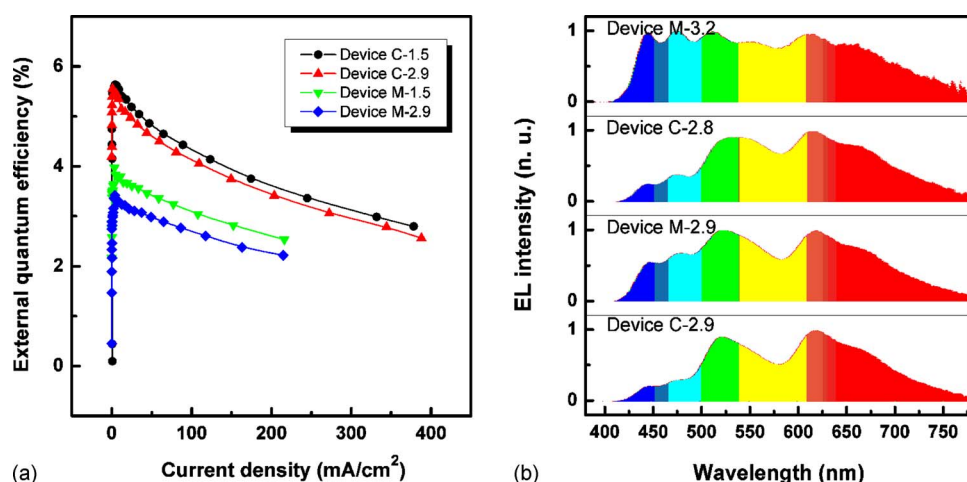


**Figure 2.** (Color online) (a) The current density and luminance curve as a function of applied voltage. (b) The current efficiency and power efficiency curve as a function of luminance.

those of the previous devices, whereas device C-2.8 shows slightly higher characteristics than device C-2.9, and device M-3.2 shows slightly lower characteristics than device M-2.9, which is as expected by the red Ir complex doping concentration, as this reduces the performance of the device at higher than 1%. Therefore, in this article, the *J-V-L* characteristics of the optimized devices are excluded for easier understanding.) Figure 4 shows the CIE 1931 color coordinates of optimized WOLEDs as a function of applied current density. Device C-2.8 shows a high CRI of 93 with CIE 1931 (0.42, 0.44) and a highly color stable property of  $\Delta = \pm (0.01, 0.01)$ , and device M-3.2 shows a high CRI of 93 with CIE 1931 (0.33, 0.35) and a highly stable color property of  $\Delta = \pm (0.00, 0.01)$ . Although the doping process in the polymer OLEDs has some drawbacks of

**Table I.** Device configurations.

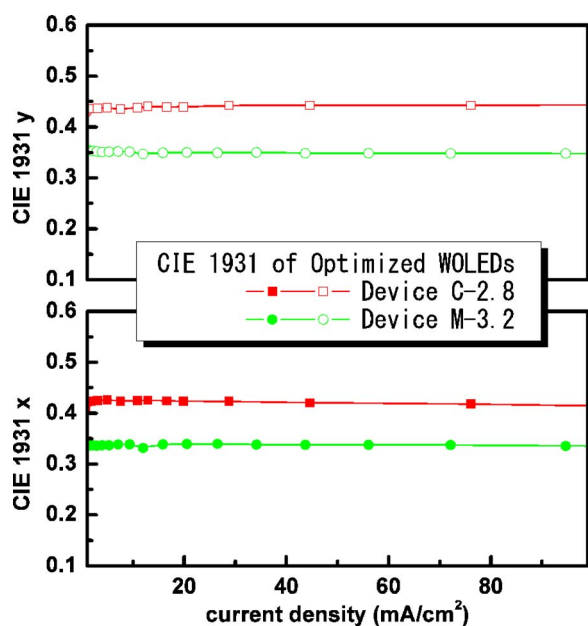
Device configuration	Yellow soluble EML	Blue EML
Device C-1.5	PVK:PBD:Ir(Czppy) <sub>3</sub> 10 wt %: Ir(Czpiq) <sub>2</sub> (acac) 1.5 wt %	CBP:BCzVBi
Device C-2.9	PVK:PBD:Ir(Czppy) <sub>3</sub> 10 wt %: Ir(Czpiq) <sub>2</sub> (acac) 2.9 wt %	CBP:BCzVBi
Device M-1.5	PVK:PBD:Ir(Czppy) <sub>3</sub> 10 wt %: Ir(Czpiq) <sub>2</sub> (acac) 1.5 wt %	mCP:BCzVBi
Device M-2.9	PVK:PBD:Ir(Czppy) <sub>3</sub> 10 wt %: Ir(Czpiq) <sub>2</sub> (acac) 2.9 wt %	mCP:BCzVBi



**Figure 3.** (Color online) (a) The EQE as a function of applied current density. (b) EL spectra of devices C-2.9 and M-2.9 and the optimized devices of C-2.8 and M-3.2. All of the EL spectra were taken at the luminance level of 1000 cd/m<sup>2</sup>.

**Table II.** Summary of the device performances of the fabricated WOLEDs.

Device configuration	At peak EQE		At 20 mA/cm <sup>2</sup>		At 5000 cd/m <sup>2</sup>			
	EQE (%)	<i>J</i> (mA/cm <sup>2</sup> )	EQE (%)	PE (lm/W)	EQE (%)	PE (lm/W)	CRI	CIE 1931
Device C-1.5	5.63	4.2	5.25	3.17	4.85	2.68	66	0.38,0.52
Device C-2.9	5.58	1.0	5.07	1.94	2.78	0.86	84	0.42,0.44
Device M-1.5	3.97	3.3	3.65	1.55	3.35	1.32	64	0.35,0.49
Device M-2.9	3.42	3.9	3.16	0.86	2.22	0.52	91	0.36,0.39



**Figure 4.** (Color online) The CIE 1931 color coordinates of optimized WOLEDs as a function of applied current density.

solubility, it has the strong point of being easier to specify the doping concentration than in thermally deposited small molecular dye-doping systems. This characteristic becomes much stronger in highly soluble dye-doping systems. This process gives good characteristics of a highly soluble dye-doping system and derives a much easier emission tuning of the OLEDs, which has contributed to the development of the highly color stable, high CRI WOLEDs.

In summary, the highly color stable, high CRI WOLEDs obtained by utilizing a simple-hybrid structure of the highly soluble

phosphorescent yellow polymer EML/thermally deposited fluorescent blue EML device structure have been successfully demonstrated. When using the CBP for blue EML hosts, the CIE 1931 color coordinate of (0.42, 0.44) with a high CRI of 93 has been demonstrated, and the device with mCP for the blue EML host shows the CIE 1931 color coordinate of (0.33, 0.35) with a high CRI of 93. The phosphorescent yellow and fluorescent blue double-EML structure is a simple and effective method for developing WOLEDs. Also, having a highly soluble phosphorescent yellow layer would improve the WOLED development process.

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