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A white organic light-emitting diode with ultra-high color rendering index, high efficiency, and extremely low efficiency roll-off

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Ultra-high color rendering index (>90) is considered to be crucial for lighting in museum, studio, art gallery, medical, and wide-color-gamut display. However, all relevant works reported to exhibit ultra-high color rendering index suffer from a low efficiency and serious efficiency roll-off. In order to achieve ultra-high color rendering index and maintain high device efficiency at the same time, we incorporate four organic dyes in an elaborate device structure. The resulting white organic light-emitting diode exhibits rather high color rendering index up to 94 over a wide brightness range and yields electroluminescence efficiencies up to 14.2%, 26.0 cd/A, and 21.9 lm/W at the brightness of 1000 cd m^{-2} , which are the highest values reported to date for such ultra-high color rendering index white organic light-emitting diodes. In addition, the current issue on efficiency roll-off is well resolved in the resulting device. The critical current density j_c is as high as 203 mA cm^{-2} . © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4890217>]

White organic light-emitting diodes (WOLEDs) have been recognized as a promising alternative display and solid-state lighting technology due to the advantages of low-voltage operation, fast response, wide-viewing angle, easy fabrication of potentially large-area, good color rendering, and mechanical flexibility.¹ Usually, a WOLED constructed by three primary colors (red, green, and blue) is used to present acceptable chromatic behavior, i.e., the performance in color rendering index (CRI) and correlated color temperature (CCT). Such an approach enables CRI around 80–90, which is acceptable in common applications including outdoor lighting, general indoor lighting, or backlights in liquid crystal display with low color gamut. However, to meet the strict requirement in some areas, such as lighting for museum, studio, art gallery, medical application, and wide-color-gamut display, it is difficult to satisfy the high criteria ($\text{CRI} > 90$) in color performance for such a white light source. To generate white light with high CRI, emission spectrum should be as broad and continuous as possible in the visible-light spectral region.

A lot of endeavors in obtaining ultra-high CRI (>90) have been made. For example, Kalinowski *et al.* reported a WOLED with maximum external quantum efficiency (EQE) of 6.5% and then reduced to 3% at 500 cd m^{-2} with a CRI of 90 based on the utilization of excimer and exciplex emission.² Wu *et al.* demonstrated an efficient three-component phosphorescent WOLED with CRI up to 94. The peak EQE and power efficiency (PE) were 10.3% and 16.4 lm W^{-1} . The efficiencies at the luminance of 100 cd m^{-2} dropped to

8.0% and 7.0 lm W^{-1} .³ Jou *et al.* fabricated a very-high CRI five-spectrum OLED with double white emission layers (EMLs), achieving a CRI of 98 with an efficacy of 8.3 lm/W at 100 cd m^{-2} , or CRI of 96 with 5.2 lm W^{-1} at 1000 cd m^{-2} .⁴ Then, they developed a hybrid WOLED, exhibiting a 93 CRI with a PE of 23.3 lm W^{-1} at 100 cd m^{-2} , or 14.3 lm W^{-1} at 1000 cd m^{-2} .⁵ Unfortunately, all these WOLEDs with ultra-high CRI suffered from low efficiencies and serious efficiency roll-off, presenting a significant barrier to practical use. Therefore, there is still considerable room for improvement in the efficiency for the ultra-high CRI WOLEDs.

In this paper, through employment of four organic dyes in a high-efficiency device structure, we develop a hybrid WOLED with CRI up to 94 and while maintaining electroluminescence (EL) efficiencies as high as 14.2%, 26.0 cd/A, and 21.9 lm/W at the brightness of 1000 cd m^{-2} .

The starting point for constructing the hybrid WOLEDs demonstrated below was the selection of appropriate emitters. The emission spectra of the emitters should compensate each other so that the white spectrum can cover the whole visible-light region. Because the emission stemming from commonly used red emitter iridium(III)bis(2-methyldibenzo-[f,h]quinoxaline)(acetylacetonate) ($\text{Ir}(\text{MDQ})_2(\text{acac})$) is subject to insufficient light output in the longer wavelength region, which would be detrimental to obtaining ultra-high CRI. We employed a deep-red dye, tris[1-phenylisoquinolinato-C2,N]iridium(III) ($\text{Ir}(\text{piq})_3$), serving as a saturated red emitter. For blue and green emitters, we incorporated highly efficient dyes, *N,N'*-di-1-naphthalenyl-*N,N'*-diphenyl-[1,1':4',1'':4'',1''':4'''-quaterphenyl]-4,4'''-diamine (4P-NPD) and

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bis(2-phenylpyridine)iridium acetylacetonate ($\text{Ir}(\text{ppy})_2(\text{acac})$). To prevent the undesirable deep valley present between the green and red peaks, it is thus necessary to adopt a yellow dye, here we use bis(2-phenyl-1,3-benzothiazololato- $\text{N},\text{C}^{2'}$)iridium (acetylacetonate) ($\text{Ir}(\text{bt})_2(\text{acac})$).

The optimized WOLED structure is indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene): poly(4-styrenesulfonate) (PEDOT:PSS) (40 nm)/ N,N' -diphenyl- N,N' -bis(1-naphthylphenyl)-1,1'-biphenyl-4,4'-diamine (NPB) (15 nm)/4,4',4''-tri(N -carbazolyl)triphenylamine (TCTA): 4 wt. % $\text{Ir}(\text{piq})_3$ (3.5 nm)/TCTA: 4 wt. % $\text{Ir}(\text{bt})_2(\text{acac})$ (4 nm)/TCTA: 25 wt. % 1,3,5-tri(m -pyrid-3-yl-phenyl)benzene (TmPyPb): 2 wt. % 4P-NPD (7 nm)/TmPyPb (4 nm)/TmPyPb: 5 wt. % $\text{Ir}(\text{ppy})_2(\text{acac})$ (3 nm)/TmPyPb (15 nm)/TmPyPb: 4 wt. % cesium carbonate (Cs_2CO_3) (35 nm)/ $\text{Cs}_2\text{CO}_3/\text{Al}$. The device was grown on a precleaned ITO substrate with a sheet resistance of $10\ \Omega/\text{sq}$. All these layers were grown by thermal evaporation in a high vacuum system. The current-voltage-luminance characteristics were measured by using a Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a calibrated silicon photodiode. The EL spectra were measured by spectrascan PR650 spectrophotometer.

Figure 1 shows the energy-level diagram of the WOLEDs. Due to the dominant hole and electron transport characteristics of TCTA and TmPyPb, respectively, it can be assumed that the main exciton recombination zone is located across the whole blue EML where the majority of injected holes and electrons meet with each other. Majority of the singlet excitons generated in the blue EML would undergo a direct radiative decay to ensure sufficient blue emission required for white light. Only few singlet excitons at the interface could be transferred via Förster transfer to the yellow EML. Due to the lower triplet energy level of $\text{Ir}(\text{bt})_2(\text{acac})$ (2.19 eV),⁶ majority of the generated triplet excitons that cannot radiate in the blue EML are captured diffusively by the yellow phosphor $\text{Ir}(\text{bt})_2(\text{acac})$ and then sequentially harvested by the red emitters, forming a cascade energy transfer mechanism. In other words, the loss of non-radiative triplet excitons on the fluorescent blue emitter can be completely avoided by efficiently transferring otherwise lost triplet excitons from 4P-NPD to $\text{Ir}(\text{bt})_2(\text{acac})$, which cannot realize in our previous work.⁷ Because majority of triplet excitons generated in the blue EML reside on the

TmPyPb molecules,⁷ the part of triplet excitons diffuse to the cathode side, which are originally wasted in our previous device,⁷ can transfer their triplet energy to the green dopants through pure TmPyPb interlayer, leading to green emission component from $\text{Ir}(\text{ppy})_2(\text{acac})$ in the resulting white EL spectrum. Overall, with such a working mechanism, all generated excitons, i.e., both singlet and triplet excitons can be harvested, realizing nearly 100% exciton harvesting.

From Figure 2, it can be seen that the resulting hybrid WOLED affords a high-quality white light, yielding an ultra-high CRI up to 94 and CIE coordinates close to the warm white point A throughout a wide luminance range. Moreover, high efficiencies and extremely low efficiency roll-off are achieved. The maximum EQE, current efficiency (CE), and PE in the forward direction are 14.3%, 26.1 $\text{cd}\ \text{A}^{-1}$, and 26.2 $\text{lm}\ \text{W}^{-1}$, respectively. At the practical brightness of 1000 $\text{cd}\ \text{m}^{-2}$, they yet remain as high as 14.2%, 26.0 cd/A and 21.9 $\text{lm}\ \text{W}^{-1}$, representing the highest efficiency values ever reported for such ultra-high CRI WOLEDs. The device possesses superior critical current density j_c up to 203 $\text{mA}\ \text{cm}^{-2}$ which outperforms other reported devices with low efficiency roll-off in literature.⁸ However, considering nearly 100% exciton harvesting, the device efficiencies are unsatisfactory compared with our previous work.⁷ That is originated from low photoluminescent quantum yield (PLQY) of $\text{Ir}(\text{piq})_3$.⁹ Thus, if $\text{Ir}(\text{piq})_3$ is replaced by a deep-red dye which allows for a high PLQY, it can be expected that the device efficiencies would be further improved.

An important element in our structure is the thin TmPyPb interlayer inserted between the blue EML and green EML. Because the singlet state of 4P-NPD (2.9 eV) is higher than that of $\text{Ir}(\text{ppy})_2(\text{acac})$ (2.6 eV) and its triplet state (2.4 eV).¹⁰ If there is no TmPyPb interlayer, the singlet excitons formed in the blue EML region are prone to transferred to the green dopant via Förster transfer or diffusion, such that no white emission is possible. Due to the fact that the singlet state of TmPyPb is higher than that of 4P-NPD and short Förster transfer radius (3 nm),¹¹ a 4 nm TmPyPb interlayer can prevent this Förster-type transfer effectively, ensuring sufficient blue emission for white light. According to our previous report, majority of triplet excitons generated in the blue EML reside on the TmPyPb molecules. The insertion of a thin interlayer would not inferior to triplet excitons

FIG. 1. Energy level scheme for materials used in the hybrid WOLED, and exciton (S_0 , S_1 , and T_1) energy diagram of the emitter layers. The gray filled rectangle represents the main exciton generation zone. R, Y, G, B, and Tm represent $\text{Ir}(\text{piq})_3$, $\text{Ir}(\text{bt})_2(\text{acac})$, $\text{Ir}(\text{ppy})_2(\text{acac})$, 4P-NPD, and TmPyPb, respectively.

FIG. 2. (a) Current density–luminance–voltage characteristics of the resulting WOLEDs with luminance measured in forward direction without outcoupling enhancement techniques. The inset shows the components of the EMLs. (b) External quantum efficiency and power efficiency versus luminance characteristics of the optimized device. (c) Normalized EL spectra at different luminance levels.

diffusing all the way through it via a sequence of Dexter energy transfers, and then contributing to the green emission. To further understand the importance of the interlayer, device W without interlayer was fabricated and its spectrum was compared to that of the optimized device. Apart from eliminating of the interlayer, all other parameters of device W were kept unchanged to allow optimum comparability. It is remarkable that the spectrum of device W strongly differs from that of the optimized device, as shown in Figure 3. There is barely any light emission in the blue spectral range for device W. The reason for this phenomenon is that most of the singlet excitons in the blue EML region were quenched by Ir(ppy)₂(acac), such that the EL spectrum is not balanced to reach the warm white light region and the CRIs

FIG. 3. Normalized EL spectra of device W at different luminance levels.

are much lower than 90. However, in the optimized structure, the interlayer prevents the singlet excitons formed in the blue EML to transfer to the lower energy green phosphors efficiently, allowing for a balanced white light over a large operation range of driving voltage. Thus, in our device structure, the thin TmPyPb interlayer plays a key role in the white light emission.

In addition to the realization of high-quality white light and high efficiency, we note here that the current issue on efficiency roll-off is well resolved in our device. The critical current density j_c , where EQE declines by half from its peak,¹² is as high as 203 mA cm⁻². Especially, from the brightness at maximum EQE to brightness of 1000, 5000, and 10 000 cd m⁻², the efficiency roll-off is only 0.7%, 7.7%, and 19.6%, exhibiting extremely low efficiency roll-off. In recent years, the efficiency roll-off behavior gains much attention.¹³ Because it plays a dominant role in quenching the radiative excitons at high luminance, which detrimentally degrades the device performance for practical applications. Triplet–triplet annihilation (TTA) and triplet–polaron quenching (TPQ) have been proposed to be responsible for efficiency roll-off.¹⁴ Fortunately, the two processes can be suppressed in our structure. First, the bipolar property in the blue EML renders a broad exciton recombination zone across the whole blue EML, which yields a lower equilibrium triplet exciton density, thus relieving both TTA and TPQ effects at high luminance. In addition, the higher triplet energy levels of NPB and TmPyPb compared with adjacent triplet emitters, i.e., Ir(piq)₃ and Ir(ppy)₂(acac) which locate at the edges of the EML, create an energetic well-like emissive region, thus well confining the generated excitons within the EML, which contribute to the reduction of efficiency roll-off. Finally, according to various previous reports,¹⁵ it has been well recognized that direct exciton formation following charge trapping on guest sites can reduce triplet and carrier accumulation in the main exciton recombination zone, which is beneficial to the formation of an extended exciton recombination zone, hence decreasing the efficiency roll-off. In order to investigate whether there exists directly charge carrier trapping on the dopants in the EL process, we fabricated a group of hole- and electron-only devices. Figure 4(a), a decreased hole current upon doping of 4 wt. % Ir(piq)₃ into TCTA than that of the non-doped layer can be observed due to the hole trapping function of Ir(piq)₃.

FIG. 4. Current-density-voltage characteristics of (a) an hole-only device: ITO/MoO₃ (10 nm)/NPB (40 nm)/TCTA: *x* wt. % Ir(piq)₃ (*x* = 0, 4) (40 nm)/MoO₃ (10 nm)/Al; (b) an hole-only device: ITO/MoO₃ (10 nm)/NPB (40 nm)/TCTA: 4 wt. % Ir(piq)₃ (3 nm)/TCTA: *x* wt. % Ir(bt)₂(acac) (*x* = 0, 4) (40 nm)/MoO₃ (10 nm)/Al; (c) an electron-only device: Al/LiF (1 nm)/TmPyPb (25 nm)/TCTA: *x* wt. % Ir(piq)₃ (*x* = 0, 4) (40 nm)/TCTA: 4 wt. % Ir(bt)₂(acac) (3 nm)/TmPyPb (25 nm)/LiF (1 nm)/Al; (d) an electron-only device: Al/LiF (1 nm)/TmPyPb (25 nm)/TCTA: *x* wt. % Ir(bt)₂(acac) (*x* = 0, 4) (40 nm)/TCTA: TmPyPb: 4P-NPD = 73 wt. %; 25 wt. %; 2 wt. % (3 nm)/TmPyPb (25 nm)/LiF (1 nm)/Al.

Whereas doping 4 wt. % Ir(bt)₂(acac) into TCTA had almost no influence on hole transport compared to that of the device with an intrinsic 40-nm-thick TCTA layer (see Figure 4(b)). Figures 4(c) and 4(d) show the current density versus voltage curves of the electron-only devices, significant decreases were found in current density in the TCTA layers doped with 4 wt. % Ir(piq)₃ and 4 wt. % Ir(bt)₂(acac) with respect to that of the pure TCTA layer, which indicate that Ir(piq)₃ and Ir(bt)₂(acac) in TCTA served as trapping sites for electrons. As shown in above phenomena strongly proves that direct exciton formation following charge trapping on Ir(piq)₃ and Ir(bt)₂(acac) sites indeed contributes to the red and yellow emissions, which not only decrease the density of triplet excitons in the main exciton recombination zone but also reduce the charge carrier accumulation in that region, resulting in further suppression of TTA and TPQ. All aspects mentioned above are greatly in favor of the improvement in efficiency roll-off.

In conclusion, based on employment of four organic dyes to broaden emission spectrum, we developed a hybrid WOLED with ultra-high CRI up to 94 over a wide brightness range, accompanied by CIE coordinates close to the warm white point A. Moreover, through a delicate design of the device structure, nearly all generated excitons are harnessed for light emission, resulting in a high efficiency and extremely low efficiency roll-off. At the practical brightness of 1000 cd m⁻², EQE, CE, and PE reach as high as 14.2%, 26.0 cd/A, and 21.9 lm W⁻¹, which are the highest values reported to date for such ultra-high CRI WOLEDs. This work paves the way for simultaneous achievement of ultra-high CRI and high efficiency in a WOLED.

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