

Color-Tunable Hybrid White Organic Light-Emitting Diodes with Double Interlayers

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How to cite this paper: Gao, C.H., Xiong, Z.Y., Yu, F.X., Ma, X.J., Zhang, Y., Zeng, X. and Xiong, Z.H. (2017) Color-Tunable Hybrid White Organic Light-Emitting Diodes with Double Interlayers. *Optics and Photonics Journal*, 7, 99-105.
<https://doi.org/10.4236/opj.2017.78B014>

Received: July 3, 2017

Accepted: August 7, 2017

Published: August 10, 2017

Abstract

An efficient color-tunable hybrid white organic light-emitting diode is demonstrated with double interlayers of 2,7-bis(carbazol-9-yl)-9,9-ditoylfluorene/2-(diphenylphosphoryl) spirofluorene (DMFL-CBP/SPPO1) inserted between blue fluorescent and yellow phosphorescent-emitting layers, and exhibits Commission Internationale de l'Eclairage (CIE1931) ranging from warm white (0.4368, 0.4497) to cool white (0.2781, 0.2896) with driving current density from 0.2 to 40 mA/cm². The recombination of singlet and the triplet excitons in blue fluorescent-emitting layer and yellow phosphorescent-emitting layer, respectively, can be modulated by both the thickness of these double interlayers and the applied current densities.

Keywords

Hybrid White Organic Light-Emitting Diode, Color-Tunable, Singlet Exciton, Triplet Exciton, Phosphor

1. Introduction

Since the first practical sandwiched organic light-emitting diode (OLED) was reported by C.W. Tang in 1980s [1], thirty years have been passed with more and more attention paid in this field due to low cost, flexibility, area source, and high response [2] [3] [4]. Hybrid white organic light-emitting diodes (HWOLEDs), which possess the advantage of both the long stability of the blue fluorescent-emitting layer based OLEDs and the high electroluminescent efficiency of yellow phosphorescent-emitting layer based OLEDs, have been considered as a potential light source for lighting and display [5].

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To prevent the singlet energy transfer from the blue fluorescent emitting layer to the complement phosphorescent emitting layer and to blocking the triplet excitons in the complement phosphorescent emitting layer, interlayer with high singlet and triplet energy should be inserted between the two layers. [6] Kinds of structures have been created to get high efficiency and long stability HWOLEDs. In 2006, Sun *et al.* proposed HWOLEDs with the blue fluorophor-single interlayer-complementary phosphor-single interlayer-blue fluorophor (F-SI-P-SI-F) structure, which can harvest both singlet and triplet excitations and exhibit a highly efficiency of 23.8 lm/W at 500 cd/m² [7]. To simplify the device structure, the architectures of HWOLEDs with the complementary phosphor-single interlayer-blue fluorophor (P-SI-F) structure [8] [11], and the complementary phosphor-mixed single interlayer-blue fluorophor (P-MSI-F) structure [14] [15] are developed to manipulate the charges and excitations in the HWOLEDs. Normally, hole transporting material (N,N-dicarbazoyl-3,5-benzene, mCP) [8], electron transporting material (2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline, BCP) [9], and ambipolar transporting material (4,4'-bis(carbazol-9-yl)biphenyl, CBP) [10] [12] [13] are used as the single interlayers. The mixed single interlayer is formed by co-evaporating hole transporting material (or ambipolar transporting material) and electron transporting material to modulate the shape of the color spectra [16] [17]. However, seldom double interlayers are reported in the HWOLEDs.

In this work, double interlayers of 2,7-bis(carbazol-9-yl)-9,9-ditoylfluorene/2-(diphenylphosphoryl) spirofluorene (DMFL-CBP/SPPO1) is inserted between the blue fluorescent emitting layer and yellow phosphorescent emitting layer to manipulate the exciton generating zone. An efficient HWOLED is realized with a large chrominance span from CIE (0.4368, 0.4497) to CIE (0.2781, 0.2896).

2. Experimental Section

After the 110 nm indium-tin-oxide (ITO) film coated glass substrate with a sheet resistance of 15 Ω/□ is treated by UV-ozone for 15 min, layers of molybdenum trioxide (MoO₃), N,N'-di(1-naphthyl)-N,N'-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB), 2-methyl-9,10-bis(naphthalen-2-yl)anthracene: 4,4'-bis(9-ethyl-3-carbazovinyleno)-1,1'-biphenyl (MADN: BCzVBi), DMFL-CBP/SPPO1, 2',2''-(1,3,5-benzinetriyl)-Tris(1-phenyl-1-H-benzimidazole):Iridium(III)bis(4-phenyl thieno[3,2-c]pyridinato-N,C2')acetylacetonate (TPBi: PO-01), bathophenanthroline (Bphen), tris-8-hydroxyquinoline aluminium (Alq), lithium quinolate/aluminum (Liq/Al), which are used as hole injecting layer, hole transporting layer, blue fluorescent emitting layer, double interlayers, yellow phosphorescent emitting layer, hole blocking layer, electron transporting layer, and bilayer cathode, respectively, are subsequently thermally evaporated under a base pressure of 2 × 10⁻⁶ Torr. The device structures of the HWOLEDs consist of ITO/MoO₃(10 nm)/NPB (75 nm) /MADN: BCzVBi 7 wt% (20 nm)/DMFL-CBP (X)/SPPO1 (Y)/TPBi: PO-0.13 wt% (20 nm)/Bphen (10 nm)/Alq (20 nm)/Liq (2

nm)/Al (120 nm), where the sum of “X” and “Y” is 5, and “X” stands for 3 nm, 2.5 nm, 2 nm in Device A, Device B, and Device C, respectively. The schematic diagram and the energy level graph of device structures are shown in **Figure 1(a)** and **Figure 1(b)** [18] [19] [20].

The oscillating quartz thickness monitor was used to measure the deposition rate and thickness of the layers in situ. The Photo Research Spectra Scan PR655 photometer and Keithley 2400 Source Meter were connected by software to measure the current-voltage curves, luminance, the electroluminescent (EL) spectra, CIE coordinates of the HWOLEDs. The active area of each HWOLED is 0.1 cm².

3. Results and Discussion

Figure 2 shows the EL performance of the three HWOLEDs with different double interlayers of DMFL-CBP (3 nm)/SPPO1 (2 nm), DMFL-CBP (2.5 nm)/SPPO1 (2.5 nm), DMFL-CBP (2 nm)/SPPO1 (3 nm) for Devices A, B and C, respectively. In current density-voltage-luminance curves in **Figure 2(a)**, it can be found the thicker the DMFL-CBP, the lower the current density and the larger the luminance at each applied voltage. In **Figure 2(b)** and **Figure 2(c)**, current efficiency and power efficiency increased with the increasing DMFL-CBP thickness at each applied current density, and shows the highest current efficiency and power efficiency of 36.5 cd/A and 30.1 lm/W driving at 0.2 mA/m² in Device A. And Devices B (C) shows maximum current efficiency and power efficiency of 27 cd/A (24.5 cd/A) and 22.7 lm/W (20.1 lm/W), respectively. These are attributed to that the exciton generation interface should be located at the interface of DMFL-CBP/SPPO1 owing to the high lowest unoccupied molecular orbital (LUMO) of DMFL-CBP (−1.29 eV) and the low *highest* occupied molecular orbital (HOMO) of SPPO1 (−6.5 eV) which can block electrons and holes at this interface, respectively.

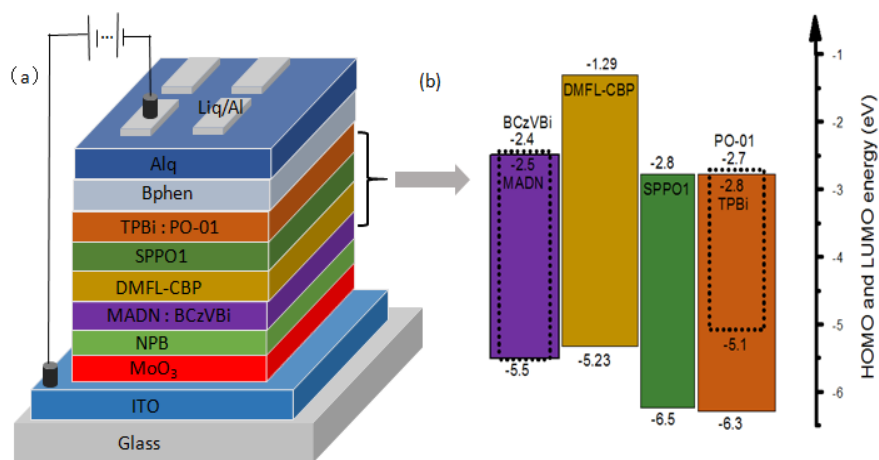


Figure 1. The schematic diagram (a) and energy level graph of the HWOLEDs (Devices A, B and C).

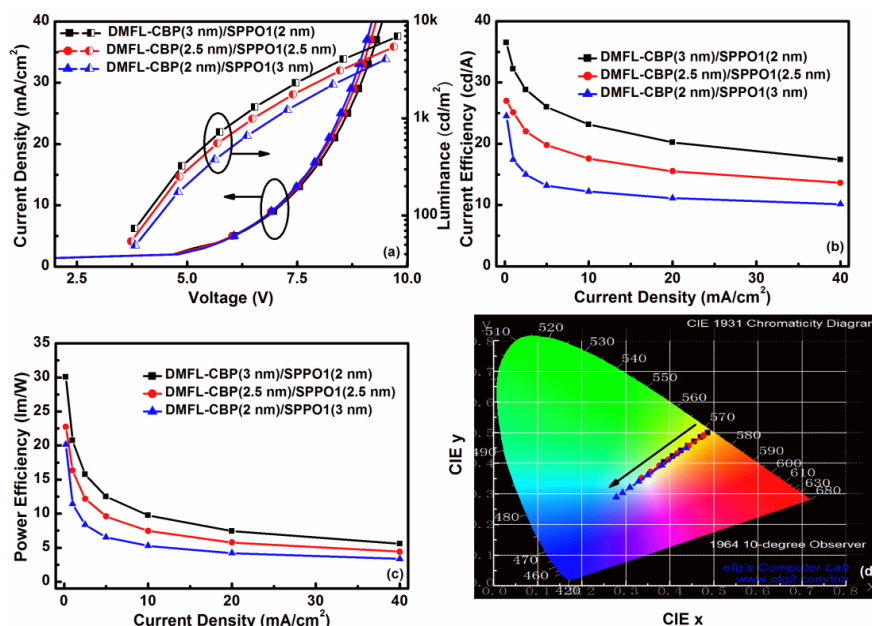


Figure 2. (a) The current density-voltage-luminance relationship; (b) The current efficiency-current density relationship; (c) The power efficiency-current density relationship of Devices A, B, and C with double interlayers of DMFL-CBP (3 nm)/SPPO1 (2 nm), DMFL-CBP (2.5 nm)/SPPO1 (2.5 nm), DMFL-CBP (2 nm)/SPPO1 (3 nm), respectively; (d) The CIE(x,y) coordinates of the three devices, in which the square, circle, and triangle symbol for Devices A, B, and C, respectively. And the direction of the arrow represents the increase trend of applied current density from 0.2 to 1, 2.5, 5, 10, 20, 40 mA/cm².

When the thickness of DMFL-CBP increased, the exciton generation interface shift closer to the yellow EML, and thus leading to more exciton diffused into the yellow EML. Moreover, the efficiency of OLEDs with PO-01 as emitter is larger than that of OLEDs with BCzVBi as emitter. So, higher efficiency are achieved in the HWOLEDs with thicker DMFL-CBP. In **Figure 2(d)**, when the driving current density increase from 0.2 mA/cm² to 40 mA/cm², the CIE coordinates of Device A with DMFL-CBP (3 nm)/SPPO1 (2 nm) changes from yellow (0.4858, 0.5000) to warm white (0.3865, 0.4038). And the CIE coordinates of the Device B with DMFL-CBP (2.5 nm)/SPPO1 (2.5 nm) changes from yellow (0.4766, 0.4909), across warm white (0.4171, 0.4324) to white (0.336, 0.3524). For the Device C with DMFL-CBP (2 nm)/SPPO1 (3 nm), the CIE coordinates ranges from warm white (0.4368, 0.4497), across white (0.3289, 0.3408) to cool white (0.2781, 0.2896). The color-tunable HWOLEDs may satisfy people's different demands in different situations.

To investigate the physical mechanism in these HWOLEDs, the normalized EL spectra driving at 0.2, 1, 2.5, 5, 10, 20, 40 mA/cm² are studied and exhibited in **Figure 3** with the yellow emission peak intensity (located at 560 nm) as the reference intensity. The proportion of blue emission part in the normalized EL spectrum increased with the increasing current density in each devices. This may be caused by that more singlets are captured by the blue fluorescent emitter

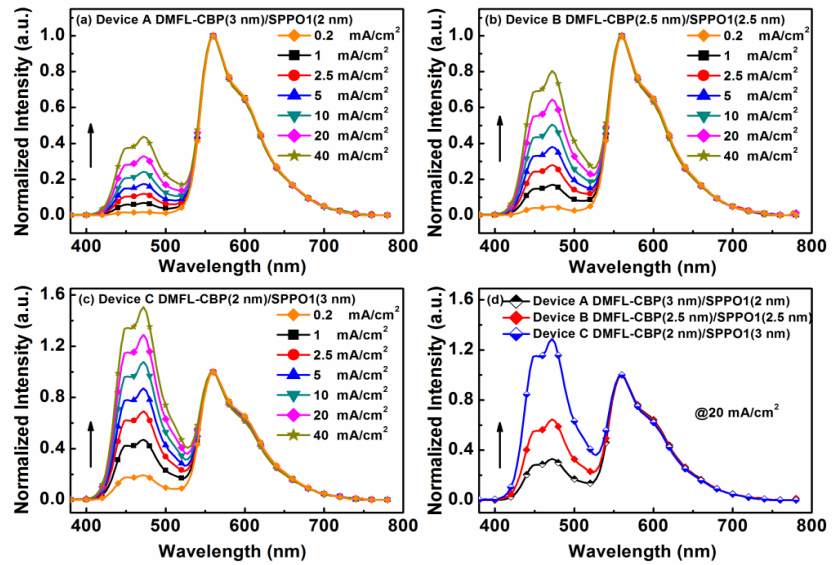


Figure 3. (a) The normalized EL spectra of Device A; (b) The normalized EL spectra of Device B; (c) The normalized EL spectra of Device C driving at current densities of 0.2, 1, 2.5, 5, 10, 20, 40 mA/cm²; (d) The EL spectra of Devices A, B, and C driving at 20 mA/cm².

BCzVBi, due to more electrons injected in to the blue EML across the DMFL-CBP layer with the increased current density. When the current density is fixed such as 20 mA/cm², the proportion of blue emission part goes up with the thickness of the DMFL-CBP decreasing, because the exciton generation interface DMFL-CBP/SPPO1 is closer to the blue fluorescent EML and thus more singlet emitting blue light via BCzVBi.

4. Conclusion

A hybrid white light-emitting diode with widespread color CIE coordinates ranging from warm white (0.4368, 0.4497), across white (0.3289, 0.3408) to cool white (0.2781, 0.2896) is demonstrated by incorporating double interlayers of DMFL-CBP/SPPO1 between the blue fluorescent emitting layer and the yellow phosphorescent emitting layer. The color CIE coordinates can be tuned by both the thickness ratio of DMFL-CBP to SPPO1 and the driving current density, due to relative change of the exciton generation interface and the electron injecting ability into blue fluorescent emitting layer.

Acknowledgements

We acknowledge financial support from the Natural Science Foundation of China (Grant Nos. 61404108), and Fundamental Research Funds for the Central Universities (Grant Nos. XDJK2017D140).

Conflict

The authors declare no competing financial interest.

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