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Research Article

The Investigation on Color Purity of Blue Organic Light-Emitting Diodes (BOLED) by Hole-Blocking Layer

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Organic light-emitting diodes (OLEDs) with triple hole-blocking layer (THBL) structure, which consist of 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP), 4,4'-bis(2,2'diphenyl vinil)-1,1'-biphenyl (DPVBi), and (4,4'-N,N'-dicarbazole)biphenyl (CBP), have been fabricated. Regardless of applied voltage variation, the luminous efficiency of the OLEDs with THBL structure was increased by 41% as compared with the dual hole-blocking layer (DHBL) structure. The CIE coordinates of (0.157, 0.111) of device with THBL structure are close to pure blue emission than that of other devices of DHBL. There is a coordinate with the slight shift of $\pm \Delta_{x,y} = (0.001, 0.008)$ for the device with THBL structure during the applied voltage of 6–9 V. The results indicate that the excitons can be effectively confined in the emitting layer of device, leading to an enhancement of luminance efficiency and more stable coordinate.

1. Introduction

Recently, organic light-emitting diodes (OLEDs) have attracted much attention due to their superior characteristics such as high luminance, wide range of colors, and wide viewing angles. OLEDs have been regarded as the next generation display technology. Many approaches have been tried to realize full-color displays [1], and it requires three basic emitting colors, red, green, and blue. However, the blue OLEDs still have inherent problems of low efficiency, poor color purity, and short lifetime in comparison with other red or green OLEDs [2]. It is important to keep the color stability with applied voltage in the blue OLEDs. So far, there is still a distance to achieve standard Commission International de L'Eclairage (CIE) coordinates (0.14, 0.08) of blue OLEDs. Generally, there are many structures of emitting layer to obtain blue OLEDs, such as single emitting layer, double emitting layer, blue guest doped layer, or multiple-quantumwell (MQW) layer [3-12]. Shi et al. utilized MQW structure

of [4,4-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB)/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP)] $_n$ that makes hole-electron balance in emitting layer, resulting in the CIE coordinates of (0.1747, 0.1059) [11]. Bang et al. used 4,4'-bis(2,2'diphenyl vinil)-1,1'-biphenyl (DPVBi) and tris-8-hydroxy-quinoline aluminum (Alq $_3$) as double emitting layer, leading to CIE coordinates of (0.150, 0.137) at 11 V [2]. However, the CIE coordinates are not stable by using the above structure or method, resulting from the shift of the excitons recombination zone.

In this study, we improved the color purity of OLEDs by using structure of triple-hole blocking layer (THBL), which consisted of alternate (4,4'-N,N'-dicarbazole) biphenyl (CBP) and BCP. Both of them have excellent electron mobility and holes barrier ability. The CBP layer compared with BCP layer has better electron mobility and is $3 \times 10^{-4} \, \mathrm{cm}^2 / \, \mathrm{V}^{-1} \, \mathrm{s}^{-1}$, which will favor electron injection from BPhen layer to emitting layer [13]. The BCP layer can effectively control the carrier in the emitting layer that will enable an increase

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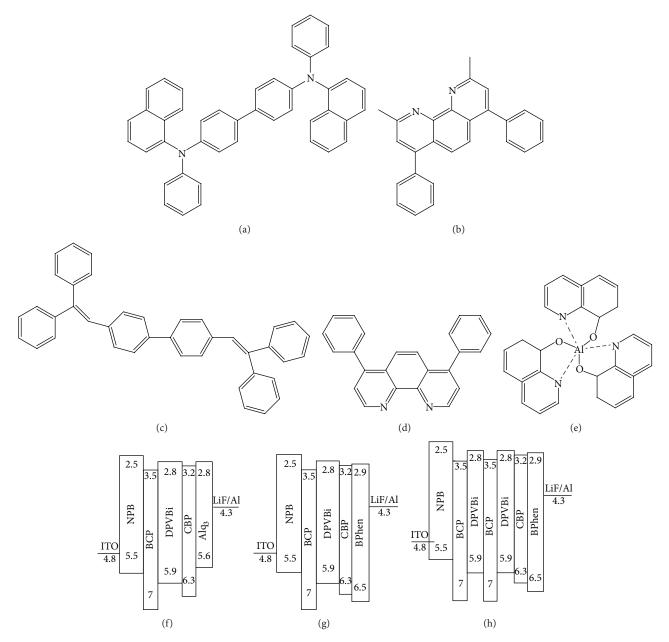


FIGURE 1: The chemical structures of (a) NPB, (b) BCP, (c) DPVBi, (d) BPhen, and (e) Alq₃ and the schematic energy band diagrams of (f) device A, (g) device B, and (h) device C.

in the recombination of emission region. It is due to the fact that there is a higher occupied molecular orbital (HOMO) of 7.0 eV for BCP. However, we used the THBL structure so that the carrier can be effectively confined and controlled in the DPVBi emitting layer, resulting in the fact that the CIE color coordinates of devices can be considerably improved. Simultaneously, the mechanism of the color stability of device with various structures is presented.

2. Experimental

Indium tin oxide (ITO) coated glass with a sheet resistance of approximately 15 Ω/\Box was consecutively cleaned in ultrasonic bath containing detergent water, acetone, ethanol, and

deionized (DI) water for 20 min each and then dried with a nitrogen (N_2) flow. The chemical structures of organic materials used and the schematic energy band diagrams of the devices A–C in this study are shown in Figure 1. These structures were as follows:

- (A) ITO/NPB (40 nm)/BCP (8 nm)/DPVBi (40 nm)/CBP (2 nm)/Alq₃ (20 nm)/LiF (0.5 nm)/Al (100 nm);
- (B) ITO/NPB (40 nm)/BCP (8 nm)/DPVBi (40 nm)/CBP (2 nm)/BPhen (10 nm)/LiF (0.5 nm)/Al (100 nm);
- (C) ITO/NPB (40 nm)/BCP (I) (6 nm)/DPVBi (20 nm)/BCP (II) (2 nm)/DPVBi (20 nm)/CBP (2 nm)/BPhen (10 nm)/LiF (0.5 nm)/Al (100 nm).

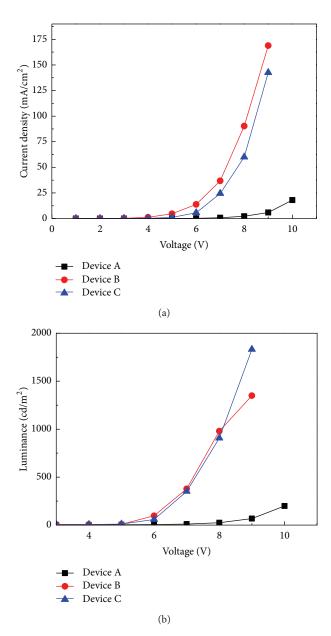


FIGURE 2: The (a) current density-voltage and (b) luminance-voltage characteristics of the devices A–C.

In the device, NPB is used as hole-injecting layer and hole-transport layer. The BCP and CBP are used as hole-blocking layer inserted between DPVBi and HTL. The DPVBi acts as blue-emitting layer. The Alq $_3$ and 4,7-diphenyl-1,10-phenanthroline (BPhen) are used as a HTL. LiF and Al are used as an electron-injection layer and cathode, respectively. All of the deposition procedures involved thermal evaporation in a vacuum chamber (<10 $^{-6}$ Torr). The organic layers were sequentially evaporated onto the ITO coated-glass substrate. The deposition rates of all organic materials were monitored by an oscillating quartz thickness monitor (Sigma, SID-142). The deposition rates were about 0.2~0.5 Å/s for organic layers and 5 Å/s for cathode (Al) and LiF. CIE coordinates, electroluminescence (EL) spectra, and luminance were measured by the photospectrometer (Kollmorgen

Instrument PR-655, USA) and the current-voltage characteristics were simultaneously measured by the programmable power source (Keithley SourceMeter 2400, USA), controlled using computer software (Chief I-V-L system, Taiwan). All measurements were carried out at room temperature under ambient.

3. Results and Discussion

Figures 2(a) and 2(b) show the current density-voltage and luminance-voltage characteristics of devices, respectively. Generally, the applied voltage is defined as turn-on voltage when the current density of device reaches the one-tenth of maximum value of the current density. In Figure 2(a), the turn-on voltages of device A-C are 9.5, 5.5, and 6.2 V, respectively. The turn-on voltage of device A is higher than that of devices B-C. It is attributed to the different electron transport layers (Alq₃, BPhen) in the devices. It is well known that the electron mobility of BPhen (2.4 $\times\,10^{-4}\,\text{cm}^2/\text{V}^{-1}\,\text{s}^{-1})$ is higher than that of Alg_3 (4.7 × 10⁻⁶ cm²/V⁻¹ s⁻¹) [14, 15]. However, the ability of electron injection from cathode to the emitting layer can increase via using an electron transport layer with higher electron mobility, resulting in a decrease in the turn-on voltage of device. In addition, from the turn-on voltages of devices B and C, there is a high turn-on voltage in device C due to inserting a thin BCP layer in the emitting layer to form the THBL structure.

By comparing devices A and B, the luminance is greatly improved from $68 \, \text{cd/m}^2$ to $1350 \, \text{cd/m}^2$ at $9 \, \text{V}$. It can be ascribed to using the BPhen to replace the Alq_3 , resulting in the fact that more electrons inject from the cathode to the DPVBi and generate more excitons in the DPVBi layer.li The luminance of devices B and C are $1350 \, \text{and} \, 1831 \, \text{cd/m}^2$ at $9 \, \text{V}$, respectively. The enhancement of the luminance in device C as compared with device B can be attributed to three factors: (1) THBL has better excitons confinement ability, (2) an increase in excitons formation in EML, and (3) there is formation both emitting layers (DPVBi) in THBL of device C.

In the devices, holes are accumulated and confined in the BCP layers because of the existence of the barrier at the interface of NPB/BCP. Hence, the hole mobility in the multilayer is expected to be decreased, and the performance of device C is expected to be enhanced. The luminous efficiency-current density characteristic is shown in Figure 3. The major parameters are summarized in Table 1. The maximum luminous efficiencies of devices B and C are 1.08 and 1.52 cd/A, respectively. The luminous efficiency of device C is 41% higher than that of device B. It shows that there is a better carrier balance in device C. The electrons are confined and accumulated in the lowest unoccupied molecular orbital (LUMO) level of DPVBi/BCP/DPVBi potential well in device C due to the existence of potential barrier at the DPVBi/BCP (II) and BCP (I)/DPVBi heterointerfaces. As a result, the balance of electron and hole can be improved by inserting a thin BCP layer in device C, forming the THBL structure, and thus more carriers will be confined within both the DPVBi layers.

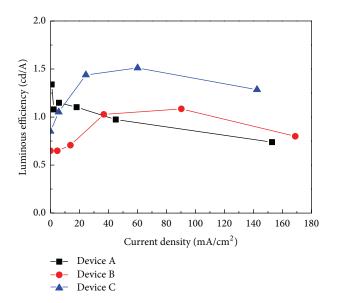


FIGURE 3: The luminance efficiency-current density characteristics of the devices A–C.

TABLE 1: Performance of devices A-C.

	Device A	Device B	Device C
Turn-on voltage (V)	9.5	5.5	6.2
Luminance (cd/m ²)	68	1350	1830
Luminance efficiency (cd/A)	1.15	0.8	1.28
Wavelength peak (nm)	448	444	436
FWHM (nm)	68	65	65
$CIE \pm \Delta_{x,y}$	(0.010, 0.005)	(0.000, 0.004)	(0.001, 0.008)

Figure 4 shows the normalized electroluminescent (EL) spectra of the devices A-C at the same current density of 10 mA/cm². The EL peaks of devices A-C are at 448, 444, and 436 nm, respectively. The EL peaks of devices A and B are near to the PL peak of NPB (446 nm) [16]. It can be directly inferred and demonstrated that major holes were accumulated in the interface of the NPB/BCP and electronhole recombination occurred in NPB layer, resulting in the blue emission from the NPB layer. The EL peak of the device C is consistent with the PL peak of DPVBi (436 nm) [17]. It is due to the insertion of thin BCP layer in the middle of DPVBi layer, leading to the fact that the major holes were confined in the HOMO level of BCP (I)/DPVBi/BCP (II) potential well in device C and recombined with electron in the DPVBi layer. From the energy level of Figure 1, the LUMO of BCP is -3.5 eV and is higher than that of DPVBi, so the inserted thin BCP (II) layer can confine the electron and prevent it from transporting from blue DPVBi layer to NPB layer. In Figure 4, the full width at half maximum (FWHM) of devices A-C are 68, 65, and 65 nm, respectively. It can be seen in Figure 4 that there is a little color-mixing phenomenon of device A and it emits blue emission because the spectrum has a broad peak.

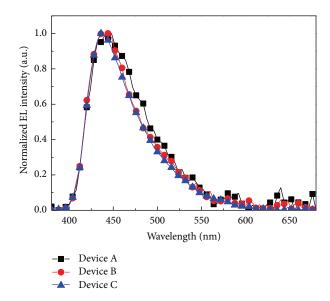


FIGURE 4: The normalized EL intensity spectra characteristics of the devices A–C at the current density of 10 mA/cm².

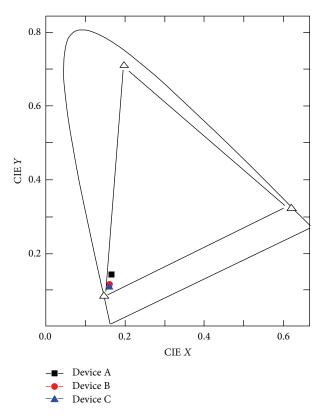


FIGURE 5: The CIE chromaticity coordinates of devices A–C at the applied voltage of 9 V.

That is to say, the blue emission in device C is more pure than that of other devices.

Figure 5 shows the CIE coordinates of devices as a function of applied voltage. The CIE coordinates of device A, device B, and device C at 9 V are (0.166, 0.135), (0.157, 0.113), and (0.157, 0.111), respectively. The CIE coordinates

of device A are far away from the pure blue emission with CIE coordinates of (0.14, 0.08). It is attributed to the portion of carriers recombination at the Alq₃ layer, generating slight green emission. The phenomenon is consistent with the result obtained in Figure 4. Besides, the CIE coordinates of device A are (0.166, 0.135) at 9 V. The difference in CIE coordinates of device A is $\pm \Delta_{x,y} = (0.010, 0.005)$. Thus, the function of BPhen to replace the Alq3 is not only due to its higher mobility but also due to its higher HOMO energy level which can avoid the hole to cross the CBP barrier layer, keeping off a mix of green light and far CIE coordinates of pure blue light. The CIE coordinates of devices B-C are (0.157, 0.113) and (0.157, 0.111) at 9 V, respectively. By comparison with device A, the color coordinates of the devices B-C are close to the pure blue region. The variation in CIE coordinates of devices B-C is only $\pm \Delta_{x,y} = (0.000, 0.004)$ and (0.001, 0.008), respectively. All the above results reveal that the displacement of the CIE coordinates for the device C at high voltage is obviously small in comparison with that of devices A-B, indicating that there is a better stability and more standard CIE coordinate of blue emission in device C. This is because the emission region of the OLEDs with a THBL consisting of BCP layer and CBP layer is confined within both DPVBi layers. Hence, the performance of near blue emitting devices with a THBL structure of BCP and CBP is greatly improved which is attributed to the improved hole-electron balance and that can be further attributed to hole barrier in the HBL structure. So by adjusting the number of multiple HBL, the amount of hole injection into emitting layer can be controlled. In this study, THBL structure outperforms dual-HBL (DHBL) structure in both luminance and efficiency, enabling electrons to be injected into the device easily and increasing electronhole pair recombination in EML to achieve carrier effective balance.

4. Conclusions

In summary, the OLEDs with THBL and DHBL structure are fabricated and investigated. With THBL structure, it can efficiently control the holes transport and achieve an electronhole pair balance, the efficiency of which is improved by 41% as compared with the DHBL. The optimized blue CIE coordinates of (0.157, 0.111) at 9 V and coordinates shift only $\pm\Delta_{x,y}=(0.001,\,0.008)$ were obtained. These results indicate that the enhancement of luminance efficiency and the more stable purity of blue color can be achieved by using THBL due to excitons confined in the EML.

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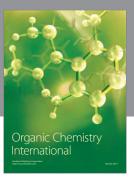
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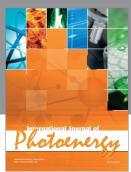
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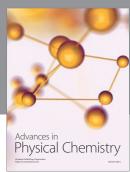
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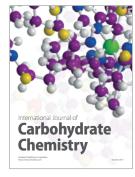
















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