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The Structure Optimization of White Organic Light-Emitting Diodes for High Efficiency

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We have demonstrated the structure optimization of white organic light-emitting diodes with two separated emissive layers for better efficiency. For efficient optimization of white organic light-emitting diodes, we carried out a insertion of hole buffer layer, a thickness change of emission layer, and a change of electron transport layer. The optimized device showed maximum luminance of 29 600 cd/m² at 14V and the maximum luminous efficiency of $14.30 \, \text{cd/A}$ at $7.34 \, \text{mA/cm}^2$, respectively. All devices showed white emission at operating voltages without change of $\text{CIE}_{x,y}$ coordinates.

Keywords: optimization; two separated emissive layers; white organic light-emitting diodes

INTRODUCTION

Organic light-emitting diodes (OLEDs) have attracted increasing attention in recent years [1–5]. Today, OLEDs are considered to be one of the flat-panel displays for the next generation due to low-voltage operation, wide-viewing angle, a high contrast ratio, and mechanical flexibility [6]. Multi-layered OLEDs (which consist of several layers such as HTL, ETL, EML, etc.) have been studied [7]. White Organic light-emitting diodes (WOLEDs) have drawn increasing

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attention as a solid-state light source and backlights in liquid-crystal displays and full-color OLEDs due to their light weight, low operating voltage and high contrast ratio [8-13]. So many device structures have been proposed to realize WOLED. Many researchers consider small molecule for WOLED because of lower efficiency, a difficult stack of organic molecules, and absence of high efficiency emission materials in polymer OLED (PLED). In this letter, We demonstrated a efficient WOLED with a fluorescent and phosphorescent emissive dopants and two different hosts, 2-methyl-9,10-di(2-naphthyl)anthracene (MADN) and bis(2-methyl-8-quinolinato)-4-phenylphenolate (BAlq). To achieve high efficiency, we controlled hole-electron recombination probability, emission layer thickness, and a simplified process. The optimized device showed maximum luminance of 29,600 cd/m² at 14 V, the maximum luminous efficiency of 14.30 cd/A at 7.34 mA/cm², and Commission Internationale de L'eclairage (CIEx,y) coordinates of (0.28, 0.35) at 6 V, respectively. The optimized device showed higher efficiency of about 1.8 times than control device. The various characteristics of WOLEDs were studied and emission mechanism was also investigated.

EXPERIMENTAL

Fabrication of OLED

Indium tin oxide (ITO) coated glass was cleaned in an ultrasonic bath by regular sequence: in acetone, methanol, distilled water and isopropyl alcohol. Hereafter, pre-cleaned ITO was treated by O₂ plasma treatment whose condition were 2×10^{-2} Torr, 125 W and 2 min [14]. WOLEDs were fabricated using the high vacuum (5 \times 10⁻⁷ Torr) thermal evaporation of organic materials onto the surface of ITO (30 Ω/sq , emitting area is 3 mm × 3 mm) coated glass substrate. All organic materials were purified by temperature-gradient sublimation except dopants in vacuum. The doping rates were 1.0-1.1 Å/sec on organic materials and 0.1 Å/sec on lithium quinolate (Liq), approximately. After the deposition of the organic layers and without a vacuum break, the Al cathode was deposited at a rate of 10 A/sec. The device structures are as follows: ITO as a anode, 4,4',4"-tris[2-naphthyl-(phenyl)amino] triphenylamine (2-TNATA) as a hole buffer, N,N'bis-(1-naphyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) as a hole transporting, MADN as a blue host, 1,4-bis[2-(4'-diphenylaminobiphenyl-4-yl)vinyl]benzene (DABPV-ph) as a blue dopant, BAlq as a red host and hole blocking, bis(5-acetyl-2-phenylpyridinato-C,N) iridium(III) acetylacetonate ((acppy)₂Ir(acac)) as a red dopant, 4,7-diphenyl-1,10phenanthroline (BPhen) as a hole blocking and electron transporting, tris-(8-hydroxy-quinolinato) aluminum (Alq₃) as a electron transporting, Liq as electron injection, and Al as a cathode material. The optimized doping concentrations for DABPV-ph in MADN and (acppy)₂Ir(acac) in BAlq were 7% and 8% concentration, respectively.

Measurements

With the DC voltage bias, the optical and electrical properties of WOLEDs such as the current density, luminance, luminous efficiency, and $CIE_{x,y}$ coordinates characteristics were measured with keithley 236 and CHROMA METER CS-100A, respectively.

RESULTS AND DISCUSSION

Figure 1 shows the device structures of OLEDs fabricated in this study. Device A, B, C, and D fabricated as the following sequence: ITO as anode/2-TNATA (device B, C, and D) as hole buffer layer (HBL)/NPB as hole transporting layer (HTL)/DABPV-ph: MADN (7%) as blue emitting layer (BEML)/(acppy)₂Ir(acac): BAlq (8%) as red emitting layer (REML)/BAlq (device A, B, and C) as hole and exciton blocking layer (HEBL)/Alq₃ (device A, B, and C) or Bphen (device D) as electron transporting layer (ETL)/Liq (20 Å) as electron injection layer (HIL)/Al as cathode.

Figure 2 shows the energy level diagrams for device A, B, C, and D, respectively. Compared to device A, device B had new insertion of HBL, 2-TNATA, between ITO and NPB. EML of device C is thicker than that of device B by 100 Å. Device D replaced from HEBL of BAlq and ETL of Alq₃ by BPhen for more simple process. Device B controlled hole-electron recombination probability and device C had broader emission zone for more confined excition. Device D also changed from BAlq as HEBL and Alq₃ as ETL to BPhen as HEBL and ETL for a more simple process and higher electron mobility.

Figure 3 shows the current density-voltage characteristics of device A, B, C, and D. They had the maximum current density of 585, 276, 239, and $250\,\mathrm{mA/cm^2}$ at $14\,\mathrm{V}$, respectively. Device A showed the highest current density at operating voltages, which seemed to be due to the absence of 2-TNATA as hole buffer layer for high efficiency between ITO and NPB in device A. Device C showed lower current density on the whole voltages than device B due to thicker EML of device C. Device D showed higher current density than device C at the operating voltages from 0 V to 13.5 V. Even though BPhen had a higher electron mobility of $5\times10^{-4}\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ than it of Alq₃, BPhen prevented more holes flowing through device D due to the higher

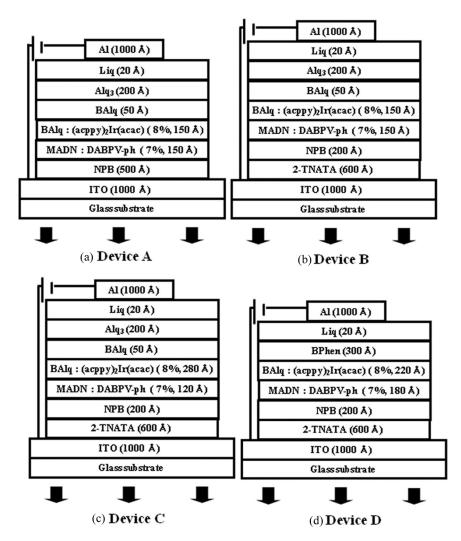


FIGURE 1 Structures of device A, B, C, and D.

highest occupied molecular orbital (HOMO) level of Bphen [15,16]. It was shown in inset Figure 3 that the devices A, B, C, and D had the maximum luminance of 38,100, 21,100, 23,600, and $29,600\, {\rm cd/m^2}$ at 14 V, respectively. Device A also showed the highest luminance than the other devices.

Figure 4 shows the luminous efficiency-current density characteristics of device A, B, C, and D. The devices showed the maximum

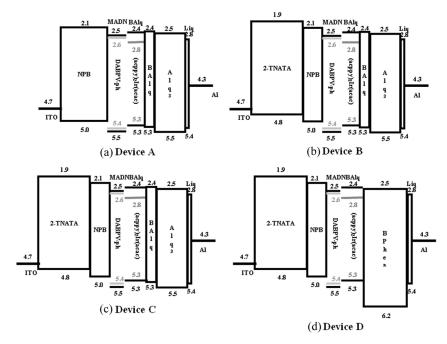


FIGURE 2 Energy-level diagrams for the device A, B, C, and D.

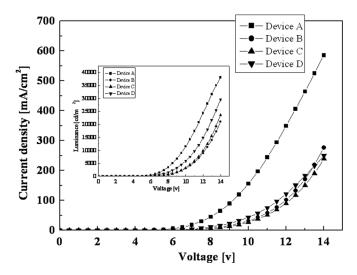


FIGURE 3 Current density (J) versus voltage (V) characteristics of device A, B, C, and D. Inset: luminance (L) versus voltage (V) characteristics of device A, B, C, and D.

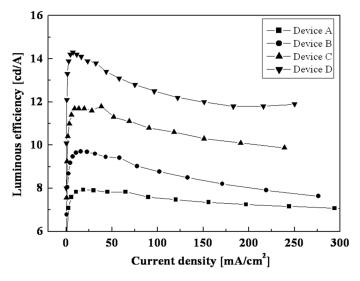


FIGURE 4 Luminous efficiency (LE) versus current density (J) characteristics of device A, B, C, and D.

luminous efficiency of 7.93, 9.70, 11.80, and 14.30 cd/A at 18.60, 15.90, 38.40, and 7.34 mA/cm², respectively. At nearby 20 mA/cm², the devices showed 7.93, 9.68, 11.7, and 13.9 cd/A at 18.6, 22.7, 19.8, and 23.5 mA/cm², respectively. The luminous efficiency for all devices did not decrease conspicuously, as the current density increased. The hole-electron recombination probability of device A seemed to be lower than the other devices because NPB and Alq3 possessed a considerable high hole mobility of $10^{-4} \, \mathrm{cm}^2 \, \mathrm{V}^{-1} \, \mathrm{S}^{-1}$ and low electron mobility of 10⁻⁶ cm² V⁻¹ S⁻¹, respectively [17]. Consequently, the efficiency of device A was affected by charge unbalance between hole mobility in NPB and electron mobility in Alq₃. Therefore we attempted inserted HBL, 2-TNATA, between ITO and NPB for the balance of hole-electron recombination in device B, C, and D. The luminous efficiency of device B improved about 1.2 times than device A as shown in Figure 4. So, device B as inserted 2-TNATA showed better charge balance than device A without 2-TNATA. To improve efficiency, device C was fabricated with thicker EML which had BEML of 120 Å and REML of 280 Å than EML of device B which had BEML of 150 A and REML of 150 A for expansion of hole-electron recombination zone and also showed the improved luminous efficiency of about 1.2 times than device B. Device D replaced had BAlq as HEBL and Alq₃ as ETL by BPhen as ETL in device C. BPhen had a higher electron mobility and HOMO than

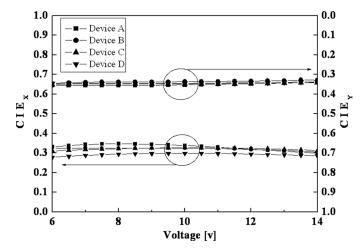


FIGURE 5 CIE $_{x,y}$ coordinates device A, B, C, and D with operation bias from 6 V to 14 V.

Alq₃ as mentioned before. Therefore device D showed higher efficiency of about 1.2 and 1.8 times than device C and device A, respectively. Although operating voltage increased as inserted 2-TNATA, we expect that the WOLEDs fabricated using device structures and materials described here may be applicable for backlights in liquid crystal displays for increasing luminous efficiency.

Figure 5 shows the $CIE_{x,y}$ coordinate changes of four devices as a function of the applied voltage from 6V to 14V. Device A, B, C, and D showed emission of $CIE_{x,y}$ coordinates from $(x=0.33,\ y=0.36)$, $(x=0.33,\ y=0.35)$, $(x=0.31,\ y=0.35)$, and $(x=0.28,\ y=0.35)$ at 6V to $(x=0.30,\ y=0.34)$, $(x=0.31,\ y=0.34)$, $(x=0.31,\ y=0.34)$, and

TABLE 1 The Characteristics of all Devices, Including Max. Luminance, Max. Luminous Efficiency, Luminous Efficiency at $20\,\text{mA/cm}^2$, and $\text{CIE}_{x,v}$ Coordinates Operating Bias at $10\,\text{V}$

Device	${ m Max.}$ luminance ${ m [cd/m^2]}$ at ${ m 14V}$	Max. luminous efficiency [cd/A]	$\begin{array}{c} Luminous\\ efficiency\\ [cd/A]\ at\ nearby\\ 20\ mA/cm^2 \end{array}$	CIE _{x,y} at 10 V
A	38,100	7.93	7.93	0.34, 0.35
В	21,100	9.70	9.68	0.33, 0.35
C	23,600	11.80	11.70	0.33, 0.35
D	29,600	14.30	13.90	0.30, 0.35

 $(x=0.29,\ y=0.34)$ at 14 V, respectively. It was found CIE_x of all devices decreased gradually as the applied voltage increased above the voltage of 8.0, 5.5, 10.0, and 9.5 V because of a triplet-triplet annihilation effect of the Ir derivatives at high injection current. Device D showed the most bluish white emission among other devices.

Table 1 showed various characteristics of all devices, including maximum luminance, maximum luminous efficiency, luminous efficiency at $20\,\text{mA/cm}^2$, and $\text{CIE}_{x,y}$ coordinates at $10\,\text{V}$.

CONCLUSIONS

In summary, we have fabricated WOLED with two separated emissive materials, blue fluorescent and red phosphorescent materials by controlling hole-electron recombination probability, emission layer thickness, and a more simple process for achieving high efficiency. It was found that device B controlling the balance of hole-electron recombination showed about 1.2 times efficiency than device A and device C expanding hole-electron recombination zone showed about 1.2 times efficiency than device B. Device D using more simple process showed 1.2 times efficiency than device C. Conclusively a luminous efficiency of WOLED fabricated showed about 1.8 times than that of device A. It was also shown that the CIE $_{\rm x,y}$ coordinates of white emission characteristics of device D did not change at operating voltage.

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