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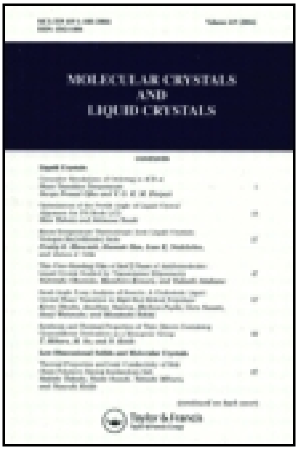

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Fabrication of White Organic Light-Emitting Diodes Using Two Complementary Color Methods

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Fabrication of White Organic Light-Emitting Diodes Using Two Complementary Color Methods

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White organic light-emitting diodes (WOLEDs) were fabricated using two complementary color methods with two emissive layer structure such as NPB (500 Å)/DPVBi (65 Å)/MADN:DCM2-0.5% (170 Å)/Bphen (300 Å)/Liq (20 Å)/Al (1000 Å). A deep blue and orange emissions were obtained from DPVBi layer and MADN host doped with a red fluorescent DCM2 dopant each. White emission was achieved through controlling hole-electron recombination by optimization of emissive layers thicknesses and DCM2 concentrations. Optimized WOLED device shows emission efficiency of 5.04 cd/A, current density of 2.64 mA/cm^2 and luminance of 938 cd/m^2 at 6 V with CIE$_{x,y}$ color coordinate of (0.338, 0.330). These results indicate that DPVBi layer effects on hole blocking and the exciton generation due to difference of energy level in highest occupied molecular orbital. The CIE$_{x,y}$ coordinates of device was slightly changed change from (0.338, 0.330) at 6 V to (0.341, 0.333) at 12 V, which is almost independent on driving voltages.

Keywords: OLED; two complementary colors; white emission

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INTRODUCTION

Since Tang and VanSlyke reported the stacked organic light-emitting diode (SOLED) [1], a number of further studies [2–4] have been conducted. The organic light-emitting diode (OLED), a self-emitting device, is drawing a lot of attention [5–8] as a next-generation display that may overwhelm liquid crystal displays (LCDs) and plasma display panels (PDPs). More than that, it has been evaluated to have superiority over the existing flat panel display (FPD) in terms of its capacity for high quality representation. It also has many advantages in terms of price competitiveness through its simple production process, improved optical view-angle, faster response-speed, and thinner film type. Several companies of home and abroad, have already reached such a commercial stage that OLEDs are now being installed in cellular phones and other kinds of displays. It is predicted that more applications will be made to other display devices, such as IMT 2000 cellular phones, personal data assistants (PDAs), and car navigation systems (CNSs) [9].

The OLED generally has a stacked structure mainly composed of an organic small molecular material. In a promising bid to represent full color through the OLED, researchers have mostly focused on studies like the dot-matrix method by separating emission for RGB and the white-emissive OLED with color filter [10,11]. Many studies have been taking place on the white-emission method because of it's proven processibility for backlight and color filter in LCD technology [12]. White-emission methods can be divided into two types. One is the 3-wavelength type [13] that mixes the existing RGB and the other is the 2-wavelength type that mixes blue and either orange or red [14–16]. The 2-wavelength type has emerged in order to make up for some drawbacks in the 3-wavelength type, but a substitute for the 2-wavelength method is urgently needed because it still uses the same blue-emitting material in the 3-wavelength method.

Therefore, this study aims to represent white emission of a 2-wavelength type. Afterwards, we achieved blue emission by using the existing DPVBi and then finally achieved orange emission by using MADN as a host material and DCM2 as a guest material. To fabricate the 2-wavelength type of white OLEDs having an emitting layer (EML) with a blue/orange emitting layer structure, we evaluated the characteristics of OLEDs as varying the thickness of the emitting layer, creating an optimal structure, changing DCM2 concentrations with MADN, and then established the high efficient the 2-wavelength type of white OLEDs.

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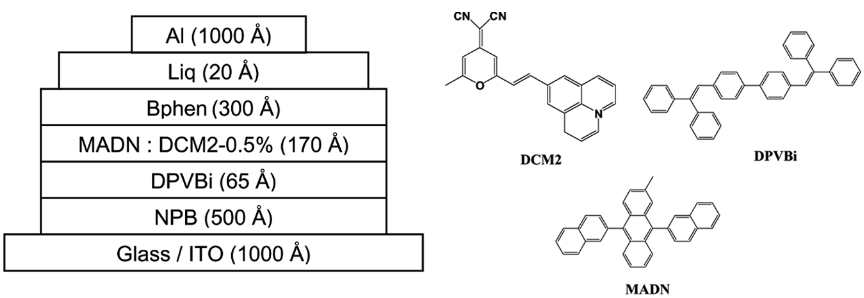
EXPERIMENT

ITO coated glass was cleaned in ultrasonic bath by regular sequence: in acetone, methanol, diluted water and isopropyl alcohol. Hereafter, pre-cleaned ITO was treated by O_2 plasma under condition of 2×10^{-2} Torr, 125 W and 2 min [17]. White OLEDs were fabricated by thermal evaporation under the high vacuum (1.0×10^{-6} Torr).

Figure 1(a) shows the schematic configuration of the WOLEDs prepared in this study, and Figure 1(b) does the molecular structures of the chromophores in the devices. The multiplayer structure of WOLED device with blue fluorescence DPVBi were as follows: ITO/ N,N′-bis-(1-naphyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) as a hole transport layer/4,4′-bis(2,2′-diphenylyinyl)-1,1′-biphenyl (DPVBi) as a blue emissive layer/ultrathin [2-methyl-6-[2-(2,3,6,7-tetrahydro-1H,5H-benzo[ij] quinolizin-9-yl)ethenyl]-4H-pyran-4-ylidene] propane-dinitrile (DCM2) in 2-methyl-9,10-di(2-naphthyl) anthracene (MADN) as a red emissive layer/8-hydroxy quinolino!aluminum (Alq_3) as an electron transport layer/lithium quinolate (Liq) as an electron injection layer/ aluminum (Al) as a cathode. Two kinds of devices were fabricated using doped DCM2 0.5% and 1.0% to MADN as adapting different thickness of blue and red emissive layers. With different DC voltage bias, the optical and electrical properties of WOLEDs such as the current density, luminance, luminous efficiency, Commission Internationale de L'eclairage (CIE) coordinates and electroluminescence characteristics were measured with Keithley 236, CHROMA METER CS-100A and JBS OLED analysis system IVL-200 respectively.



(a) (b)

FIGURE 1 (a) Device structure of the WOLED. (b) Used organic materials as the WOLED.

RESULT AND DISCUSSION

Figure 2 shows UV-vis. absorption and photo- luminescence(PL) spectra of DPVBi, MADN, and DCM2. As shown Figure 2(a), in UV/vis. absorption spectrum of DCM2 as dopant material and photoluminescence

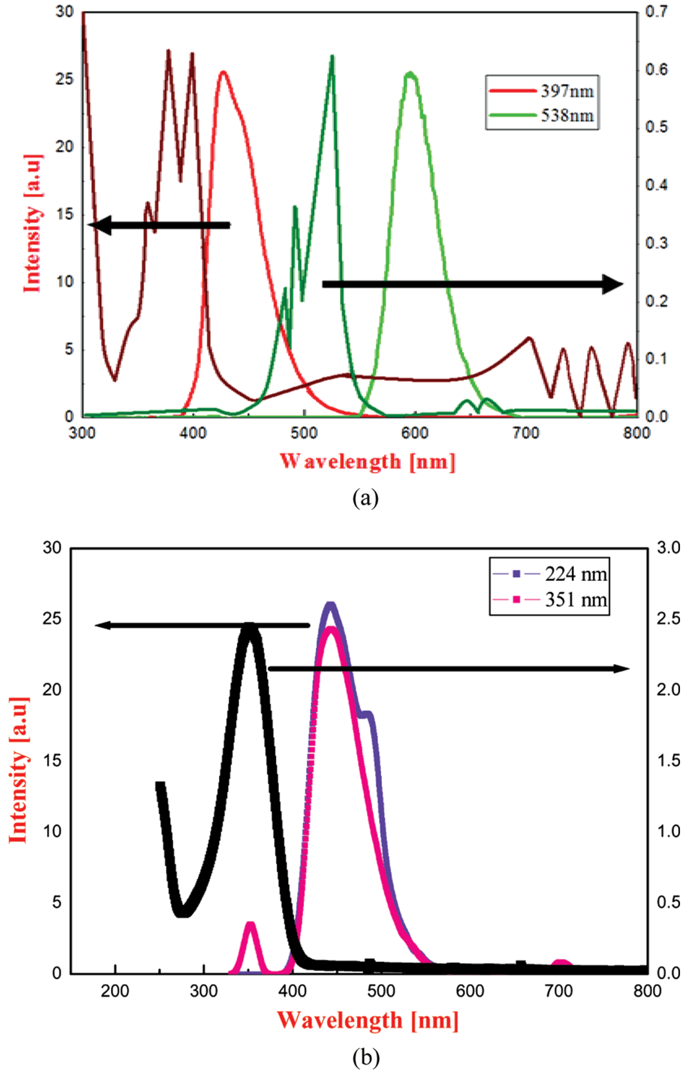


FIGURE 2 UV absorption and photoluminescence(PL) spectrum of (a) MADN and DAM2. (b) DPVBi.

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of MADN as host material, we predicted energy transfer from the MADN to the DCM2 considering spectrum overlapping between the UV/vis absorption of DCM2 and phosphorescence(photoluminescence?) of MADN. White OLEDs are also expected by photoluminescence according to sum of two materials which DCM2 using as red emission material and DPVBi using as blue emission material.

UV-vis absorption spectrum of the DCM2 and photoluminescence of the MADN were overlapped in the region of 450 nm to 550 nm and it explains labile energy transfer from MADN to DCM2. As shown Figure 2(b), white emission is estimated sum of photoluminescence of DPVBi at 445 nm and that of DCM2 at 595 nm with maximum intensities.

We compared CIEx,y coordinates of blue and red OLED devices using DPVBi, MADN, and DCM2. The CIEx,y coordinates of blue OLED device with DPVBi layer was (0.159, 0.191) and these of red OLED devices B1, B2, B3, B4, and B5 with doped 0.25, 0.5, 1, 2, 4% DCM2 to MADN were (0.426, 0.440), (0.468, 0.453), (0.503, 0.455), (0.542, 0.427), and (0.571, 0.401), respectively. Figure 3 shows that CIEx,y coordinates of red OLED devices with increased DCM2 concentration was approached to deep red region while that with decreased DCM2 concentration was shifted to yellow-orange region due to Förster energy transfer from MADN to DCM2. Considering perfect

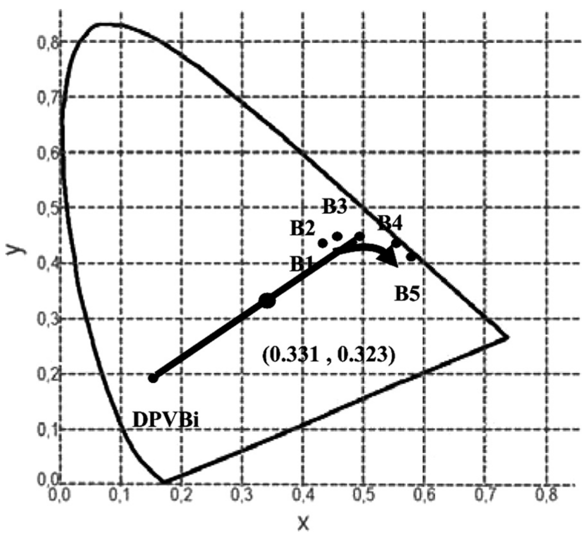

FIGURE 3 CIE coordinate of DPVBi and MADN:DCM2 with DCM2 concentration.

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white color coordinate (0.33, 0.33), white OLED device with 1% of DCM2 in MADN obtained best CIE white coordinate of (0.331, 0.323) with optimum mixture of red CIE coordinate of (0.503, 0.455) and blue CIE coordinate of (0.159, 0.191).

Figure 4 shows CIEx,y coordinates of white OLEDs as its emission layers were confined 150 Å through thickness optimization of blue and red emission layers. Thickness ratio between blue emissive DPVBi and red emissive MADN:DCM2-1% layers of four devices C1, C2, C3, and C4 were defined such as 50/100 Å, 55/95 Å, 60/90 Å, and 80/70 Å, respectively. Among these four white OLED devices, CIEx,y coordinates of the C2 was closer to perfect white emission of (0.33, 0.33). Increased DPVBi thickness from 55 Å to 80 Å, indeed, white OLED shifted to DPVBi's blue CIEx,y coordinates indicating hole-electron recombination was enormously depended on DPVBi thickness. In experiment, however, color characteristics of white OLED device C2 were different from calculation of CIE coordinates of blue and red emission. The reason for CIE coordinates difference between experimental and calculated values is due to deviation caused by optical property of organic multi layers and organic surface morphology such as micro-cavity and electromer.

Thickness of DPVBi layer was fixed at 55 Å and then that of MADN:DCM2-0.5% layer was increased such as 95 Å, 145 Å, 170 Å,

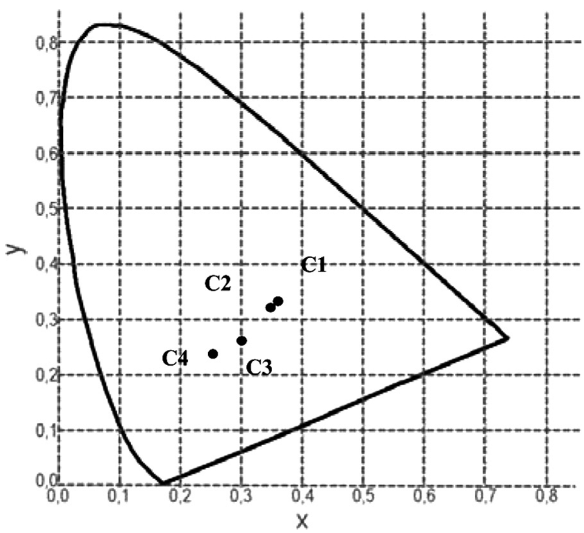

FIGURE 4 CIE coordinate of white OLEDs, C1, C2, C3, and C4 with various thickness ratio of DPVBi and MADN:DCM2-1% layer.

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and 210 Å. CIEx,y coordinates of white devices D1, D2, D3, and D4 as increasing thickness of MADN:DCM2-0.5% layer were (0.330, 0.320), (0.338, 0.323), (0.347, 0.334), and (0.381, 0.377), respectively. As shown in Figure 5, considering recombination zone in the device of MADN: DCM2 and DPVBi, CIE coordinates was not changed much from (0.33, 0.32) to (0.34, 0.32) although thickness of red emissive layer was changed from 95Å to 145Å while white coordinates was shifted to (0.35, 0.34) and (0.38, 0.38) as thickness of MADN layer changed to 170 Å and 215 Å. In other words, there was no significant change of CIEx,y coordinate of white emission until thickness of MADN reached at 145 Å but it was shifted to red emissive region after 215 Å because recombination occurs more in MADN layer than DPVBi of white OLED device with MADN:DCM2-0.5%.

Figure 6 shows energy diagram of fabricated WOLEDs. Change of CIE color coordinates in white OLED devices with blue emissive DPVBi layer and red emissive MADN:DCM2 layer was effected by thickness of each layer and also depended on energy levels of layer materials. DPVBi's HOMO energy level is 0.9 eV higher than that of NPB and this makes hole-electron recombination mainly occur in DPVBi. Increased bias voltages, therefore, CIEx,y coordinates shifted to red emission due to extension of recombination to the MADN:DCM2 layers. As a result, DPVBi layer thickness sensitively influenced on the white emission rather than that of MADN:DCM2 layer according to very low HOMO energy.

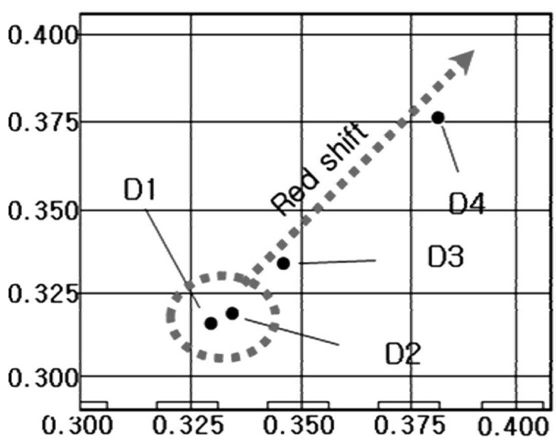

FIGURE 5 CIEx,y coordinates of white OLEDs with increasing MADN:DCM2 layer thickness.

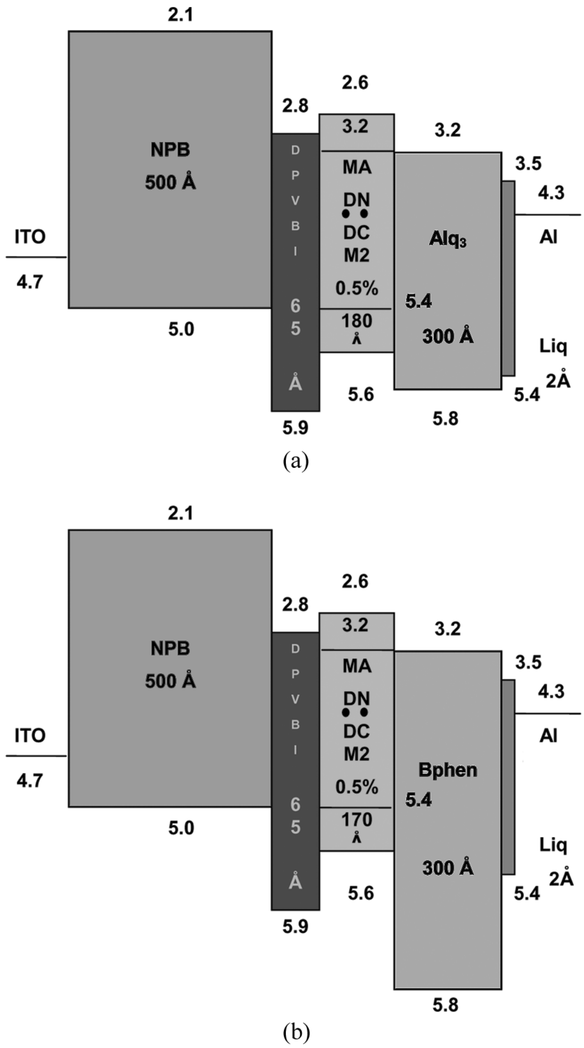

FIGURE 6 Energy level diagrams and structures of white OLED with optimized white emission. (a) Alq_3 was used as ETL (b) Bphen was used as ETL

WOLEDs using Bphen as electron transport layer had higher current efficiency than WOLEDs using Alq_3 because HOMO levels of Bphen is 0.8 eV lower than that of Alq_3 and electron mobility of Bphen is 100 times higher than Alq_3 due to hole blocking effect as well as fast electron transfer in Bphen layer.

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TABLE 1 Electrical and Optical Characteristics of Optimized White OLEDs

Optimized WOLED	CIE coordinate (x, y)	Current density (mA/cm^2)	Max luminance (cd/m^2)	Efficiency (cd/A)
Device E1	(0.332, 0.3191)	46.1	11900	2.30
Device E2	(0.325, 0.328)	31.4	18100	3.51
Device E3	(0.330, 0.335)	30.9	13400	3.17
Device E4	(0.338, 0.330)	18.6	15200	5.04

At 1000 cd/m^2.

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Best white OLED devices were prepared through optimization of emission layer thickness and efficient electron transport layer materials. White OLED devices E1, E2, and E3 were fabricated with combination of different thicknesses in DPVBi and MADN:DCM2-0.5 % such as 55 Å/95 Å, 65 Å/180 Å, and 75 Å/215 Å. Table 1 shows performance characteristics of optimized white OLEDs. In overall, device E1 having thin emissive layer has highest current density but lowest luminous efficiency whereas white device E3 having thicker emissive layer does different characteristics. This result can be explained hole and electron have better chance to move anode and cathode than recombination in thin thickness of emitting layer, which shows lower brightness at higher current density. In the device E3, probability of hole-electron recombination is decreased due to larger recombination region under limited electron-hole generation considering suitable DPVBi 65 Å and MADN:DCM2-0.5 % 180 Å of emissive layer thickness, therefore, device E2 was optimized better than other white OLED. Electron transport layer of optimized WOLED was finally changed from Alq_3 to Bphen on device E4 with same layer thickness. Red emissive layer's thickness was increased in 170 Å for better white emission because of fast electron injection by Bphen. That's way WOLED using Bphen have higher luminous efficiency than using Alq_3 as shown Table 1.

Table 2 shows CIEx,y coordinates, electrical and optical characteristics of optimized white OLED with increasing voltages. Current

TABLE 2 Electrical and Optical Characteristics of Optimized White OLED

Optimized WOLED	CIE coordinate (x, y)	Current density (mA/cm^2)	Luminance (cd/m^2)	Efficiency (cd/A)
6 V	(0.338, 0.330)	18	938	5.04
9 V	(0.337, 0.330)	153	67500	4.40
12 V	(0.341, 0.333)	335	1520	4.30

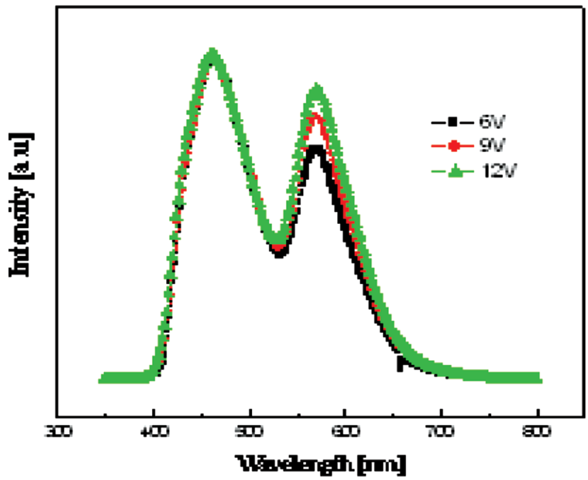

FIGURE 7 Electroluminescence of optimized white OLED with different driving voltages.

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density and luminous efficiency of optimized white OLED were 18 mA/cm^2 and 5.04 cd/A at 6 V and maximum luminous was 15200 cd/m^2 at 12 V. According to voltage increase from 6 V to 12 V, CIEx,y coordinates very slightly changes from (0.338, 0.330) to (0.341, 0.333) which means optimized white OLED was independent on bias voltages.

Electroluminescence of optimized white OLED with different bias voltages was shown in Figure 7. In UV/vis. absorption and photoluminescence spectra of DPVBi, MADN, and DCM2, combination of DPVBi's peak at 450 nm and DCM2's peak at 595 nm by Förster energy transfer from MADN to DCM2 can generate white emission. Increasing voltages, DCM2's emission peak has higher intensity comparing with DPVBi's due to shifting main recombination zone from DPVBi and MADN:DCM2-0.5 % to MADN:DCM2-0.5% layer.

CONCLUSION

Electrical and optical characteristics of white OLEDs using DPVBi of the blue layer and DCM2 doped MADN of the red layer were demonstrated. Combination of DPVBi and DCM2 0.5 % doped MADN as emitting layers generated white emission with perfectly white CIEx,y coordinates of (0.338, 0.330). Hole-electron recombination of fabricated white OLED was mainly happened between DPVBi and MADN:DCM2 layers due to DPVBi's higher HOMO levels and color coordinates of white OLED was sensitively effected by DPVBi's layer thickness but not by MADN:DCM2 layer thickness. CIEx,y coordinates of optimized white OLED was very slightly changed as increasing voltages such as from (0.338, 0.330) at 6 V to (0.341, 0.333) at 12 V. Luminous efficiency and maximum brightness of optimized white OLED were 6.2 cd/A and 15200 cd/m^2, respectively. These results indicates that WOLED fabricated with two emitting layered structure with DCM2 as red-dopant under MADN along with blue emitting layer using DPVBi holds promising potentials for backlight sources in LCDs and full color OLED displays in near future.

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