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Energy Transfer Efficiency Control from the Blue Emitting Aromatics to the Yellow Fluorescent Intramolecular Charge Transfer Type Dye for Electroluminescence

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Energy Transfer Efficiency Control from the Blue Emitting Aromatics to the Yellow Fluorescent Intramolecular Charge Transfer Type Dye for Electroluminescence

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An intramolecular charge transfer type yellow fluorescent material, (Z)-4-(2-[4,4″-bis(dimethylamino)-1,1′:4′,1″-terphenyl-2′-yl]-2′-1-cyanovinyl)benzonitrile (BDAT-CP) was synthesized for the implimentation to organic light emitting diodes (OLEDs). The molecules of the this catagory often have electro-conductivity and show high luminescence at longer wavelength in visible region. The BDAT-CP exhibited a large Stokes shift from 329 nm (UV absorption maximum) to 628 nm of photoluminescence(PL) in chloroform solvent. Solid powder PL showed yellow emission peaked at 582 nm. The device structures were indium tin oxide (ITO) / N,N′-bis-(1-naphyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB, 50 nm) / 2-methyl-9,10-bis(naphthalen-2-yl)anthracene (MADN, 30 nm) : BDAT-CP (x %) / 4,7-diphenyl-1,10-phenanthroline (BPhen, 30 nm) / Liq(2 nm) / Al. The maximum luminance measured were 14700 cd/m^2 (x = 5%), 9825 cd/m^2 (x = 10%) and 7901 cd/m^2 (x = 15%) for each BDAT-CP concentration. The luminance efficiency and quantum efficiency observed were 7.14 (x = 5%), 4.72 (x = 10%) and 3.19 (x = 15%), and 2.77 (x = 5%), 2.09 (x = 10%) and 1.59 (x = 15%), respectively. The device emitted yellow light corresponding to Commission Internationale de I'Eclairage (CIE$_{xy}$) coordinates of (0.492, 0.494) (x = 5%) and (0.523, 0.470) (x = 10%) and (0.545, 0.451) (x = 15%) at 8 V. This study showed that the energy transfer from MADN to BDAT-CP was dependent on the concentration of yellow dye, which affected the hole transporting properties of the OLED.

Keywords BDAT-CP; intramolecular charge transfer; yellow light-emitting diode

1. Introduction

Organic light-emitting devices (OLEDs) have attracted significant attention in recent years and are considered with self-emission, low-voltage operation, fast response time, wide-angle of field, high contrast and mechanical flexibility to be realized in flat-panel display industry [1–3]. White organic light-emitting diodes (WOLEDs) have application to backlights of liquid-crystal display (LCD), large area solid-state lighting and full color flat-panel display [4–7]. In order to achieve white color, triple emission layers from red, green and

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blue emitter were developed [8,9]. Another method to achieve WOLEDs is the two complementary colors way like as blue mixed with yellow or orange color [10–12]. This method has advantages such as low material cost and simple fabrication process [10,13]. For this reasons, there exists a continuing need and innovative opportunity for the development of highly efficient yellow dopant materials for various OLED display applications. Notwithstanding many previous researches, yellow emitting materials still showed low luminous and quantum efficiency and luminance maximum [14–16].

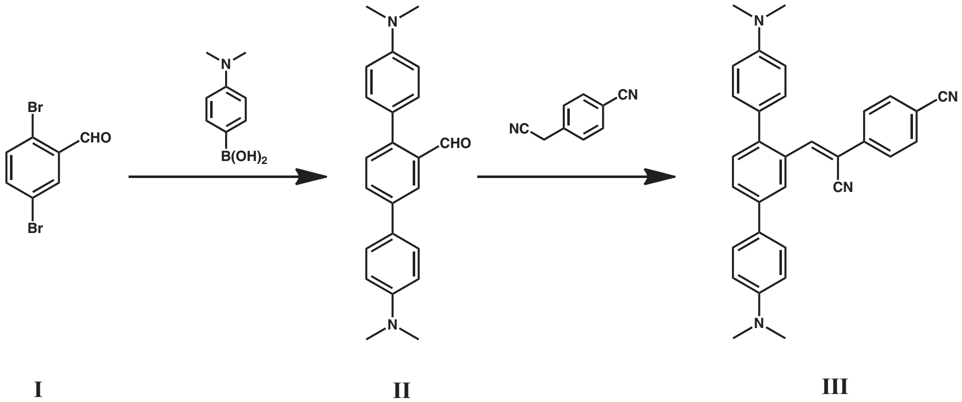
In this study, we designed and synthesized a new yellow fluorescent material, (2Z)-3-[4,4″-bis(dimethylamino)-1,1′:4′,1″-terphenyl-2′-yl]-2-(4-cyanophenyl)acetonitrile (BDAT-CP), that has intramolecular charge transfer (ICT) property for use in yellow OLEDs. In order to achieve yellow light emission with high device performance, device using BDAT-CP as a dopant was fabricated and its device performance was measured.

2. Experimental

2.1. Synthesis

Synthesis of BDAT-CP was shown in Scheme 1. All solvents involved in the experiments were reagent grade and were purified by the usual methods before use. 2,5-Dibromobenzaldehyde (I) was commercially available (Aldrich). The molecular structure of BDAT-CP was confirmed by ^{1}H and ^{13}C-NMR spectroscopy, MS (FAB) spectrum and elemental analysis.

4,4″-bis(dimethylamino)-1,1′:4′,1″-terphenyl-2′-carbaldehyde (II) was obtained from the reaction of 2,5-dibromobenzalaldehyde (I) with 4-(dimethylamino)phenylboronic acid by Suzuki coupling. The mixture of 2,5-dibromobenzalaldehyde (I) (1.0 g, 3.79 mmol), 4-(dimethylamino)phenylboronic acid (1.375 g, 8.33 mmol), $Pd(PPh_3)_4$ (175 mg), Na_2CO_3 (1.20 g) and Aliquat336 (0.693 ml) in mixed solvent of toluene (80 ml)-H_2O (50 ml)-THF (20 ml) was mildly refluxed under N_2 atmosphere with stirring for 3 hr. The reaction mixture was cooled, filtered and washed with mixture of ethyl acetate and n-hexane (1 : 5). Removal of the solvents and drying under high vacuum afforded 835 mg (2.42 mmol)



Scheme 1. Synthesis of BDAT-CP.

of the product (II) as a green solid and further purification was not required. Yield: 62%; ^{1}H-NMR (500 MHz, $CDCl_3$) δ (ppm) 10.093 (s, 1H), 8.210 (s, 1H), 7.831 (d, 1H), 7.586(d, 1H), 7.270 (d, 2H), 7.588 (d, 2H), 6.849 (d, 4H), 3.001 (s, 12H); ^{13}C-NMR (125 MHz, $CDCl_3$) δ (ppm) 193.57, 150.19, 143.91, 139.43, 133.69, 131.35, 131.11, 128.97, 128.28, 127.46, 127.40, 125.17, 124.63, 112.65, 112.01, 40.45, 40.37.

(Z)-4-(2-[4,4′′-bis(dimethylamino)-1,1′:4′,1′′-terphenyl-2′-yl]-2′-1-cyanovinyl)benzonitrile (BDAT-CP) (III) was prepared from the Knoevenagel condensation reaction of 4,4′′-bis(dimethylamino)-1,1′:4′,1′′-terphenyl-2′-carbaldehyde (II) with 4-cyanophenylacetonitrile. The mixture of 4,4′′-bis(dimethylamino)-1,1′:4′,1′′-terphenyl-2′-carbaldehyde (494 mg, 1.43 mmol), 4-cyanophenylacetonitrile (0.8149 g, 5.73 mmol) and sodium ethoxide (prepared by the reaction of 79 mg of Na with 15 ml of absolute EtOH) in 300 ml of EtOH was stirred at room temperature for 7 days. The Orange yellow solid formed in reaction mixture was filtered and washed with distillated water, EtOH and MeOH. Removal of the solvents and drying under vacuum afforded 577 mg (1.23 mmol) of the product (III) as a yellow solid. Mp. 258°C (DSC); Yield: 86.0%; ^{1}H-NMR (500 MHz, $CDCl_3$) δ (ppm) 8.405 (*d*, 1H, aromatic), 8.204 (m, 1H, aromatic), 7.858 (*s*, 1H, vinyl), 7.718 (m, 1H, aromatic), 7.615 (*d*, 2H, aromatic), 7.471 (*d*, 1H, aromatic), 7.441 (*t*, 2H, aromatic), 7.332 (*t*, 1H, aromatic), 7.305 (*d*, 2H, aromatic), 6.842 (*d*, 2H, aromatic), 6.899 (m, 2H, aromatic), 3.062 (*s*, 12H, methyl); ^{13}C-NMR (125 MHz, $CDCl_3$) δ (ppm) 150.319, 146.411, 133.018, 131.123, 130.646, 128.069, 126.674, 113.097, 112.836, 112.276, 77.522, 77.268, 77.014, 40.776, 40.661; Anal. calcd. for $C_{32}H_{28}N_4$: C: 82.02, H: 6.09, N: 11.96. Found: C: 81.93, H: 5.97, N: 11.82. MS (FAB) calcd for $C_{31}H_{29}N_3$ (M^+) m/z 468.23, found 468.23.

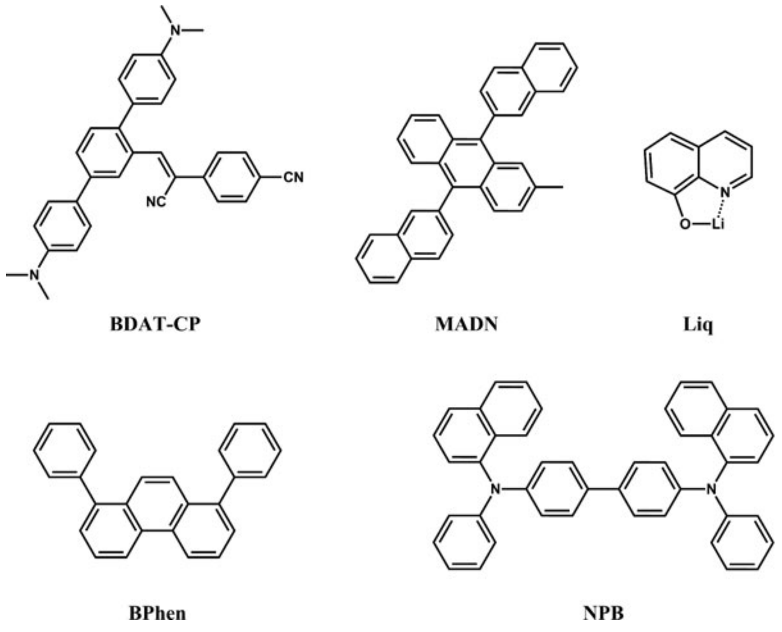


Figure 1. The molecular structure of BDAT-CP, MADN, Liq, BPhen, and NPB.

2.2. Measurement

^{1}H-NMR and ^{13}C-NMR spectra were recorded on a Varian Unity INOVA 500 spectrometer operating at 499.761 MHz and 125.701 MHz, respectively. Elemental analysis was performed on a CE instrument EA1112 analyzer. Mass spectrum (FAB-MS) was measured on JEOL, JMS-AX505WA, HP 5890 Series II Hewlett-Packard 5890A (capillary column)

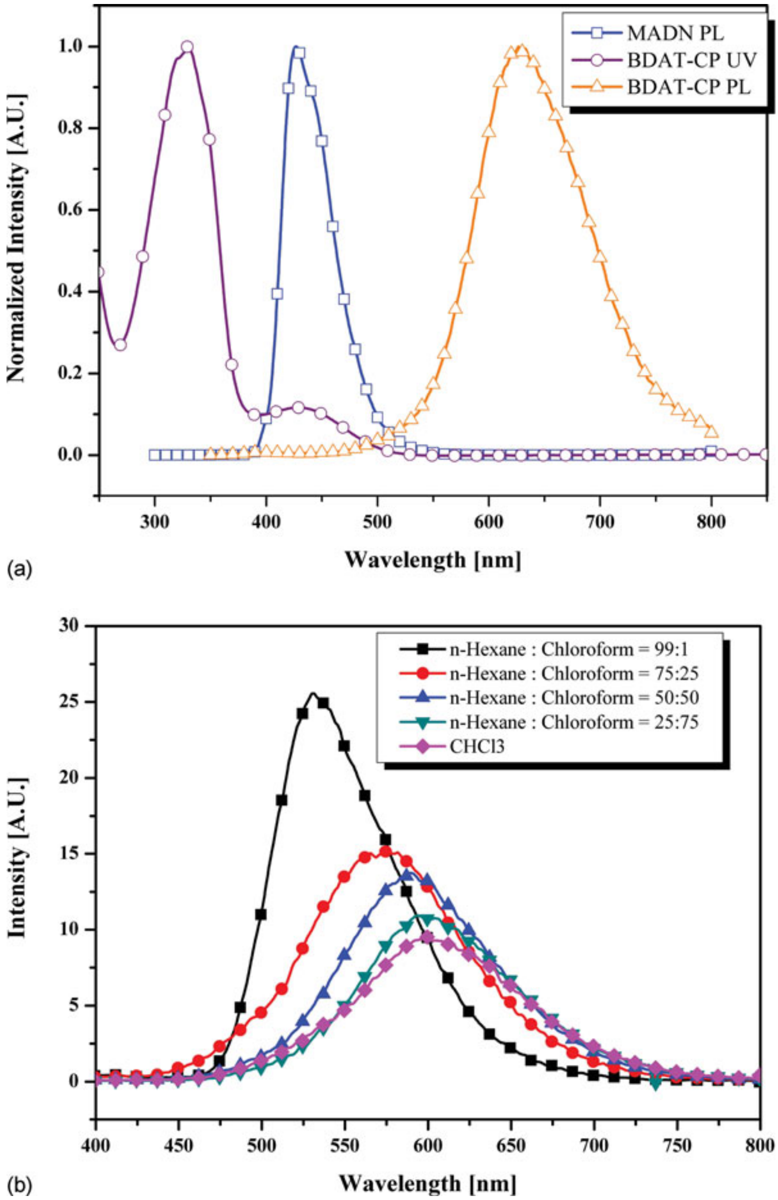


Figure 2. (a) Normalized UV-Visible absorption and PL spectra of BDAT-CP in chloroform solution and PL spectrum of MADN in film, (b) ICT property of BDAT-CP in solvents of different polarities and (c) the HOMO and LUMO energy levels of materials used in device with the structure of ITO/NPB/MADN:BDAT-CP/BPhen/Liq/Al. *(Continued)*

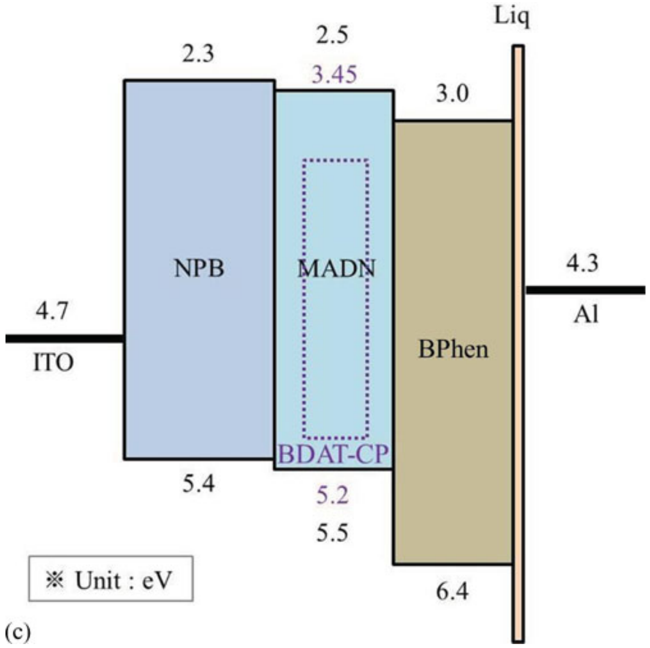


Figure 2. (Continued)

using standard conditions. The melting point was measured to be 258°C by differential scanning calorimetry (DSC) using a Seiko Exstar 7000 (DSC7020) with a scan rate of 10°C/min at the temperature range of 40 ∼ 300°C. UV-visible absorption and photoluminescence (PL) spectra were measured by HP model 8453 and Perkin Elmer LS55, respectively.

The HOMO energy level of 5.2 eV for BDAT-CP was measured by cyclic voltammetric method using a EpslianEC-2000 at a scan rate of 100 mV/s and LUMO energy level of 3.45 eV was calculated from measured HOMO energy level. The value of optical band gap was observed as 1.75 eV which was obtained from UV-visible absorption spectrum. Electroluminescence (EL) spectra and brightness-current-voltage characteristics of the device were measured by using keithley 2400, CHROMA METER CS-1000A. All the measurements were carried out at room temperature.

2.3. Fabrication of OLED

The OLED was fabricated by the high vacuum thermal deposition (8×10^{-7} torr) of organic materials onto the surface of indium tin oxide (ITO) coated in glass substrate. An ITO coated glass was cleaned in an ultrasonic bath by regular sequences: in acetone, methanol, distilled water and isopropyl alcohol. The cleaned substrate was immediately loaded into the deposition chamber in order to prevent air contamination. The device has the structure of ITO (100 nm)/NPB (50 nm)/MADN:BDAT-CP (x %) (30 nm)/BPhen (30 nm)/Liq (2 nm)/Al (100 nm), in which NPB, BDAT-CP, BPhen and Liq were used as a hole transporting layer (HTL), an emitter layer (EML), a hole blocking and electron transporting layer (HBL, ETL) and an electron injection layer (EIL), respectively. Figure 1 showed molecular structures of BDAT-CP, MADN, Liq, BPhen, and NPB for fabrication of devices.

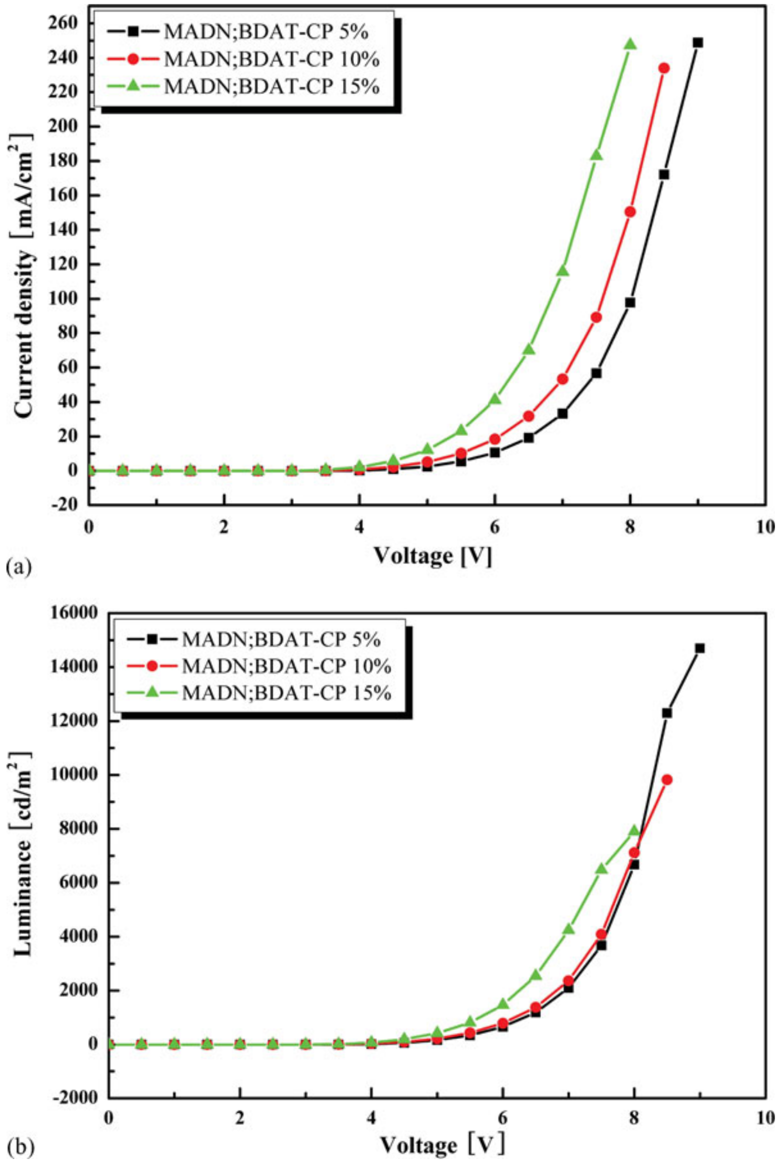


Figure 3. (a) Current density vs. voltage and (b) luminance vs. voltage characteristics with variation of doping concentration.

3. Results and Discussion

Figure 2(a) showed UV absorption and PL spectra of BDAT-CP in chloroform and PL spectrum of MADN in film. They were used as the dopant and the host materials, respectively. The UV absorption spectrum of BDAT-CP showed a peak at 329 nm and shoulder at 429 nm. The PL spectrum of BDAT-CP showed yellow emission with a maximum peak at 620 nm. The intramolecular charge transfer from amino groups to cyano groups made large Stokes shift and the emission usually came from the twisted excited state [17,18].

The large Stokes shift hardly caused an overlap between absorption and emission bands. Although the two bands were not fully overlapped, MADN was used as the host material for OLEDs using BDAT-CP as the dopant. The overlap between UV-visible absorption band of BDAT-CP and the PL band of MADN was favorable for the Förster energy transfer from MADN to BDAT-CP. Figure 2 (b) showed photoluminescence(PL) of BDAT-CP in the Chloroform and n-Hexane mixture. The fluorescence wavelength and intensity were changed

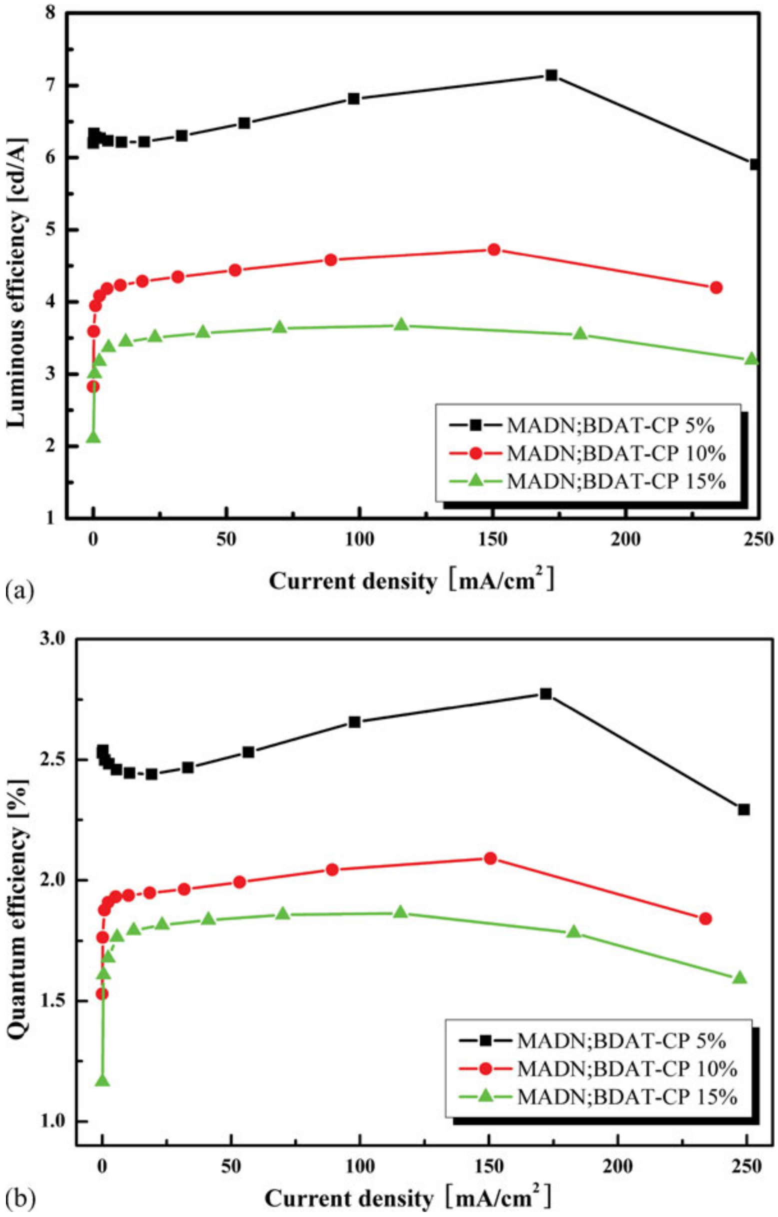


Figure 4. (a) Current density vs. luminous efficiency and (b) quantum efficiency vs. current density characteristics with variation of doping concentration.

by ratio of n-hexane in the solvent. The molecules such as BDAT-CP and (Z)-3-(4,4″-bis(dimethylamino)-[1,1′:4′,1″-terphenyl]-2′-yl)-2-phenylacrylonitrile(BDAT-P) [14] have extended conjugation of electron donating (donor) and accepting (acceptor) moieties within molecule show a strong intramolecular charge transfer (ICT) character. PL wavelength of a fluorophore with ICT character can be controlled by modification of donor or acceptor moiety which changes the degree of ICT. As the electron withdrawing ability in acceptor is increased, intramolecular charge separation in excited state is enhanced. As concentration of chloroform increased, PL intensity decreased, which was due to increase in radiationless transition caused by extended twisted state and energy orbital overlap. The dramatic

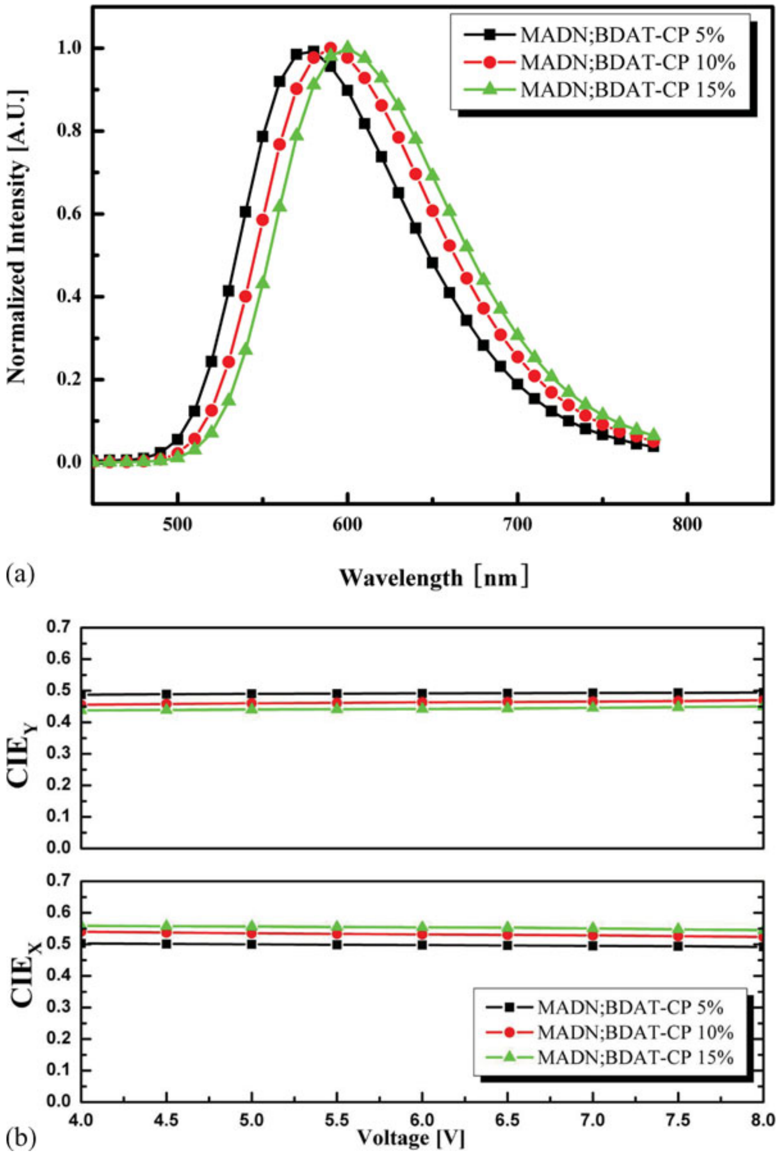


Figure 5. (a) Normalized EL spectra, (b) measured CIExy coordinates and (c) chromaticity diagram measured with variation of doping concentration. *(Continued)*

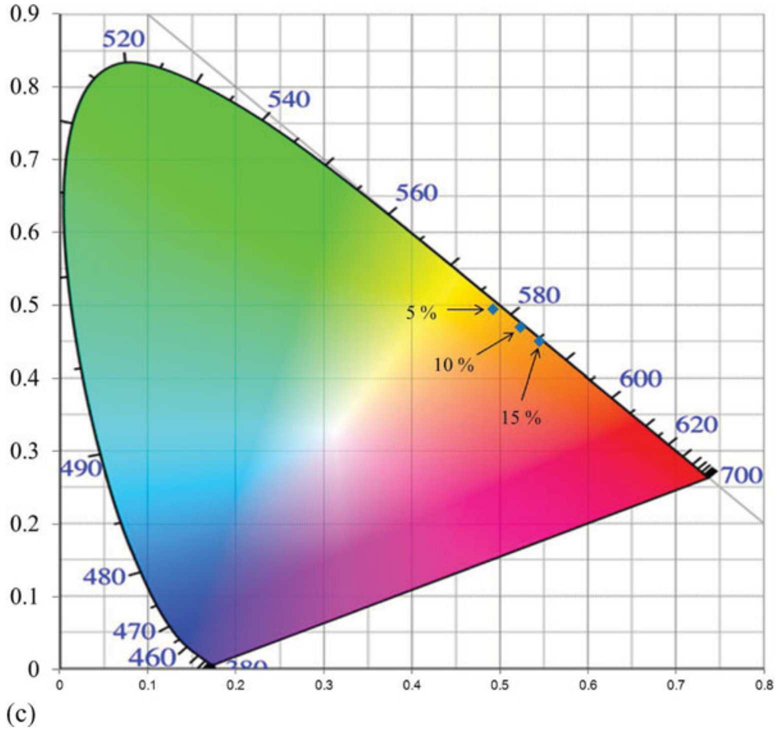


Figure 5. (Continued)

bathochromic shift observed from BDAT-CP compared to BDAT-P can be easily understood by stronger ICT character. Figure 2 (c) represented the HOMO and LUMO energy levels of the materials used for the device with the structure of ITO/NPB/MADN:BDAT-CP/BPhen/Liq/Al. It showed that the correlation of energy band gap of MADN with that of BDAT-P was suitable to Förster energy transfer from MADN to BDAT-CP.

Figure 3(a) and (b) showed the current density vs. voltage and current density vs. luminance of BDAT-CP doped OLEDs at various doping concentration of 5, 10 and 15%. The luminance maxima were 14700 cd/m^2 (x = 5%), 9825 cd/m^2 (x = 10%) and 7901 cd/m^2 (x = 15%), respectively. As doping concentration increased from 5% to 15%, luminance was decreased, which was ascribed to the mismatching of holes and electrons to make excitons. The holes can travel to the Bphen because the BDAT-CP have higher HOMO level than MADN.

As shown in Fig. 4(a) and (b), device doped with the new yellow fluorescent BDAT-CP represented suitable luminous efficiency and quantum efficiency for OLEDs. The maximum luminous and quantum efficiency values were observed to be 7.14 (x = 5%), 4.72 (x = 10%) and 3.19 (x = 15%), and 2.77 (x = 5%), 2.09 (x = 10%) and 1.59 (x = 15%), respectively. Based on Figs 3 and 4, EL emission peak was much more shifted to short wavelength compared to that of PL. The device with doping concentration of 5% was observed to be the most stable and efficient.

Figure 5(a) showed the EL spectra of BDAT-CP with variation of doping concentration (x = 5, 10 and 15%). EL emission peaks were shifted to longer wavelength as doping concentration was increased and they were measured to be 576 (x = 5%), 590

Table 1. EL characteristics of yellow OLEDs, including the reported maximum luminance (*L*max), luminous efficiency (*è*), and CIE

Material/method	*L*max (cd/m^2)	*è* (cd/A)	CIE (*x*, *y*)	ref
BDAT-CP	14,700	7.13[a]	(0.49, 0.49)	This work
4-(dicyanomethylene)-2-*tert*-butyl-6-(*p*-diphenylaminostyryl)-4*H*pyran (DCTP) in Alq3 (2 wt%)	19,383	5.3[b]	(0.47, 0.51)	19
rubrene in TPDS (5 mol%)		1.87[c]		20
rubrene in o-TPD:m-OXD (0.4 wt%)		2.30[d]		21
rubrene in NPB (5 wt%)	27,700	4.20[e]		22
rubrene in Alq_3 (10 mol%)	4,200	1.58[e]		23

[a] Maximum efficiency. [b] At current density $J = 20$ mA/cm^2. [c] At current density $J = 10$ mA/cm^2. [d] At current density $J = 100$ mA/cm^2. [e] Maximum efficiency.

(x = 10%) and 598(x = 15%) nm, respectively. Yellow light was more pronounced with the increase of doping concentration. Many other of EL properties observed from the device with doping concentration of 5% were superior to those observed from the device with doping concentration of 10% and 15%. Yellow light emission is not for full color display but for WOLEDs so it is assumed that doping concentration of 5% is more favorable than that of 10 and 15%. As shown in Fig. 5(b), Commission Internationale de I'Eclairage (CIE_{xy}) coordinates of device were measured to be (0.492, 0.494) (x = 5%) and (0.523, 0.470) (x = 10%) and (0.545, 0.451) (x = 15%) at 8 V, respectively. Observed CIE_{xy} coordinates represented that the device with doping concentration from 5 to 15% emitted yellowish green EL emission, which was shown in Fig. 5(c). This yellow emitting OLED made with BDAT-CP are concentration dye concentration dependent, which is influenced by hole-transporting properties. The energy transfer of MADN to BDAT-CP is very efficient.

As mentioned above, BDAT-CP has better properties than other existing yellow fluorescent materials. Table 1 showed Characteristics of BDAT-CP and several established yellow OLEDs, Including the reported maximum luminance, luminous efficiency, and CIE coordinates. It showed that BDAT-CP has higher luminous efficiency. But luminance maximum is normal level as compared with other materials. However, researches like alterations of device structures and improvement of external quantum efficiency will be able to easily solve this problem.

4. Conclusions

A yellow fluorescent material, (2Z)-3-[4,4″-bis(dimethylamino)-1,1′:4′,1″-terphenyl-2′-yl]-2-(4-cyanophenyl)acetonitrile (BDAT-CP), was synthesized for use in OLED application. This fluorescent material has two acceptors that made intramolecular charge transfer (ICT) characters. As the electron withdrawing ability in acceptor is increased, intermolecular charge separation in excited state is enhanced, and thus, dramatic shift in PL emission of BDAT-CP to much longer wavelength compared to BDAT-P can be achieved. The maximum luminous and quantum efficiency of device that the devices fabricated by BDAT-CP as emitting layer material was measured to be 7.14 cd/A and quantum efficiency of 2.77%, respectively. Based on both of device efficiency and stability of the efficiency, device

A which was using BDAT-CP resulted in the better efficient device than device B which was using BDAT-P. Therefore, it can be concluded that a newly synthesized fluorescent BDAT-CP may be a good candidate for use in fabrication of WOLEDs.

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