High Efficiency White Organic Light-Emitting Diodes from One Emissive Layer

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In this study, two-wavelength white organic light-emitting devices (WOLEDs) were fabricated using a single doped layer, which was obtained by forming a recombination zone in a single emissive layer. In this emissive layer, both blue emission and yellow emission were obtained using a 4,4′-bis(2,2′-diphenylethen-1-yl)biphenyl (DPVBi) host doped with a red fluorescent dye, 4-(dicyanomethylene)-2-*tert*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4*H*-pyran (DCJTB). The white light resulted from the excitation in this single emissive layer by controlling the concentration of fluorescent dye in the emissive layer. In addition, a higher efficiency could be obtained using organic layers/Li/Al in the WOLED structure instead of organic layers/LiF/Al. The electroluminescent (EL) spectrum of the device was not sensitive to the driving voltage of the device. The obtained WOLEDs showed a maximum luminance of approximately $64,200\,cd/m^2$ and the device began to emit light (at luminance of $0.1\,cd/m^2$) at 3.4 V. At a luminance of approximately $100\,cd/m^2$, the external quantum and the power efficiencies were 2.47% and 4.27 lm/W, respectively.   [DOI: 10.1143/JJAP.46.806]

KEYWORDS: WOLEDs, two wavelength, one emissive layer, doping

1. Introduction

Organic light-emitting devices (OLEDs), using either small molecules or polymers, have attracted considerable attention due to their ease of production, low cost, low operating voltage, wide viewing angles, tunability of the color emission, rapid response time, and compatibility with flexible substrates, etc.[1–3] Among the OLEDs, white OLEDs (WOLEDs) have become particularly attractive due to their potential applications as backlight sources in full-color displays combined with color filters,[4,5] white solid-state light sources,[6] etc.

White light-emission can be obtained by mixing two complementary colors (e.g., red/bluish-green, blue/orange, or green/magenta) or three primary colors (red, green, and blue) from small-molecules and/or polymers.[6–14] In order to obtain white emission from an OLED, the excitation of more than one molecular species is necessary because the luminescence of a single organic molecule does not span the entire visible spectrum. To realize white OLEDs with stable performance characteristics, the unchangeable color coordinates and stable exciton recombination formation with respect to the driving voltage in the emitting layers are essential.[15] There are several reports on WOLEDs, which suggested WOLEDs using a multilayer structure with two or more emitting layers and with a hole and electron blocking layer. However, their results appear to show serious problems such as variations in the Commission Internationale de l'Eclairage (CIE) chromaticity coordinates with the change in current (or voltage) and low luminance efficiency even with an optimum current density.[13,14]

In this study, one-layer emitting WOLEDs with two wavelengths composed of glass/tin-doped indium oxide (ITO; 150 nm)/4,4′,4″-tris(2-naphthylphenyl-1-phenylami-no)triphenylamine (2-TNATA; 60 nm)/*N*,*N*′-bis(1-naph-thyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB; 15 nm)/4,4′-bis(2,2′-diphenylethen-1-yl)biphenyl (DPVBi):4-(dicyanomethylene)-2-*tert*-butyl-6-(1,1,7,7-tetramethyljulo-lidyl-9-enyl)-4*H*-pyran (DCJTB; 20 nm, x%)/tris(8-quinoli-nolato) aluminum(III) (Alq$_3$; 35 nm)/lithium (Li) or lithium

fluoride (LiF; 1.0 nm)/aluminium (Al; 100 nm) ($x = 0–0.3$) were fabricated by forming an exciton recombination zone in a single emissive layer, generating a blue and yellow color. One of aims in this study was to achieve WOLEDs with stable performance characteristics without affecting its bipolar transporting property to obtain almost unchangeable color coordinates in white emission for our WOLED structure similar to other studies on WOLEDs with a single emissive layer.[16] The other was to fabricate a balanced white light with CIE chromaticity coordinates of (0.33, 0.33) by controlling doping levels of red dopant DCJTB in the blue host DPVBi emitting two different colors consisting of blue and yellow.

2. Experimental Procedure

Figures 1(a) and 1(b) show a schematic diagram of the WOLEDs fabricated in this study, and the molecular structures of the devices, respectively. OLED devices composed of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, x%)/Alq$_3$ (35 nm)/Li or LiF (1.0 nm)/Al (100 nm) ($x = 0–0.3$) were fabricated using a vapour phase deposition method. The ITO (anode) coated on glass (0.7 mm) had a sheet resistance of approximately $12–15\,\Omega$/square. 2-TNATA and NPB act as the hole injection layer (HIL) and the hole transport layer (HTL), respectively. DPVBi doped with DCJTB (red fluorescent dye) is the blue and yellow emissive layer. Alq$_3$, Li or LiF, and Al function as the electron transport layer (ETL), the electron injection layer (EIL), and the cathode, respectively. Before loading the ITO glass into the deposition chamber, the ITO coated on glass was cleaned with detergents and deionized water, and dried in the oven at $120\,°C$ for 2 h. The devices were fabricated by evaporating organic materials, Li or LiF, and Al onto the ITO coated on glass substrate at deposition rates of $0.03–0.06$, $0.01–0.02$, and $0.03–0.2$ nm/s, respectively and sequentially, at a pressure $< 1 \times 10^{-6}$ Torr. Doping in each emitting layer was achieved by co-evaporating both the host and the dopant. The emissive active area of the devices was $2 \times 2\,mm^2$.

The electroluminescent (EL) spectra of the as-fabricated devices were measured by optical emission spectroscopy (PCM-420, SC Technology) and the CIE chromaticity

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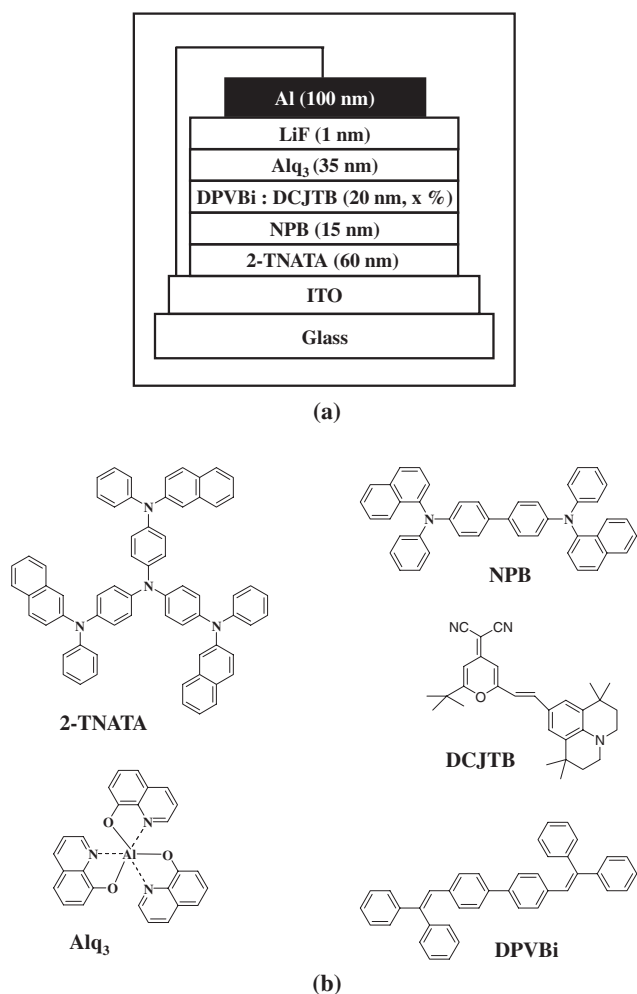*Corresponding author. E-mail address: gyyeom@skku.edu

(a)

(b)

Fig. 1. (a) Schematic diagram of the WOLED device investigated. (b) Molecular structures.
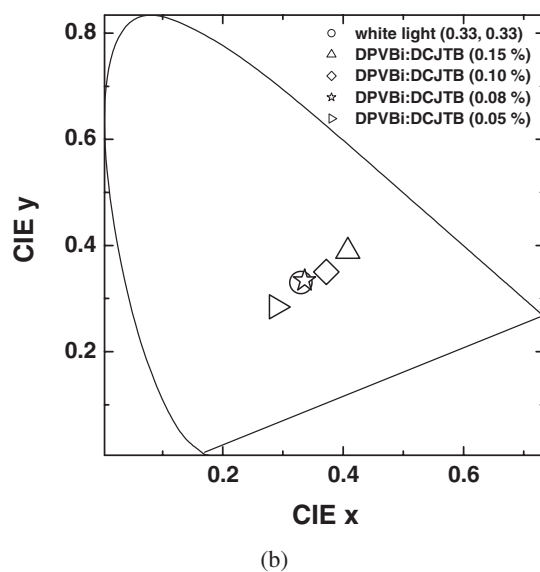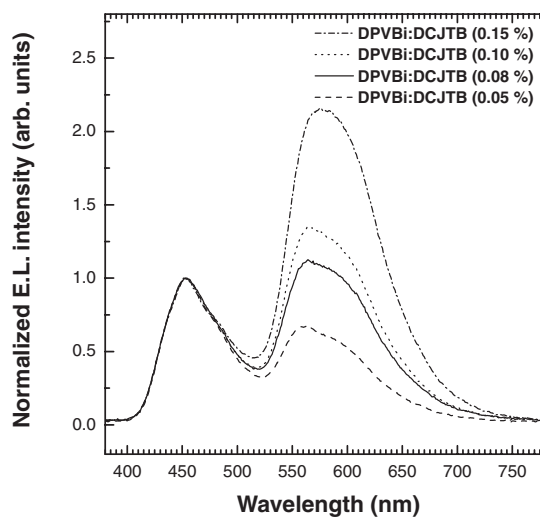

(a)

(b)

Fig. 2. (a) Normalized electroluminescent spectra and (b) CIE chromaticity diagram of the devices consisting of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, $x\%$)/Alq$_3$ (35 nm)/Li (1.0 nm)/Al (100 nm) ($x = 0.15$, 0.1, 0.08, and 0.05, respectively) at the luminance of $100 \, \text{cd/m}^2$.

coordinates were determined using a chroma meter (CS-100, Konica Minolta). The current-voltage-luminance characteristics were measured using a source-measure unit (2400, Keithley Instrument), and the intensities from the emission of OLEDs devices were measured using the photocurrent induced on silicon photodiodes using a picoammeter (485, Keithley Instrument).

3. Results and Discussion

In the OLED devices, the addition of dopants such as the DCM-series (for example: DCJTB, DCM2,[14] etc.) to organic materials such as Alq$_3$ alters the peak intensity of the emission spectra to a longer wavelength, and therefore shows a red-shift with increasing dopant concentration as a result of the solid-state solvation effect.[17] However, it was reported that for the doping of Alq$_3$ with DCM2, the external quantum efficiency (η_{ext}) and the power efficiency (η_{PE}) decreased with increasing doping concentration $> 0.5\%$ as a result of the decrease in hole mobility due to concentration quenching and hole trapping processes.[18] The addition of the red dopant, DCJTB, to the blue host DPVBi had a similar effect. Therefore, in this study, the concentration of DCJTB was kept to $<0.5\%$. In a previous experiment, when the DCJTB concentration was varied as 0, 0.1, 0.2, and 0.3%, the EL spectra showed white light-like characteristics for the

device containing 0.1% DCJTB added to DPVBi (data not shown). Accordingly, the DCJTB concentration was varied from 0.05 to 0.15%. In addition, in order to obtain an improved luminous efficiency, Li was used as the EIL layer and a device composed of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, $x\%$)/Alq$_3$ (35 nm)/Li (1.0 nm)/Al (100 nm) was fabricated for white light emission as shown in Fig. 1(a).

Figures 2(a) and 2(b) show the EL spectra and CIE chromaticity diagram of the WOLED devices fabricated as a function of the DCJTB doping concentration (0.05–0.15%), respectively. As shown in Fig. 2(a), The EL spectra showed two distinctive emission bands consisting of a blue band originating from DPVBi and yellow band originating from DCJTB. The blue band showed a maximum peak (λ_{max}) at 453 nm regardless of the dopant concentration. In the case of the yellow band, when the DCJTB concentration was $<0.1\%$, the peak (λ_{max}) was observed at near 565 nm.
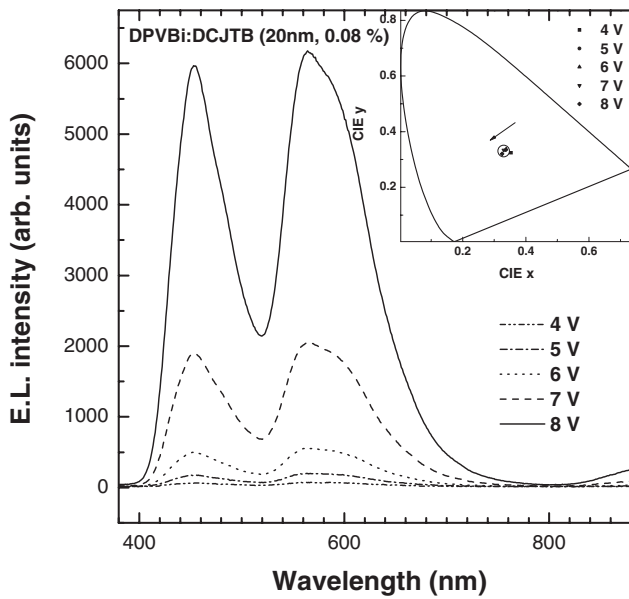
Fig. 3. Electroluminescent spectra of the device consisting of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, 0.08%)/Alq$_3$ (35 nm)/Li (1.0 nm)/Al (100 nm) as a function of the voltage (4 to 8 V). The inset shows the corresponding CIE chromaticity diagram of the same device. The white circle point indicates the CIE chromaticity coordinates of (0.33, 0.33).

Table I. Doping level (%) of DCJTB-doped DPVBi and the current density (*J*), power efficiency (η_{PE}), external quantum efficiency (η_{ext}), CIE coordinates, and maximum luminance (L_{max}) of WOLEDs.

	Device 1	Device 2	Device 3	Device 4	Device 5
Electron injection layer	Li	Li	Li	Li	LiF
Doping level of DCJTB (wt %)	0.15	0.1	0.08	0.05	0.08
J (mA/cm^2)[a]	1.77	1.80	1.79	1.74	1.72
η_{PL} (lm/W)[a]	4.32	3.60	4.27	4.06	2.93
η_{ext} (%)[a]	2.23	2.17	2.47	2.09	2.07
CIE coordinates[a]	0.41, 0.39	0.37, 0.35	0.34, 0.33	0.29, 0.28	0.34, 0.33
L_{max} (cd/m^2)	62,000	60,500	64,200	59,200	59,300

a) At 100 cd/m^2.

However, when the dopant concentration was increased to 0.15%, the λ_{max} was shifted to 575 nm. The blue-shift of DCJTB (100% DCJTB itself has a peak at 630 nm) obtained in these experiments through the doping of DCJTB with an extremely low concentration of <0.15% appears to be related to the strong solid state solvation effect as reported by other researchers,[19] even though an increase in the dopant concentration from 0.05 to 0.15% shows color shift towards red, possibly due to more energy transfer from DPVBi to the dopant DCJTB. An optimized white emission could be obtained at a DCJTB concentration of 0.08% due to the change in color with increasing DCJTB doping concentration, as shown by the CIE chromaticity of Fig. 2(b). When the dopant concentration was >0.08%, the coordinates deviate from the optimum white emission due to too much energy transfer from the host, DPVBi, to the dopant, DCJTB. Therefore, in these experiments, white light emission could be obtained in a single emissive layer through energy transfer from the blue host, DPVBi, to red dopant, DCJTB, by controlling the DCJTB concentration in DPVBi.

Figure 3 shows EL spectra of the optimized white OLED device (0.08% DCJTB) as a function of the voltage, and the insert shows its CIE chromaticity diagram. As shown in Fig. 3, the EL intensities increased similarly for both blue emission and yellow emission with increasing voltage from 4 V (4.2 cd/m^2 at 0.09 mA/cm^2) to 8 V (2,800 cd/m^2 at 42.7 mA/cm^2). Therefore, no significant color shift could be observed. In addition, as shown the CIE chromaticity diagram inserted, the chromaticity measured as a function of voltage remained similar. The no significant change in CIE chromaticity obtained in these results could be explained by two reasons. First, the device is composed of a single emission layer and two neighboring blocking layers.

Therefore, the electrons/holes entering the device can be effectively confined and recombined within the emission layer due to the presence of the two neighboring electron-blocking and hole-blocking layers.[20–22] Second, all WO-LEDs investigated have comparatively low doping-concentrations. Less exciton quenching would have occurred and much less color variation was observed with increasing current density.[23,24]

Table I shows the photometric properties of the devices as a function of the DCJTB doping levels. Devices 1, 2, 3, and 4 correspond to a DCJTB doping concentration of 0.15, 0.1, 0.08, and 0.05%, respectively. Device 5 was fabricated with the structure of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, 0.08%)/Alq$_3$ (35 nm)/LiF (1.0 nm)/Al (100 nm). Therefore, the Li of the EIL was replaced with LiF for the device containing 0.08% DCJTB doped DPVBi in order to compare the EIL properties of Li with the conventionally used LiF. As shown in the table, the maximum luminance (L_{max}) of devices 1–5 were 62,000, 60,500, 64,200, 59,200, and 59,300 cd/m^2, respectively, and the turn-on voltages (at the luminance of 0.1 cd/m^2) of the devices were 3.2, 3.4, 3.4, 3.2, and 3.6 V, respectively. The external quantum efficiencies (η_{ext}) at 100 cd/m^2 were 2.23, 2.17, 2.47, 2.09, and 2.07%, respectively, and the power efficiencies (η_{PE}) were 4.32, 3.60, 4.27, 4.06, and 2.93 lm/W, respectively. As shown in the table, device 3 containing DPVBi doped with 0.08% DCJTB showed best white balance, and the CIE chromaticity coordinate was (0.34, 0.33).

Figure 4 shows (a) the current density–voltage–luminance (*J*–*V*–*L*), (b) the external quantum efficiency–current density–power efficiency (η_{ext}–*J*–η_{PE}) for devices 3 and 5 which used Li and LiF as the EIL, respectively, at the same doping concentration of 0.08% DCJTB in DPVBi. As shown in Fig. 4(a), device 3 showed a lower turn-on voltage and a higher luminance at a lower voltage compared with device 5. In addition, as shown in Fig. 4(b) and Table I, the highest η_{PE} and η_{ext} of device 3 was 4.27 lm/W and 2.47% (at 1.79 mA/cm^2 and 100 cd/m^2), respectively, while the highest η_{PE} and η_{ext} of device 5 was 2.93 lm/W and 2.07% (at 1.72 mA/cm^2 and 100 cd/m^2), respectively. Therefore, higher efficiencies could be also obtained with device 3 compared with device 5. As shown in Table I, the CIE coordinates were the same at (0.34, 0.33). Therefore, both devices showed white light emission close to the ideal white
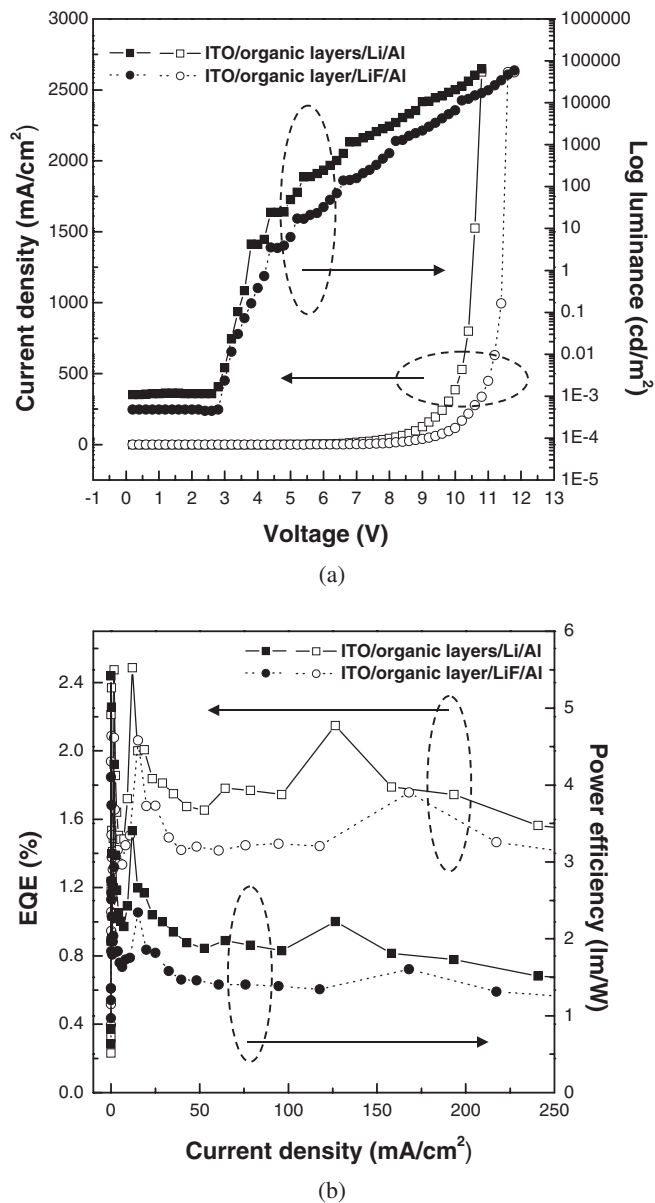
(a)

(b)

Fig. 4. (a) The current density–voltage–luminance and (b) the external quantum efficiency–current density–power efficiency characteristics of the device consisting of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, 0.08%)/Alq$_3$ (35 nm)/Li or LiF (1.0 nm)/Al (100 nm).

light emission of (0.33, 0.33). As a result, the use of Li as the EIL instead of LiF showed not only a higher η_{ext} and η_{PL} but also a lower turn-on voltage, which is possibly due to the lower work function.

4. Conclusions

In this study, two-wavelength WOLEDs were fabricated using a single doped layer in the device structure consisting of glass/ITO (150 nm)/2-TNATA (60 nm)/NPB (15 nm)/DPVBi:DCJTB (20 nm, x%)/Alq$_3$ (35 nm)/Li (1.0 nm)/Al

(100 nm). White light could be obtained in a single emissive layer by varying the doping concentration of DPVBi in DCJTB, and a higher luminous efficiency could be obtained using Li as the EIL instead of the conventionally used LiF. By using a DCJTB (red dopant) concentration of 0.08% in the blue host DPVBi, the peak heights of the blue emission and yellow emission of the EL spectra were similar and the CIE chromaticity coordinate was (0.34, 0.33), which is close to the ideal value of (0.33, 0.33). The optimum device showed a maximum luminance of 64,200 cd/m^2 and a turn-on voltage of 3.4 V. The current density, external quantum efficiency, and power efficiency at 100 cd/m^2 were 1.79 mA/cm^2, 2.47%, and 4.27 lm/W, respectively. This device showed no significant change in the EL spectra and CIE chromaticity coordinates with increasing driving voltage.

1) H. Kanno, R. J. Holmes, Y. Sun, S. K. Cohen, and S. R. Forrest: Adv. Mater. **18** (2006) 339.
2) C. C. Changa, J. F. Chen, S. W. Hwang, and C. H. Chen: Appl. Phys. Lett. **87** (2005) 253501.
3) W. Zhao and J. M. White: Appl. Phys. Lett. **87** (2005) 103503.
4) H. Kanno, Y. Sun, and S. R. Forrest: Appl. Phys. Lett. **86** (2005) 263502.
5) P. Destruel, P. Jolinat, R. Clergereaux, and J. Farenc: J. Appl. Phys. **85** (1999) 397.
6) B. W. D'Andrade and S. R. Forrest: Adv. Mater. **16** (2004) 1585.
7) C. W. Tang, S. A. Van Slyke, and C. H. Chen: J. Appl. Phys. **65** (1989) 3610.
8) J. Kido, W. Ikeda, M. Kimura, and K. Nagai: Jpn. J. Appl. Phys. **35** (1996) L394.
9) J. Kido, M. Kimura, and K. Nagai: Science **267** (1995) 1332.
10) R. H. Jordan, A. Dodabalapur, M. Strukelj, and T. M. Miller: Appl. Phys. Lett. **68** (1996) 1192.
11) R. S. Deshpande, V. Bulovic, and S. R. Forrest: Appl. Phys. Lett. **75** (1999) 888.
12) C. W. Ko and Y. T. Tao: Appl. Phys. Lett. **79** (2001) 4234.
13) G. Li and J. Shinar: Appl. Phys. Lett. **83** (2003) 5359.
14) H. S. Oh, J. Y. Cho, and S. B. Yoon: J. Korean Phys. Soc. **45** (2004) 460.
15) M. Berggren, A. Dodabalapur, R. E. Slusher, and Z. Bao: Nature **389** (1997) 466.
16) X. Y. Zheng, W. Q. Zhu, Y. Z. Wu, X. Y. Jiang, R. G. Sun, Z. L. Zhang, and S. H. Xu: Displays **24** (2003) 121.
17) V. Bulovic, A. Shoustikov, M. A. Baldo, E. Bose, V. G. Kozlov, M. E. Thompson, and S. R. Forrest: Chem. Phys. Lett. **287** (1998) 455.
18) K. O. Cheon and J. Shinar: Appl. Phys. Lett. **81** (2002) 1738.
19) V. Bulovic, R. Deshpande, M. E. Thompson, and S. R. Forrest: Chem. Phys. Lett. **308** (1999) 317.
20) C. H. Chuen, Y. T. Tao, F. I. Wu, and C. F. Shu: Appl. Phys. Lett. **85** (2004) 4609.
21) C. H. Chuen and Y. T. Tao: Appl. Phys. Lett. **81** (2002) 4499.
22) Y. Shao and Y. Yang: Appl. Phys. Lett. **86** (2005) 073510.
23) Y. T. Tao, C. W. Ko, and E. Balasubramaniam: Thin Solid Films **417** (2002) 61.
24) J. H. Jou, Y. S. Chiu, R. Y. Wang, H. C. Hu, C. P. Wang, and H. W. Lin: Org. Electron. **7** (2006) 8.