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Diphenylaminofluorene-Based Donor-Acceptor Molecules for Blue Light-Emitting Diodes

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*A series of blue fluorescent 2-diphenylaminofluorene-7-ylstyrene derivatives end-capped with electron-accepting groups, such as methylpyridine, qunoline, isoqunoline, phenanthridine were synthesized and characterized. To examine the electroluminescent properties of these molecules, multilayer devices were fabricated. The device **D** exhibited highly efficient blue emission with a luminous efficiency (LE), power efficiency (PE) and CIE*x,y *coordinates of 7.13 cd/A, 2.80 lm/W at 20 mA/cm^2 and (0.148, 0.191) at 7.0 V, respectively. In addition, the device **A** showed efficient deep-blue emission with a LE, PE and CIE*x,y *coordinates of 6.85 cd/A, 2.68 lm/W at 20 mA/cm^2, and (0.147, 0.152) at 7.0 V, respectively.*

Keywords Blue fluorescent materials; deep-blue emitting materials; diphenylaminofluorene derivatives; organic light-emitting diode

Introduction

Full-color organic light-emitting diode (OLED) display applications require a set of primary RGB emitters with sufficiently high luminous efficiency and properly balanced color chromaticity, as well as adequate operational stability. Recently, OLED doped with blue emitters, such as anthracene, fluorene, carbazole and pyrene derivatives, were reported [1–8]. Nevertheless, the efficiency, lifetime and color purity of these blue emitters should be further improved.

Many emissive electron donor-acceptor type molecules have been reported, and EL colors spanning the entire visible region, including green [9], yellow [10], and red [11], have been achieved from OLEDs based on these materials. However, only a few electron donor-acceptor type molecules have been developed as blue emitters in OLEDs [12–13]. Therefore, more systematic studies of the structure-electroluminescent property relationships of electron donor-acceptor type molecules will be essential for the design of next-generation ambipolar emissive materials for OLEDs.

This paper reports the synthesis and electroluminescent properties of four donor-acceptor (D-A) type blue-emitting molecules containing 2-diphenylaminofluorene as a

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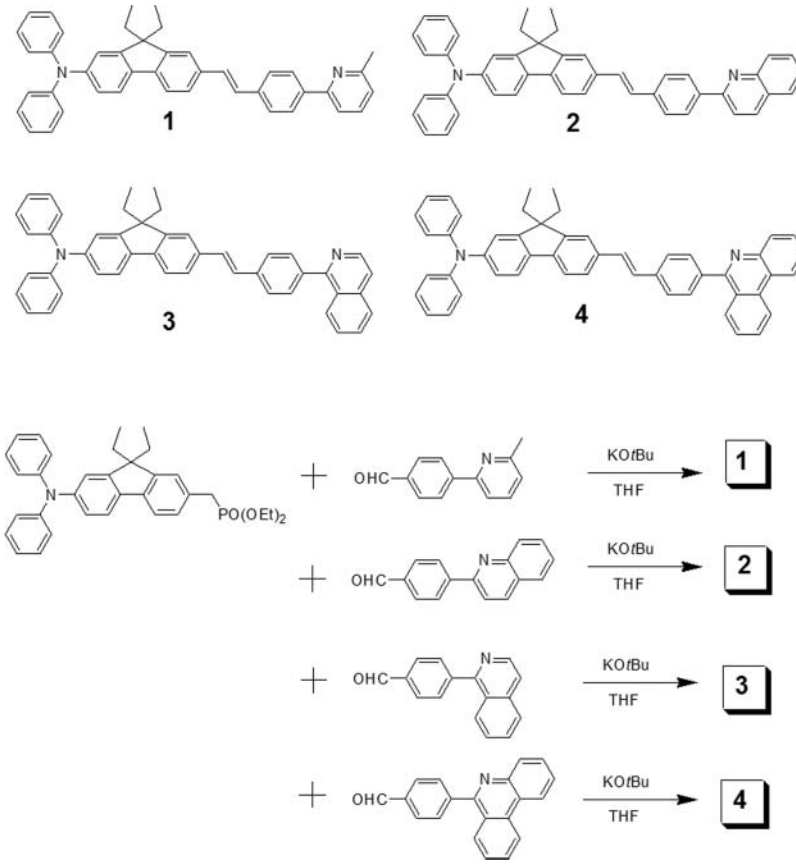

Figure 1. Synthesis of blue fluorescent materials (**1–4**).

electron donor and the various pyridine derivatives, including methylpyridine, qunoline, isoqunoline, and phenanthridine, as electron acceptors. Figure 1 shows the molecular structures of the 2-diphenylaminofluorene molecules, 7-(4-(6-methylpyridin-2-yl)styryl)-9,9-diethyl-N,N-diphenyl-9H-fluoren-2-amine **(1)**, 7-(4-(quinolin-2-yl)styryl)-9,9-diethyl-N,N-diphenyl-9H-fluoren-2-amine (**2**), 7-(4-(isoquinolin-1-yl)styryl)-9,9-diethyl-N, N-diphenyl-9H-fluoren-2-amine (**3**), and 7-(4-(phenanthridin-6-yl)styryl)-9,9-diethyl-*N,N*-diphenyl-9H-fluoren-2-amine (**4**). This paper reports the electron acceptor structure property relationships in D-A molecules relevant to the photophysical properties, HOMO/LUMO energy levels and solid-state electronic devices.

Experimental

Synthesis

All commercially available reagents were purchased from Aldrich and TCI and used without further purification. (7-(Diphenylamino)-9,9-diethylfluoren-2-yl)methylphosphonate [1], 4-(6-methylpyridin-2-yl)benzaldehyde [14], 4-(quinolin-2-yl)benzaldehyde [15], 4-(isoquinolin-1-yl)benzaldehyde [15], and 4-(phenanthridin-6-yl)benzaldehyde [15] was

synthesized by elsewhere reported procedures. The blue compounds were properly purified and dried. ^{1}H and ^{13}C NMR spectra were recorded on a Varian (Unity Inova 300Nb) spectrometer. Fourier transform infrared (FT-IR) spectra were recorded using a Buker VERTEX70 FT-IR spectra spectrometer. Low- and high-resolution mass spectra were measured using a JEOL JMS-AX505WA spectrometer in the Fast atom bombardment (FAB) mode and a JMS-T100TD (AccuTOF-TLC) spectrometer in the positive-ion mode. The UV-Vis absorption and photoluminescence spectra of these newly designed dopants were measured in dichloromethane (10^{-5} M) using Sinco S-3100 and Amincobrowman series 2 luminescence spectrometer. The fluorescent quantum yield were determined in dichloromethane solution at 293K against 4,4′-bis(4-(diphenyl-amino)styryl)biphenyl (BDAVBi) as a reference ($\Phi = 0.86$) [16]. The HOMO energy levels were measured with a low energy photo-electron spectrometry (Riken-Keiki AC-2). LUMO energy levels were estimated by subtracting the energy gap from the HOMO energy levels.

General procedure for the Horner-Wadsworth-Emmons reaction: To a mixture of phosphonate intermediate (0.82 mmol) and corresponding aldehyde (0.82 mmol) in 20 mL of anhydrous THF at 0°C was added dropwise 1.2 mL of 1.0 M KO*t*-Bu (1.2 mmol) in THF under N_2. The reaction mixture was stirred for 10 min at 0°C, followed by 1 hr at room temperature and quenched with water. The solution mixture was filtered with water. The filtered solid was dissolved in CH_2Cl_2, dried over Na_2SO_4, filtered with silica gel, and the solvent removed under reduced pressure to afford a crud product recrystallized from $CHCl_3$/EtOH, affording product (**1–4**).

7-(4-(6-Methylpyridin-2-yl)styryl)-9,9-diethyl-N,N-diphenyl-9H-fluoren-2-amine (**1**). Yield = 50%. ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm): 8.00 (d, $J = 8.3$ Hz, 2H), 7.63 (d, $J = 8.3$ Hz, 4H), 7.59 (d, $J = 5.4$ Hz, 1H), 7.56–7.48 (m, 3H), 7.47 (s, 1H), 7.24–7.22 (m, 4H), 7.12 (d, $J = 8.7$ Hz, 6H), 7.10–6.99 (m, 4H), 2.64 (s, 3H), 1.99–1.92 (m, 4H), 0.38 (t, $J = 7.3$ Hz, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ (ppm): 158.4, 156.5, 151.6, 150.4, 148.0, 147.3, 141.3, 138.6, 138.1, 136.9, 136.4, 135.5, 129.8, 129.3, 129.2, 127.3, 127.2, 126.8, 126.0, 123.9, 123.7, 122.5, 121.6, 120.6, 120.4, 119.4, 117.4, 56.1, 32.8, 24.8, 8.6; MS (FAB$^+$): 582 [M$^+$]; HRMS (FAB$^+$): [M$^+$] calculated for $C_{43}H_{38}N_2$, 582.3035, found: 582.3029.

7-(4-(Quinolin-2-yl)styryl)-9,9-diethyl-N,N-diphenyl-9H-fluoren-2-amine (**2**). Yield = 54%. ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm): 8.25–8.17 (m, 4H), 7.93 (d, $J = 8.6$ Hz, 1H), 7.83 (d, $J = 8.1$ Hz, 1H), 7.75 (d, $J = 8.1$ Hz, 1H), 7.71 (d, $J = 8.2$ Hz, 2H), 7.61 (t, $J = 7.8$ Hz, 2H), 7.56 (d, $J = 7.4$ Hz, 2H), 7.49 (s, 1H), 7.29–7.24 (m, 4H), 7.14–7.11 (m, 6H), 7.06–6.99 (m, 4H), 2.01–1.93 (m, 4H), 0.39 (t, $J = 7.3$ Hz, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ (ppm): 151.7, 150.5, 148.0, 147.3, 141.4, 138.7, 138.4, 136.7, 135.4, 130.2, 129.7, 129.2, 127.9, 127.5, 127.4, 127.2, 127.0, 126.9, 126.3, 123.9, 123.7, 122.6, 120.7, 120.5, 119.3, 118.8, 56.1, 32.8, 8.6; MS (FAB$^+$): 618 [M$^+$]; HRMS (FAB$^+$): [M$^+$] calculated for $C_{46}H_{38}N_2$, 618.3035, found: 618.3031.

7-(4-(Isoquinolin-1-yl)styryl)-9,9-diethyl-N,N-diphenyl-9H-fluoren-2-amine (**3**). Yield = 53%. ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm): 8.63 (d, $J = 5.7$ Hz, 1H), 8.19 (d, $J = 8.4$ Hz, 1H), 7.90 (d, $J = 8.1$ Hz, 1H), 7.72 (d, $J = 2.6$ Hz, 4H), 7.69–7.64 (m, 4H), 7.61–7.49 (m, 8H), 7.29–7.49 (m, 5H), 7.14–7.11 (m, 2H), 7.06–7.02 (m, 2H), 1.99–1.90 (m, 4H), 0.39 (t, $J = 7.2$ Hz, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ (ppm): 151.7, 150.5, 148.3, 147.3, 142.3, 137.9, 136.9, 136.3, 135.4, 130.4, 130.1, 130.0, 129.2, 127.6, 127.2, 127.1, 126.7, 126.3, 126.1, 123.9, 123.7, 122.5, 120.7, 120.4, 119.9, 119.3, 56.1, 32.7, 8.6; MS (FAB$^+$): 618 [M$^+$]; HRMS (FAB$^+$): [M$^+$] calculated for $C_{46}H_{38}N_2$, 618.3035, found: 618.3028.

7-(4-(Phenanthridin-6-yl)styryl)-9,9-diethyl-N,N-diphenyl-9H-fluoren-2-amine (**4**). Yield = 38%. ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm): 8.73 (d, J = 8.3 Hz, 1H), 8.64 (dd, J = 0.9, 8.3 Hz, 1H), 8.26 (dd, J = 1.2, 8.0 Hz, 1H), 8.20 (d, J = 7.8 Hz, 1H), 7.89 (td, J = 1.2, 7.1 Hz, 1H), 7.79–7.63 (m, 8H), 7.61 (d, J = 7.8 Hz, 1H), 7.57–7.51 (m, 3H), 7.30–7.24 (m, 4H), 7.15–7.11 (m, 5H), 7.08–6.99 (m, 4H), 2.06–1.89 (m, 4H), 0.40 (t, J = 7.3 Hz, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ (ppm): 160.9, 151.6, 150.5, 147.9, 147.3, 143.9, 141.4, 138.7, 138.1, 136.3, 135.4, 133.5, 130.6, 130.4, 130.3, 130.1, 129.2, 129.1, 128.9, 127.2, 127.1, 126.9, 126.4, 126.1, 125.2, 123.9, 123.7, 123.6, 122.5, 122.3, 121.9, 120.7, 120.4, 119.3, 56.1, 32.7, 8.6; MS (FAB$^+$): 668 [M$^+$]; HRMS (FAB$^+$): [M$^+$] calculated for $C_{50}H_{40}N_2$, 668.3191, found: 668.3188.

Fabrication of OLED and Measurements

For fabricating OLEDs, indium-tin-oxide (ITO) thin films coated on glass substrates were used, 30 Ω/square of the sheet resistivity with a thickness of 100 nm. The ITO-coated glass was cleaned in an ultrasonic bath by the following sequence: acetone, methyl alcohol, distilled water, storage in isopropyl alcohol for 48 h, and dried by N_2 gas gun. The substrates were treated by O_2 plasma under conditions of 2×10^{-2} Torr at 125 W for 2 min. All organic materials and metals were deposited under high vacuum (5×10^{-7} Torr). The OLEDs were fabricated in the following sequence: ITO/2-TNATA (60 nm)/NPB (20 nm)/2% dapant: MADN (30 nm)/Alq_3 (20 nm)/Liq (2 nm)/Al (100 nm). The current density (J), luminance (L), luminous efficiency (LE), and the CIE chromaticity coordinates of the OLEDs were measured with Keithly 2400, Chroma meter CS-1000A. Electroluminescence was measured using a Roper Scientific Pro 300i.

Results and Discussion

Figure 1 shows the structures of the electron donor-acceptor (D-A) type fluorescent materials. The (7-(diphenylamino)-9,9-diethylfluoren-2-yl)methylphosphonate [1] and aldehyde intermediates, such as 4-(6-methylpyridin-2-yl)benzaldehyde [13], 4-(quinolin-2-yl)benzal dehyde [14], 4-(isoquinolin-1-yl)benzaldehyde [14], and 4-(phenanthridin-6-yl)benzal dehyde [14] were prepared according to the methods reported in the literature. The Horner-Emmons reaction between 7-(Diphenylamino)-9,9-diethylfluoren-2-yl)methylphosphonate and the corresponding aldehyde compounds afforded the blue-emitting materials (**1**–**4**) in moderate yield. After conventional purification, such as column chromatography and recrystallization, these newly synthesized materials (**1–4**) were purified further by train sublimation under reduced pressure ($<10^{-3}$ torr). The molecular structures of the blue-emitting materials (**1–4**) were fully characterized by ^{1}H- and ^{13}C-nuclear magnetic resonance (NMR), and low- and high-resolution mass spectroscopy.

Figure 2 presents the UV-Vis absorption and PL emission spectra of blue fluorescent materials **1**–**4** in dichloromethane and on quartz plates. All compounds exhibited a charge-transfer (CT) absorption band in the dichloromethane solution over the range, 385–395 nm. The UV-vis absorption spectra of the blue materials (**1–4**) overlapped well with the PL emission spectra of a common blue host, MADN. This suggests that energy transfer between the MADN and these materials is quite efficient and MADN serves well as a host in OLED devices using these materials as dopants. The emission spectra of the D-A molecules increased in the order compounds **1** (495 nm) < **3** (520 nm) < **4** (525 nm) < **2** (529 nm). The emission spectra of the D-A molecule showed a gradual red shift with increasing π-conjugation length of the electron acceptor. For example, compared to compound **1** with

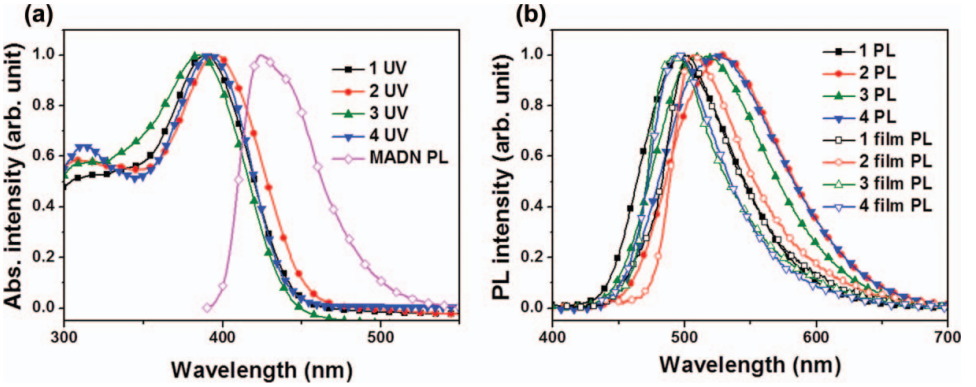

Figure 2. (a) UV-Vis absorption spectra and (b) PL spectra of materials (**1–4**) (solution; closed symbols and film; opened symbols).

the pyridine moiety, the observed red shifts in the emission spectra of compounds **2–4** may arise from the extended π-conjugation in the electron accepting moieties. Compound **3** had a shorter wavelength (520 nm) than compound **2** (529 nm), which may be rationalized from the fact that the dihedral angle between the phenyl group and isoqunoline in compound **3** is more distorted than that between phenyl group and quinoline in compound **2**, resulting in a wide band gap. Table 1 lists the photophysical properties of the D-A materials. Interestingly, compounds **1–4** all showed a maximum emission wavelength that was blue-shifted (3–30 nm) in the film compared to the solution. The emission quantum yield of these materials was high ($\Phi = 0.50$–0.79), suggesting that these materials have highly efficient electroluminescent properties in OLED devices.

The highest occupied molecular orbital (HOMO) levels were measured using a photoelectron spectrometer (Riken-Keiki AC-2), and the lowest unoccupied molecular orbital (LUMO) levels were calculated by subtracting the corresponding optical band gap from the HOMO values. The HOMO and LUMO energy levels of compounds **1–4** ranged from -5.41 to -5.61 eV and -2.68 to -2.80 eV, respectively (Table 1). The calculated HOMO-LUMO energy gaps for compounds **1–4** were 2.81, 2.73, 2.81 and 2.77 eV, respectively. Figure 3 shows the HOMO and LUMO energy levels of the blue fluorescent materials,

Table 1. Physical properties of blue materials (**1–4**)

Compound	UV_{Max} (nm)[a]	PL_{Max} (nm)[a]/[b]	FWHM (nm)[a]/[b]	HOMO/LUMO (eV)[c]	E_g	Φ [d]
1	389	495/492	86/62	−5.58/−2.77	2.81	0.79
2	395	529/506	100/64	−5.41/−2.68	2.73	0.60
3	385	520/503	99/62	−5.61/−2.80	2.81	0.50
4	392	525/495	102/63	−5.56/−2.79	2.77	0.56

[a]Maximum absorption or emission wavelength in 1,2-dichloroethane (1×10^{-5} M). [b]Thin films. [c]Obtained from AC-2 and UV-Vis absorption measurements. [d]Using BDAVBi as a standard; $\lambda_{ex} = 360$ nm ($\Phi = 0.86$ in 1,2-dichloroethane).

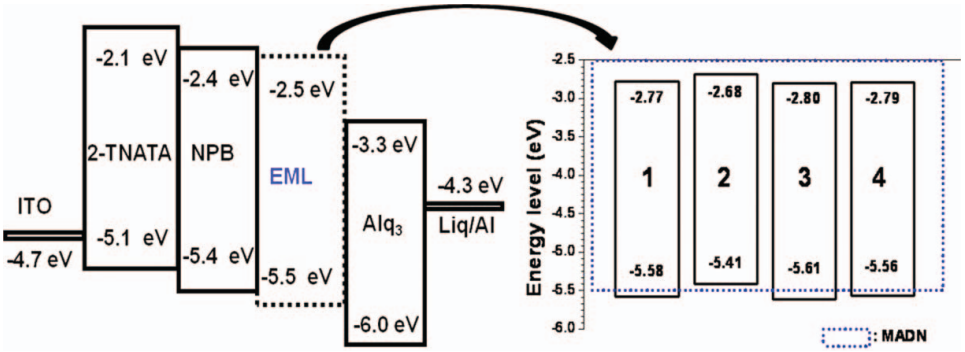

Figure 3. Energy level diagram of the materials used in devices.

along with those of other materials used in the electroluminescence devices, including ITO, 2-TNATA, NPB, MADN, Alq_3 and Liq:Al.

To examine the EL properties of these materials, the devices were fabricated with the following structure: ITO/2-TNATA (60 nm)/NPB (20 nm)/MADN: 2% dapant (30 nm)/Alq_3 (20 nm)/Liq (2 nm)/Al (100 nm). Figure 5 shows the luminous efficiencies and luminance of the devices, and the data is summarized in Table 2. Devices **A-D** showed high luminous efficiencies of 6.85, 5.20, 6.90 and 7.13 cd/A, and power efficiencies of 2.68, 2.04, 2.80, and 2.74 lm/W at 20 mA/cm^2, respectively. This suggests that the EL performance of devices **A–D** is sensitive to the structural changes of the acceptor groups in the emitting layers. For example, compared to device **B** using compound **2** with a quinoline end-capping group, the luminous and power efficiency of device **D** using compound **4** with a phenanthridine end-capping group increased by 37.1 and 34.3% at 20 mA/cm^2, respectively. Interestingly, the HOMO energy level of compound **2** was 0.15∼0.20 eV higher than that of compounds **1**, **3** and **4**. Presumably, this high HOMO energy level of dopant **2** and effective hole-trapping in device **B** would reduce the EL efficiencies of device **B** compared to devices **A**, **C** and **D**.

Molecular mechanics (MM2) energy minimization simulations of compounds **1–4** were carried out using ChemDraw to explain the observed differences in electroluminescent properties of devices **A–D** at the molecular level of the dopant materials. Figure 4 shows the energy-minimized structures of compounds **1–4**. The calculated dihedral angles between the phenyl and methylpyridyl (qunolinyl) moiety of compounds **1** and **2** were 0.54 and 2.30°, respectively. However, the angles between the phenyl and isoqunolinyl (phenanthridyl)

Table 2. EL performance characteristic of devices (**A–D**)

	Dopant (%)	L[a] (cd/m^2)	V_{on}[b] (V)	LE[c]/[d] (cd/A)	PE[c]/[d] (lm/W)	EL[e] (nm)/fwhm	CIE[e](x,y)
A	**1**	7522	4.0	6.85/6.85	2.68/3.64	460/48	(0.147, 0.152)
B	**2**	6785	4.5	5.20/5.23	2.04/2.33	464/56	(0.154, 0.156)
C	**3**	6308	4.5	6.90/6.99	2.74/3.15	464/64	(0.148, 0.179)
D	**4**	6441	4.5	7.13/7.16	2.80/3.36	466/64	(0.148, 0.191)

[a]Value of the luminance at 11 V. [b]Turn-on voltage at 1.0 cd/m^2. [c]Value measured at 20 mA/cm^2. [d]Maximum values. [e]Value measured at 7.0 V.

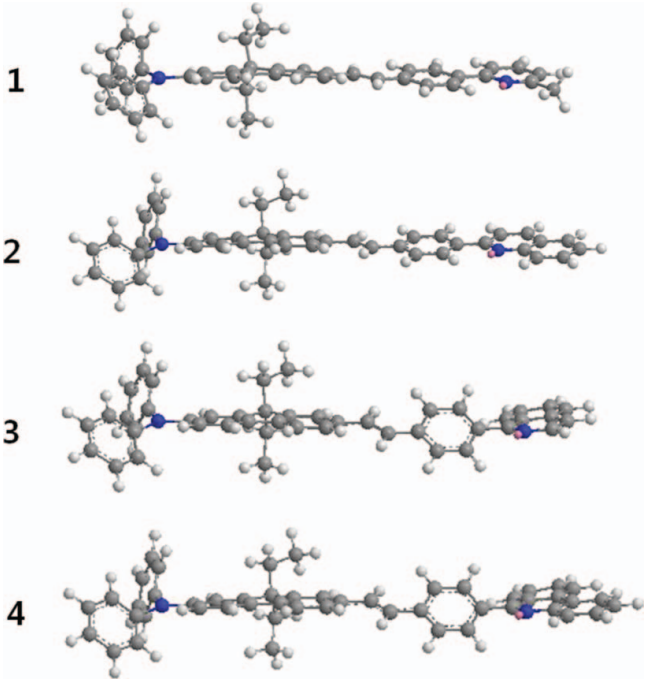

Figure 4. Energy-minimized structure of materials (**1–4**) by MM2 calculation.

moieties of compounds **3** and **4** were 41.6 and 43.7°, respectively. These results suggest that the molecular aggregation between compounds **1** and **2** is more effective than that between compounds **3** and **4**. Accordingly, devices **A** and **B** using compounds **1** and **2** would show lower EL efficiency through concentration quenching compared to devices **C** and **D** using compounds **3** and **4**. Presumably, the higher quantum yield of compound **1** ($\Phi = 0.79$) than compound **2** ($\Phi = 0.60$) plays an important role in the improved EL efficiency of device **A** using compound **1** compared to device **B** using compound **2**.

As shown in Figure 6, devices **A-D** emitted blue electroluminescence in the blue region at 460, 464, 466, and 464 nm, respectively. The CIE coordinates of devices **A**-**D**

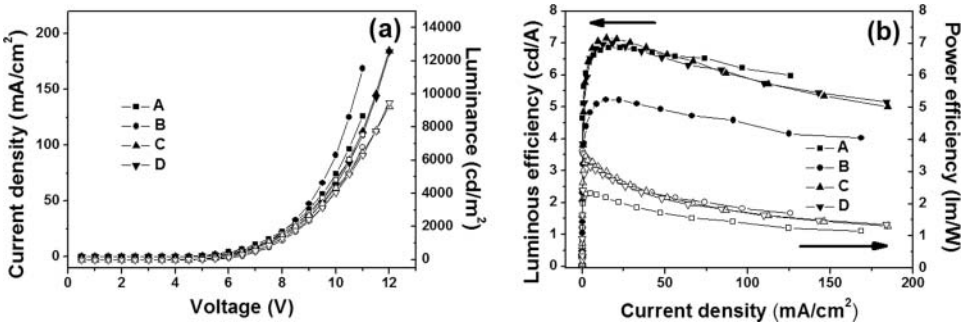

Figure 5. (a) *J-V-L* characteristics of devices and (b) luminous efficiency and power efficiency vs. current density relationship of devices **A–D**.

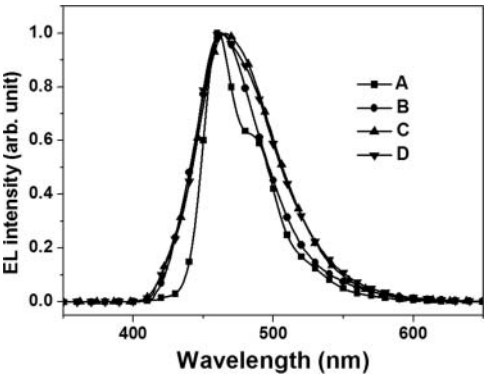

Figure 6. EL spectra of blue OLEDs at 7.0 V.

were (0.147, 0.152), (0.154, 0.156), (0.148, 0.179) and (0.148, 0.191), respectively. Device **A** using compound **1** showed deep blue emission with a Commission Internationale de l'Eclairage (CIE*y*) coordinates of approximately 0.15 at 7.0 V. Given that the electron donor moiety in compounds **1**–**4** is the same, the observed blue shift in device **A** may be due to the less π-conjugation of the electron acceptor moiety of compound **1** in the emitting layer of device **A**.

Conclusions

This paper reported a molecular design strategy of combining a 2-diphenylaminofluorene-7-ylstyrene as an electron donor and pyridine derivatives as an electron acceptor to give the new electron donor-acceptor type blue materials. In particular, device **D** exhibited highly efficient blue emission with a luminous efficiency, power efficiency and CIE*x,y* coordinates of 7.13 cd/A, 2.80 lm/W at 20 mA/cm^2 and ($x = 0.148$, $y = 0.191$) at 7.0 V, respectively. In addition, device **A** showed efficient deep-blue emission with a luminous efficiency, power efficiency and CIE*x,y* coordinates of 6.85 cd/A, 2.68 lm/W at 20 mA/cm^2 and ($x = 0.147$, $y = 0.152$) at 7.0 V, respectively. This clearly suggests that the materials based on the donor-acceptor structures have excellent properties suitable for blue-emitting materials in OLEDs

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