

White-Light-Emitting Diodes from Single Polymer Systems Based on Polyfluorene Copolymers with Quinoxaline Derivatives

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ABSTRACT: White polymeric light-emitting diodes (WPLEDs) from a single polymer system were developed by incorporating 2,3-bis(4-methoxyphenyl)quinoxaline and 5,8-bis(*N,N*-diphenylamino)-2,3-bis(4-methoxyphenyl)quinoxaline into poly(9,9-dioctylfluorene). All copolymers showed good thermal stability with 5% weight loss temperatures above 420 °C, and glass transition temperatures ranged from 86 to 143 °C. Electroluminescence (EL) devices with indium tin oxide/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonic acid)/emission layer/Ca/Ag were found to emit white light with Commission Internationale de l'Eclairage (CIE) coordinates of (0.33, 0.40) and (0.30, 0.36). These devices exhibit a highest brightness of 12 300 cd/m² and a maximum current efficiency of 2.56 cd/A.

Introduction

White polymeric light-emitting diodes (WPLEDs) have recently attracted a great of attention due to their potential applications in back-lighting for liquid-crystal displays (LCDs), full-color light-emitting diodes (LEDs) with color filters, and next-generation lighting sources.¹ The general way to obtain WPLED is to use dye/polymer blending systems² or polymer/polymer blending systems.³ However, the blending systems always have a color instability problem due to phase separation. The single polymer systems have been considered as a feasible approach to improve this problem. Several studies have successfully synthesized single polymers containing red-, green-, and blue- emission units in the backbone of polyfluorene.⁴

In this work, we developed new white polymeric light-emitting diodes from a single polymer system. The single polymer systems were prepared from the copolymerization of dioctylfluorene monomer with monomers having 2,3-bis(4-methoxyphenyl)quinoxaline (moQ) and 5,8-bis(*N,N*-diphenylamino)-2,3-bis(4-methoxyphenyl)quinoxaline (DPAmoQ) units. The DPAmoQ system exhibits a longer wavelength emission because of internal charge transfer (ICT),⁵ where the biphenylamine group acts as an electron donor and the quinoxaline unit acts as an electron acceptor. By adjusting the molar ratios of the monomers, we were able to balance the light color of the copolymers and obtain white-light emission.

Experimental Section

Materials. 4,7-Dibromo-2,1,3-benzothiadiazole (**1**), 4,4'-dimethoxybenzil (**3**), 2,7-dibromo-9,9-dioctylfluorene (**9**), palladium(II) acetate (Pd(OAc)₂), dioctylfluorene-2,7-diboronic acid bis(1,3-propanediol) ester (**8**), and *o*-xylene were obtained from Aldrich Chemicals. Tetrakis(triphenylphosphine)palladium (Pd(PPh₃)₄), Aliquat 336, and *N*-bromosuccinimide (NBS) were purchased from Acros Organics. Tri-*tert*-butylphosphine (P(Bu₃)^t) was obtained from Strem Chemicals. 4-Biphenylamine (**5**) was purchased from Alfa Aesar. Sodium *tert*-butoxide (^tBuONa) was obtained from TCI. Toluene, chloroform, and ammonium hydroxide were obtained from TEDIA. All reagents were used as received.

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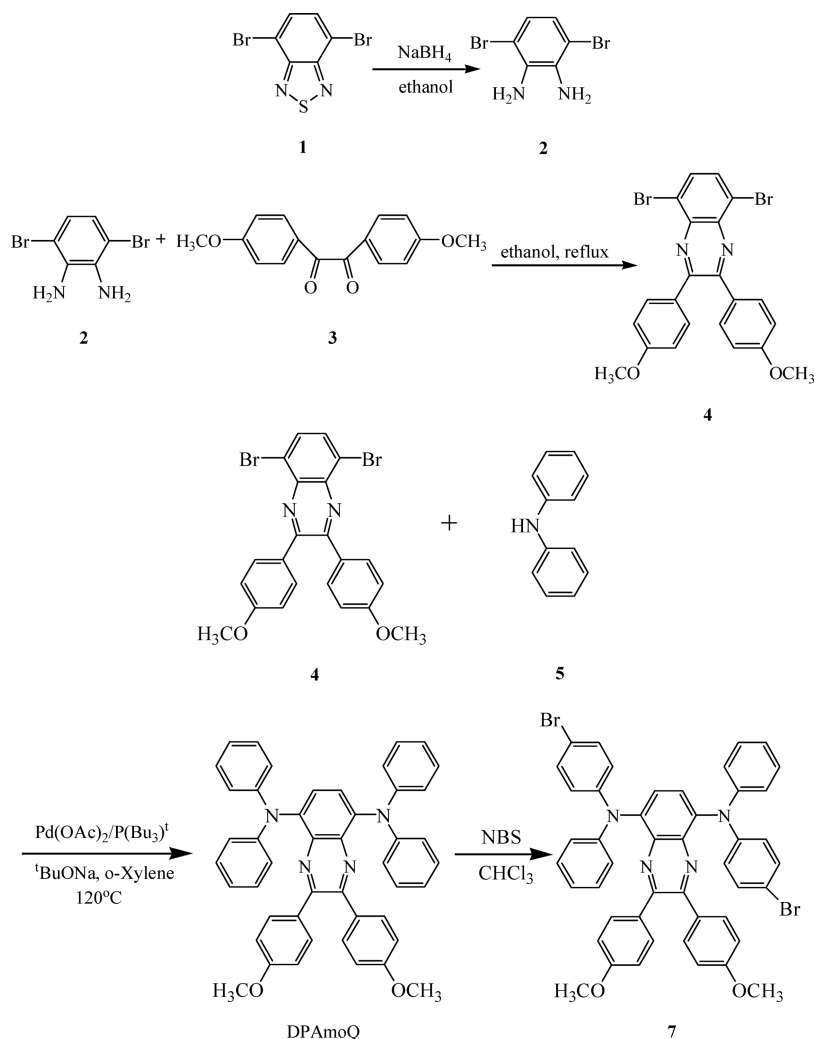
Measurements and Characterization. ¹H NMR spectra were recorded on a Bruker Advance 600 spectrometer, and the deuterated CDCl₃ and DMSO were used as solvents and the chemical shifts were reported in ppm. The molecular weights and distributions of polymers were obtained by using a Waters GPC 2414 in tetrahydrofuran (THF) via a calibration curve of polystyrene standards. Thermal stability was analyzed using a TA Instruments thermogravimetric analyzer (TGA) Q500 at a heating rate of 10 °C/min under nitrogen. The glass transition temperature (*T*_g) was determined on a Perkin-Elmer Instruments model 4000 differential scanning calorimeter (DSC) at a heating rate of 10 °C/min under nitrogen. UV–vis absorption spectra were recorded on a Hitachi U-2001 spectrophotometer. Photoluminescence spectra (PL) were recorded on a Hitachi F-4500 fluorometer. Electroluminescence (EL) spectra and luminance of the devices were measured using a Photoresearch PR650. The voltage–current was controlled by a Keithley 2400 power source. Cyclic voltammetry (CV) was carried out on a potentiostat/galvanostat Autolab PGSTAT30 with a platinum electrode at a scan rate of 50 mV/s against Ag/AgCl reference electrode with a nitrogen-saturated solution of 0.1 M tetrabutylammonium hexafluorophosphate in acetonitrile.

Synthesis of 3,6-Dibromobenzene-1,2-diamine (2**).** Compound **1** (2 g, 6.8 mmol) was suspended in 100 mL of ethanol and then cooled to 0 °C under nitrogen. NaBH₄ (3.65 g, 96.6 mmol) was added into the solution slowly, and the mixture was stirred at room temperature overnight. After removing ethanol, the product was poured into deionized water. The precipitated powder was collected and washed with deionized water and methanol several times. The yield was 88% (1.59 g). ¹H NMR (600 MHz, DMSO): 6.63 (s, 2H), 4.99 (s, 4H).

Synthesis of 5,8-Dibromo-2,3-bis(4-methoxyphenyl)quinoxaline (Br2-moQ) (4**).** A mixture of compound **2** (1.58 g, 6 mmol), compound **3** (1.62 g, 6 mmol), and ethanol (50 mL) was refluxed under nitrogen for 12 h. The precipitate was collected by filtration, washed with ethanol, and then vacuum-dried. The yield was 43% (1.29 g). ¹H NMR (600 MHz, CDCl₃): 7.85 (s, 2H), 6.95 (d, 4H), 6.88 (d, 4H), 3.85 (s, 6H). Elemental analysis (%) calcd: C, 52.83; H, 3.22; N, 5.60. Found: C, 52.61; H, 3.36; N, 5.75.

Synthesis of 5,8-Bis(*N,N*-diphenylamino)-2,3-bis(4-methoxyphenyl)quinoxaline (DPAmoQ). Pd(OAc)₂ (9 mg, 0.04 mmol),

Scheme 1. Synthesis of Monomers



PBu₃¹ (0.64 mL, 50 mg/mL in *o*-xylene, 0.16 mmol), compound **4** (1 g, 2 mmol), compound **5** (0.68 g, 4 mmol), sodium *tert*-butoxide (0.43 g, 4.4 mmol), and *o*-xylene (30 mL) were mixed at room temperature in a three-neck round-bottom flask and heated to 120 °C for overnight under nitrogen. After addition of water and extractive workup with Et₂O, *o*-xylene was stripped off in vacuum. The product was washed with deionized water and methanol several times. The yield was 71% (0.96 g). ¹H NMR (600 MHz, CDCl₃): 7.46 (s, 2H), 7.29 (m, 8H), 7.03 (m, 12H), 6.71(d, 4H), 6.81 (d, 4H), 3.71 (s, 6H).

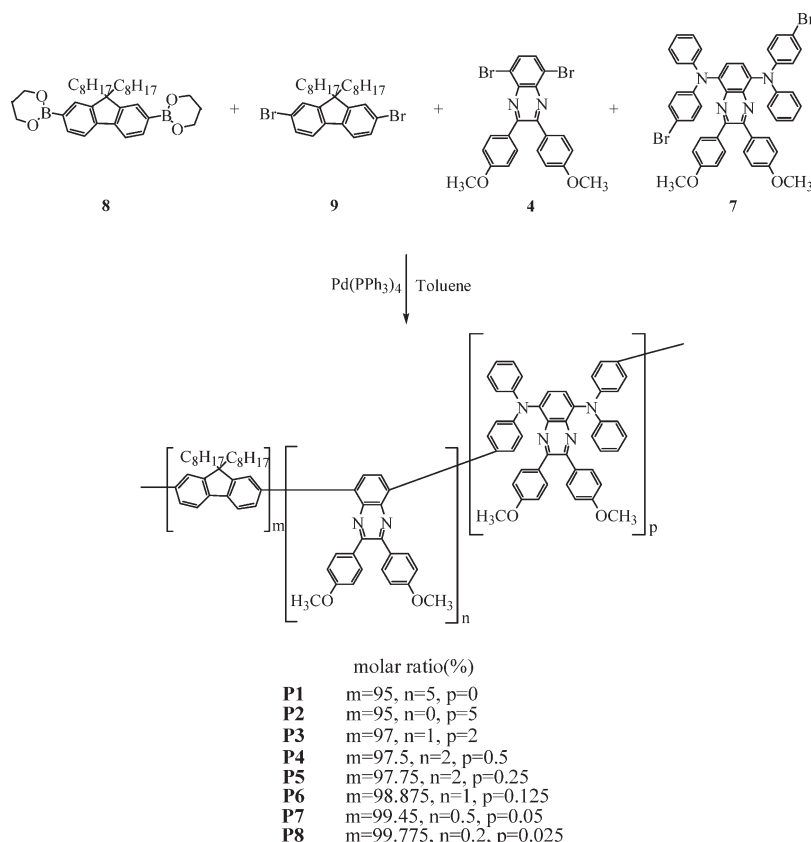
Synthesis of 5,8-Bis(*N*-(4-bromophenyl)anilino)-2,3-bis(4-methoxyphenyl)quinoxaline (Br₂-DPAmoQ) (7). DPAmoQ (0.81 g, 1.2 mmol) was dissolved in 50 mL of chloroform under an argon atmosphere. NBS (0.43 g, 2.4 mmol) was dissolved in 50 mL of chloroform, and the solution was added to the DPAmoQ solution and stirred at room temperature overnight. Aqueous K₂CO₃ solution was added and stirred for several hours. The organic layer was washed with deionized water twice. The crude product was recrystallized from a mixture of dichloromethane and methanol at room temperature. The product is a mixture of *N*⁵,*N*⁸-bis(4-bromophenyl)-2,3-bis(4-methoxyphenyl)-*N*⁵,*N*⁸-diphenylquinoxaline-5,8-diamine and *N*⁵,*N*⁵-bis(4-bromophenyl)-2,3-bis(4-methoxyphenyl)-*N*⁸,*N*⁸-diphenylquinoxaline-5,8-diamine. The yield was 53% (0.53 g). ¹H NMR (600 MHz, DMSO): 7.46 (s, 2H), 7.43 (m, 4H), 7.32 (m, 6H), 7.06 (m, 4H), 6.94 (d, 4H), 6.83 (d, 4H), 6.73 (d, 4H), 3.72 (s, 6H). Elemental analysis (%) calcd: C, 66.20; H, 4.11; N, 6.71. Found: C, 65.72; H, 4.00; N, 6.60. MS (FAB): *m/z* 834 (M⁺) (calcd 834.6).

General Procedure of Copolymer Synthesis. The synthetic route is shown in Scheme 2. The synthesis of copolymer **P1** is given as an example. To a 100 mL round-bottom flask, compound **8** (0.28 g, 0.5 mmol), compound **9** (0.25 g, 0.45 mmol), and compound **4** (0.025 g, 0.05 mmol) were dissolved in 10 mL of toluene and purged with nitrogen for 30 min. Subsequently, Pd(PPh₃)₄ (10 mg, 0.0086 mmol), Aliquat 336 (several drops), and 2 M aqueous K₂CO₃ solution (10 mL) were added to the flask. The reaction mixture was stirred at 90 °C for 2 days. The end-cappers (bromobenzene and phenylboronic acid) dissolved in toluene were added and stirred for 12 h. After being cooled to room temperature, the reaction mixture was poured into a mixture of methanol and water. The solid was filtered and redissolved in chloroform. Ammonium hydroxide solution was added and stirred overnight. The organic layer was washed with deionized water twice. Then it was precipitated in a large amount of methanol. The solid was further purified by Soxhlet extraction for 24 h using acetone as a solvent and dried in a vacuum oven at 70 °C overnight. The yield was 72%. ¹H NMR (600 MHz, CDCl₃): 7.80–7.96 (broad), 7.64–7.71 (broad), 7.48 (m), 6.87 (d), 3.83 (s), 2.13 (m), 1.14–1.22 (broad), 0.80 (m).

P2. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.25 g, 0.45 mmol), and compound **7** (0.042 g, 0.05 mmol). The yield was 69%. ¹H NMR (600 MHz, CDCl₃): 7.79–7.84 (broad), 7.59–7.69 (broad), 7.48 (m), 7.08 (d), 6.92 (d), 6.62 (s), 3.74 (s), 2.13 (broad), 1.11–1.21 (broad), 0.80 (broad).

P3. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.257 g, 0.47 mmol), compound **4** (0.005 g,

Scheme 2. Synthesis of Copolymers



0.01 mmol), and compound **7** (0.017 g, 0.02 mmol). The yield was 68%. $^1\text{H NMR}$ (600 MHz, CDCl_3): 7.85 (broad), 7.69 (broad), 2.13 (broad), 1.14–1.21 (broad), 0.81 (broad).

P4. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.26 g, 0.475 mmol), compound **4** (0.01 g, 0.02 mmol), and compound **7** (0.0042 g, 0.005 mmol). The yield was 82%. $^1\text{H NMR}$ (600 MHz, CDCl_3): 7.85 (broad), 7.70 (broad), 2.13 (broad), 1.14–1.21 (broad), 0.81 (broad).

P5. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.262 g, 0.4775 mmol), compound **4** (0.01 g, 0.02 mmol), and compound **7** (0.0021 g, 0.0025 mmol). The yield was 74%. $^1\text{H NMR}$ (600 MHz, CDCl_3): 7.85 (broad), 7.70 (broad), 2.13 (broad), 1.14–1.22 (broad), 0.81 (broad).

P6. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.268 g, 0.48875 mmol), compound **4** (0.005 g, 0.01 mmol), and compound **7** (0.001 g, 0.00125 mmol). The yield was 64%. $^1\text{H NMR}$ (600 MHz, CDCl_3): 7.85 (broad), 7.70 (broad), 2.13 (broad), 1.14–1.22 (broad), 0.82 (broad).

P7. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.271 g, 0.4945 mmol), compound **4** (0.0025 g, 0.005 mmol), and compound **7** (0.00042 g, 0.0005 mmol). The yield was 77%. $^1\text{H NMR}$ (600 MHz, CDCl_3): 7.85 (broad), 7.70 (broad), 2.13 (broad), 1.14–1.22 (broad), 0.81 (broad).

P8. Monomer feed ratio: compound **8** (0.28 g, 0.5 mmol), compound **9** (0.273 g, 0.49775 mmol), compound **4** (0.001 g, 0.002 mmol), and compound **7** (0.00021 g, 0.00025 mmol). The yield was 78%. $^1\text{H NMR}$ (600 MHz, CDCl_3): 7.85 (broad), 7.70 (broad), 2.13 (broad), 1.14–1.22 (broad), 0.81 (broad).

EL Device Fabrication. Patterned ITO slides were cleaned with detergent, deionized water, acetone, and 2-propanol in an ultrasonic bath. The ITO slides were dried on a hot plate at 150 °C for 30 min followed by UV ozone treatment for 30 min before use. A hole injection layer of poly(styrenesulfonic acid)-doped poly(ethylene dioxythiophene) [PEDOT:PSS] (Baytron P VP AI 4083) was spin-coated on the ITO and was dried on the hot plate for 30 min at 150 °C. The emitting polymer was

spin-coated from its solution in chloroform (10 g/L) and dried on a hot plate. Finally, a thin layer of calcium (20 nm) was deposited by thermoevaporation under a vacuum of 10^{-5} Torr, followed by a layer of silver (150 nm). The device measurement was carried out in a glovebox at room temperature.

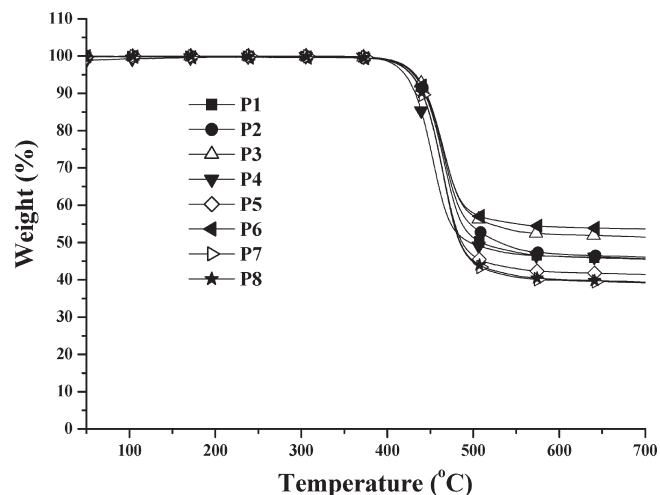
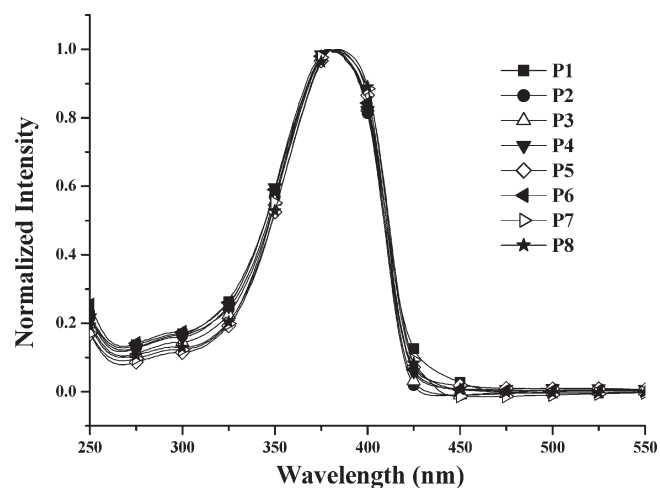
Results and Discussion

Synthesis and Characterization. The monomer **7** (Br2-DPAmoQ) was prepared by the bromination reaction with NBS. The dibrominated product is most probably a mixture of regioisomers. The bromination with NBS is not regioselective among the various N-activated phenyl rings. By using only $^1\text{H NMR}$, one cannot distinguish between N^5, N^8 -bis(4-bromophenyl)-2,3-bis(4-methoxyphenyl)- N^5, N^8 -diphenylquinoxaline-5,8-diamine and N^5, N^8 -bis(4-bromophenyl)-2,3-bis(4-methoxyphenyl)- N^5, N^8 -diphenylquinoxaline-5,8-diamine. The monomer **7** uses biphenylamine instead of regular thiophene to act as an electron donor, which could avoid the fluorescence quenching effect. The synthesis of light-emitting copolymers using palladium-catalyzed Suzuki cross-coupling⁶ is depicted in Scheme 2. The molar ratios of compounds **4** and **7** in the copolymers were in the range of 0.025–5%. The number-average molecular weights of the synthesized copolymers varied in the range of 21 600–25 900 g/mol with polydispersity index ranging from 1.79 to 1.87 (Table 1). All copolymers are soluble in common organic solvents, such as chloroform, THF, and toluene. The thermal stability of these copolymers was determined by TGA under a nitrogen atmosphere as shown in Figure 1. The thermal decomposition temperatures (T_{d5} , 5% weight loss) of the copolymers are all above 420 °C, which indicates that all copolymers have good thermal stability. The glass-transition temperatures (T_g s) of the copolymers were investigated by DSC under nitrogen. The T_g s of the copolymers

Table 1. Molecular Weights and Thermal Properties of Copolymers

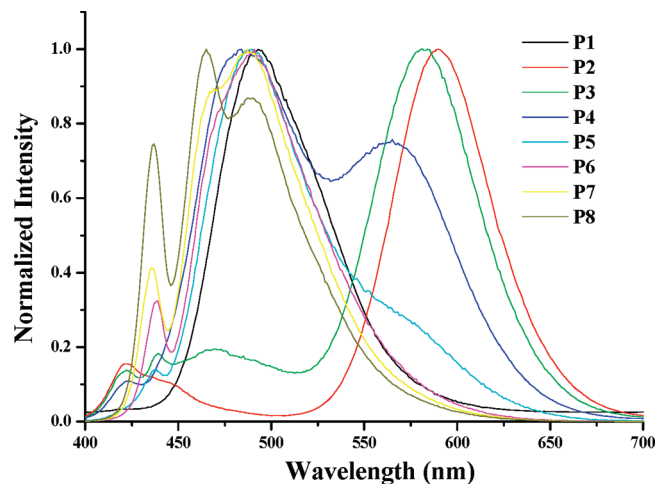
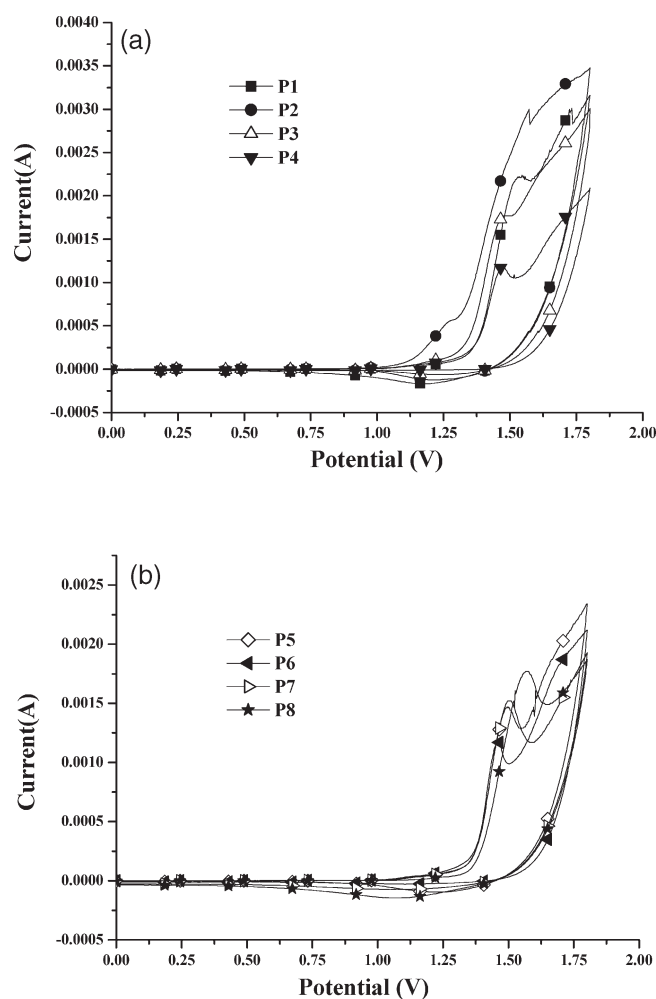
copolymer	M_n (g/mol)	PDI ^a	T_g (°C)	T_d (°C) ^b
P1	24 900	1.79	133	432
P2	21 600	1.85	143	432
P3	23 800	1.85	119	433
P4	23 700	1.87	116	424
P5	25 900	1.86	112	429
P6	23 000	1.83	106	432
P7	24 200	1.84	86	429
P8	21 700	1.87	86	429

^a Polydispersity. ^b Temperature of 5% weight loss measured by TGA in nitrogen.

**Figure 1.** TGA curves of copolymers.**Figure 2.** UV-vis spectra of copolymers.**Table 2. Optical Properties of Copolymers**

copolymer	$\lambda_{\text{Abs, film}}$ (nm)	$\lambda_{\text{PL, film}}$ (nm)
P1	380	493
P2	380	422, 590
P3	380	422, 439, 470, 584
P4	380	423, 484, 568
P5	383	438, 490, 564
P6	380	438, 490
P7	380	435, 468, 487
P8	383	437, 465, 489

range from 86 to 143 °C (in Table 1). Because of the incorporation of rigid quinoxaline segments into the polymer chains, the copolymers have higher glass transition

**Figure 3.** PL spectra of copolymers (excitation: 380 nm).**Figure 4.** Cyclic voltammograms of copolymers.

temperatures than poly(9,9-dioctylfluorene) (72 °C)⁷ even at such a low molar ratio. Copolymer **P2** displays the highest T_g value, which is due to the incorporation of rigid monomer **7** at the highest molar ratio. By contrast, **P7** and **P8** display the lowest T_g values which are very close to the reported T_g for poly(9,9-dioctylfluorene).

Optical Properties. The UV-vis absorption spectra of the copolymer films are shown in Figure 2. All copolymers exhibit a strong peak at 380 nm (listed in Table 2), which

Table 3. Electrochemical Properties of Copolymers

copolymer	$E_{\text{ox}}^{\text{onset}}$ (V)	HOMO (eV) ^a	$E_{\text{g}}^{\text{opt}}$ (eV) ^b	LUMO (eV) ^c
P1	1.36	−5.67	2.92	−2.75
P2	1.27	−5.58	2.94	−2.64
P3	1.33	−5.64	2.95	−2.69
P4	1.35	−5.66	2.95	−2.71
P5	1.36	−5.67	2.93	−2.74
P6	1.36	−5.67	2.93	−2.74
P7	1.37	−5.68	2.92	−2.76
P8	1.38	−5.69	2.94	−2.75

^a $E_{\text{HOMO}} = -e(E_{\text{ox}}^{\text{onset}} - E_{\text{FOC}} + 4.8 \text{ V})$, where $E_{\text{FOC}} = 0.488 \text{ V}$ vs Ag/Ag^+ . ^b Optical band gaps were estimated from the onset of absorption. ^c LUMO values were obtained from HOMO values and optical band gaps.

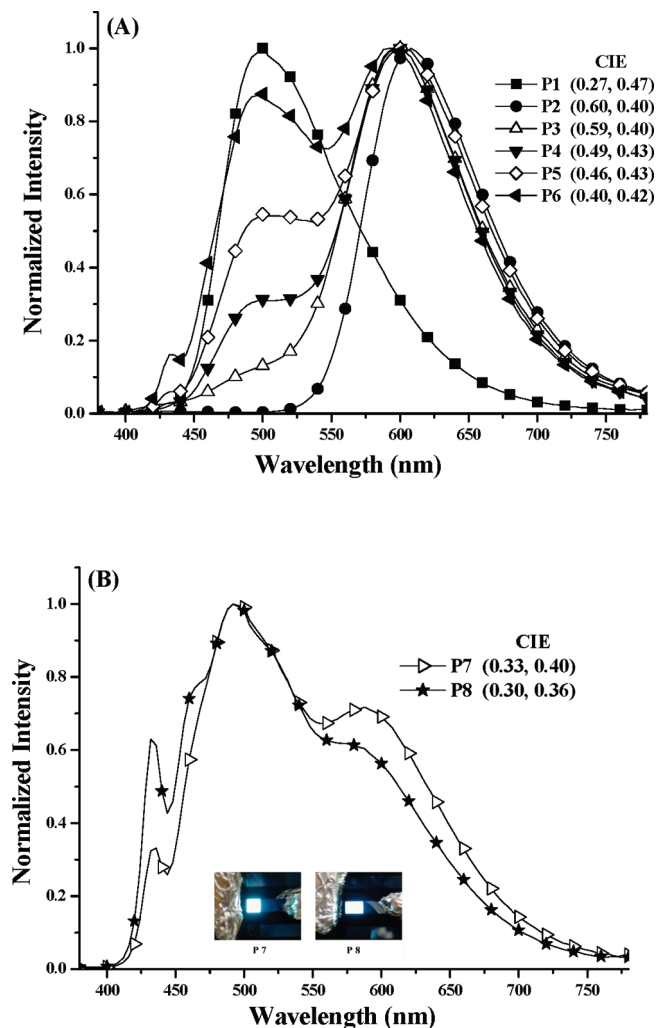


Figure 5. EL spectra of copolymers with corresponding CIE coordinates: (A) P1, P2, P3, P4, P5, and P6; (B) P7 and P8 (inset: photographs of EL devices during operation).

belongs to the absorption of homopolydioctylfluorene.⁸ The absorption peaks of moQ and DPAmoQ segments cannot be found because of their low contents in the polymer backbone.

The photoluminescence spectra of the copolymers in the solid state are shown in Table 2 and Figure 3. In the spectrum of P1, the intense emission peak at 493 nm is attributed to moQ unit, and the emission of the fluorene segment in the polymer chain could not be found. The PL spectrum of P2 exhibits a major emission peak at 590 nm and a small one at 422 nm, corresponding to the DPAmoQ and the

Table 4. EL Device Performances of Copolymers

copolymer	λ_{max} (nm)	maximum brightness (cd/m ²)	current efficiency (cd/A)	CIE (x, y)
P1	500	2924	0.88	(0.27, 0.47)
P2	608	2880	0.77	(0.60, 0.40)
P3	600	4468	1.37	(0.59, 0.40)
P4	500, 600	7740	2.43	(0.49, 0.43)
P5	500, 600	5285	1.66	(0.46, 0.43)
P6	432, 500, 596	8784	2.56	(0.40, 0.42)
P7	432, 492, 588	12300	2.02	(0.33, 0.40)
P8	432, 464, 492, 588	7259	1.87	(0.30, 0.36)

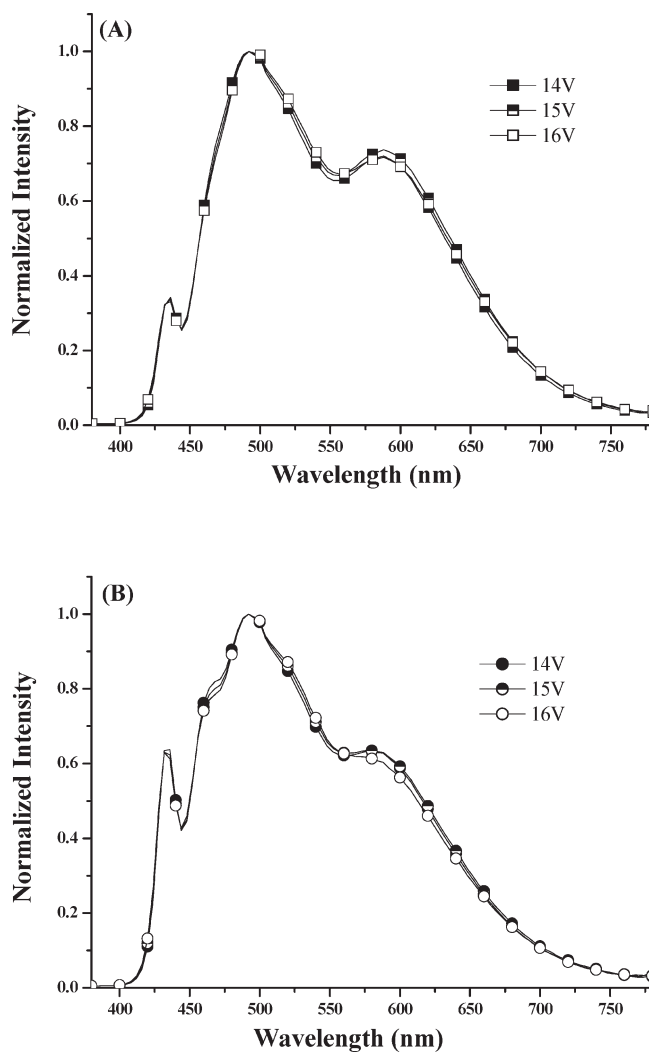


Figure 6. EL spectra of the devices of (A) P7 and (B) P8 at different biases.

polydioctylfluorene backbone.⁹ The results reveal that the energy transfer from the polydioctylfluorene backbone to moQ is more efficient than to DPAmoQ. From the PL spectra of P3, P4, P5, P6, P7, and P8, the different intensities of the blue, green, and red emission peaks can be observed when adjusting the molar ratios of the monomers. The incomplete energy transfer from polydioctylfluorene (blue emission) to moQ (green emission) and DPAmoQ (red emission) can provide white-light emission covering a wide range in the visible region.

Electrochemical Properties. The electrochemical behaviors of the copolymers were investigated by cyclic voltammetry (CV) and anodically scanned cyclic voltammograms (Figure 4).

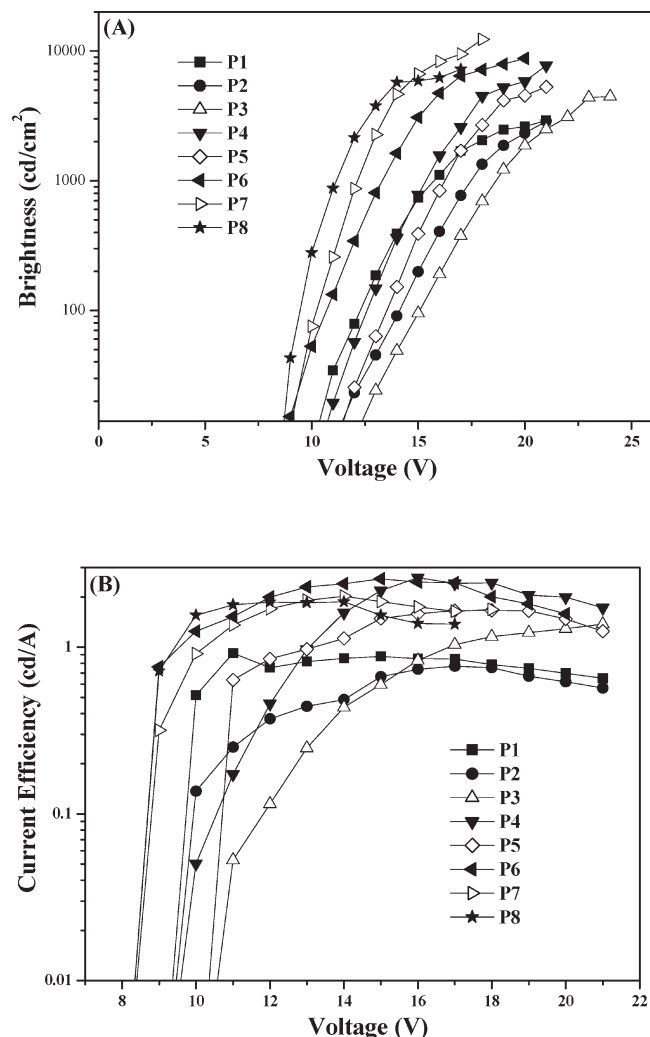


Figure 7. Voltage–brightness (A) and voltage–current efficiency (B) curves of copolymers.

No reduction wave for either copolymers **P1**–**P8** was observed in the range 0 to -2.5 eV vs Ag/Ag^+ . The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energy values of the copolymers are listed in Table 3. The onset oxidation ($E_{\text{ox}}^{\text{onset}}$) of the copolymers were found to range from 1.27 to 1.38 V, corresponding to the HOMO levels of -5.69 to -5.58 . **P2** has the highest HOMO value because it contains the largest amount of compound **7** incorporated into the polymer. Furthermore, diphenylamine is an electron-donating group that can raise the HOMO level of the copolymer. Because of their similar absorption spectra, the optical band gaps (E_g^{opt}) of the copolymers are very close.

EL Properties. The copolymer EL devices with the configuration of ITO/PEDOT:PSS/emission layer/Ca/Ag were fabricated. Figure 5 shows the EL spectra of the copolymers, and the detail emitting peaks are listed in Table 4. The EL spectrum of **P1** only exhibits green emission ($\lambda_{\text{max}} = 500$ nm) from the moQ unit. **P2** and **P3** both exhibit red emission ($\lambda_{\text{max}} = 608$ and 600 nm) from the DPAmoQ segment, and **P3** has a small emission shoulder at about 470 nm. **P4** and **P5** show green ($\lambda_{\text{max}} = 500$ nm) and red ($\lambda_{\text{max}} = 600$ nm) emission in their EL spectra, but **P5** has higher intensity of green emission due to its lower molar ratio of compound **7**. We can find the contribution of fluorene segment ($\lambda_{\text{max}} = 432$ nm) from the spectra of **P6**, **P7**, and **P8** and the coexistence of blue, green, and red emissions in those spectra.

The CIE coordinates of **P7** and **P8** are (0.33, 0.40) and (0.33, 0.36), respectively, and are close to the pure white-light emission of (0.33, 0.33).¹⁰ We can observe white emission from the devices of **P7** and **P8**, presented in the inset of Figure 5B. The stronger red and green emission in the EL spectra than in the PL spectra is attributed to the charge-trapping mechanism,¹¹ and the incomplete energy transfer from the polydioctylfluorene backbone to moQ and DPAmoQ segments provides EL spectra with red, green, and blue emission. The EL spectra of **P7** (Figure 6A) and **P8** (Figure 6B) are stable during increasing applied voltage. The results show that the copolymers in this study are suitable for display applications. Figure 7 shows the voltage–brightness characteristic and voltage–current efficiency of the EL devices, and their related performances are listed in Table 4. The maximum brightness and maximum current efficiency of the EL devices range from 2880 to $12\,300$ cd/m^2 and 0.77 to 2.56 cd/A . The device of **P7** emits near-white light with maximum brightness of $12\,300$ cd/m^2 , current efficiency of 2.02 cd/A , and a CIE coordinate of (0.33, 0.40). The devices of **P7** and **P8** exhibited higher brightness and more stable EL spectra during different bias than other polyfluorene-based copolymers containing red-, green-, and blue-emitting units in the backbone, which have been reviewed by Leclerc et al.¹²

Conclusions

We have obtained white-light-emitting devices from single polymer systems based on fluorene, moQ, and DPAmoQ units. The spectra of the EL devices can be adjusted to white emission by changing the molar ratios of the monomers. The devices of **P7** and **P8** exhibit maximum brightness of $12\,300$ and 7256 cd/m^2 as well as a maximum current efficiency of 2.02 and 1.87 cd/A with CIE coordinates of (0.33, 0.40) and (0.30, 0.36), respectively. This study demonstrates a useful method to prepare white-light-emitting diodes based on single polymer systems.

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