

## Supporting Information

### Dual-functional Conjugated Polymers Based on Trifluorene-2-yl-amine for RGB Organic Light-Emitting Diodes

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#### Materials.

All reagents were obtained from Sigma Aldrich Chemical Co., Alfa Aesar Chemical Co., Aladdin Chemical Co., and HuiCheng Chemical Co., and they were used as received unless otherwise specified. All manipulations involving air-sensitive reagents were performed in the atmosphere of dry argon. The solvents (THF, toluene) were purified by routine procedure and distilled under dry argon before being used.

#### Synthesis.

2-Nitrofluorene (**1**),<sup>1</sup> 3,6-di-*tert*-butyl-9H-carbazole,<sup>2</sup> 2,7-dibromo-9,9-dioctylfluorene,<sup>3</sup> and 2,7-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)-9,9-dioctylfluorene (**16**)<sup>4</sup> were prepared according to the reference procedures.

*2-Nitro-9,9-dioctylfluorene (2)*. To a mixture of **1** (5.30 g, 25 mmol), tetrabutyl ammonium bromide (0.45 g, 1.25 mmol), and octylbromide (12.8 g, 66.3 mmol) in 100 ml toluene, 50% (in weight) aqueous solution of NaOH (40 ml) was added rapidly under stirring. Then the mixture was heated to 60 °C and was stirred for 8 hours under a nitrogen atmosphere. After cooling to room temperature, 1 M hydrochloric acid was poured into the solution to quench and neutralize the reaction. Then 100 ml water was added and the mixture was extracted with ethyl acetate. The extract was washed with brine and dried over anhydrous magnesium sulfate. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give a pale yellow oil (10.0 g) in 91.5% yield. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.30-8.22 (dd, 1H), 8.20 (d, 1H), 7.80-7.78 (m, 2H), 7.43-7.38 (m, 3H), 2.05-1.20 (m, 4H), 1.28-1.03 (br, 20H), 0.80 (t, 6H), 0.59-0.52 (m, 4H).

*2-Amino-9,9-dioctylfluorene (3)*. **2** (4.38 g, 10 mmol) was dissolved in 50 ml EtOH at room temperature. To this was added 1.05 g of 5% Pd/C, and the mixture was stirred at room temperature under an argon atmosphere. Then hydrazine monohydrate (2.4 ml) was added dropwise via syringe over 20 min. The reaction mixture was stirred for 10 hours at 80 °C. After cooling to room temperature, the precipitate was separated by filtration and the solvent was removed by rotary evaporation, providing 4.08 g white solid in 100% yield. <sup>1</sup>H NMR (300 MHz, DMSO), δ (ppm): 7.50 (d, 1H), 7.40 (d, 1H), 7.25-7.09 (m, 3H), 6.55 (s, 1H), 6.50 (d, 1H), 5.18 (s, 2H), 1.92-1.70 (m, 4H), 1.23-0.90 (br, 20H), 0.80 (t, 6H), 0.65-0.40 (m, 4H).

*2-Bromine-7-nitro-9,9-dioctylfluorene (4)*. A mixture of **2** (5.46 g, 12.5 mmol) and iron (0.14 g, 2.44 mmol) in 50 ml chloroform was cooled with an ice bath. Then 1.3 ml liquid bromine (18.8 mmol) was added dropwise into

the mixture via dropping funnel. The mixture was stirred at 0 °C for 9 h. The solution of sodium thiosulfate was then poured into the mixture to quench the reaction. After that, the mixture was extracted with dichloromethane. The extract was washed with brine and dried over anhydrous magnesium sulfate. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give a pale yellow solid (5.42 g) in 84.0% yield. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.28-8.24 (dd, 1H), 8.18 (d, 1H), 7.80-7.74 (d, 1H), 7.67-7.62 (m, 1H), 7.55-7.52 (m, 2H), 2.08-1.92 (m, 4H), 1.24-1.04 (br, 20H), 0.81 (t, 6H), 0.70-0.50 (m, 4H).

**2-Cyano-7-nitro-9,9-dioctylfluorene (5).** Under an argon atmosphere, a mixture of **4** (2.05 g, 4 mmol) and cuprous cyanide (0.39 g, 4 mmol) in 30 ml dimethyl formamide (DMF) was heated to 150 °C and was stirred for 15 h. After cooling to room temperature, the mixture was poured into water and extracted with dichloromethane. The extract was washed with brine and dried over anhydrous magnesium sulfate. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give a reddish brown solid (1.53 g) in 83.2% yield. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.32 (d, 1H), 8.24 (s, 1H), 7.89 (d, 2H), 7.70 (m, 2H), 2.08-2.01 (m, 4H), 1.25-1.06 (br, 20H), 0.80 (t, 6H), 0.60-0.40 (m, 4H).

**9-(7-Nitro-9,9-dioctyl-9H-fluorene-2-yl)-9H-carbazole (6).** Under an argon atmosphere, a mixture of **4** (1.03 g, 2 mmol), carbazole (0.33 g, 2 mmol), potassium carbonate (0.85 g, 6 mmol) and copper (5 g, 78 mmol) in 30 ml dimethyl sulfoxide (DMSO) was heated to 150 °C and was stirred for 12 h. After cooling to room temperature, the mixture was poured into water and extracted with dichloromethane. The extract was washed with brine and dried over anhydrous magnesium sulfate. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give a reddish brown solid (0.32 g) in 26.7% yield. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.36-8.34 (d, 1H), 8.28 (s, 1H), 8.20 (d, 2H), 8.04-8.02 (d, 1H), 7.91-7.88 (d, 1H), 7.67-7.63 (m, 2H), 7.45 (m, 4H), 7.37-7.32 (m, 2H), 2.09 (m, 4H), 1.28-1.12 (br, 20H), 0.90-0.60 (m, 10H).

**3,6-Di-tert-butyl-9-(7-nitro-9,9-dioctyl-9H-fluorene-2-yl)-9H-carbazole (7).** Under an argon atmosphere, a mixture of 3,6-di-tert-butyl-9H-carbazole (1.86 g, 6.63 mmol), **4** (3.09 g, 6.02 mmol), CuI (0.14 g, 0.74 mmol), K<sub>2</sub>CO<sub>3</sub> (1.25 g, 9.02 mmol) and 18-crown-6 (0.16 g, 0.61 mmol) in 5 ml *N,N'*-dimethylpropylene urea (DMPU) was heated to 180 °C and stirred for 24 h. After cooling to room temperature, the mixture was poured into water and extracted with dichloromethane. The extract was washed with brine and dried over anhydrous magnesium sulfate. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give orange solid (3.09 g) in 76.0% yield. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.33-8.30 (dd, 1H), 8.25 (d, 1H), 8.17 (d, 2H), 7.99-7.97 (d, 1H), 7.87-7.84 (d, 1H), 7.64 (d, 1H), 7.61 (s, 1H), 7.50 (d, 1H), 7.47 (d, 1H), 7.41 (s, 1H), 7.38 (s, 1H), 2.10-2.00 (m, 4H), 1.48 (s, 18H), 1.30-1.02 (br, 20H), 0.83-0.78 (t, 6H), 0.77-0.60 (m, 4H).

**2-Amino-7-cyano-9,9-dioctylfluorene (8).** **8** was synthesized by following the similar procedure of **3**. Yield: 72.3%. <sup>1</sup>H NMR (300 MHz, DMSO), δ (ppm): 7.75 (s, 1H), 7.70-7.60 (m, 2H), 7.53 (d, 1H), 6.60-6.50 (m, 2H), 5.55 (s, 2H), 2.05-1.73 (m, 4H), 1.22-0.90 (br, 20H), 0.78 (t, 6H), 0.60-0.32 (m, 4H).

**9-(7-Amino-9,9-dioctyl-9H-fluorene-2-yl)-9H-carbazole (9).** **9** was synthesized by following the similar procedure of **3**. Yield: 58.9%. <sup>1</sup>H NMR (300 MHz, DMSO), δ (ppm): 8.25 (m, 2H), 7.77 (m, 1H), 7.52 (m, 2H), 7.41 (m, 3H),

7.30 (m, 4H), 6.60 (m, 2H), 5.30 (s, 2H), 2.10-1.75 (m, 4H), 1.40-0.90 (br, 20H), 0.90-0.50 (m, 10H).

*7-(3,6-Di-tert-butyl-carbazol-9-yl)-9,9-dioctyl-9H-fluoren-2-ylamine (10)*. **10** was synthesized by following the similar procedure of **3**. Yield: 96.4%. <sup>1</sup>H NMR (300 MHz, DMSO),  $\delta$  (ppm): 8.29 (s, 2H), 7.75-7.72 (d, 1H), 7.52-7.32 (m, 5H), 7.28 (s, 1H), 7.22 (s, 1H), 6.61-6.57 (m, 2H), 5.28 (s, 2H), 2.06-1.70 (m, 4H), 1.41 (s, 18H), 1.20-1.00 (br, 20H), 0.80-0.50 (m, 10H).

*Bis-(7-bromo-9,9-dioctyl-9H-fluoren-2-yl)-(9,9-dioctyl-9H-fluoren-2-yl)-amine (11)*. A mixture of Pd<sub>2</sub>(dba)<sub>3</sub> (0.576 g, 0.0628 mmol), diphenylphosphino-ferrocene (DPPF) (70.4 mg, 0.125 mmol), 2,7-dibromo-9,9-dioctylfluorene (2.23 g, 4.77 mmol), **3** (0.92 g, 2.27 mmol), and *t*-BuONa (0.88 g, 9.13 mmol) was stirred in toluene (20 ml) for 24 h at 110 °C under an argon atmosphere. After cooling to room temperature, the reaction mixture was neutralized with 1 M HCl solution. Then the reaction mixture was extracted with ethyl acetate. The organic extracts were combined and washed with brine and dried with anhydrous MgSO<sub>4</sub>. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give a pale yellow and viscous liquid 2.14 g. Yield: 80.0%. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>),  $\delta$  (ppm): 7.70-7.40 (m, 10H), 7.38-7.27 (m, 4H), 7.24-7.20 (m, 2H), 7.02 (d, 3H), 1.96-1.78 (m, 12H), 1.25-1.07 (br, 60H), 0.87-0.67 (m, 30H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 75 MHz),  $\delta$  (ppm): 152.66, 152.05, 151.67, 150.41, 147.43, 146.77, 140.87, 140.03, 136.47, 134.88, 129.95, 126.80, 126.39, 126.03, 123.24, 122.90, 122.75, 120.43, 120.31, 120.12, 119.07, 118.56, 117.91, 55.41, 55.10, 40.38, 40.29, 31.84, 30.11, 30.03, 29.41, 29.28, 23.93, 22.63, 14.09.

*7-[Bis-(7-bromo-9,9-dioctyl-9H-fluoren-2-yl)-amino]-9,9-dioctyl-9H-fluorene-2-carbonitrile (12)*. **12** was synthesized by following the similar procedure of **11**. Yield: 78.8%. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>),  $\delta$  (ppm): 7.66-7.60 (m, 2H), 7.60-7.50 (m, 4H), 7.50-7.42 (m, 5H), 7.25-7.20 (m, 4H), 7.01 (d, 3H), 2.07-1.81 (m, 12H), 1.27-1.09 (br, 60H), 0.89-0.69 (m, 30H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 75 MHz),  $\delta$  (ppm): 152.88, 152.69, 151.83, 150.97, 148.63, 146.92, 145.58, 139.79, 135.69, 133.55, 131.45, 130.05, 126.23, 126.08, 123.66, 121.54, 120.60, 120.45, 120.00, 119.31, 118.75, 116.89, 108.83, 55.45, 40.24, 40.11, 31.83, 31.79, 30.33, 30.00, 29.69, 29.39, 29.25, 23.94, 22.62, 14.07.

*Bis-(7-bromo-9,9-dioctyl-9H-fluoren-2-yl)-(7-carbazol-9-yl-9,9-dioctyl-9H-fluoren-2-yl)-amine (13)*. **13** was synthesized by following the similar procedure of **11**. Yield: 68.9%. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>),  $\delta$  (ppm): 8.18 (d, 2H), 7.80 (d, 1H), 7.61 (d, 1H), 7.54-7.42 (m, 12H), 7.32-7.26 (m, 4H), 7.26-7.25 (m, 3H), 7.05-7.03 (d, 3H), 1.88 (m, 12H), 1.25-1.09 (br, 60H), 0.86-0.68 (m, 30H).

*Bis-(7-bromo-9,9-dioctyl-9H-fluoren-2-yl)-[7-(3,6-di-tert-butyl-carbazol-9-yl)-9,9-dioctyl-9H-fluoren-2-yl]-amine (14)*. **14** was synthesized by following the similar procedure of **11**. Yield: 50.6%. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>),  $\delta$  (ppm): 8.17 (s, 2H), 7.87-7.72 (m, 1H), 7.67-7.34 (m, 15H), 7.33-7.27 (m, 1H), 7.25-7.16 (m, 2H), 7.15-6.82 (m, 3H), 2.03-0.80 (m, 12H), 1.48 (s, 18H), 1.30-1.00 (br, 60H), 0.91-0.55 (m, 30H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 75 MHz),  $\delta$  (ppm): 152.70, 152.36, 152.14, 151.76, 147.37, 147.10, 142.76, 140.00, 139.77, 139.45, 136.26, 135.57, 135.12, 130.00, 126.07, 125.47, 123.54, 123.37, 123.13, 121.40, 120.52, 120.38, 120.23, 120.01, 118.19, 116.30, 109.21, 67.97, 55.45, 55.42, 40.31, 34.75, 32.05, 31.91, 31.86, 30.13, 30.05, 29.71, 29.54, 29.43, 29.35, 29.29, 25.63, 24.20, 23.97, 22.66, 14.11.

*Bis-(7-bromo-9,9-dioctyl-9H-fluoren-2-yl)-[9,9-dioctyl-7-(5-phenyl-[1,3,4]oxadiazol-2-yl)-9H-fluoren-2-yl]-amine (15)*. Under an argon atmosphere, a mixture of **12** (0.74 g, 0.54 mmol), sodium azide (0.22 g, 3.45 mmol), and

ammonium chloride (0.20 g, 3.65 mmol) was stirred in 30 ml dry DMF for 36 h at 100 °C. After cooling to room temperature, HCl solution was added into the reaction mixture to acidity. Then the reaction mixture was extracted with ethyl acetate. The organic extracts were combined and washed with brine and dried with anhydrous MgSO<sub>4</sub>. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column give a reddish brown intermediates tetrazolium 0.37 g. The obtained tetrazolium (0.34 g, 0.24 mmol) was dissolved in 40 ml anhydrous pyridine under an argon atmosphere. 2 ml of benzoyl chloride (4.27 mmol) was then added dropwise into the mixture via syringe. After that, the mixture was heated to reflux and stirred for 3 days. After cooling to room temperature, the reaction mixture was extracted with ethyl acetate. The organic extracts were combined and washed with brine and dried with anhydrous MgSO<sub>4</sub>. The precipitate was separated by filtration. The solvent was removed under reduced pressure, and the residue went through a silica-gel column to give reddish brown viscous liquid 0.41 g. Yield: 51.4%. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.20-8.17 (m, 2H), 8.11 (s, 1H), 8.09 (s, 1H), 7.74 (d, 1H), 7.61-7.43 (m, 11H), 7.27-7.26 (m, 2H), 7.25-7.24 (m, 2H), 7.03 (dt, 3H), 2.07-1.80 (m, 12H), 1.43-1.08 (br, 60H), 0.99-0.69 (m, 30H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 75 MHz), δ (ppm): 172.12, 164.36, 152.67, 151.75, 147.09, 139.84, 133.75, 131.66, 130.20, 129.99, 129.35, 129.07, 128.46, 126.97, 126.23, 126.04, 124.02, 123.44, 121.21, 120.42, 119.42, 118.43, 55.49, 55.43, 40.27, 31.83, 31.81, 30.02, 29.41, 29.27, 24.02, 23.93, 22.63, 22.60, 14.09, 14.05.

*General procedure for polymer synthesis.* Carefully purified **16** (0.5 mmol), 0.5 mmol **11** or **12** or **13**, or **14**, Pd(PPh<sub>3</sub>)<sub>4</sub> (0.5-2.0 mol%), 1 ml Et<sub>4</sub>NOH and 1 ml deionized water were dissolved in 8-15 ml toluene. The mixture was heated to 90-100 °C with vigorous stirring for 48 h under an argon atmosphere. At the end of polymerization, polymers were sequentially end-capped with phenylboronic acid and bromobenzene in order to avoid a carrier trap and luminescence quenching center formation by end groups in OLEDs.<sup>5</sup> The mixture was then poured into methanol. The precipitated material was recovered by filtration and was extracted with methanol and acetone utilizing a Soxhlet extractor to remove oligomers and catalyst residues. The resulted polymers were dried under vacuum. Yields: 50-80%.

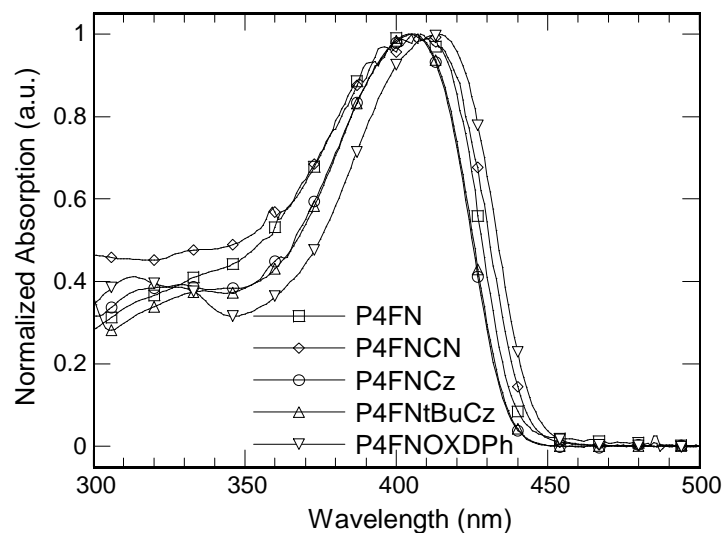
*P4FN.* <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 7.90-7.80 (m, 2H), 7.79-7.50 (m, 14H), 7.43-7.31 (br, 5H), 7.20-7.03 (br, 3H), 2.30-1.75 (m, 16H), 1.40-1.00 (m, 80H), 0.99-0.50 (m, 40H). Anal. Calcd. for [C<sub>116</sub>H<sub>161</sub>N]<sub>n</sub>: C, 88.77; H, 10.34; N, 0.89. Found: C, 85.01; H, 10.35; N, 0.91.

*P4FNCN.* <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 7.88-7.78 (m, 2H), 7.77-7.52 (br, 15H), 7.37-7.27 (m, 4H), 7.17-7.02 (br, 3H), 2.30-1.75 (m, 16H), 1.40-1.00 (m, 80H), 0.99-0.48 (m, 40H). Anal. Calcd. for [C<sub>117</sub>H<sub>160</sub>N<sub>2</sub>]<sub>n</sub>: C, 88.13; H, 10.11; N, 1.76. Found: C, 87.00; H, 10.70; N, 1.68.

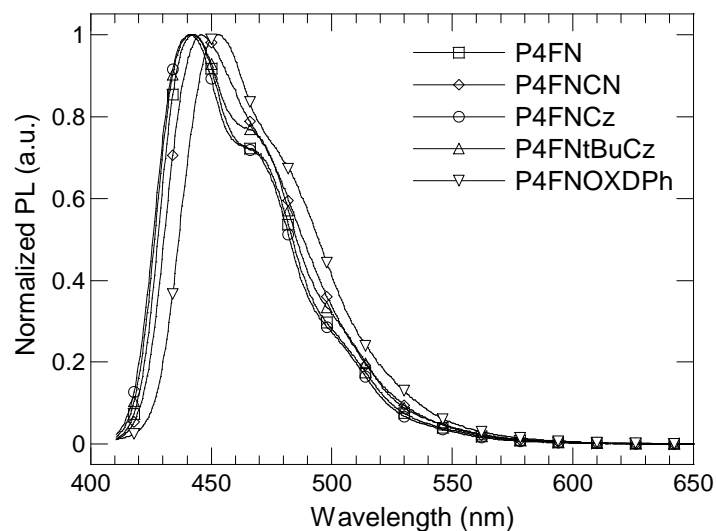
*P4FNCz.* <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.20-8.16 (d, 2H), 7.90-7.75 (m, 4H), 7.74-7.57 (br, 12H), 7.56-7.48 (m, 3H), 7.47-7.37 (br, 5H), 7.36-7.27 (br, 4H), 7.20-6.90 (br, 2H), 2.30-1.80 (m, 16H), 1.40-1.00 (br, 80H), 0.99-0.50 (m, 40H). Anal. Calcd. for [C<sub>128</sub>H<sub>168</sub>N<sub>2</sub>]<sub>n</sub>: C, 88.62; H, 9.76; N, 1.61. Found: C, 82.55; H, 9.89; N, 1.46.

*P4FNtBuCz.* <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>), δ (ppm): 8.17 (s, 2H), 7.87-7.77 (m, 3H), 7.76-7.57 (m, 12H), 7.56-7.44 (m, 5H), 7.43-7.30 (m, 5H), 7.19-7.02 (m, 3H), 2.25-1.80 (m, 16H), 1.49 (s, 18H), 1.40-1.02 (br, 80H), 0.99-0.55 (m, 40H). Anal. Calcd. for [C<sub>136</sub>H<sub>184</sub>N<sub>2</sub>]<sub>n</sub>: C, 88.44; H, 10.04; N, 1.52. Found: C, 86.83; H, 11.07; N, 1.45.

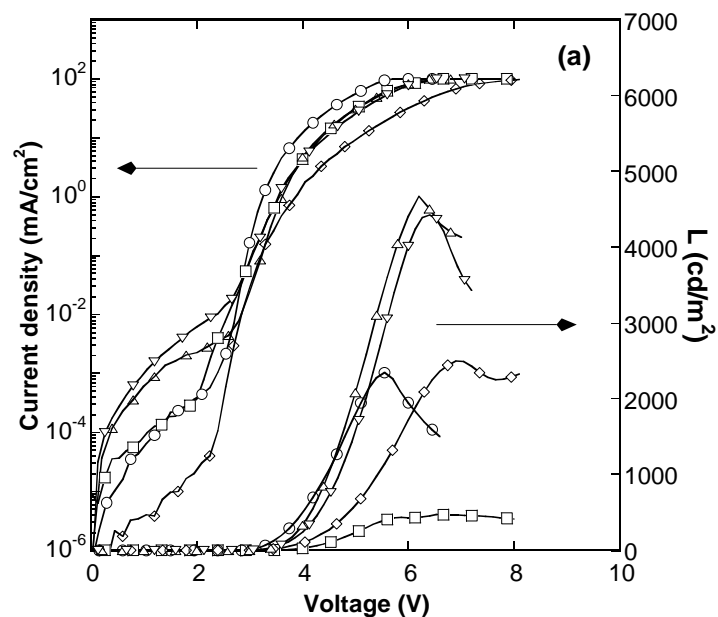
**P4FNOXDPh.** P4FNOXDPh was synthesized by following the former general procedure, except  $\text{Pd}(\text{PPh}_3)_4$  was replaced by  $\text{Pd}(\text{OAc})_2$  and tricyclohexyl phosphine ( $\text{PCy}_3$ ).  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ),  $\delta$  (ppm): 8.24-8.04 (m, 4H), 7.87-7.50 (br, 18H), 7.40-7.27 (br, 4H), 7.16-7.03 (br, 3H), 2.30-1.76 (m, 16H), 1.40-1.00 (br, 80H), 0.99-0.43 (m, 40H). Anal. Calcd. for  $[\text{C}_{124}\text{H}_{165}\text{ON}_3]_n$ : C, 86.91; H, 9.71; N, 2.45. Found: C, 86.28; H, 9.72; N, 2.43.



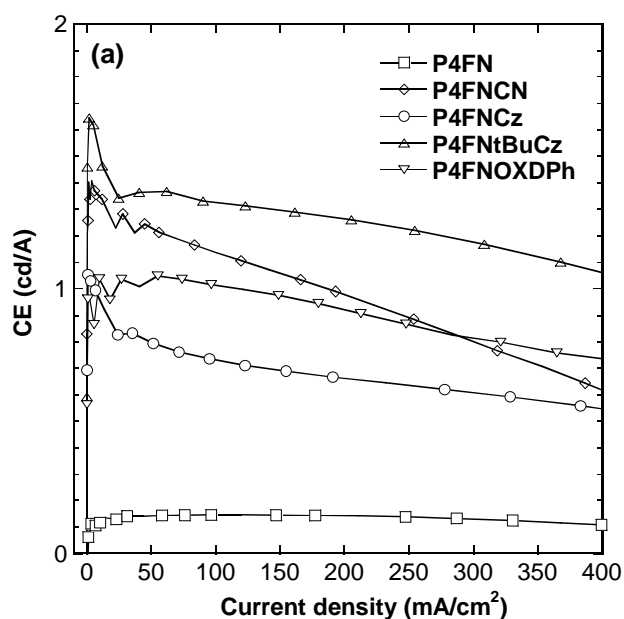
**Figure S1.** Normalized UV-vis absorption spectra of the polymers in dilute toluene solutions.



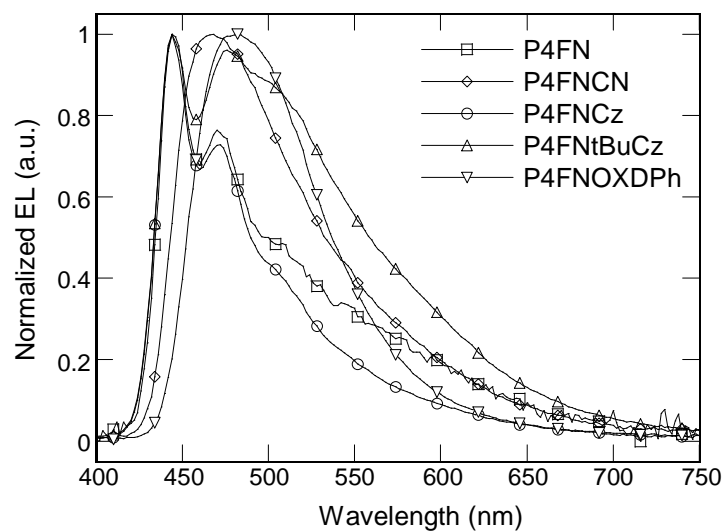
**Figure S2.** Normalized PL spectra of the polymers in dilute toluene solutions.



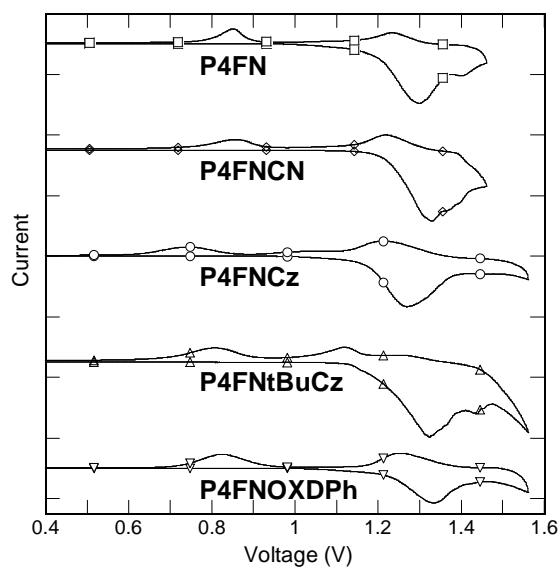
**Figure S3.** Current density and luminance versus voltage characteristics of the single-layer devices with the configuration of ITO / PEDOT:PSS (40 nm) / EML (70~80 nm) / CsF (1.5 nm) / Al. EML: P4FN (□), P4FNCN (◇), P4FNCz (○), P4FNtBuCz (▽), and P4FNOXDPh (△).



**Figure S4.** Current efficiency (CE) versus current density characteristics of the single-layer devices with the configuration of ITO / PEDOT:PSS (40 nm) / EML (70~80 nm) / CsF (1.5 nm) / Al.



**Figure S5.** EL spectra of the single-layer devices based on the polymers as an EML in the configuration of ITO / PEDOT:PSS (40 nm) / EML (70~80 nm) / CsF (1.5 nm) / Al.



**Figure S6.** Cyclic voltammograms of the polymers in thin films coated from their toluene solutions measured in 0.1 M Bu<sub>4</sub>NPF<sub>6</sub> in anhydrous acetonitrile at a scan rate of 50 mV s<sup>-1</sup>. The potentials are calibrated to the Fc<sup>+</sup>/Fc external standard.

**Table S1.** Summary of the electroluminescent performance of the single-layer devices with the configuration of ITO / PEDOT:PSS (40 nm) / EML (70~80 nm) / CsF (1.5 nm) / Al.

EML	$V_{\text{on}}^a$ (V)	$L_{\text{max}}$ (cd m <sup>-2</sup> )	$\text{CE}_{\text{max}}$ (cd A <sup>-1</sup> )	$\text{EQE}_{\text{max}}$ (%)	CIE (x, y)
P4FN	3.3	478	0.15	0.16	(0.23, 0.25)
P4FNCN	3.0	2500	1.41	1.07	(0.22, 0.30)
P4FNCz	2.8	2274	1.05	1.08	(0.19, 0.20)
P4FNtBuCz	3.0	4675	1.64	0.96	(0.24, 0.32)
P4FNOXDPh	3.0	4556	1.05	0.66	(0.19, 0.35)

<sup>a</sup> Turn-on voltage for electroluminescence (a luminance of 1 cd m<sup>-2</sup> was detected).

## References

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