

# Three-Color White Electroluminescence from a Single Polymer System with Blue, Green and Red Dopant Units as Individual Emissive Species and Polyfluorene as Individual Polymer Host\*\*

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White polymer light emitting diodes (WPLEDs) have attracted considerable attention because of their potential utilization in full color flat-panel displays, backlights and solid-state lighting source.<sup>[1–25]</sup> An ideal white emission from polymer light emitting diodes should have not only good CIE (Commission Internationale d'Eclairage) coordinates and high electroluminescence (EL) efficiencies but also excellent color rendering index (CRI) (CRI is a numerical measurement of how “true” color looks when viewed with the lighting source. CRI values range from 0 to 100 with 100 represents true color reproduction.) and long device operational lifetime.

Recent research efforts in the field of WPLEDs have focused on white EL from a single polymer owing to its advantage of high EL efficiency, good color stability and no phase separation.<sup>[7–25]</sup> Our group has proposed two strategies to develop white electroluminescent single polymers. One is to incorporate an orange chromophore to a blue light emitting polymer host for blue-orange two-color white EL.<sup>[11]</sup> The color purity of white EL of these polymers is enhanced by tuning the emission wavelength of the orange chromophore for the matched wavelength of the orange component and the blue component.<sup>[12]</sup> The EL efficiencies of these polymers are improved by enhancing the photoluminescence (PL) quantum efficiency of the orange species,<sup>[13]</sup> by covalently attaching the orange chromophore to the side chain of the blue polymer host to form intramolecular dopant/host system,<sup>[14]</sup> and by enhancing the efficiency and redshifting the emission spectrum of the blue species<sup>[15]</sup>. As a result, a luminous efficiency of 12.8 cd A<sup>-1</sup>, CIE coordinates of (0.31, 0.36) and CRI of 75 is achieved with

their single-layer devices.<sup>[15]</sup> However, EL spectra of these polymers cannot cover the whole visible range and have CRI values of lower than 80, which are not sufficient for practical applications. The other strategy of ours is to attach a red chromophore and a green chromophore to a blue emissive polymer host for blue-green-red three-color white EL.<sup>[16]</sup> We also find that EL performance of these polymers can be improved by covalently attaching both the green dopant unit and the red dopant unit to the side chain of the blue polymer host.<sup>[17]</sup> After the improvement, a luminous efficiency of 7.3 cd A<sup>-1</sup> is demonstrated.<sup>[17]</sup> Although ideal EL spectra with high CRI values have been demonstrated, white EL of this kind suffers from relatively low EL efficiencies. Recently, based on this strategy, Cao et al.<sup>[19]</sup> and Shu et al.<sup>[20]</sup> have both reported white electroluminescent polymers with a green unit and a red unit incorporated into the backbone of a blue polymer host. Luminous efficiencies of 6.1 cd A<sup>-1</sup> and 4.8 cd A<sup>-1</sup> have been demonstrated. The same two groups have also reported white electroluminescent polymers with triplet emission.<sup>[21,22]</sup> Owing to the involvement of triplet emission, devices based on these polymers exhibit high luminous efficiencies of 6.9 cd A<sup>-1</sup> and 8.2 cd A<sup>-1</sup>. Considering the virtues and drawbacks of the above two strategies, we think that optimization of white electroluminescent single polymer's efficiency and EL spectral quality tradeoff is an important issue for the field of WPLEDs. Most importantly, both the aforementioned two strategies need the polymer backbone function as both the polymer host and the blue light-emitting species. Unfortunately, most of polymer hosts themselves cannot emit light efficiently. Until now, only polyfluorene can meet these requirements although polyfluorene itself suffers from poor spectral stability and short device operational lifetime.<sup>[26,27]</sup>

Here, we propose a new approach to realize three-color white EL from a single polymer with both high EL efficiencies and high CRI values by using blue, green and red dopant units as individual emissive species and polyfluorene as individual polymer host. This is achieved by covalently attaching both a blue emissive dopant unit, a green emissive dopant unit and a red emissive dopant unit to the side chain of a polymer host (see Fig. 1). White EL is expected to come from the individual emission from the three dopant units without the contribution of the polymer host. The difference between this novel approach and our previous two strategies is that white emission comes from the dopant units in this approach but from both

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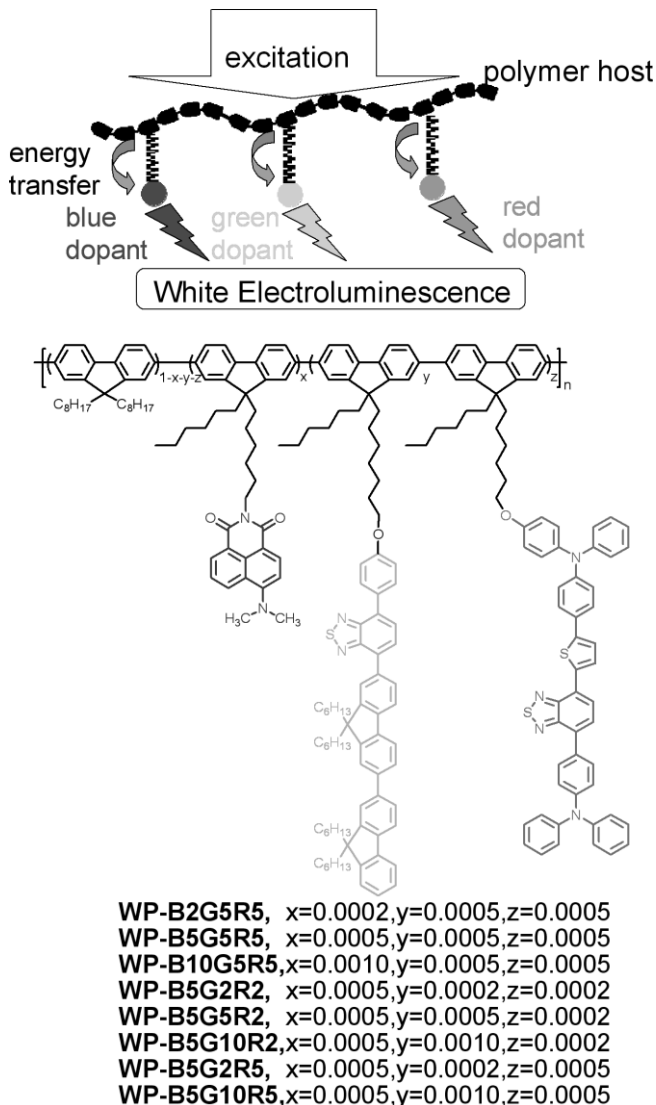


Figure 1. Schematic illustration and chemical structures of the polymers.

the dopant units and the polymer host in the previous two strategies. Our motivation of this approach is 3-folds and is explained as follows. (i) Since the emission comes from the three dopant units, high CRI values can be obtained after tuning the emission wavelength of the three dopant units. (ii) Because the blue emission comes from the blue dopant unit instead of from the polymer host, we can expect enhancement of EL efficiencies through the utilization of a blue dopant with high PL quantum efficiency. (iii) Because this approach does not require the polymer host to emit blue light, this approach will benefit from easy availability of polymer hosts besides polyfluorene<sup>[28,29]</sup> in the future. As a result, single layer device of the resulting polymers emit high-quality white light with CIE coordinates of (0.33, 0.36), CRI as high as 88, luminous efficiency up to 8.6 cd A<sup>-1</sup> and power efficiency as high as 5.3 lm W<sup>-1</sup>.

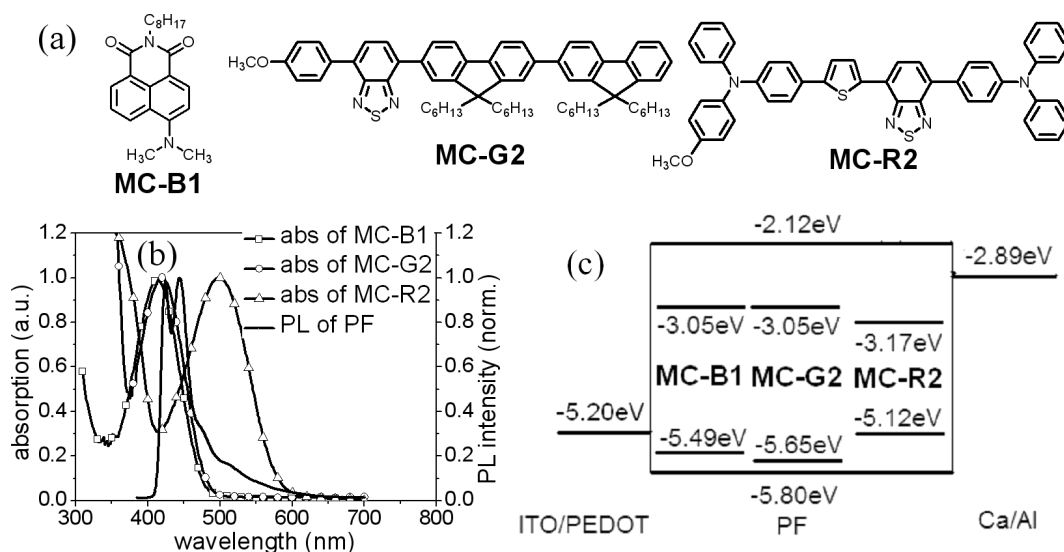
The chemical structures of the designed polymers are shown in Figure 1. We select polyfluorene as the polymer host

because of its good charge-carrier-transporting and film-forming properties as well as large bandgap.<sup>[27]</sup> DMAN unit<sup>[30]</sup> (its model compound: MC-B1, see Figure 2a. PL quantum efficiency:  $\Phi_{PL} = 0.84$ ), P-BT-FF unit (its model compound: MC-G2, see Fig. 2a.  $\Phi_{PL} = 0.73$ ) and TPA-T-BT-TPA unit (its model compound: MC-R2, see Fig. 2a.  $\Phi_{PL} = 0.59$ ) are chosen as the blue, green and red light-emitting dopant units, respectively. All the three dopant units have high PL quantum efficiencies and can form dopant/host system with polyfluorene in terms of energy levels and spectral overlap.<sup>[31]</sup> The dopant units are all covalently attached to the side chain of polyfluorene by alkyl spacers. In order to obtain blue, green and red emission balance, the contents of the dopant units are tuned within the range from 0.02 mol % to 0.10 mol %. For WP-B2G5R5, WP-B5G5R5 and WP-B10G5R5, the contents of the green dopant unit and red dopant unit are both fixed to be 0.05 mol % while the content of the blue dopant unit is adjusted as 0.02 mol %, 0.05 mol % and 0.10 mol %, respectively. Similar adjustment in dopant units' contents has also been carried out for the green dopant unit in WP-B5G2R2, WP-B5G5R2 and WP-B5G10R2 as well as for the red dopant unit in WP-B5G2R2, WP-B5G2R5 and WP-B5G2R10. All these polymers are soluble in common organic solvents, such as toluene, chloroform, tetrahydrofuran (THF), etc. Their number average molecular weights, as determined by gel permeation chromatography (GPC) with polystyrene as standards, range from  $7.03 \times 10^4$  to  $8.65 \times 10^4$  with polydispersity (PDI) ranging from 2.41 to 2.92.

Figure 3 shows the PL spectra of the polymers in solid films. All the polymers show dominant blue emission from the polymer backbone and weak emission from the blue, green, and red dopant units. The emission from the dopant units comes from the Förster energy transfer from the polymer host to the three dopant units because of the overlap of the emission spectrum of polyfluorene and absorption spectra of the three model compounds as shown in Figure 2b.

Single layer devices of the polymers were fabricated with the configuration of ITO/poly(styrene sulfonic acid):poly(ethylenedioxythiophene) (PEDOT:PSS) (40 nm)/polymer (90 nm)/Ca (10 nm)/Al (100 nm). The EL spectra of the devices are shown in Figure 4. The corresponding CIE coordinates and CRI values are listed in Table 1. All their EL spectra exhibit a very broad emission band covering the whole visible range from 400 nm to 700 nm with four emission peaks locating at about 593 nm, 508 nm, 475 nm, and 421 nm. The peak at 593 nm is due to the TPA-T-BT-TPA unit, the peak at 508 nm is attributed to the P-BT-FF unit, the peak at 475 nm arises from the DMAN unit and the peak at 420 nm originates from the polyfluorene backbone. The emission bands of the three dopant units are much stronger in the EL spectra compared to in the PL spectra because of the charge trapping effect of the three dopant units.<sup>[32]</sup> This is favored by the fact that the LUMO and HOMO energy levels of all the three model compounds lie between those of polyfluorene (see Fig. 2c).<sup>[31,32]</sup>

The relative intensity of the peaks in EL spectra can be tuned by adjusting the contents of the three dopant units. As shown



**Figure 2.** a) Chemical structures of the model compounds, **MC-B1**, **MC-G2**, and **MC-R2**. b) Absorption spectra of **MC-B1**, **MC-G2**, **MC-R2**, and PL spectrum of **PF**. c) LUMO and HOMO energy levels of **MC-B1**, **MC-G2**, **MC-R2**, and **PF**.

**Table 1.** EL performance of the devices of the polymers.

Polymer	Turn-on Voltage [V]	Luminous Efficiency [ $\text{cd A}^{-1}$ ]	Power Efficiency [ $\text{lm W}^{-1}$ ]	Maximum Brightness [ $\text{cd m}^{-2}$ ]	CIE Coordinates (x, y)	CRI
<b>WP-B2G5R5</b>	3.5	9.0	5.7	17700	(0.39, 0.40)	90
<b>WP-B5G5R5</b>	3.5	10.8	7.6	20510	(0.40, 0.42)	84
<b>WP-B10G5R5</b>	3.5	7.1	4.2	14590	(0.35, 0.38)	88
<b>WP-B5G2R2</b>	3.5	7.2	4.5	11230	(0.36, 0.36)	91
<b>WP-B5G5R2</b>	3.5	8.6	5.4	11510	(0.33, 0.36)	88
<b>WP-B5G10R2</b>	3.5	8.7	5.0	25450	(0.36, 0.46)	71
<b>WP-B5G2R5</b>	3.5	7.3	4.6	15690	(0.37, 0.37)	93
<b>WP-B5G2R10</b>	3.5	5.0	3.3	27320	(0.49, 0.38)	60

in Figure 4a, for **WP-B2G5R5**, **WP-B5G5R5** and **WP-B10G5R5** with the same contents of the green dopant unit and red dopant unit but different content of the blue dopant unit, the relative intensity of the blue emission band in the EL spectra increases with enhancing content of the blue dopant unit. This is due to the more excitons transferring from the polymer host to the blue dopant unit and the more charge carrier recombination directly on the blue dopant unit resulting from the higher content of the blue dopant unit.<sup>[19]</sup> Similar phenomenon has also been observed for the green emission band (Fig. 4b) and the red emission band (Fig. 4c). By adjusting the relative intensity of the blue, green and red emission band in the EL spectra, high quality white EL is obtained. For example, the CIE coordinates of **WP-B10G5R5**, **WP-B5G2R2**, **WP-B5G5R2**, and **WP-B5G2R5** are (0.35, 0.38), (0.36, 0.36), (0.33, 0.36), and (0.37, 0.37), respectively, which are very close to (0.33, 0.33) of standard white emission. The CRI values of them are 91, 88, 88 and 93, respectively, which are obviously higher than 65–75 of white electroluminescent polymers with an orange chromophore incorporated into a blue polymer

host.<sup>[11–15]</sup> The high CRI values are attributed to the matched emission wavelength of the three dopant units. As for the EL spectra of **WP-B2G5R5**, **WP-B5G5R5**, **WP-B5G10R2**, and **WP-B5G2R10**, the green emission band or the red emission band is much stronger than the blue emission band because of the relatively high content of the green or red dopant units. Their CIE coordinates are (0.39, 0.40), (0.40, 0.42), (0.36, 0.46), and (0.49, 0.38), respectively, indicating that their emission colors are light green or light orange. The residual emission from the polyfluorene backbone in the EL spectra cannot be completely diminished even when the contents of the dopant units are high.

All the devices of the polymers exhibit good spectral stability. Figure 5 shows the EL spectra of the device of **WP-B10G5R5** under different voltage. When the bias increases from 5 V to 10 V and the brightness increases from 91  $\text{cd m}^{-2}$  to 10750  $\text{cd m}^{-2}$ , the EL spectral shape keeps unchanged. The bias independent EL spectra are due to the molecular dispersion of the three dopant units in the polymer host.

The EL performance of the devices of the polymers is listed in Table 1. All the devices of the four white light emitting polymers, **WP-B10G5R5**, **WP-B5G2R2**, **WP-B5G5R2** and **WP-B5G2R5**, show excellent EL performance with the luminous efficiencies from 7.1  $\text{cd A}^{-1}$  to 8.6  $\text{cd A}^{-1}$  and power efficiencies from 4.2  $\text{lm W}^{-1}$  to 5.4  $\text{lm W}^{-1}$ . The supporting information shows the typical voltage-current density-brightness characteristic and current density-luminous efficiency-power efficiency curve of the device of **WP-B5G5R2**, which shows a turn on voltage of 3.5 V, luminous efficiency of 8.6  $\text{cd A}^{-1}$ , power efficiency of 5.4  $\text{lm W}^{-1}$  and maximum brightness of 11510  $\text{cd m}^{-2}$  with the CIE coordinates of (0.33, 0.36). This performance is somewhat better than that of white electrolu-

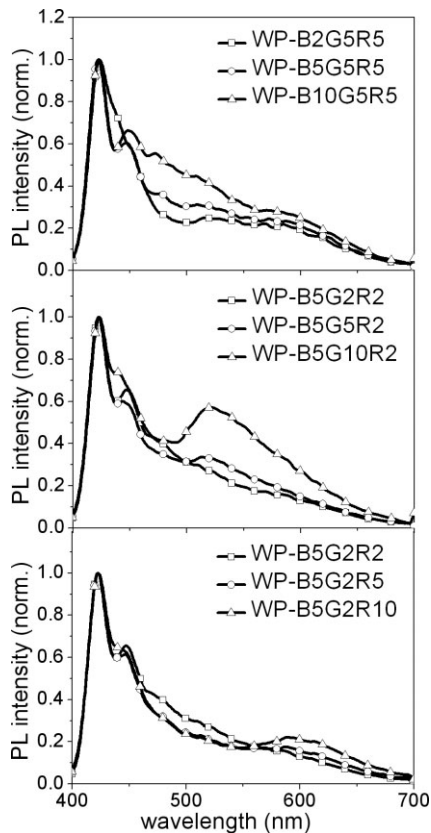


Figure 3. PL spectra of the polymers in solid films.

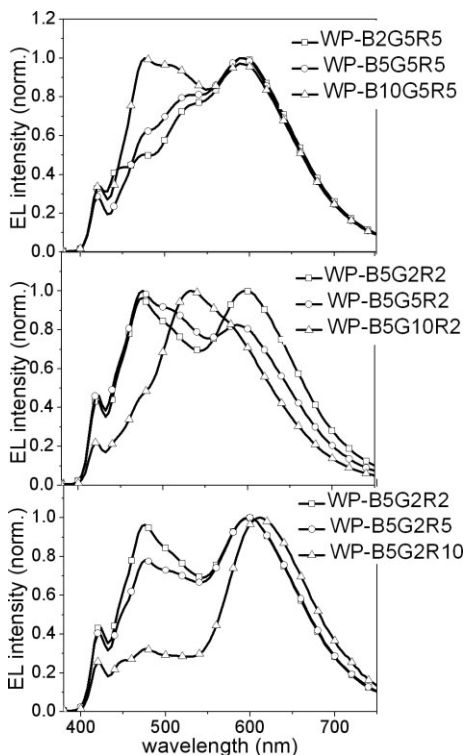


Figure 4. EL spectra of the devices of the polymers.

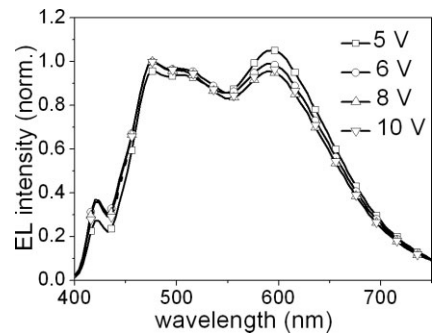


Figure 5. EL spectra of the device of WP-B10G5R5 at different bias.

minescent polymers with a green chromophore and a red chromophore incorporated into a blue polymer host in literatures.<sup>[16–25]</sup> We attribute the excellent EL performance of these polymers to the highly efficient blue emission from the blue DMAN dopant unit instead of from the polymer host.

In conclusion, by using blue, green and red dopant units as the individual emissive species and using polyfluorene as the individual polymer host, we have developed a series of white electroluminescent single polymers through covalently attaching the three dopant units to the side chain of the polymer host. Since white EL comes from the individual emission from the three dopant units, excellent EL performance and high CRI values are achieved. Single layer devices of these polymers emit white light with luminous efficiency of  $8.6 \text{ cd A}^{-1}$ , power efficiency of  $5.4 \text{ lm W}^{-1}$ , CIE coordinates of (0.33, 0.36) and CRI value of 88. These polymers seem to provide an avenue to design white electroluminescent single polymers with both excellent color quality and high EL efficiencies. Most importantly, the approach to design these polymers does not require the polymer host to emit blue light efficiently, therefore, this approach will benefit from easy availability of polymer host besides polyfluorene and is expected to realize long device operational lifetime in the future. Further investigation on white electroluminescent polymers without polyfluorene as the polymer host is in progress in our lab.

## Experimental

**General Procedure for Suzuki Polymerization:** To a mixture of 2,7-dibromo-9,9-dioctylfluorene (**1**), 9,9-dioctyl-2,7-bis(trimethyleneborate)fluorene (**2**),  $\text{Pd}(\text{PPh}_3)_4$  (0.0115 g, 0.01 mmol) under dry argon atmosphere was added one drop of Aliquat 336, degassed toluene (6 mL) and aqueous 2 M potassium carbonate (2.0 mL, 4 mmol). Solutions of  $5 \times 10^{-4} \text{ M}$  9-hexyl-9-(6-(4-(dimethylamino)-1,8-naphthalimide-9-yl)hexyl)-2,7-dibromofluorene (**Monomer-B1**) in toluene,  $5 \times 10^{-4} \text{ M}$  9-hexyl-9-(6-(4-(4-(2-(9,9-dihexylfluorene-2-yl)-9,9-dihexylfluorene-2-yl)-2,1,3-benzothiadiazole-7-yl)-phenoxy)-hexyl)-2,7-fluorene (**Monomer G2**) in toluene and  $5 \times 10^{-4} \text{ M}$  9-hexyl-9-(6-(4-(*N*-phenyl-*N*-(4-(5-(4-(4-(diphenylamino)phenyl)-2,1,3-benzothiadiazole-7-yl)-thienyl-2-yl)phenyl)amino)phenoxy)hexyl)-2,7-dibromofluorene (**Monomer-R2**) in toluene with corresponding volumes were also added to this mixture. The mixture was heated to  $90^\circ\text{C}$  in dark and kept stirred for 8 hours. Then 6 mL toluene was added to the reaction mixture to facilitate stirring. The resulting mixture was kept stirred for another

16 hours. Then the polymer was endcapped by adding **2** (3 mL 0.1 M solution in degassed toluene) followed by stirring for 6 hours, and by adding bromobenzene (1 mL 1 M solution in degassed toluene) followed by stirring for 8 hours. The whole mixture was poured into methanol. The precipitate was collected by filtration and then dissolved in chloroform. The solution was washed with water, dried with anhydrous Na<sub>2</sub>SO<sub>4</sub> and then concentrated to an appropriate volume. The fiber-like polymer was obtained by pouring the concentrated solution into methanol. After washing with acetone for 24 hours in a Soxhlet apparatus, the polymer was further purified with reprecipitation in chloroform/methanol for several times. The final product was obtained after drying in vacuum with a yield of 60–70%.

**PF: 1** (0.2792 g, 0.5000 mmol), **2** (0.2742 g, 0.5000 mmol) were used in the polymerization. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>, δ): 7.87 (d, 2H), 7.72 (br, 4H), 2.10 (br, 4H), 1.14 (br, 24H), 0.81 (t, 6H). Anal. calcd: C, 89.69; H, 10.31. Found: C, 89.08; H, 10.02. GPC: *M<sub>n</sub>* = 1.28 × 10<sup>5</sup>, PDI = 2.39. **WP-B2G5R5: 1** (0.2734 g, 0.4985 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol), **Monomer-G2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) and **Monomer-R2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 7.85 × 10<sup>4</sup>, PDI = 2.41. **WP-B5G5R5: 1** (0.2734 g, 0.4985 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol), **Monomer-G2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) and **Monomer-R2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 8.17 × 10<sup>4</sup>, PDI = 2.72. **WP-B10G5R5: 1** (0.2731 g, 0.4980 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (2.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 1 × 10<sup>-3</sup> mmol), **Monomer-G2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) and **Monomer-R2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 7.19 × 10<sup>4</sup>, PDI = 2.92. **WP-B5G2R2: 1** (0.2737 g, 0.4990 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol), **Monomer-G2** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol) and **Monomer-R2** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 8.65 × 10<sup>4</sup>, PDI = 2.50. **WP-B5G5R2: 1** (0.2734 g, 0.4985 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol), **Monomer-G2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) and **Monomer-R2** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 7.25 × 10<sup>4</sup>, PDI = 2.91. **WP-B5G10R2: 1** (0.2731 g, 0.4980 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol), **Monomer-G2** (2.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 1 × 10<sup>-3</sup> mmol) and **Monomer-R2** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 7.27 × 10<sup>4</sup>, PDI = 2.80. **WP-B5G2R5: 1** (0.2734 g, 0.4985 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol), **Monomer-G2** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol) and **Monomer-R2** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 7.03 × 10<sup>4</sup>, PDI = 2.63. **WP-B5G2R10: 1** (0.2731 g, 0.4980 mmol), **2** (0.2792 g, 0.5000 mmol), **Monomer-B1** (1.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 5 × 10<sup>-4</sup> mmol), **Monomer-G2** (0.4 mL 5 × 10<sup>-4</sup> M solution in toluene, 2 × 10<sup>-4</sup> mmol) and **Monomer-R2** (2.0 mL 5 × 10<sup>-4</sup> M solution in toluene, 1 × 10<sup>-3</sup> mmol) were used in the polymerization. GPC: *M<sub>n</sub>* = 8.28 × 10<sup>4</sup>, PDI = 2.47.

**Device Fabrication:** The indium-tin oxide (ITO) glass plates were degreased in ultrasonic solvent bath and then dried at 120 °C for 15 minutes. The PEDOT:PSS layer with an approximate thickness of 40 nm was spin-coated on the treated ITO at 3000 rpm for 60 s and then baked for 15 minutes at 120 °C. The polymer layer (approximate 90 nm) was then spin-coated on top of the PEDOT/ITO coated glass substrate with its fresh toluene solution (10 mg/ml) at ambient atmosphere. Finally, a thin layer of calcium (10 nm) followed by a layer of aluminum (100 nm) was deposited in a vacuum thermal evaporator through a shadow mask at a pressure of 5 × 10<sup>-3</sup> Pa. The active area of the devices was 10 mm<sup>2</sup>. The EL spectra, CIE coordinates, current-voltage and brightness-voltage characteristics of the devices were recorded

with a Spectrascan PR650 spectrophotometer at the forward direction and a computer-controlled Keithley 2400 under ambient condition.

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