

Electronic Supporting Information for:

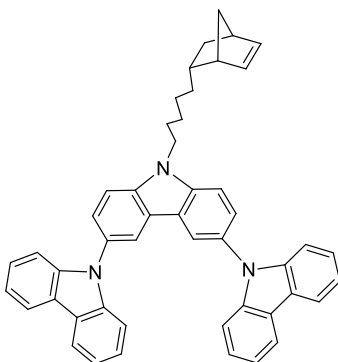
Phosphorescent light-emitting diodes using triscarbazole/bis(oxadiazole) hosts: comparison of homopolymer blends and random and block copolymers

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Contents

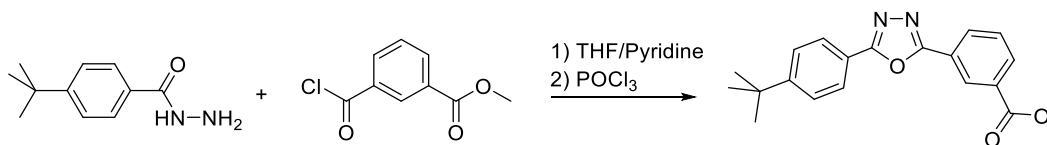
I. Synthesis of Monomers 1 and 2	pS1
II. Synthesis of Sty ₂ TCz	pS7
III. Solid-State NMR Spectra for ¹ H Spin-Diffusion Experiments	pS10
IV. Spin-Spin Relaxation and ¹ H Spin Diffusion Coefficients	pS12
V. Summary of Optical Data for Polymeric Hosts	pS13
VI. Performance Details of the Green-Emitting Devices	pS13
VII. References	pS16

I. Synthesis of Monomers **1** and **2**



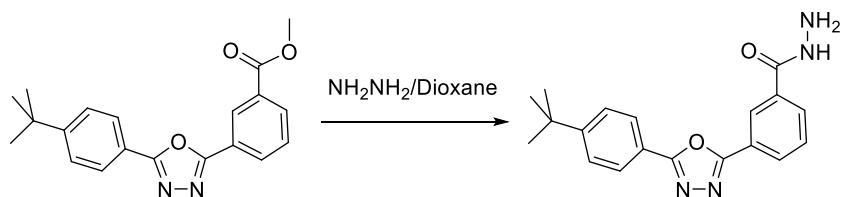
Monomer 1: K₂CO₃ (13.99 g, 101.22 mmol) was added to a mixture of triscarbazole¹ (10.15 g, 20.40 mmol), 5-(5-bromopentyl)-norbornene² (10 mL, 50.29 mmol, ca. 3:17 mixture of *exo*- and *endo*-isomers), and DMF (100 mL) under N₂. The reaction was heated to 100 °C for 60 h. The reaction mixture was then cooled to room temperature and water (50 mL) was added to precipitate a yellow solid. The crude product was isolated by filtration and purified by

recrystallization from dichloromethane and column chromatography (silica gel, eluting with CH₂Cl₂) to give a white solid (12.09 g, 90%). ¹H NMR (400 MHz, CDCl₃): δ 8.21 (s, br, 2H), 8.15 (d, *J* = 7.6 Hz, br, 4H), 7.65 (s, br, 4H), 7.38 (q, *J* = 8 Hz, 4H), 7.37 (s, br, 4H), 7.27 (dd, *J* = 6.0, 2.0 Hz, 2H), 7.25 (dd, *J* = 6.0, 2.0 Hz, 2H), 6.10 (dd, *J* = 6.0, 2.8 Hz, C=C-H, 0.85 H, endo), 6.07 (dd, *J* = 5.6, 3.2 Hz, C=C-H, 0.15 H, exo), 6.00 (dd, *J* = 5.6, 3.2 Hz, C=C-H, 0.15 H, exo), 5.90 (dd, *J* = 6.0, 2.8 Hz, C=C-H, 0.85 H, endo), 4.47 (t, *J* = 7.2 Hz, 2H), 2.75 (s, br, 2H), 2.06~1.94 (m, 3H), 1.85 (dd, *J*₁ = 11.2 Hz, *J*₂ = 4.0 Hz, 1H), 1.53~1.06 (m, 8H), 0.49 (ddd, *J*₁ = 11.2 Hz, *J*₂ = 4.0 Hz, *J*₃ = 2.8 Hz, 1H). ¹³C{¹H}NMR (100 MHz, CDCl₃): δ 142.06, 140.39, 137.29, 132.52, 129.51, 126.16, 126.05, 123.59, 123.31, 120.48, 120.04, 119.82, 110.32, 109.94, 49.82, 45.66, 43.91, 42.74, 38.97, 34.94, 32.68, 29.40, 28.74, 27.89. Anal. Calcd. for C₄₈H₄₁N₃: C, 87.37; H, 6.26; N, 6.37. Found: C, 87.28; H, 6.30; N, 6.37. EI-MS: *m/z* 659.3 (M⁺).



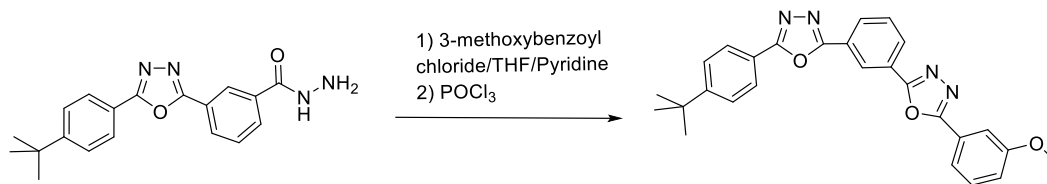
Methyl 3-(5-(4-*tert*-butylphenyl)-1,3,4-oxadiazol-2-yl)benzoate: To a solution of 4-*tert*-butylbenzohydrazide (37.0 g, 192 mmol) in dry tetrahydrofuran (500 mL) cooled in ice-water was slowly added methyl 3-(chlorocarbonyl)benzoate (37.0 g, 186 mmol). During the addition, a white solid was appeared. The reaction mixture was then stirred at room temperature for 17 h.; pyridine (100 mL) was added and the reaction mixture was stirred for 1 h. The reaction mixture was poured into water (1500 mL) and the white solid was collected by filtration and dried overnight under vacuum. This solid, methyl 3-(2-(4-*tert*-butylbenzoyl)hydrazinecarbonyl)-benzoate, was used in the next step without characterization or purification; it was suspended in POCl₃ (200 mL) and heated to 90 °C and for 6 h. After cooling to room temperature the reaction mixture was poured into ice-water (4000 mL) and the resulting brown-colored solid was

collected by vacuum filtration. The crude product was dried and purified by column chromatography (silica gel, 19:1 dichloromethane/ethyl acetate as eluent). After removal of solvents and recrystallization from acetone/water, a white solid was obtained (40.2 g, 64%). ^1H NMR (400 MHz, CDCl_3): δ 8.77 (t, $J = 1.2$ Hz, 1H), 8.36 (dt, $J_1 = 7.6$ Hz, $J_2 = 1.2$ Hz, 1H), 8.22 (dt, $J_1 = 7.6$ Hz, $J_2 = 1.2$ Hz, 1H), 8.09 (d, $J = 8.8$ Hz, 2H), 7.64 (t, $J = 7.6$ Hz, 1H), 7.56 (d, $J = 8.8$ Hz, 2H), 4.00 (s, 3H, OCH_3), 1.38 (s, 9H, $^t\text{Bu CH}_3$). $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 166.05, 164.97, 163.56, 155.54, 132.43, 131.17, 131.00, 129.30, 127.82, 126.84, 126.07, 124.44, 120.82, 52.48, 35.09, 31.08. MS(EI): m/z 336 (M^+). Anal. Calcd for $\text{C}_{20}\text{H}_{20}\text{N}_2\text{O}_3$: C, 71.41; H, 5.99; N, 8.33. Found: C, 71.45; H, 6.01; N, 8.33.



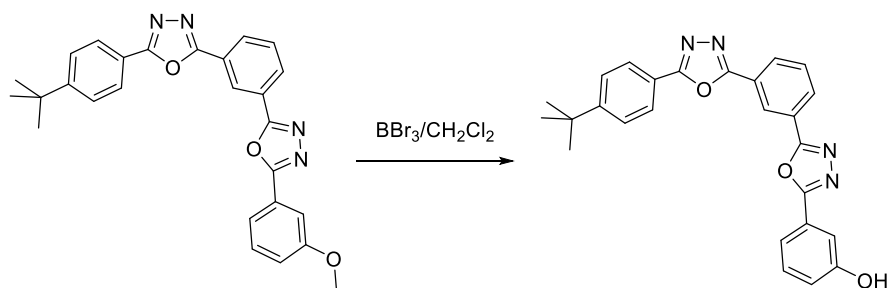
3-(5-(4-tert-Butylphenyl)-1,3,4-oxadiazol-2-yl)benzohydrazide: To a solution of methyl 3-(5-(4-tert-butylphenyl)-1,3,4-oxadiazol-2-yl)benzoate (34.0 g, 101 mmol) in dioxane (300 mL) and ethanol (100 mL) was added hydrazine hydrate (50.0 mL). The reaction mixture was heated to 100 °C for 20 h. The reaction mixture was cooled down to room temperature and water (500 mL) was added. The resulting white solid was collected by filtration and dried under vacuum (33.0 g, 97%). ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.07 (s, br, 1H, NH), 8.55 (t, $J = 1.6$ Hz, 1H), 8.25 (dt, $J_1 = 8.0$ Hz, $J_2 = 1.6$ Hz, 1H), 8.06 (d, $J = 8.8$ Hz, 2H), 7.69 (t, $J = 8.0$ Hz, 1H), 7.65 (d, $J = 8.8$ Hz, 2H), 4.60 (s, br, 2H, NH_2), 1.33 (s, 9H, $^t\text{Bu CH}_3$). $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, $\text{DMSO}-d_6$): δ 165.61, 165.13, 164.35, 156.00, 135.24, 131.13, 130.54, 129.93, 127.53, 127.15, 126.11, 124.55,

121.49, 35.76, 31.73. MS(MALDI): m/z 337 (MH^+), Anal. Calcd for $C_{19}H_{20}N_4O_2$: C, 67.84; H, 5.99; N, 16.66. Found: C, 67.79; H, 5.96; N, 16.56.

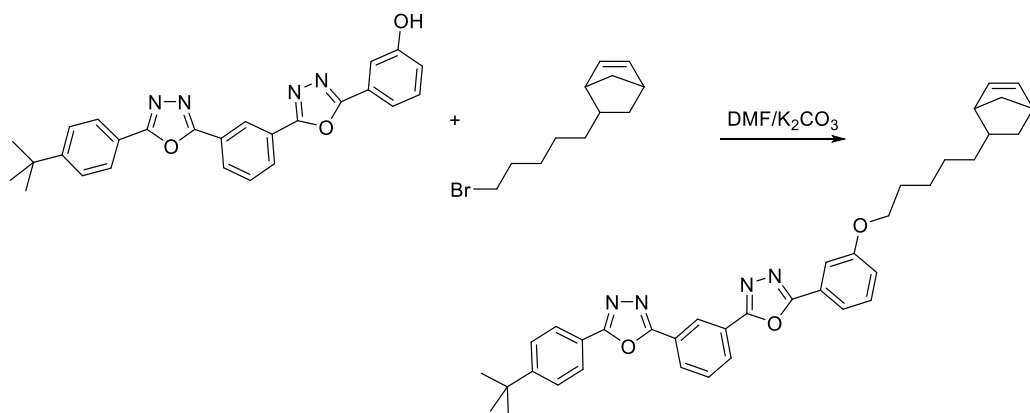


2-(4-*tert*-Butylphenyl)-5-(3-(5-(3-methoxyphenyl)-1,3,4-oxadiazol-2-yl)phenyl)-1,3,4-

oxadiazole: This compound was synthesized as a white powder (13.8 g, 51%) from 2,4-(5-(4-*tert*-butylphenyl)-1,3,4-oxadiazol-2-yl)benzohydrazide (20.0 g, 59.5 mmol) and 3-methoxybenzoyl chloride (11.0 g, 64.4 mmol), using a procedure analogous to that for methyl 3-(5-(4-*tert*-butylphenyl)-1,3,4-oxadiazol-2-yl)benzoate. ¹H NMR (400 MHz, CDCl₃) δ 8.86 (t, J = 1.6 Hz, 1H), 8.34 (dt, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 2H), 8.11 (d, J = 8.4 Hz, 2H), 7.73 (m, 3H), 7.57 (d, J = 8.4 Hz, 2H), 7.47 (t, J = 7.6 Hz, 1H), 7.32 (dd, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 3.93 (s, 3 H, OCH₃), 1.39 (s, 9 H, ^tBu CH₃). ¹³C{¹H} NMR (100 MHz, CDCl₃): δ 165.11, 164.94, 163.62, 163.34, 159.95, 155.64, 130.26, 129.97, 129.74, 126.89, 126.10, 125.10, 124.92, 124.90, 124.65, 120.70, 119.42, 118.42, 111.60, 55.56, 35.10, 31.08. MS(EI): m/z 452 (M^+). Anal. Calcd for $C_{27}H_{24}N_4O_3$: C, 71.67; H, 5.35; N, 12.38. Found: C, 71.37; H, 5.22; N, 12.39.



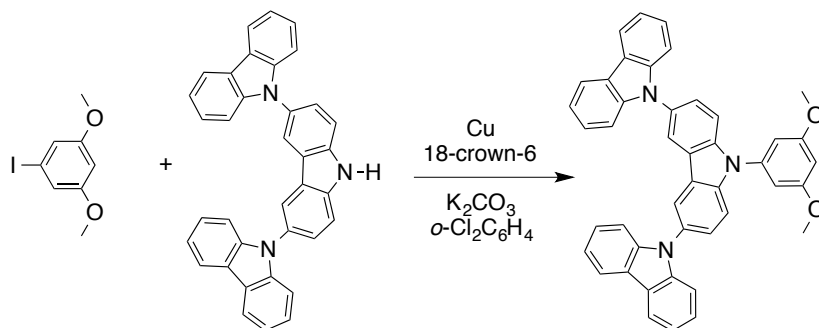
3-(5-(3-(5-(4-*tert*-Butylphenyl)-1,3,4-oxadiazol-2-yl)phenyl)-1,3,4-oxadiazol-2-yl)phenol: To a solution of 2-(4-*tert*-butylphenyl)-5-(3-(5-(3-methoxyphenyl)-1,3,4-oxadiazol-2-yl)phenyl)-1,3,4-oxadiazole (12.0 g, 26.5 mmol) in dichloromethane (320 mL) cooled to at $-78\text{ }^{\circ}\text{C}$ (dry-ice/acetone bath) was dropwise added BBr_3 (100 mL of a 1 M solution in dichloromethane) under nitrogen. The reaction was stirred at room temperature for 19.5 h and then poured into ice-water (600 mL). Dichloromethane was evaporated under reduced pressure and the resulting white solid was collected by filtration and washed with water. The product was purified by recrystallization from acetone and drying under vacuum as a white powder (11.5 g, 99%). $^1\text{H-NMR}$ (400 MHz, DMSO-d_6): δ 10.00 (s, 1 H), 8.68 (s, 1H), 8.31 (m, 2H), 8.07 (d, $J = 8.4$ Hz, 2H), 7.86 (t, $J = 8.0$ Hz, 1H), 7.63 (d, $J = 8.4$ Hz, 2H), 7.58 (d, $J = 7.6$ Hz, 1H), 7.53 (s, 1H), 7.42 (t, $J = 7.6$ Hz, 1H), 7.03 (dd, $J_1 = 7.6$ Hz, $J_2 = 1.6$ Hz, 1H), 1.32 (s, 9 H). $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO-d_6): δ 165.27, 165.25, 163.89, 163.80, 158.88, 156.02, 131.55, 130.46, 127.54, 127.08, 125.36, 125.20, 125.06, 124.88, 121.34, 120.22, 118.42, 114.04, 35.74, 31.71. MS(EI): m/z 438 (M^+). Anal. Calcd for $\text{C}_{26}\text{H}_{22}\text{N}_4\text{O}_3$: C, 71.22; H, 5.06; N, 12.78. Found: C, 71.05; H, 5.08; N, 12.64.



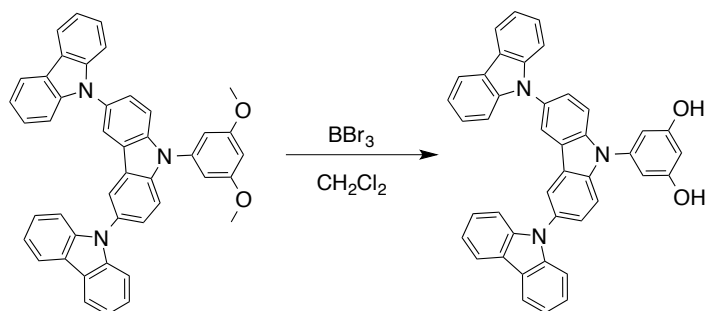
Monomer 2: K_2CO_3 (10.0 g, 72.4 mmol) was added to a stirred solution of 3-(5-(3-(5-(4-*tert*-butylphenyl)-1,3,4-oxadiazol-2-yl)phenyl)-1,3,4-oxadiazol-2-yl)phenol (8.0 g, 18 mmol) and 5-(5-bromopentyl)-norbornene² (5.0 g, 21 mmol) in DMF (100 mL) at room temperature. The reaction was heated to 60 °C for 18 h. After cooling, water (500 mL) was added and the resulting white solid product was collected by filtration. After drying, the crude product was purified by column chromatography (silica gel, 9:1 dichloromethane/ethyl acetate as eluent). After removal of solvents, recrystallization from dichloromethane/methanol, and drying in vacuum, a white solid product was obtained (7.1 g, 65%). ¹H NMR (400 MHz, CDCl₃): δ 8.83 (m, 1H), 8.30 (m, 2H), 8.08 (d, *J* = 8.4 Hz, 2H), 7.70-7.65 (m, 3H), 7.56 (d, *J* = 8.4 Hz, 2H), 7.42 (t, *J* = 8.0 Hz, 1H), 7.06 (m, 1H), 6.08 (dd, *J* = 6.0, 2.8 Hz, C=C-H, 0.85H, endo), 6.06 (dd, *J* = 5.6, 3.2 Hz, C=C-H, 0.15H, exo), 5.98 (dd, *J* = 5.6, 3.2 Hz, C=C-H, 0.15H, exo), 5.89 (dd, *J* = 6.0, 2.8 Hz, C=C-H, 0.85H, endo), 4.03 (t, *J* = 6.8 Hz, 2H, OCH₂), 2.72 (m, br, 1.7H), 2.49 (s, br, 0.3H), 1.96 (m, 1H), 1.81 (m, 2.5 H), 1.46-1.03 (m, 7.5H), 0.47 (m, 1H). ¹³C{¹H} NMR (100 MHz, CDCl₃): δ 165.12, 165.01, 163.60, 163.34, 159.56, 155.65, 136.91, 132.33, 130.21, 129.96, 129.71, 129.66, 126.90, 126.10, 125.13, 124.98, 124.91, 124.61, 120.74, 119.23, 118.79, 112.31, 68.34, 49.52, 45.37, 42.48, 38.65, 35.10, 34.65, 32.38, 31.09, 29.18, 28.38, 26.24 ppm. MS-EI: *m/z*

600.3 (M^+). Anal. Calcd for $C_{38}H_{40}N_4O_3$: C, 75.97; H, 6.71; N, 9.33. Found: C, 75.96; H, 6.76; N, 9.24.

II. Synthesis of Sty_2TCz

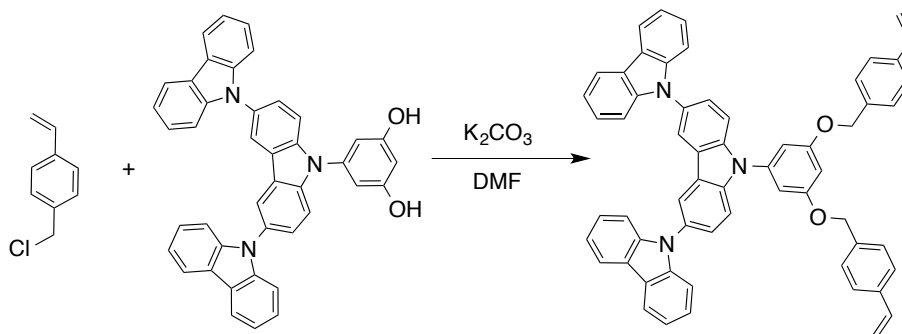


9'-(3,5-Dimethoxyphenyl)-9'H-9,3':6',9''-tercarbazole: 1-Iodo-3,5-dimethoxybenzene (2.42 g, 9.14 mmol),³ triscarbazole¹ (3.51 g, 7.03 mmol), 18-crown-6 ether (0.059 g, 0.21 mmol), copper powder (5.85 g, 92.0 mmol), and *o*-dichlorobenzene (75 mL) were combined in a flask under a nitrogen atmosphere. Potassium carbonate (11.53 g, 83.4 mmol) was added and the reaction was heated to 180 °C for 24 h. After cooling, solids were removed by filtration and washed, and the solvents were removed from the filtrate under reduced pressure. The crude material was purified by column chromatography (silica gel; 4:1 hexanes/ethyl acetate) to afford an off-white powder after recrystallization from hot dichloromethane/methanol (3.20 g, 72%). 1H NMR (300 MHz, $CDCl_3$): δ 8.27-8.26 (m, 2H), 8.16 (d, $J = 7.7$ Hz, 4H), 7.75-7.70 (m, 2H), 7.64-7.59 (m, 2H), 7.42-7.37 (m, 8H), 7.31-7.26 (m, 4H), 6.88 (d, $J = 2.2$ Hz, 2H), 6.66 (t, $J = 2.2$ Hz, 1H), 3.92 (s, 6H). $^{13}C\{^1H\}$ (75 MHz, $CDCl_3$): δ 162.1, 141.8, 140.6, 138.8, 130.5, 126.3, 125.9, 124.0, 123.2, 120.2, 119.7, 111.5, 109.7, 105.5, 100.3, 55.7. IE-MS: m/z 633.2 (M^+).



5-(9'H-[9,3':6',9''-Tercarbazol]-9'-yl)benzene-1,3-diol: 9'-(3,5-Dimethoxyphenyl)-9'H-

9,3':6',9''-tercarbazole (4.01 g, 6.33 mmol) was dissolved in anhydrous dichloromethane (40 mL) under a nitrogen atmosphere and cooled to $-78\text{ }^{\circ}\text{C}$ (dry-ice/acetone bath). Boron tribromide (50 mL of a 1.0 M solution in dichloromethane, 50 mmol) was added dropwise to the cooled solution. The cooling bath was then removed and after ca. 4 h, the mixture was added into ice-water (150 mL) with stirring. The biphasic mixture was separated and the aqueous phase was extracted with dichloromethane ($2 \times 100\text{ mL}$). The combined organic phase was dried over magnesium sulfate, filtered, and evaporated under reduced pressure. The crude product was purified by column chromatography (silica gel, toluene) and recrystallization from acetone/methanol to afford a grayish white powder (1.55 g, 41%). ^1H (300 MHz, $\text{DMSO-}d_6$): δ 9.91 (s, 2H), 8.66 (d, $J = 1.9\text{ Hz}$, 2H), 8.27-8.21 (m, 4H), 7.82-7.75 (m, 2H), 7.72-7.66 (m, 2H), 7.46-7.36 (m, 8H), 7.30-7.22 (m, 4H), 6.66 (d, $J = 2.1\text{ Hz}$, 2H), 6.51 (t, $J = 2.1\text{ Hz}$, 1H). $^{13}\text{C}\{^1\text{H}\}$ (75 MHz, $\text{DMSO-}d_6$): δ 160.3, 141.5, 140.2, 138.3, 129.9, 126.6, 126.4, 124.0, 122.9, 120.9, 120.7, 120.2, 112.1, 110.2, 105.1, 102.8. EI-MS m/z 605.2 (M^+).



Sty2-TCz: 5-(9'*H*-[9,3':6',9''-Tercarbazol]-9'-yl)benzene-1,3-diol (1.00 g, 1.65 mmol), DMF (25 mL), and potassium carbonate (2.78 g, 20.1 mmol) combined in a flask. 1-(chloromethyl)-4-vinylbenzene (0.607 g, 3.98 mmol) added dropwise to mixture. After 24h, deionized water (100 mL) added to flask and white precipitate formed. The crude material was isolated by filtration and purified by column chromatography (silica gel; 4:1 hexanes/ethyl acetate). The resulting oil was precipitated into methanol, collected by filtration, and dried to afford a white powder (0.559 g, 41%). ^1H (300 MHz, $\text{DMSO-}d_6$): δ 8.64 (d, $J = 1.5$ Hz, 2H), 8.23 (d, $J = 7.7$ Hz, 4H), 7.58-7.31 (m, 20H), 7.26 (t, $J = 7.4$ Hz, 4H), 6.99 (d, $J = 2.2$ Hz, 2H), 6.91 (t, $J = 2.0$ Hz, 1H), 6.71 (dd, $J = 17.7, 11.0$ Hz, 2H), 5.83-5.77 (d, $J = 17.6$ Hz, 2H), 5.25 (s, 4H), 5.18 (d, $J = 10.9$ Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ (75 MHz, $\text{DMSO-}d_6$): δ 162.1, 160.8, 141.4, 140.0, 137.2, 136.9, 136.7, 130.0, 128.4, 128.2, 126.8, 126.6, 124.1, 122.9, 120.9, 120.2, 115.0, 110.1, 105.9, 69.8. EI-MS m/z 837.2 (M^+). Anal. Calcd. for $\text{C}_{60}\text{H}_{43}\text{N}_3\text{O}_2$: C, 86.00; H, 5.17; N, 5.01. Found: C, 85.76; H, 5.14; N, 5.08.

III. Solid-State NMR Spectra for ^1H Spin-Diffusion Experiments

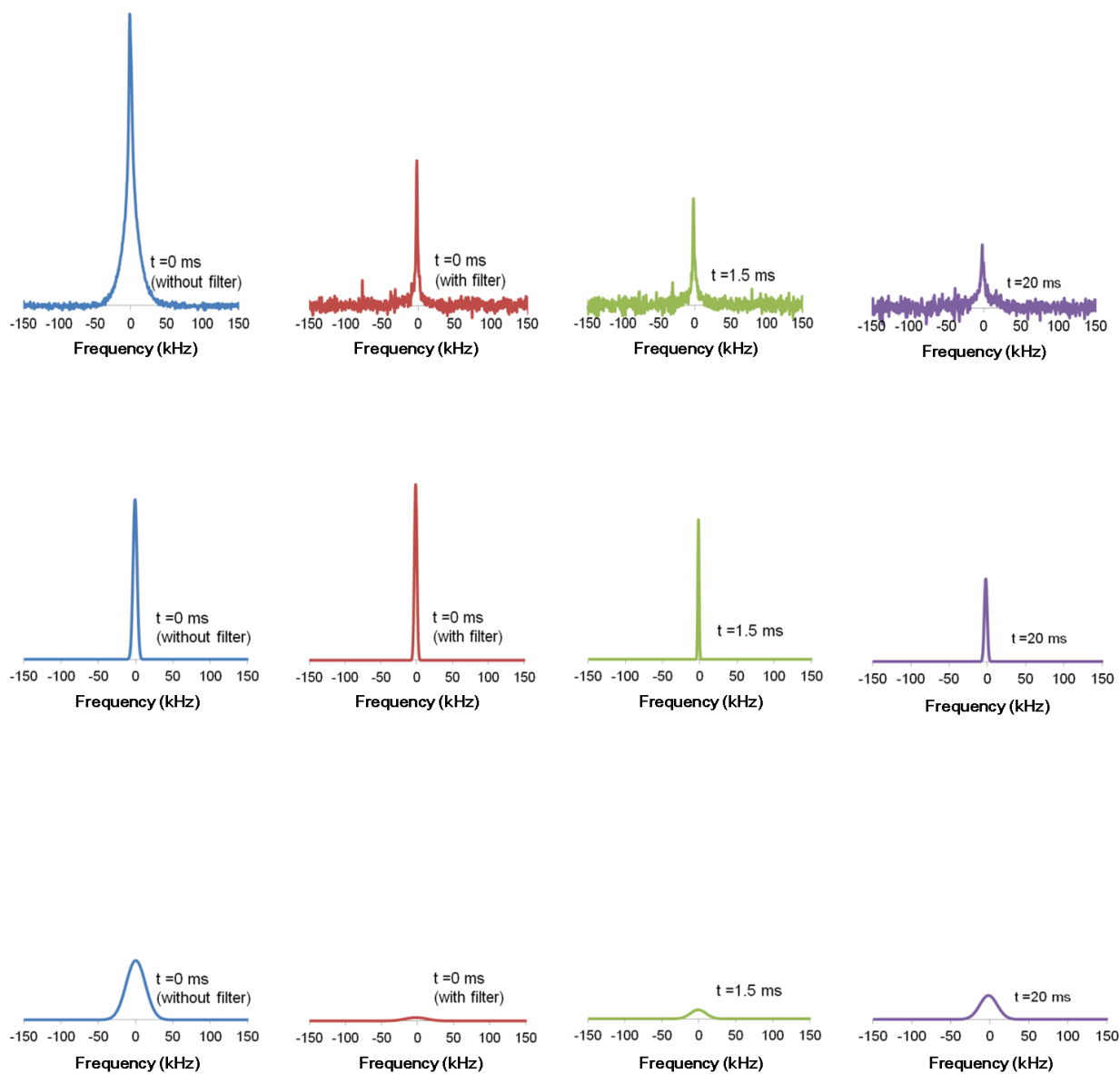


Figure S1. Representative raw and fitted ^1H spectra at different spin-diffusion times for the 1:1 poly-1/poly-2 physical blend. The first row, second, and third rows show the raw spectra, fitted curves for the mobile phase, and fitted curves for the rigid phase respectively.

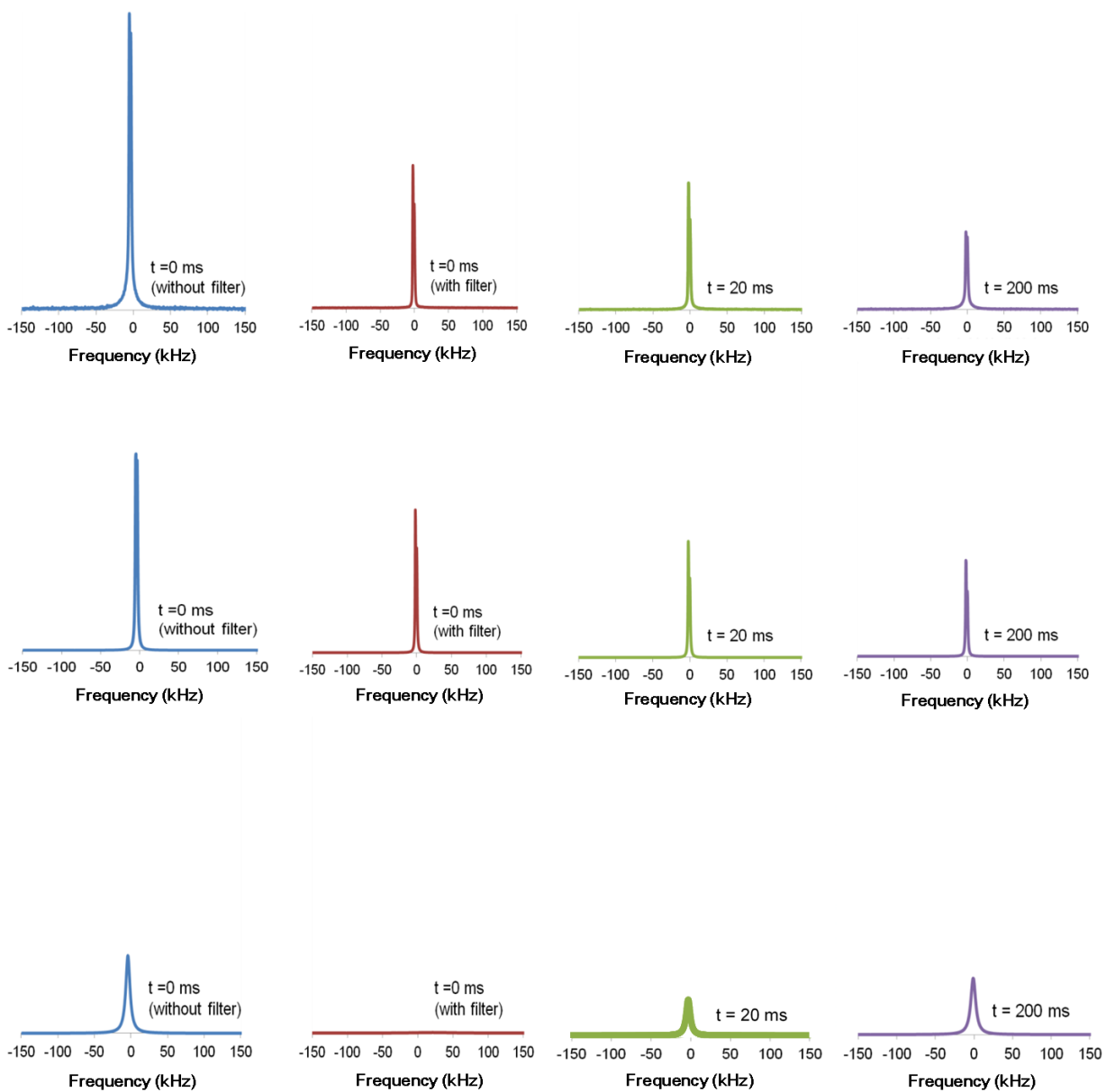


Figure S2. Representative raw and fitted ^1H spectra at different spin-diffusion times for the 1:1 poly-1/poly-2 physical blend. The first row, second, and third rows show the raw spectra, fitted curves for the mobile phase, and fitted curves for the rigid phase respectively.

IV. Spin-Spin Relaxation and ^1H Spin Diffusion Coefficients

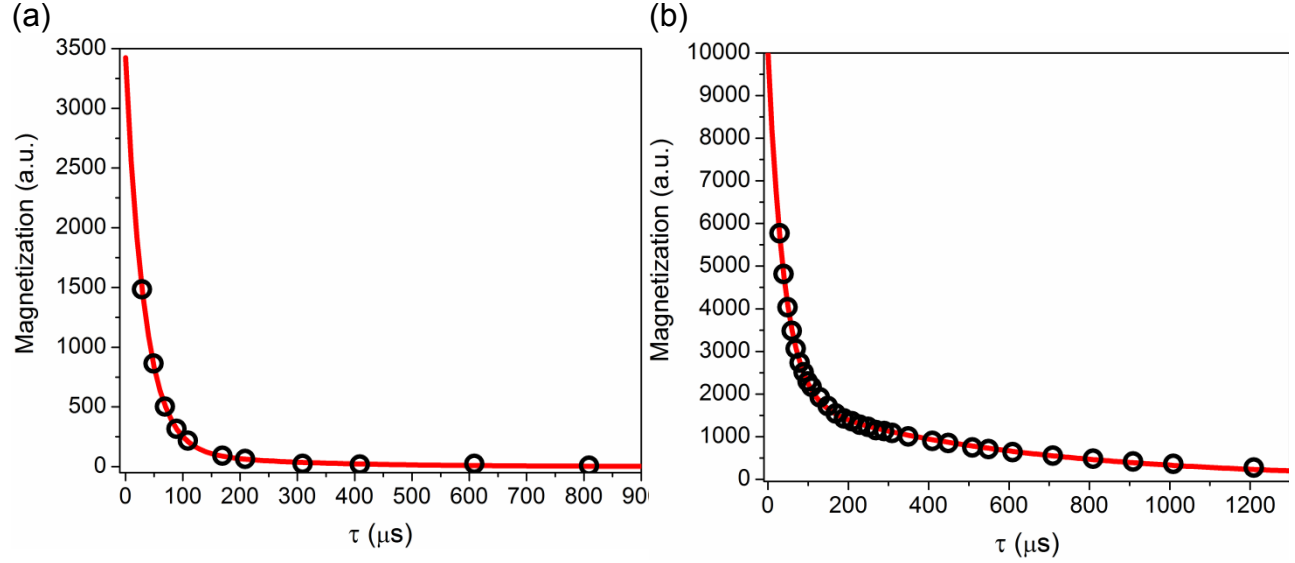


Figure S3. Spin-spin relaxation experimental results (black circle) and fitted curves (red curve) for (a) poly-1-b-2 and (b) the poly-1/poly-2 physical blend. The molar ratio between the oxadiazole and tricarbazole units in both samples is ca. 1:1.

The fitting for the spin-spin relaxation curves follows the two-exponential-decay function:

$$M = M_{rigid} \cdot \exp(-\tau / T_{2,rigid}) + M_{mobile} \cdot \exp(-\tau / T_{2,mobile}) \quad (\text{S1})$$

where M is the observed magnetization, τ is the echo time, M_{rigid} and M_{mobile} are the pre-exponential factor for the rigid and mobile phase respectively, and $T_{2,rigid}$ and $T_{2,mobile}$ are the characteristic time for the spin-spin relaxation for the rigid and mobile phase respectively. As discussed in the context, the rigid phase is assigned to the Tcz domain and the mobile phase is assigned to the Oxz domain. The spin diffusion coefficients were then estimated according the following empirical correlation:⁴

$$D = 4.5 \times 10^{-5} T_2^{-1} + 0.26 \quad (\text{S2})$$

where D is the diffusion coefficient in a unit of nm^2/ms and T_2 is in a unit of second. The deduced T_2 values and the diffusion coefficients are summarized in Table S1.

Table S1. Parameters Deduced from the Spin-Spin Relaxation

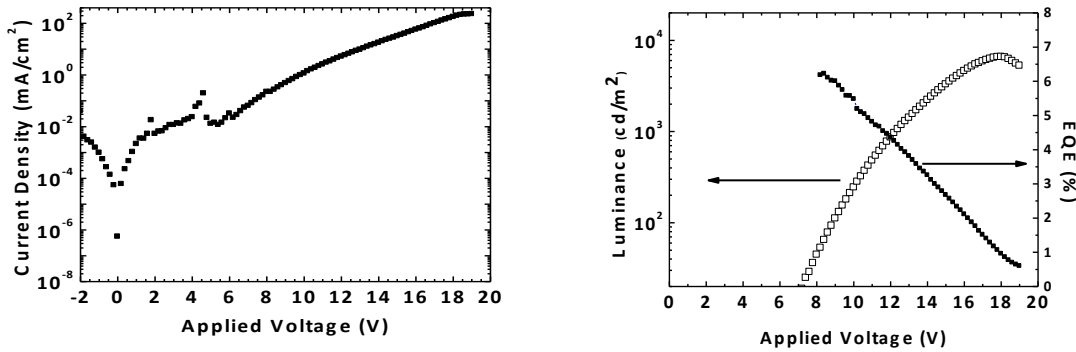
	M_{rigid} (a.u.)	M_{mobile} (a.u.)	$T_{2,rigid}$ (μ s)	$T_{2,mobile}$ (μ s)	D_{rigid} (nm^2/ms)	D_{mobile} (nm^2/ms)
Block Copolymer	3268	155	32.8	209.6	1.63	0.47
Physical Blend	8149	1902	39.5	572.7	1.40	0.34

V. Summary of Optical Data for Polymeric Hosts

Table S2. Absorption and emission maxima of the polymers in dichloromethane at room temperature.

Polymer	Absorption		Emission	
	λ_{max} (nm)	E_{max} (eV)	λ_{max} (nm)	E_{max} (eV)
poly-1	227, 289	5.46, 4.29	366	3.39
poly-2	239, 294, 342	5.19, 4.22, 3.63	391, 408	3.17, 3.04
poly-1-co-2	238, 294, 343	5.21, 4.22, 3.62	390, 407, 475	3.18, 3.05, 2.61
poly-1-b-2	238, 294, 342	5.21, 4.22, 3.63	366, 389	3.39, 3.19
1:1 poly-1/poly-2	238, 294, 342	5.21, 4.22, 3.63	366, 387	3.39, 3.20

VI. Performance Details of the Green-Emitting Devices

**Figure S4.** Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using a poly-1-co-2 host with Ir(ppy)₃ (6 wt%).

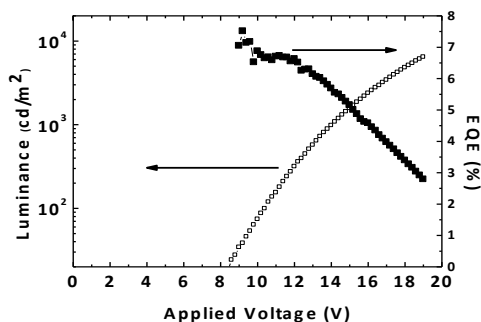
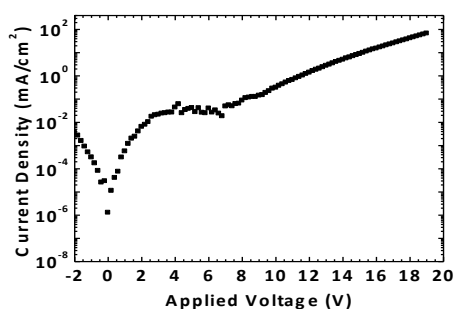


Figure S5. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using poly-1-co-2 host with Ir(ppy)₃ (12 wt%).

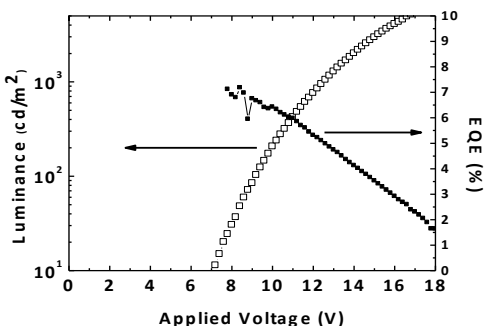
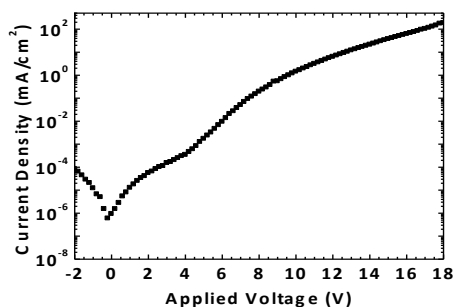


Figure S6. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using poly-1-b-2 host with Ir(ppy)₃ (6 wt%).

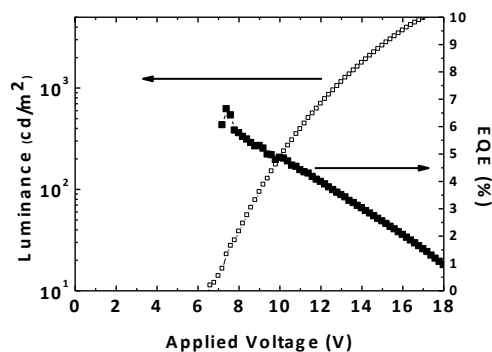
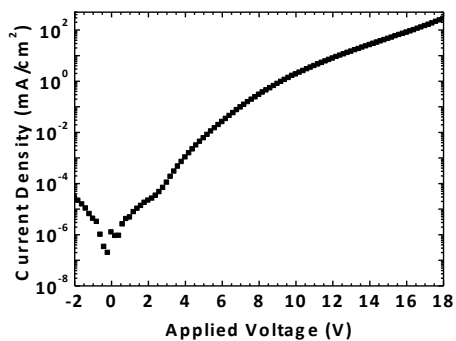


Figure S7. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using poly-1-b-2 host with Ir(ppy)₃ (12 wt%).

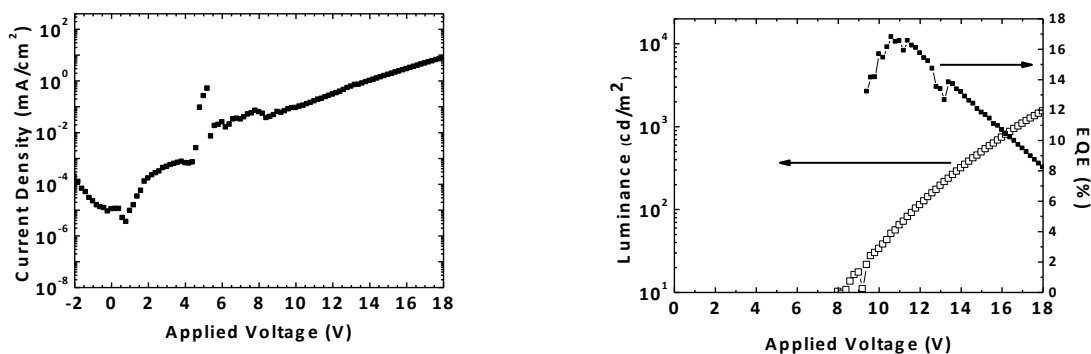


Figure S8. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using 1:1 poly-1/poly-2 host with Ir(ppy)₃ (6 wt%).

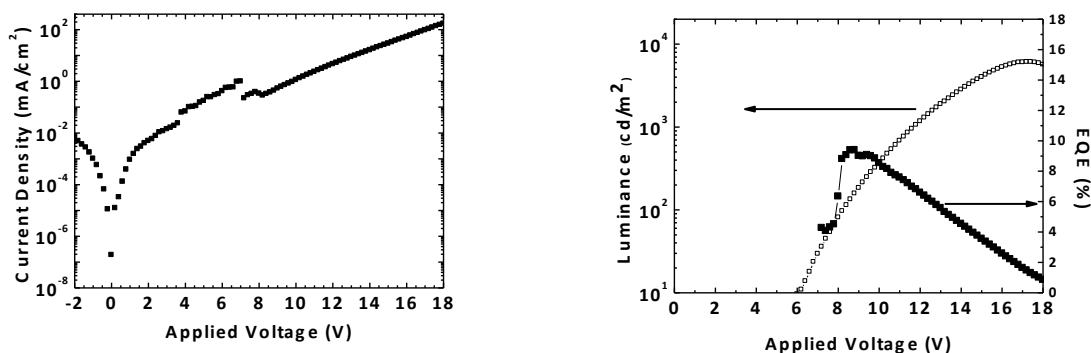


Figure S9. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using 1:1 poly-1/poly-2 host with Ir(ppy)₃ (12 wt%).

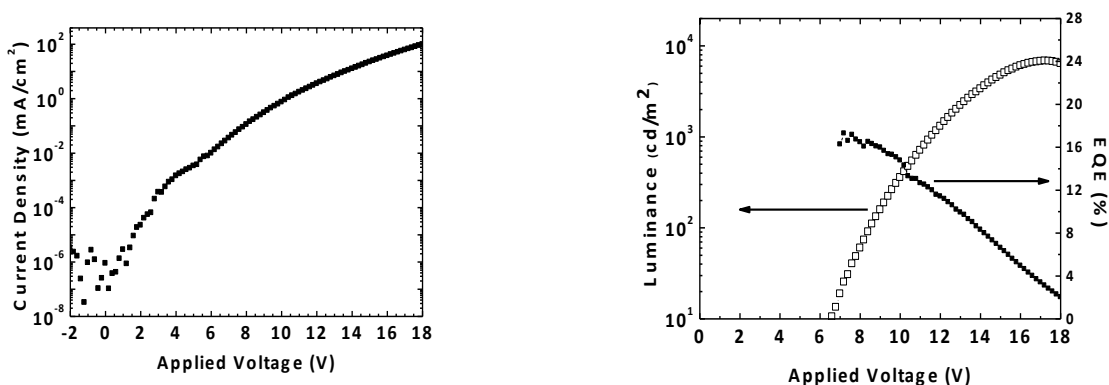


Figure S10. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using 1:1.5 poly-1/poly-2 host with Ir(ppy)₃ (6 wt%).

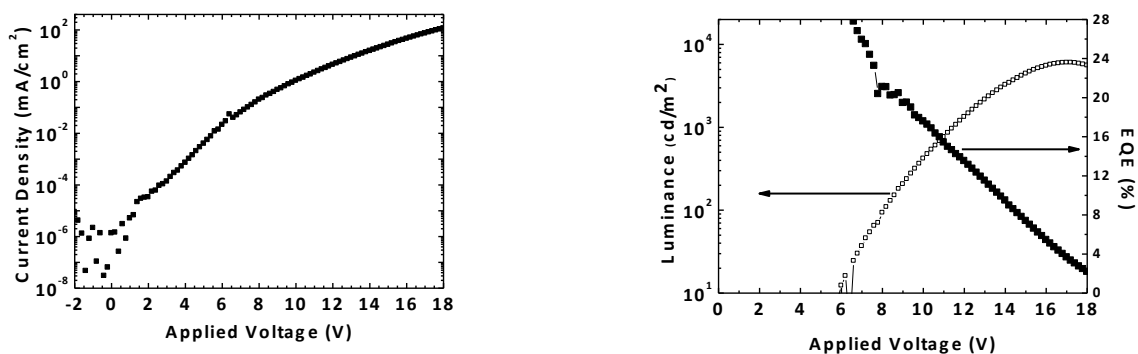


Figure S11. Plots showing current density–voltage characteristics, luminance vs. applied voltage, and external quantum efficiency vs. the applied voltage for an OLED using 1.5:1 poly-1/poly-2 host with Ir(ppy)₃ (6 wt%).

VII. References

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