Supporting Information

Dark Current Reduction Strategy *via* a Layer-by-Layer Solution Process for a High-Performance All-Polymer Photodetector

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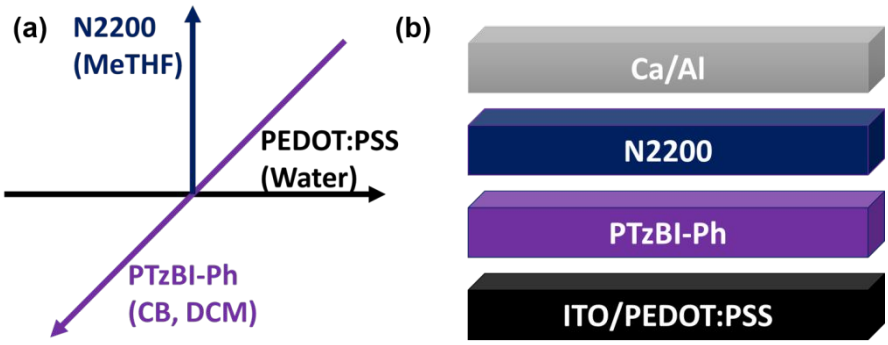

Figure S1. (a) The semi-orthogonal relationship of PEDOT:PSS, PTzBI-Ph and N2200. (b) The processing sequence of gradient bulk heterojunction device.

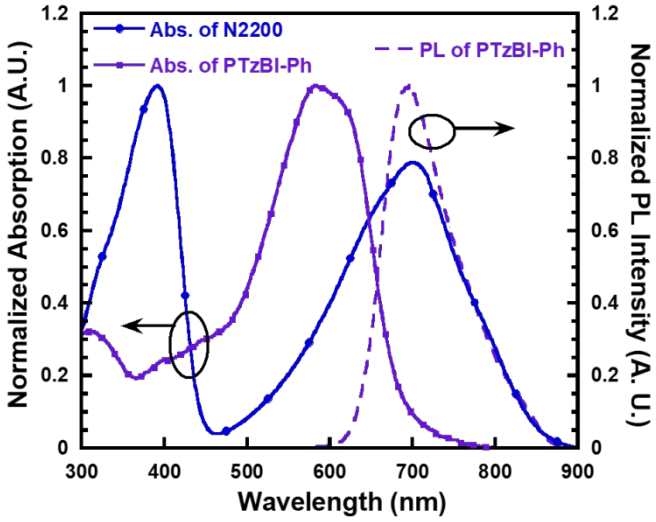

Figure S2. The normalized absorption and PL spectra of PTzBI-Ph, and the normalized absorption spectrum of N2200 in films.

Table S1. Performance of LBL and blend-processed OPDs with various thickness configurations.

Configuration[a]	Average[b] (Best) Values @-0.1 V			
	J_d /nA cm^{-2}	EQE_{max} /%	R_{max} /A W^{-1}	D^*_{max}[c] /×10^{12} Jones
PTzBI-Ph (60 nm)	7.37 ± 2.17	52.20 ± 3.58	0.23 ± 0.02	5.00 ± 1.23
/N2200 (60 nm)	(3.74)	(55.50)	(0.25)	(7.22)
PTzBI-Ph (80 nm)	5.68 ± 2.36	56.43 ± 2.06	0.25 ± 0.01	6.37 ± 1.58
/N2200 (40 nm)	(2.35)	(54.48)	(0.25)	(0.90)
PTzBI-Ph (80 nm)	3.77 ± 2.87	55.31 ± 2.34	0.25 ± 0.01	9.07 ± 4.05
/N2200 (60 nm)	(1.25)	(53.15)	(0.25)	(12.54)
PTzBI-Ph (80 nm)	3.22 ± 1.37	44.92 ± 6.30	0.21 ± 0.01	6.99 ± 1.73
/N2200 (80 nm)	(1.24)	(42.04)	(0.20)	(9.82)
PTzBI-Ph (100 nm)	3.51 ± 2.02	33.52 ± 6.00	0.15 ± 0.03	5.08 ± 1.34
/N2200 (60 nm)	(1.30)	(26.14)	(0.13)	(6.21)
PTzBI-Ph:N2200	75.97 ± 45.96	8.89 ± 0.12	0.04 ± 0.01	0.32 ± 0.10
(2:1wt, 80 nm)	(29.68)	(9.02)	(0.044)	(0.45)
PTzBI-Ph:N2200	10.46 ± 8.14	14.06 ± 0.59	0.06 ± 0.01	1.31 ± 0.42
(2:1wt, 100 nm)	(5.08)	(14.72)	(0.067)	(1.65)
PTzBI-Ph:N2200	6.74 ± 4.56	7.11 ± 0.74	0.04 ± 0.01	0.89 ± 0.34
(2:1wt, 120 nm)	(3.13)	(7.31)	(0.037)	(1.17)

[a] The thickness of N2200 was estimated from difference value between total thickness and thickness of PTzBI-Ph.

[b] Each value is the average of 8 devices.

[c] Calculated from equation (1), 1 Jones = 1 cm Hz$^{1/2}$ W^{-1}.

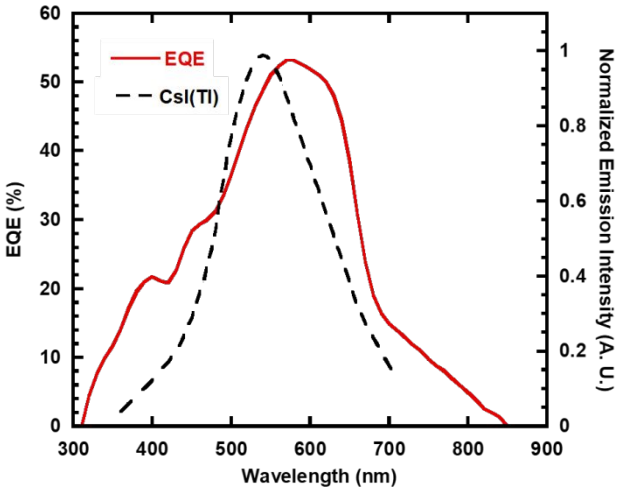

Figure S3. The EQE of all polymer photodetector with configuration of ITO/PEDOT:PSS (10 nm)/PTzBI-Ph (80 nm)/N2200 (60 nm)/Ca (5 nm)/Al (200 nm) and CsI (Tl)-based scintillator's emission spectrum.

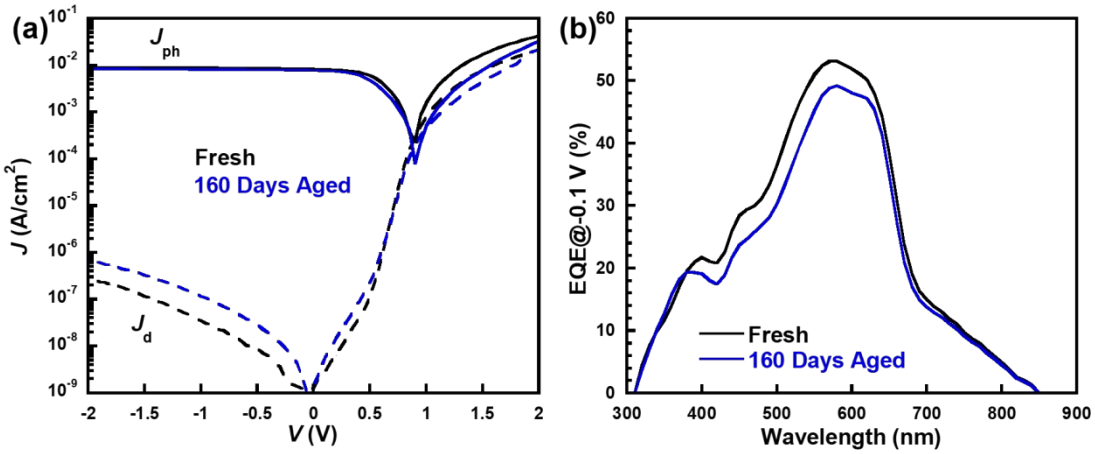

Figure S4. (a) The *J*-*V* and (b) EQE characteristics of champion device before and after 160 days ageing.

Table S2. Performance of LBL-processed OPDs before and after 160 days stored in ambient and at room temperature.

PTzBI-Ph (80 nm) /N2200 (60 nm)[a]	Average[b] (Best) Data @-0.1 V			
	J_d /nA cm^{-2}	EQE_{max} /%	R_{max} /A W^{-1}	D^*_{max}[c] /×10^{12} Jones
Fresh	3.77 ± 2.87 (1.25)	55.31 ± 2.34 (53.15)	0.25 ± 0.01 (0.25)	9.07 ± 4.05 (12.54)
After 160 days	7.25 ± 5.60 (2.25)	46.63 ± 3.73 (49.17)	0.22 ± 0.02 (0.24)	5.56 ± 2.50 (8.79)

[a] The thickness of N2200 was estimated from difference value between total thickness and thickness of PTzBI-Ph.

[b] Each value is the average of 8 devices.

[c] Calculated from equation (1), 1 Jones = 1 cm Hz$^{1/2}$ W^{-1}.