Supporting Information

Enhancing the Performance of Organic Solar Cells by Using PDINN-doped PEDOT:PSS as the Hole Transport Layer

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Experimental Section

Materials and Instruments

The ITO glass was purchased from the South China Science & Technology Company. The PM6 and L8-BO were purchased from Derthon. The PDINN and PNDIT-F3N was purchased from Organtec Ltd. All these solvents used here were commercially available from Sigma-Aldrich. The PEDOT:PSS (Clevios P VP AI 4083) was purchased from Heraeus. The Y6 and D18 were synthesized by our own laboratory according to the procedure reported in literature.[1,2] All materials were used as received without further purification.

UV-visible absorption spectra were obtained on a PerkinElmer UV-vis spectrometer (model Lambda 750). Atomic force microscopy (AFM) measurements were performed under ambient conditions using a Digital Instrument Multimode Nanoscope IIIA operating in the tapping mode. The thickness of the blend films and transport layer were determined by a Dektak 6 M surface profile meter. Ultraviolet photoelectron spectroscopy (UPS) experiments were carried out on a Thermo Scientific ESCALab 250Xi spectrometer. The gas discharge lamp was used for UPS with helium gas admitted and the He I (21.22 eV) emission line employed. The helium pressure in the analysis chamber during analysis was about 2e-8 mbar. The data were acquired with -10 V bias. Contact angle characterization was performed on DSA100, Kruss. Raman spectra was collected with a confocal Raman spectrometer (HORIBA, LABRAM HR-800) using a 532 nm laser as the excitation source. The X-ray photoelectron spectroscopy (XPS) was evaluated via employment of the Thermo Scientific* ESCALAB 250Xi instrument.

Photoluminescence quenching spectroscopy was conducted using PG2000-Pro (400-1000 nm) and NIR1700 (900-1700 nm) Scientific Grade Spectrometer (idea optics, China). Impedance spectrogram analysis uses a cold light source of halogens LG-150 model instrument to simulate a light source.

Preparation of PEDOT:PSS with PDINN

PEDOT:PSS-PDINN solution was prepared by adding 1 wt% PDINN aqueous solution with 0.2% acetic acid into PEDOT:PSS solution.

OSC Device Fabrication and Characterization

The conventional devices were fabricated with the structure of ITO/PEDOT:PSS, PEDOT:PSS-PDINN/PM6:Y6/PNDIT-F3N/Ag. Substrates are quartz or indium tin oxide (ITO) coated glass, depending on the type of measurement. All substrates were cleaned in ultrasonic bath with washing solution, deionized water, isopropyl alcohol, and deionized water, consecutively. The substrates were dried on a hotplate at 160 °C for 10 min, and were then treated with UV-ozone for 25 min. PEDOT:PSS solution with/without PDINN filtered by polyether sulfone filter (0.45 um) was spin-coated on these cleaned ITO substrates at the speed of 3500 rpm for 20 s to obtain a film thickness of 30 nm, followed by annealing at 150 ℃ for 15 min in air before being transferred into a glovebox. The blend solutions of PM6:Y6 (D/A weight ratio, 1:1.2) mixtures were all processed with chloroform (CF) with a polymer concentration of 6.5 mg/mL. Chloronaphthalene (CN) was added to the active layer solution at 0.5 vol% of the solution, and then spin-coated at 1700 rpm for 30 s to obtain a film thickness of 100 nm on the top of the HTL, and then annealing 100 ℃ for 5 min. Methanol solution of PNDIT-F3N (1 mg/mL) was then deposited atop the active layer at 3000 rpm for 30 s. The Ag layer (100 nm) electrodes were sequentially evaporated onto the electron transport layer, at a vacuum pressure below 4.0×10^{-4} pa. The blend solution of D18:L8-BO (D/A weight ratio, 1:1) mixtures were all processed with chloroform (CF) with a polymer concentration of 4 mg/mL, and then spin-coated at 2000 rpm for 30 s to obtain a film thickness of 100 nm on the top of the HTL. Methanol solution of PNDIT-F3N (1 mg/mL) was then deposited atop the active layer at 3000 rpm for 30 s. The Ag layer (100 nm) electrodes were sequentially evaporated onto the electron transport layer, at a vacuum pressure below 4.0×10^{-4} pa. Both of the two active layers solutions were heated at 40 ℃ for 2 hours to obtain well-mixed blend solutions. The effective device area, defined by the overlap region of ITO and Ag electrodes, was 0.04 cm^2. The *J-V* curves of photocurrents were recorded in glove box at approximately 25 ℃ using an instrument from Enli Technology Ltd., Taiwan (SS-F53A) under AM 1.5G illumination (AAA class solar simulator, with an intensity of 100 mW cm^{-2} calibrated with a standard single crystal Si photovoltaic cell). External quantum efficiency (EQE) measurements were conducted in air without encapsulation. The EQE data were obtained using a solar cell spectral response measurement system

(QER3011, Enli Tech-nology Co.Ltd), and the intensity was calibrated with a standard single-crystal Si photovoltaic cell.

Space-Charge-Limited-Current Measurement

The hole and electron-only devices with structures of ITO/HTL/active layer/MoO_3/Ag and ITO/ZnO/active layer/PDINN/Ag were fabricated, respectively. Dark *J-V* curves of the hole/electron devices were measured by the space-charge limited current (SCLC) method. Dark *J-V* curves were fitted by using the Mott-Gurney equation: $J = 9\varepsilon_o\varepsilon_r\mu V/8d^3$, where J is the space charge limited current, ε_0 is the vacuum permittivity ($\varepsilon_o = 8.85 \times 10^{-12}$ F/m), ε_r is the permittivity of the active layer ($\varepsilon_r \approx 3$), μ is charge mobility, and d is the thickness of the active layer.

Table S1. Photovoltaic properties of multi-component OPV cells in incorporation with PM6:Y6-based active layer

Active layer	D/A ratio	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)	Ref.
PM6:Y6	1:1.2	0.83	25.3	74.8	15.7	[1]
PM6:J71:Y6	0.9:0.1:1.2	0.85	25.55	76.0	16.5	[4]
PM6:PDHP-Th:Y6	1:0.1:1.2	0.85	26.6	71.7	16.2	[5]
PM6:PDHP-Ph:Y6	1:0.03:1.2	0.823	26.58	68.2	15.4	[5]
PM6:TPD-3F:Y6	0.9:0.1:1.2	0.893	25.83	73.7	17.0	[6]
PM6:PBDB-T-SF:Y6	0.85:0.15:1.2	0.86	25.21	76.11	16.4	[7]
PM6:Phl-Th:Y6	1:0.15:1.2	0.85	24.8	72.1	15.2	[8]
PM6:Phl-Se:Y6	1:0.15:1.2	0.85	25.7	75.5	16.4	[8]
PM6:Phl-Se:Y6:$PC_{71}BM$	1:0.15:1.2:0.2	0.85	26.3	76.8	17.2	[8]
PM6:S3:Y6	0.8:0.2:1.2	0.86	25.86	79.17	17.53	[9]
PM6:SM1:Y6	0.85:0.15:1.2	0.83	25.7	77.5	16.55	[10]
PM6:BTR:Y6	0.95:0.05:1.2	0.84	25.8	76.7	16.6	[11]
PM6:BPR-SCl:Y6	0.75:0.25:1	0.87	25.77	75	16.7	[12]
PM6:ECTBD:Y6	0.85:0.15:1.2	0.85	25.54	76.24	16.52	[13]
PM6:DRTB-T-C4:Y6	0.9:0.1:1.2	0.85	24.74	81.3	17.13	[14]
PM6:P1:Y6	0.6:0.4:1.2	0.87	25.45	73.23	16.2	[15]
PM6:Y6:$PC_{61}BM$	1:1.2:0.2	0.85	25.40	77	16.5	[16]
PM6:Y6: $PC_{71}BM$	1:1.2:0.1	0.861	25.10	77.20	16.70	[17]
PM6:Y6:$PC_{71}BM$	1:1.0:0.2	0.85	25.70	76.35	16.67	[18]

PM6:Y6:$PC_{71}BM_{(flexible)}$	1:1.0:0.2	0.828	23.57	72.03	14.06	[18]
PM6:Y6:$PC_{71}BM$	1:1.0:0.2	0.85	27.5	73.1	17.1	[19]
PM6:Y6:$PC_{71}BM$	1:0.96:0.24	0.83	26.0	75.9	16.3	[20]
PM6:Y6:IDIC	1:1.0:0.2	0.87	25.39	74.92	16.51	[21]
PM6:Y6:MF1	1:1.08:0.12	0.853	25.68	78.61	17.2	[22]
PM6:Y6:IN-4F	1:1.2:0.1	0.85	25.70	74.5	16.3	[23]
PM6:Y6:IT-4F	1:1.2:0.2	0.844	25.4	75.9	16.27	[24]
PM6:Y6:ITCPTC	1:1.14:0.06	0.86	25.67	78.8	17.42	[25]
PM6:Y6:N7IT	1:1.14:0.06	0.85	25.53	77.7	16.96	[25]
PM6:Y6:PF2-DTC	1:1.14:0.06	0.85	25.69	77.5	17.04	[25]
PM6:Y6:3TP3T-4F	1:1.02:0.18	0.85	25.9	74.9	16.7	[26]
PM6:Y6:3TP3T-IC	1:1.02:0.18	0.86	25.2	71.6	15.6	[26]
PM6:Y6:O-IDTBR	1:0.85:0.15	0.85	25.75	76	16.6	[27]
PM6:Y6:C8-DTC	1:1.08:0.12	0.873	26.50	75.61	17.52	[28]
PM6:Y6:Y-Th2	1:1.05:0.15	0.851	25.33	71.44	16.01	[29]
PM6:Y6:FBTIC	1:1.0:0.2	0.866	24.6	77.9	16.7	[30]
PM6:Y6:BTP-M	1:1.2:0.2	0.88	26.56	73.46	17.03	[31]
PM6:Y6:BTP-S2	1:0.96:0.24	0.88	26.20	75.80	17.4	[32]
PM6:Y6:SY3	1:1:0.2	0.855	25.51	78.2	17.1	[33]
PM6:Y6:$N2200_{(spin)}$	1:1.2:0.12	0.83	26.3	76	16.6	[34]
PM6:Y6:$N2200_{(blade)}$	1:1.2:0.12	0.83	26.3	74	16.0	[34]
PM6:Y6:BTF	1:1.2:0.1	0.853	26.11	74.22	16.5	[35]
PM6:Y6:DFBT-TT6	1:1.2:0.066	0.845	26.56	76	17.1	[36]
PM6:Y6:DIBC	1:1.2:0.1	0.83	25.61	77.12	16.4	[37]
PM6:Y6:APDC-TPDA	1:1.2:0.1	0.84	25.98	77.5	17.0	[38]
PM6:Y6:IDMIC-4F	1:1.08:0.12	0.864	25.60	74.20	16.60	[39]
PM6:Y6:PIDTC-T	1:1.2:0.02	0.847	25.50	77.60	16.76	[40]
PM6:Y6:IDIC:$PC_{71}BM$	1:1:0.2:0.1	0.87	26.19	75.29	17.07	[41]
PM6:Y6:Br-ITIC	1:1.08:0.12	0.854	25.5	75.1	16.4	[42]

PM6:Y6:Br-ITIC:$PC_{71}BM$	1:1.08:0.12:0.12	0.85	25.8	76.4	16.8	[42]
PM6:PM7:Y6:$PC_{71}BM$	0.8:0.2:1.2:0.25	0.859	26.55	79.23	18.1	[43]

Table S2. Photovoltaic parameters of PM6:Y6-based OSCs subjected to different treatment processing

Active layer	Processing conditions	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)	Ref.
PM6:Y6	CB 80 ℃	0.796	21.12	72.15	12.2	[44]
PM6:Y6	CF 80 ℃	0.835	26.52	76.21	16.88	[44]
PM6:Y6	CB solution/substrate temperature (80/80 ℃)	0.82	25.9	70.0	15.2	[45]
PM6:Y6	o-XY solution/substrate temperature (100/100 ℃)	0.81	26.6	70.3	15.6	[45]
PM6:Y6	TMB solution/substrate temperature (110/110 ℃)	0.80	26.4	70.9	15.4	[45]
PM6:Y6	Solvent-flushing (0.5% Acac)	0.857	25.50	75.57	16.5	[46]
PM6:Y6:$PC_{71}BM$	Single-annealed 100 ℃	0.84	25.40	75	16.0	[47]
PM6:Y6:$PC_{71}BM$	Single-annealed 120 ℃	0.83	25.90	74	15.9	[47]
PM6:Y6:$PC_{71}BM$	Double-annealed 100/120 ℃	0.83	26.60	76	16.8	[47]
PM6:Y6	0.01 wt% BCF	0.84	25.96	73.47	16.0	[48]
PM6:Y6:$PC_{71}BM$	0.004 wt% BV	0.84	26.3	77	17.1	[49]
PM6:Y6	1 wt% GCl	0.840	26.09	79.05	17.3	[50]
PM6:Y6	1.5% CN 400 nm	0.81	26.3	68.1	14.4	[51]
PM6:Y6	A3	0.820	26.50	76.05	16.50	[52]
PM6:Y6	0.5% PN	0.820	25.90	71.80	15.40	[53]
PM6:Y6	PHJ	0.830	26.52	70.88	15.41	[54]
PM6:Y6	SA-4	0.804	25.7	73.4	15.2	[55]
PM6:Y6	INB-1F	0.81	27.0	72.0	15.7	[56]
PM6:Y6	INB-3F	0.81	27.1	72.2	15.8	[56]

PM6:Y6	INB-5F	0.81	27.7	74.3	16.4	[56]
PM6:Y6	Ferrocene	0.838	26.71	76.00	17.40	[57]
PM6:Y6	MeOH(vapor atmosphere)	0.86	26.43	77.39	17.59	[58]
PM6:Y6	BBS	0.858	27.1	75.5	17.6	[59]
PM6:Y6	PCN3	0.860	26.73	77.35	17.80	[60]

Table S3. Photovoltaic parameters of PM6:Y6-based OSCs with different interfacial layers/electrodes

Active layer	Interfacial layer/electrode	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)	Ref.
PM6:Y6	g-C_3N_4:PEDOT:PSS	0.84	26.71	73	16.40	[61]
PM6:Y6	PEDOT:PSS-DA	0.84	25.52	77.1	16.6	[62]
PM6:Y6	PEDOT:PSS(TEMPO$^+$Br$^-$)	0.82	27.18	72.59	16.1	[63]
PM6:Y6	PEDOT:PSS-Nafion (4 : 1)	0.84	25.76	75.40	16.3	[64]
PM6:Y6	PEDOT:PSS-0.75%SG	0.841	26.58	78.21	17.4	[65]
PM6:Y6	PEDOT:PSS/$Ti_3C_2T_X$	0.830	25.63	68.40	14.5	[66]
PM6:Y6	CPA-PEDOT:PSS	0.84	25.5	77.0	16.5	[67]
PM6:Y6	CuSCN/TFB	0.85	24.45	72.69	15.1	[68]
PM6:Y6	WS_2	0.84	25.9	73	15.8	[69]
PM6:Y6:$PC_{71}BM$	WS_2	0.84	26.0	78	17	[69]
PM6:Y6	Aqueous MoO_x (200°C)	0.845	27.43	73.8	17.1	[70]
PM6:Y6	TMO	0.830	26.53	72.00	15.7	[71]
PM6:Y6	POE	0.830	25.14	68.11	13.8	[72]
PM6:Y6	BiOCl NPs	0.83	27.07	71.70	16.1	[73]
PM6:Y6	S-FrGO	0.77	24.64	69.9	13.3	[74]
PM6:Y6:$PC_{71}BM$	NDI-NI	0.86	25.40	76.14	16.86	[75]
PM6:Y6:$PC_{71}BM$	TiO_xN_y	0.850	26.18	76.41	17.02	[76]
PM6:Y6:$PC_{71}BM$	Phen-NaDPO:$Sn(SCN)_2$	0.850	26.20	70.00	15.60	[77]
PM6:Y6:$PC_{71}BM$	PEDOT:$PSS_{(wash)}$	0.850	25.86	76.20	16.75	[78]
PM6:Y6:$PC_{71}BM$	$PCPDTK_{0.50}H_{0.50}$-TT	0.854	25.10	75.9	16.3	[79]

($0.04\ cm^2$)						
PM6:Y6:$PC_{71}BM$ ($1\ cm^2$)	$PCPDTK_{0.50}H_{0.50}$-TT	0.872	17.33	67.5	10.2	[79]
PM6:Y6	PEIE-DBO	0.840	27.26	69.00	15.7	[80]
PM6:Y6	TEA-capped ZnO	0.820	29.19	65.20	15.6	[81]
PM6:Y6	ZnO:PBI-SO_3H	0.84	24.67	73.46	15.4	[82]
PM6:Y6	ZnO/KOH	0.85	27.1	68.1	15.7	[83]
PM6:Y6	ZnO/EDT	0.856	27.84	76.0	17.4	[84]
PM6:Y6	ZnO NPs/ASP-wrapped SD SWNTs	0.870	24.90	66.09	14.3	[85]
PM6:Y6	$ZnO/Ti_3C_2T_x$	0.830	26.38	75.40	16.51	[86]
PM6:Y6	PDINO-G	0.85	25.65	75.78	16.5	[87]
PM6:Y6	PDINO	0.821	25.58	72.24	15.17	[88]
PM6:Y6	PDINN	0.847	25.89	78.59	17.23	[88]
PM6:Y6	PDINO (15 nm)/Ag	0.861	26.15	77.80	17.48	[89]
PM6:Y6	PDINOH	0.849	77.95	26.23	17.60	[90]
PM6:Y6	QxTPPO1	0.86	26.37	75.00	16.83	[91]
PM6:Y6	QxTPPO2	0.85	25.23	74.00	15.94	[91]
PM6:Y6	POSSFN-G	0.849	25.34	73.93	15.9	[92]
PM6:Y6	ADMAFN-G	0.845	25.45	74.91	16.1	[92]
PM6:Y6	In_2O_3	0.834	26.40	73.44	16.17	[93]
PM6:Y6	Ga_2O_3	0.825	26.03	74.55	16.01	[93]
PM6:Y6	PFBP-Br	0.83	26.12	73.49	16.2	[94]
PM6:Y6	SnO_2 (10 nm)	0.831	26.63	72.75	16.1	[95]
PM6:Y6	SnO_2 (160 nm)	0.819	25.28	63.14	13.1	[95]
PM6:Y6	*n*-SnO_2/InP/ZnS QDs	0.845	25.48	70.63	15.22	[96]
PM6:Y6	crosslinked $Hf(ACBV)_4$	0.83	25.69	73.1	15.1	[97]
PM6:Y6	$Al(acac)_3$	0.817	20.71	73.34	12.4	[98]
PM6:Y6	HDSID	0.84	25.84	71.84	15.6	[99]

PM6:Y6	Os≡C (30)	0.87	25.21	74.19	16.28	[100]
PM6:Y6	PET/AgNWs-GV	0.826	25.14	73.60	15.28	[101]
PM6:Y6	T2-CNORH	0.863	25.50	70.70	15.50	[102]
PM6:Y6	ZnO/OSiND	0.830	25.31	76.93	16.16	[103]
PM6:Y6$_{(ST\text{-}OSCs)}$	AgNW-BM	0.716	20.84	61.26	9.12	[104]
PM6:Y6:$PC_{71}BM_{(ST\text{-}OSCs)}$	Ag/ITO	0.88	17.9	65.2	10.2	[105]
PM6:Y6$_{(ST\text{-}OSCs)}$	1 nm Au/20 nm Ag	0.854	22.12	72.27	14.20	[106]
PM6:Y6$_{(ST\text{-}OSCs)}$	MDM (B)	0.860	20.90	74.00	13.30	[107]
PM6:Y6$_{(FST\text{-}OSCs)}$	D-PEDOT:PSS	0.80	19.28	68.36	10.5	[108]
PM6:Y6$_{(flexible)}$	GR&AgNWs/PH1000	0.830	23.20	69.98	13.44	[109]
PM6:Y6$_{(flexible)}$	$Ti_3C_2T_x$/PEDOT:PSS	0.83	24.78	64.0	13.15	[110]
PM6:Y6	CNP-20/Glass/ITO	0.860	26.53	71.22	16.17	[111]
PM6:Y6	LBL H_2SO_4/EG	0.785	19.55	72.50	11.12	[112]
PM6:Y6$_{(flexible)}$	PI@GR	0.840	25.80	10.20	15.20	[113]
PM6:Y6$_{(flexible)}$	Em-Ag/AGNWs:AZO-SG	0.832	25.05	72.97	15.2	[114]
PM6:Y6$_{(flexible)}$	Treated cPI/AgN	0.830	26.03	70.00	15.12	[115]

Table S4. Contact angle of water(wat) and glycerol (oil) and surface tension of PEDOT:PSS and PEDOT:PSS-PDINN

HTL	θ_{wat} [deg]	θ_{oil} [deg]	γ_s [mN m^{-1}]
PEDOT:PSS	17.9	37.3	70.94
PEDOT:PSS-PDINN	18.4	44.9	73.85

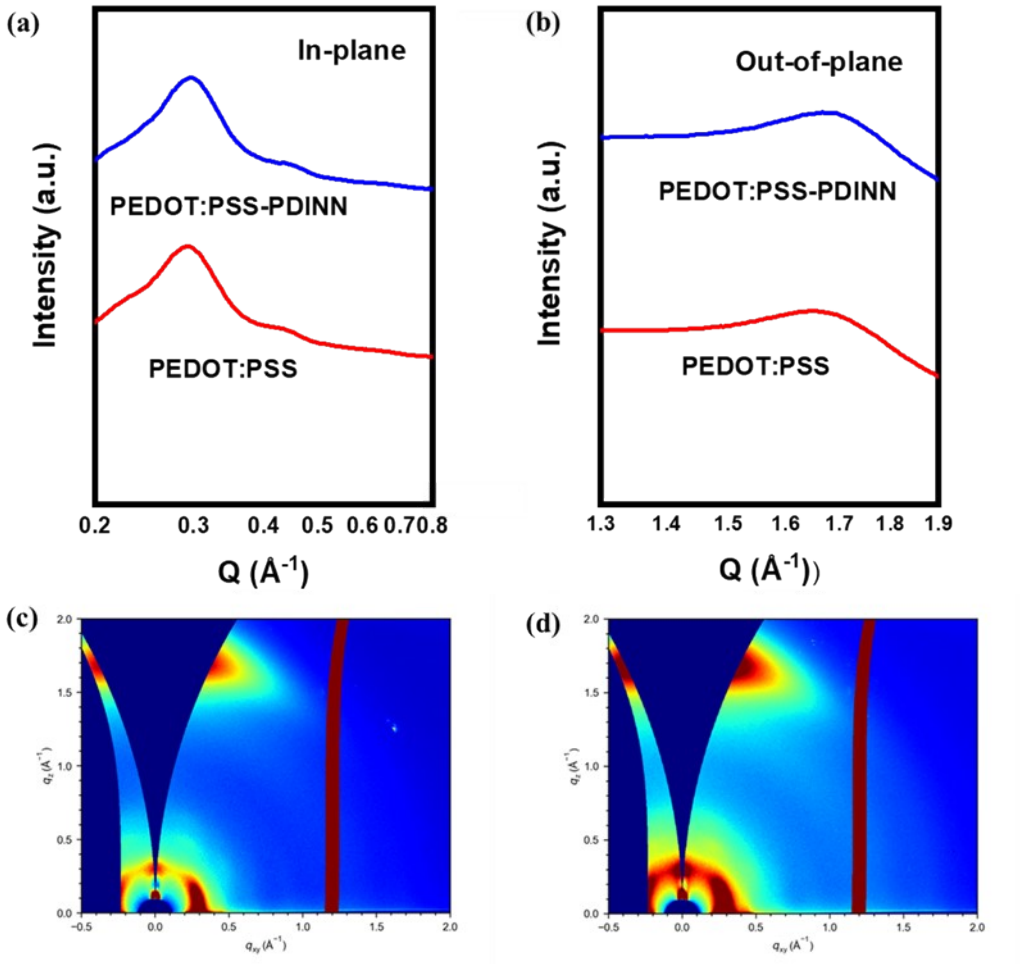

Figure S1. 1D profiles along the in-plane (a) and out-of-plane (b) directions and 2D GIWAXS patterns of the blend films based (c) PEDOT:PSS and (d) PEDOT:PSS-PDINN.

Table S5. Crystal Coherence lengths of the (010) peak and the d-spacing for the PM6:Y6 system prepared under two HTLs.

Out of Plane (010)				
HTL	location ($Å^{-1}$)	d-spacing(Å)	FWHM	CCL(Å)
PEDOT:PSS	1.64	3.82	0.15	37.26
PEDOT:PSS-PDINN	1.66	3.78	0.14	39.92

Two Terminal Conductivity Study

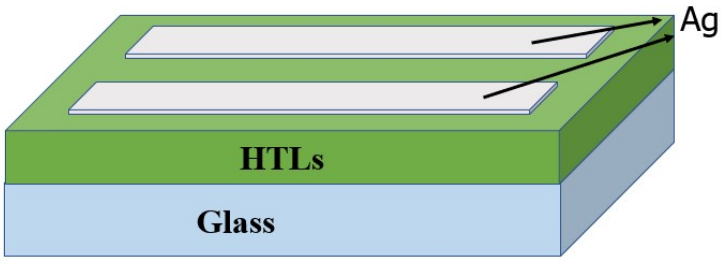

Figure S2. The schematic diagram of the device to measure the conductivity of HTL films.

The quartz sheet was coated with hole transport layer materials, and then 100 nm silver was deposited with a special mold. The calculation method is consistent with the paper.[3]

Calculation of Flory-Huggins interaction parameters.

The surface energy γ_s values could be calculated according to Wu model on the neat films by the Equation:

$$\gamma = \gamma^d + \gamma^p \quad (1)$$

$$\gamma_L(1+\cos\theta) = \frac{4\gamma_S{}^d\gamma_L{}^d}{\gamma_S{}^d + \gamma_L{}^d} + \frac{4\gamma_S{}^P\gamma_L{}^P}{\gamma_S{}^2 + \gamma_L{}^p} \quad (2)$$

And the two different contact angles of water and glycerol are measured to achieve the γ_s, And the γ is the sum of dispersion (d) and polar (p) components.

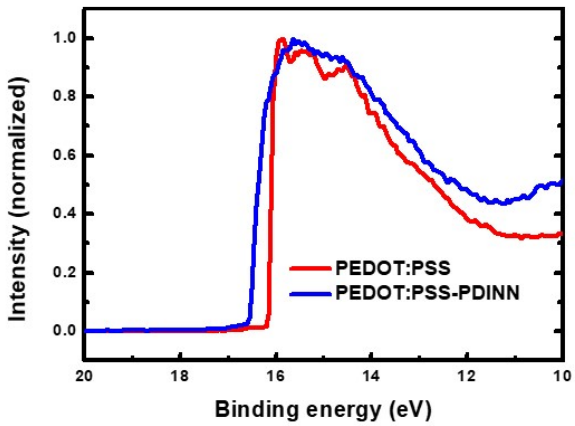

Figure S3. PEDOT:PSS and PEDOT:PSS-PDINN thin-film UPS. All UPS characterizations were taken from samples on ITO substrates.

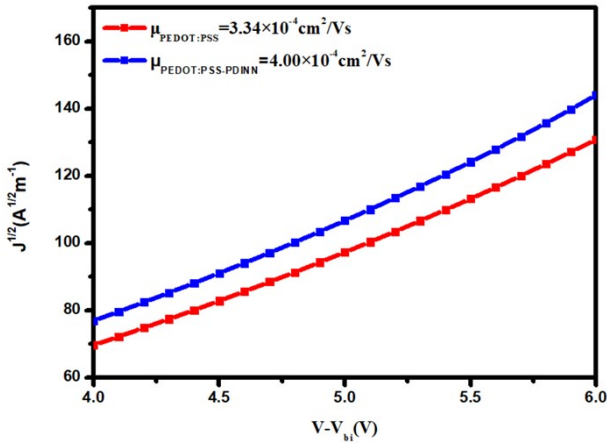

Figure S4. SCLC plots of BHJ films prepared on different HTLs

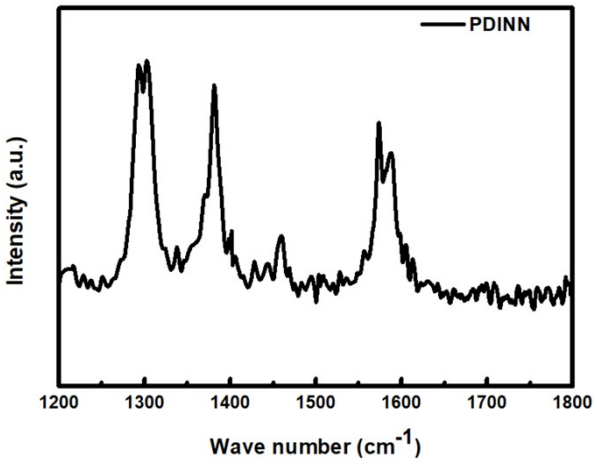

Figure S5. Raman spectrum of PDINN.

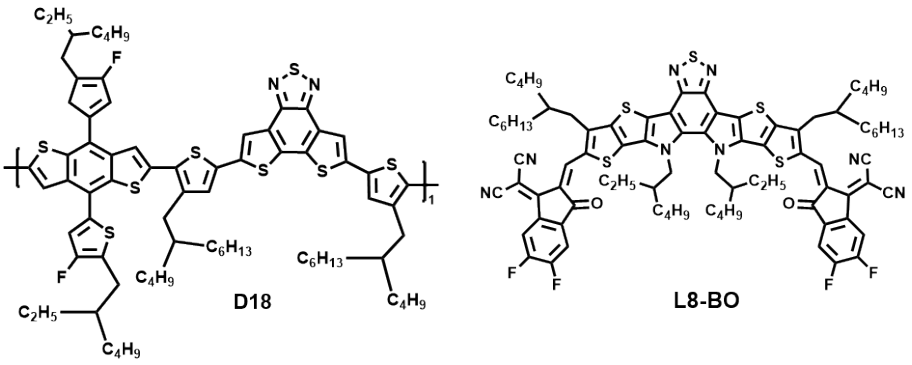

Figure S6. Molecular structures of D18 and L8-BO.

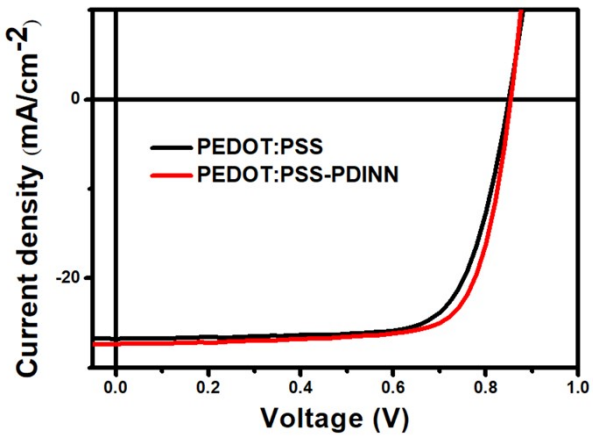

Figure S7. *J-V* diagram of the optimized D18:L8-BO system.

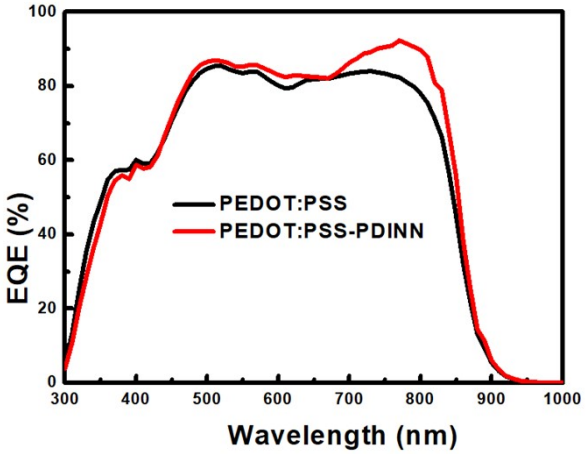

Figure S8. The EQE spectra of the devices based on optimized D18:L8-BO system.

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