

Supporting Information

A wide-bandgap copolymer donor based on phenanthridin-6(5H)-one unit

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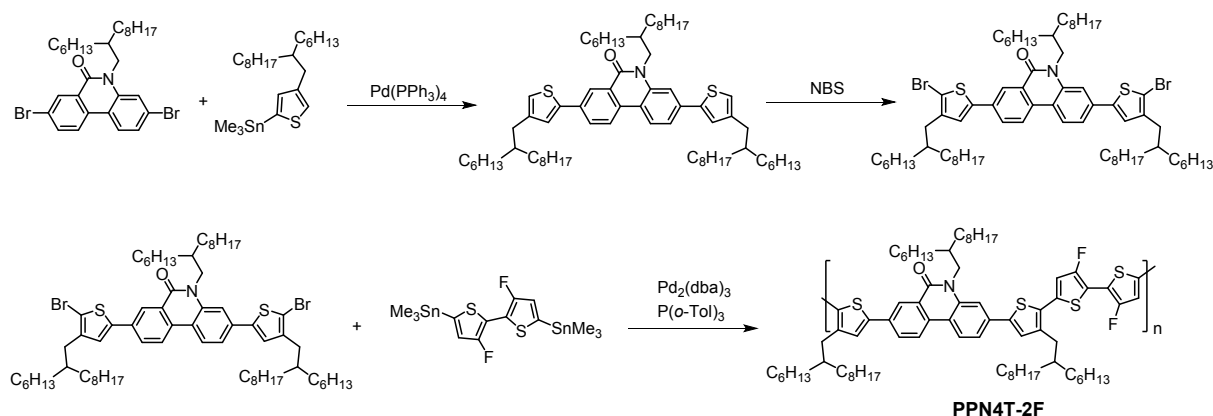
11. A summary for the photovoltaic performance of reported ultra-wide-bandgap copolymer donors

1. General characterization

^1H and ^{13}C NMR spectra were measured on a Bruker Avance-400 spectrometer. Absorption spectra were recorded on a Shimadzu UV-1800 spectrophotometer. Cyclic voltammetry was done by using a Shanghai Chenhua CHI620D voltammetric analyzer under argon in an anhydrous acetonitrile solution of tetra-*n*-butylammonium hexafluorophosphate (0.1 M). A glassy-carbon electrode was used as the working electrode, a platinum-wire was used as the counter electrode, and a Ag/Ag^+ electrode was used as the reference electrode. The copolymer was coated onto glassy-carbon electrode and all potentials were corrected against Fc/Fc^+ . AFM was performed on a Multimode microscope (Veeco) using tapping mode.

2. Synthesis

All reagents were purchased from J&K Co., Aladdin Co., Innochem Co., Derthon Co., SunaTech Co. and other commercial suppliers. All reactions dealing with air- or moisture-sensitive compounds were carried out by using standard Schlenk techniques. 3,8-Dibromo-5-(2-hexyldecyl)phenanthridin-6(5H)-one,^[1] and (3,3'-difluoro-[2,2'-bithiophene]-5,5'-diyl)bis(trimethylstannane)^[2] were synthesized according to literature.



Scheme S1 The synthetic route for PPN4T-2F.

5-(2-Hexyldecyl)-3,8-bis(4-(2-hexyldecyl)thiophen-2-yl)phenanthridin-6(5H)-one. To a solution of 3,8-dibromo-5-(2-hexyldecyl)phenanthridin-6(5H)-one (50 mg, 0.087 mmol) and (4-(2-hexyldecyl)thiophen-2-yl)trimethylstannane (122 mg, 0.26 mmol) in toluene (2.5 mL) and DMF (0.5 mL) was added $\text{Pd}(\text{PPh}_3)_4$ (10 mg, 0.0087 mmol) under N_2 . The mixture was heated to reflux and stirred overnight. After cooling to room temperature, the reaction mixture

was poured into water followed by extraction with CH₂Cl₂ three times. The combined organic layer was dried over anhydrous Na₂SO₄. After removal of the solvent, the crude product was purified via column chromatography (silica gel) by using CH₂Cl₂:petroleum ether (1:4) as eluent to give **5-(2-hexyldecyl)-3,8-bis(4-(2-hexyldecyl)thiophen-2-yl)phenanthridin-6(5H)-one** as a light yellow oil (65 mg, 72%). ¹H NMR (CDCl₃, 400 MHz, δ/ppm): 8.76 (s, 1H), 8.22 (d, *J* = 8.5 Hz, 2H), 7.95 (d, *J* = 8.4 Hz, 1H), 7.62 (s, 1H), 7.54 (d, *J* = 8.3 Hz, 1H), 7.34 (s, 1H), 7.23 (s, 1H), 6.91 (d, *J* = 6.6 Hz, 2H), 4.45 (br, 2H), 2.59-2.56 (m, 4H), 2.07 (br, 1H), 1.66 (s, 2H), 1.43-1.27 (m, 72H), 0.88-0.85 (m, 18H). ¹³C NMR (CDCl₃, 100 MHz, δ/ppm): 162.01, 143.42, 143.04, 142.50, 137.69, 135.47, 134.28, 132.02, 129.67, 125.88, 125.70, 125.31, 123.80, 122.19, 121.26, 121.21, 119.78, 118.39, 112.49, 46.53, 38.91, 36.75, 35.25, 35.18, 33.36, 31.92, 31.88, 31.83, 30.08, 30.05, 30.02, 29.78, 29.70, 29.65, 29.56, 29.34, 29.29, 26.95, 26.63, 22.68, 22.63, 14.12, 14.07. MALDI-TOF MS (*m/z*): 1032.5 (M + H⁺).

3,8-bis(5-bromo-4-(2-hexyldecyl)thiophen-2-yl)-5-(2-hexyldecyl)phenanthridin-6(5H)-one. To a solution of 5-(2-hexyldecyl)-3,8-bis(4-(2-hexyldecyl)thiophen-2-yl)phenanthridin-6(5H)-one (134 mg, 0.13 mmol) in a mixed solvent of CHCl₃ (5 mL) and DMF (1.7 mL) was added NBS (57.8 mg, 0.325 mmol). The mixture was stirred at room temperature for two hours. The mixture was poured into water and extracted with CH₂Cl₂ three times. The combined organic layer was dried over anhydrous Na₂SO₄. After removal of the solvent, the crude product was purified via column chromatography (silica gel) by using CH₂Cl₂:petroleum ether (1:4) as eluent to give **3,8-bis(5-bromo-4-(2-hexyldecyl)thiophen-2-yl)-5-(2-hexyldecyl)phenanthridin-6(5H)-one** as a yellow oil (100 mg, 65%). ¹H NMR (CDCl₃, 400 MHz, δ/ppm): 8.64 (s, 1H), 8.16-8.13 (m, 2H), 7.79 (d, *J* = 8.4 Hz, 1H), 7.46 (s, 1H), 7.42 (d, *J* = 8.3 Hz, 1H), 7.16 (s, 1H), 7.06 (s, 1H), 4.38 (br, 2H), 2.53-2.49 (m, 4H), 2.00 (br, 1H), 1.71 (s, 2H), 1.42-1.27 (s, 72H), 0.88-0.83 (m, 18H). ¹³C NMR (CDCl₃, 100 MHz, δ/ppm): 161.75, 142.79, 142.74, 142.57, 142.03, 137.74, 134.64, 133.43, 132.10, 129.21, 125.69, 125.39, 125.20, 124.95, 123.94, 122.33, 119.27, 118.50, 112.14, 109.94, 109.89, 46.48, 38.64, 38.61, 36.79, 34.46, 34.42, 33.39, 31.92, 31.90, 31.87, 30.11, 30.04, 30.01, 29.81, 29.71, 29.69, 29.63, 29.58, 29.35, 29.33, 29.31, 27.02, 26.55, 22.68, 22.65, 14.11. MALDI-TOF MS (*m/z*): 1190.3 (M + H⁺).

PPN4T-2F. To a mixture of 3,8-bis(5-bromo-4-(2-hexyldecyl)thiophen-2-yl)-5-(2-hexyldecyl)phenanthridin-6(5H)-one (106 mg, 0.085 mmol), (3,3'-difluoro-[2,2'-bithiophene]-

5,5'-diyl)bis(trimethylstannane) (56 mg, 0.085 mmol), Pd₂(dba)₃ (3.1 mg, 0.0034 mmol) and P(*o*-Tol)₃ (8.3 mg, 0.027 mmol) in a Schlenk flask was added toluene (2 mL) under argon. The mixture was heated to reflux for 24 h. Then the solution was cooled to room temperature and added into 150 mL methanol dropwise. The precipitate was collected and further purified via Soxhlet extraction by using methanol, hexane and chloroform in sequence. The residue was extracted by boiling chlorobenzene. The chlorobenzene fraction was concentrated and added into methanol dropwise. The precipitate was collected and dried under vacuum overnight to give **PPN4T-2F** as a black solid (56 mg, 46%). The *M_n* for PPN4TFBT is 65.6 kDa, with a PDI of 2.16. ¹H NMR (400 MHz, CDCl₃, δ/ppm): 8.21-6.81 (br, aromatic protons), 4.35 (br, aliphatic protons), 2.69 (br, aliphatic protons), 1.75-0.87 (br, aliphatic protons).

3. NMR

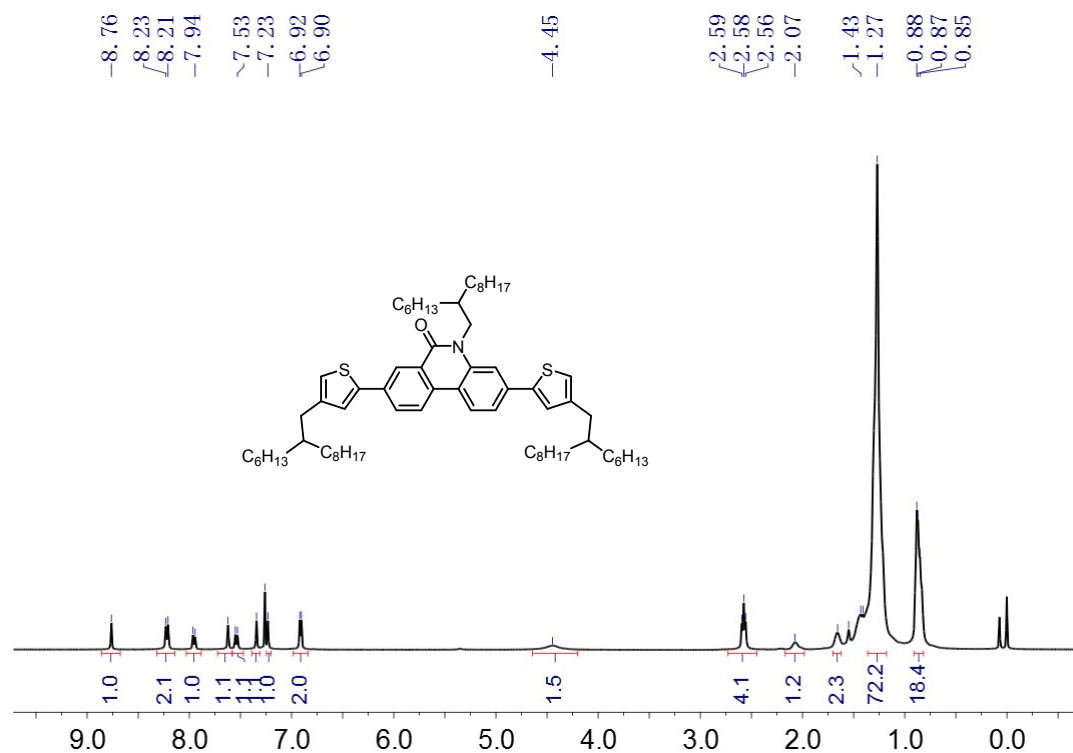


Fig. S1 ¹H NMR spectrum of 5-(2-hexyldecyl)-3,8-bis(4-(2-hexyldecyl)thiophen-2-yl)phenanthridin-6(5H)-one.

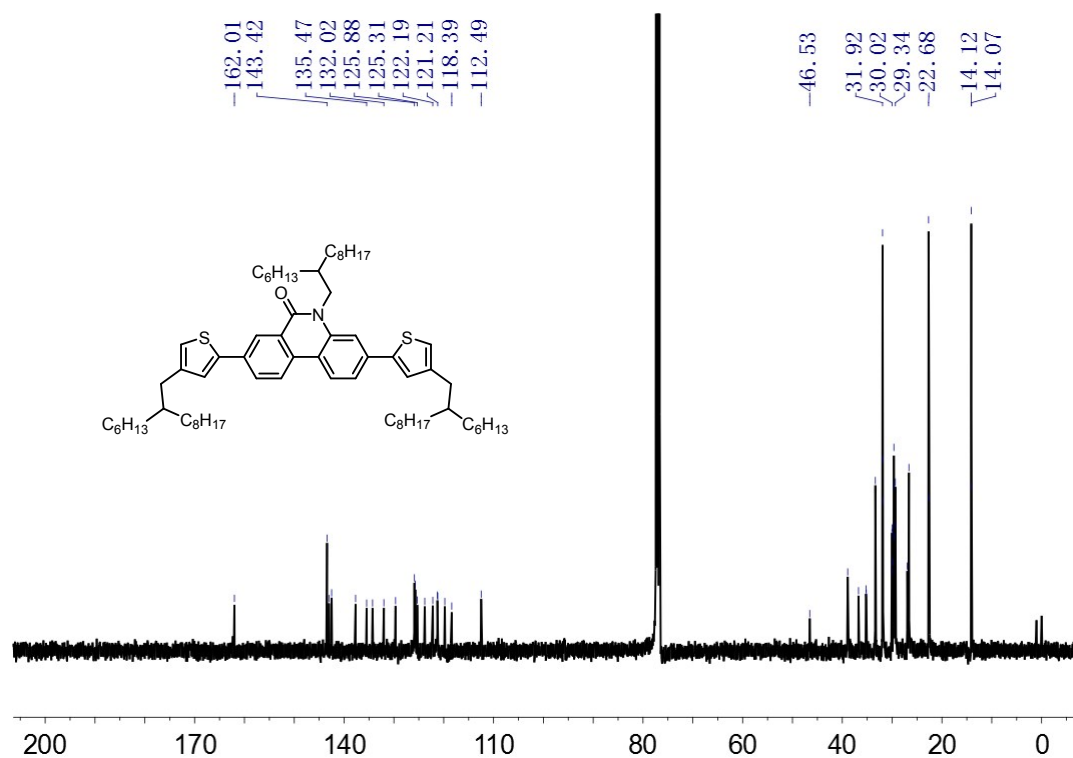


Fig. S2 ¹³C NMR spectrum of 5-(2-hexyldecyl)-3,8-bis(4-(2-hexyldecyl)thiophen-2-yl)phenanthridin-6(5H)-one.

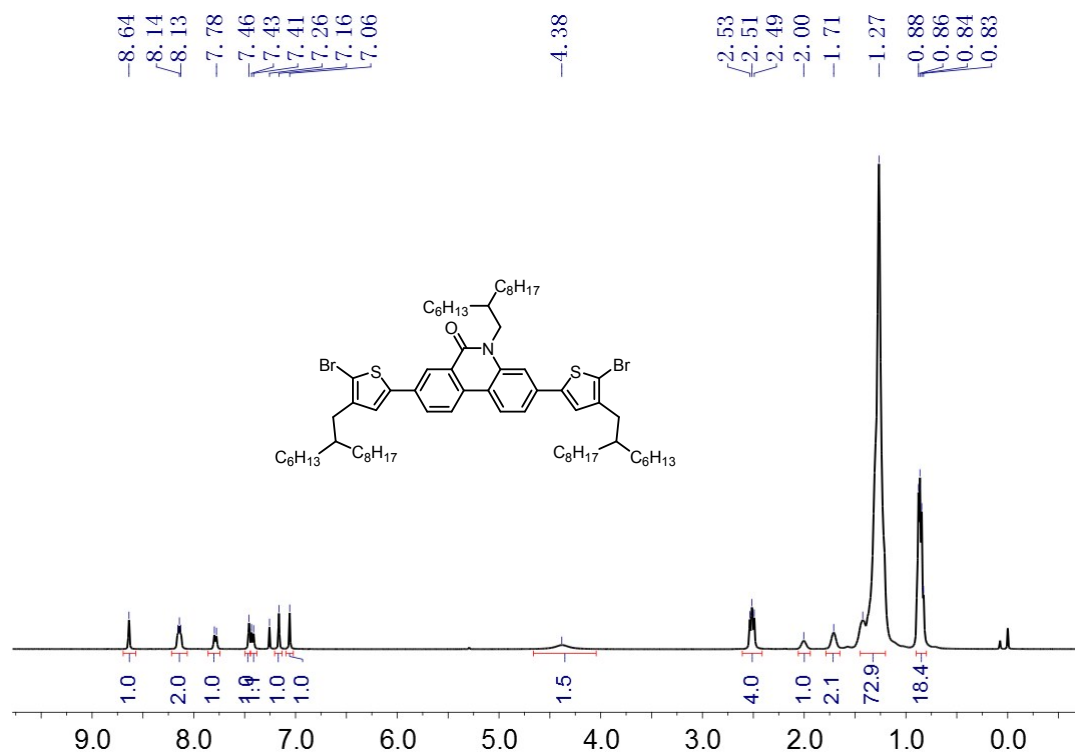


Fig. S3 ¹H NMR spectrum of **3,8-bis(5-bromo-4-(2-hexyldecyl)thiophen-2-yl)-5-(2-hexyldecyl)phenanthridin-6(5H)-one**.

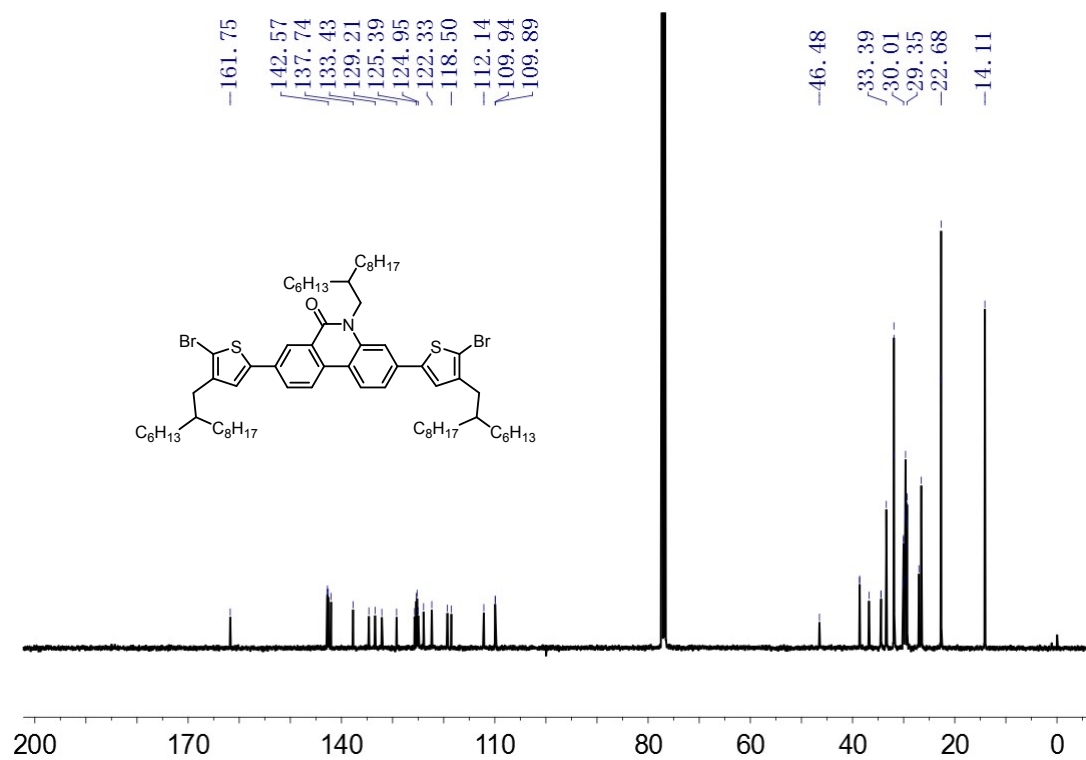


Fig. S4 ¹³C NMR spectrum of **3,8-bis(5-bromo-4-(2-hexyldecyl)thiophen-2-yl)-5-(2-hexyldecyl)phenanthridin-6(5H)-one**.

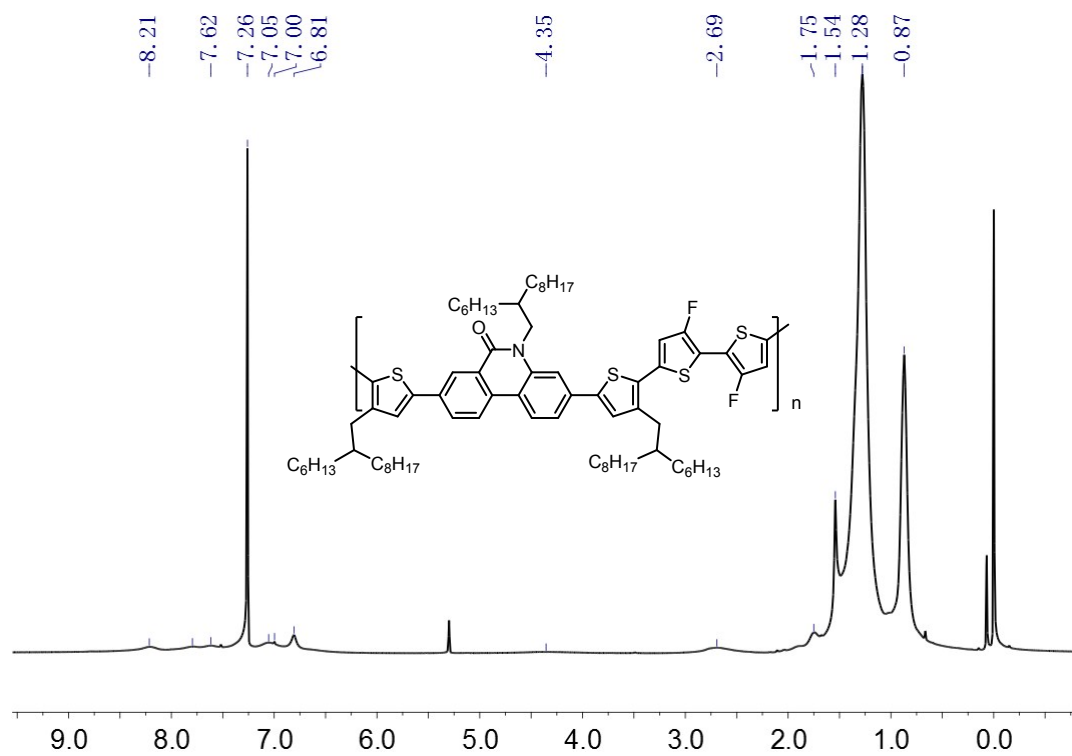


Fig. S5 ¹H NMR spectrum of PPN4T-2F.

4. CV

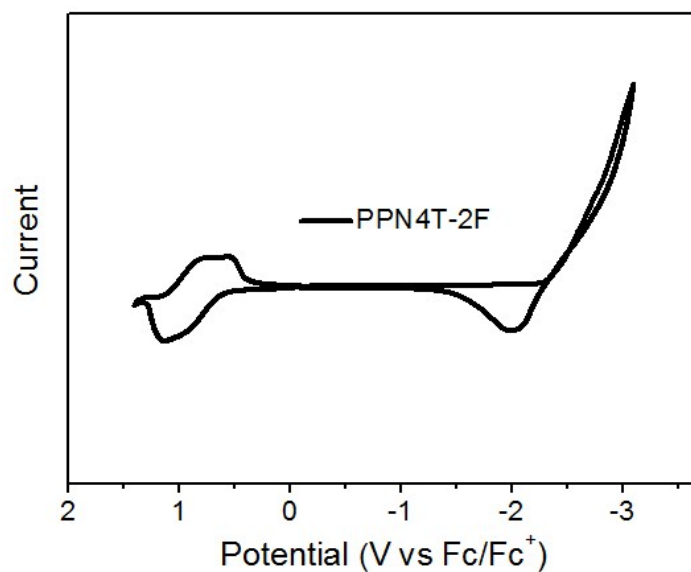


Fig. S6 Cyclic voltammogram for PPN4T-2F.

5. PL quenching

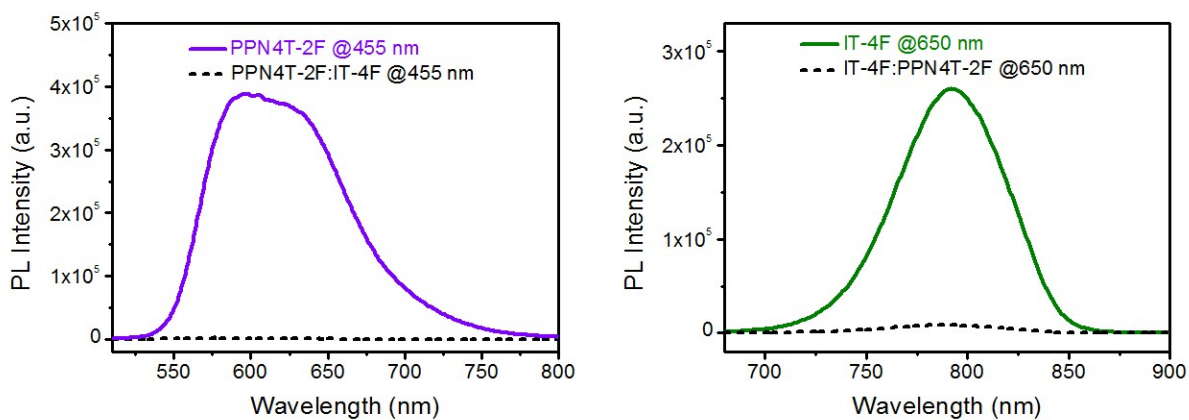


Fig. S7 PL quenching between PPN4T-2F and IT-4F. The weight ratio between PPN4T-2F and IT-4F in the blend film is 1:1.

6. Device fabrication and measurements

Inverted solar cells

The ZnO precursor solution was prepared according to literature.^[3] It was spin-coated onto ITO glass (4000 rpm for 30 s). The films were annealed at 200 °C in air for 30 min. ZnO film thickness is ~30 nm. A PPN4T-2F:IT-4F blend in chlorobenzene (CB) was spin-coated onto ZnO layer. MoO₃ (~6 nm) and Ag (~80 nm) was successively evaporated onto the active layer through a shadow mask (pressure ca. 10⁻⁴ Pa). The effective area for the devices is 4 mm². The thicknesses of the active layers were measured by using a KLA Tencor D-120 profilometer. *J-V* curves were measured by using a computerized Keithley 2400 SourceMeter and a Xenon-lamp-based solar simulator (Enli Tech, AM 1.5G, 100 mW/cm²). The illumination intensity of solar simulator was determined by using a monocrystalline silicon solar cell (Enli SRC2020, 2cm×2cm) calibrated by NIM. The external quantum efficiency (EQE) spectra were measured by using a QE-R3011 measurement system (Enli Tech).

Hole-only devices

The structure for hole-only devices is ITO/PEDOT:PSS/PPN4T-2F:IT-4F/MoO₃/Al. A 30 nm thick PEDOT:PSS layer was made by spin-coating an aqueous dispersion onto ITO glass (4000 rpm for 30 s). PEDOT substrates were dried at 150 °C for 10 min. A PPN4T-2F:IT-4F blend in CB was spin-coated onto PEDOT layer. Finally, MoO₃ (~6 nm) and Al (~100 nm) was successively evaporated onto the active layer through a shadow mask (pressure ca. 10⁻⁴ Pa). *J-V* curves were measured by using a computerized Keithley 2400 SourceMeter in the dark.

Electron-only devices

The structure for electron-only devices is Al/PPN4T-2F:IT-4F/Ca/Al. Al (~80 nm) was evaporated onto a glass substrate. A PPN4T-2F:IT-4F blend in CB was spin-coated onto Al. Ca (~5 nm) and Al (~80 nm) were successively evaporated onto the active layer through a shadow mask (pressure ca. 10⁻⁴ Pa). *J-V* curves were measured by using a computerized Keithley 2400 SourceMeter in the dark.

7. Optimization of device performance

Table S1 Optimization of D/A ratio for PPN4T-2F:IT-4F inverted solar cells.^a

D/A [w/w]	V_{oc} [V]	J_{sc} [mA/cm ²]	FF [%]	PCE [%]
1:0.6	0.86	12.53	57.9	6.23 (6.03) ^b
1:0.8	0.87	12.79	58.9	6.56 (6.33)
1:1	0.85	13.51	62.5	7.16 (7.08)
1:1.2	0.87	12.98	57.8	6.51 (6.39)

^a Blend solution: 18 mg/mL in CB with 0.5 vol% CN; spin-coating: 1500 rpm for 60 s.

^b Data in parentheses stand for the average PCEs for 10 cells.

Table S2 Optimization of the active layer thickness for PPN4T-2F:IT-4F inverted solar cells.^a

Thickness [nm]	V_{oc} [V]	J_{sc} [mA/cm ²]	FF [%]	PCE [%]
138	0.86	13.05	58.3	6.52 (6.28) ^b
112	0.85	13.51	62.5	7.16 (7.08)
82	0.87	13.10	62.3	7.10 (6.98)
61	0.85	12.87	58.2	6.35 (6.11)

^a D/A ratio: 1:1 (w/w); blend solution: 18 mg/mL in CB with 0.5 vol% CN.

^b Data in parentheses stand for the average PCEs for 10 cells.

Table S3 Optimization of CN content for PPN4T-2F:IT-4F inverted solar cells.^a

CN [vol%]	V_{oc} [V]	J_{sc} [mA/cm ²]	FF [%]	PCE [%]
0	0.87	13.35	57.1	6.66 (6.43) ^b
0.5	0.85	13.51	62.5	7.16 (7.08)
1	0.87	13.71	65.9	7.86 (7.63)
1.5	0.85	12.79	64.5	7.04 (6.78)
2	0.87	11.95	63.6	6.60 (6.44)

^a D/A ratio: 1:1 (w/w); blend solution: 18 mg/mL in CB; spin-coating: 1500 rpm for 60 s.

^b Data in parentheses stand for the average PCEs for 10 cells.

Table S4 Optimization of DIO content for PPN4T-2F:IT-4F inverted solar cells.^a

DIO [vol%]	V_{oc} [V]	J_{sc} [mA/cm ²]	FF [%]	PCE [%]
0	0.87	13.35	57.1	6.66 (6.43)
0.3	0.84	15.50	58.8	7.62 (7.44)
0.6	0.82	16.07	59.6	7.81 (7.58)
0.9	0.82	14.71	61.8	7.41 (7.12)
1.2	0.80	14.35	57.5	6.59 (6.17)

^a D/A ratio: 1:1 (w/w); blend solution: 18 mg/mL in CB; spin-coating: 1500 rpm for 60 s.

^b Data in parentheses stand for the average PCEs for 10 cells.

Table S5 Optimization of DIO content for PPN4T-2F:IT-4F (with 1 vol% CN) inverted solar cells.^a

DIO [vol%]	V_{oc} [V]	J_{sc} [mA/cm ²]	FF [%]	PCE [%]
0	0.87	13.71	65.9	7.86 (7.63) ^b
0.3	0.85	15.24	63.3	8.19 (7.95)
0.6	0.82	16.27	64.2	8.54 (8.32)
0.9	0.83	15.39	64.1	8.20 (7.92)
1.2	0.84	13.95	57.4	6.74 (6.16)

^a D/A ratio: 1:1 (w/w); blend solution: 18 mg/mL in CB with 1 vol% CN; spin-coating: 1500 rpm for 60 s.

^b Data in parentheses stand for the average PCEs for 10 cells.

8. Exciton dissociation probabilities

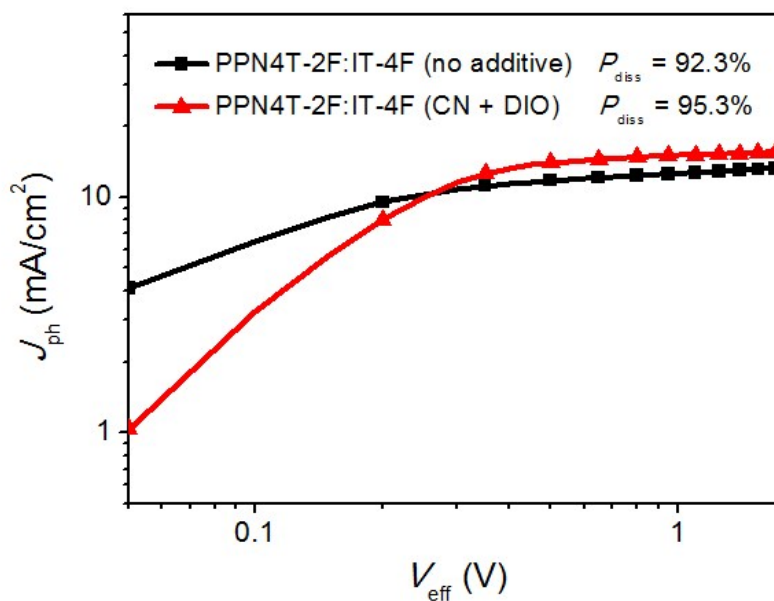


Fig. S8 J_{ph} - V_{eff} plots.

9. Bimolecular recombination

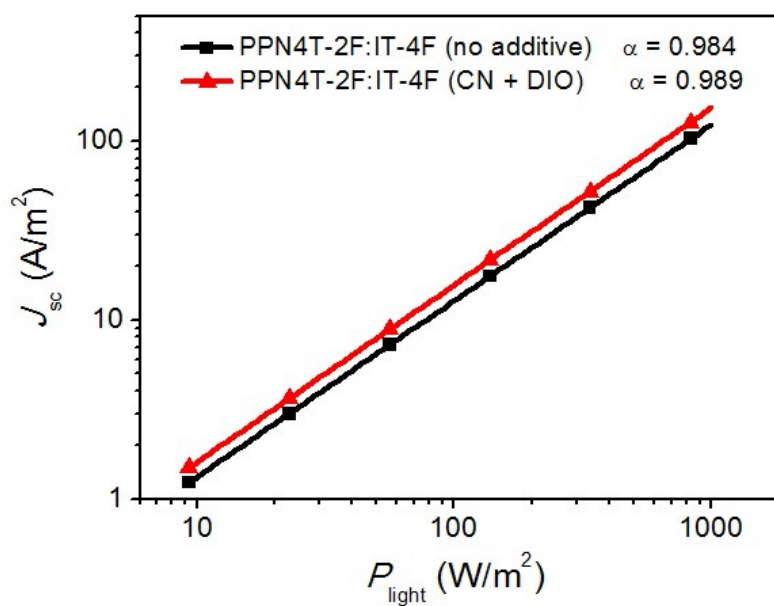


Fig. S9 J_{sc} - P_{light} plots.

10. SCLC

Charge carrier mobility was measured by SCLC method. The mobility was determined by fitting the dark current to the model of a single carrier SCLC, which is described by:

$$J = \frac{9}{8} \varepsilon_0 \varepsilon_r \mu \frac{V^2}{d^3}$$

where J is the current density, μ is the zero-field mobility of holes (μ_h) or electrons (μ_e), ε_0 is the permittivity of the vacuum, ε_r is the relative permittivity of the material, d is the thickness of the blend film, and V is the effective voltage ($V = V_{\text{appl}} - V_{\text{bi}}$, where V_{appl} is the applied voltage, and V_{bi} is the built-in potential determined by electrode work function difference). Here, $V_{\text{bi}} = 0.1$ V for hole-only devices, $V_{\text{bi}} = 0$ V for electron-only devices.^[4] The mobility was calculated from the slope of $J^{1/2}$ - V plots.

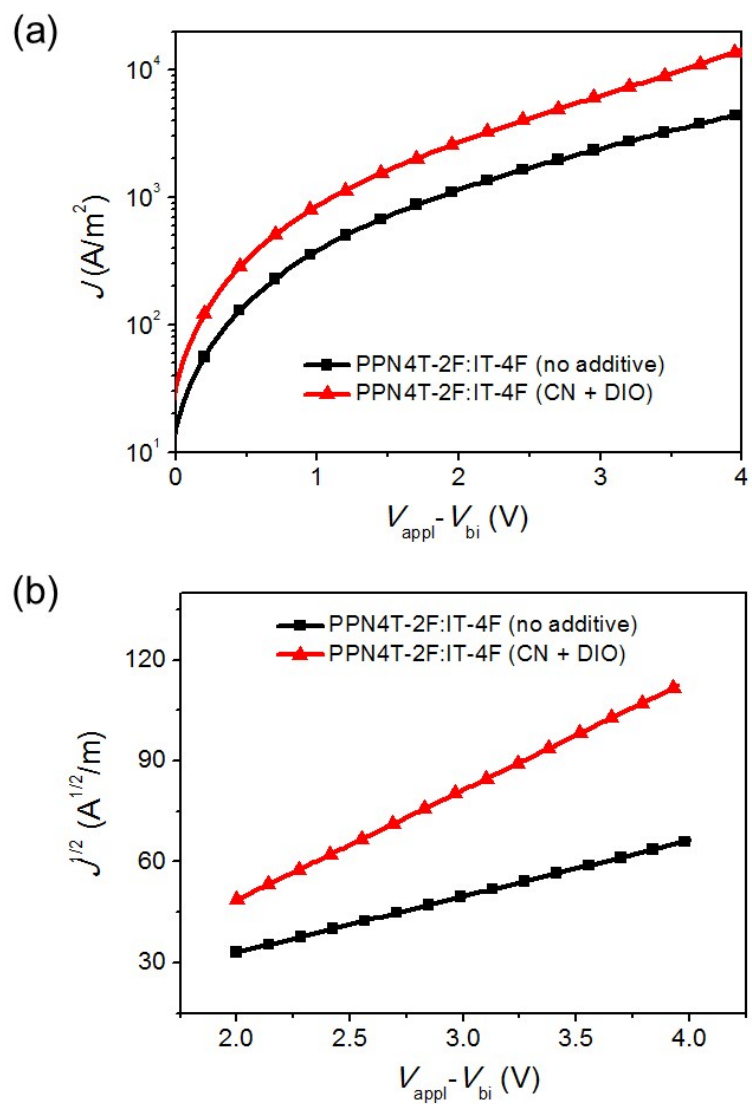


Fig. S10 J - V curves (a) and corresponding $J^{1/2}$ - V plots (b) for the hole-only devices (in dark). The thicknesses for PPN4T-2F:IT-4F blend films without and with additives are 99 nm and 102 nm, respectively.

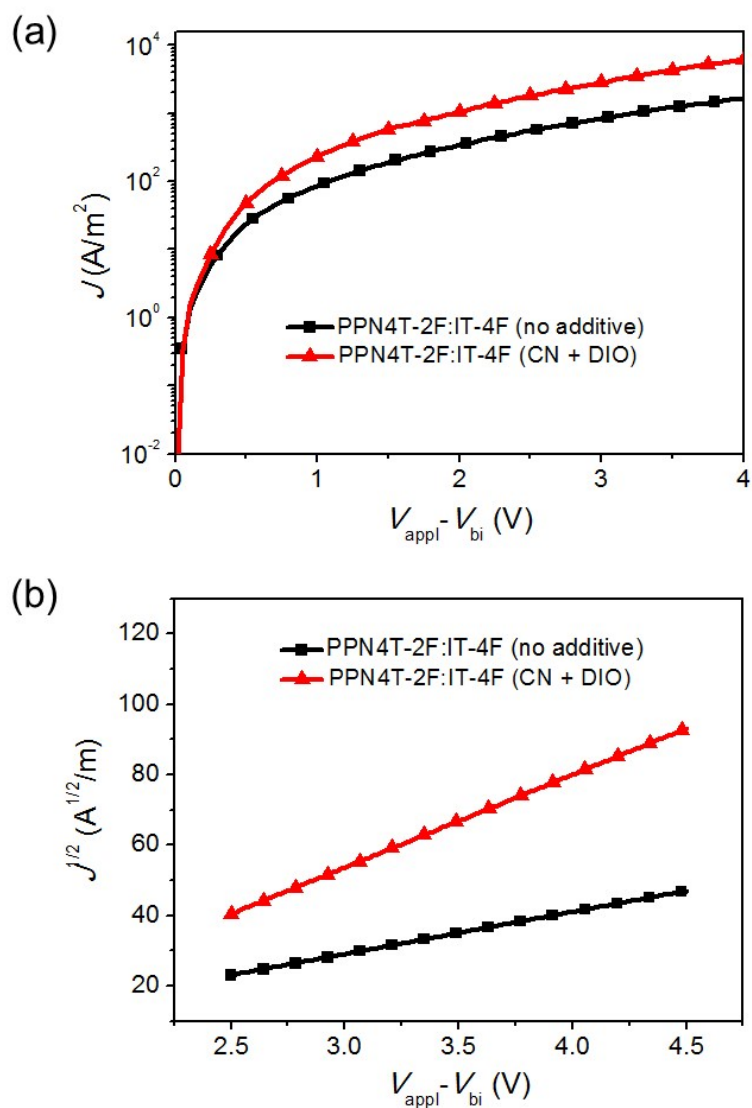


Fig. S11 J - V curves (a) and corresponding $J^{1/2}$ - V plots (b) for the electron-only devices (in dark). The thicknesses for PPN4T-2F:IT-4F blend films without and with additives are 92 nm and 96 nm, respectively.

Table S6. Hole and electron mobilities for PPN4T-2F:IT-4F blend films without and with additives.

Additive	μ_h [cm ² V ⁻¹ s ⁻¹]	μ_e [cm ² V ⁻¹ s ⁻¹]	μ_h/μ_e
none	9.06×10^{-5}	2.86×10^{-5}	3.17
1 vol% CN + 0.6 vol% DIO	3.83×10^{-4}	1.58×10^{-4}	2.42

11. A summary for the photovoltaic performance of reported ultra-wide-bandgap copolymer donors

Table S7 Solar cell performance of reported ultra-wide-bandgap copolymer donors ($E_g^{\text{opt}} > 2.07$ eV).

Polymer	E_g^{opt} (eV)	Device structure	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)	Reference
O-PBDT	2.21	ITO/PEDOT:PSS/O-PBDT:PC ₇₁ BM/Ca/Al	4.18	0.83	45.0	1.56	<i>Chem. Mater.</i> , 2012, 24 , 2534.
PBDTT	2.13	ITO/PEDOT:PSS/PBDTT:PC ₇₁ BM/LiF/Al	11.95	0.93	55.0	6.12	<i>Chem. Mater.</i> , 2015, 27 , 2653.
PBDTN	2.22	ITO/PEDOT:PSS/PBDTN:PC ₇₁ BM/LiF/Al	9.20	0.83	41.0	3.20	<i>ACS Appl. Mater. Inter.</i> , 2014, 6 , 15774.
PBDT-BT	2.10	ITO/MoO ₃ /PBDT-BT:PC ₇₁ BM/LiF/Al	5.86	0.82	45.8	2.20	<i>Chem. Mater.</i> , 2015, 27 , 6858.
PBDT[2H]T	2.10	ITO/PEDOT:PSS/PBDT[2H]T:PC ₇₁ BM/Ca/Al	10.70	0.90	72.0	7.00	<i>Chem. Mater.</i> , 2015, 27 , 4184.
PBDT[2F]T	2.10	ITO/PEDOT:PSS/PBDT[2F]T:PC ₇₁ BM/Ca/Al	6.30	0.80	58.0	2.90	<i>Chem. Mater.</i> , 2015, 27 , 4184.
PCDTTPD-ic	2.21	ITO/PEDOT:PSS/PCDTTPD-ic:[60]PCBM/Al	0.56	1.01	25.0	0.14	<i>Macromolecules.</i> , 2012, 45 , 1833.
P1	2.12	ITO/PEDOT:PSS/P1:PC ₇₁ BM/LiF/Al	4.40	0.96	53.0	2.23	<i>Macromol. Rapid Commun.</i> , 2013, 34 , 1163.
P2	2.11	ITO/PEDOT:PSS/P2:PC ₇₁ BM/LiF/Al	3.00	0.89	53.0	1.42	<i>Macromol. Rapid Commun.</i> , 2013, 34 , 1163.
P2	2.10	ITO/PEDOT:PSS/P2:PC ₇₁ BM/Ca/Al	3.43	0.80	54.7	1.50	<i>Polym. Chem.</i> , 2013, 4 , 2174.
Pa	2.13	ITO/PF3N-OX/Pa:PC ₇₁ BM/MoO ₃ /Al	8.19	0.92	60.2	4.54	<i>J. Mater. Chem. A.</i> , 2014, 2 , 321.
Pb	2.19	ITO/PEDOT:PSS/Pb:PC ₇₁ BM/Ca/Al	1.51	0.66	46.2	0.46	<i>J. Mater. Chem. A.</i> , 2014, 2 , 321.
Pc	2.21	ITO/PEDOT:PSS/Pc:PC ₇₁ BM/Ca/Al	5.46	0.66	59.6	2.15	<i>J. Mater. Chem. A.</i> , 2014, 2 , 321.
PBDTT-LTPD	2.19	ITO/PEDOT:PSS/PBDTT-LTPD:PC ₇₁ BM/Ca/Al	14.32	1.00	52.0	7.59	<i>Adv. Sci.</i> , 2015, 2 , 1500021.
PID2	2.12	ITO/PEDOT:PSS/PID2:PC ₇₁ BM/Ca/Al	5.94	0.88	58.6	3.05	<i>Adv. Funct. Mater.</i> , 2014, 24 , 3432.
PV-BDTC2	2.09	ITO/PEDOT:PSS/PV-BDTC2:PC ₇₁ BM/Ca/Al	10.37	1.03	70.0	7.49	<i>J. Mater. Chem. A.</i> , 2016, 4 , 18792.
PIDT-SO	2.27	ITO/PFN/PIDT-SO:PC ₇₁ BM/MoO ₃ /Al	6.96	0.97	34.0	2.31	<i>Org. Electron.</i> , 2014, 15 , 2950.
PIDT-DHTSO	2.20	ITO/PFN/PIDT-DHTSO:PC ₇₁ BM/MoO ₃ /Al	8.20	0.95	48.0	3.81	<i>Org. Electron.</i> , 2014, 15 , 2950.
PBDTTTP	2.11	ITO/PEDOT:PSS/PBDTTTP:PC ₇₁ BM/Ca/Al	8.12	0.98	69.5	5.53	<i>Polym. Chem.</i> , 2016, 7 , 1027.
PBDTT	2.13	ITO/PEDOT:PSS/PBDTT-IDIC/PFN/Al	13.7	0.88	43.9	5.3	<i>Macromol. Rapid Commun.</i> , 2018, 1800660.
PBDT-ODZ	2.12	ITO/CuSCN/PBDT-ODZ:ITIC-Th/PFN-Br/Al	15.52	1.08	60.4	10.12	<i>Adv. Mater.</i> , 2018, 30 , 1800737.
PBDTS-TDZ	2.09	ITO/ZnO/PBDTS-TDZ:ITIC/MoO ₃ /Ag	17.78	1.10	65.4	12.8	<i>Adv. Mater.</i> , 2018, 30 , 1703973.

P(o-FDBND-2T)	2.29	ITO/V ₂ O ₅ /P(o-FDBND-2T):PC ₇₁ BM/Ca/Al	2.63	0.87	62	1.44	<i>Chem. Mater.</i> , 2017, 29 , 9162.
P(o-2FDBND-2T)	2.29	ITO/V ₂ O ₅ /P(o-2FDBND-2T):PC ₇₁ BM/Ca/Al	0.89	1.50	56	0.79	<i>Chem. Mater.</i> , 2017, 29 , 9162.
P(p-FDBND-2T)	2.29	ITO/V ₂ O ₅ /P(p-FDBND-2T):PC ₇₁ BM/Ca/Al	10.12	0.89	66	6.55	<i>Chem. Mater.</i> , 2017, 29 , 9162.
P(p-2FDBND-2T)	2.27	ITO/V ₂ O ₅ /P(p-2FDBND-2T):PC ₇₁ BM/Ca/Al	7.86	0.90	67	5.27	<i>Chem. Mater.</i> , 2017, 29 , 9162.
PTNT	2.20	ITO/PEDOT:PSS/PTNT:PC ₇₁ BM/LiF/Al	8.1	0.90	63	5.0	<i>J. Am. Chem. Soc.</i> , 2014, 136 , 11578.
PIDTT-TT	2.14	ITO/ZnO/PEIE/PIDTT-TT:PC ₇₁ BM/MoO ₃ /Ag	11.2	0.96	66	7.10	<i>J. Mater. Chem. A.</i> , 2017, 5 , 712.

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