

# A Universal Fluorinated Polymer Acceptor Enables All-Polymer Solar Cells with Efficiency Over 15%

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## Experimental section

### Materials

PBDB-T, PM6, 2,5-bis(trimethylstannyl)thiophene, 1-bromo-2-fluoro-4,5-dimethylbenzene and other reagents and solvents were purchased from Dongguan Alda New Materials Technology Co. Ltd, and used directly without further purification. Compound 5, PTzBI-oF and PY5T were synthesized according to the previous reports.<sup>1,2</sup>

### Synthesis of Compound 1

1-Bromo-2-fluoro-4,5-dimethylbenzene (5.00 g, 24.63 mmol), potassium hydroxide (1.97 g, 49.26 mmol) and deionized water (150 mL) were heated to reflux. Then potassium permanganate (15.57 g, 98.52 mmol) was carefully added into the reaction and the mixture was kept reflux overnight. After cooling and filtering, the filtrate was extracted with ethyl acetate (100 mL) for three times. Compound 1 (6.18 g, 23.50 mmol, yield = 95%) was obtained after evaporating the organic phase as a white solid. <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>) δ (ppm): 13.77 (s, 2H), 8.06 (d, *J* = 6.7 Hz, 1H), 7.69 (d, *J* = 8.9 Hz, 1H); <sup>13</sup>C NMR (101 MHz, DMSO-d<sub>6</sub>) δ (ppm): 167.72, 167.19, 161.42, 158.93, 136.20, 136.13, 135.19, 131.26, 117.94, 117.69, 111.55, 111.34.

### Synthesis of compound 2

Compound 1 (6.00 g, 22.81 mmol) and acetic anhydride (50 mL) were heated to reflux for 4 h. The reaction solution was evaporated to afford compound 2 (5.30 g, 21.63 mmol, yield = 92%) as a white solid. <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>) δ (ppm): 8.05 (d, *J* = 6.7 Hz, 1H), 7.68 (d, *J* = 8.9 Hz, 1H); <sup>13</sup>C NMR (101 MHz, DMSO-d<sub>6</sub>) δ (ppm): 166.81,

166.28, 160.24, 158.25, 135.18, 135.12, 134.11, 130.21, 116.87, 116.67, 110.60, 110.43.

### **Synthesis of compound 3**

Compound 2 (5.00 g, 20.40 mmol), tert-butyl acetoacetate (3.55 g, 22.44 mmol) and trimethylamine (5 mL) were added into acetic anhydride (60 mL) and stirred at room temperature overnight. Then the reaction solution was carefully poured into 300 mL hydrochloric acid ice solution (1 M) and then heated to 80 °C for 2 h. After cooling, compound 3 (4.26 g, 17.54 mmol, yield = 86%) was gained by filtering as a gray solid. <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>) δ (ppm): 8.23 (d, *J* = 5.9 Hz, 1H), 7.64 (d, *J* = 6.6 Hz, 1H), 3.27 (s, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ (ppm): 195.13, 194.34, 173.32, 144.34, 139.92, 137.46, 129.53, 110.39, 44.94.

### **Synthesis of compound 4 (4a and 4b)**

Compound 3 (4.00 g, 16.46 mmol), malononitrile (1.30 g, 19.75 mmol), sodium acetate (1.62 g, 19.75 mmol) and ethanol (100 mL) were stirred at room temperature overnight. After acidizing to pH ≈ 1 by dilute hydrochloric acid, the mixture was extracted with ethyl acetate (200 mL) and washed by sodium chloride aqueous solution. The organic phase was evaporated. The crude product was purified by column chromatography with petroleum ether/dichloromethane (1:2) as eluent to give compound 4 (blend of 4a and 4b) (4.08 g, 14.02 mmol, yield = 85%) as a caesious powder. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ (ppm): 8.89 (d, *J* = 5.6 Hz, 0.47H), 8.34 (d, *J* = 7.9 Hz, 0.53H), 8.21 (d, *J* = 6.3 Hz, 0.53H), 7.65 (d, *J* = 6.4 Hz, 0.47H), 3.76 (s, 2H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ (ppm): 192.63, 191.94, 164.66, 163.99, 163.43, 162.06, 142.92, 141.97, 138.97,

137.28, 131.60, 130.39, 119.87, 119.64, 113.28, 113.01, 111.76, 111.62, 111.55, 80.81, 79.95, 43.41, 43.17.

### Synthesis of Y5-F-Br

Compound 4 (1.18 g, 4.06 mmol), compound 5 (2.00 g, 1.35 mmol), pyridine (6 mL), and chloroform (200 mL) were heated to reflux for 12 h. After cooling, the reaction solution was poured into methanol (200 mL) and then filtered. The residue was further purified by column chromatography with petroleum ether/dichloromethane (8:1) as eluent to give Y5-F-Br (2.36 g, 1.17 mmol, yield = 86%) a dark blue powder. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ (ppm): 9.18 (s, 2H), 8.95 (d, *J* = 5.6 Hz, 0.65H), 8.45 (d, *J* = 8.6 Hz, 1.35H), 8.11 (d, *J* = 6.4 Hz, 1.35H), 7.60 (d, *J* = 6.3 Hz, 0.65H), 4.76 (d, *J* = 7.5 Hz, 4H), 3.23 (t, *J* = 7.6 Hz, 4H), 2.10 (d, *J* = 4.5 Hz, 2H), 1.93 – 1.82 (m, 4H), 1.53 (dt, *J* = 15.0, 7.3 Hz, 4H), 1.42 – 0.73 (m, 126H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ (ppm): 186.33, 185.97, 163.88, 163.71, 161.83, 161.64, 158.89, 158.66, 154.13, 154.09, 147.63, 145.39, 140.87, 140.79, 138.74, 138.68, 137.85, 136.58, 136.42, 136.32, 135.71, 135.63, 134.41, 133.88, 133.86, 133.69, 133.61, 131.25, 130.96, 128.96, 120.13, 119.97, 117.40, 117.22, 116.97, 116.79, 115.16, 114.75, 114.68, 113.78, 113.54, 113.32, 111.09, 110.89, 68.96, 55.81, 39.3, 32.10, 32.07, 31.40, 30.69, 30.04, 30.01, 29.92, 29.85, 29.82, 29.78, 29.71, 29.69, 29.61, 29.56, 29.51, 29.49, 25.72, 22.86, 22.84, 14.27; <sup>19</sup>F NMR (471 MHz, CDCl<sub>3</sub>) δ (ppm): -93.22 (s, 0.67F), -94.26 (s, 0.33F).

### Synthesis of PFA1

Y5-F-Br (202.3 mg, 0.1 mmol), 2,5-bis(trimethylstannyl)thiophene (41.0 mg, 0.1 mmol) and toluene (2 mL) were added into a sealed tube. After bubbling with N<sub>2</sub> for 15 min.,

tris(dibenzylideneacetone)dipalladium (2 mg) and tris(2-methylphenyl)phosphine (4 mg) were added into the solution. The mixture was sealed and then allowed to react at 130 °C for 24 h. After cooling to room temperature, the reaction solution was dropwise added into 150 mL methanol and the precipitate was filtered out. The crude polymer was washed with methanol, acetone and n-hexane in a Soxhlet apparatus, then redissolved in chloroform. The solution was concentrated to 15 mL and reprecipitated in 150 mL methanol. The anticipated PFA1 was obtained by filtration and drying under vacuum as a dark solid (yield = 91%).

### **Measurements**

<sup>1</sup>H NMR, <sup>13</sup>C NMR and <sup>19</sup>F NMR spectra were recorded on a Bruker DRX 400 spectrometer. Molecular weights of the polymers were measured on an Acquity Advanced Polymer Chromatography (Waters) with high temperature chromatograph, using 1,2,4-trichlorobenzene as the mobile phase at 150 °C. UV-vis absorption spectra and photoluminescence (PL) spectra were measured on a Shimadzu UV-3600 spectrometer and a HORIBA scientific fluoromax-4 spectro-fluorometer, respectively. The polymer films used for absorption coefficient measurement were spin-coated from chloroform solution (with concentration of about 6 mg mL<sup>-1</sup>) onto quartz plate substrate (with the thickness of ~100 nm), then annealing at 100°C for 10 min. The measurement of absorption coefficient of polymers in chloroform solution was carried out by dissolving polymer in chloroform with concentration of about 1.0×10<sup>-5</sup> g mL<sup>-1</sup>. After subtracting the signal of blank sample, UV-vis absorption intensity was measured on a

Shimadzu UV-3600 spectrometer. Cyclic voltammetry (CV) measurements were performed on a CHI800C electrochemical workstation equipped with a glass carbon working electrode, a saturated calomel reference electrode and a Pt sheet counter electrode, using Bu<sub>4</sub>NPF<sub>6</sub> acetonitrile solution (0.1 M) as the electrolyte. The ferrocene/ferrocenium redox couple (Fc/Fc<sup>+</sup>) was used as the external standard and the redox potential of Fc/Fc<sup>+</sup> ( $E_{\text{Fc/Fc}^+}$ ) was determined to be 0.38 V. With regard to energy level of the ferrocene reference (4.8 eV to vacuum), the highest occupied molecular orbital energy level ( $E_{\text{HOMO}}$ ) and the lowest unoccupied molecular orbital energy level ( $E_{\text{LUMO}}$ ) are calculated according to the equations of  $E_{\text{HOMO}} = -e(E_{\text{ox}} + 4.8 - E_{\text{Fc/Fc}^+})$  and  $E_{\text{LUMO}} = -e(E_{\text{red}} + 4.8 - E_{\text{Fc/Fc}^+})$ , respectively, where  $E_{\text{ox}}$  and  $E_{\text{red}}$  are onset of the first oxidation and reduction potentials. Atomic force microscopy (AFM) images were recorded on a Bruker Multimode 8 microscope in the tapping mode. Transmission electron microscopy (TEM) images were obtained using a JEM-2100F instrument. Grazing incidence wide-angle X-ray scattering (GIWAXS) measurements was performed on a Xenocs Xeuss 2.0 system with an Excillum MetalJet-D2 X-ray source operated at 70.0 kV, 2.8570 mA, and a wavelength of 1.341 Å. The sample to detector distance was ~223 mm and the incidence angle was set at 0.20°. Scattering images were recorded using a DECTRIS PILATUS3 R 1M area detector with a pixel size of 0.172 mm by 0.172 mm. Samples were prepared by spin-coating the films on PEDOT:PSS/silicon wafer substrates.

## **Device fabrication and characterization**

Organic solar cells with a conventional structure of ITO/PEDOT:PSS/active layer/PFN-Br/Ag were fabricated according to a well-established operating procedures. Patterned indium tin oxide (ITO) glass substrates were cleaned with acetone, isopropanol, detergent, deionized water and isopropanol in succession. After dried in vacuum at 80 °C overnight, ITO substrates underwent 2 min oxygen plasma treatment. A 40 nm thick PEDOT:PSS layer was spin-coated on the ITO substrates and then baked at 150 °C for 20 min. The PTzBI-oF:PY5T and PTzBI-oF:PFA1 blends with the ratio of 1:1 were dissolved in chloroform (CF) or chloroform containing 0.3~0.7% 1-chloronaphthalene (CN), and the concentration of polymers were 6 mg mL<sup>-1</sup>. The blend films were formed by spin-coating atop PEDOT:PSS layer at 700 rpm for 20 s and annealed at 100 °C for 10 min. The film thicknesses of photoactive layers were maintained at about 100 nm. Then, 5 nm of PFN-Br was spin-coated from methanol:ethanol mixed solution as the cathode interface. The thicknesses of the organic films were measured by profilometry with a Tencor Alfa-Step 500. At last, 100 nm of argentum (Ag) layer (thicknesses monitored by an STM-100/MF-Sycon quartz crystal) was thermally deposited onto the organic layer through a shadow mask under a vacuum of  $1 \times 10^{-6}$  Pa. The effective area of the devices was 0.04 cm<sup>2</sup>.

The current density-voltage (J-V) characteristics were measured under a computer controlled Keithley 2400 sourcemeter under 1 sun, AM 1.5G solar simulator (Taiwan, Enlitech SS-F5). The EQE spectra was recorded by a Enlitech QE-R3011 EQE measurement system.

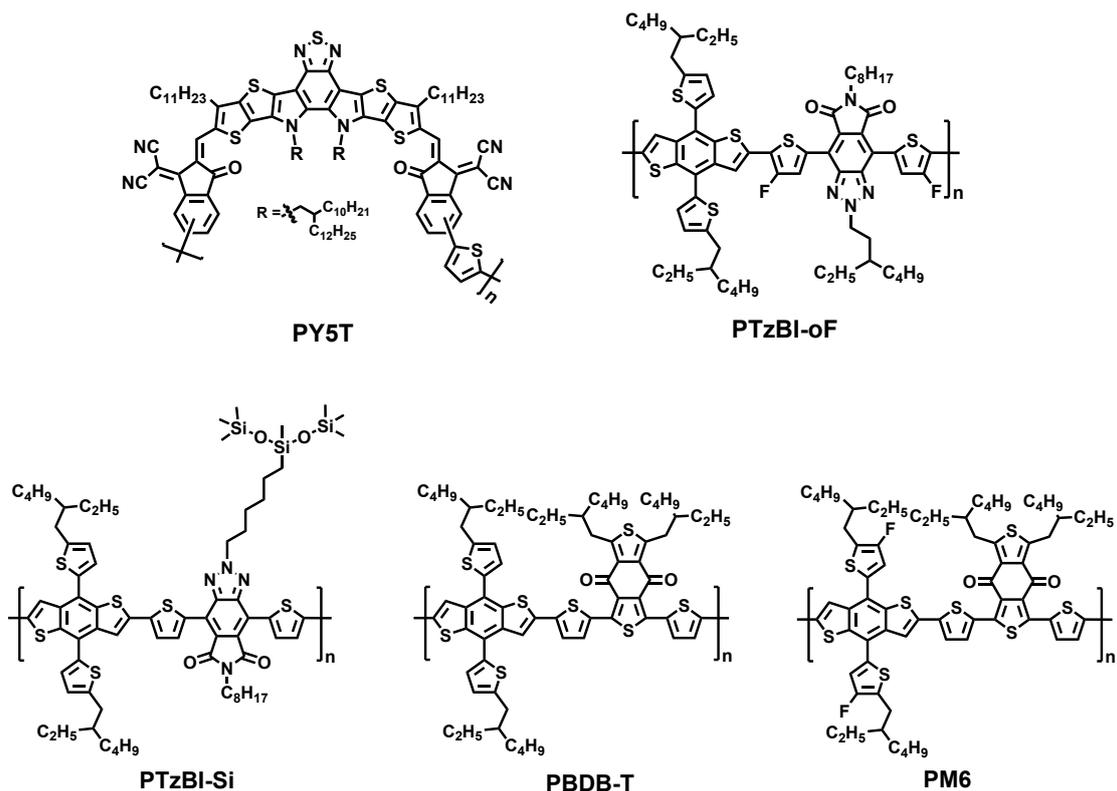
### Single-carrier devices fabrication and characterization

The hole-only and electron-only devices were fabricated with the structures of ITO/PEDOT:PSS/active layer/MoO<sub>3</sub>/Ag and ITO/ZnO/active layer/PFN-Br/Al, respectively. For the hole-only device, 10 nm of MoO<sub>3</sub> was vacuum-deposited on the active layer as the cathode. For the electron-only device, 40 nm of ZnO layer was spin-coated from the ZnO sol-gel. The other layers were prepared similar to the corresponding solar cells.

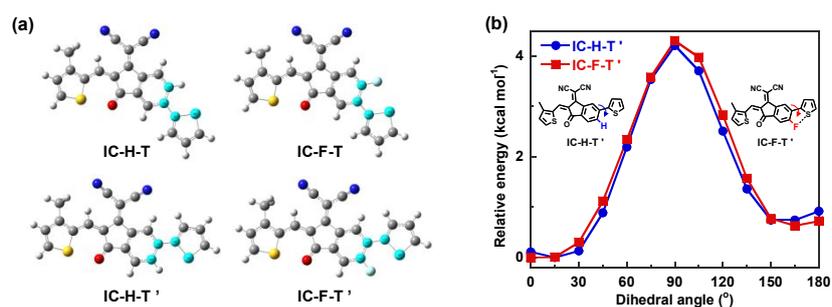
The mobility  $\mu$  was determined by fitting the dark current to the model of a single carrier SCLC, which is described by the following equation:

$$J = (9/8)\epsilon_0\epsilon_r\mu V^2/d^3,$$

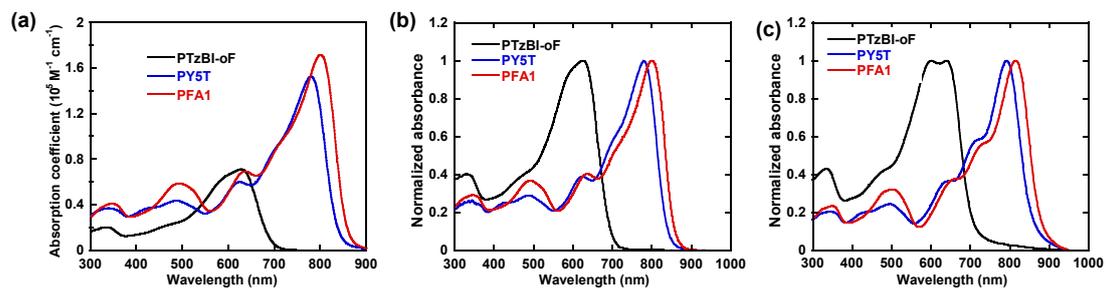
where  $J$  is the current density,  $\epsilon_0$  is the free-space permittivity,  $\epsilon_r$  is the relative permittivity of the material,  $V$  is the effective voltage  $V-V_{bi}$  and  $d$  is the thickness of the active layer. The charge (hole or electron) mobilities are calculated from the y-intercept of the  $J-V$  curves.



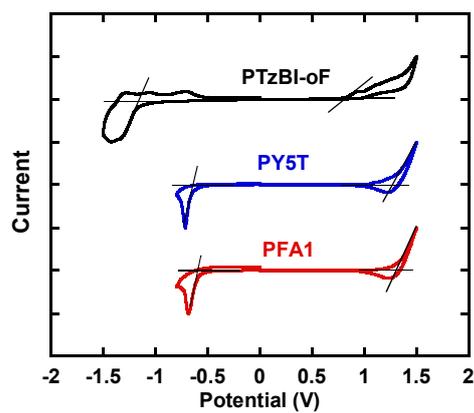
**Figure S1.** Chemical structures of PY5T, PTzBI-oF, PTzBI-Si, PBDB-T and PM6.



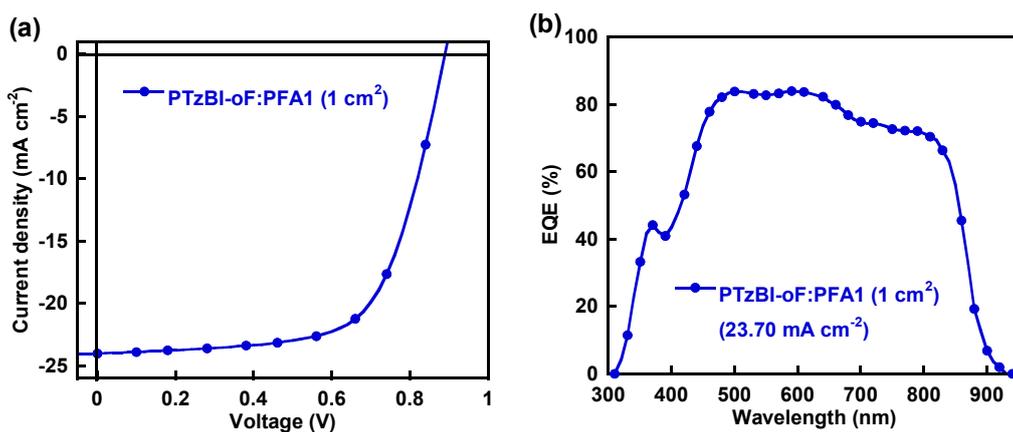
**Figure S2.** (a) Optimized geometry for model fragments of PY5T and PFA1. (b) PES scan of model compounds IC-H-T' and IC-F-T' for the other conformation of PY5T and PFA1, respectively.



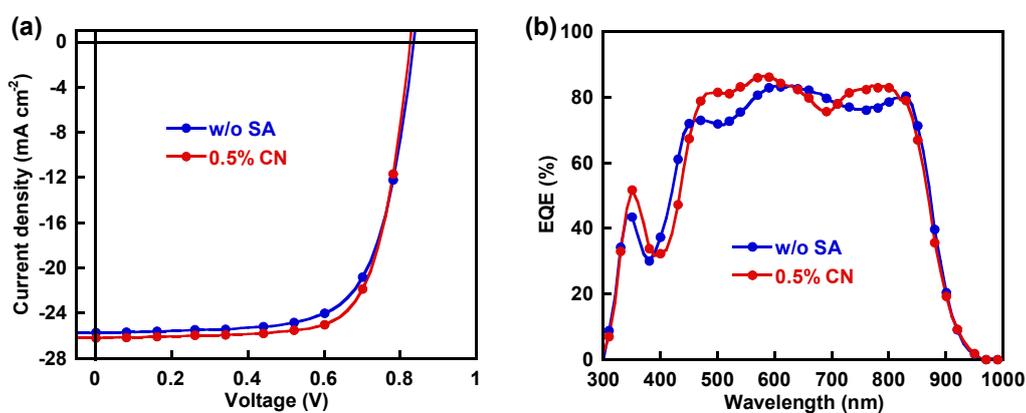
**Figure S3.** (a) Absorption coefficient of polymers in chloroform with concentration of  $1.0 \times 10^{-5} \text{ g mL}^{-1}$ . The normalized UV-vis absorption spectra of polymers (b) in dilute chloroform solution and (c) thin film.



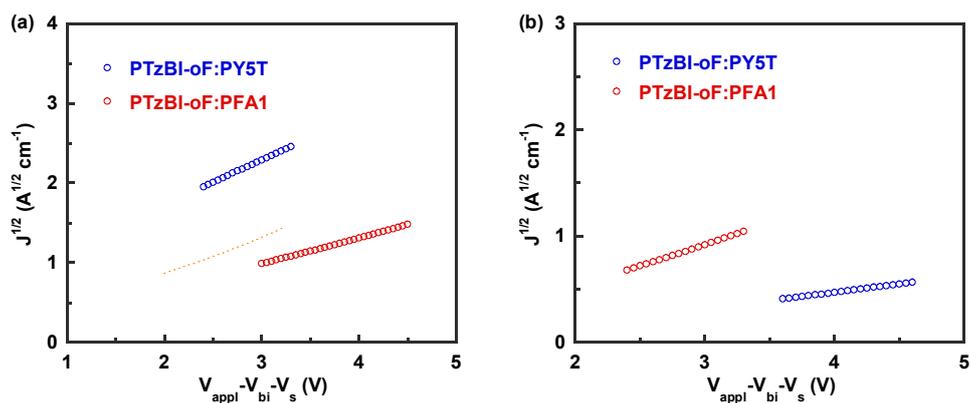
**Figure S4.** CV curves of polymers (versus saturated calomel electrode, SCE).



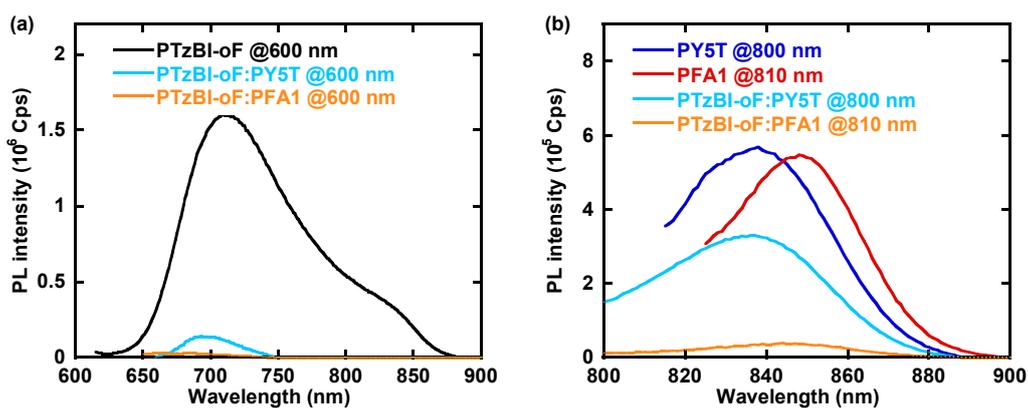
**Figure S5.** (a)  $J$ - $V$  characteristics and (b) EQE spectra of large area all-PSC based on PTzBI-oF:PFA1 with an effective area of  $1 \text{ cm}^2$ . The device structure is ITO/PEDOT:PSS/PTzBI-oF:Y5-F-Br/PFN-Br/Ag. Inset is the photo of  $1 \text{ cm}^2$  cell on an ITO layout.



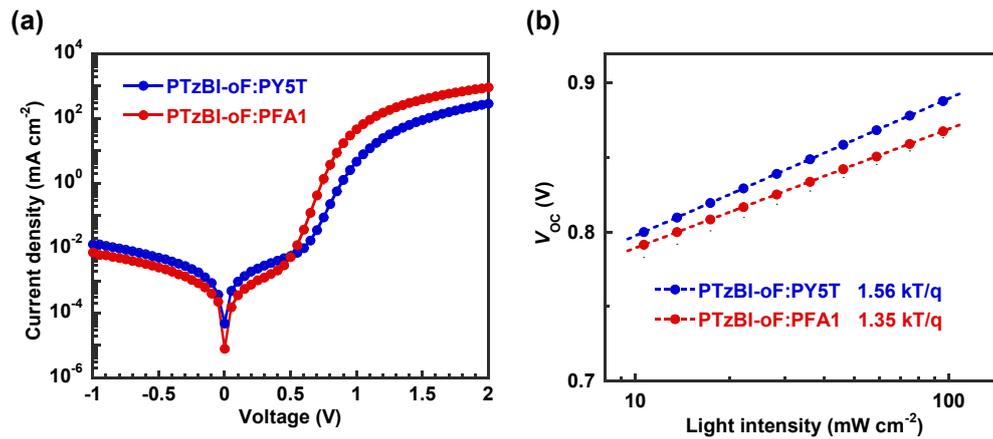
**Figure S6.** (a)  $J$ - $V$  characteristics and (b) EQE spectra of devices based on PTzBI-oF:Y5-F-Br with or without solvent additive. The device structure is ITO/PEDOT:PSS/PTzBI-oF:Y5-F-Br/PFN-Br/Ag. The effective area of the devices is  $0.04 \text{ cm}^2$ .



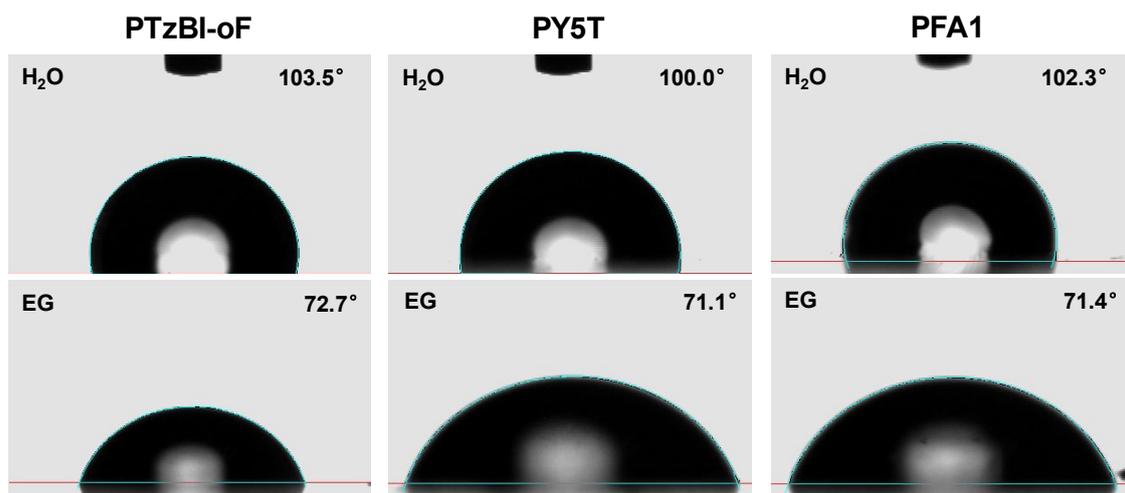
**Figure S7.** Dark  $J^{1/2}$ - $V$  characteristics for (a) hole-only and (b) electron-only devices based on blend films with the structure of ITO/PEDOT:PSS/active layer/ $M_0O_3$ /Ag and ITO/ZnO/active layer/PFN-Br/Al



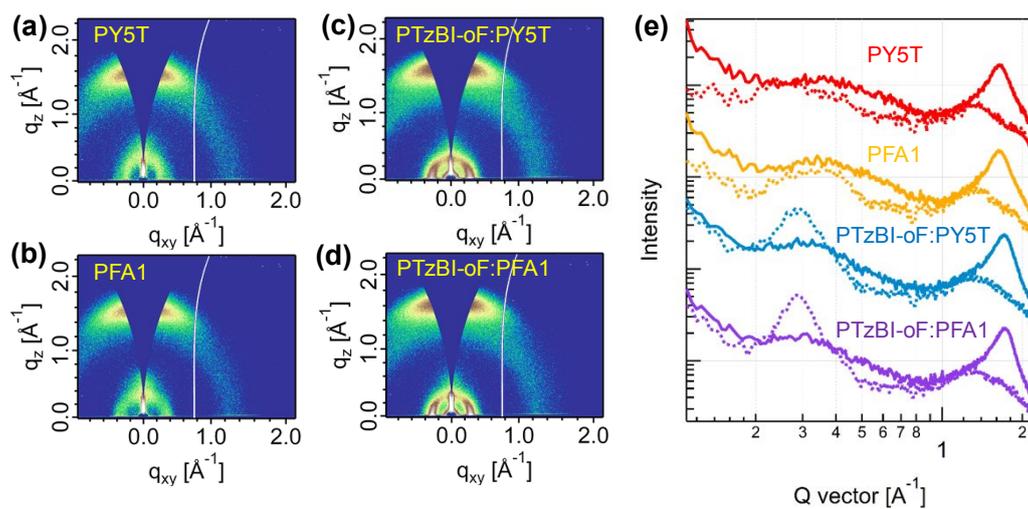
**Figure S8.** PL responses for polymer acceptors and corresponding blends excited at (a) 600 nm and (b) around 800 nm.



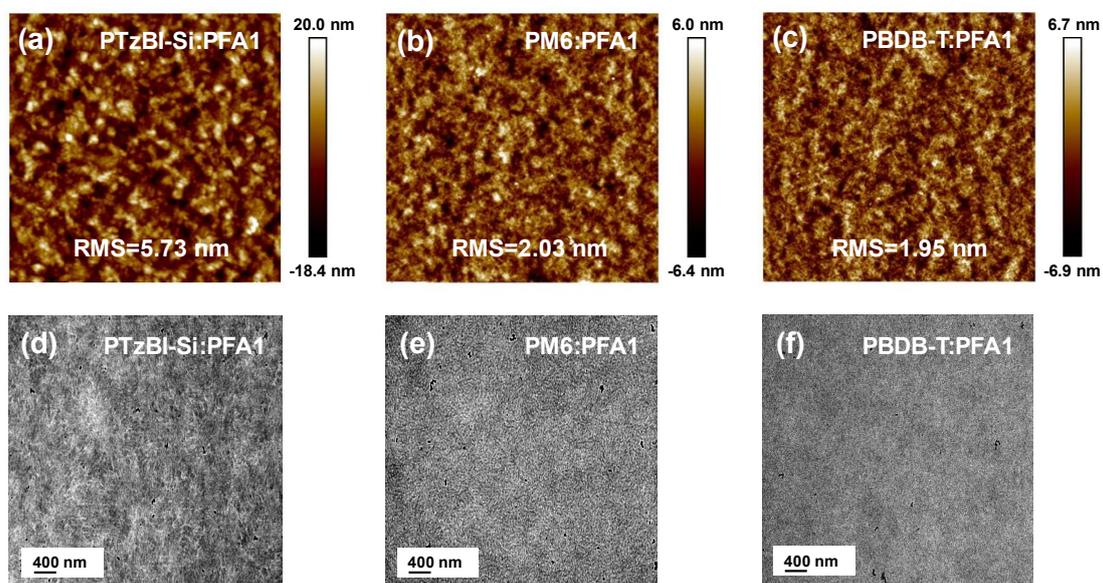
**Figure S9.** (a) Dark  $J$ - $V$  characteristics and (b)  $V_{OC}$  versus  $P_{light}$  characteristics for all-PSCs based on PTzBI-oF:PY5T and PTzBI-oF:PFA1.



**Figure S10.** Contact angle measurements of the pristine PTzBI-oF, PY5T and PFA1 films cast from chloroform solution. The measurements are carried out by using deionized water (H<sub>2</sub>O) and ethylene glycol (EG) as the wetting liquid.



**Figure S11.** (a-d) GIWAXS 2D images and (e) sector averaged curves in the OOP (solid lines) and IP (dotted lines) directions of pristine films and BHJ films.



**Figure S12.** (a, b, c) AFM height images (images size:  $5 \mu\text{m} \times 5 \mu\text{m}$ ) and (d, e, f) TEM images of the blend films.

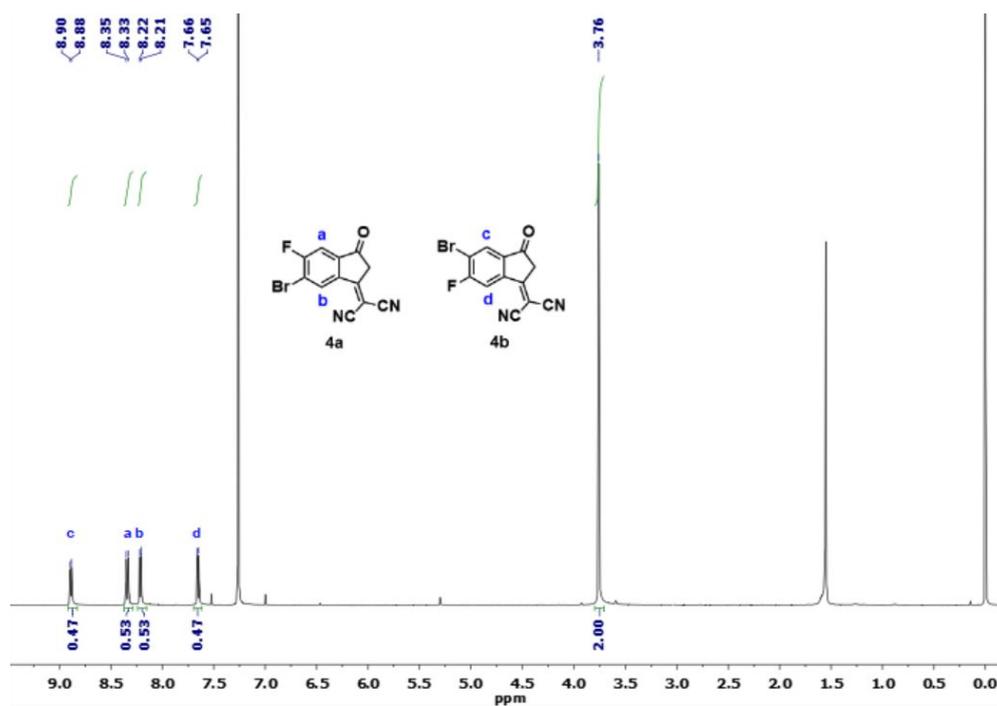


Figure S13.  $^1\text{H}$  NMR spectrum of 4a and 4b.

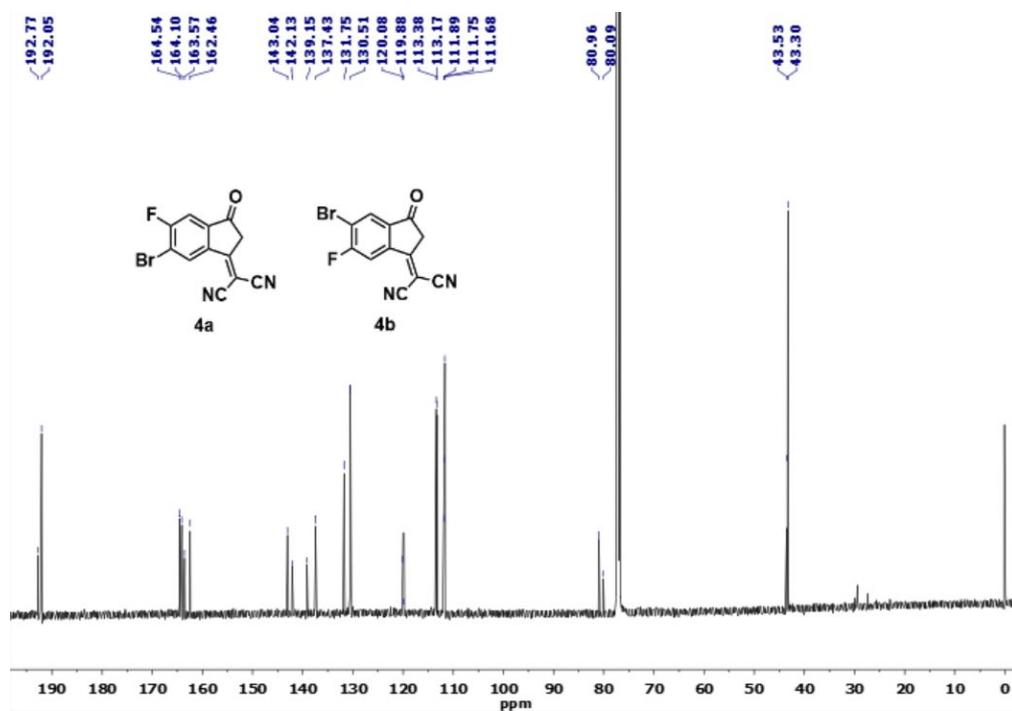


Figure S14.  $^{13}\text{C}$  NMR spectrum of 4a and 4b.

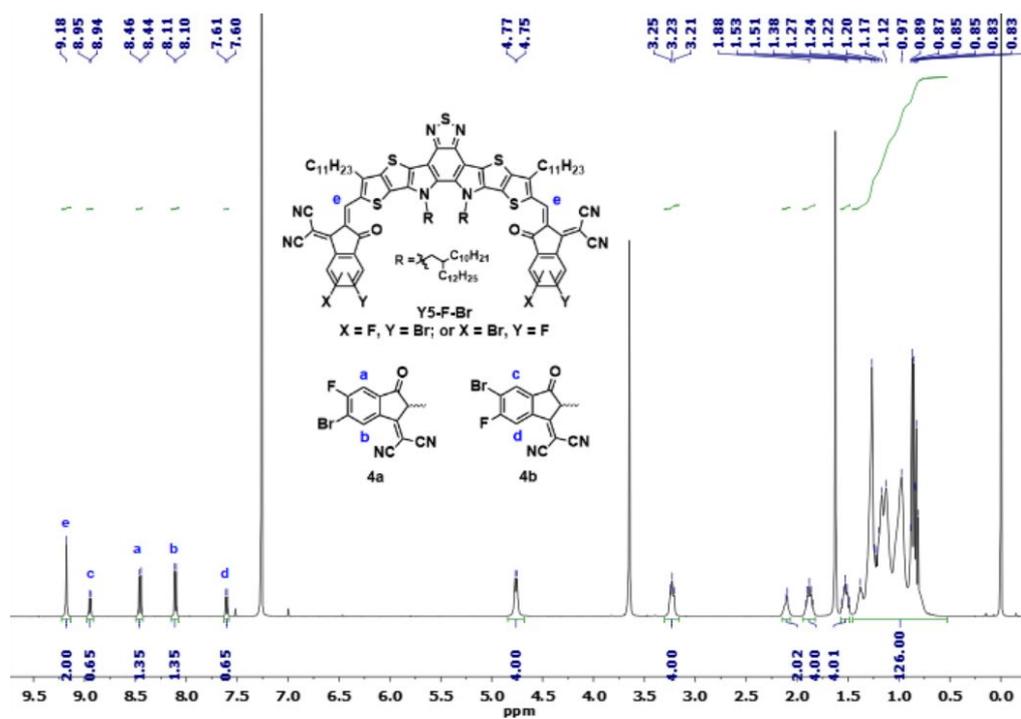


Figure S15. <sup>1</sup>H NMR spectrum of Y5-F-Br.

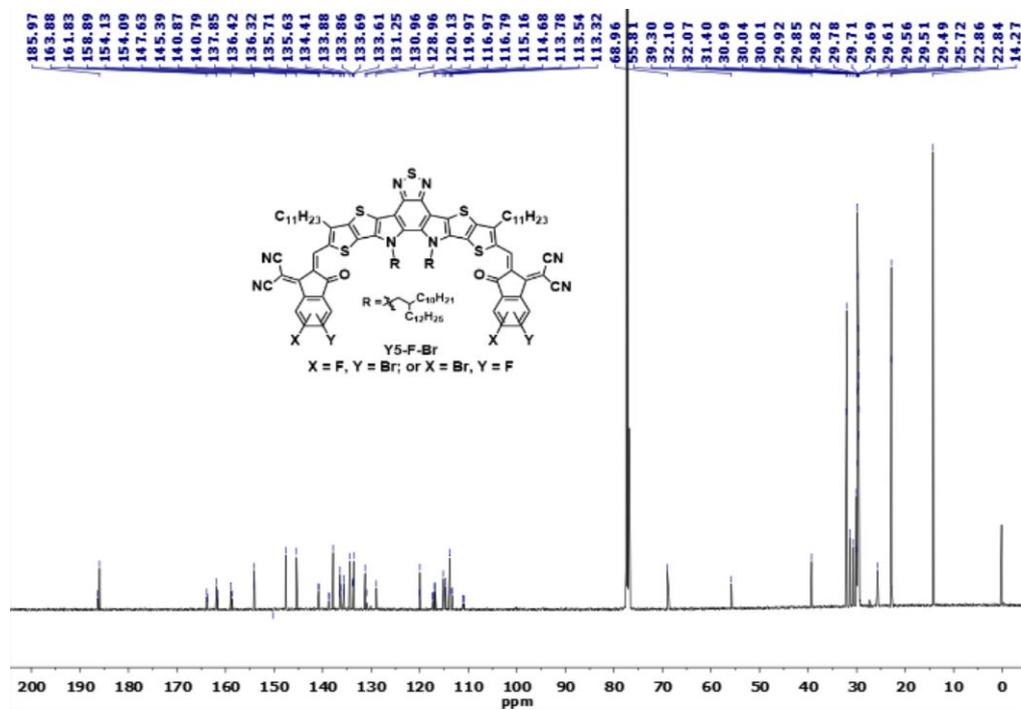
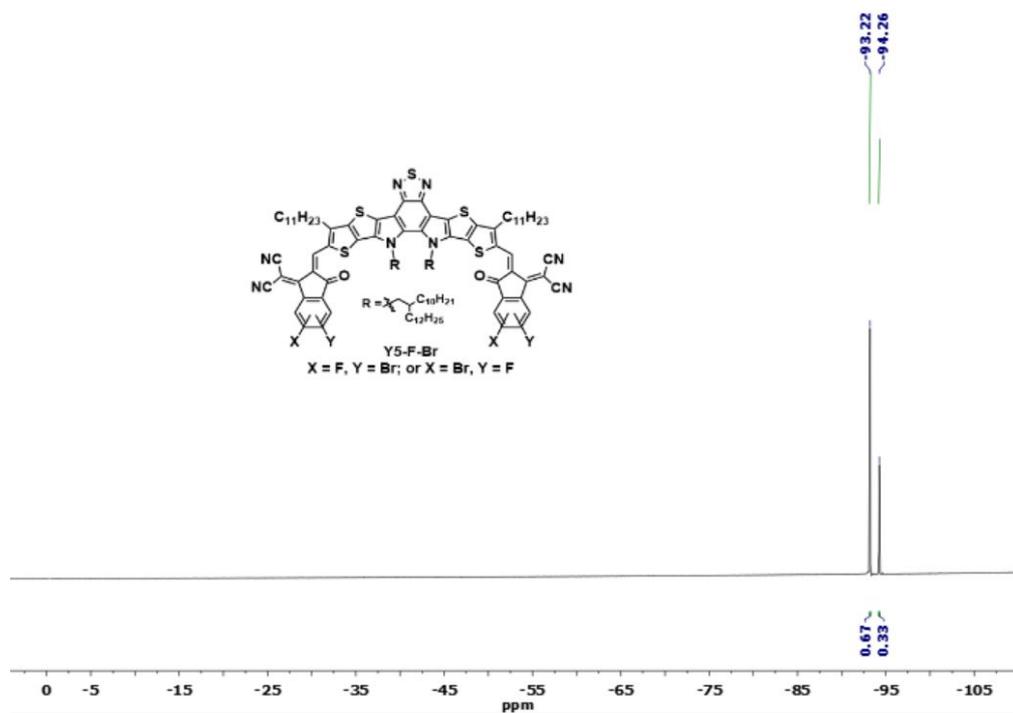


Figure S16. <sup>13</sup>C NMR spectrum of Y5-F-Br.



**Figure S17.**  $^{19}\text{F}$  NMR spectrum of Y5-F-Br.

**Table S1.** Optical and electrical properties of polymers.

Polymer	$\lambda_{\max}^a$ (nm)	$\epsilon_{\max}^a$ ( $\times 10^5 \text{ M}^{-1} \text{ cm}^{-1}$ )	$\lambda_{\max}^b$ (nm)	$\epsilon_{\max}^b$ ( $\times 10^4 \text{ cm}^{-1}$ )	$\lambda_{\text{onset}}^b$ (nm)	$E_g^c$ (eV)	HOMO <sup>d</sup> (eV)	LUMO <sup>d</sup> (eV)
PTzBI-oF	625	1.7	639	7.4	710	1.75	-5.25	-3.25
PY5T	778	1.5	792	8.2	852	1.46	-5.71	-3.76
PFA1	800	0.7	815	9.4	881	1.41	-5.74	-3.84

<sup>a</sup> Measured in dilute chloroform solution. <sup>b</sup> Measured in thin film spin-coated from chloroform solution. <sup>c</sup> Calculated by the equation of  $1240/\lambda_{\text{onset}}$ . <sup>d</sup> Estimated from cyclic voltammetry measurements.

**Table S2.** Photovoltaic parameters for PTzBI-oF:PY5T-based all-PSCs processed with/without solvent additive.

Blend	SA	$V_{\text{OC}}$ (V)	$J_{\text{SC}}$ ( $\text{mA cm}^{-2}$ )	FF (%)	PCE (%)
PTzBI-oF:PY5T	/	0.90	9.72	46.78	4.09
	0.5% CN	0.91	9.91	44.42	4.01

**Table S3.** Photovoltaic parameters for PTzBI-*o*F:PFA1-based all-PSCs processed with different solvent additives.

Blend	SA	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
PTzBI- <i>o</i> F:PFA1	/	0.87	23.53	64.76	13.23
	0.5% DIO	0.86	24.10	65.96	13.72
	0.5% DBE	0.87	24.60	63.05	13.43
	0.5% CN	0.87	23.96	72.67	15.11

**Table S4.** Photovoltaic parameters for PTzBI-*o*F:PFA1-based all-PSCs processed with different amount of chloronaphthalene.

Blend	SA	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
PTzBI- <i>o</i> F:PFA1	/	0.87	23.53	64.76	13.23
	0.3% CN	0.86	23.93	68.91	14.23
	0.5% CN	0.87	23.96	72.67	15.11
	0.7% CN	0.87	23.95	69.69	14.56

**Table S5.** Photovoltaic parameters for PTzBI-oF:Y5-F-Br-based PSCs.

Blend	SA	$V_{OC}$ (V)	$J_{SC}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	$J_{SC, EQE}$ (mA cm <sup>-2</sup> )
PTzBI-oF:PY5T	/	0.83	25.70	69.54	14.92	24.55
	0.5% CN	0.82	26.15	72.60	15.66	25.01

**Table S6.** Relative parameters obtained from  $J_{ph}-V_{eff}$ ,  $V_{OC}-P_{light}$ ,  $J_{SC}-P_{light}$  and Dark  $J-V$  curves.

Acceptor	$R_s$ ( $\Omega$ cm <sup>2</sup> )	$R_{sh}$ ( $\Omega$ cm <sup>2</sup> )	n ( $V_{OC}-P_{light}$ )	$\alpha$ ( $J_{SC}-P_{light}$ )	$J_{sc}$ (mA cm <sup>-2</sup> )	$J_{sat}$ (mA cm <sup>-2</sup> )	P (E,T) (%)
PY5T	2.18	$1.12 \times 10^5$	1.56 kT/q	0.85	9.23	14.66	63.01
PFA1	0.90	$2.63 \times 10^5$	1.35 kT/q	0.98	24.12	25.24	95.59

**Table S7.** The contact angles and surface free energy parameters of the PTzBI-oF, PY5T and PFA1 pristine films.

Film	Contact Angle (°)		$\gamma_l^a$ (mJ m <sup>-2</sup> )	$\chi_{donor-acceptor}^b$ (K)
	H <sub>2</sub> O	EG		
PTzBI-oF	103.5	72.7	38.62	NaN
PY5T	100	71.1	34.13	1.38×10 <sup>-1</sup>
PFA1	102.3	71.4	38.78	1.65×10 <sup>-4</sup>

<sup>a</sup>  $\gamma$  represents the surface energy of PTzBI-oF, PY6T and PFA1 pristine films. <sup>b</sup> Flory-Huggins interaction parameter  $\chi_{donor-acceptor}$  estimated from the equation of  $\chi_{donor-acceptor} = K (\sqrt{\gamma_{donor}} - \sqrt{\gamma_{acceptor}})^2$ ,  $K$  is a constant.

**Table S8.** Peak fitting parameters of GIWAXS line-cut profiles.

Sample	peak	Type	Location	Amplitude	Area	FWHM
			[Å <sup>-1</sup> ]	[a.u.]	[a.u.]	[Å <sup>-1</sup> ]
PY5T	0	Gauss	0.25	4.42	0.44	0.094
	1	Gauss	0.36	2.50	0.47	0.175
PFA1	0	Gauss	0.25	2.74	0.26	0.089
	1	Gauss	0.36	7.25	1.27	0.165
PTzBI-oF:PY5T	0	Gauss	0.29	29.75	3.03	0.096
PTzBI-oF:PFA1	0	Gauss	0.29	37.04	3.04	0.077

**Table S9.** Photovoltaic parameters for all-PSCs based on PFA1 and three commercial polymer donors.

Blend	Solvent additive	Annealing	$V_{OC}$ (V)	$J_{SC}$ (mA cm <sup>-2</sup> )	$FF$ (%)	PCE (%)
PTzBI-Si:PFA1	2%DBE	100 °C/10min	0.84	21.29	66.88	11.94
PBDB-T:PFA1	1%CN	100 °C/10min	0.83	24.26	70.18	14.20
PM6:PFA1	0.5%CN	100 °C/10min	0.90	22.41	68.10	13.75

## References

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