## Supporting Information

# Regulating the Optoelectronic Properties of Small Molecular Donors with Multiple Alternative Electron-Donor and Acceptor Units for Organic Solar Cells

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## Synthesis and characterization

## Materials:

All reagents were used as received from commercial sources. All reagents were weighed and handled in air, and refilled with an inert atmosphere of  $N_2$  at room temperature. DMF and DMSO were distilled under reduced pressure from CaH<sub>2</sub>. The DMSO was stored with the powder of 4 Å molecular sieves. Toluene and 1,4-Dioxane was distilled from sodium and benzophenone immediately before use. Compound 2,5-bis(2-ethylhexyl)-3,6-di(thiophen-2-yl)-2,5-dihydropyrrolo[3,4-*c*]pyrrole-1,4-dione (1), 4-bromo-5,6-difluoro-2-hexyl-2*H*-benzo[*d*][1,2,3]triazole (2), and 4-decyl-2,7-bis(tributylstannyl)dithieno[3,2-*b*:2',3'-*d*]pyridin-5(4*H*)-one (4) was synthesized according to the reported literature. Compound 2,5-bis(trimethylstannyl)thiophene (5), (4,4,9,9-tetrakis(4-hexylphenyl)-4,9-dihydro-*s*-

indaceno[1,2-*b*:5,6-*b*']dithiophene-2,7-diyl)bis(trimethylstannane) (**6**), and **7** were purchased from Derthon Optoelectronic Materials Science Technology.

#### Synthesis of 3

To a 25 mL of Schlenk tube were added compound 1 (0.41 g, 0.78 mmol), 2 (1.0g, 3.13 mmol), Pd(TFA)<sub>2</sub> (13 mg, 0.039 mmol) and Ag<sub>2</sub>O (0.73 mg, 3.13 mmol) under N<sub>2</sub>, followed by DMSO (10 mL) with stirring. The reaction mixture was stirred at 80 °C (preheated oil bath) for 5 h. Then the reaction mixture was cooled to room temperature, filtered and diluted with dichloromethane, washed with brine, dried over Na<sub>2</sub>SO<sub>4</sub>, filtered and concentrated. The product was purified with silica gel chromatography as a purple solid. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 9.08 (dd, J = 4.3, 1.9 Hz, 1H), 8.89 (dd, J = 3.9, 1.1 Hz, 1H), 8.40-8.38 (m, 1H), 7.57 (dd, J = 5.0, 1.1 Hz, 1H), 7.24-7.19 (m, 1H), 4.73-4.69 (m, 2H), 4.07-3.95 (m, 4H), 2.13-1.80 (m, 2H), 1.93-1.77 (m, 2H), 1.54 (s, 2H), 1.35-1.19 (m, 2OH), 0.88-0.76 (m, 15H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 161.72, 161.68, 140.70, 139.85, 139.80, 136.81, 136.72, 135.93, 135.69, 131.93, 131.84, 131.61, 131.54, 130.88, 129.81, 128.55, 110.75, 110.62, 108.86, 108.16, 57.44, 46.11, 45.91, 39.38, 39.10, 31.18, 30.22, 30.19, 30.01, 28.41, 28.34, 26.21, 23.61, 23.49, 23.16, 23.11, 23.06, 22.45, 14.08, 14.07, 14.00, 10.53, 10.47. MS (EI) m/z: 839 (M<sup>+</sup>); Anal. Calcd for C<sub>42</sub>H<sub>52</sub>BrF<sub>2</sub>N<sub>5</sub>O<sub>2</sub>S<sub>2</sub> : C, 59.99; H, 6.23; N, 8.33. Found: C, 59.50; H, 5.98; N, 8.60.

### Synthesis of BFD

In a 50 mL round bottom flask, a mixture of compound **3** (0.5 g, 0.60 mmol), compound **4** (0.27 g, 0.24 mmol),  $Pd_2(dba)_3$  (15 mg) and  $P(o-tol)_3$  (10 mg) were added and toluene (20 mL) were added into the mixture under N<sub>2</sub>. The reaction mixture was stirring at 110 °C for 48 h. Then the reaction was cooled to room temperature. Distilled water (50 mL) was added and the mixture was extracted with  $CH_2Cl_2$  for several times. The combined organci phases were dried with anhydrous Na<sub>2</sub>SO<sub>4</sub> and concentrated. The crude product was subjected column chromatography on silica gel chromagraphy with PE:CH<sub>2</sub>Cl<sub>2</sub> (v/v: 1:1) and obtained blue

solid. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 8.95 (m, 4H), 8.45 (br, 1H), 8.15 (br, 3H), 7.36 (s, 2H), 7.06 (s, 2H), 4.88 (s, 4H), 4.38 (br, 2H), 4.01 (br, 4H), 3.81 (br, 4H), 2.31 (d, J = 6.7 Hz, 4H), 2.10-1.75 (m, 6H), 1.68-1.19 (m, 59H), 1.10-0.82 (m, 32H). <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 160.99, 160.46, 157.16, 147.90, 139.07, 136.55, 135.61, 134.91, 132.69, 131.09, 130.00, 129.54, 128.05, 127.52, 109.48, 107.61, 100.00, 57.21, 46.02, 45.61, 39.46, 39.03, 32.05, 31.57, 31.53, 30.03, 29.84, 29.70, 29.56, 28.34, 28.22, 27.56, 26.65, 26.61, 23.47, 23.19, 22.80, 14.29, 14.21, 14.13, 10.48, 10.31. MS (MALDI-TOF) m/z: 1865.83; Found: 1865.85 (M<sup>+</sup>) Anal. Calcd for C<sub>103</sub>H<sub>127</sub>F<sub>4</sub>N<sub>11</sub>O<sub>5</sub>S<sub>6</sub> : C, 66.24; H, 6.85; N, 8.25. Found: C, 66.32; H, 7.01; N, 8.60.

## Synthesis of TFD

Target molecule TFD was synthesized using the same method with the synthesis of BFD by replacing compound **4** with compound **5**. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 9.13 (s, 2H), 8.94 (d, *J* = 3.1 Hz, 2H), 8.43-8.16 (m, 4H), 7.58-7.40 (m, 2H), 7.21-7.11 (m, 2H), 4.95-4.72 (m, 4H), 4.22-4.01 (m, 4H), 4.00-3.76 (m, 4H), 2.43-2.21 (m, 4H), 2.08-1.97 (m, 2H), 1.86 (s, 2H), 1.53-1.21 (m, 42H), 1.11-0.75 (m, 32H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 161.24, 160.74, 148.51, 146.12, 139.66, 139.07, 137.44, 136.95, 136.45, 135.44, 134.75, 130.92, 130.55, 130.13, 129.69, 128.19, 110.50, 108.69, 107.78, 56.97, 46.04, 45.64, 39.46, 39.04, 31.47, 30.11, 29.86, 28.37, 28.27, 26.55, 23.51, 23.41, 23.32, 23.17, 22.68, 14.24, 14.17, 14.10, 10.49, 10.37. MS (MALDI-TOF) m/z: 1602.69; Found: 1603.33 (M<sup>+</sup>) Anal. Calcd for C<sub>88</sub>H<sub>106</sub>F<sub>4</sub>N<sub>10</sub>O<sub>4</sub>S<sub>5</sub> : C, 65.89; H, 6.66; N, 8.73. Found: C, 65.55; H, 6.20; N, 8.56.

## Synthesis of IDTFD

Target molecule IDTFD was synthesized using the same method with the synthesis of BFD by replacing compound **4** with compound **6**. <sup>1</sup>H NMR (400 MHz, CD<sub>2</sub>Cl<sub>2</sub>)  $\delta$  (ppm) 9.08 (d, *J* = 2.7 Hz, 2H), 8.82 (d, *J* = 3.0 Hz, 2H), 8.42 (d, *J* = 4.3 Hz, 2H), 8.27 (s, 2H), 7.59 (d, *J* = 5.0 Hz, 2H), 7.55 (s, 2H), 7.21 (m, 10H), 7.07 (m, 8H), 4.74 (t, *J* = 7.2 Hz, 4H), 4.15-3.87 (m, 8H), 2.55-2.47 (m, 8H), 2.10 (m, 6H), 1.99-1.86 (m, 6H), 1.78 (br, 3H), 1.29-1.15 (m, 75H),

0.82-0.74 (m, 36H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 161.65, 156.55, 154.08, 145.15, 141.77, 141.73, 140.14, 137.51, 137.43, 136.41, 135.61, 134.99, 130.99, 128.48, 128.03, 126.59, 126.03, 118.10, 114.85, 108.51, 106.86, 63.12, 57.10, 46.14, 45.86, 39.39, 39.11, 35.65, 31.76, 31.43, 31.28, 30.27, 30.19, 29.92, 29.23, 28.45, 28.35, 26.29, 23.63, 23.49, 23.21, 23.14, 22.64, 22.53, 14.15, 14.11, 14.09, 10.56, 10.49. MS(MALDI-TOF) m/z: 2425.21; Found: 2425.04 (M<sup>+</sup>) Anal. Calcd for C<sub>148</sub>H<sub>176</sub>F<sub>4</sub>N<sub>10</sub>O<sub>4</sub>S<sub>6</sub> : C, 73.23; H, 7.31; N, 5.77. Found: C, 72.92; H, 7.28; N, 5.75.

### Synthesis of IDTTFD

Target molecule IDTTFD was synthesized using the same method with the synthesis of BFD by replacing compound **4** with compound **7**. <sup>1</sup>H NMR (400 MHz, CD<sub>2</sub>Cl<sub>2</sub>)  $\delta$  (ppm) 9.19 (s, 2H), 8.93 (s, 2H), 8.78 (s, 2H), 8.54 (s, 2H), 7.67 (s, 4H), 7.31 (m, 10H), 7.20 (m, 8H), 4.88 (t, *J* = 7.2 Hz, 4H), 4.44-3.85 (m, 8H), 2.73-2.50 (m, 14H), 2.25 (dd, *J* = 13.5, 6.3 Hz, 6H), 2.05 (m, 4H), 1.90 (br, 2H), 1.35-1.25 (m, 72H), 0.91-0.87 (m, 36H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  (ppm) 161.57, 153.95, 146.68, 146.46, 145.40, 142.47, 142.02, 140.09, 139.97, 137.70, 137.52, 137.45, 137.37, 136.43, 136.31, 135.63, 133.37, 131.24, 131.08, 130.61, 129.77, 128.65, 128.39, 128.16, 123.99, 117.32, 111.90, 111.77, 108.80, 108.67, 108.43, 108.03, 63.07, 57.13, 46.12, 45.87, 39.39, 39.11, 35.69, 31.75, 31.37, 31.28, 30.27, 30.18, 29.98, 29.27, 28.47, 28.36, 26.33, 23.63, 23.49, 23.22, 23.15, 22.64, 22.56, 14.14, 14.12, 14.09, 10.55, 10.49. MS(MALDI-TOF) m/z: 2538.16; Found: 2538.39 (M<sup>+</sup>) Anal. Calcd for C<sub>152</sub>H<sub>176</sub>F<sub>4</sub>N<sub>10</sub>O<sub>4</sub>S<sub>8</sub> : C, 71.89; H, 6.99; N, 5.52. Found: C, 72.22; H, 7.16; N, 5.47.

**General measurement and characterization:** Nuclear magnetic resonance (NMR) spectra were collected on a Bruker AV-500 (400 MHz) in deuterated chloroform with tetramethylsilane as a reference. MALDI-TOF mass spectrometric measurements were carried out on Bruker Bifiex III MALDI TOF. Elemental analyses were performed with a Harrios elemental analysis instrument. Thermo-gravimetric analysis (TGA) were analyzed on a TA

Instrument Model SDT Q600 simultaneous TGA/DSC analyzer with a heating rate of 10 °C min<sup>-1</sup> and a N<sub>2</sub> flow rate of 90 mL min<sup>-1</sup>. UV-vis absorption spectra were obtained using a Shimadzu UV-2700 UV-vis-NIR spectrometer. Cyclic voltammetry (CV) data were collected on a CHI630E electrochemical workstation equipped with a saturated calomel electrode as the reference electrode, a platinum wire counter electrode, and a glassy carbon working electrode. The carried measurements were with anhydrous acetonitrile out tetrabutylammoniumhexafluorophosphate (0.1 M) as the supporting electrolyte at a scan rate of 100 mV s<sup>-1</sup> under an argon atmosphere. Potentials were referenced to the ferrocene/ferrocenium couple by using ferrocene as the standard. The oxidation potential of ferrocene was set at -4.80 eV with respect to zero vacuum level. The HOMO and LUMO energy levels were calculated according to the equation of  $E_{\text{HOMO}} = -[4.8 + (E_{\text{ox}} - E_{\text{Fc/Fc+}})]$ (eV) and  $E_{LUMO}$  = - [4.8 + ( $E_{red}$  -  $E_{Fc/Fc+}$ )] (eV). X-ray diffraction (XRD) patterns of the materials were recorded on a Rigaku X-ray diffractometer operated in reflection geometry at 30 mA, 40 kV with Cu Ka radiation. Atomic force microscopy (AFM) images were obtained by using Oxford atomic microscope at tapping mode. TEM specimens were prepared following identical conditions as the actual devices, but were drop-casted onto 40 nm PEDOT:PSS covered substrate. After drying, the substrates were transferred to deionized water and the floated films were transferred to TEM grids. TEM images were obtained on JEM-2100(HR) (200 kV) TEM. GIXD measurements were performed at the MetalJet-D2 Excillum source in a Xenocs-SAXS/WAXS system with a X-ray wavelength of 1.34144 Å and the samples were treated at a fixed angle of 0.2°. 2D detector (Pilatus3R 1M from Dectris) was used with a framing rate of 10 Hz to record the scattering pattern. The samples were prepared on silicon wafer substrates with the same experimental conditions as those for solar cells. Space charge limited current (SCLC) is described by  $J = 9\varepsilon_0\varepsilon_{\rm T}\mu V^2/8L^3$ , where J is the current density,  $\varepsilon_r$  is the relative dielectric constant of the transport medium,  $\varepsilon_0$  is the permittivity of free space (8.85 × 10<sup>-12</sup> F m<sup>-1</sup>),  $\mu$  is the hole or electron mobility, L is the film

thickness of active layer, V is the internal voltage in the device and  $V = V_{appl} - V_{bi} - V_{a}$ , where  $V_{appl}$  is the applied voltage to the device,  $V_{bi}$  is the built-in voltage due to the relative work function difference of the two electrodes and  $V_{a}$  is the voltage drop due to contact resistance and series resistance across the electrodes.

**Device fabrication and characterization:** The device structure is ITO/ZnO/active layer/MoO<sub>3</sub>/Ag. ITO(coated glass with a sheet resistance of  $< 15 \ \Omega sq^{-1}$ ) was used as the substrate, which was cleaned by sequential sonication in H<sub>2</sub>O containing detergent, deionized H<sub>2</sub>O, acetone, and isopropanol followed by UV/ozone (BZS250GF-TC, HWOTECH, Shenzhen) treatment for 15 min. The ZnO cathode buffer layer (20 nm) was spin-coated on an ITO substrate at 3000 rpm for 30s and annealed at 250 °C for 30 minutes. The substrates were then transferred to a nitrogen filled glove box for spin casting the active layer. Finally, a bilayer anode structure of MoO<sub>3</sub> layer (10 nm)/Ag (100 nm) was deposited atop the active layer by thermal evaporation in a vacuum of  $2 \times 10^{-6}$  Torr.

The thicknesses of the active layers were measured using Vecco Dektak 150 surface profiler meter. The active areas of the devices were 0.045 cm<sup>2</sup> defined by shadow mask. The current density-voltage (*J-V*) characteristics were measured with Keithley 2400 source measurement unit under AM 1.5G (100 mW cm<sup>-2</sup>) irradiation from Newport solar simulator. Light intensity was calibrated with a standard silicon solar cell. External quantum efficiencies (*EQE*) of solar cells were analyzed by certified Newport incident photon conversion efficiency (*IPCE*) measurement system.



Fig. S1. TGA curves of BFD, TFD, IDTFD and IDTTFD.



Fig. S2. Cyclic voltammograms of BFD, TFD, IDTFD and IDTTFD.



**Fig. S3**. AFM height (a, b, c, d), and phase (e, f, g, h) images for BFD:PC<sub>61</sub>BM (a, e), TFD:PC<sub>61</sub>BM (b, f), IDTFD:PC<sub>61</sub>BM (c, g) and IDTTFD:PC<sub>61</sub>BM (d, h) blended films.



**Fig. S4.** GIXD profiles of neat films BFD (a), TFD (b), IDTFD (c), IDTTFD (d) and blended films BFD:PC<sub>61</sub>BM (e), TFD:PC<sub>61</sub>BM (f), IDTFD:PC<sub>61</sub>BM (g) and IDTTFD:PC<sub>61</sub>BM (h) BFD, TFD, IDTFD, IDTFD with 5% CN as additives.



Fig. S5. The in-plane (a) and out-of-plane (b) line-cuts of GIXD.

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Compound	$ heta_1$	$ heta_2$	$ heta_3$	$ heta_4$	$ heta_5$	$ heta_6$	$ heta_7$	$\theta_8$
BFD	10.0°	6.04°	0.10°	0.58°	0.80°	0.30°	2.56°	0.64°
TFD	6.16°	4.75°	0.04°	0.29°	0.30°	0.30°	4.65°	6.14°
IDTFD	6.86°	2.64°	0.32°	0.27°	0.81°	0.59°	2.59°	5.72°
IDTTFD	6.76°	3.10°	0.66°	0.36°	0.08°	0.62°	2.79°	6.30°

**Table S1**. The dihedral angles of the four small molecules between donor units and acceptor units.

**Table S2**. Photovoltaic performances of OSCs based on donor:  $PC_{71}BM$  under theillumination of AM 1.5G, 100 mW cm<sup>-2</sup>.

Donor:PC <sub>71</sub> BM	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-</sup> <sup>2</sup> )	FF (%)	PCE <sub>max</sub> (%)	PCE <sub>ave</sub> (%)
BFD	0.57	1.51	58	0.37	0.28
BFD <sup>a</sup>	0.59	1.08	55	0.35	0.29
TFD	0.55	1.30	58	0.41	0.35
TFD <sup>a</sup>	0.51	1.33	48	0.32	0.29
IDTFD	0.65	2.21	48	0.69	0.62
<b>IDTFD</b> <sup>a</sup>	0.52	2.21	37	0.43	0.37
IDTTFD	0.84	1.51	48	0.57	0.51
<b>IDTTFD</b> <sup>a</sup>	0.84	1.93	33	0.53	0.48

a ; thermal annealing at 100 °C for ten minutes.

Donor:PC <sub>71</sub> BM	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-</sup> <sup>2</sup> )	FF (%)	PCE <sub>max</sub> (%)	PCE <sub>ave</sub> (%)
BFD <sup>a</sup>	0.71	3.85	47	1.29	1.23
BFD <sup>a,b</sup>	0.76	3.00	46	1.07	1.02
TFD <sup>a</sup>	0.76	8.89	47	3.20	3.14
TFD <sup>a,b</sup>	0.77	9.31	46	3.36	3.29
<b>IDTFD</b> <sup>a</sup>	0.81	9.26	50	3.84	3.75
IDTFD <sup>a,b</sup>	0.81	8.63	53	3.74	3.69
<b>IDTTFD</b> <sup>a</sup>	0.83	11.93	54	5.33	5.25
IDTTFD <sup>a,b</sup>	0.85	10.96	54	5.05	4.99

**Table S3**. Photovoltaic performances of OSCs based on donor:  $PC_{71}BM$  under the illumination of AM 1.5G, 100 mW cm<sup>-2</sup>.

a: 3% DIO. b: solvent annealing with chloroform for 30s.

Table S4. Photovoltaic performances of OSCs based on donor: PC71BM with different	j
additives under the illumination of AM 1.5G, 100 mW cm <sup>-2</sup> .	

D:A	additive	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE <sub>max</sub> (%)	PCE <sub>ave</sub> (%)
IDTTFD:PC <sub>71</sub> BM	1% DIO	0.83	8.33	53	3.69	3.62
IDTTFD:PC <sub>71</sub> BM <sup>a</sup>	1% DIO	0.84	8.42	51	3.63	3.58
IDTTFD:PC71BM	3% DIO	0.83	11.93	54	5.33	5.25
IDTTFD:PC71BM	1% DPE	0.83	4.98	50	2.06	2.00
IDTTFD:PC <sub>71</sub> BM <sup>a</sup>	1% DPE	0.81	4.85	52	2.03	1.97
IDTTFD:PC71BM	3% DPE	0.81	8.98	56	4.06	3.98

a: solvent annealing with dichloromethane for 30s.

Donor	additive	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-</sup> <sup>2</sup> )	FF (%)	PCE <sub>max</sub> (%)	PCE <sub>ave</sub> (%)
IDTTFD	3% DIO	0.85	11.71	55	5.49	5.43
IDTTFD	4% DIO	0.86	10.90	57	5.40	5.30
IDTTFD	5% DIO	0.87	11.11	55	5.39	5.32
IDTTFD	5% DPE	0.86	14.08	58	7.06	7.01
IDTFD	3% DIO	0.86	10.01	52	4.45	4.38
IDTFD	3% DPE	0.85	11.83	53	5.34	5.29

**Table S5**. Photovoltaic performances of OSCs based on donor:  $PC_{61}BM$  with different additives under the illumination of AM 1.5G, 100 mW cm<sup>-2</sup>.

**Table S6**. Photovoltaic performances of OSCs based on donor:  $PC_{61}BM$  with different D:A ratios under the illumination of AM 1.5G, 100 mW cm<sup>-2</sup>.

Donor	D:A ratio	additive	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE <sub>max</sub> (%)	PCE <sub>ave</sub> (%)
IDTTFD	1:1.2	5% DPE	0.85	13.62	56	6.48	6.42
IDTTFD	1:1.2	5%DPE	0.84	13.23	50	5.59	5.51
IDTTFD	1:1.8	5% DPE	0.84	13.46	53	6.06	5.99
IDTTFD	1:1.8	5% DPE	0.85	12.36	54	5.67	5.62
IDTFD	1:1.5.	1% CN	0.85	10.28	50	4.35	4.29
IDTFD	1:1.5	2% CN	0.88	3.91	55	1.87	1.82
IDTTFD	1:1.5	1% CN	0.82	9.77	50	4.01	3.95
IDTTFD	1:1.5	2%CN	0.84	11.57	54	5.22	5.14