Supporting Information

Mechanochemical synthesis of nonfullerene small molecular acceptors

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1. Experimental Section

1.1 Materials and Characterization

All chemicals and reagents were purchased from commercial sources and used without further purification unless otherwise noted. 2,7-dibromo-9,9-dihexyl-9H-fluorene^[1] and **B1**^[2], **A2**^[3], **A3**^[4], ethyl 4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzoate^[5] and 1,4-dibromo-2,5-bis((2-hexyldecyl)oxy)benzene^[6], **B7**^[7] were synthesized according to the published procedures. PBDB-T and PM6 were purchased from Fuan optoelectronic, PNDIT-F3N was purchased from eFlexPV Shenzhen.

Nuclear magnetic resonance (NMR) spectra were measured on a Bruker AVANCE NEO 600 spectrometer. Mass spectra (MS) were obtained from a Bruker Solarix XR FTMS Mass Spectrometer. Thermogravimetric analysis (TGA) was performed using NETZSCH STA449C analyzer under nitrogen gas flow with a heating rate of 20 °C/min. Photoluminescence (PL) spectra were taken on Horiba Jobin yvon FluoroMax-4 spectrometer. X-ray diffraction (XRD) measurement was recorded in the range of 20= 3-32° on a desktop X-ray diffractometer (Rigaku MiniFlex 600) with Cu K_{α} radiation (λ = 1.5406 Å). UV-vis absorption spectra were taken on Agilent Cary 50 UV-vis spectrometer. Cyclic voltammetry (CV) measurements were carried out on an Ingsens IGS1100 electrochemical analyzer with a three-electrode cell in a deoxygenated anhydrous acetonitrile solution of *n*-Bu₄NPF₆ (0.1 M). A platinum disk electrode, platinum-wire, and Ag/AgNO₃ electrode were used as a working electrode, a counter electrode, and a reference electrode, respectively. The CV curves were calibrated using ferrocene/ferrocenium (Fc/Fc⁺) redox couple as an external standard, which was measured under the same condition before and after the measurement of samples. During CV measurements, the films were drop-cast on the working electrode from chloroform solution. Because SMA-3 can be easily dissolved in acetonitrile, its acetonitrile solution was used to evaluate its electrochemical energy levels. E_{1/2} Fc/Fc+ = 0.09 V. The HOMO and LUMO energy levels were respectively estimated according to the equations: $E_{HOMO} = -e(E_{ox} + 4.71)$ (eV) and $E_{LUMO} = -e(E_{red} + 4.71)$ (eV), where E_{ox} is the first onset oxidation, $E_{\rm red}$ is the first onset reduction and the unit of potential is V versus Ag/AgNO₃. The surface morphology of the films were obtained by atomic force microscopy (Advanced Scanning Probe Microscope, XE-100, PSIA).

1.2 Synthesis of intermediates via direct arylation reactions



Intermediate A1: Under the protection of argon, 2,7-dibromo-9,9-dihexyl-9H-fluorene (0.55 mmol, 0.2708 g), K_2CO_3 (0.83 mmol, 0.1147 g), trimethylacetic acid (0.17 mmol, 0.0174 g), 2-thiophenecarboxaldehyde (1.65 mmol, 0.185 g), deoxygenated toluene (5 mL) were added to a schlenk tube. Then the ligand PCy₃·HBF₄ (0.022 mmol, 0.0081 g) and palladium acetate (0.011 mmol, 0.0025 g) were added, and the reaction was carried out at 80°C and the reaction process was tracked via thin layer chromatography (TLC). Twenty hours later, the PCy₃·HBF₄ (0.022 mmol, 0.0081 g) and palladium acetate (0.011 mmol, 0.0025 g) were added again,

and the reaction mixture was heated at 120°C for 24 hours. Then the mixture was poured into ice water to quench the reaction. It was extracted with dichloromethane. The combined organic extracts were washed with water and brine and then dried over MgSO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and dichloromethane (7:13, v/v) as the eluent to afford a yellow solid (0.2241 g, 73.4%), $R_f = 0.27$ (petroleum ether: dichloromethane 1:3, v/v). ¹**H NMR** (400 MHz, CDCl₃, ppm): δ 9.92(s, 2H), 7.79-7.74(m, 4H), 7.70(d, J = 6.9 Hz, 2H), 7.64(s, 2H), 7.49(d, J = 3.9 Hz, 2H), 2.07-2.01(m, 4H), 1.09 (dd, J = 14.8, 8.5 Hz, 12H), 0.76 (t, J = 6.7 Hz, 6H), 0.72-0.62 (m, 4H). ¹³**C NMR** (100 MHz, CDCl₃, ppm): δ 182.72, 154.80, 152.26, 142.26, 141.64, 137.46, 132.43, 125.75, 124.07, 120.78, 120.71, 77.36, 77.04, 76.72, 55.56, 40.27, 31.42, 29.58, 23.78, 22.54, 13.97.



Ethyl 4-(2-formylthiophen-3-yl)benzoate: Under the protection of argon, ethyl 4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzoate (5.31 mmol, 1.4664 g), 3-bromothiophene-2-carboxaldehyde (4 mmol, 0.7642 g), and deoxygenated tetrahydrofuran (65 mL), Na₂CO₃ (20.52 mmol, 2.1749 g), deoxygenated water (25 mL) and Pd(PPh₃)₄ (0.086 mmol, 0.0994 g) was added to a flask in turn. The reaction was carried out at 80°C and the reaction process was tracked via TLC. Nineteen hours later, the mixture was poured into water and extracted with ethyl acetate. The organic layer was separated, washed with brine dried over MgSO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and ethyl acetate (6:1, v/v) as the eluent and the crude product was then recrystallized with methanol to afford a white solid (0.7675 g, 73.7%), *R*_f = 0.29 (petroleum ether: ethyl acetate 6:1, v/v). ¹**H NMR** (400 MHz, CDCl₃, ppm): δ 9.88 (s, 1H), 8.16 (d, *J* = 8.3 Hz, 2H), 7.78 (d, *J* = 5.0 Hz, 1H), 7.56 (d, *J* = 8.3 Hz, 2H), 7.26 (d, *J* = 5.0 Hz, 1H), 4.43 (q, *J* = 7.1 Hz, 2H), 1.43 (t, *J* = 7.1 Hz, 3H). ¹³**C NMR** (100 MHz, CDCl₃, ppm): δ 183.67, 166.02, 149.99, 139.16, 138.19, 134.43, 130.75, 130.48, 130.00, 129.55, 61.30, 14.35.



Intermediate A4: Under the protection of argon, 1,4-dibromo-2,5-bis((2-hexyldecyl)oxy)benzene (0.63 mmol, 0.4516 g), ethyl 4-(2-formylthiophen-3-yl)benzoate (1.38 mmol, 0.3592 g), K_2CO_3 (0.9 mmol, 0.1244 g), trimethylacetic acid (0.314 mmol, 0.0321 g), anhydrous toluene (25 mL), anhydrous dimethylformamide (5 mL) were added to a Schlenk tube. The mixture was deoxygenated by bubbling argon for 30 min. The PCy₃·HBF₄ (0.06 mmol, 0.0221 g) and Pd(PPh₃)₄ (0.028 mmol, 0.0324 g) were then added. The tube was sealed and the reaction was carried out at 100°C for 25 hours and 120°C for 16 hours. The mixture was then poured into water and extracted with ethyl acetate. The combined organic extracts were washed with water and brine and then dried over MgSO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and ethyl acetate (13:1, v/v) as the eluent and the crude product was then recrystallized with methanol to afford a yellow solid (0.3321 g, 49.0%), R_f = 0.31 (petroleum ether: ethyl acetate 13:1, v/v). ¹**H NMR** (600 MHz, CDCl₃, ppm): δ 9.89 (s, 2H), 8.18 (d, *J* = 8.3 Hz, 4H), 7.73 (s, 2H), 7.61 (d, *J* = 8.4 Hz, 4H), 7.33 (s, 2H), 4.43 (t, *J* = 7.2 Hz, 4H), 4.03 (d, *J* = 5.5 Hz, 4H), 1.94-1.91 (m, 2H), 1.44 (m, 12H), 1.30 (m, 42H), 0.85 (m, 12H). ¹³**C NMR** (100 MHz, CDCl₃, ppm): δ 183.61, 166.04, 150.29, 149.74,

147.64, 138.47, 137.94, 130.74, 130.01, 129.48, 129.10, 123.10, 112.60, 72.53, 61.32, 38.24, 31.90, 31.84, 31.63, 31.58, 30.03, 29.71, 29.59, 29.33, 26.97, 22.69, 14.36, 14.24.

1.3 Synthesis of small molecular acceptors via ball milling reactions

Mechnochemical experiments were carried out in a planetary ball mill FOCUCY F-P400E that was equipped with steel jars (28 mL, 3.8 mm inner diameter, 6 mm outer diameter) with stainless steel spheres inside (see Figure S1). To equilibrate the system, another parallel vessel was used. The reactions were performed in air at room temperature unless otherwise noted. Each reaction progress was monitored by TLC, and was superadded the catalyst or reactant according to the results of TLC. *Note*: If necessary, the reaction was assisted via the thermal preservation processes (80°C). In this case, the ball milling reaction was suspended. The jar was opened and was then taken into a vacuum pump. Then the pump operated for 1 hour (80°C, 0.09 MPa).

SMA-1 was chosen as the model target compound for studying the ball milling reactions, and **A1** and **B1** were the raw materials. The general procedure for the synthesis of **SMA-1** was described below: **A1** (0.2 mmol), **B1** (0.6 mmol) and catalyst (1.2 mmol) were added to a stainless steel grinding jar equipped with a certain amount of stainless steel ball in air at room temperature. The jar was then sealed. The mixture was milled with specific ball milling speed and the reaction was monitored by TLC. The catalyst or **B1** was superadded to promote the reaction in the process and Figure S2 showed the detailed information. The resulting crude product mixture was dissolved in ethyl acetate. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, the resulting residue was recrystallized with methanol to afford **SMA-1** as a black solid. The optimized parameters and corresponding results were listed in Table 1. $R_f = 0.38$ (petroleum ether: dichloromethane 1:3, v/v). **1H NMR** (600 MHz, CDCl₃, ppm): δ 8.93 (s, 2H), 8.73 (d, *J* = 7.7 Hz, 2H), 7.99 (d, *J* = 6.9 Hz, 2H), 7.92 (d, *J* = 3.9 Hz, 2H), 7.87 (d, *J* = 7.9 Hz, 2H), 7.80 (dd, *J* = 16.3, 8.3 Hz, 8H), 7.62 (d, *J* = 3.9 Hz, 2H), 7.92 (d, *J* = 3.9 Hz, 2H), 7.87 (d, *J* = 7.9 Hz, 2H), 7.80 (dd, *J* = 16.3, 8.3 Hz, 8H), 7.62 (d, *J* = 3.9 Hz, 2H), 2.13 (m, 4H), 1.11 (m, *J* = 7.2 Hz, 6H), 1.05 (m, 4H), 0.75 (m, *J* = 7.1 Hz, 6H), 0.66 (t, 6H). ¹³C NMR (150 MHz, CDCl₃, ppm): δ 188.60, 160.88, 154.36, 152.59, 146.42, 142.40, 140.13, 138.08, 137.06, 136.57, 135.36, 134.68, 132.86, 126.26, 125.41, 125.01, 123.72, 122.74, 121.26, 120.88, 114.53, 114.45, 69.96, 55.90, 40.85, 31.50, 29.59, 23.81, 21.63, 13.99.



Figure S1. (a) and (b): Planetary ball mill (FOCUCY F-P400E); (c): stainless balls; (d): stainless jars.



Figure S2. The details of the ball milling reactions for synthesizing the model small molecular acceptor SMA-1.

SMA-2 ~ SMA-10 were also synthesized via ball milling reactions. The results were listed in Table S1 and the details were described below.

Table S1. Scope of small molecular acceptors synthesized via ball milling reactions^[a]

Entry	product	X:Y:Z ^[b]	Milling time (min) ^[c]	Yield (%) ^[d]
1	SMA-2	1:3:12	160	50.9
2	SMA-3	1:3:12	160	86.2
3	SMA-4	1:3:24	160	31.0
4	SMA-5	1:3:9	80	98.9
5	SMA-6	1:21:45	320	46.0
6	SMA-7	1:15:33	240	39.3
7	SMA-8	1:3:36	240	75.0
8	SMA-9	1:3:36	240	73.4
9	SMA-10	1:3:36	240	66.0

^[a] Reaction conditions: the initial mole ratio of the dialdehyde aromatic derivative: methylene-activated derivative: catalyst is 1:3:6, in air. See the corresponding experimental description below for details of the procedure. ^[b] The final mole ratio of the dialdehyde aromatic derivative (X): methylene-activated derivative (Y): catalyst(Z). ^[c] Monitored by TLC. ^[d] Isolated yield.



SMA-2: The reaction was carried out with **A1** (0.2 mmol, 0.1110 g, 1 equivalent), 1,3-indanedione (**B2**) (0.6 mmol, 0.0877 g) and CH₃COONH₄ (1.2 mmol, 0.0925 g). The mixture was milled for 40 min in air at room temperature, 3 equivalents of CH₃COONH₄ were then added. The mixture was milled for 40 min again in air at room temperature, and then another 3 equivalents of CH₃COONH₄ were added. Subsequently, the mixture was milled for another 80 min in air at room temperature and the reaction was completed (monitored by TLC). The resulting crude product was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, the resulting residue was recrystallized with methanol to afford **SMA-2** as a bright red solid (0.0825 g, 50.9%), $R_f = 0.16$ (petroleum ether: dichloromethane 1:3, v/v). ¹H NMR (600 MHz, CDCl₃, ppm): δ 8.01 (m, 8H), 7.85-7.74 (m, 10H), 7.57 (d, J = 4.1 Hz, 2H), 2.11 (m, 4H), 1.15-1.00 (m, 12H), 0.75 (t, J = 7.2 Hz, 6H), 0.66 (m, 4H). ¹³**C NMR** (150 MHz, CDCl₃, ppm): δ 190.59, 190.10, 158.10, 152.53, 143.99, 142.20, 142.07, 140.70, 136.75, 136.45, 135.24, 135.05, 132.87, 126.10, 125.04, 123.97, 123.21, 122.99, 120.96, 120.86, 77.37, 77.16, 76.95, 55.93, 40.49, 31.60, 29.75, 23.93, 22.71, 14.12.



SMA-3: The reaction was carried out with **A1** (0.2 mmol, 0.1110 g, 1 equivalent), malononitrile (**B3**) (0.6 mmol, 0.0396 g) and CH₃COONH₄ (1.2 mmol, 0.0925 g). The mixture was milled in air at room temperature. After ball milling for 40 min and 80 min respectively, 3 equivalents of CH₃COONH₄ were added. The reaction was completed by ball milling for 160 min. The resulting crude product was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, the resulting residue was recrystallized with methanol to afford **SMA-3** as a bright red solid (0.1122 g, 86.2%), $R_{\rm f} = 0.66$ (dichloromethane). ¹H **NMR** (600 MHz, CDCl₃, ppm): δ 7.83 (s, 2H), 7.80 (d, *J*=7.9 Hz, 2H), 7.76-7.73 (m, 4H), 7.64 (d, *J*=1.8 Hz, 2H), 7.54 (d, *J*=4.1 Hz, 2H), 2.09-2.07 (m, 4H), 1.13-1.00 (m, 12H), 0.75 (t, *J*=7.2 Hz, 6H), 0.66-0.60 (m, 4H). ¹H-NMR spectra data can be identified as the same with the literature^[8].



SMA-4: The reaction was carried out with **A1** (0.2 mmol, 0.1110 g, 1 equivalent), 3-ethylrhodanine (**B4**) (0.6 mmol, 0.0968 g) and CH₃COONH₄ (1.2 mmol, 0.0925 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 80 min and 120 min, respectively, the mixture was treated with the abovementioned thermal preservation processes, followed by 6 equivalents of CH₃COONH₄ were added each. After ball milling for another 40 min, the reaction was completed. The resulting crude product was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, the resulting residue was recrystallized with methanol to afford **SMA-4** as a red solid (0.0521 g, 31.0%), R_f = 0.81 (dichloromethane). ¹H **NMR** (600 MHz, CDCl₃, ppm): δ 7.89 (s, 2H), 7.75 (d, *J* = 8.0 Hz, 2H), 7.68 (d, *J* = 7.7 Hz, 2H), 7.61 (s, 2H), 7.49 (d, *J* = 3.9 Hz, 2H), 7.44 (d, *J* = 4.0 Hz, 2H), 4.29-4.14 (m, 4H), 2.11-2.03 (m, 4H), 1.31 (t, *J* = 7.2 Hz, 6H), 1.15-0.99 (m, 8H), 0.75 (m, 8H), 0.67 (m, 6H). ¹³C **NMR** (150 MHz, CDCl₃, ppm): δ 192.23, 167.49, 153.25, 152.42, 141.56, 137.12, 135.72, 132.87, 125.48, 125.43, 124.86, 120.87, 120.65, 120.29, 77.37, 77.16, 76.95, 55.75, 40.46, 40.09, 31.60, 29.72, 23.92, 22.68, 14.15, 12.44.



SMA-5: The reaction was carried out with **A2** (0.09 mmol, 0.1108 g, 1 equivalent), malononitrile (**B3**) (0.27 mmol, 0.0178 g) and CH₃COONH₄ (0.54 mmol, 0.0416 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 3 equivalents of CH₃COONH₄ were added. After ball milling for another 40 min, the reaction was completed. The resulting crude product was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, the resulting residue was washed with methanol to gain **SMA-5** as a black solid (0.1182 g, 98.9%), $R_f = 0.48$ (petroleum ether: dichloromethane 1:2, v/v). ¹H NMR (600 MHz, CDCl₃, ppm): δ 8.76 (s, 2H), 8.73 (d, *J* = 8.0

Hz, 2H), 8.27 (s, 2H), 7.97 (d, J = 8.0 Hz, 2H), 7.60 (s, 2H), 7.24 (d, J = 8.3 Hz, 8H), 7.12 (d, J = 8.4 Hz, 8H), 2.63-2.54 (m, 8H), 1.63-1.57 (m, 8H), 1.37 -1.24 (m, 24H), 0.86 (t, J = 6.9 Hz, 12H). ¹H-NMR spectra data can be identified as the same with the literature^[9].



SMA-6: The reaction was carried out with **A2** (0.09 mmol, 0.1108 g, 1 equivalent), 3-ethylrhodanine (**B4**) (0.27 mmol, 0.0435 g) and CH₃COONH₄ (0.54 mmol, 0.0416 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 3 equivalents of CH₃COONH₄ were added. Then, after ball milling for every 40 min, the mixture was treated with the above-mentioned thermal preservation processes, followed by 6 equivalents of CH₃COONH₄ and 3 equivalents of 3-ethylrhodanine were added each. When the mixture was milled for total 320 min, the reaction was completed. The resulting mixture was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and dichloromethane (7:4, v/v) as the eluent and the crude product was then recrystallized with methanol to afford **SMA-6** as a black solid (0.0629 g, 46.0%), *R*_f = 0.27 (petroleum ether: dichloromethane 7:4, v/v). **1H NMR** (600 MHz, CDCl₃, ppm): δ 8.49 (s, 2H), 8.17 (s, 2H), 7.94 (d, *J* = 7.7 Hz, 2H), 7.67 (d, *J* = 7.7 Hz, 2H), 7.57 (s, 2H), 7.26-7.24 (m, 8H), 7.11 (d, *J* = 8.5 Hz, 8H), 4.24 (q, *J* = 7.2 Hz, 4H), 2.59-2.55 (m, 8H), 1.63-1.53 (m, 8H), 1.36-1.25 (m, 30H), 0.88-0.85 (m, 12H). ¹H-NMR spectra data can be identified as the same with the literature^[3].



SMA-7: The reaction was carried out with **A3** (0.1 mmol, 0.1040 g, 1 equivalent), 3-ethylrhodanine (**B4**) (0.3 mmol, 0.0484 g) and CH₃COONH₄ (0.6 mmol, 0.0462 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 3 equivalents of CH₃COONH₄ were added. Then, after ball milling for every 40 min, the mixture was treated with the above-mentioned thermal preservation processes, followed by 6 equivalents of CH₃COONH₄ and 3 equivalents of 3-ethylrhodanine were added each. When the mixture was milled for total 240 min, the reaction was completed. The resulting mixture was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and dichloromethane (2:3, v/v) as the eluent and the crude product was then recrystallized with methanol to afford **SMA-7** as a black solid (0.0521 g, 39.3%), *R*_f = 0.74 (petroleum ether: dichloromethane 1:2, v/v). ¹H NMR (600 MHz, CDCl₃, ppm): δ 8.53 (s, 2H), 8.22 (s, 2H), 8.01 (d, *J* = 7.7 Hz, 2H), 7.73 (d, *J* = 8.5 Hz, 2H), 7.42 (s, 2H), 4.26 (q, *J* = 7.2 Hz, 4H), 2.14-1.94 (m, 8H), 1.34 (t, *J* = 7.2 Hz, 6H), 1.29-1.07 (m, 40H), 0.92-0.82 (m, 8H), 0.78 (t, *J* = 7.0 Hz, 12H). ¹H-NMR spectra data can be identified as the same with the literature^[4].



SMA-8: The reaction was carried out with 2-(5,6-dichloro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (B5) (0.3 mmol, 0.0789 g), A4 (0.1 mmol, 0.1076 g, 1 equivalent) and CH₃COONH₄ (0.6 mmol, 0.0462 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 6 equivalents of CH₃COONH₄ were added. Then, after ball milling for every 40 min, the mixture was treated with the above-mentioned thermal preservation processes, followed by 6 equivalents of CH₃COONH₄ were added each. When the mixture was milled for total 240 min, the reaction was completed. The resulting mixture was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and dichloromethane (5:8, v/v) as the eluent and the crude product was then recrystallized with methanol to afford **SMA-8** as a black solid (0.1174 g, 75.0%), $R_f = 0.29$ (petroleum ether: dichloromethane 1:2, v/v). ¹H NMR (600 MHz, CDCl₃, ppm): δ 8.74 (s, 2H), 8.67 (s, 2H), 8.22 (d, J = 8.3 Hz, 4H), 7.96 (s, 2H), 7.85 (s, 2H), 7.56 (d, J = 8.3 Hz, 4H), 7.46 (s, 2H), 4.44 (q, J = 7.1 Hz, 4H), 4.15 (d, J = 5.3 Hz, 4H), 2.07 (m, 2H), 1.64-1.60 (m, 4H), 1.46 (t, J = 7.2 Hz, 6H), 1.38 (m, 10H), 1.26-1.19 (m, 34H), 0.85-0.79 (m, 12H). ¹³C NMR (150 MHz, CDCl₃, ppm): δ 185.11, 166.04, 158.88, 156.92, 152.80, 151.07, 139.79, 139.44, 138.84, 138.60, 137.81, 136.15, 133.89, 131.23, 130.29, 130.07, 129.43, 126.86, 125.22, 124.08, 122.83, 114.15, 113.42, 112.28, 73.00, 70.84, 61.35, 38.23, 31.92, 31.49, 31.45, 30.08, 29.75, 29.69, 29.39, 27.03, 22.71, 22.67, 14.35, 14.12. FT-MS (m/z): calcd for C₉₀H₉₄Cl₄N₄O₈S₂: 1563.5340 Found: 1563.5365. TGA (5% weight loss): 346°C. PL: excitation λ_{max} 532 nm; emission λ_{max} 816 nm.



SMA-9: The reaction was carried out with 2-(5,6-difluoro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (**B6**) (0.3 mmol, 0.0691 g), **A4** (0.1 mmol, 0.1076 g, 1 equivalent) and CH₃COONH₄ (0.6 mmol, 0.0462 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 6 equivalents of CH₃COONH₄ were added. Then, after ball milling for every 40 min, the mixture was treated with the above-mentioned thermal preservation processes, followed by 6 equivalents of CH₃COONH₄ were added each. When the mixture was milled for total 240 min, the reaction was completed. The resulting mixture was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and dichloromethane (1:4, v/v) as the eluent to afford **SMA-9** as a black solid (0.1101 g, 73.4%), *R*_f = 0.23 (petroleum ether: dichloromethane 1:4, v/v). ¹**H NMR** (600 MHz, CDCl₃, ppm): δ 8.65 (s, 2H), 8.51 (dd, *J* = 9.9, 6.4 Hz, 2H), 8.22 (d, *J* = 8.3 Hz, 4H), 7.84 (s, 2H), 7.69 (t, *J* = 7.4 Hz, 2H), 7.55 (d, *J* = 8.3 Hz, 4H), 7.46 (s, 2H), 4.44 (q, *J* = 7.1 Hz, 4H), 4.15 (d, *J* = 5.3 Hz, 4H), 2.08 (m, 2H), 1.61 (m, 4H), 1.53-1.51 (m, 2H), 1.45 (t,

J = 7.1 Hz, 6H), 1.38 (m, 8H), 1.29-1.15 (m, 34H), 0.82 (q, *J* = 7.1 Hz, 12H). ¹³**C NMR** (150 MHz, CDCl₃, ppm): δ 185.19, 166.18, 159.13, 156.90, 155.55, 153.80, 152.67, 151.17, 139.03, 137.36, 133.72, 131.33, 130.41, 130.18, 129.54, 124.18, 122.94, 115.07, 114.29, 113.46, 112.81, 112.69, 112.47, 73.07, 70.88, 61.47, 38.36, 32.04, 31.62, 30.20, 29.86, 29.80, 29.51, 27.13, 22.81, 14.48, 14.23. **FT-MS** (m/z): calcd for C₉₀H₉₄F₄N₄O₈S₂: 1499.6522 Found: 1499.6478. **TGA** (5% weight loss): 340°C. **PL**: excitation λ_{max} 532 nm; emission λ_{max} 813 nm.



SMA-10: The reaction was carried out with 2-(5-bromo-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (B7) (0.3 mmol, 0.0891 g), A4 (0.1 mmol, 0.1076 g, 1 equivalent) and CH₃COONH₄ (0.6 mmol, 0.0462 g). The mixture was milled in air at room temperature. After ball milling for 40 min, 6 equivalents of CH₃COONH₄ were added. Then, after ball milling for every 40 min, the mixture was treated with the above-mentioned thermal preservation processes, followed by 6 equivalents of CH₃COONH₄ were added each. When the mixture was milled for total 240 min, the reaction was completed. The resulting mixture was dissolved in dichloromethane. The organic phase was washed with water, brine, and dried with anhydrous Na₂SO₄. After removing the solvent, silica gel column chromatography was used to purify the product with the mixture of petroleum ether and ethyl acetate (4:1, v/v) as the eluent and the crude product was then recrystallized with methanol to afford SMA-10 as a black solid (0.1046 g, 66.0%), R_f = 0.24 (petroleum ether: dichloromethane 1:9, v/v). ¹H NMR (600 MHz, CDCl₃, ppm): δ 8.66 (s, 2H), 8.51 (d, J = 8.5 Hz, 2H), 8.22 (d, J = 8.1 Hz, 4H), 8.03 (d, J = 2.0 Hz, 2H), 7.86 (dd, J = 8.5, 2.0 Hz, 2H), 7.84 (s, 2H), 7.56 (d, J = 8.2 Hz, 4H), 7.45 (s, 2H), 4.44 (q, J = 7.1 Hz, 4H), 4.15 (d, J = 5.3 Hz, 4H), 2.08 (m, 2H), 1.64-1.59 (m, 4H), 1.53 (m, 2H), 1.45 (t, J = 7.2 Hz, 6H), 1.41-1.36 (m, 10H), 1.28-1.18 (m, 40H), 0.83-0.80 (m, 12H). ¹³C NMR (150 MHz, CDCl₃, ppm): δ 185.91, 166.09, 160.18, 156.54, 152.37, 151.02, 138.98, 138.42, 137.87, 137.49, 133.77, 131.13, 130.26, 130.07, 129.70, 129.36, 126.42, 124.04, 123.10, 114.51, 113.68, 112.33, 72.95, 70.29, 61.33, 38.24, 31.92, 31.51, 30.08, 29.75, 29.68, 29.38, 27.04, 22.69, 14.35, 14.12. **FT-MS (m/z)**: calcd for C₉₀H₉₆Br₂N₄O₈S₂: 1583.5109 Found: 1583.5098. TGA (5% weight loss): 344°C. PL: excitation λ_{max} 532 nm; emission λ_{max} 814 nm.

1.4 Device fabrication and characterization

The inverted structure is ITO/ZnO/active layer/MoO₃/Ag and the conventional structure is ITO/PEDOT:PSS/active layer/PFN-F3N/Ag. For the device fabrication, the ITO substrates were ultrasonic cleaned in acetone, water, acetone, and isopropyl alcohol. First, for the inverted device, sol-gel ZnO was spin-coated onto the pre-cleaned ITO substrate at 4000 rpm for 40 s and then the substrates were baked at 200°C for 10 min under ambient condition; for the conventional one, the PEDOT:PSS spin-coated onto the pre-cleaned ITO substrate at 4000 rpm for 30 s and then the substrates were baked at 150°C for 10 min under ambient condition. Subsequently, polymer:**SMA-8** (10-16 mg/mL in chloroform) solutions with or without 1 vol% CN were deposited onto the ZnO or PEDOT:PSS layer for 40 s, at spin rate of 2000-3000 rpm in the nitrogen glove box. After thermal annealing at 100°C for 10 min in the glove box, finally, for the inverted device, MoO₃

at a speed of 0.2 Å/s (8 nm), and Ag at a speed of 2 Å/s (100 nm) layers were thermally evaporated at a pressure of 5 × 10⁻⁶ mbar to accomplish the device fabrication; for the conventional device, PNDIT-F3N (0.5 mg/mL in methanol) was spin-coated onto the active layer at 2000 rpm for 30 s and Ag at a speed of 2 Å/s (100 nm). The J-V characterization was performed by Keithley 2400 digital source meter under simulated AM 1.5G solar irradiation at 100 mW cm⁻². The device area is 0.0725 cm², solar cell devices were measured in forward scan (-1.0 V \rightarrow 1.0 V, step 0.0125 V, scan rate: 0.1 V s⁻¹) without an illumination mask in the glovebox.

We firstly constructed an inverted device configuration of ITO/ZnO/polymer donor: SMA-8/MoO₃/Ag under 100 mW cm⁻² AM 1.5 G illumination. The composition of the active layer was adjusted from 1/1.8 to 1/1.2 by weight, and chloroform is selected as the processing solvent. The optimization conditions were summarized in Table S3-S4. The as-cast PBDB-T:SMA-8 device (1/1.8, w/w) showed a PCE of 2.27%, which was significantly lower compared to the one in PM6:SMA-8 device (4.23%) under the similar condition. Hence, we further tuned the concentration of the PM6:SMA-8, and CN was used as a processing additive to optimize the blend morphology. As can we noted, the optimized PM6:SMA-8 device (1/1.8, w/w) at a concentration delivering a PCE of 5.13% with an open-circuit voltage (V_{oc}) of 0.887 V, a short-circuit current density (J_{sc}) of 9.90 mA cm⁻² and a fill factor (FF) of 0.58. As shown in Table S5, by further adding 1 vol% CN to the processing solvent, the corresponding PM6:SMA-8 affords a significantly much higher PCE of 8.72% together with an enhanced J_{sc} of 15.92 mA cm⁻², a FF of 0.67 and a slightly decreased V_{oc} of 0.817 V. It has been reported that the conventional device structure can further improve device performance due to favorable vertical phase separation and energy level alignment^[10]. We finally thought to adopt the reported structure (ITO/PEDOT:PSS/PM6:SMA-8/PNDIT-F3N/Ag) to further optimize the photovoltaic performance of devices based on PM6:SMA-8. As shown in Fig. 3 and Table S6, the conventional devices with the same active layer exhibited improved performance relative to those using inverted structure. The PCE can be further increased to 10.02%.

2. Some experimental data



Figure S3. ¹H NMR spectrum of ethyl 4-(2-formylthiophen-3-yl)benzoate.



Figure S4. ¹³C NMR spectrum of ethyl 4-(2-formylthiophen-3-yl)benzoate.



Figure S5.¹H NMR spectrum of A4.



Figure S6.¹³C NMR spectrum of A4.





Figure S8.13C NMR spectrum of SMA-8.



Figure S11.¹H NMR spectrum of SMA-10.







 Meas.m/z
 # ion Formula
 Score
 m/z
 err [ppm]
 Mean err [ppm]
 mSigma
 rdb
 e⁻⁻ Conf
 N-Rule

 1565.536529
 1
 C90H95CHN406852
 100.00
 1563.533996
 -1.6
 2.3
 112.3
 60.0
 even
 ok

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Figure S13. Mass spectrum (FT-MS) of SMA-8.



 Meas.m/z
 #
 Ion Formula
 Score
 m/z
 err [ppm]
 Mean err [ppm]
 mSigma
 rdb
 e⁻Conf
 N-Rule

 1499.647845
 1
 090H95F4N40852
 100.00
 1499.652197
 2.9
 1.3
 167.9
 48.0
 even
 ok

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Figure S14. Mass spectrum (FT-MS) of SMA-9.



 Meas. m/z
 #
 Ion Formula
 Score
 m/z
 err [ppm]
 Mean err [ppm]
 mSigma
 rdb
 e[®] Conf
 N-Rule

 1583.509842
 1
 090H97Br2N40682
 100.00
 1583.510910
 0.7
 -1.3
 100.1
 52.0
 even
 ok

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Figure S15. Mass spectrum (FT-MS) of SMA-10.



Figure S16. UV-vis absorption spectra of small molecular acceptors (the insert shows the corresponding photograph of sample).



Figure S17. Cyclic voltammetry diagram of small molecular acceptors.

 Table S2. Optical properties and electrochemical properties.

product	٤ 10⁴ M⁻¹cm⁻¹	λ _{max} sol [nm]	λ _{max} ^{film} [nm]	λ _{onset} ^{film} [nm]	<i>E_g^{opt}</i> [eV] ^[a]	E _{red} / E _{LUMO} (V/ eV) ^[b]	Е _{ох} ∕ Е _{номо} (V/ eV) ^[с]
SMA-1	7.0	579	637	740	1.68	-0.80/ -3.91	1.00/ -5.71
SMA-2	11.5	511	529	598	2.07	-1.04/ -3.67	1.14/ -5.85
SMA-3	4.4	475	484	568	2.18	-1.18/ -3.53	1.08/ -5.79
SMA-4	6.6	490	490	576	2.15	-1.05/ -3.66	0.94/ -5.65
SMA-5	15.2	659	673	770	1.61	-1.00/ -3.71	0.89/ -5.60
SMA-6	11.3	629	665	735	1.69	-1.09/ -3.62	0.76/ -5.47
SMA-7	9.9	644	691	782	1.59	-0.98/ -3.73	0.70/ -5.41
SMA-8	9.7	645	761	825	1.50	-0.72/ -3.99	0.89/ -5.60
SMA-9	10.2	638	730	772	1.61	-0.74/-3.97	1.11/-5.82
SMA-10	11.9	642	665	790	1.57	-0.77/-3.94	1.09/-5.80

^[a] Calculated by the equation: $E_{g}^{\text{film}} = 1240 / \lambda_{\text{onset}}^{\text{film}} \text{ eV}$. ^[b] $E_{\text{LUMO}} = -(E_{\text{red}} - E_{1/2}^{\text{ForFc+}} + 4.8) \text{ eV}$. ^[c] $E_{\text{HOMO}} = -(E_{\text{ox}} - E_{1/2}^{\text{ForFc+}} + 4.8) \text{ eV}$.

 $E_{1/2}^{\text{Fc/Fc+}} = 0.09 \text{ V}.$



Figure S18. The TGA curve of SMA-8.



Figure S19. The TGA curve of SMA-9.



Figure S20. The TGA curve of SMA-10.



Figure S21. Photoluminescence spectrum of SMA-8 (excitation: λ_{max} =532 nm).



Figure S22. Photoluminescence spectrum of SMA-9 (excitation: λ_{max} =532 nm).



Figure S23. Photoluminescence spectrum of SMA-10 (excitation: λ_{max} =532 nm).



Figure S24. X-ray diffraction pattern of SMA-8 thin film (d = 21.07 Å).



Figure S25. X-ray diffraction pattern of SMA-9 thin film (d = 21.12 Å).



Figure S26. X-ray diffraction pattern of SMA-10 thin film (d = 22.09 Å).



Figure S27. Chemical structures of (a) PBDB-T and (b) PM6 polymer donors.

Table S3. Photovoltaic data based on PBDB-T:SMA-8 inverted structure solar cells (blend solution of 16 mg/mL and spin rate of 3000 rpm).

1 ,					
Active layer	Weight ratio	V _{oc} (V)	J _{sc} (mA/cm²)	FF (%)	PCE (%)
PBDB-T:SMA-8	1/1.5	0.734	6.22	0.40	1.82
PBDB-T:SMA-8	1/1.8	0.756	6.84	0.44	2.27
PBDB-T:SMA-8	1/1.2	0.763	6.30	0.44	2.11

Table S4. Photovoltaic data based on PM6:SMA-8 inverted structure solar cells (blend ratio of 1/1.8).

Active layer	Concertation	Spin-rate	V _{oc} (V)	J _{sc} (mA/cm²)	FF (%)	PCE (%)
PM6: SMA-8	10 mg/mL	3000	0.870	8.84	0.57	4.35
PM6: SMA-8	12 mg/mL	3000	0.887	9.90	0.58	5.13
PM6: SMA-8	14 mg/mL	3000	0.864	10.45	0.49	4.45
PM6: SMA-8	16 mg/mL	3000	0.840	10.26	0.49	4.23
PM6: SMA-8	12 mg/mL	2000	0.855	8.94	0.40	3.01
PM6: SMA-8	12 mg/mL	2500	0.877	10.53	0.49	4.52

Table S5. Photovoltaic data based on PM6:SMA-8 inverted structure solar cells with and without 1 % CN (blend solution of 12 mg/mL

and spin rate of 3000 rpm).							
Active layer	Additive	V _{oc} (V)	J _{sc} (mA/cm²)	FF (%)	PCE (%)		
PM6: SMA-8	0 % CN	0.887	9.90	0.58	5.13		
PM6: SMA-8	1% CN	0.817	15.92	0.67	8.72		

 Table S6. Photovoltaic data based on PM6:SMA-8 conventional structure solar cells with and without 1 % CN (blend solution of 12 mg/mL and spin rate of 3000 rpm).

Active layer	Additive	V _{oc} (V)	J _{sc} (mA/cm²)	FF (%)	PCE (%)
PM6: SMA-8	0 % CN	0.887	10.89	0.58	5.60
PM6: SMA-8	1 % CN	0.831	17.46	0.69	10.02

Material		Quantity consumed	Cost (¥)	Total (¥)
	2-hexyldecan-1-ol	128.8 g	128.8	
	Bromine	157 g	219.8	
	PPh ₃	141.1 g	70.1	
	hydroquinone	22.5 g	6.8	
	Potassium carbonate	80.7 g	3.2	
	ethyl 4-bromobenzoate	117.9 g	353.7	
	Bis(pinacolato)diboron	103.7 g	238.5	
	Potassium acetate	151.4 g	148.4	
	PdCl ₂ (dppf)	20.3 g	1015	
	Dichloromethyl methyl ether	108.3 g	1083	
Reagents	Titanium tetrachloride	274.5 g	82.4	
rteagents	3-bromothiophene	80.8 g	142.2	
	Sodium carbonate	276.2	11	
	Pd(PPh ₃) ₄	21.5 g	922.3	
	Pivalic acid	8.8 g	2.4	
	PCy ₃ ·HBF ₄	6.1 g	73.2	
	5,6-dichloroisobenzofuran-1,3-dione	119.5 g	5616.5	
	Tert-butyl acetoacetate	108.8 g	86	21211 5
	Hydrochloric acid	0.75 L	35.9	21211.5
	Malononitrile	46.7 g	24.3	
	Sodium acetate	43.5 g	4.2	
	Ammonium acetate	236.4 g	26	
	Dichloromethane	0.52 L	5.8	
	N,N-Dimethylformamide	1.69 L	92.9	
	1,4-dioxane	2.06 L	251.3	
	Tetrahydrofuran	8.2 L	615	
Solvent	Toluene	6.87 L	302.2	
	Acetic anhydride	0.82 L	90.2	
	Triethylamine	0.55 L	33	
	Ethanol	1.06 L	29.6	
	Acetic acid	0.2 L	8	
	NaSO ₄	495 g	33.8	
	Eluent and extraction solvent	440.6 L	5727	
Purification	SiO ₂	58.4 kg	2920	
	Methanol	19 L	313	
1	Acetic acid	12.4	496	

Table S7. Survey of calculated chemical synthesis costs for SMA-8 (100 g) $^{\mbox{\tiny [1]}}.$

^[1] The chemical synthesis cost was calculated according to the model reported in the previous literature^[11].

Table S8. Comparison of the synthetic costs for acceptor materials.

Compound	C _g (¥ per g) ^a	Reference
SMA-8	212.1	This work
IDIC ^b	268.4	[11][12]
ITIC ^b	245.1	[11][13]
ITIC-4F ^b	434.4	[11][14]
6TIC-4F b	730.1	[15][16]
Y6 ^b	779.4	[15][17]

 ${}^{a}C_{g}$ cost-per-gram, and unit cost converted from the calculation; b Some representative fused-ring acceptors, and their molecular

structures are shown in Figure. S28.



Figure S28. Chemical structures of acceptor molecules involved in Table S8.

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