Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Electronic Supplementary Information for:

Fused octacyclic electron acceptor isomers for organic solar cells

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Materials

Unless stated otherwise, all the chemical reagents and solvents used were obtained commercially and were used without further purification. Chloroform (99.9%), 1,8-diiodooctane (DIO) (97.0%) and 2-methoxyethanol (99.8%) were purchased from TCI and J&K Chemical Inc.; $Zn(CH_3COO)_2 \cdot 2H_2O$ (99.0%), ethanolamine (99.5%) and MoO_3 were purchased from Sigma-Aldrich Inc. Toluene and tetrahydrofuran (THF) were distilled from sodium benzophenone under nitrogen. Compounds **1**, **3** and PM6 ($M_n = 20.2$ kg mol^{-1}, PDI = 2.0) were purchased from Sunatech Ltd, TCI and 1-Material Inc, respectively. 2FIC was synthesized according to our reported procedure.[S1]

Synthesis of BTIC and NTIC

Compound 2. Compound **1** (492 mg, 1.0 mmol), ethyl 2-bromothiophene-3-carboxylate (550 mg, 2.2 mmol), anhydrous K_2CO_3 (2.8 g, 20 mmol), Aliquat 336 (1 drop), and $Pd(PPh_3)_4$ (115 mg, 0.1 mmol) were dissolved in deoxygenated toluene/H_2O (50 mL, 2/1, v/v). The solution mixture was stirred at 110 °C for 48 h, and then cooled to room temperature. 100 mL of water was added and the mixture was extracted with dichloromethane (2 × 100 mL). The precipitate was filtered and washed with methanol, *n*-hexane and dichloromethane. The product of compound **2** was obtained as a yellow solid (450 mg, 82%). This compound was directly used for next step reaction without further purification due to limited solubility. MS (MALDI-TOF) calcd for $C_{28}H_{20}O_4S_4$: 548.0244; found: 547.9 (M).

BT. The Grignard reagent was prepared by the following procedure: to a suspension of magnesium turnings (144 mg, 6 mmol) and 3-4 drops of 1,2-

dibromoethane in dry THF (6 mL) was slowly added 1-bromo-4-hexylbenzene (1.44 g, 6 mmol) dropwise, and the mixture was stirred for 2 h. BT was synthesized by two steps: firstly, to a solution of compound **2** (280 mg, 0.5 mmol) in dry THF (10 mL) under nitrogen was added the prepared Grignard reagent dropwise at room temperature. The resulting mixture was stirred at reflux for 12 h. 100 mL of ammonium chloride saturated solution was added and the mixture was extracted with dichloromethane (2 × 100 mL). The combined organic layer was dried over anhydrous $MgSO_4$. After removal of the solvent under reduced pressure, a brown solid was obtained and directly used for next step reaction without further purification. Secondly, to a solution of the brown solid in octane (50 mL) was added acetic acid (10 mL) and sulfuric acid (0.05 mL) slowly. The resulting solution was stirred at 70 °C for 5 h. After removal of the octane under reduced pressure, the residue was washed with sodium carbonate saturated aqueous solution (50 mL × 3) and extracted by diethyl ether (50 mL × 2). Then, after removal of the solvent under reduced pressure, the residue was purified by column chromatography on silica gel using a mixture solvent as eluent (petroleum ether/dichloromethane, v/v = 10/1) to give a yellow solid (333 mg, 2 steps overall yield 61%). Pure ^{1}H NMR and ^{13}C NMR were not obtained because the product cannot be separated with a small amount of isomeric compound. MS (MALDI-TOF) calcd for $C_{72}H_{76}S_4$: 1068.4830; found: 1069.4 (M^+).

BT-CHO. A Vilsmeier reagent was prepared firstly. 4 mL anhydrous DMF was added to a dry 100 mL two-necked round bottom flask, and the solution was cooled to 0 °C and stirred when 0.4 mL phosphorous oxychloride ($POCl_3$) was added by syringe under argon protection. The mixture kept at 0 °C for 2 h, and then BT (255 mg, 0.2 mmol) in dry dichloroethane (DCE) (20 mL) was added. Then, the mixture solution was allowed to reflux overnight. After cooling to room temperature, 100 mL water was added to quench the reaction. The mixture was extracted with dichloromethane, and the organic layer was collected, washed with water and dried with anhydrous $MgSO_4$. After removal of the solvent under reduced pressure, the residue was purified by column chromatography on silica gel using a mixture solvent as eluent (petroleum ether/dichloromethane, v/v = 1/1) to give a yellow solid (233 mg, 87%). ^1H NMR (400 MHz, CD_2Cl_2): δ 9.82 (s, 2H), 7.85 – 7.60 (m, 6H), 7.16 (d, J = 8.2 Hz, 8H), 7.10 (d, J = 8.1 Hz, 8H), 2.56 (t, J = 7.8 Hz, 8H), 1.60 – 1.50 (m, 8H), 1.35 – 1.26 (m, 24H), 0.87 – 0.81 (m, 12H). ^{13}C NMR (100 MHz, CD_2Cl_2): δ 183.2, 158.5, 150.9, 150.1, 146.9, 143.1, 139.2, 137.5, 136.3, 133.8, 133.6, 132.0, 129.1, 122.6, 119.0, 64.3, 36.0, 32.3, 31.9, 29.7, 23.2, 14.4. MS (MALDI-TOF) calcd for $C_{74}H_{76}O_2S_4$: 1124.4728; found: 1125.5 (M^+). Anal. calcd for $C_{74}H_{76}O_2S_4$: C, 78.96; H, 6.81. Found: C, 78.87; H, 6.77.

BTIC. To a three-necked round bottom flask were added BT-CHO (113 mg, 0.1 mmol), 2FIC (70 mg, 0.3 mmol), pyridine (0.15 mL) and chloroform (20 mL). The mixture was deoxygenated with nitrogen for 20 min and then stirred at reflux for 12 h. After cooling to room temperature, the mixture was poured into methanol (200 mL) and filtered. The residue was purified by column chromatography on silica gel using a mixture solvent as eluent (petroleum ether/dichloromethane, v/v = 1/1) to give a blue solid (120 mg, 77%). ^1H NMR (500 MHz, $CDCl_3$): δ 8.87 (s, 2H), 8.54 (dd, J = 9.7, 6.1 Hz, 2H), 7.90 (d, J = 8.2 Hz, 2H), 7.83 (d, J = 8.2 Hz, 2H), 7.76 (s, 2H), 7.69 (t, J = 7.4

Hz, 2H), 7.13 (d, *J* = 8.0 Hz, 8H), 7.08 (d, *J* = 8.1 Hz, 8H), 2.55 (t, *J* = 7.8 Hz, 8H), 1.62 – 1.53 (m, 8H), 1.32 – 1.23 (m, 24H), 0.90 – 0.81 (m, 12H). ^{13}C NMR (125 MHz, $CDCl_3$): δ 186.2, 160.8, 159.3, 158.5, 155.7, 155.6, 153.6, 153.5, 151.2, 142.8, 140.8, 139.8, 139.3, 139.0, 137.3, 136.6, 134.7, 134.5, 133.5, 128.8, 128.7, 128.6, 122.9, 121.2, 119.5, 115.3, 115.1, 114.4, 114.3, 112.9, 112.7, 69.7, 63.8, 35.7, 31.8, 31.4, 29.2, 22.7, 14.2. HRMS (MALDI) calcd for $C_{98}H_{80}F_4N_4O_2S_4$: 1549.5134; found: 1549.5142. Anal. calcd for $C_{98}H_{80}F_4N_4O_2S_4$: C, 75.94; H, 5.20; N, 3.61. Found: C, 75.99; H, 5.17; N, 3.66.

Compound 4. Compound **4** was synthesized according to the synthetic procedure of compound **2**. Compound **4** was obtained as a yellow solid (430 mg, 78%). This compound was directly used for next step reaction without further purification due to limited solubility. MS (MALDI-TOF) calcd for $C_{28}H_{20}O_4S_4$: 548.0244; found: 548.1 (M).

NT. Compound NT was synthesized according to the synthetic procedure of BT. Compound NT was obtained as a yellow solid (300 mg, 2 steps overall yield 55%). ^{1}H NMR (400 MHz, CD_2Cl_2): δ 7.49 (d, *J* = 11.9 Hz, 4H), 7.33 (d, *J* = 5.2 Hz, 4H), 7.24 (d, *J* = 7.7 Hz, 8H), 7.12 (d, *J* = 7.7 Hz, 8H), 2.59 (t, *J* = 7.9 Hz, 8H), 1.66 – 1.56 (m, 8H), 1.41 – 1.32 (m, 24H), 0.97 – 0.88 (m, 12H). ^{13}C NMR (100 MHz, CD_2Cl_2): δ 161.5, 152.6, 142.6, 141.6, 140.0, 137.3, 136.7, 132.6, 129.5, 129.1, 128.7, 127.5, 126.5, 123.5, 121.6, 121.1, 63.2, 36.1, 32.3, 32.0, 29.7, 23.2, 14.5. MS (MALDI-TOF) calcd for $C_{72}H_{76}S_4$: 1068.4830; found: 1069.5 (M^+). Anal. calcd for $C_{77}H_{76}S_4$: C, 80.85; H, 7.16. Found: C, 80.86; H, 7.11.

NT-CHO. Compound NT-CHO was synthesized according to the synthetic procedure of compound BT-CHO. Compound NT-CHO was obtained as a yellow solid (235 mg, 88%). ^{1}H NMR (300 MHz, CD_2Cl_2): δ 9.84 (s, 2H), 7.79 (s, 2H), 7.25 (d, J = 7.8 Hz, 10H), 7.16 (d, J = 8.1 Hz, 8H), 6.97 (d, J = 10.9 Hz, 2H), 2.56 (t, J = 7.7 Hz, 8H), 1.64 – 1.52 (m, 8H), 1.33 – 1.25 (m, 24H), 0.91 – 0.84 (m, 12H). ^{13}C NMR (75 MHz, CD_2Cl_2): δ 183.0, 161.5, 156.5, 147.1, 145.7, 143.4, 143.1, 138.7, 135.9, 132.3, 131.7, 129.3, 128.6, 126.5, 122.2, 121.5, 63.4, 36.0, 32.2, 32.0, 29.7, 23.2, 14.4. MS (MALDI-TOF) calcd for $C_{74}H_{76}O_2S_4$: 1124.4728; found: 1125.5 (M^+). Anal. calcd for $C_{74}H_{76}O_2S_4$: C, 78.96; H, 6.81. Found: C, 78.94; H, 6.79.

NTIC. Compound NTIC was synthesized according to the synthetic procedure of compound BTIC. Compound NTIC was obtained as a blue solid (126 mg, 81%). ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.88 (s, 2H), 8.33 (s, 2H), 7.76 (s, 2H), 7.57 (t, J = 7.3 Hz, 2H), 7.39 (s, 2H), 7.26 – 7.02 (m, 18H), 2.59 (t, J = 7.9 Hz, 8H), 1.63 – 1.55 (m, 8H), 1.29 – 1.20 (m, 24H), 0.89 – 0.82 (m, 12H). ^{13}C NMR (125 MHz, $CDCl_3$): δ 185.9, 162.0, 158.8, 157.9, 157.6, 155.5, 155.4, 153.4, 153.3, 145.1, 143.0, 140.2, 139.5, 138.5, 137.8, 136.3, 134.4, 132.2, 129.0, 128.3, 126.6, 121.9, 121.7, 120.0, 115.0, 114.8, 114.5, 114.4, 112.6, 112.4, 68.8, 63.2, 35.7, 31.8, 31.5, 29.2, 22.7, 14.2. HRMS (MALDI) calcd for $C_{98}H_{80}F_4N_4O_2S_4$: 1549.5134; found: 1549.5137. Anal. calcd for $C_{98}H_{80}F_4N_4O_2S_4$: C, 75.94; H, 5.20; N, 3.61. Found: C, 75.85; H, 5.18; N, 3.57.

Molecular modelling

Density functional theory (DFT) calculations were performed with the Gaussian 09 program,[S2] using the B3LYP functional.[S3, S4] All-electron double-ξ valence basis sets with polarization functions 6-31G* were used for all atoms.[S5] Geometry optimizations were performed with full relaxation of all atoms in gas phase without solvent effects. Vibration frequency calculation was performed to check that the stable structures had no imaginary frequency. Charge distribution of the molecules was calculated by Mulliken population analysis.

Characterization

The ^{1}H NMR and ^{13}C NMR spectra were measured by Bruker AVANCE 300, 400 or 500 MHz spectrometer. Mass spectra were performed by Bruker Daltonics Biflex III MALDI-TOF Analyzer in the MALDI mode. Elemental analyses were measured through a Flash EA 1112 elemental analyzer. Thermogravimetric analysis (TGA) measurements were carried out using a Shimadzu thermogravimetric analyzer (Model DTG-60) under flowing nitrogen gas at a heating rate of 10 °C min^{-1}. The ultraviolet-visible light (UV-vis) absorption spectra were measured using the JASCO-570 spectrophotometer (JASCO. Inc., Japan) in solution (chloroform) and the thin film (on a quartz substrate). Electrochemical measurements were carried out under nitrogen in a solution of tetra-*n*-butylammonium hexafluorophosphate ($[^{n}Bu_4N]^{+}[PF_6]^{-}$) (0.1 M) in CH_3CN employing a computer-controlled CHI660C electrochemical workstation, a glassy carbon working electrode coated with BTIC or NTIC film, a Ag/AgCl reference electrode, and a platinum-wire auxiliary electrode. The potentials were referenced to a ferrocenium/ferrocene ($FeCp_2^{+/0}$) couple using ferrocene as an external standard. Atomic force microscope (AFM) images were measured on Multimode 8 scanning probe microscopy (Bruker Daltonics, United States) in the tapping mode.

GIWAXS measurements

GIWAXS measurements were accomplished with a Xeuss 2.0 SAXS/WAXS laboratory beamline using a Cu X-ray source (8.05 keV, 1.54 Å) and a Pilatus3R 300K detector. The incidence angle is 0.2^{o}.

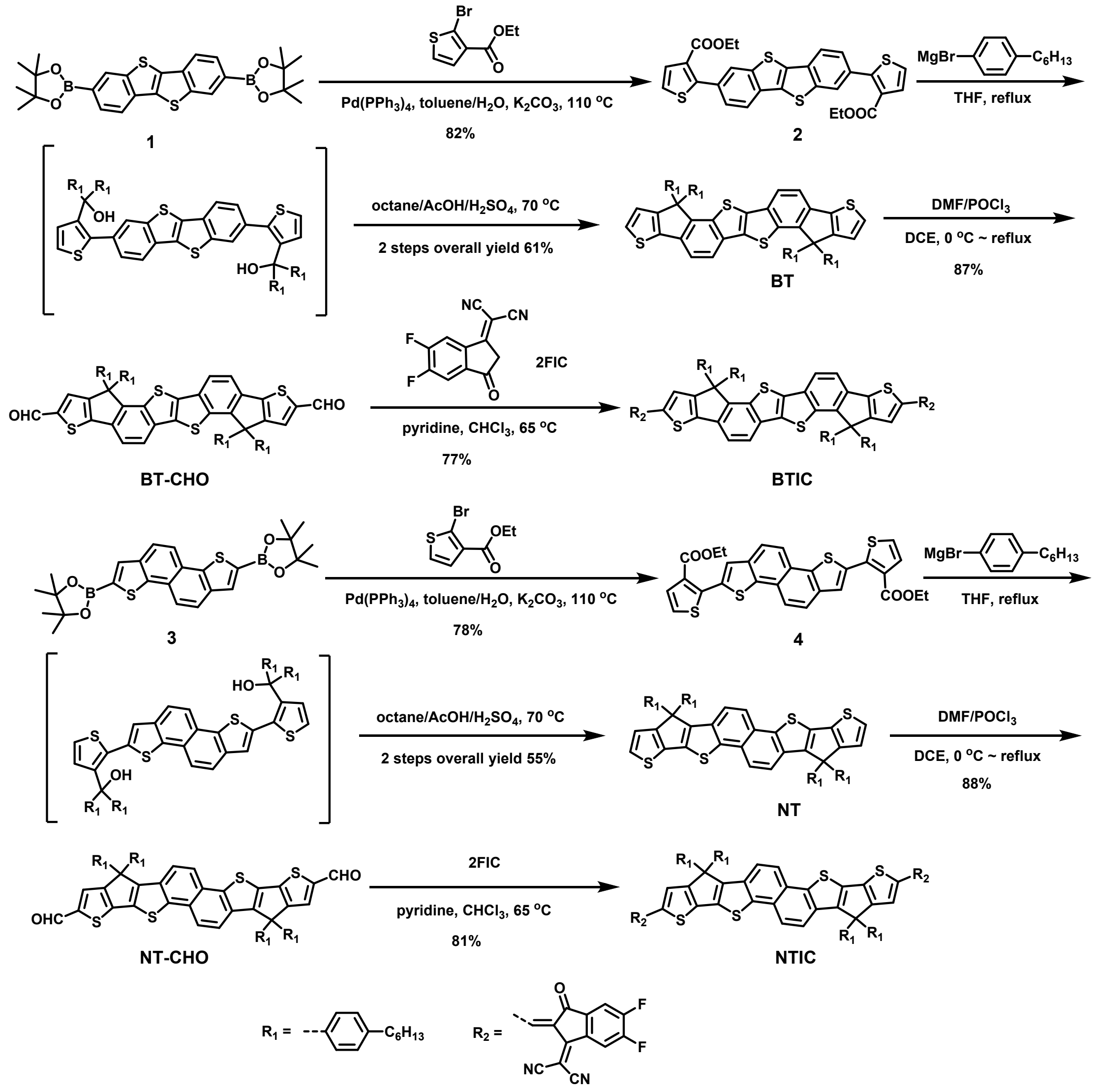
Device fabrication and characterization

All the devices are based on an inverted sandwich structure, the patterned indium-tin oxide glass (ITO glass)/zinc oxide (ZnO) (*ca.* 30 nm)/PM6:acceptor/MoO_3 (*ca.* 5 nm)/Ag (*ca.* 80 nm). First, ITO glass (sheet resistance = 15 $\Omega\cdot\square^{-1}$) was continuously pre-cleaned in the ultrasonic bath with de-ionized water, acetone and isopropanol. Then, a ZnO layer was spin-coated at 4000 $r\cdot min^{-1}$ onto the ITO glass from ZnO precursor solution (prepared by dissolving 0.1 g of zinc acetate dihydrate ($Zn(CH_3COO)_2\cdot 2H_2O$) and 0.03 mL of ethanolamine ($NH_2CH_2CH_2OH$) in 1 mL of 2-methoxyethanol ($CH_3OCH_2CH_2OH$)), followed by baking at 200 °C for 30 min. The active layer blend was spin-coated at 2000 $r\cdot min^{-1}$ onto the ZnO layer to form the photoactive layer. A MoO_3 layer and Ag layer were then evaporated under vacuum (*ca.* 10^{-5} Pa) to form the anode electrode. The *J*-*V* curves were measured using a computer-controlled B2912A Precision Source/Measure Unit (Agilent Technologies, United States). An XES-70S1 (SAN-EI Electric Co., Ltd., Japan) solar simulator (AAA grade, 70 mm × 70 mm) coupled with AM 1.5G solar spectrum filters was used as the light source, and the optical power at the sample was 100 mW cm^{-2}. The measured area of the active device was 4 mm^2. A 2 cm × 2 cm monocrystalline silicon reference cell (SRC-1000-TC-QZ) was purchased from VLSI Standards Inc. The external quantum efficiency (EQE) spectra were measured using a Solar Cell Spectral Response Measurement System QE-R3011 (Enlitech Co., Ltd.). The light intensity at each wavelength was calibrated through standard single crystal Si photovoltaic cell.

SCLC measurements

Hole-only or electron-only devices were fabricated as follows, ITO/PEDOT:PSS/active layer/Au for holes and ITO/ZnO/active layer/Ca/Al for electrons. The mobility was extracted by fitting the *J-V* curves using space charge limited current (SCLC) method,[S6] which follows $J = (9/8)\mu\varepsilon_r\varepsilon_0 V^2\exp(0.89(V/E_0L)^{0.5})/L^3$.

Here, J refers to the current density, μ is hole or electron mobility, ε_r is relative dielectric constant of the transport medium, which is equal to 3, ε_0 is the permittivity of free space (8.85×10^{-12} F m^{-1}), $V = V_{\mathrm{appl}} - V_{\mathrm{bi}}$, where V_{appl} is the applied voltage to the device, and V_{bi} is the built-in voltage due to the difference in work function of the two electrodes (for hole-only diodes, V_{bi} is 0.2 V; for electron-only diodes, V_{bi} is 0 V). E_0 is characteristic field, L is the thickness of the active layer and was measured by Dektak XT (Bruker).



Scheme S1 Synthetic routes to BTIC and NTIC.

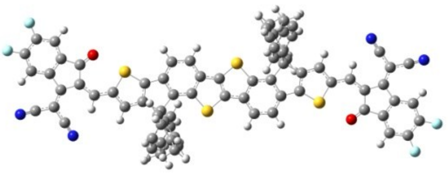
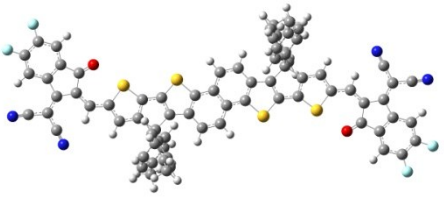
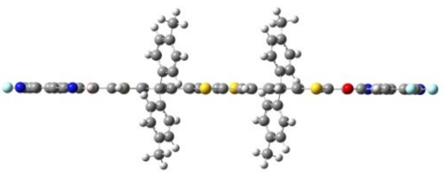
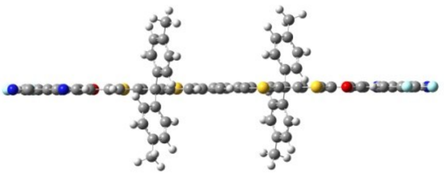
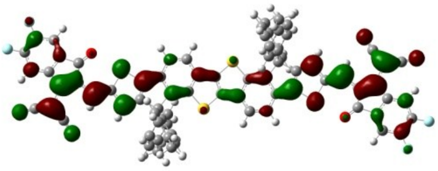
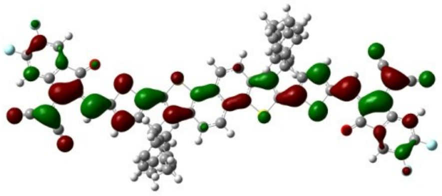
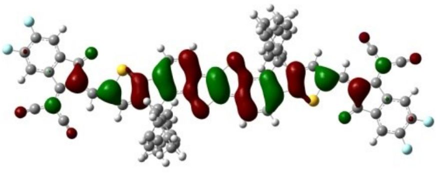
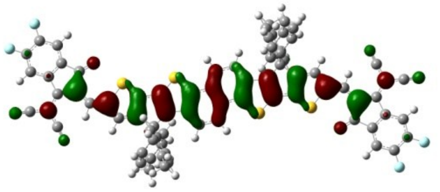
	BTIC	NTIC
top view		
side view		
LUMO	 –3.40 eV	 –3.42 eV
HOMO	 –5.62 eV	 –5.56 eV

Fig. S1 The optimal geometries and LUMO and HOMO distribution of BTIC and NTIC (alkyls are simplified to methyl) calculated at B3LYP/6-31G* level.

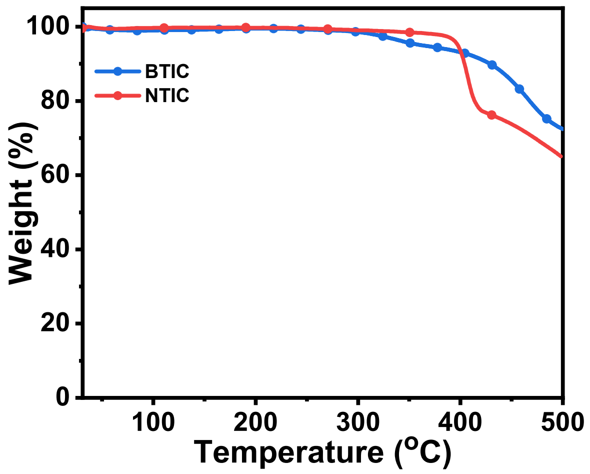


Fig. S2 TGA curves of BTIC and NTIC.

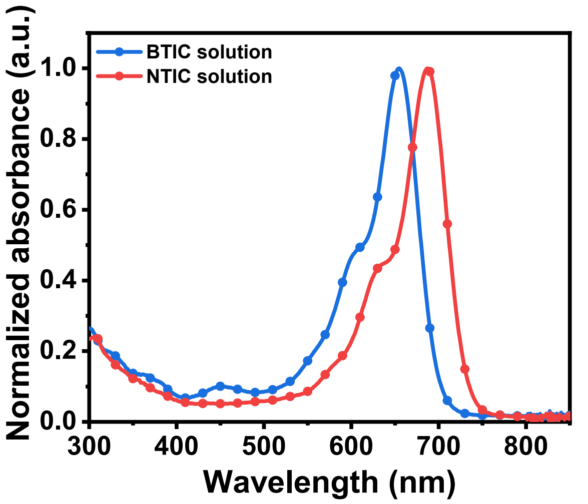


Fig. S3 BTIC and NTIC absorption spectra in chloroform solution.

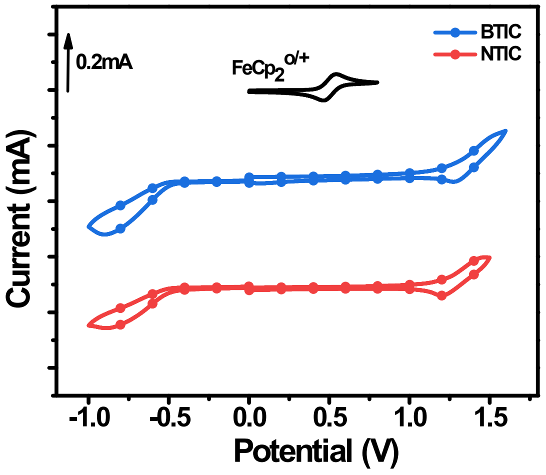


Fig. S4 Cyclic voltammograms for BTIC and NTIC in CH_3CN/0.1 M Bu_4NPF_6 at 100 mV s^{-1}; the horizontal scale refers to an Ag/AgCl electrode as a reference electrode.

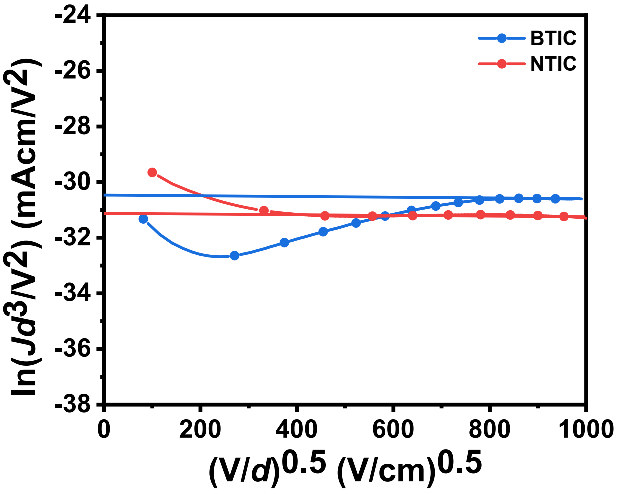


Fig. S5 *J-V* characteristics in the dark for electron-only devices based on BTIC and NTIC.

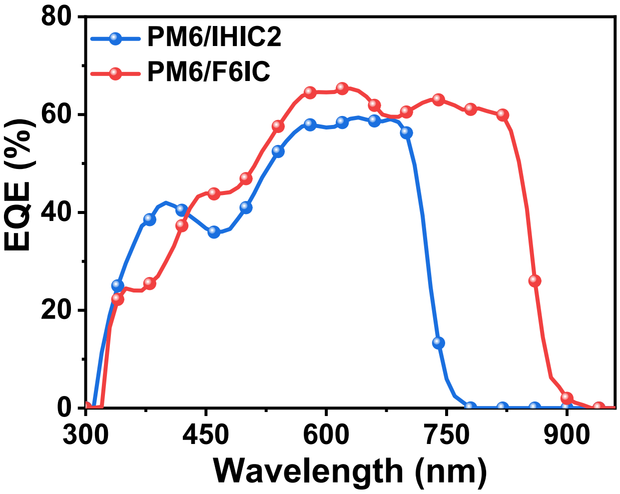


Fig. S6 EQE spectra of the optimized binary-blend OSCs.

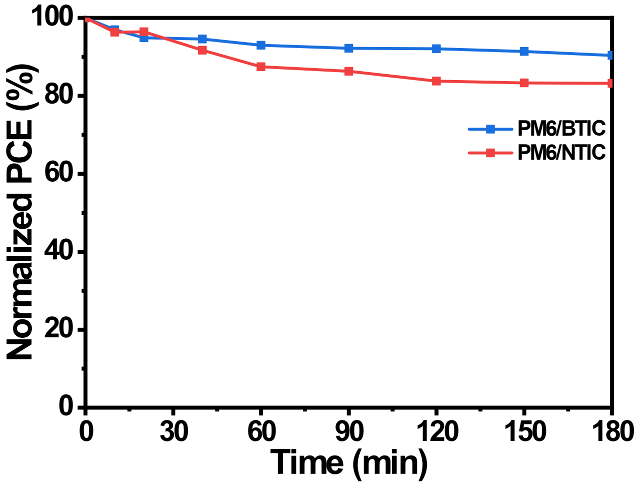


Fig. S7 Thermal stability curves of OSCs under continuous heating at 80 °C for 180 min.

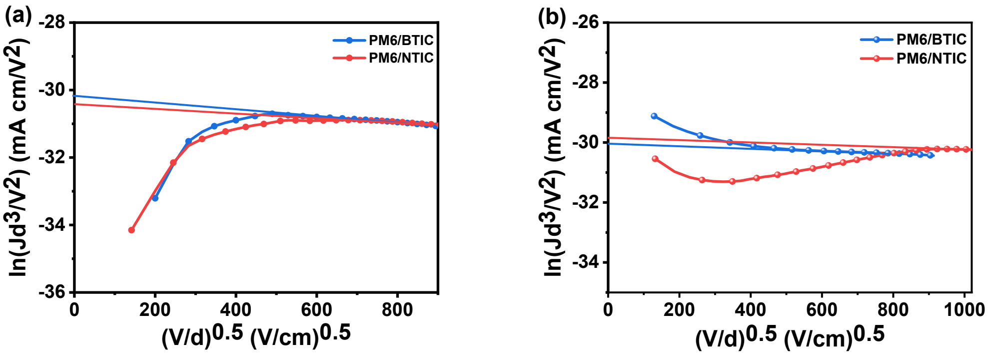


Fig. S8 *J-V* characteristics in the dark for (a) hole-only and (b) electron-only devices based on PM6/acceptor blends.

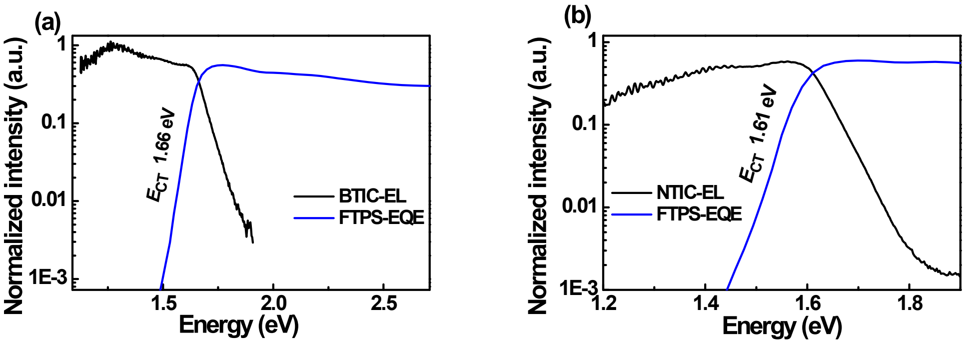


Fig. S9 Electroluminescence and FTPS-EQE spectra of (a) BTIC and (b) NTIC-based OSCs.

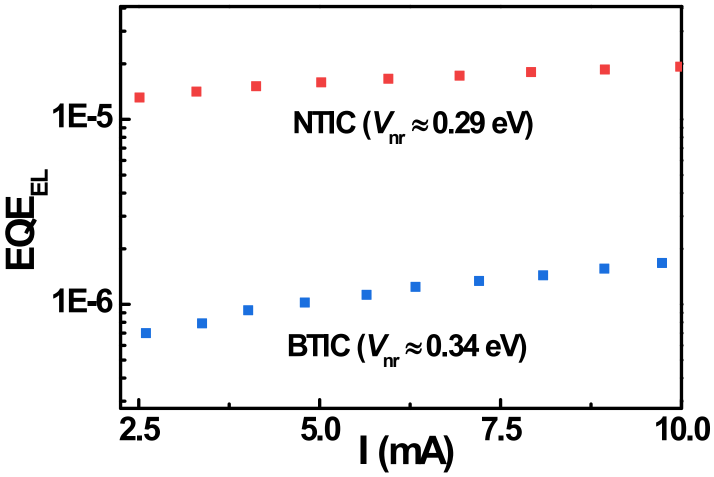


Fig. S10 Electroluminescence external quantum efficiency of the BTIC and NTIC-based OSCs (V_{nr}: non-radiative recombination loss).

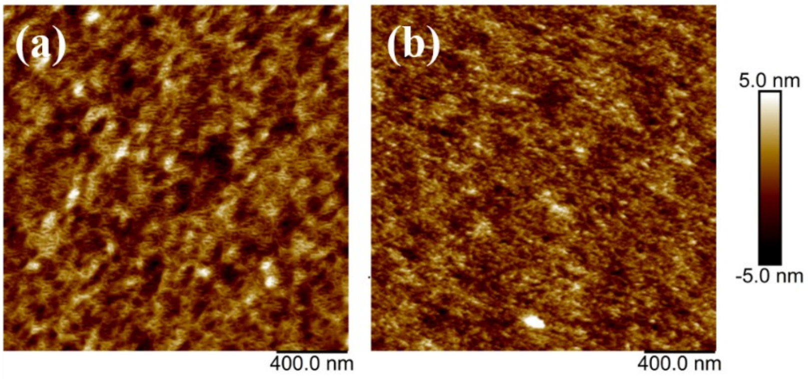


Fig. S11 AFM height images of (a) PM6/BTIC and (b) PM6/NTIC blends under optimized device conditions.

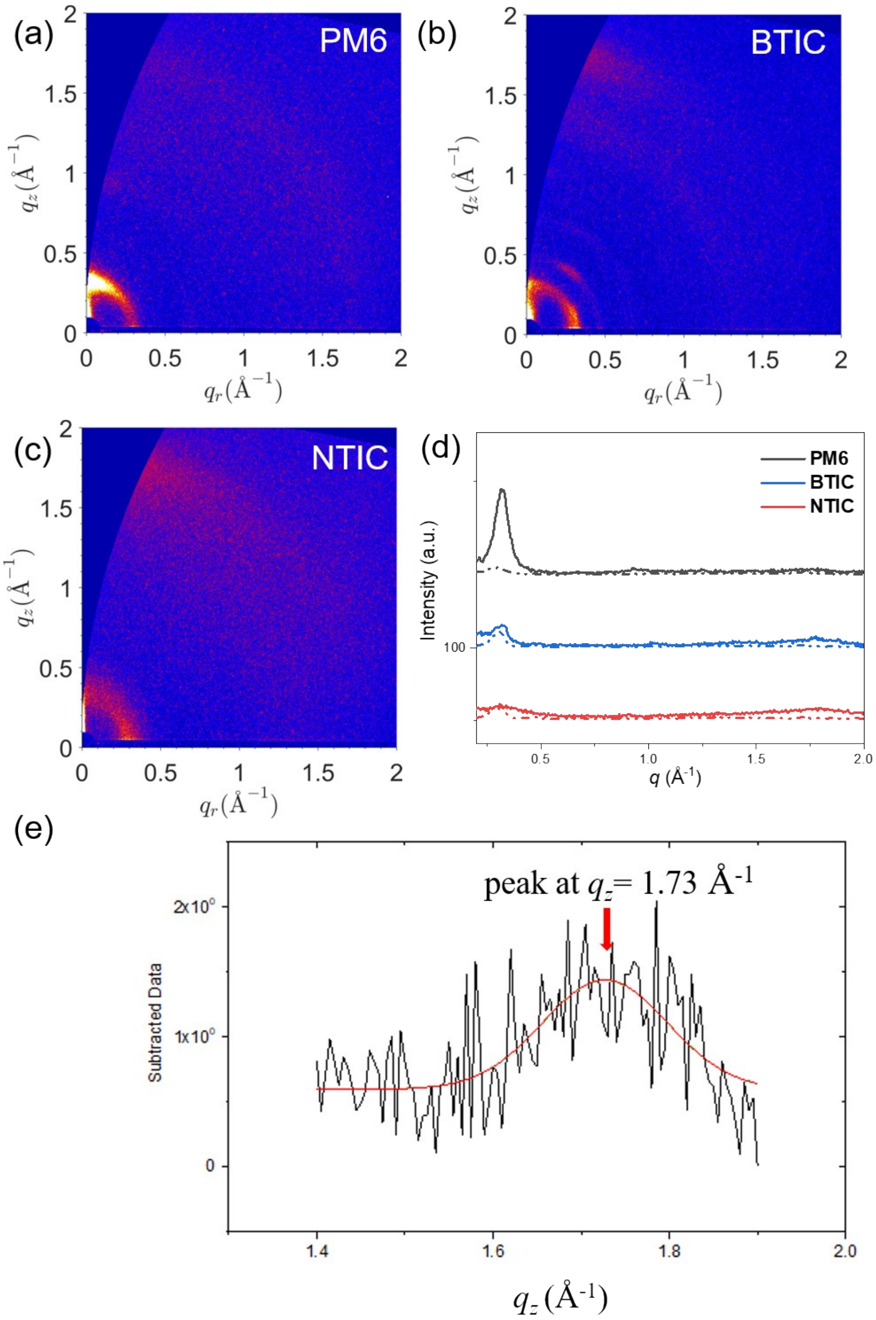


Fig. S12 2D GIWAXS patterns of (a) PM6, (b) BTIC and (c) NTIC pure films; (d) the corresponding intensity profiles along the in-plane (dashed line) and out-of-plane (solid line) directions; (e) π-π peak fitting based on the out-of-plane line-cut of PM6 film.

Table S1. Device data of as-cast OSCs based on PM6/acceptor with different D/A ratio.

acceptor	D/A (w/w)	V_{OC}^a (V)	J_{SC}^a (mA cm^{-2})	FF^a (%)	PCE^a (%)
BTIC	1.2/1	0.979 (0.982±0.003)	13.3 (12.6±0.3)	67.6 (68.7±2.1)	8.79 (8.52±0.29)
	1/1	0.989 (0.986±0.003)	13.3 (13.2±0.1)	71.1 (69.7±1.4)	9.34 (9.16±0.18)
	1/1.2	0.979 (0.979±0.003)	11.8 (11.3±0.4)	68.9 (69.2±1.2)	7.97 (7.67±0.23)
NTIC	1.2/1	0.946 (0.944±0.003)	13.7 (13.4±0.3)	69.9 (69.0±1.1)	9.07 (8.73±0.22)
	1/1	0.943 (0.948±0.004)	15.4 (14.7±0.4)	68.4 (68.8±1.1)	9.91 (9.55±0.20)
	1/1.2	0.948 (0.949±0.004)	13.1 (12.9±0.2)	70.8 (69.7±1.7)	8.80 (8.77±0.28)

[a] Average values (in parenthesis) are obtained from 20 devices.

Table S2. Device data of the OSCs based on PM6/acceptor (D/A=1/1) with different DIO content in chloroform.

acceptor	DIO (%)	V_{OC}[a] (V)	J_{SC}[a] (mA cm^{-2})	FF[a] (%)	PCE[a] (%)
BTIC	0.1	0.961 (0.965±0.004)	15.0 (14.6±0.2)	70.5 (67.8±1.7)	10.2 (9.51±0.28)
	0.2	0.962 (0.951±0.008)	16.6 (16.1±0.3)	71.9 (69.8±1.1)	11.5 (10.7±0.4)
	0.3	0.939 (0.936±0.004)	16.4 (16.2±0.2)	69.2 (68.3±0.8)	10.7 (10.4±0.2)
NTIC	0.2	0.941 (0.936±0.003)	18.0 (17.8±0.2)	71.6 (70.1±0.8)	12.2 (11.7±0.2)

[a] Average values (in parenthesis) are obtained from 20 devices.

Table S3. Charge mobilities of pure and blend films measured by SCLC method.

active layer	μ_h (cm^2 V^{-1} s^{-1})	μ_e (cm^2 V^{-1} s^{-1})	μ_h/μ_e
BTIC	-	3.4×10^{-4}	-
NTIC	-	1.8×10^{-4}	-
PM6/BTIC	4.6×10^{-4}	5.5×10^{-4}	0.84
PM6/NTIC	3.4×10^{-4}	6.7×10^{-4}	0.51

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