Supporting Information

Highly Efficient Fused Ring Electron Acceptors Based on a New Undecacyclic Core

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1. General characterization

¹H and ¹³C NMR spectra were measured on a Bruker Advance-400 spectrometer. Absorption spectra were recorded on a Lambda 950 spectrophotometer (PerkinElmer, U.S.A.). Cyclic voltammetry was done by using a Shanghai Chenhua CHI660C voltammetric analyzer under argon in a acetonitrile solution of tetra-*n*-butylammonium hexafluorophosphate (0.1 M). A glassy-carbon electrode was used as the working electrode, a platinum-wire was used as the counter electrode, and a Ag/Ag⁺ electrode was used as the reference electrode. The polymer and acceptors were coated onto glassy-carbon electrode and all potentials were corrected against Fc/Fc⁺. AFM was performed on a Dimension 3100 microscope (Veeco) using tapping mode. TEM was performed on a Tecnai G2 T20 U-TWIN operated at 200 kV. GIWAXS measurement was performed at beamline 7.3.3¹ at the Advanced Light Source, Berkeley, U.S.A, with a Dectris Pilatus 2 M photon counting detector to detect the scattered X-ray. GIWAXS samples were prepared on Si substrates coated with ZnO. Two dimensional scattering patterns were collected on an in-vacuum CCD camera (Princeton Instrument PI-MTE). The beam size at the 5 samples is 100 mm.

2. Synthesis

All reagents were purchased from J&K Co., Innochem Co., HWRK Chem Co. and other commercial suppliers. Dithieno[3,2-*b*:2',3'-*d*]thiophene, (4,8-bis((2-ethylhexyl)oxy)benzo[1,2-*b*:4,5-*b*']dithiophene-2,6-diyl)bis(trimethylstannane) and (4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-*b*:4,5-*b*']dithiophene-2,6-diyl)bis(trimethylstannane) were purchased from SunaTech Inc.. All reactions dealing with air- or moisture-sensitive compounds were carried out using standard Schlenk techniques. The compound **1**, ethyl 2-bromodithieno[3,2-*b*:2',3'-*d*]thiophene-3-carboxylate, was prepared according to the literature.²⁻³



Scheme S1. The synthetic routes for IUIC-O and IUIC-T.

Ethyl 2-bromodithieno[3,2-*b***:2',3'-***d***]thiophene-3-carboxylate. ¹H NMR (CDCl₃, 400 MHz, δ/ppm): 7.40 (d,** *J* **= 5.2 Hz, 1H), 7.31 (d,** *J* **= 5.2 Hz, 1H), 4.46 (q,** *J* **= 7.1 Hz, 2H), 1.49 (t,** *J* **= 7.1 Hz, 3H). ¹³C NMR (CDCl₃, 100 MHz, δ/ppm): 160.92, 142.59, 140.52, 130.15, 128.87, 126.85, 125.01, 120.65, 119.22, 61.59, 14.24.**

Diethyl 2,2'-(4,8-bis((2-ethylhexyl)oxy)benzo[1,2-b:4,5-b']dithiophene-2,6diyl)bis(dithieno[3,2-b:2',3'-d]thiophene-3-carboxylate) (Compound 3a). To a solution ethyl 2-bromodithieno[3,2-b:2',3'-d]thiophene-3-carboxylate (730 mg, 2.1 mmol) and (4,8bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b']dithiophene-2,6-

diyl)bis(trimethylstannane) (772 mg, 1.0 mmol) in fresh distilled toluene (20 mL) in a twonecked flask was added Pd(PPh₃)₄ (65 mg) under Ar. The mixture was heated to reflux and stirred overnight. Then it was cooled down to room temperature and added to 150 mL methanol dropwise. The precipitate collected to give diethyl 2,2'-(4,8-bis((2was ethylhexyl)oxy)benzo[1,2-b:4,5-b']dithiophene-2,6-diyl)bis(dithieno[3,2-b:2',3'-d]thiophene-3-carboxylate) without further purification (964 mg, 98%). ¹H NMR (CDCl₃, 400 MHz, δ /ppm): 7.96 (s, 2H), 7.44 (d, J = 4.7 Hz, 2H), 7.35 (d, J = 5.2 Hz, 2H), 4.45 (g, J = 7.1 Hz, 4H), 4.35 (s, 4H), 1.75 (s, 2H), 1.46-1.25 (m, 22H), 0.88 (t, J = 6.7 Hz, 12H). MALDI-TOF MS (m/z): 979.3 (M + H⁺)

2,2'-(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b']dithiophene-2,6-Diethyl divl)bis(dithieno[3,2-b:2',3'-d]thiophene-3-carboxylate) (Compound 3b). To a solution ethyl 2-bromodithieno[3,2-b:2',3'-d]thiophene-3-carboxylate (730 mg, 2.1 mmol) and (4,8bis((2-ethylhexyl)oxy)benzo[1,2-*b*:4,5-*b*']dithiophene-2,6-diyl)bis(trimethylstannane) (904 mg, 1.0 mmol) in fresh distilled toluene (20 mL) in a two-necked flask was added Pd(PPh₃)₄ (66 mg) under Ar. The mixture was heated to reflux and stirred overnight. Then it was cooled down to room temperature and added to 150 mL methanol dropwise. The precipitate was diethyl 2,2'-(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5collected to give b']dithiophene-2,6-diyl)bis(dithieno[3,2-b:2',3'-d]thiophene-3-carboxylate) without further purification (1.08 g, 97%). ¹H NMR (CDCl₃, 400 MHz, δ /ppm): 8.08 (s, 2H), 7.42 (d, J = 5.2Hz, 2H), 7.37 (d, J = 3.5 Hz, 2H), 7.33 (d, J = 5.2 Hz, 2H), 6.91 (d, J = 3.4 Hz, 2H), 4.39 (q, J) = 7.1 Hz, 4H), 2.87 (d, J = 6.7 Hz, 4H), 1.74-1.64 (m, 2H), 1.47-1.29 (m, 22H), 0.93 (m, 12H). MALDI-TOF MS (m/z): 1111.6 (M + H⁺)

Compound 4a. To a solution of 1-bromo-4-hexylbenzene (1.21 g, 5 mmol) in dry THF (10 mL) was added magnesium (120 mg, 5 mmol) under argon, and the mixture was heated to reflux for \sim 1 h until magnesium disappeared. Then the Grignard reagent was added to a suspension of diethyl 2,2'-(4,8-bis((2-ethylhexyl)oxy)benzo[1,2-b:4,5-b']dithiophene-2,6-

diyl)bis(dithieno[3,2-b:2',3'-d]thiophene-3-carboxylate) (490 mg, 0.5 mmol) in THF (10 mL) at room temperature under argon. The mixture was heated to reflux overnight and then allowed to cool down to room temperature. It was poured into water (~100 mL) and extracted with CH₂Cl₂ twice. The organic phase was dried over anhydrous Na₂SO₄. After removing the solvent, the residue was resolved in octane (15 mL) and acetic acid (1.5 mL) again, and the concentrated H₂SO₄ (0.2 mL) was added dropwise. The solution was heated to reflux for 1 h and then quenched with water. It was extracted with CH₂Cl₂ twice and the combined organic phase was dried over anhydrous Na₂SO₄. After removing the solvent, the crude product was purified via column chromatography (silica gel) using petroleum ether/CH₂Cl₂ (5:1, v/v) as eluent to give a yellow solid compound 4a (345 mg, 46%). ¹H NMR (CDCl₃, 400 MHz, δ/ppm): 7.39 (d, J = 8.0 Hz, 8H), 7.27 (d, J = 5.3 Hz, 2H), 7.15 (d, J = 5.2 Hz, 2H), 7.05 (d, J = 8.0 Hz, 8H), 3.61 (s, 4H), 2.53 (t, J = 7.7 Hz, 8H), 1.63-1.48 (m, 8H), 1.43-1.14 (m, 42H), 0.96 (m, 12H), 0.86 (m, 12H). ¹³C NMR (CDCl₃, 100 MHz, δ/ppm):154.11, 149.91, 144.41, 141.71, 141.18, 137.86, 136.61, 136.22, 135.38, 134.59, 132.37, 131.44, 128.68, 128.17, 126.56, 125.41, 120.66, 73.35, 64.06, 35.57, 31.94, 31.70, 31.24, 29.65, 29.41, 29.15, 25.80, 22.77, 22.58, 14.18, 14.05, 11.24.

Compound 4b. To a solution of 1-bromo-4-hexylbenzene (1.21 g, 5 mmol) in dry THF (10 mL) was added magnesium (120 mg, 5 mmol) under argon, and the mixture was heated to reflux for ~1 h until magnesium disappeared. Then the Grignard reagent was added to a suspension of 2,2'-(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b']dithiophene-2,6diethyl diyl)bis(dithieno[3,2-b:2',3'-d]thiophene-3-carboxylate) (556 mg, 0.5 mmol) in THF (10 mL) at room temperature under argon. The mixture was heated to reflux overnight and then allowed to cool down to room temperature. It was poured into water (~100 mL) and extracted with CH₂Cl₂ twice. The organic phase was dried over anhydrous Na₂SO₄. After removing the solvent, the residue was resolved in octane (15 mL) and acetic acid (1.5 mL) again, and the concentrated H₂SO₄ (0.2 mL) was added dropwise. The solution was heated to reflux for 1 h and then quenched with water. It was extracted with CH₂Cl₂ twice and the combined organic phase was dried over anhydrous Na₂SO₄. After removing the solvent, the crude product was purified via column chromatography (silica gel) using petroleum ether/CH₂Cl₂ (5:1, v/v) as eluent to give a yellow solid compound **4b** (220 mg, 27%). ¹H NMR (CDCl₃, 400 MHz, δ/ppm): 7.10 (s, 2H), 7.03-6.90 (m, 18H), 6.46 (s, 2H), 6.13 (dd, *J* = 3.04, 7.49 Hz, 2H), 2.80-2.69 (m, 4H), 2.54 (d, J = 6.34 Hz, 8H), 1.58 (s, 10H), 1.38-1.27 (m, 40H), 0.99 (m, 12H), 0.88 (m, 12 12H).¹³C NMR (CDCl₃, 100 MHz, δ/ppm): 149.53, 146.28, 146.22, 141.34, 141.13, 141.09, 136.55, 135.25, 135.18, 134.87, 130.30, 128.46, 128.43, 128.40, 128.38, 128.22, 128.15,

128.01, 124.46, 124.33, 120.53, 63.63, 41.52, 41.29, 35.62, 35.50, 34.21, 34.07, 32.52, 31.77, 31.45, 29.70, 29.21, 29.16, 28.88, 28.83, 25.76, 25.68, 23.24, 23.15, 22.65, 22.63, 14.32, 14.29, 14.11, 10.93, 10.79.

Compound 5a. To a solution of compound **4a** (300 mg, 0.20 mmol) in dry THF (20 mL) was added 1.6M *n*-butyllithium (0.50 mL, 0.8 mmol) at -78 °C under argon, and it stirred at the temperature for ~1 h. Then the solution was warmed to -50 °C. Anhydrous DMF (0.50 mL) was added to the solution and it continued to stir at -50 °C for ~1 h. Then it was quenched with water and extracted with CH₂Cl₂ several times. The organic phase was dried over anhydrous Na₂SO₄. After removing the solvent, the crude product was purified via column chromatography (silica gel) using petroleum ether/CH₂Cl₂ (1:1, v/v) as eluent to give a red solid compound **5a** (283 mg, 91%). ¹H NMR (CDCl₃, 400 MHz, δ /ppm): 9.86 (s, 2H), 7.75 (s, 2H), 7.37 (d, *J* = 8.2 Hz, 8H), 7.06 (d, *J* = 8.2 Hz, 8H), 3.63 (s, 4H), 2.53 (m, 8H), 1.61-1.49 (m, 10H), 1.38-1.13 (m, 40H), 0.95 (m, 12H), 0.85 (m, 12H).¹³C NMR (CDCl₃, 100 MHz, δ /ppm): 182.47, 153.91, 151.65, 144.77, 142.97, 142.11, 140.77, 140.48, 139.54, 138.56, 137.70, 135.97, 134.92, 132.16, 129.98, 128.56, 128.33, 126.79, 73.51, 64.16, 35.55, 31.93, 31.67, 31.23, 29.63, 29.39, 29.12, 25.79, 22.76, 22.57, 14.17, 14.04, 11.22.

Compound 5b. To a solution of compound **4b** (200 mg, 0.12 mmol) in dry THF (15 mL) was added 1.6M *n*-butyllithium (0.30 mL, 0.48 mmol) at -78 °C under argon, and it stirred at the temperature for ~1 h. Then the solution was warmed to -50 °C. Anhydrous DMF (0.30 mL) was added to the solution and it continued to stir at -50 °C for ~1 h. Then it was quenched with water and extracted with CH₂Cl₂ several times. The organic phase was dried over anhydrous Na₂SO₄. After removing the solvent, the crude product was purified via column chromatography (silica gel) using petroleum ether/CH₂Cl₂ (1:1, v/v) as eluent to give a red solid compound **5b** (177 mg, 87%). ¹H NMR (CDCl₃, 400 MHz, δ /ppm): 9.82 (s, 2H), 7.71 (s, 2H), 7.04-6.93 (m, 16H), 6.47 (s, 2H), 6.13 (m, 2H), 2.75 (m, 4H), 2.55 (d, *J* = 6.4 Hz 8H), 1.58 (s, 8H), 1.36 (m, 42H), 0.98 (m, 12H), 0.87 (m, 12H). ¹³C NMR (CDCl₃, 100 MHz, δ /ppm): 182.44, 155.69, 152.31, 150.11, 146.63, 146.57, 142.97, 141.72, 141.51, 141.47, 140.49, 139.54, 139.01, 138.52, 136.14, 134.58, 134.51, 135.25, 130.84, 130.44, 129.82, 128.39, 128.16, 128.08, 128.01, 125.16, 124.53, 124.40, 63.61, 41.52, 41.30, 35.60, 35.48, 34.18, 34.03, 32.49, 31.74, 31.44, 29.18, 29.13, 28.86, 28.81, 25.76, 25.65, 23.23, 23.14, 22.64, 22.62, 14.32, 14.29, 14.10, 10.91, 10.79.

IUIC-O. To a solution of compound **5a** (80 mg, 51 μ mol) in CHCl₃ (10 mL) were added 2- (5,6-difluoro-3-oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile (59 mg, 0.25 mmol) and pyridine (1 mL). It was heated to reflux for 1 h and then cooled down to room temperature. The

solution was poured into column chromatography (silica gel) directly and the crude product was purified by using CHCl₃ as eluent to give a dark solid IUIC-O (86 mg, 85%). ¹H NMR (CDCl₃, 400 MHz, δ /ppm): 8.87 (s, 2H), 8.52 (dd, *J* = 9.8, 6.6 Hz, 2H), 7.90 (s, 2H), 7.66 (t, *J* = 7.5, 4H), 7.37 (d, *J* = 8.2 Hz, 8H), 7.08 (d, *J* = 8.2 Hz, 8H), 3.65 (s, 4H), 2.54 (m, 8H), 1.56 (m, 8H), 1.37-1.18 (m, 42H), 0.96 (t, *J* = 6.8 Hz, 12H), 0.85 (t, *J* = 6.4, 12H). ¹³C NMR (CDCl₃, 100 MHz, δ /ppm): 186.06, 158.12, 155.75, 154.33, 153.30, 146.74, 145.14, 143.31, 143.15, 142.70, 142.37, 138.08, 137.75, 137.72, 136.63, 135.50, 135.25, 134.46, 132.59, 128.50, 128.46, 127.13, 120.98, 115.02, 114.82, 114.32, 114.27, 112.68, 112.48, 73.65, 69.43, 64.25, 35.55, 31.93, 31.67, 31.23, 29.71, 29.62, 29.40, 29.11, 25.79, 22.76, 22.57, 14.18, 14.04. MALDI-TOF MS (m/z): 1979.7 (M⁺).

IUIC-T. To a solution of compound **5b** (80 mg, 47 µmol) in CHCl₃ (10 mL) were added 2-(5,6-difluoro-3-oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile (55 mg, 0.24 mmol) and pyridine (1 mL). It was heated to reflux for 1 h and then cooled down to room temperature. The solution was poured into column chromatography (silica gel) directly and the crude product was purified by using CHCl₃ as eluent to give a dark solid IUIC-T (88 mg, 89%). ¹H NMR (CDCl₃, 400 MHz, δ /ppm): 8.84 (s, 2H), 8.51 (m, 2H), 7.84 (s, 2H), 7.64 (t, *J* = 7.3 Hz, 2H), 6.96 (m, 16H), 6.48 (s, 2H), 6.14 (s, 2H), 2.72 (m, 4H), 2.56 (s, 8H), 1.56 (a, 8H), 1.49-1.24 (m, 42H), 1.00 (d, *J* = 6.1 Hz, 12H), 0.88 (d, *J* = 5.0 Hz, 12H). ¹³C NMR (CDCl₃, 100 MHz, δ /ppm): 185.99, 158.11, 156.02, 153.40, 150.72, 146.91, 146.81, 146.65, 142.61, 142.41, 142.38, 141.98, 141.76, 141.61, 138.05, 137.74, 137.58, 136.58, 134.46,134.12, 133.80, 132.99, 131.04, 130.58, 128.50, 128.28, 128.04, 127.98, 125.64, 124.63, 120.98, 114.30, 114.24, 112.65, 63.71, 41.53, 41.34, 35.60, 35.48, 34.20, 34.01, 32.54, 31.74, 31.44, 29.70, 29.25, 29.12, 28.87, 25.79, 25.69, 23.25, 23.17, 22.65, 22.62, 14.35, 14.31, 14.10, 10.92, 10.79. MALDI-TOF MS (m/z): 2112.6 (M + H⁺).

3. NMR spectra



-3.65

 $\begin{array}{c} -2.56\\ -2.52\\ -2$









Fig. S3 Absorption spectra of IUIC-O and IUIC-T in CHCl₃ (10⁻⁵ M).





Fig. S4 Cyclic voltammograms of PBDB-T, IUIC-O and IUIC-T.

	$\lambda_{sol.}$	λ_{film}	λ_{onset}	$E_{\rm g}^{\rm opt}$	HOMO	LUMO	$E_{\rm g}^{\rm ec}$
	[nm]	[nm]	[nm]	$[eV]^a$	[eV]	[eV]	$[eV]^b$
PBDB-T	-	622	686	1.81	-5.16	-3.02	2.14
IUIC-O	768	812	928	1.34	-5.16	-3.93	1.23
IUIC-T	757	795	902	1.37	-5.26	-3.92	1.34
	1 -						

 ${}^{a}E_{g}^{opt} = 1240/\lambda_{onset}; {}^{b}E_{g}^{ec} = LUMO-HOMO.$



Fig. S5 UPS spectra of the inelastic cut-off region (a) and the HOMO band region (b) of IUIC-O, IUIC-T and PBDB-T thin films.

6. Device fabrication and measurements

Inverted solar cells

The ZnO precursor solution was prepared according to the literature.^[4] It was spin-coated onto ITO glass (4000 rpm for 30 s). The films were annealed at 200 °C in air for 30 min. ZnO film thickness is ~30 nm. A PBDB-T:acceptor blend in ODCB with additive was spin-coated onto ZnO layer. Then the film was annealed at 100 °C in N₂ for 10 min. MoO₃ (~5 nm) and Ag (~80 nm) was successively evaporated onto the active layer through a shadow mask (pressure ca. 10^{-4} Pa). The effective area of the cells is 4 mm². *J-V* characteristics of devices were measured under AM 1.5G (100 mW cm⁻²) by using a Newport Thermal Oriel 91159A solar simulator. Light intensity is calibrated with a Newport Oriel PN 91150 V Si-based solar cell. *J-V* characteristics were recorded by using a Keithley 2400 source meter unit. EQEs were performed in air on an Oriel Newport system (Model 66902) equipped with a standard Si diode.

Electron-only devices

The structure for electron-only devices is ZnO/active layer/Ca/Al. The ZnO precursor was spin-coated onto ITO glass and annealed at 200 °C in air for 30 min. A PBDB-T:acceptor blend in ODCB with additive was spin-coated onto ZnO. Then the film was annealed at 100 °C in N₂ for 10 min. Ca (~5 nm) and Al (~100 nm) were successively evaporated onto the active layer through a shadow mask (pressure ca. 10^{-4} Pa). *J-V* characteristics were measured in the dark and recorded by using a Keithley 2400 source meter unit.

Hole-only devices

The structure for hole-only devices is PEDOT:PSS/active layer/MoO₃/Ag. The PEDOT:PSS was spin-coated onto ITO glass and annealed at 140 °C in air for 20 min. A PBDB-T:acceptor blend in ODCB with additive was spin-coated onto PEDOT:PSS. Then the film was annealed at 100 °C in N₂ for 10 min. MoO₃ (~5 nm) and Ag (~100 nm) were successively evaporated onto the active layer through a shadow mask (pressure ca. 10^{-4} Pa). *J-V* characteristics were measured in the dark and recorded by using a Keithley 2400 source meter unit.

7. PL



Fig. S6 Photoluminescence (PL) spectra of the PBDB-T, IUIC-O and IUIC-T neat films as well as PBDB-T:IUIC-O and PBDB-T:IUIC-T blend films excited at 622 nm (a), 800 nm (b) and 800 nm (c), respectively.

8. EQE_{EL}



Fig. S7 Electroluminescence quantum efficiency (EQE_{EL}) of PBDB-T:IUIC-O and PBDB-

T:IUIC-T devices.

9. SCLC

The space charge limited current (SCLC) was studied by using electron-only devices to find the charge-transport properties. The electron-only devices, consisting of active layer sandwiched between a ZnO coated ITO electrode and Ca/Al counter-electrode as the holeblocking contact, were fabricated. From the current density as a function of voltage data, the electron mobility in the trap-free SCLC region can be estimated using the Mott-Gurney equation:

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

where ε_0 is the permittivity of the vacuum, ε_r is the relative permittivity of the material, μ is the charge-carrier mobility, V is the effective voltage, $V = V_{appl} - V_{bi}$, where V_{appl} is the applied voltage, and V_{bi} is the built-in potential which results from the difference in the work function of the anode and the cathode, and d is the sample thickness. Using this expression, the electron mobilities of IUIC-O and IUIC-T in the blend films were calculated.



Fig. S8 The $J^{1/2}$ -V curves of electron-only devices (a) and hole-only devices (b) based on PBDB-T:IUIC-O and PBDB-T:IUIC-T blend films (under dark).

Table S2. Electron and hole mobilities of PBDB-T:IUIC-O and PBDB-T:IUIC-T blend films.

	$\mu_{\rm e} [{\rm cm}^2 {\rm V}^{-1} {\rm s}^{-1}]$	$\mu_h [{ m cm}^2 { m V}^{-1} { m s}^{-1}]$
PBDB-T:IUIC-O	1.25×10^{-4}	2.86×10^{-4}
PBDB-T:IUIC-T	1.60×10^{-4}	6.79×10^{-4}

10. AFM



Fig. S9 AFM images of PBDB-T:IUIC-O and PBDB-T:IUIC-T blend films: height images (left) and phase images (right).

11. GIWAXS



Fig. S10 2D GIWAXS patterns of PBDB-T, IUIC-O and IUIC-T neat films.

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