

## Supporting Information

### Effects of Fused-Ring Regiochemistry on the Properties and Photovoltaic Performance of *n*-type Organic Semiconductor Acceptors

Xiaojun Li,<sup>a b</sup> He Huang,<sup>b</sup> Zhengxing Peng,<sup>c</sup> Chenkai Sun,<sup>a b</sup> Dengchen Yang,<sup>a</sup> Jiadong Zhou,<sup>e</sup> Alex Liebman-Pelaez,<sup>d</sup> Chenhui Zhu,<sup>d</sup> Zhi-Guo Zhang,<sup>a</sup> Zhanjun Zhang,<sup>b</sup> Zengqi Xie,<sup>e</sup> Harald Ade,<sup>c</sup> \*Yongfang Li<sup>a b f</sup>\*

<sup>a</sup> CAS Research/Education Center for Excellence in Molecular Sciences, CAS Key Laboratory of Organic Solids, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China;

<sup>b</sup> School of Chemical Science, University of Chinese Academy of Sciences, Beijing 100049, China;

<sup>c</sup> Department of Physics and Organic and Carbon Electronics Lab (ORaCEL), North Carolina State University, Raleigh, North Carolina 27695, USA;

<sup>d</sup> Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA.

<sup>e</sup> Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, China.

<sup>f</sup> Laboratory of Advanced Optoelectronic Materials, College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou, Jiangsu 215123, China;

## Experimental Section

### Synthesis of TPIC and TPTIC:

All chemicals and solvents were purchased from J&K, Alfa Aesar and TCI Chemical Co. respectively. Compound **1**, **2**, (see Scheme 1) and the PTQ10 was synthesized according to the procedure reported in the literatures<sup>1-3</sup>. The synthetic route of TPIC and TPTIC was shown in Scheme 1 and detailed synthesis processes are described in the following.

**Synthesis of Compound 3a:** Compound **1a** (1.95 g, 5 mmol), Compound **2a** (2.56 g, 11 mmol), Pd (OAc)<sub>2</sub> (0.56 mg, 0.25 mmol) and tBu<sub>3</sub>PHBF<sub>4</sub> (144 mg, 0.5 mmol) were dissolved in acetone (60 mL). After stirred for 5 min, NaOH aqueous solution (2M) was added, and the reaction mixture was stirred for 1 h, then the mixture was extracted with dichloromethane (100 mL × 3) and water (100 mL). The collected organic layer was dried over MgSO<sub>4</sub>. After removal of the solvent under reduced pressure, the residue was purified by column chromatography on silica gel to give a white product **3a** (2.05g, 92% yield). <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 7.50 (d, *J* = 5.3 Hz, 2H), 7.29 (d, *J* = 5.3 Hz, 2H), 6.92 (s, 2H), 4.18 (q, *J* = 7.1 Hz, 4H), 3.73 (s, 6H), 1.17 (t, *J* = 7.1 Hz, 6H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 163.57, 150.43, 144.81, 130.81, 129.25, 124.45, 123.72, 114.42, 60.38, 56.23, 14.02. MS (EI<sup>+</sup>) *m/z* calcd for [M]<sup>+</sup> C<sub>22</sub>H<sub>22</sub>O<sub>6</sub>S<sub>2</sub> 446, found 446.

**Synthesis of Compound 3b:** Compound **1b** (2.00 g, 4.3 mmol), Compound **2b** (2.4 g, 10.3 mmol), and Pd (OAc)<sub>2</sub> (0.25 g, 0.2 mmol) were dissolved in acetone (43 mL). The reaction mixture was stirred for 3 h, then the mixture was extracted with dichloromethane (100 mL × 3) and water (100 mL). The collected organic layer was dried over MgSO<sub>4</sub>. After removal of the solvent under reduced pressure, the residue was purified by column chromatography on silica gel to give a yellow product **3b** (1.65 g, 86% yield). <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 7.84 (s, 2H), 7.26 (d, *J* = 5.5 Hz, 2H), 6.87 (d, *J* = 5.5 Hz, 2H), 4.22 (q, *J* = 7.1 Hz, 4H), 3.77 (s, 6H), 1.18 (t, *J* = 7.1 Hz, 6H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 167.56, 153.89, 134.28, 133.09, 131.02, 123.70, 117.72, 116.19, 61.13, 58.62, 13.90. MS (EI<sup>+</sup>) *m/z* calcd for [M]<sup>+</sup> C<sub>22</sub>H<sub>22</sub>O<sub>6</sub>S<sub>2</sub>

446, found 446.

### Generally synthesis of 5

Compound **3** (2.2 g, 5 mmol) was dissolved in 30 mL dry CH<sub>2</sub>Cl<sub>2</sub> and the reaction mixture was cooled down to -78°C. Then 15 mL BBr<sub>3</sub> (1.0 M solution in CH<sub>2</sub>Cl<sub>2</sub>) was added, after 5min the reaction mixture was heated to 25°C and stirred for 12 h. After that, water was added and the precipitation was filtered. After dried, the precipitation was dissolved in 35 mL dry THF and react with 20 mL hexylmagnesium bromide (1.0 M solution in THF) at rt. for 12 h, then the mixture was extracted with dichloromethane (100 mL × 3) and water (100 mL). The collected organic layer was dried over MgSO<sub>4</sub>. After removal of the solvent under reduced pressure, the crude product was dissolved in 20 mL toluene, and *p*-toluenesulfonic acid (1.7 g, 10 mmol) was added. After being stirred at 80 °C for 12 h, the mixture was extracted with dichloromethane and water. The combined organic layer was washed with water and brine, dried over anhydrous MgSO<sub>4</sub>. After removal of solvent it was chromatographed on silica gel using petroleum ether/dichloromethane (5:1) as eluent, yielding a solid product **5**.

**5a** (2.4 g, 65% yield) <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 7.15 (d, *J* = 5.1 Hz, 2H), 6.82 (s, 2H), 6.74 (d, *J* = 5.1 Hz, 2H), 1.83 (d, *J* = 9.2 Hz, 8H), 1.21 (s, 48H), 0.85 (t, *J* = 6.7 Hz, 12H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 146.01, 137.80, 132.15, 124.95, 123.53, 119.27, 110.19, 83.77, 40.15, 31.85, 29.97, 29.48, 29.27, 23.83, 22.65, 14.10. HRMS (TOF) *m/z* calcd for [M]<sup>+</sup> C<sub>48</sub>H<sub>74</sub>O<sub>2</sub>S<sub>2</sub> 746.5124, found 746.5125.

**5b** (2.2 g, 60% yield) <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 7.04 (d, *J* = 5.3 Hz, 2H), 6.77 (s, 2H), 6.67 (d, *J* = 5.3 Hz, 2H), 2.00-1.82 (m, 8H), 1.24 (d, *J* = 10.7 Hz, 48H), 0.83 (d, *J* = 6.9 Hz, 12H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 151.87, 132.76, 126.75, 121.98, 119.42, 117.23, 114.45, 85.48, 39.22, 31.84, 29.91, 29.41, 29.26, 23.69, 22.65, 14.10. HRMS (TOF) *m/z* calcd for [M]<sup>+</sup> C<sub>48</sub>H<sub>74</sub>O<sub>2</sub>S<sub>2</sub> 746.5124, found 746.5124.

### Generally synthesis of 6

A Vilsmeier reagent, which was prepared with POCl<sub>3</sub> (0.14 mL, 1.46 mmol) in DMF (2.00 mL, 25.84 mmol), was added to a cold solution of compound **5** (215 mg,

0.29 mmol) in 1,2-dichloroethane (20 mL) at 0 °C under Ar. After being stirred at 60 °C for 12 h, the mixture was poured into ice water (100 mL), neutralized with Na<sub>2</sub>CO<sub>3</sub>, and then extracted with dichloromethane. The combined organic layer was washed with water and brine, dried over anhydrous MgSO<sub>4</sub>. After removal of solvent it was chromatographed on silica gel using petroleum ether/dichloromethane (1:1) as eluent, yielding a yellow solid **6**.

**6a** (205 mg, 88% yield) <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 9.87 (s, 2H), 7.40 (s, 2H), 6.93 (s, 2H), 1.99-1.77 (m, 8H), 1.22 (s, 48H), 0.85 (t, *J* = 6.7 Hz, 12H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 181.27, 146.35, 140.72, 140.40, 138.24, 132.46, 119.26, 110.45, 82.54, 39.05, 30.79, 28.82, 28.41, 28.20, 22.77, 21.60, 13.06. HRMS (TOF) *m/z* calcd for [M+H]<sup>+</sup> C<sub>50</sub>H<sub>75</sub>O<sub>4</sub>S<sub>2</sub> 803.5102, found 803.5101.

**6b** (200 mg, 86% yield) <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 9.82 (s, 2H), 7.31 (s, 2H), 6.94 (s, 2H), 1.93 (dd, *J* = 12.4, 7.3 Hz, 8H), 1.22 (s, 48H), 0.85 (t, *J* = 6.6 Hz, 12H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 181.22, 151.60, 137.60, 133.73, 126.53, 125.52, 123.82, 117.63, 84.99, 38.42, 30.78, 28.75, 28.35, 28.18, 22.67, 21.61, 13.06. HRMS (TOF) *m/z* calcd for [M+H]<sup>+</sup> C<sub>50</sub>H<sub>75</sub>O<sub>4</sub>S<sub>2</sub> 803.5102, found 803.5094.

**Synthesis of Compound TPTC** Compound **6a** (113 mg, 0.14 mmol) and 1,1-dicyanomethylene-3-indanone (134 mg, 0.69 mmol) were dissolved in dry CHCl<sub>3</sub> (25 mL). The mixture was deoxygenated with argon gas, and 0.6 ml pyridine were added and refluxed for 12 h. Then, the mixture was poured into water (100 mL) and extracted with CHCl<sub>3</sub> (2 × 100 mL). The organic layer was washed with water, and then dried over MgSO<sub>4</sub>. After removing the solvent, the residue was purified using column chromatography on silica gel employing petroleum ether/CHCl<sub>3</sub> (1:4) as an eluent, yielding a dark blue solid **TPTC** (137 mg, 85%). <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.82 (s, 2H), 8.71 (dd, *J* = 6.1, 2.0 Hz, 2H), 7.95 (dd, *J* = 5.8, 2.8 Hz, 2H), 7.87-7.74 (m, 4H), 7.45 (s, 2H), 7.11 (s, 2H), 1.91 (d, *J* = 6.9 Hz, 8H), 1.24 (s, 48H), 0.85 (t, *J* = 6.6 Hz, 12H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 188.27, 160.29, 148.42, 147.75, 141.47, 140.56, 139.98, 137.29, 136.94, 136.58, 135.33, 134.67, 125.39, 123.90, 122.88, 121.35, 114.55, 111.73, 83.59, 69.66, 40.32, 31.83, 29.83, 29.45, 29.25, 23.76, 22.64, 14.10. HRMS (TOF) *m/z* calcd for [M]<sup>+</sup> C<sub>74</sub>H<sub>82</sub>N<sub>4</sub>O<sub>4</sub>S<sub>2</sub> 1154.5782, found 1154.5782.

**Synthesis of Compound TPTIC:** Compound **6b** (113 mg, 0.14 mmol) and 1,1-dicyanomethylene-3-indanone (134 mg, 0.69 mmol) were dissolved in dry CHCl<sub>3</sub> (25 mL). The mixture was deoxygenated with argon gas, and 0.6 ml pyridine were added and refluxed for 12 h. Then, the mixture was poured into water (100 mL) and extracted with CHCl<sub>3</sub> (2 × 100 mL). The organic layer was washed with water, and then dried over MgSO<sub>4</sub>. After removing the solvent, the residue was purified using column chromatography on silica gel employing petroleum ether/CHCl<sub>3</sub> (1:4) as an eluent, yielding a dark blue solid **TPTIC** (143 mg, 89%). <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.79 (s, 2H), 8.73-8.67 (m, 2H), 7.95 (dd, *J* = 6.1, 2.5 Hz, 2H), 7.86-7.71 (m, 4H), 7.47 (s, 2H), 7.15 (s, 2H), 1.98 (dd, *J* = 12.9, 7.9 Hz, 8H), 1.44-1.20 (m, 48H), 0.83 (t, *J* = 6.5 Hz, 12H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 188.50, 160.10, 154.04, 140.13, 137.27, 136.91, 136.32, 135.36, 134.62, 134.15, 132.67, 132.27, 128.45, 125.44, 123.76, 123.41, 119.42, 114.51, 114.40, 86.20, 70.03, 40.34, 31.81, 29.79, 29.42, 29.24, 23.70, 22.65, 14.10. HRMS (TOF) *m/z* calcd for [M +H]<sup>+</sup> C<sub>74</sub>H<sub>83</sub>N<sub>4</sub>O<sub>4</sub>S<sub>2</sub> 1155.5850, found 1155.5856.

**Measurements:** <sup>1</sup>H NMR spectra were measured on a Bruker DMX-400 spectrometer with *d*-chloroform as the solvent and trimethylsilane as the internal reference. UV-visible absorption spectra were measured on a Hitachi U-3010 UV-vis spectrophotometer. Mass spectra were recorded on a Shimadzu spectrometer. Thermogravimetric analysis (TGA) was conducted on a Perkin-Elmer TGA-7 thermogravimetric analyzer at a heating rate of 20 °C/min and under a nitrogen flow rate of 100 mL/min. UV-vis absorption spectra of active layers of the PSCs were measured on a Hitachi U-3010 UV-vis spectrophotometer. The electrochemical cyclic voltammetry was performed on a Zahner IM6e Electrochemical Workstation, in an acetonitrile solution of 0.1 mol/L *n*-Bu<sub>4</sub>NPF<sub>6</sub> at a potential scan rate of 100 mV/s with an Ag/AgCl reference electrode and a platinum wire counter electrode. The film morphology was measured using an atomic force microscope (AFM, SPA-400) using the tapping mode.

**Device fabrication and characterization:** The PSCs were fabricated with a structure of ITO/PEDOT: PSS (40 nm)/active layer/cathode. A thin layer of PEDOT: PSS was

deposited through spin-coating on precleaned ITO-coated glass from a PEDOT: PSS aqueous solution (Baytron P VP AI 4083 from H. C. Starck) at 4000 rpm and dried subsequently at 150 °C for 15 min in air. Then the device was transferred to a nitrogen glove box, where the active blend layer of PTQ10 polymers and acceptors was spin-coated from its chloroform solution onto the PEDOT: PSS layer under a spin-coating rate of 3000 rpm. After spin-coating, the active layers were annealed at 120 °C for 5 min for the devices with thermal annealing treatment. The thickness of the active layers is *ca.* 100 nm. Then methanol solution of PDINO at a concentration of 1.0 mg mL<sup>-1</sup> was deposited atop the active layer at 3000 rpm for 30 s to afford a PDINO cathode buffer layer with thickness of *ca.* 10 nm. Finally, top Al electrode was deposited in vacuum onto the cathode buffer layer at a pressure of *ca.* 5.0 × 10<sup>-5</sup> Pa. The active area of the device was 4.7 mm<sup>2</sup>. The current density-voltage (*J-V*) characteristics of the PSCs were measured in glovebox on a computer-controlled Keithley 2450 Source-Measure Unit. Oriel Sol3A Class AAA Solar Simulator (model, Newport 94023A) with a 450 W xenon lamp and an air mass (AM) 1.5 filter was used as the light source. The light intensity was calibrated to 100 mW cm<sup>-2</sup> by a Newport Oriel 91150V reference cell. The input photon to converted current efficiency (IPCE) was measured by Solar Cell Spectral Response Measurement System QE-R3-011 (Enli Technology Co., Ltd., Taiwan). The light intensity at each wavelength was calibrated with a standard single-crystal Si photovoltaic cell.

**Mobility Measurements:** Hole and electron mobilities were measured using the the space charge limited current (SCLC) method. Device structures are ITO/PEDOT:PSS/PTQ10: acceptor (1:1.5, w/w) /Au for hole-only devices and ITO/ZnO/PTQ10: acceptor (1: 1.5, w/w) /PDINO/Al for electron-only devices. The SCLC mobilities were calculated by MOTT-Gurney equation:

$$J = \frac{9\varepsilon_r\varepsilon_0\mu V^2}{8L^3} \quad (1)$$

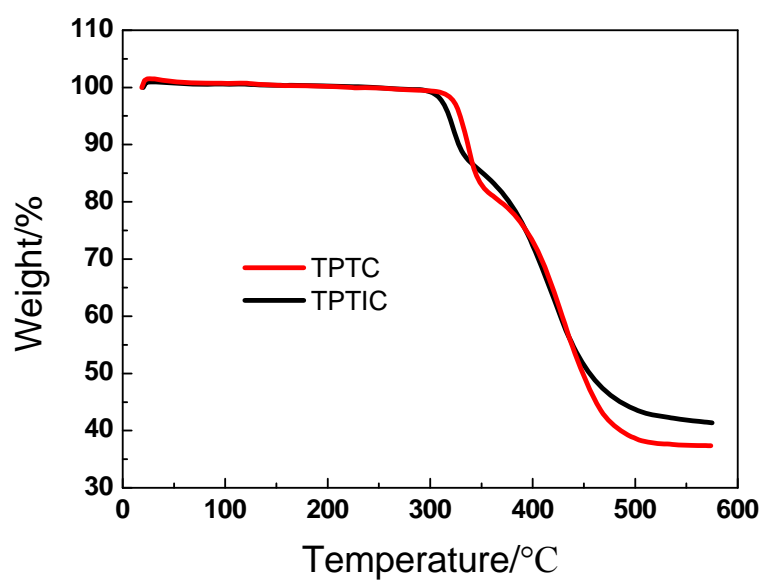
Where *J* is the current density,  $\varepsilon_r$  is the relative dielectric constant of active layer material usually 2-4 for organic semiconductor, herein we use a relative dielectric constant of 4,  $\varepsilon_0$  is the permittivity of empty space,  $\mu$  is the mobility of hole or

electron and  $L$  is the thickness of the active layer,  $V$  is the internal voltage in the device, and  $V = V_{\text{app}} - V_{\text{bi}}$ , where  $V_{\text{app}}$  is the voltage applied to the device, and  $V_{\text{bi}}$  is the built-in voltage resulting from the relative work function difference between the two electrodes (in the hole-only and the electron-only devices, the  $V_{\text{bi}}$  values are 0.2 V and 0V respectively).

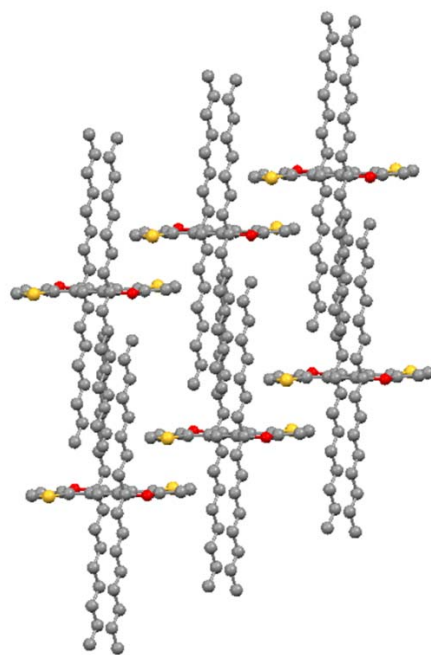
**DSC measurements.** Neat materials were dissolved in chlorobenzenen and stirred overnight at 80 °C. The solution was then drop cast on a precleaned glass substrate and kept under a nitrogen atmosphere for 48 h to remove the residual solvents. DSC heat-cool-heat measurements were carried out at a heating/cooling rate of 10 °C/min using a TA Instrument Discovery Series.

**GIWAXS Characterization:** Grazing-incidence wide-angle X-ray scattering (GIWAXS) measurements were conducted at Advanced Light Source (ALS), Lawrence Berkeley National Laboratory, Berkeley, CA at the beamline 7.3.3. Data was acquired at the critical angle ( $0.13^\circ$ ) of the film with a hard X-ray energy of 10 keV. X-ray irradiation time was 10-30 s, dependent on the saturation level of the detector. Beam center was calibrated using AgB powder and the sample-to-detector distance was about 330 mm. The  $\pi$ - $\pi$  coherence lengths ( $L$ ) are estimated based on the Scherrer Equation ( $L=2\pi K/\text{FWHM}$ ), where  $K$  is the shape factor (here we use 0.9), and FWHM is the full width at half maximum of the (010) diffraction peaks.

**Resonant soft X-ray scattering (RSoXS):** Resonant soft X-ray scattering was performed at beamline 11.0.1.2 Advanced Light Source, LBNL. Thin film samples were spin-casted on top of the PEDOT:PSS covered Si wafers under exactly the same condition as those for the fabrication of solar cell devices. Then BHJ thin films were floated and transferred onto silicon nitride membrane windows. The scattering was done in transmission mode and signals were collected in vacuum using Princeton Instrument PI-MTE CCD camera. The RSoXS profiles are Lorentz corrected and thickness normalized.

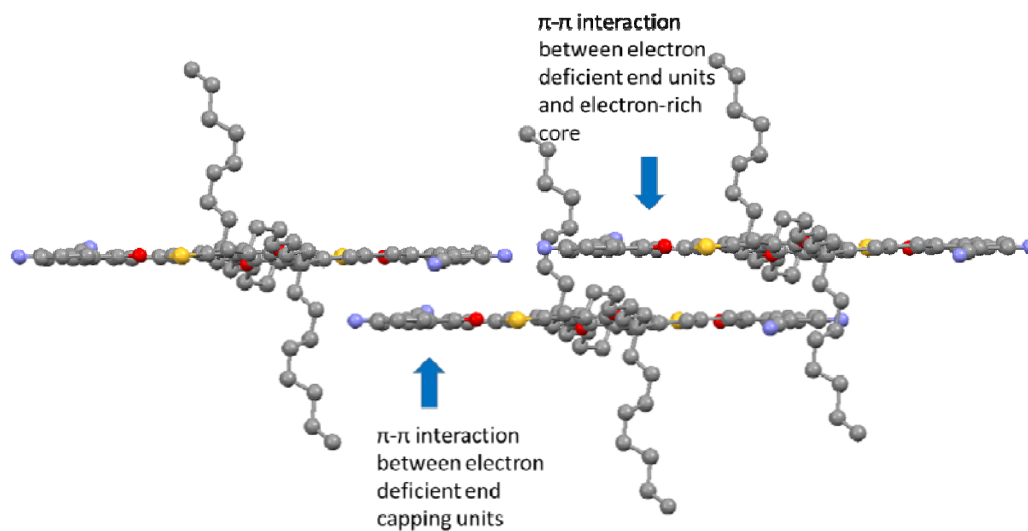


**Figure S1.** TGA plots of TPTC and TPTIC

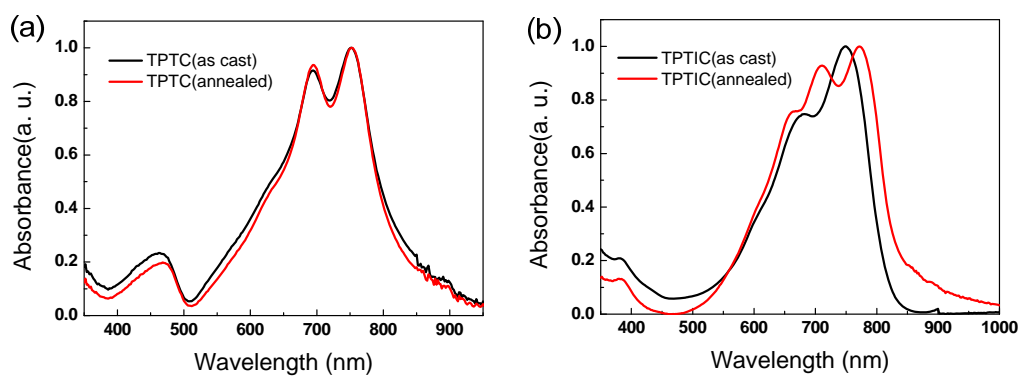


**Figure S2.** Crystal structure and packing of the core of TPTIC (compound **5b**).

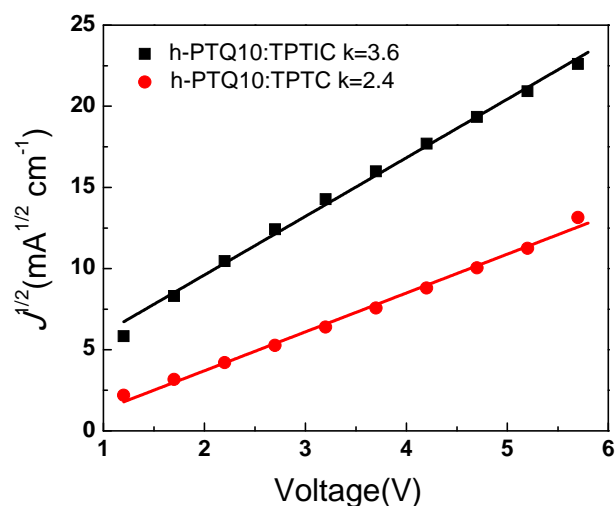




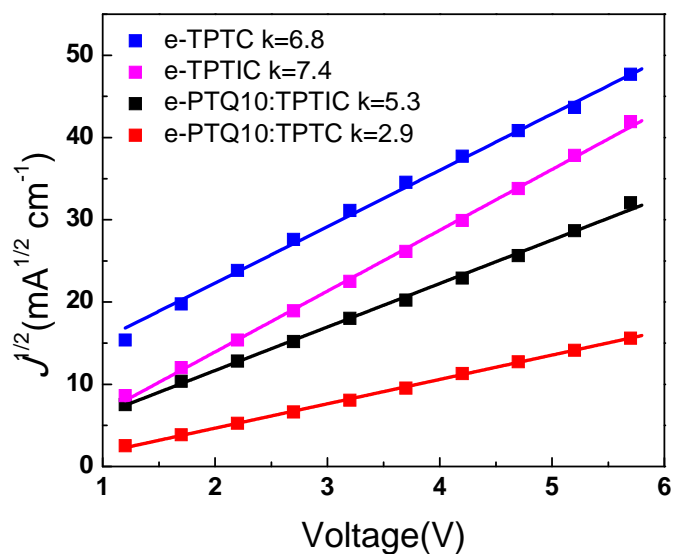
**Figure S3.** Crystal structure and packing of TPTC, and the intermolecular interactions.



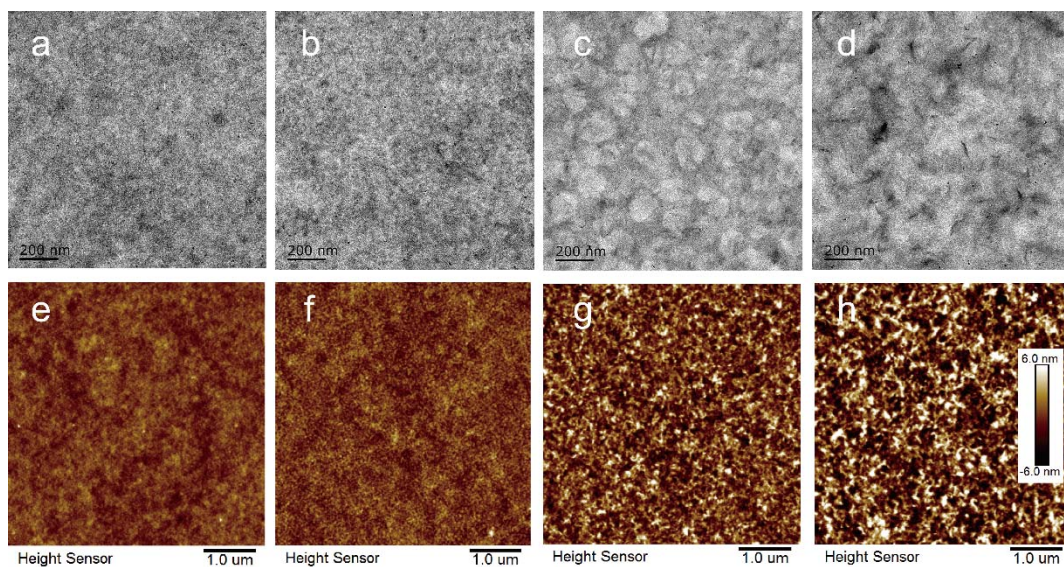
**Figure S4.** Absorption spectra of (a) TPTC and (b) TPTIC in chloroform solutions as cast and with thermal annealing at 120 °C for 5 min.



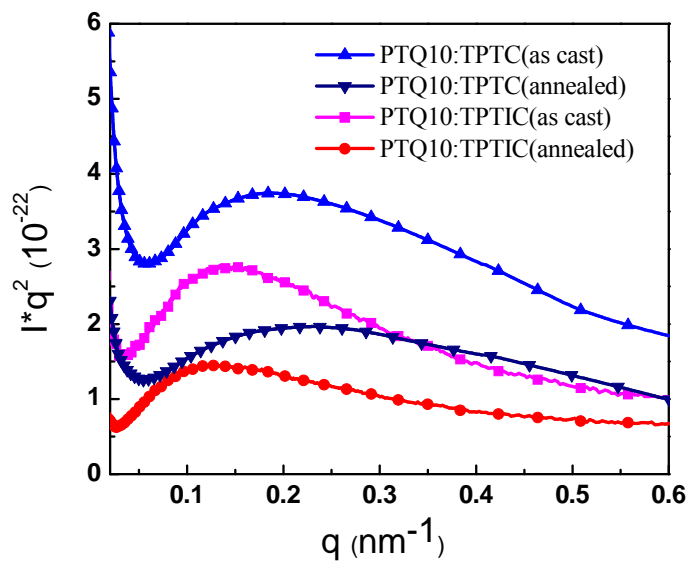
**Figure S5.**  $J_{1/2} \sim (V_{\text{appl}} - V_{\text{bi}} - V_{\text{s}})$  characteristics of hole-only devices based on the blend films of PTQ10:acceptors. Solid lines are the fitting lines of the data.



**Figure S6.**  $J_{1/2} \sim (V_{\text{appl}} - V_{\text{bi}} - V_{\text{s}})$  characteristics of the electron-only devices based on the neat films of acceptors and blend films of PTQ10:acceptors. Solid lines are the fitting lines of the data.



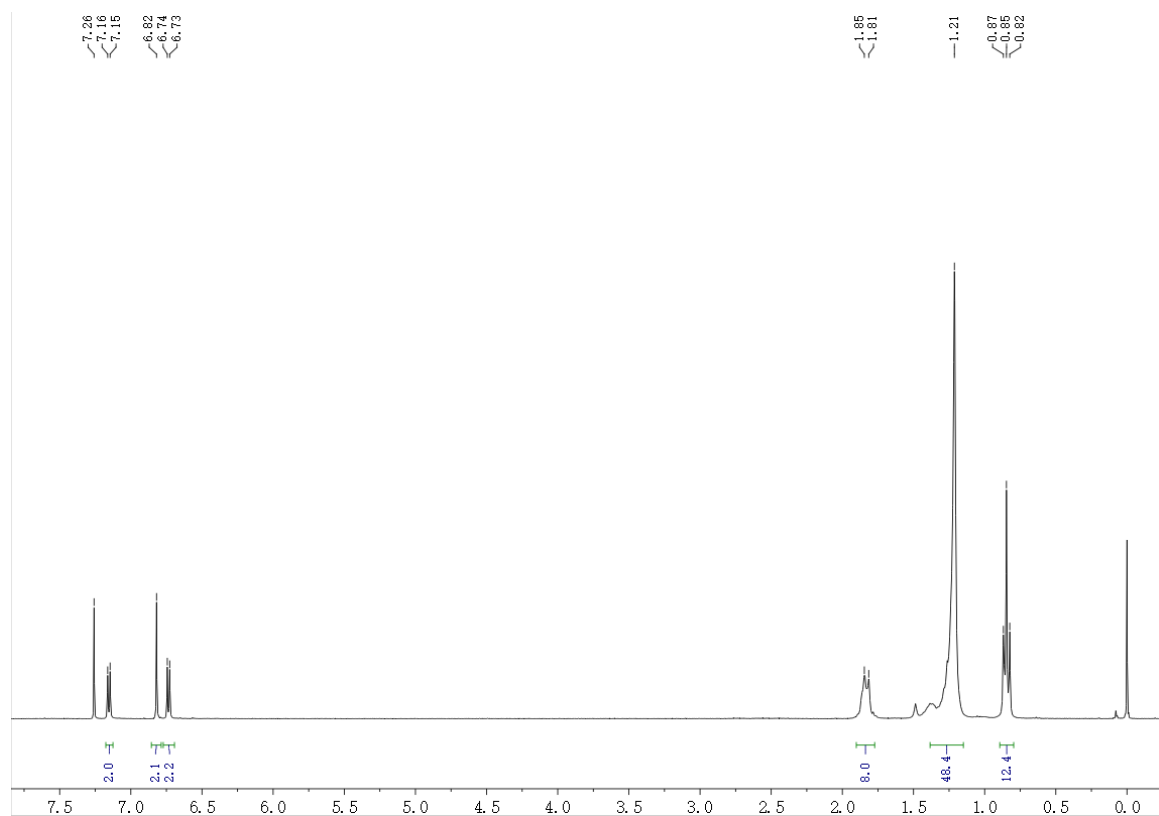
**Figure S7.** AFM and TEM images for blend films of PTQ10 and the acceptors: (a, e) PTQ10:TPTIC without extra treatment; (b, f) PTQ10:TPTIC with thermal annealing at 120 °C for 5 min; (c, g) PTQ10:TPTIC without extra treatment; (d, h) PTQ10:TPTIC with thermal annealing at 120 °C for 5 min.



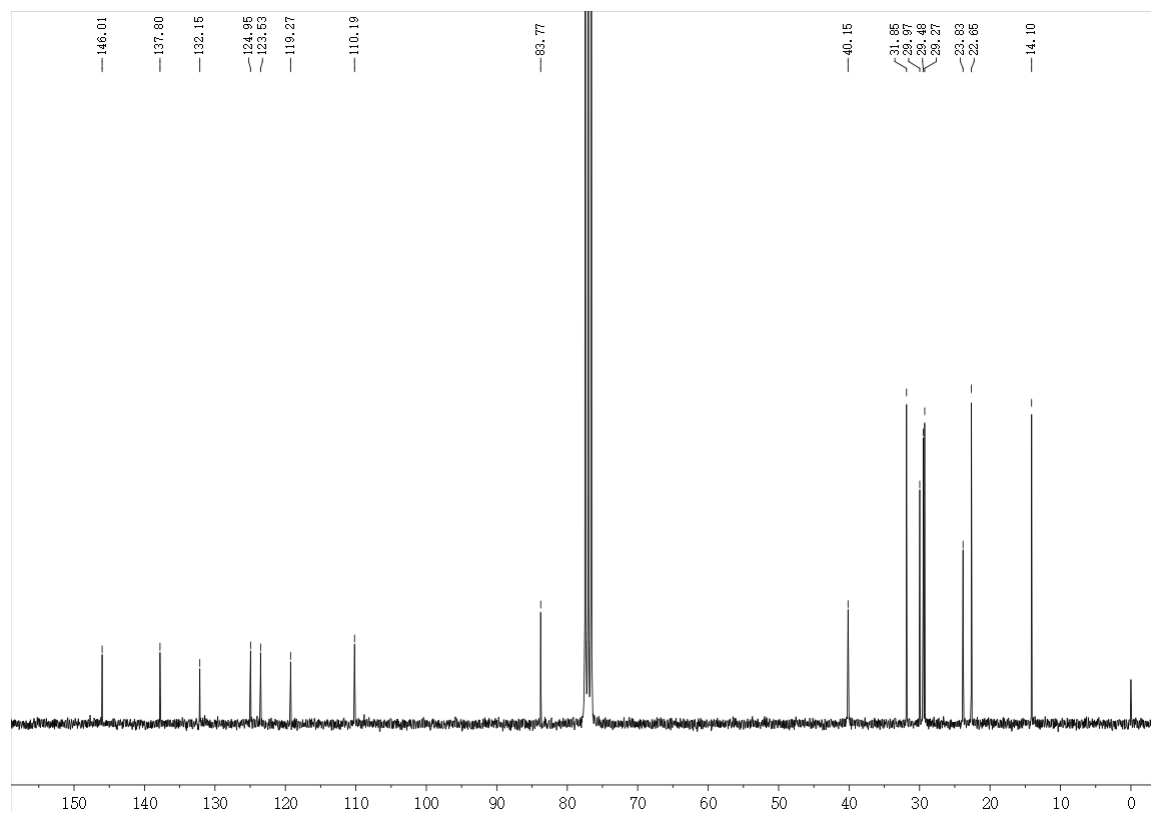
**Figure S8.** The RSoXS profiles of PTQ10:TPTIC and PTQ10:TPTIC for the as cast and thermal-annealed blend films.

**Table S1.** Charge carrier mobilities of the neat films of acceptors and the blend films of PTQ10: acceptors with thermal annealing at 120 °C for 5 min.

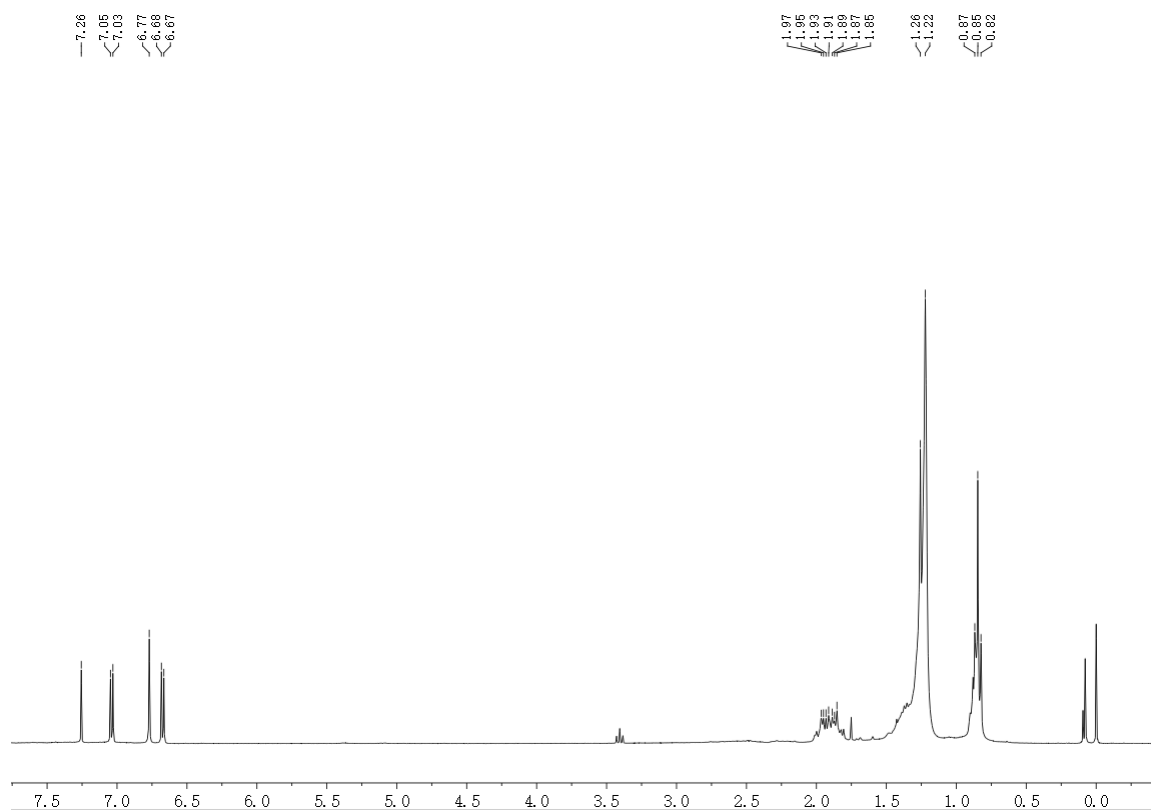
blend	$\mu_h$ (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	$\mu_e$ (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )
PTQ10:TPTC	$0.19 \times 10^{-4}$	$0.28 \times 10^{-4}$
PTQ10:TPTIC	$0.58 \times 10^{-4}$	$0.94 \times 10^{-4}$
TPTC		$0.33 \times 10^{-4}$
TPTIC		$1.83 \times 10^{-4}$



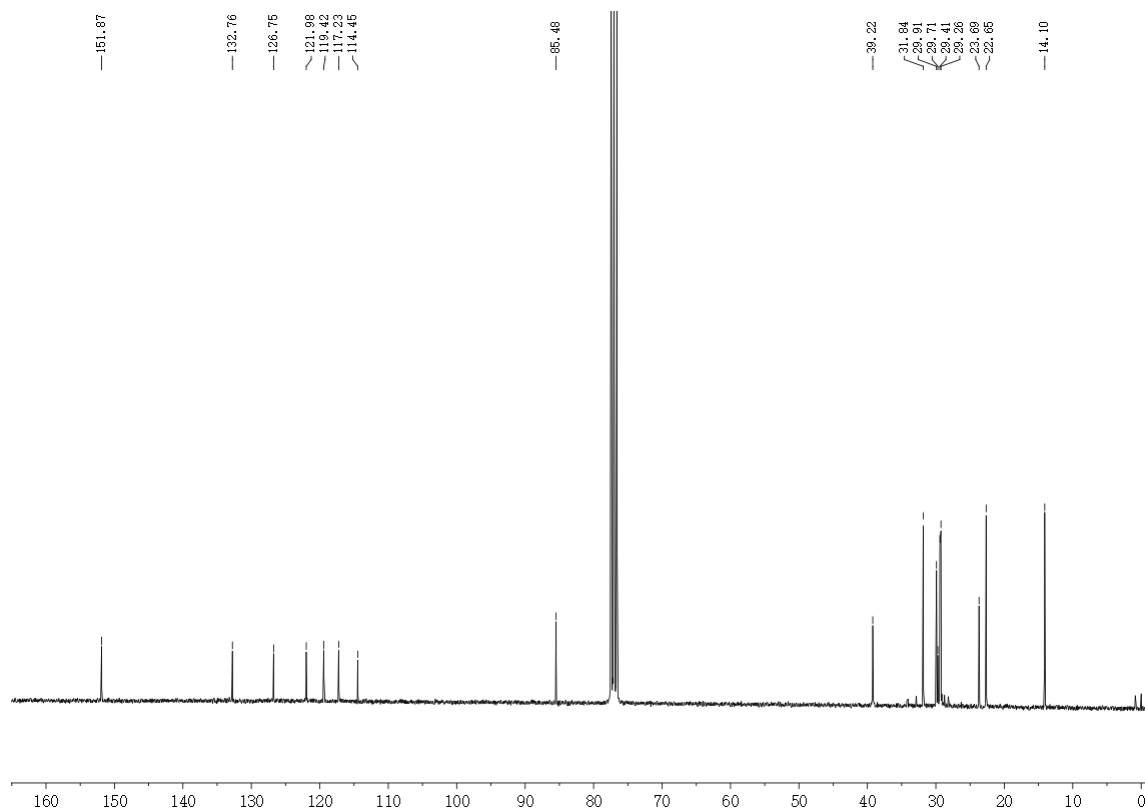
**Figure S9.** <sup>1</sup>H NMR spectrum (300 MHz, CDCl<sub>3</sub>) of **5a**.



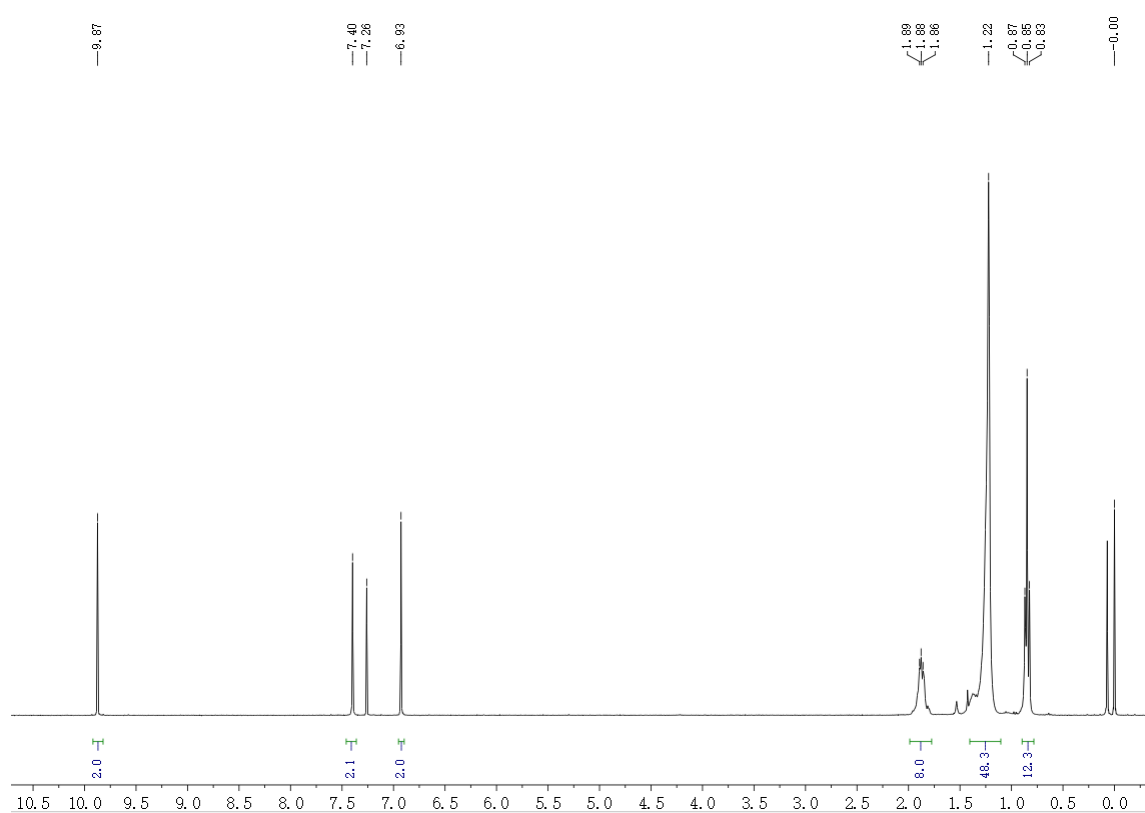
**Figure S10.**  $^{13}\text{C}$  NMR spectrum (75 MHz,  $\text{CDCl}_3$ ) of **5a**.



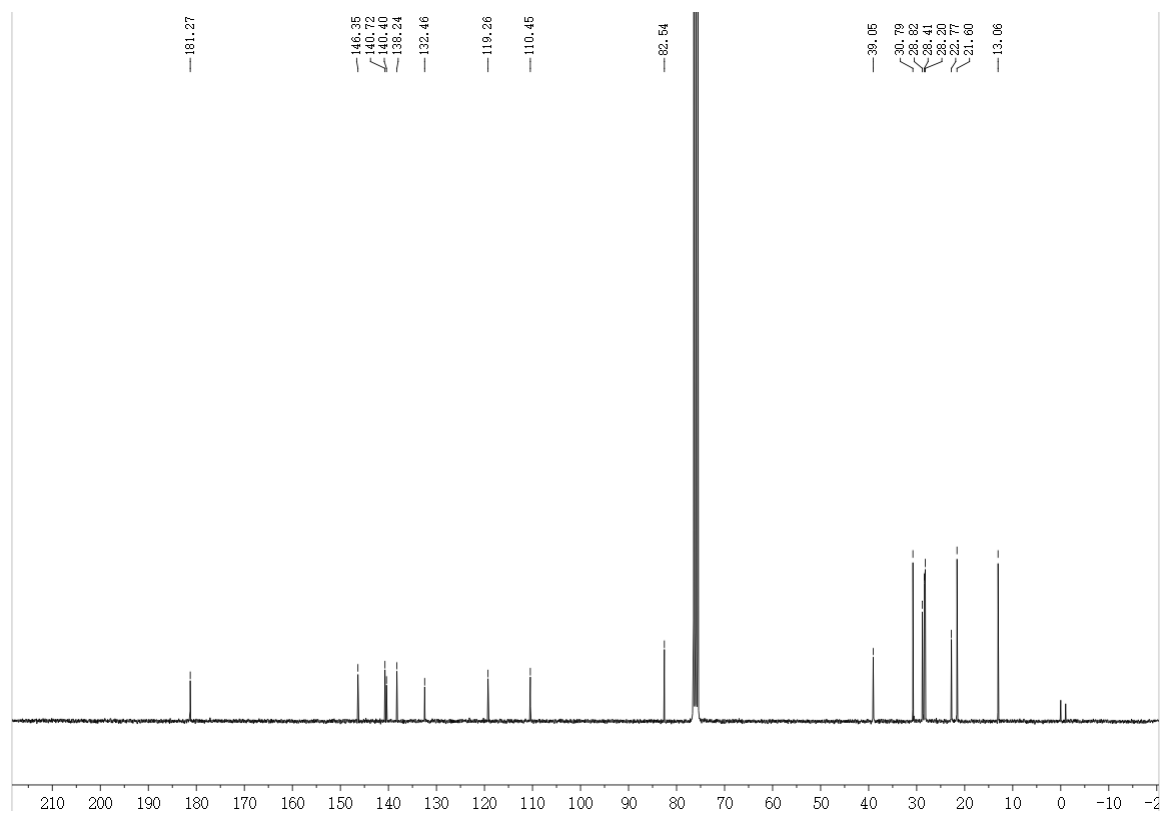
**Figure S11.**  $^1\text{H}$  NMR spectrum (300 MHz,  $\text{CDCl}_3$ ) of **5b**.



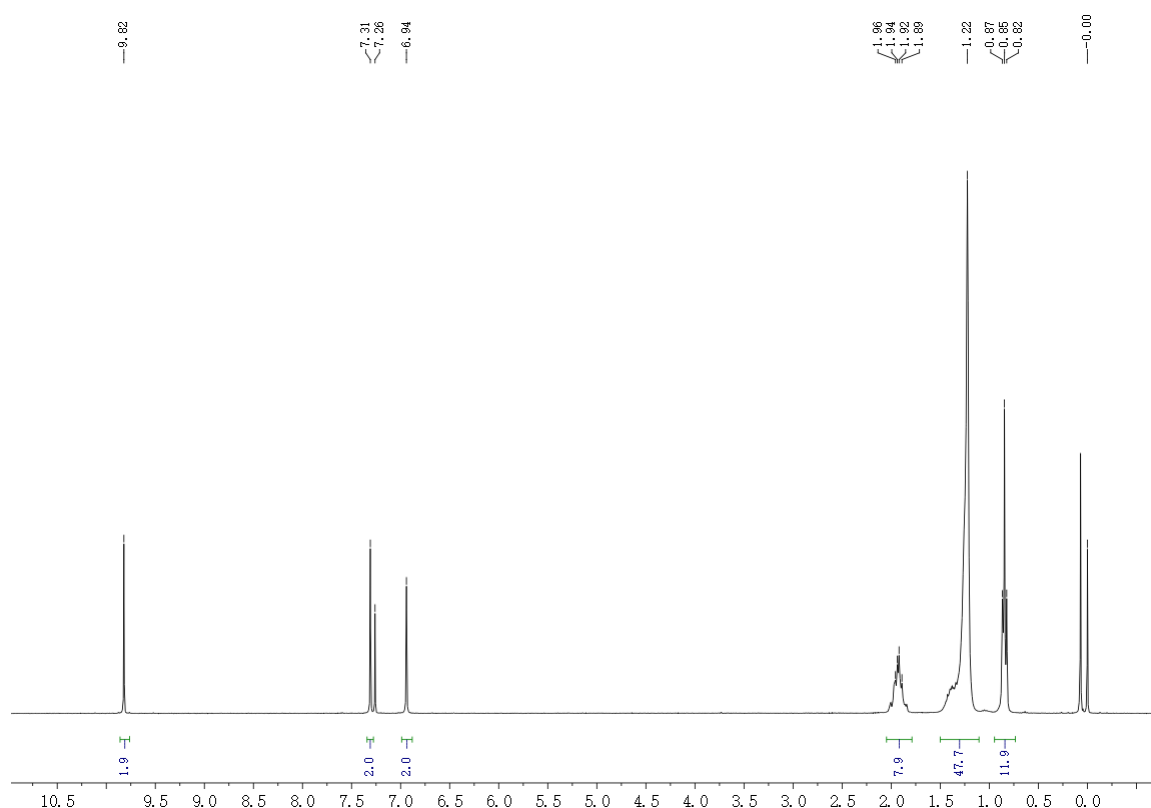
**Figure S12.**  $^{13}\text{C}$  NMR spectrum (75 MHz,  $\text{CDCl}_3$ ) of **5b**.



**Figure S13.**  $^1\text{H}$  NMR spectrum (300 MHz,  $\text{CDCl}_3$ ) of **6a**.



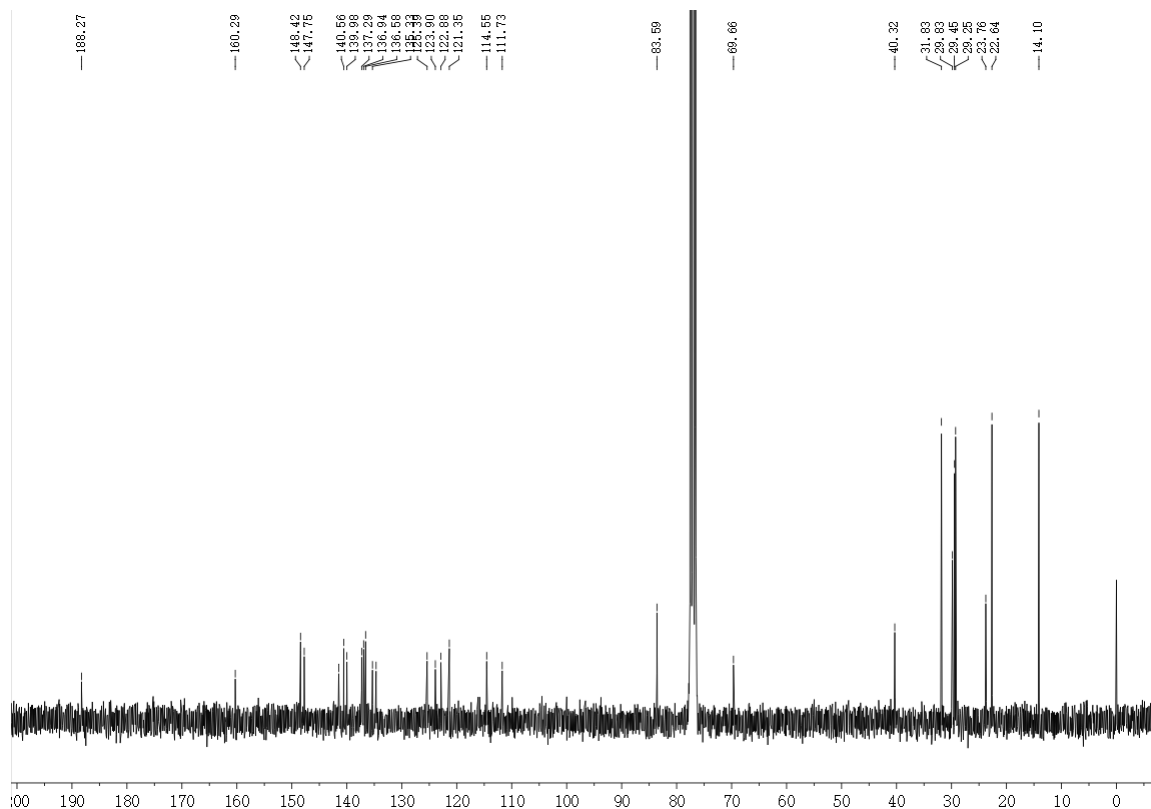
**Figure S14.**  $^{13}\text{C}$  NMR spectrum (75 MHz,  $\text{CDCl}_3$ ) of **6a**.



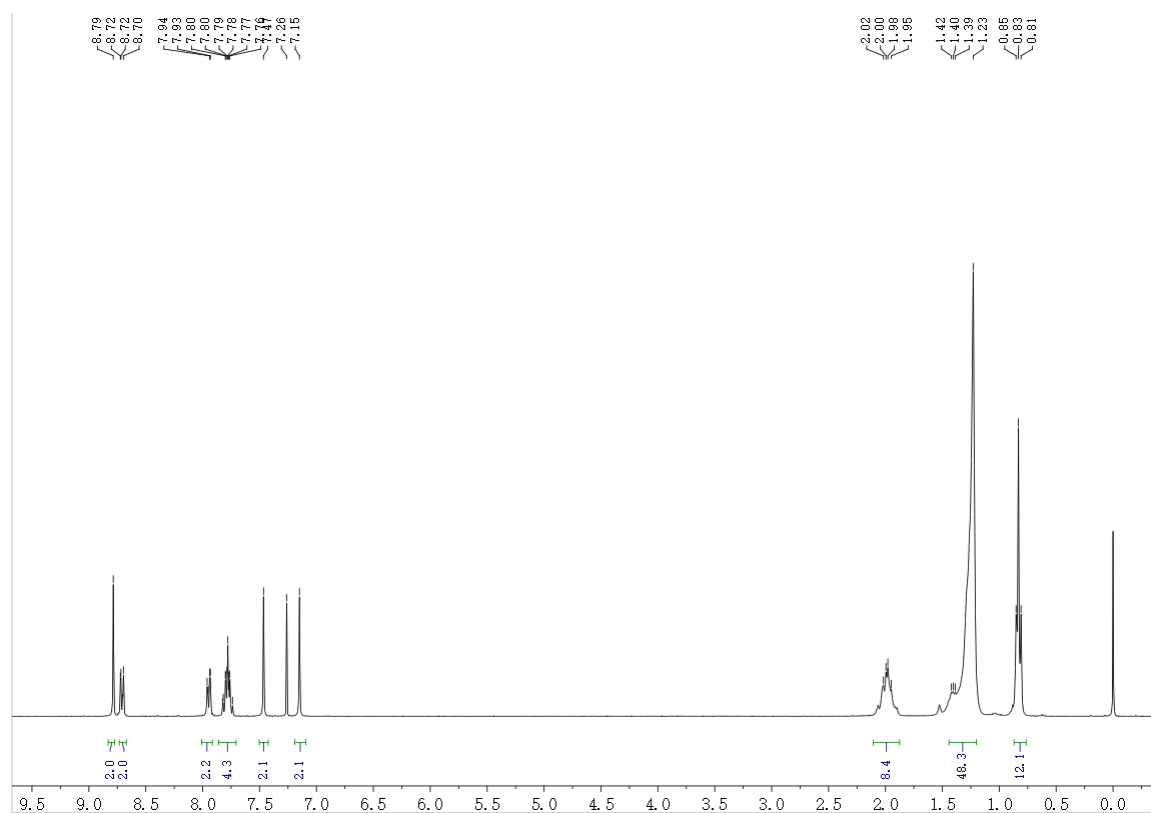
**Figure S15.**  $^1\text{H}$  NMR spectrum (300 MHz,  $\text{CDCl}_3$ ) of **6b**.



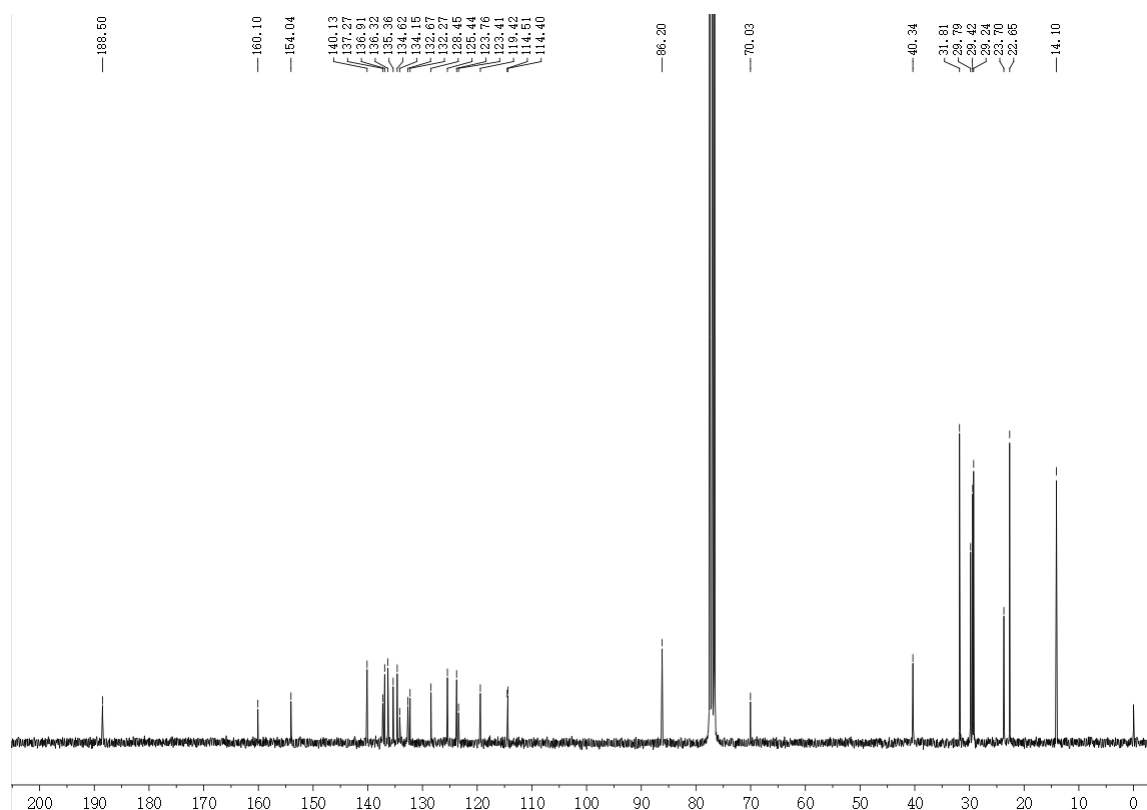




**Figure S18.**  $^{13}\text{C}$  NMR spectrum (75 MHz,  $\text{CDCl}_3$ ) of TPTC.



**Figure S19.**  $^1\text{H}$  NMR spectrum (300 MHz,  $\text{CDCl}_3$ ) of TPTIC.



**Figure S20.**  $^{13}\text{C}$  NMR spectrum (75 MHz,  $\text{CDCl}_3$ ) of TPTIC.

## References

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