

Supplementary Information

19% Efficiency All-Polymer Solar Cells via Introducing Pincer-Shaped Non-covalent Bond Interactions

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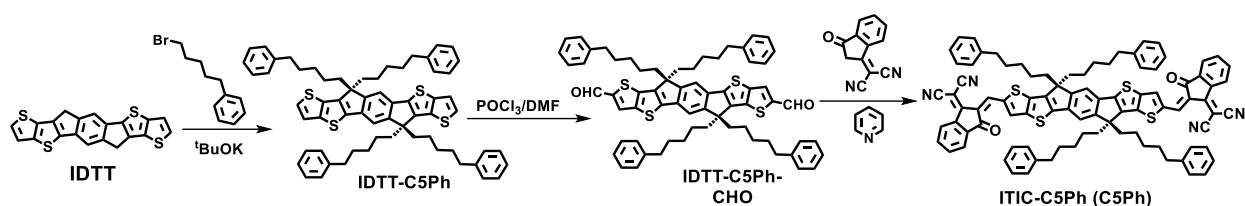
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1. Synthesis of small molecules



Scheme S1. The synthetic routes of C5Ph molecule.

C6Ph was synthesized according to our previous literature (Adv. Mater. 2023, 35, 2208986). The synthetic route of C5Ph is shown in **Scheme 1**. Detailed synthetic procedures are shown below:

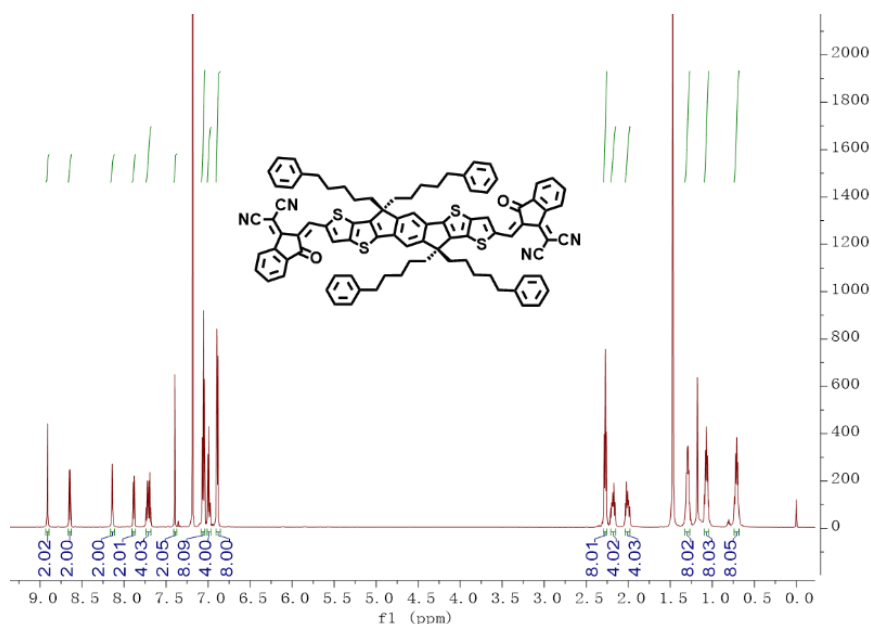
Synthesis of IDTT-C5Ph: In a 100 mL flash, indacenodithienothiophene (IDTT) (400 mg, 1.06 mmol) and 1-Bromo-5-phenylpentane (1.92 g, 8.45 mmol) were dissolved into 20 mL anhydrous THF. The ^tBuOK (948.6 mg, 8.45 mmol) was added slowly in batches. After stirring at room temperature for an additional 2 hours, the reaction mixture was extracted with dichloromethane three times and the collected organic layers were distilled under reduced pressure. The residue was purified by silica gel column chromatography to afford off-white powder. (Yield: 63%)

¹H NMR (600 MHz, Chloroform-*d*) δ 7.38 – 7.34 (m, 4H), 7.18 (t, *J* = 7.5 Hz, 8H), 7.11 (t, *J* = 7.3 Hz, 4H), 6.99 (d, *J* = 7.4 Hz, 8H), 2.37 (t, *J* = 7.7 Hz, 8H), 2.13 (td, *J* = 12.5, 4.9 Hz, 4H), 2.04 – 1.95 (m, 4H), 1.39 (p, *J* = 7.0 Hz, 8H), 1.12 (q, *J* = 7.6 Hz, 8H), 0.83 (tt, *J* = 11.8, 6.1 Hz, 8H). ¹³C NMR (151 MHz, Chloroform-*d*) δ 152.22, 145.36, 143.68, 142.60, 141.40, 136.60, 133.51, 128.32, 128.09, 125.48, 125.42, 120.57, 112.97, 54.11, 38.27, 35.60, 30.91, 29.70, 29.24, 23.77.

Synthesis of IDTT-C5Ph-CHO: N, N-dimethylformamide (DMF) (1 mL) was added to 1,2-dichloroethane (1 mL) under argon atmosphere, then POCl₃ (1.5 mL) was added dropwise at 0 °C and stirred for another 2 hours. Then IDTT-C5Ph (170 mg, 0.18 mmol) dissolved in 1,2-dichloroethane (10 mL) was dropped and refluxed for 48 hours. The mixture cooled to room temperature and poured into ice water stirred for another 4 hours. Then the mixture was extracted with dichloromethane for three times and removed solvent under vacuum. The crude product was purified on silica gel column chromatography to give orange oil. (Yield: 68%)

¹H NMR (600 MHz, Chloroform-*d*) δ 9.97 (s, 2H), 8.02 (s, 2H), 7.41 (s, 2H), 7.16 (t, *J* = 7.5 Hz, 8H), 7.10 (t, *J* = 7.3 Hz, 4H), 6.98 – 6.96 (m, 8H), 2.35 (t, *J* = 7.6 Hz, 8H), 2.16 – 2.10 (m, 4H), 2.01 (ddd, *J* = 13.6, 11.3, 5.1 Hz, 4H), 1.36 (dt, *J* = 14.1, 7.2 Hz, 8H), 1.14 – 1.09 (m, 8H), 0.77 (dddd, *J* = 16.4, 13.0, 6.7, 3.8 Hz, 8H). ¹³C NMR (151 MHz, Chloroform-*d*) δ 182.81, 153.66, 150.25, 145.94, 143.87, 142.37, 141.67, 139.85, 137.02, 130.22, 128.28, 128.14, 125.54, 114.18, 54.49, 38.12, 35.59, 30.89, 29.16, 23.90.

Synthesis of C5Ph: IDTT-C5Ph-CHO (150 mg, 0.15 mmol) and 2-(3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (200 mg, 1.03 mmol) were dissolved in 20 mL degassed chloroform. Then pyridine (1 mL) was added and the mixture was refluxed for 2.5 hours. After cooling to room temperature, the mixture was poured into methane and filtered. The residue was purified by silica gel column chromatography, yielding a blue solid. (Yield: 76%) ^1H NMR (600 MHz, Chloroform- d) δ 8.91 (s, 2H), 8.65 (d, 2H), 8.14 (s, 2H), 7.89 (d, 2H), 7.74 – 7.68 (m, 4H), 7.40 (s, 2H), 7.06 (t, $J = 7.4$ Hz, 8H), 6.99 (t, $J = 7.4$ Hz, 4H), 6.91 – 6.85 (m, 8H), 2.27 (t, $J = 7.7$ Hz, 8H), 2.21 – 2.15 (m, 4H), 2.01 (dt, $J = 13.7, 8.7$ Hz, 4H), 1.29 (pd, $J = 7.1, 2.7$ Hz, 8H), 1.09 – 1.04 (m, 8H), 0.74 – 0.68 (m, 8H).



2. Details of single crystal cultivation

The small molecule C5Ph was dissolved in chloroform ($\sim 1\text{mg/mL}$, 8 mg) and sealed in a small vial (volume: 15 mL) by aluminium-foil paper. Then several (4~6) pinholes were made on the foil paper by fine needle and put the small vial into a large-sized vial (volume: 50 mL), which contains 10 mL CH_3OH . Finally, the large-sized vial was tightly sealed and put into a quiet and dark place at room temperature for 5~7 days to culture the single crystals.

The crystal structure of C6Ph was extracted from our previous report. (Adv. Mater. 2023, 35, 2208986)

3. Device fabrication and evaluations

3.1 Device fabrication of rigid solar cells

The polymer donors PBQx-TF, PM6, and polymer acceptor PY-IT were purchased from Solarmer Materials Inc. The Molecular weights and PDIs were measured based on high temperature gel permeation chromatography measurements (trichlorobenzene, 150 °C). PBQx-TF, GPC: Mn= 38.6 kDa, Mw=72.3 kDa, PDI=1.87; PM6, GPC: Mn= 38.4 kDa, Mw=92.2 kDa, PDI=2.39; PY-IT, GPC: Mn= 7.3 kDa, Mw=13.1 kDa, PDI=1.78.

All the rigid solar cells were fabricated with a conventional device structure of Glass/ITO/PEDOT:PSS/active layer/PDINN/Ag. The patterned ITO glass (sheet resistance ~15 Ω/ square) was pre-cleaned in an ultrasonic bath of acetone and isopropyl alcohol and treated by Plasma for 100 s. Then a thin layer (about 25 nm) of PEDOT:PSS was spin-coated onto the ITO glass at 4000 rpm and baked at 150 °C for 15 min.

Active materials: PBQx-TF, PM6 and PY-IT were purchased from Solarmer-materials Co.

For the fabrication of bulk heterojunction (**BHJ**) APSCs, PBQx-TF:PY-IT based all-polymer solutions (PBQx-TF concentration: 5.3 mg/ml in Toluene) with 1.8 vol% 1-chloronaphthalene (CN) were stirred for 2.5 hrs at 85 °C before spin-coating on the PEDOT:PSS layer to form the thin active layer about 110±20 nm. PM6:PY-IT based all-polymer solutions (PM6 concentration: 6.8 mg/ml in chloroform) with 1.8 vol% 1-chloronaphthalene (CN) were stirred for 2.5 hrs at 50 °C before spin-coating on the PEDOT:PSS layer to form the thin active layer about 110±20 nm. The thickness of the thick active layer was measured using a Veeco Dektak 150 profilometer. Then PDINN (in CH₃OH, 1mg/mL) was spin-coating at 3000 rpm to form the electron transfer layer. Finally, Ag (100 nm) metal electrode was thermal evaporated under about 5×10⁻⁵ Pa and the device area was 0.0936 cm² defined by shadow mask.

For the fabrication of pseudo-planar heterojunction (**PPHJ**) APSCs, the donor(s) solutions in Toluene (PBQx-TF concentration: 5.5 mg/ml) were stirred for 2.5 hrs at 85 °C before spin-coating on the PEDOT:PSS layer to form the bottom layers about 65±10 nm. the acceptor top solutions in chloroform (PY-IT concentration: 7 mg/ml) with 1.8 vol% 1-chloronaphthalene (CN) were stirred for 2.5 hrs at 50 °C before spin-coating on donor layers to form the acceptor layers about 40±10 nm. Then PDINN (in CH₃OH, 1mg/mL) was spin-coating at 3000 rpm to form the electron transfer layer. Finally, Ag (100 nm) metal electrode was thermal evaporated under about 5×10⁻⁵ Pa and the device area was 0.0936 cm² defined by shadow mask.

3.2 Device fabrication of flexible solar cells

All the flexible solar cells were fabricated with a conventional device structure of PET/ITO(AgNWs)/PEDOT:PSS/active layer/PDINO/Al. Prepare flexible samples by sticking them onto a rigid

glass substrate to prevent device deformation from affecting film quality. The patterned ITO glass (sheet resistance $\sim 15 \Omega/\text{square}$) was pre-cleaned in an ultrasonic bath of acetone and isopropyl alcohol and treated by Plasma for 60 s. Then a thin layer (about 25 nm) of PEDOT:PSS was spin-coated onto the ITO glass at 4000 rpm and baked at 100 °C for 20 min. The preparation of the active layer is the same as that of rigid devices. Then PDINN (in CH_3OH , 1mg/mL) was spin-coating at 3000 rpm to form the electron transfer layer. Finally, Al (100 nm) metal electrode was thermal evaporated under about 5×10^{-5} Pa.

4. Instruments and characterizations

The current density-voltage (J - V) curves of all the polymer solar cells (OPVs) were measured by a Keithley 2400 unit in high-purity nitrogen-filled glove box ($\text{H}_2\text{O} < 0.1$ PPM; $\text{O}_2 < 0.1$ PPM). The AM 1.5G irradiation was provided by an XES-40S2 (SAN-EI ELECTRIC Co., Ltd) solar simulator (AAA grade, $70 \times 70 \text{ mm}^2$ photobeam size) with light intensity of 100 mW cm^{-2} . The solar simulator is calibrated by standard silicon cell (SRC-00178) before device testing. The standard silicon cell was last certified in September 2022. The dark current test is in an opaque box. The J - V curves were measured in forward scan mode (from -0.2 V to 1.2 V) with a scan step length of 0.02 V and dwell time of 50 ms. The pre sweep delay is 0.5 s.

The external quantum efficiency (EQE) spectra of OPVs were measured by a Zolix Solar Cell Scan 100. The light intensity at each wavelength was also calibrated with a standard silicon solar cell (RCS103011-E) before EQE spectra test. The EQE spectra are tested in an air environment without any packaging.

The absorption spectra of films were measured with a PERSEE TU-1810 PC spectrometer. Spin the prepared solution on the Plasma treated quartz wafer by spin coating, and the treatment method is consistent with the device.

The AFM were measured by Agilent 5400 AFM. Spin the prepared solution onto the glass substrate with PEDOT:PSS to prepare the test samples. The films treatment method is the same as that of the devices.

The hole mobility and electron mobility were measured by space-charge-limited current (SCLC) method with a device configuration of ITO/PEDOT:PSS/active layer/ MoO_3 /Ag and ITO/ ZnO /active layer/PDINN/Ag structure.

The theoretical simulations and calculations of interactions were conducted by Gaussian 09 based on B3LYPD3/def2-SVP level of theory.

GIWAXS were measured at the synchrotron radiation center of Shanghai light source. Spin the prepared solution onto the plasma treated silicon wafer. The size is 1.5 cm×1.5cm. The grazing angle were selected at 0.12°-0.16°.

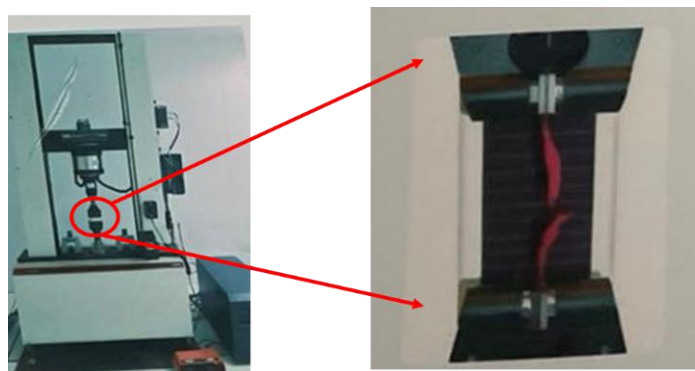
Film thickness is determined by Dektak 150 Step meter.

Electrochemical cyclic voltammetry (*C-V*) was measured by electrochemical workstation.

Contact angles are measured by the contact angle measuring instrument CSCDIC-200S.

Depth-dependent absorption spectra of the PPHJ active layers: The preparation of the active layer is the same as that of rigid devices.

In-situ stress-strain test (Adv. Energy Mater. 2022, 12, 2201614): For the tensile testing specimen, the active layers were blade-coated onto the PEDOT:PSS/glass substrate. To float the layers on the water surface, water was allowed to penetrate into the PEDOT:PSS layer. Subsequently, PEDOT:PSS was dissolved, and the active layer was delaminated from the glass substrate. By performing this process at the water surface, the floating active layer specimen could be obtained. The active layer specimen with a size of 2.54×0.5 cm was prepared by using a cutting plotter. The tensile test was performed by a linear stage with a strain rate of 1 μm/s. During the tensile test, stress and strain data were obtained through a load cell (Istron-3300, US) and a digital image correlation device. The corresponding test program is Bluehill Lite software. Elongation at break (%) = $L1$ (tensile length of sample)/ L (effective length of sample) * 100%. The schematic diagram of stretching instrument and stretching process is as follows:



Mechanical stability tests: The mechanical stability tests were carried out in high purity nitrogen in the laboratory glove box. The performance test data of flexible devices are obtained at a set speed of 2 times/s under cylinders with different curvature. (Please note that the bending rate has a great impact on the performance of the device, and ignore the attenuation of devices in a short time)

5. Supplementary Figures and Tables

5.1 Supplementary Figures

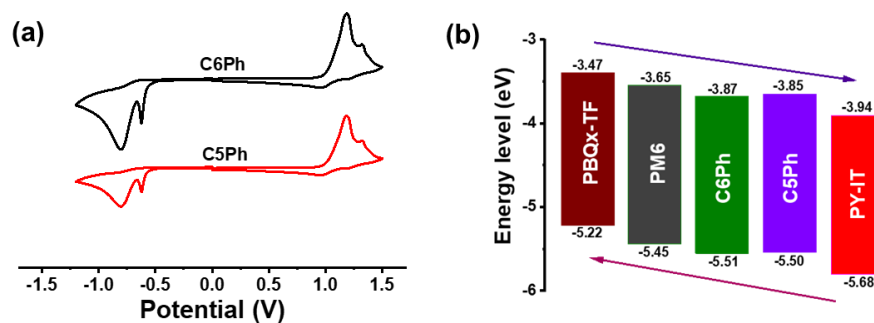


Fig. S1 (a) The C-V plots of C6Ph and C5Ph; (b) Schematic diagram of energy levels for all materials.

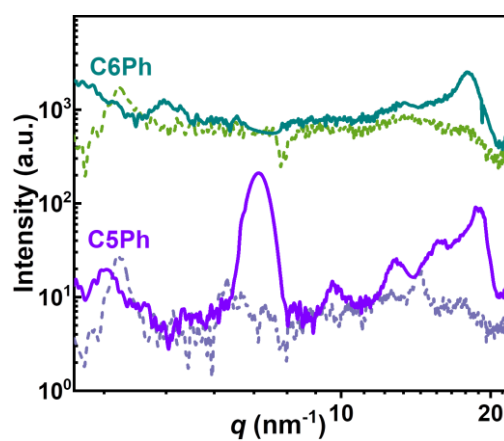


Fig. S2 The line-cut profiles of C6Ph and C5Ph neat films based on GIWAXS studies.

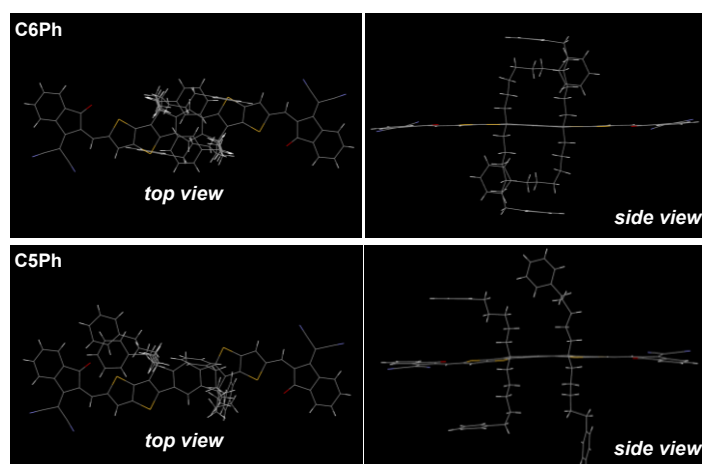


Fig. S3 The molecular conformations of C6Ph and C5Ph in single crystals.

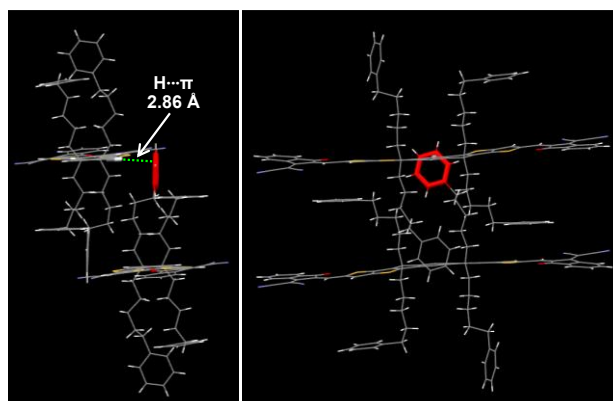


Fig. S4 The short distance H \cdots π interaction between the hydrogen atom of C5Ph₍₁₎ skeleton and the distal phenyl π -plane of C5Ph₍₀₎.

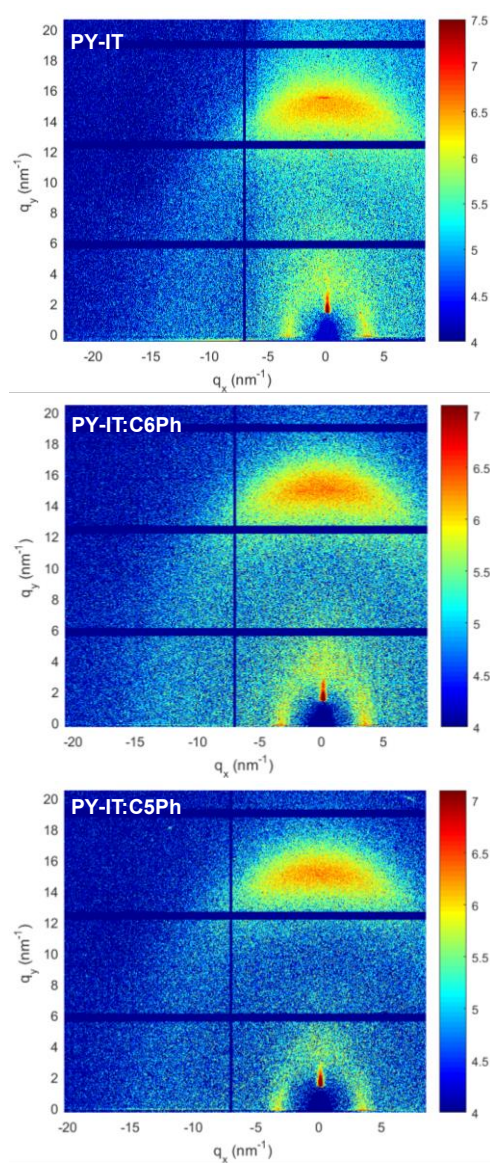


Fig. S5 2D-GIWAXS patterns of PY-IT, PY-IT:C6Ph, and PY-IT:C5Ph. The weight ratios for PY-IT and the C6Ph or C5Ph are 1.2:0.1.

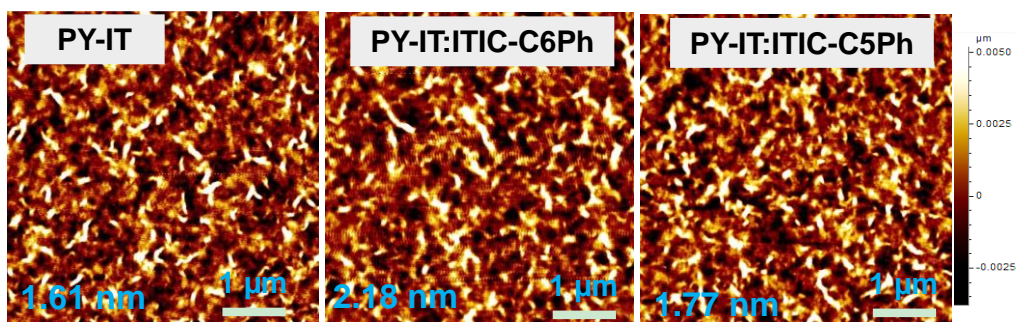


Fig. S6 AFM images of PY-IT, PY-IT:C6Ph, and PY-IT:C5Ph. The weight ratios for PY-IT and the C6Ph or C5Ph are 1.2:0.1. The inserted numbers are on behalf of the RMS values.

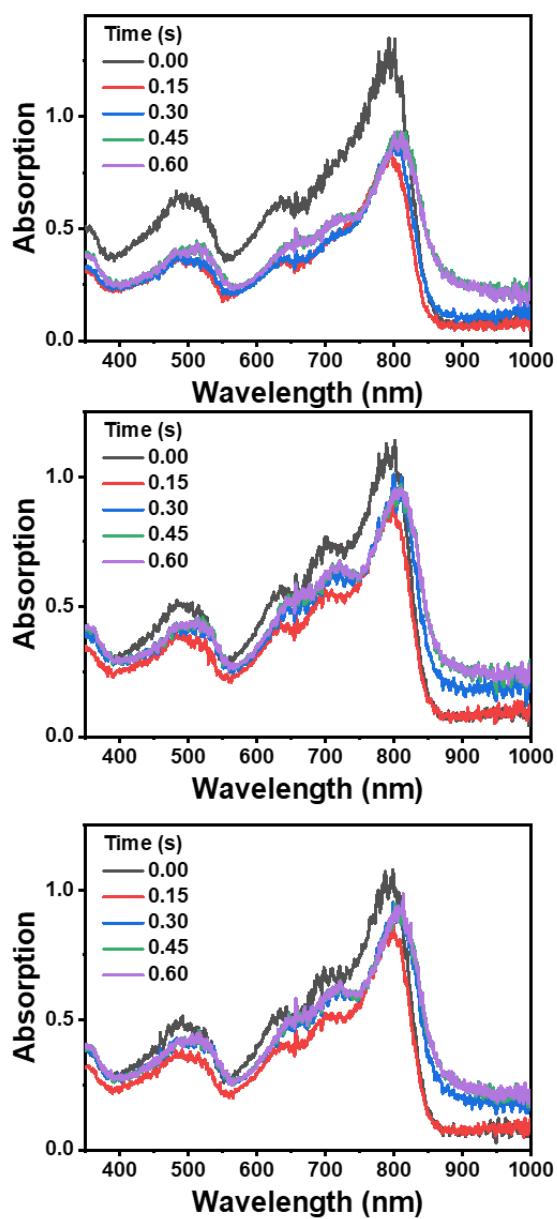


Fig. S7 The in-suit absorption spectra of PY-IT, PY-IT:C6Ph, and PY-IT:C5Ph. The weight ratios for PY-IT and the C6Ph or C5Ph are 1.2:0.1.

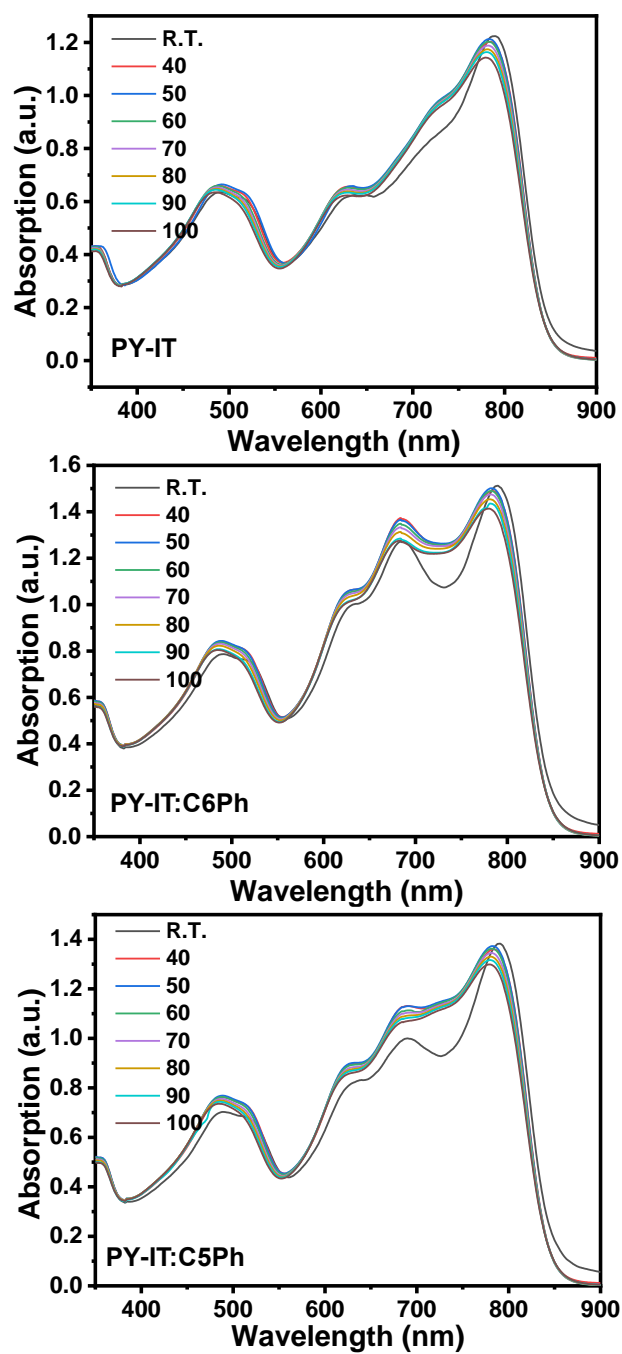


Fig. S8 The absorption spectra of PY-IT, PY-IT:C6Ph, and PY-IT:C5Ph in dilute chlorobenzene solutions with temperature dependence.

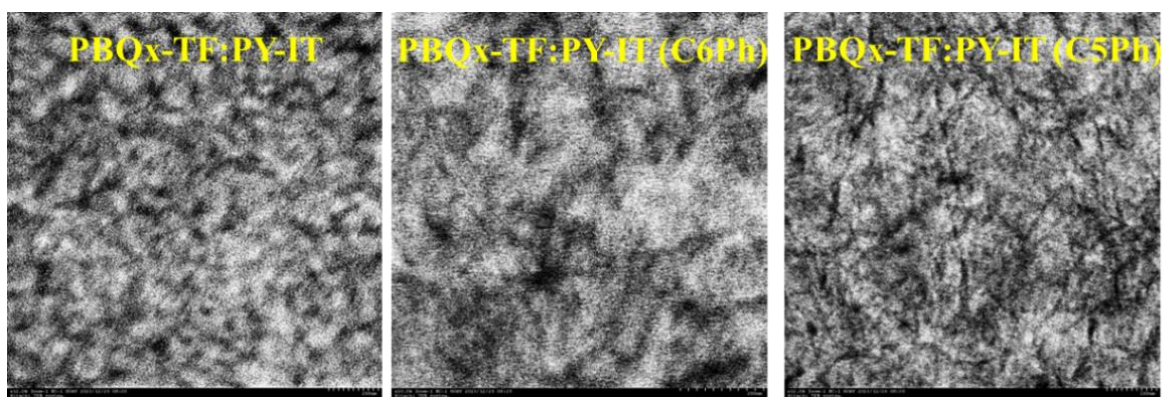


Fig. S9 TEM images of PBQx-TF:PY-IT, PBQx-TF:PY-IT (C6Ph) and PBQx-TF:PY-IT (C5Ph) blend films.

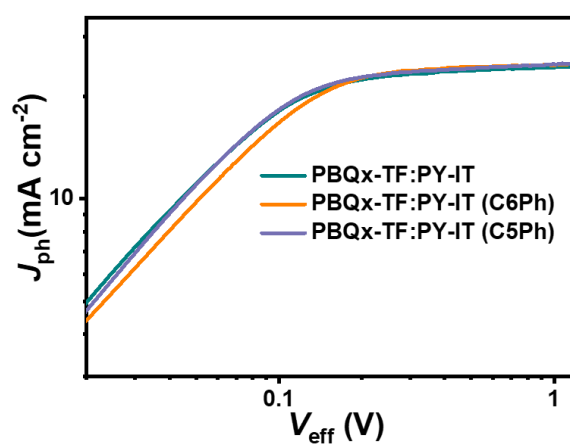


Fig. S10 The photocurrent density (J_{ph}) versus effective voltage (V_{eff}) curves of PBQx-TF:PY-IT, PBQx-TF:PY-IT (C6Ph) and PBQx-TF:PY-IT (C5Ph) based all-PSCs.

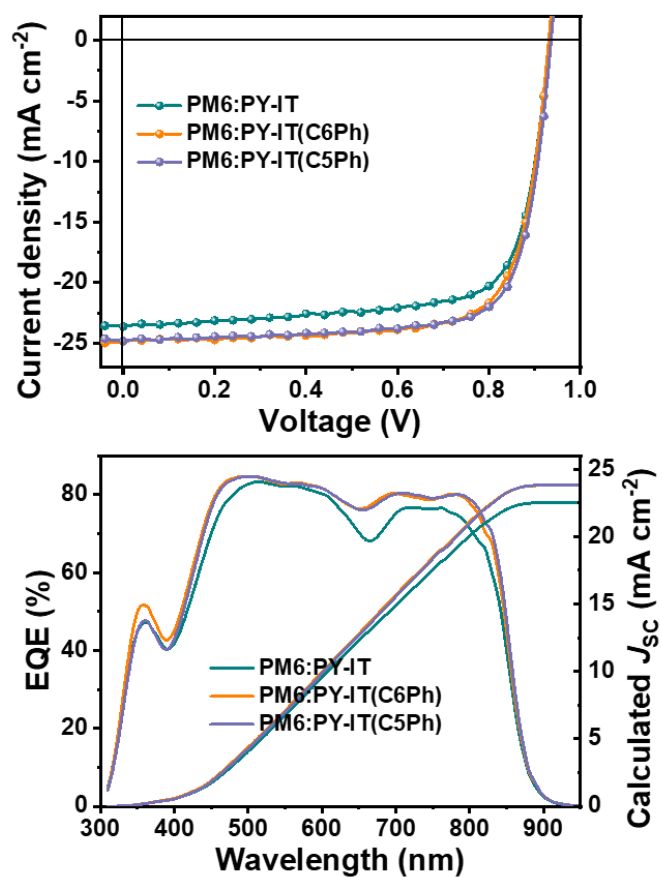


Fig. S11 Optimal J - V and EQE plots of PM6:PY-IT, PM6:PY-IT(C6Ph) and PM6:PY-IT(C5Ph) based all-PSCs.

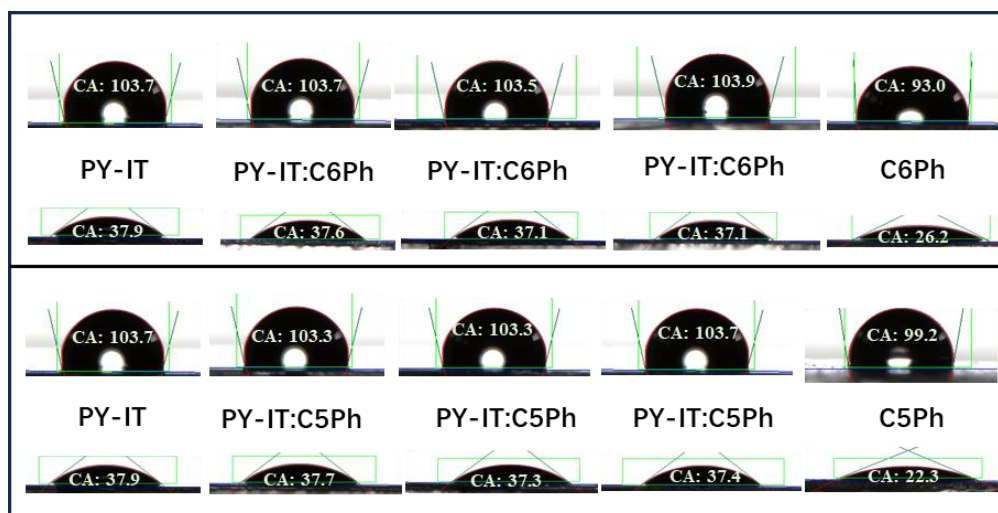


Fig. S12 Contact Angle images of the neat films and small molecular doped films.

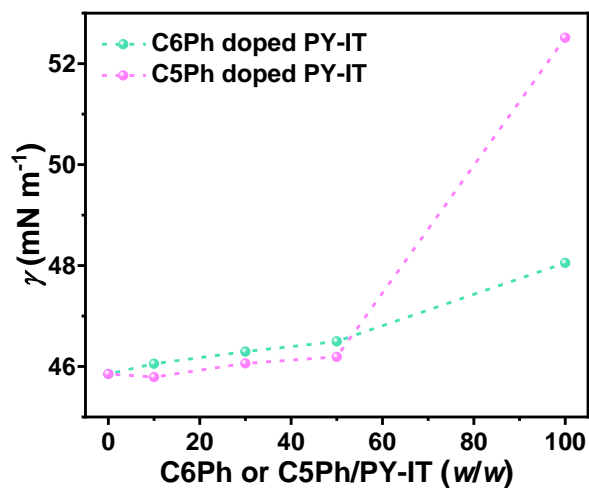


Fig. S13 Evolutions of surface energies of acceptor blends doping with small molecules with different contents.

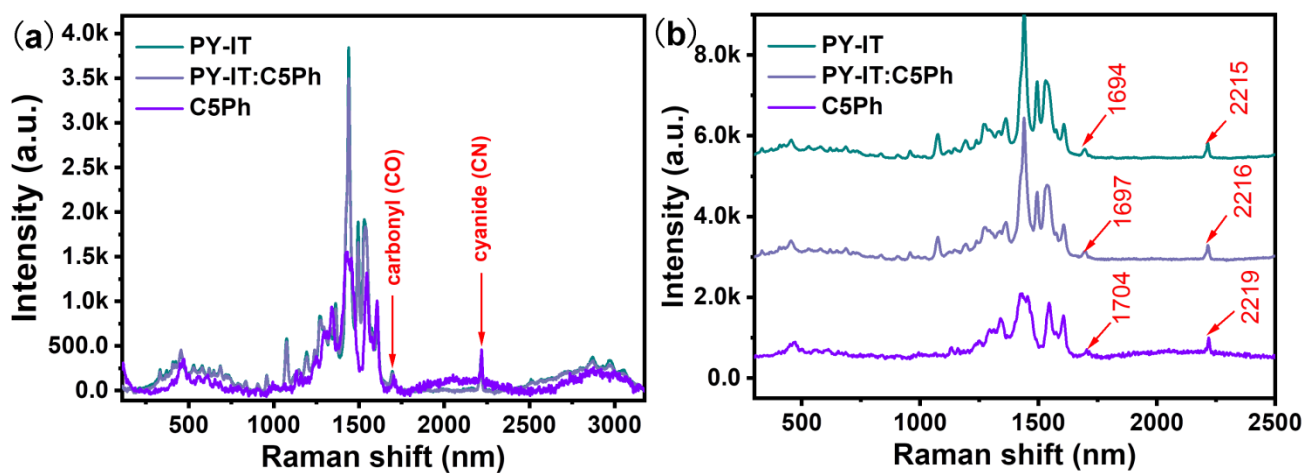


Fig. S14 The Raman spectroscopy of PY-IT, C5Ph, and PY-IT:C5Ph (1.2:0.1) films.

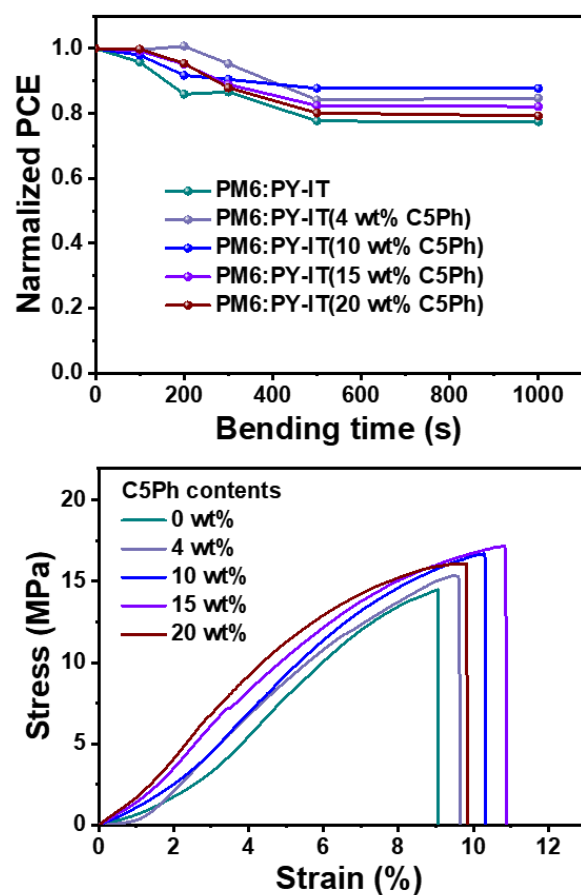


Fig. S15 The bending stability (average values from five devices) and stress-strain curves as for PM6:PY-IT based flexible devices and active layers with different C5Ph contents.

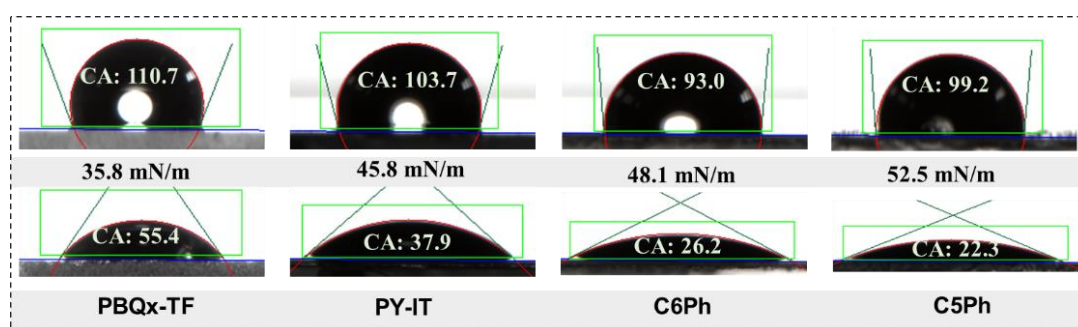


Fig. S16 Contact Angle images of PBQx-TF, PY-IT, and small molecules.

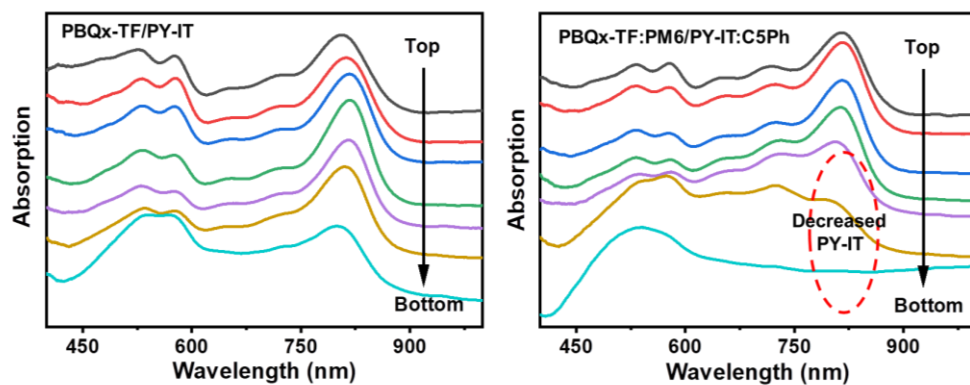


Fig. S17 Depth-dependent absorption spectra of the PPHJ active layers.

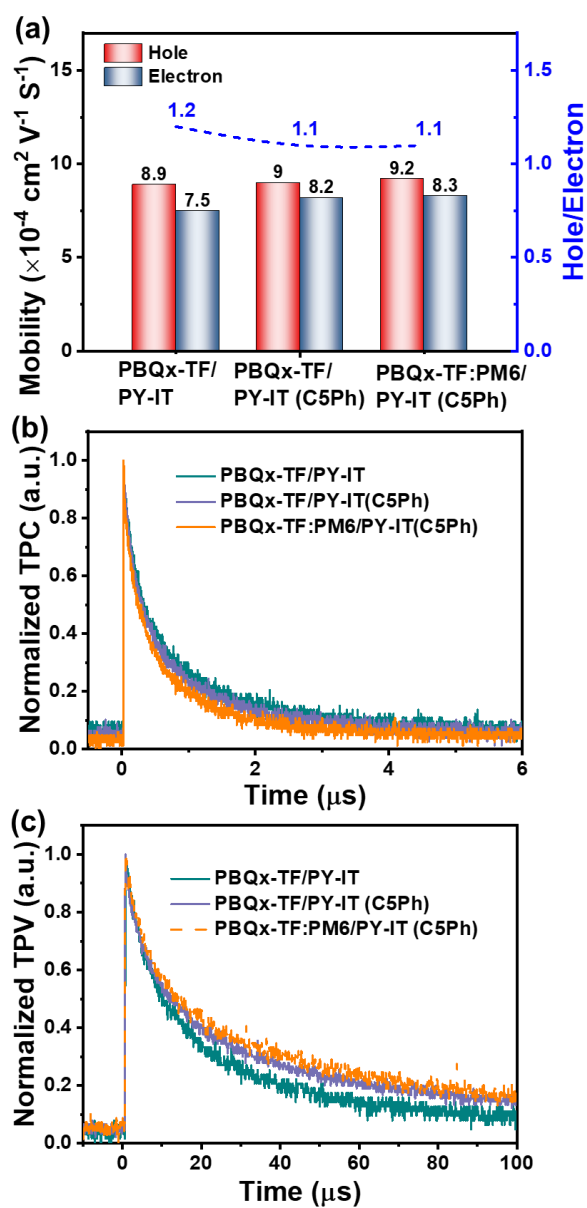


Fig. S18 (a) Hole and electron mobility obtained through SCLC method; TPC (b) and TPV (c) decays of the PBQx-TF/PY-IT, PBQx-TF/PY-IT (C5Ph) and PBQx-TF:PM6/PY-IT (C5Ph) based all-PSCs.

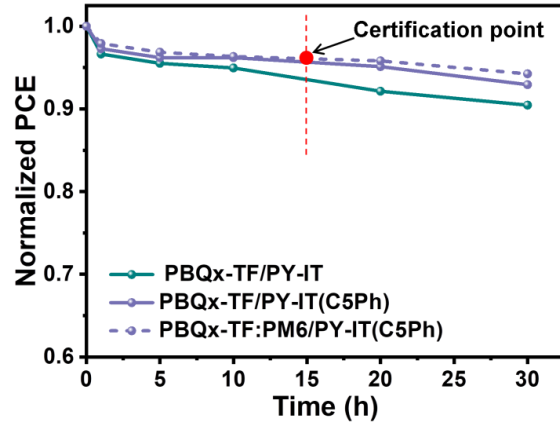


Fig. S19 The storage stability measurements of the encapsulated devices (average values from five devices).

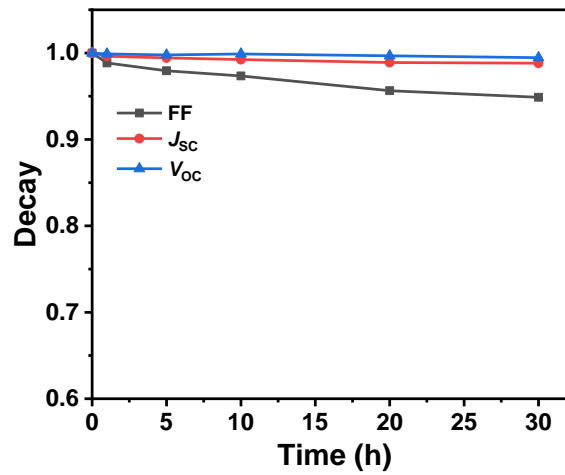


Fig. S20 The decay plots of FF, J_{sc} , and FF of the PBQx-TF:PM6/PY-IT (C5Ph) device under storage (average values from five devices).



检验检测报告 TEST REPORT

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无锡检验

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规格型号 MODEL/TYPE	有机太阳能电池 Organic Solar Cells
委托单位 APPLICANT	中国科学院青岛生物能源与过程研究所 Qingdao Institute of Bioenergy and bioprocess Technology, Chinese Academy of Sciences



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国家太阳能光伏产品质量检验检测中心
National Center of Inspection on Solar Photovoltaic Products Quality

检验检测报告
TEST REPORT

No: 2024DMCS20003

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产品名称 Sample Description	有机太阳能电池器件 Organic Solar Cells	规格型号 Model/Type	有机太阳能电池 Organic Solar Cells
		商 标 Trade Mark	-- --
标称生产单位 Nominal Producer	-- --		
委托单位名称\地址\邮编 Applicant Name \Add. \P. C.	中国科学院青岛生物能源与过程研究所\中国青岛市崂山区松岭路 189 号\-- Qingdao Institute of Bioenergy and bioprocess Technology, Chinese Academy of Sciences\No. 189 Songling Road, Laoshan District, Qingdao, China\--		
样品数量 Sample Quantity	1 片 1 piece	样品状态 Condition of Sample	符合检验要求 Comply with testing requirements
标称生产日期/批号 Nominal Date of Production /Lot No.	--/--	样品接收日期 Date of Sample Receiving	2024-01-08
检验检测日期 Testing Date(s)	2024-01-08	检验检测地点 Testing Location	本机构·新华路 CPVT·Xinhua Road
检验检测依据 Test In Accordance With	IEC 60904-1:2020 Photovoltaic devices - Part 1: Measurement of photovoltaic current-voltage characteristics		
判定依据 Decide In Accordance With	国家光伏质检中心 --		
检验结论 Conclusion	-- --	  签发日期: 2024-01-15 检验检测专用章 检验检测专用章 (2)	
备 注: Remarks:	-- --		

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Tested by
吕首首

样品描述 Sample Description

电池片 描述 Solar Cell Description	产品型号 Product model	有机太阳能电池 Organic Solar Cells
	电池片长 (mm) Length of solar cell (mm)	—
	电池片宽 (mm) Width of solar cell (mm)	—
	电池片类型 Solar cell type	其他 Others
备注: — Remark: —		



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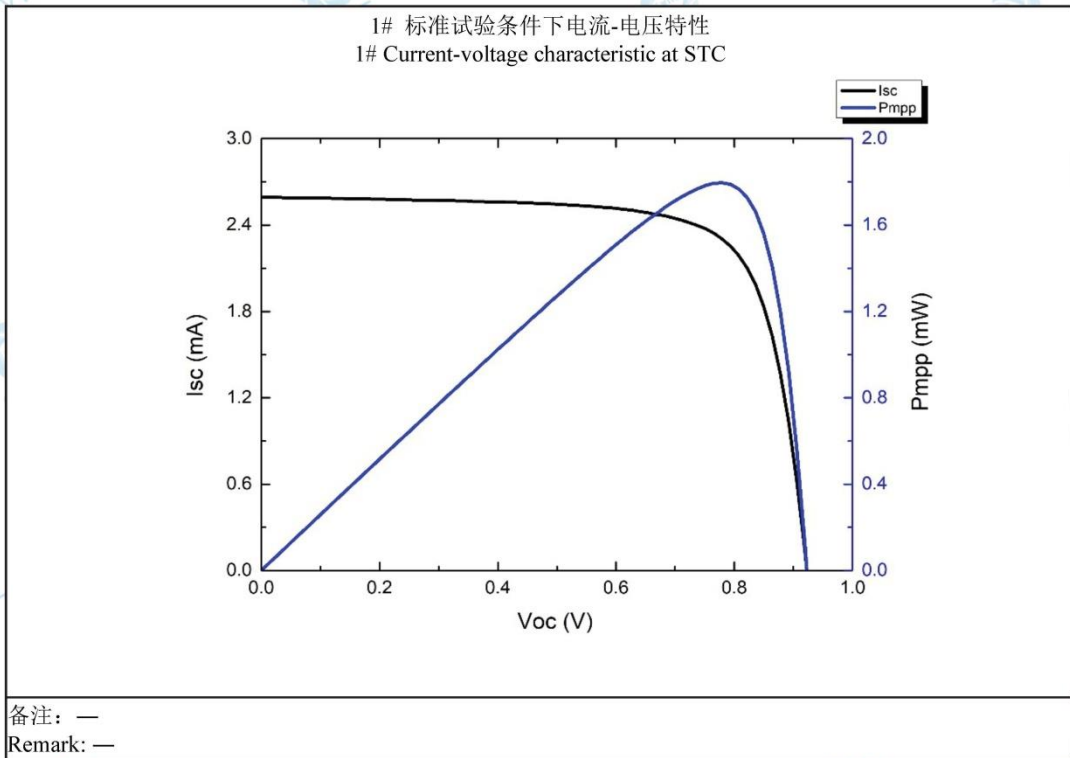
Test Results

序号 Clause	检验项目 Test item(s)	单位 Unit	技术要求 Technical requirements	结果 Results	单项评价 Verdict Pass/Fail
1	电流-电压特性的测量 (1#) Current-voltage characteristic measurement (1#)	—	在标准试验条件下（样品温度： 25℃，辐照度：1000W/m ² ，标准 太阳光谱辐照分布符合 IEC 60904-3 规定），测量样品随负荷 变化的电流-电压特性。 At STC (module temperature: 25℃, irradiance: 1000W/m ² , standard solar spectral irradiance distribution corresponds to IEC60904-3), measure the current-voltage characteristics of the cell with the variation of load.	—	—
1.1	开路电压 Voc Open-circuit voltage, Voc	V	—	0.9200	—
1.2	短路电流 Isc Short-circuit current, Isc	mA	—	2.592	—
1.3	最大功率 Pmax Maximum-power, Pmax	mW	—	1.795	—
1.4	最大功率点电压 Vmp Maximum-power voltage, Vmp	V	—	0.7799	—
1.5	最大功率点电流 Imp Maximum-power current, Imp	mA	—	2.301	—
1.6	填充因子 FF, % Fill factor FF, %	—	—	75.27	—
1.7	转换效率 η, % Conversion efficiency η, %	—	$\eta = \frac{P_{max}}{1000W/m^2 \times S} \times 100\%$ S 为样品面积/Sample area	18.20	—

备注：样品面积 S 为样品活性区域所确定的面积，S=0.099cm²
 Remark: Sample area S is determined by the active area of the sample, S=0.099cm²
 扫描方向从-0.2V~1.2V，步长为 0.014V。
 Sweep direction: -0.2V~1.2V, sweep steps: 0.014V.

检验检测结果

Test Results



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Fig. S21 The certification report from CPVT, China, as for PBQx-TF:PM6/PY-IT (C5Ph) device (after 15 hours of device preparation).

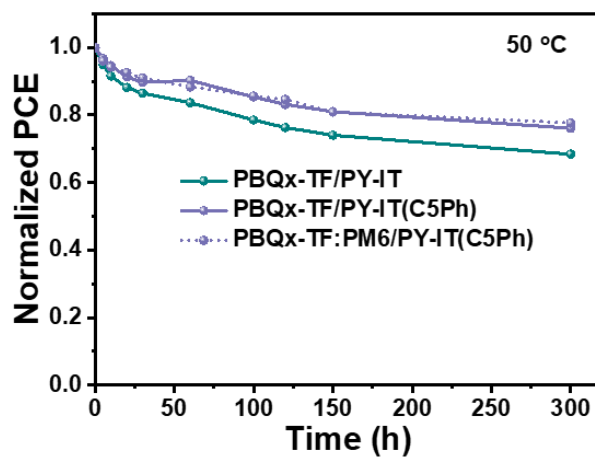


Fig. S22 Thermal stability of the all-PSCs under continuous annealing at 50 °C (average values from five devices).

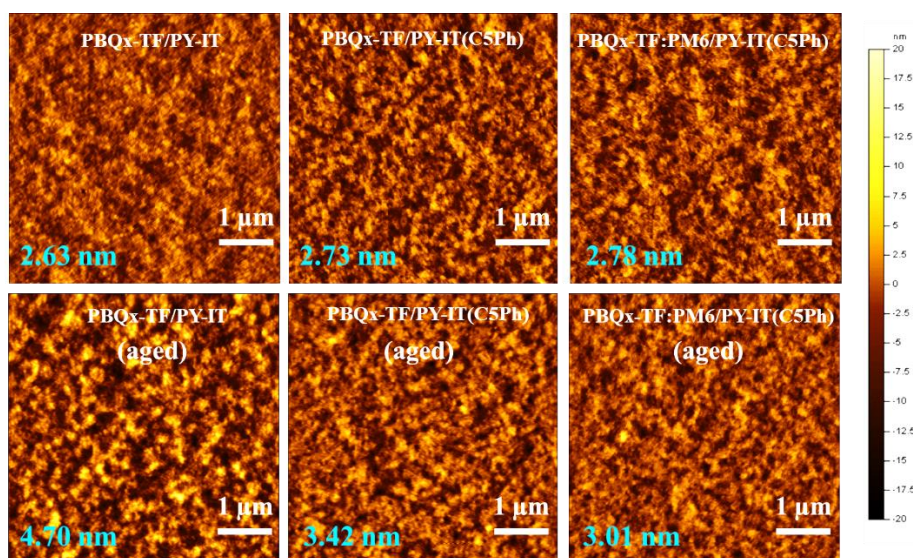


Fig. S23 AFM images of PBQx-TF/PY-IT, PBQx-TF/PY-IT (C5Ph) and PBQx-TF:PM6/PY-IT (C5Ph) films (fresh and aged for 30 hrs under continuous annealing at 50 °C) in glove box.

5.2 Supplementary Tables

Table S1 GIWAXS data of C6Ph and C5Ph neat films.

SMA	IP		OOP			
	(100) nm ⁻¹	<i>d</i> ₁₀₀ Å	(100) nm ⁻¹	(200) nm ⁻¹	(010) nm ⁻¹	<i>d</i> ₀₁₀ Å
C6Ph	3.57	17.60	-	-	18.06	3.48
C5Ph	3.58	17.55	3.43	6.85	18.99	3.31

Table S2 Crystal data and structure refinement for C6Ph.

CCDC number	2208806
Empirical formula	C ₁₀₀ H ₈₈ Cl ₁₈ N ₄ O ₂ S ₄
Formula weight	2144.08
Temperature/K	169.99(14)
Crystal system	triclinic
Space group	P-1
<i>a</i> /Å	9.8123(6)
<i>b</i> /Å	15.5877(7)
<i>c</i> /Å	18.3125(5)
α /°	71.984(4)
β /°	77.751(4)
γ /°	71.930(5)
Volume/Å ³	2511.2(2)
<i>Z</i>	1
ρ_{calc} /cm ³	1.418
μ /mm ⁻¹	5.676
F(000)	1102.0
Crystal size/mm ³	0.28 × 0.15 × 0.02
Radiation	Cu K α (λ = 1.54184)
2 Θ range for data collection/°	6.916 to 171.524
Index ranges	-12 ≤ <i>h</i> ≤ 12, -19 ≤ <i>k</i> ≤ 19, -22 ≤ <i>l</i> ≤ 23
Reflections collected	75394
Independent reflections	9976 [<i>R</i> _{int} = 0.1639, <i>R</i> _{sigma} = 0.0648]
Data/restraints/parameters	9976/0/587
Goodness-of-fit on <i>F</i> ²	2.003
Final <i>R</i> indexes [<i>I</i> >= 2 σ (<i>I</i>)]	<i>R</i> ₁ = 0.1351, <i>wR</i> ₂ = 0.3286
Final <i>R</i> indexes [all data]	<i>R</i> ₁ = 0.1749, <i>wR</i> ₂ = 0.3474
Largest diff. peak/hole / e Å ⁻³	0.91/-0.94

Table S3 Crystal data and structure refinement for C5Ph.

CCDC number	2299831
Empirical formula	C ₉ H ₇ Cl ₁₂ N ₄ O ₂ S ₄
Formula weight	1849.24
Temperature/K	169.98(10)
Crystal system	triclinic
Space group	P-1
a/Å	10.70790(10)
b/Å	19.7519(2)
c/Å	21.8828(2)
α/°	75.6190(10)
β/°	85.3700(10)
γ/°	82.8160(10)
Volume/Å ³	4442.08(8)
Z	2
ρ _{calc} /g/cm ³	1.383
μ/mm ⁻¹	4.708
F(000)	1908.0
Crystal size/mm ³	0.3 × 0.15 × 0.05
Radiation	Cu Kα (λ = 1.54184)
2θ range for data collection/°	4.174 to 151.334
Index ranges	-13 ≤ h ≤ 12, -24 ≤ k ≤ 24, -27 ≤ l ≤ 26
Reflections collected	62824
Independent reflections	17871 [R _{int} = 0.0346, R _{sigma} = 0.0310]
Data/restraints/parameters	17871/1/1073
Goodness-of-fit on F ²	1.062
Final R indexes [I ≥ 2σ (I)]	R ₁ = 0.0461, wR ₂ = 0.1235
Final R indexes [all data]	R ₁ = 0.0522, wR ₂ = 0.1286
Largest diff. peak/hole / e Å ⁻³	1.25/-0.77

Table S4 GIWAXS data of PBQ_x-TF:PY-IT, PBQ_x-TF:PY-IT (C6Ph), and PBQ_x-TF:PY-IT (C5Ph) active layers.

Active layer ^a	IP		OOP			
	(100) nm ⁻¹	d ₁₀₀ Å	(100) nm ⁻¹	d ₁₀₀ Å	(010) nm ⁻¹	d ₀₁₀ Å
PBQ _x -TF:PY-IT	3.28	19.16	3.91	16.07	17.53	3.58
PBQ _x -TF:PY-IT (C6Ph)	3.31	18.98	3.92	16.03	17.76	3.54
PBQ _x -TF:PY-IT (C5Ph)	3.32	18.93	3.91	16.07	17.78	3.53

^a The weight ratios of PBQ_x-TF:PY-IT (C6Ph or C5Ph) are 1:1.2:0.1.

Table S5 Photovoltaic parameters of rigid all-PSCs with different C6Ph or C5Ph contents.^a

Active layer (PBQx-TF:PY-IT)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
PBQx-TF:PY-IT	0.920 (0.919±0.03)	23.92 (23.85±0.25)	76.85 (76.52±0.52)	16.91 (16.68±0.16)
PBQx-TF:PY-IT (2 wt% C6Ph)	0.920 (0.920±0.03)	24.27 (24.02±0.39)	77.24 (77.05±0.42)	17.21 (17.05±0.21)
PBQx-TF:PY-IT (4 wt% C6Ph)	0.921 (0.920±0.03)	24.30 (24.12±0.22)	77.61 (77.21±0.48)	17.37 (17.19±0.14)
PBQx-TF:PY-IT (6 wt% C6Ph)	0.922 (0.921±0.03)	24.23 (24.20±0.21)	77.32 (77.14±0.31)	17.28 (17.11±0.17)
PBQx-TF:C6Ph	0.929 (0.928±0.03)	16.72 (16.55±0.23)	75.19 (74.89±0.52)	11.67 (11.52±0.26)
PBQx-TF:PY-IT (2 wt% C5Ph)	0.920 (0.921±0.03)	24.21 (24.01±0.24)	78.12 (77.77±0.51)	17.41 (17.26±0.22)
PBQx-TF:PY-IT (4 wt% C5Ph)	0.921 (0.921±0.04)	24.68 (24.53±0.27)	79.25 (78.45±0.62)	18.02 (17.81±0.16)
PBQx-TF:PY-IT (6 wt% C5Ph)	0.923 (0.922±0.04)	24.64 (24.50±0.31)	78.56 (78.16±0.45)	17.86 (17.53±0.19)
PBQx-TF:C5Ph	0.932 (0.920±0.03)	16.56 (16.41±0.19)	77.22 (76.89±0.46)	11.92 (11.76±0.21)

^a Average parameters with standard deviations obtained from ten devices.

Table S6 The exciton/charge parameters of all-PSCs.

Active layer	τ (ps)	P_{diss} (%)	P_{coll} (%)	τ_{ext} (μ s)	τ_{rec} (μ s)	n_{trap} (10^{16} cm ⁻³)
PBQx-TF:PY-IT	185	98.1	85.5	3.22	15.7	0.85
PBQx-TF:PY-IT (C6Ph)	118	98.6	87.1	2.18	16.1	0.72
PBQx-TF:PY-IT (C5Ph)	108	99.0	88.2	1.01	22.9	0.55

Table S7 The parameters of elongation at break, toughness, elastic deformation, elastic modulus, and yield strength of blend films with different C5Ph contents.

Blend films (PBQx-TF:PY-IT)	Elongation at break (%)	Toughness (MPa)	Elastic deformation (%)	Elastic modulus (MPa)	yield strength (MPa)
0 wt% C5Ph	9.80	87.25	4.80	242	11.65
4 wt% C5Ph	10.54	108.97	5.37	246	13.24
10 wt% C5Ph	11.61	137.07	6.06	247	14.98
15 wt% C5Ph	12.37	163.67	5.14	273	14.04
20 wt% C5Ph	11.02	135.14	4.86	265	12.87