Supporting information

Shamrock-shaped Non-fullerene Acceptors Enable High-Efficiency and High-Voltage Organic Photovoltaics

*Zongtao Wang[1,2#], Mengwei Ji[1,2#], Ailing Tang[2#], Mengzhen Du[1,2], Chenyu Mu[1,2], Yingliang Liu[1], Ergang Wang[3], Erjun Zhou[1,2]**

[1] School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, China

[2] National Center for Nanoscience and Technology, Beijing 100190, China

[3] Department of Chemistry and Chemical Engineering, Chalmers University of Technology, Göteborg, SE-412 96, Sweden

E-mail: zhouej@nanoctr.cn.

[#]These three authors contributed equally to this work.

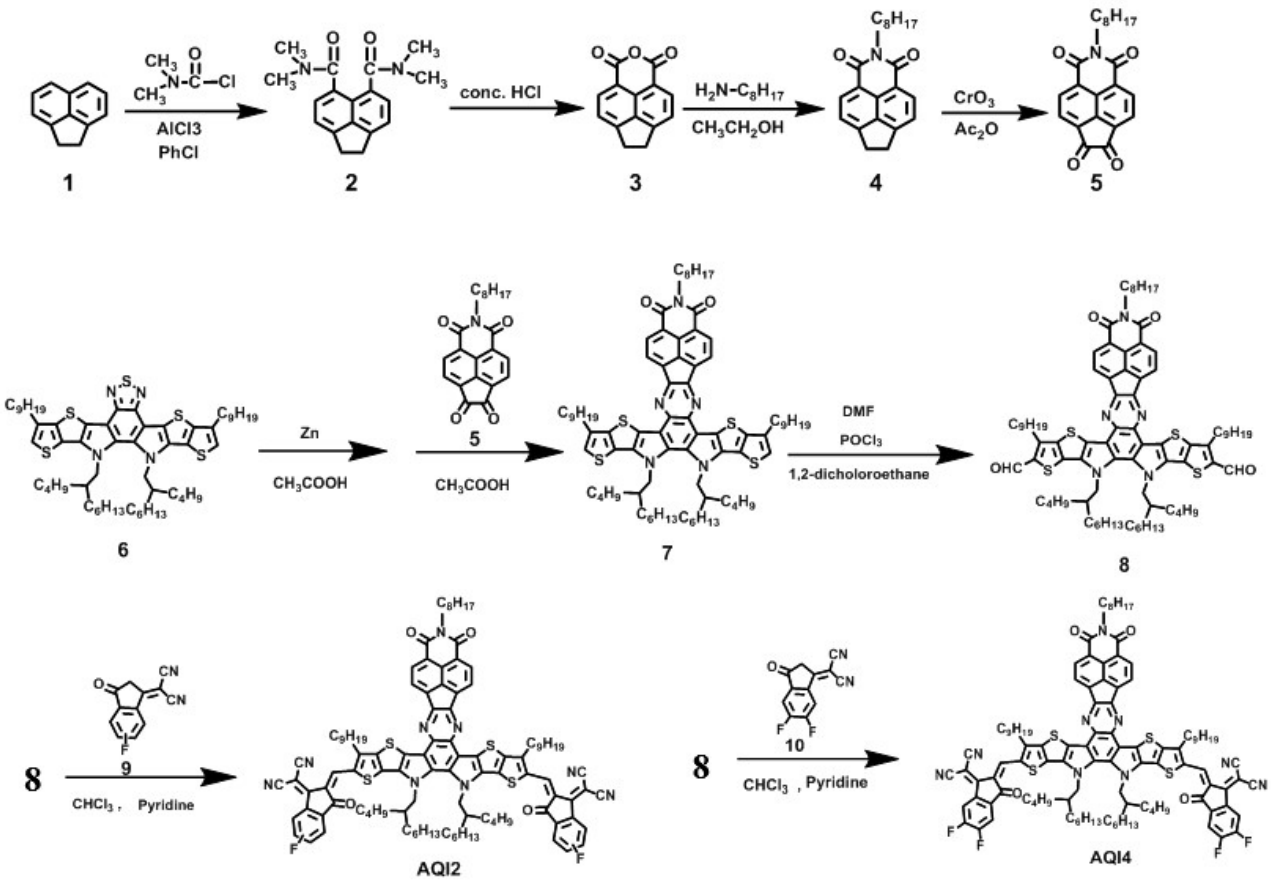
1. Materials and Synthesis

Reagents and Materials

All chemicals were obtained from commercially available sources and used without further purification. D18 and Y6 were purchased from Hyper, Inc. PNDIT-F3N were purchased from eFlexPV. PEDOT:PSS (CleviosTM P VP Al 4083) was purchased from Heraeus.

Synthesis of AQI2 and AQI4

Note: Compound 1, 6, 9 and 10 were purchased from commercial companies and used without further purifications. Compound 5 were synthesized according to the literature.[1]



Scheme S1. The synthetic routes of AQI2 and AQI4.

Synthesis of compound 7: Under nitrogen protection, zinc powder (490 mg, 4.5 mmol) was added

in the solution of compound 6 (500 mg, 0.3 mmol) in acetic acid (25 mL). Then the mixture solution was heated to 80°C and stirred for overnight. After the solution was cooled to room temperature, the solid was removed by filtration. Transfer the filtrate to a two-neck flask containing compound 5 (97 mg, 0.27 mmol), then the mixture solution was heated to 60°C for 6 h. After cooling to room temperature, washed with saturated salt water and dichloromethane. The solvent was removed under reduced pressure. The crude product was subsequently purified by column chromatography on silica gel to afford compound 7 as green solid (259 mg, 74% yield). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.65 (d, *J* = 7.3 Hz, 2H), 8.55 (d, *J* = 7.3 Hz, 2H), 7.06 (s, 2H), 4.67 (d, *J* = 7.8 Hz, 4H), 4.26-4.14 (m, 2H), 2.89 (t, *J* = 7.6 Hz, 4H), 2.23-2.13 (m, 2H), 1.92 (p, *J* = 7.5 Hz, 4H), 1.77 (p, *J* = 7.6 Hz, 2H), 1.35-1.24 (m, 35H), 1.01-0.78 (m, 40H), 0.64 (td, *J* = 7.5, 2.9 Hz, 12H).

Synthesis of compound 8: Under the protection of nitrogen, 2 mL DMF was added into the two-neck flask. Then, $POCl_3$ (0.1 mL, 1.9 mmol) was injected at 0°C. After being stirred at 0°C for 1 h, the mixture of compound 7 (259 mg, 0.19 mmol) and 1, 2-dichloroethane (5 mL) was directly injected into the two-neck flask. Next, the mixture was stirred at room temperature for overnight, then was cooled to 0 °C (50 mL), neutralized with aqueous $NaHCO_3$ for another 4 h. Washed with $NaHCO_3$ aqueous solution and dichloromethane. The solvent was removed under reduced pressure. The crude product was subsequently purified by column chromatography on silica gel to afford compound 8 as green solid (192 mg, 72% yield). ^{1}H NMR (400 MHz, Chloroform-d) δ 10.18 (s, 2H), 8.68 (dd, *J* = 11.3, 7.3 Hz, 2H), 8.58 (dd, *J* = 19.7, 7.3 Hz, 2H), 4.71 (d, *J* = 7.7 Hz, 4H), 4.22 (q, *J* = 7.6, 6.5 Hz, 2H), 3.27 (q, *J* = 7.6, 6.6 Hz, 4H), 2.15 (d, *J* = 4.9 Hz, 2H), 1.97 (q, *J* = 8.3, 7.7 Hz, 4H), 1.83-1.71 (m, 2H), 1.49-1.23 (m, 35H), 1.13-0.82 (m, 40H), 0.64 (q, *J* = 7.0 Hz, 12H).

Synthesis of AQI2: Compound 8 (85 mg, 0.06 mmol) and compound 9 (53 mg, 0.25 mmol) were dissolved into dry chloroform (10 mL) in a two-neck flask. The solution was flushed with nitrogen for 5 min. After 0.3 mL pyridine were added, the mixture was stirred at 65°C overnight. After cooling to room temperature, the reaction mixture was poured into water and extracted several times with chloroform. Then the solvent was removed under reduced pressure, and the crude product was purified by column chromatography on silica gel to yield AQI2 as red solid (85 mg, 78% yield). ^{1}H NMR (400 MHz, Chloroform-d) δ 9.09 (d, *J* = 10.1 Hz, 2H), 8.66 (d, *J* = 12.9 Hz, 1H), 8.45 (d, *J* = 7.2 Hz, 2H),

8.29 (d, *J* = 9.0 Hz, 1H), 8.15 (d, *J* = 7.2 Hz, 2H), 8.01-7.95 (m, 1H), 7.59 (s, 1H), 7.48-7.38 (m, 2H), 4.90 (d, *J* = 8.0 Hz, 4H), 4.22 (s, 2H), 3.20 (d, *J* = 8.8 Hz, 4H), 2.34 (s, 2H), 1.81 (d, *J* = 32.8 Hz, 6H), 1.40-0.99 (m, 63H), 0.84 (t, *J* = 6.8 Hz, 12H), 0.71 (d, *J* = 30.3 Hz, 12H).

Synthesis of AQI4**:** Compound 8 (91 mg, 0.07 mmol) and compound 10 (61 mg, 0.28 mmol) were dissolved into dry chloroform (15 mL) in a two-neck flask. The solution was flushed with nitrogen for 5 min. After 0.3 mL pyridine were added, the mixture was stirred at 65°C overnight. After cooling to room temperature, the reaction mixture was poured into water and extracted several times with chloroform. Then the solvent was removed under reduced pressure, and the crude product was purified by column chromatography on silica gel to yield AQI4 as red solid (84 mg, 71% yield). ^{1}H NMR (400 MHz, Chloroform-d) δ 9.07 (s, 2H), 8.52 (d, *J* = 7.2 Hz, 2H), 8.46 (dd, *J* = 9.9, 6.4 Hz, 2H), 8.30 (d, *J* = 7.2 Hz, 2H), 7.71 (t, *J* = 7.5 Hz, 2H), 4.90 (d, *J* = 8.1 Hz, 4H), 4.21 (t, *J* = 7.7 Hz, 2H), 3.19 (t, *J* = 7.8 Hz, 4H), 2.33 (s, 2H), 1.91-1.72 (m, 6H), 1.44-1.06 (m, 63H), 0.87 (d, *J* = 19.5 Hz, 12H), 0.77-0.66 (m, 12H).

2. Measurements and Characterizations

Materials Characterization

Thermo gravimetric analysis (TGA) was recorded on Diamond TG/DTA under the protection of nitrogen at a heating rate of 10 ℃ min^{-1}. UV-vis spectra were tested with UV-3600i PLUS (Shimadzu Corporation). Grazing incidence wide-angle X-ray scattering (GIWAXS) analyses were investigated at the XEUSS SAXS/WAXS equipment. The data were obtained with an area Pilatus 100k detector with a resolution of 195 × 487 pixels (0.172 mm × 0.172 mm). The X-ray wavelength was 1.54 Å, and the incidence angle was 0.2°. The samples were spin-coated onto the Si substrate.

Electrochemical Characterizations

Cyclic voltammetry (CV) was recorded on a CH instruments electrochemical workstation using glassy carbon discs as the working electrode, Pt wire as the counter electrode, Ag/AgCl electrode as the reference electrode. 0.1 mol L^{-1} tetrabutylammonium-hexafluorophosphate (Bu4NPF6) dissolved in methanol was employed as the supporting electrolyte, which was calibrated by the ferrocene/ferrocenium (Fc/Fc+) as the redox couple. Thin films were formed by drop-casting of solutions in chloroform on the working electrode and then dried in air. The HOMO and LUMO energy

level can be calculated by the equations:

$$E_{HOMO} = -\,e\,(4.8 + E_{ox} - E^{1/2}_{Fc+/Fc})\ V$$

$$E_{LUMO} = -\,e\,(4.8 + E_{red} - E^{1/2}_{Fc+/Fc})\ V$$

Device Fabrication

OPVs were fabricated with a conventional structure of ITO/PEDOT:PSS/active layers/PNDIT-F3N/Ag. The ITO-coated glass substrates were sequentially cleaned in detergent deionized water and ethanol for 15 min each at room temperature. A 20 nm thick PEDOT:PSS layer was firstly spin-casted on top of the ITO substrates at 3000 rpm for 30 s and then annealed at 150℃ for 10 min under ambient atmosphere. The solution for D18:Y6 (1:1.6, w/w, 10.92 mg mL^{-1} in total), D18:AQI2 (1:1.2, w/w, 9.24 mg mL^{-1} in total), and D18:AQI4 (1:1.5, w/w, 10.5 mg mL^{-1} in total) with was prepared in chloroform and was stirred at 80 C° for 2 hours. After cooling down for one minute, the active layer solutions was spin-coated on the top of PEDOT:PSS at a speed of 4000 rpm for 30 s and then CF solvent vapor annealing was last for 8 min in an N_2-filled glove box. Then, the methanol solution of PNDIT-F3N (1 mg mL^{-1}) was spin-coated on the top of active layer at a speed of 3000 rpm for 30 s. Finally, a 100 nm-thick metal silver electrode was thermally deposited under vacuum conditions of 3×10^{-4} Pa. The active area of device is 5 mm^2 and mask area is 3.14 mm^2.

Device Characterization

The *J-V* curves were measured using a Zolix Solar IV-150A-ZZU system, all the devices are not encapsulated and they are performed in a glovebox filled with N_2. The photocurrent was tested under AM 1.5 G illumination at 100 mW cm^{-2} using a Zolix HPS-300XA solar simulator. Light intensity was calibrated with a Zolix QE-B1 Si-based solar cell. The EQE spectrum was carried out by using a Zolix SCS10-X150-DSSC-ZZU system.

Highly sensitive EQE, EQE_{EL} and EL measurements

Highly sensitive EQE was measured using an integrated system (PECT-600, Enlitech), where the photocurrent was amplified and modulated by a lock-in instrument. Electroluminescence quantum efficiency (EQE_{EL}) and electroluminescence (EL) measurements were performed by applying external voltage/current sources through the devices (ELCT-3010, Enlitech).

Transient Absorption Spectroscopy

TA spectroscopy was tested by a regenerative amplified Ti:sapphire laser system (Coherent) as the laser source and EOS spectrometer (Ultrafast Systems LLC) as the spectrometer, pump beam excitation intensity at 15.9 μJ cm^{-2}.

Exciton diffusion length Measurements

TA spectroscopy is employed to measure exciton lifetimes as a function of excitation density. When exciton annihilation occurs in the film, the exciton decay is accelerated with increasing excitation fluence. The excitation fluences are 1.59 and 15.9 μJ cm^{-2}. The exciton decay can be globally fit to a rate equation accounting for exciton annihilation and first-order recombination of the excitons:

$$-\frac{dn(t)}{dt} = kn(t) + \gamma n^2(t) \quad (1)$$

which has the following solution:

$$n(t) = \frac{n(0)exp(-kt)}{1 + \frac{\gamma}{k}n(0)[1 - exp(-kt)]} \quad (2)$$

where, $n(t)$ is the exciton density at a decay time of t, k is the monomolecular decay rate and γ is the singlet−singlet bimolecular exciton annihilation rate. Here, the intrinsic k, was extracted from the dilute solution where diffusion and annihilation does not occur. The γ are obtained by fitting Equation 2. Then, the diffusion coefficient D can be obtained through the following equation:

$$D = \frac{\gamma}{8\pi R} \quad (3)$$

where R is the annihilation radius of singlet excitons. R is assumed to be 1 nm. Then L_D are calculated based on the

$$L_D = (D\tau)^{1/2} \quad (4)$$

Here, the intrinsic exciton lifetime, τ, is obtained from the dilute solution measurement.

Fluorescence Measurement

PL, TRPL and PLQY measurements were detected by Edinburgh Fluorescence Spectrometer (FLS 980).

AFM and TEM Measurements

AFM was tested by Bruker Dimension Icon. TEM was tested bu JEOL JEM-2100Plus. The samples prepared with the optimized device fabrication conditions.

Mobility Measurements

The electron-only and hole-only devices were fabricated by using the device structure of ITO/TIPD/active layer/PNDIT-F3N/Ag and ITO/PEDOT:PSS/active layer/Au, respectively. The electron mobilities were approximated by the Mott-Gurney equation:

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

where J stands for current density, ε_0 is the permittivity of free space (8.85×10^{-12} C V^{-1} m^{-1}), ε_r is the relative permittivity of the transport medium (assuming that of 3.0), μ is the charge mobility, V is the internal potential in the device, and L is the thickness of the active layer.

Supplementary Figures

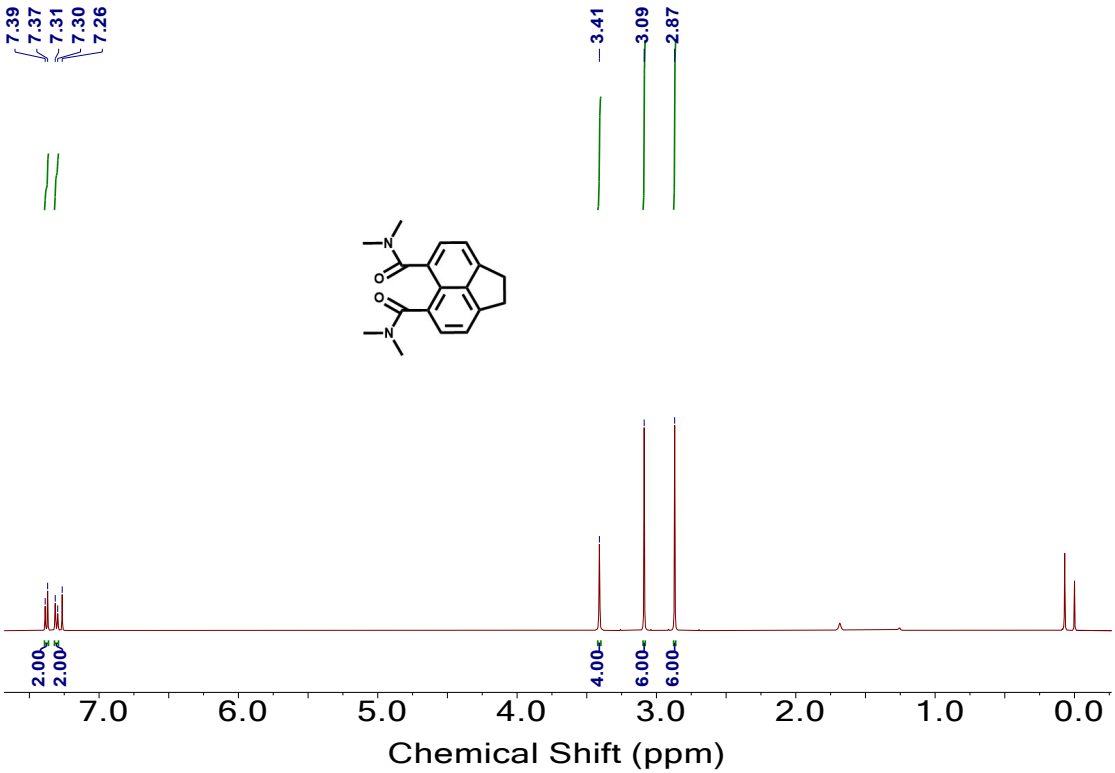


Figure S1. [1]H NMR spectrum of Compound 2.

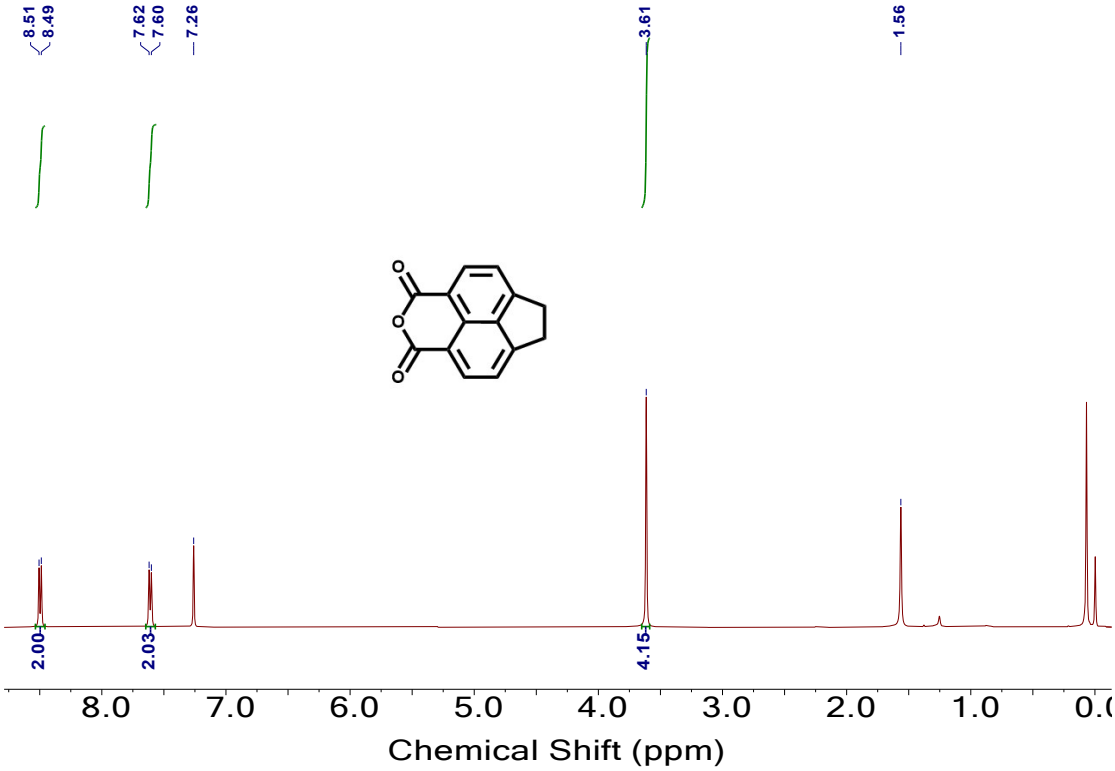


Figure S2. [1]H NMR spectrum of Compound 3.

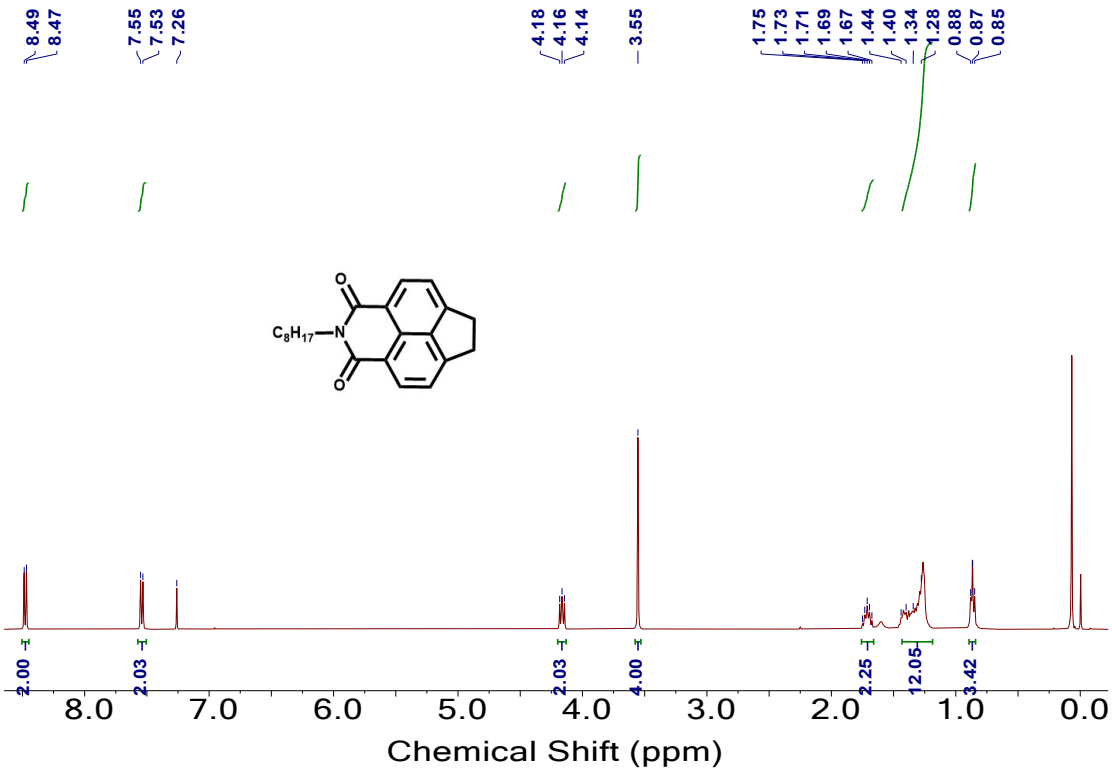


Figure S3. [1]H NMR spectrum of Compound 4.

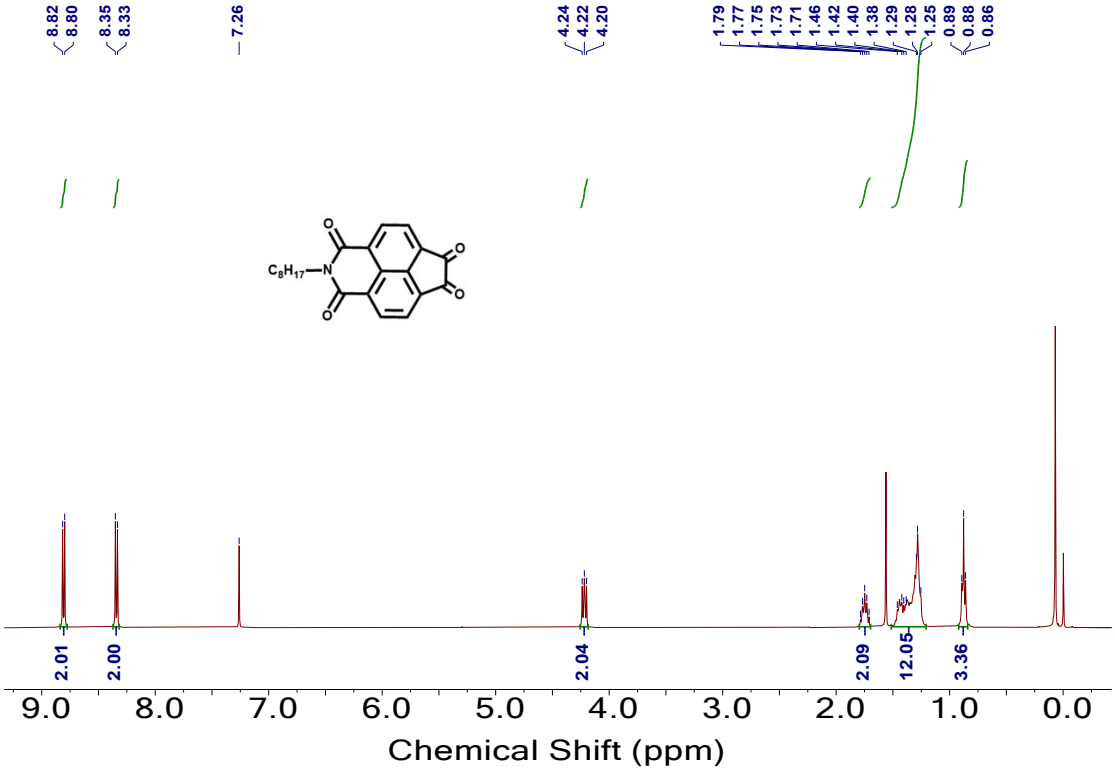


Figure S4. [1]H NMR spectrum of Compound 5.

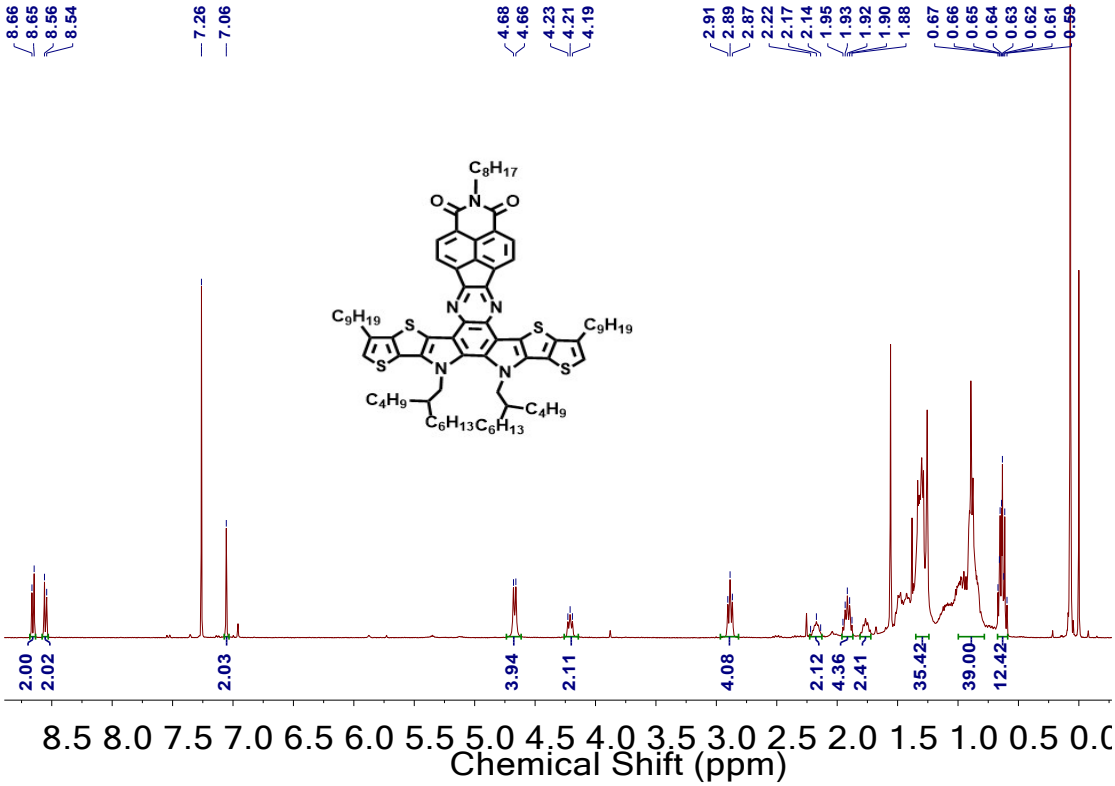


Figure S5. [1]H NMR spectrum of Compound 7.

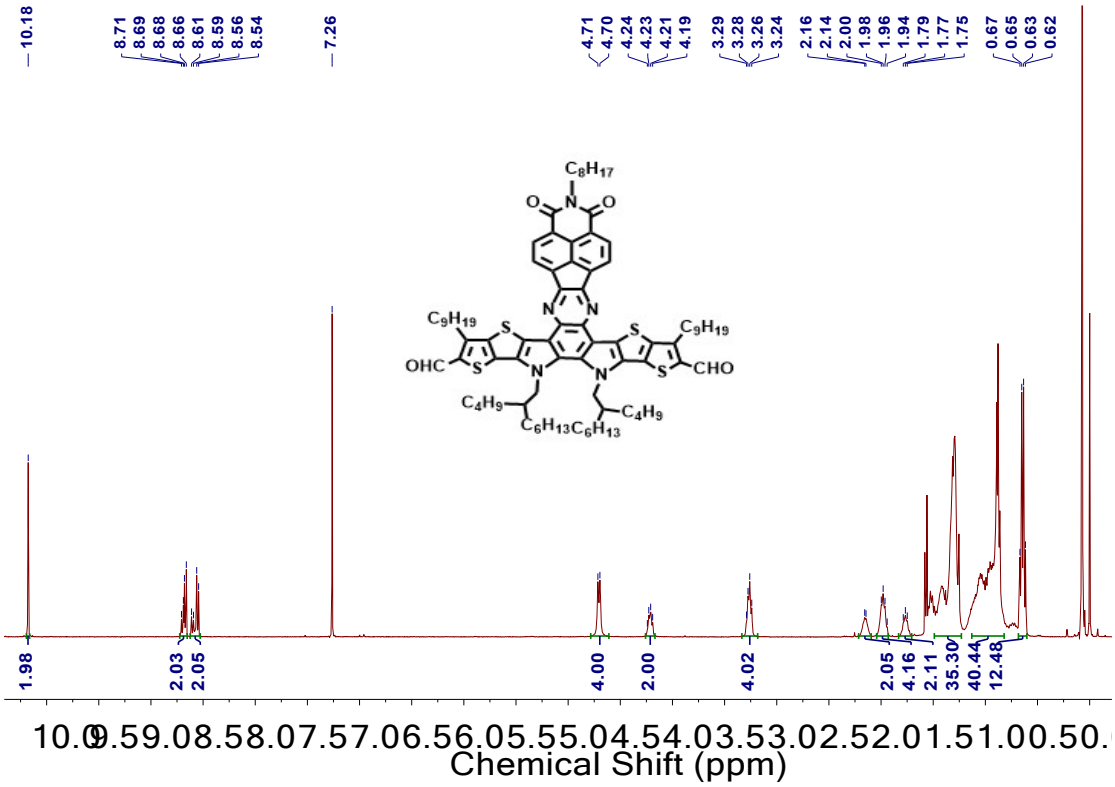


Figure S6. [1]H NMR spectrum of Compound 8.

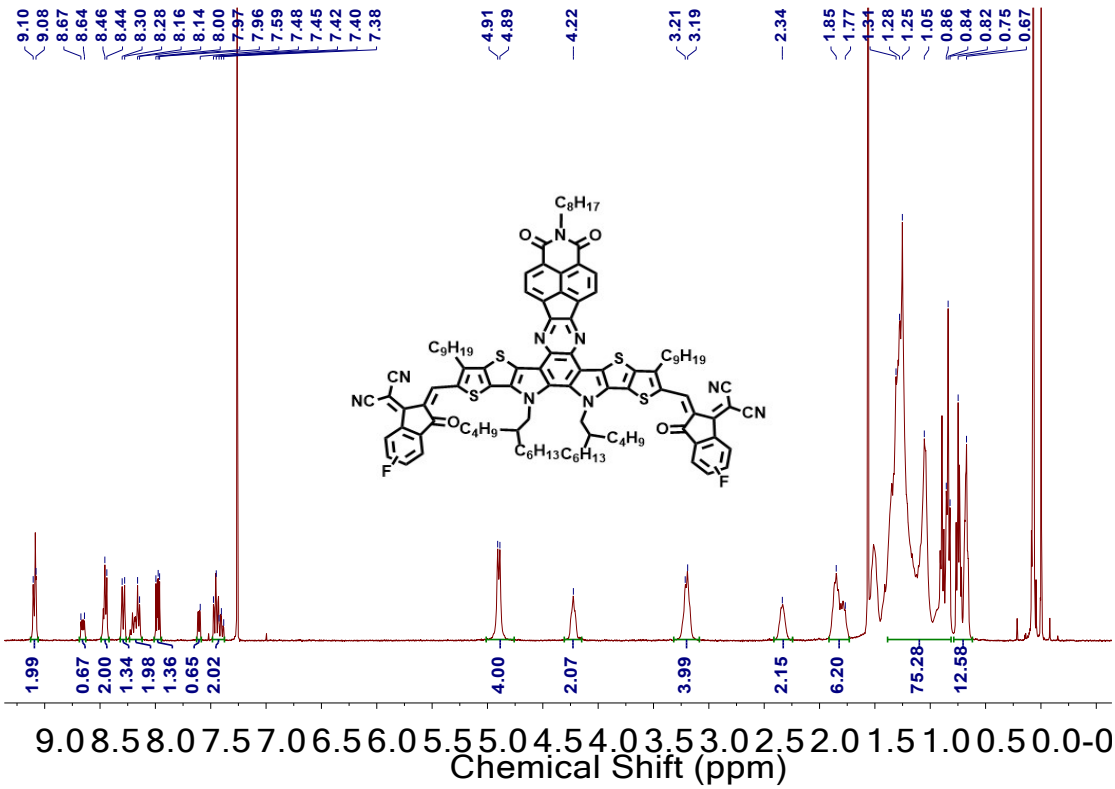


Figure S7. [1]H NMR spectrum of AQI2.

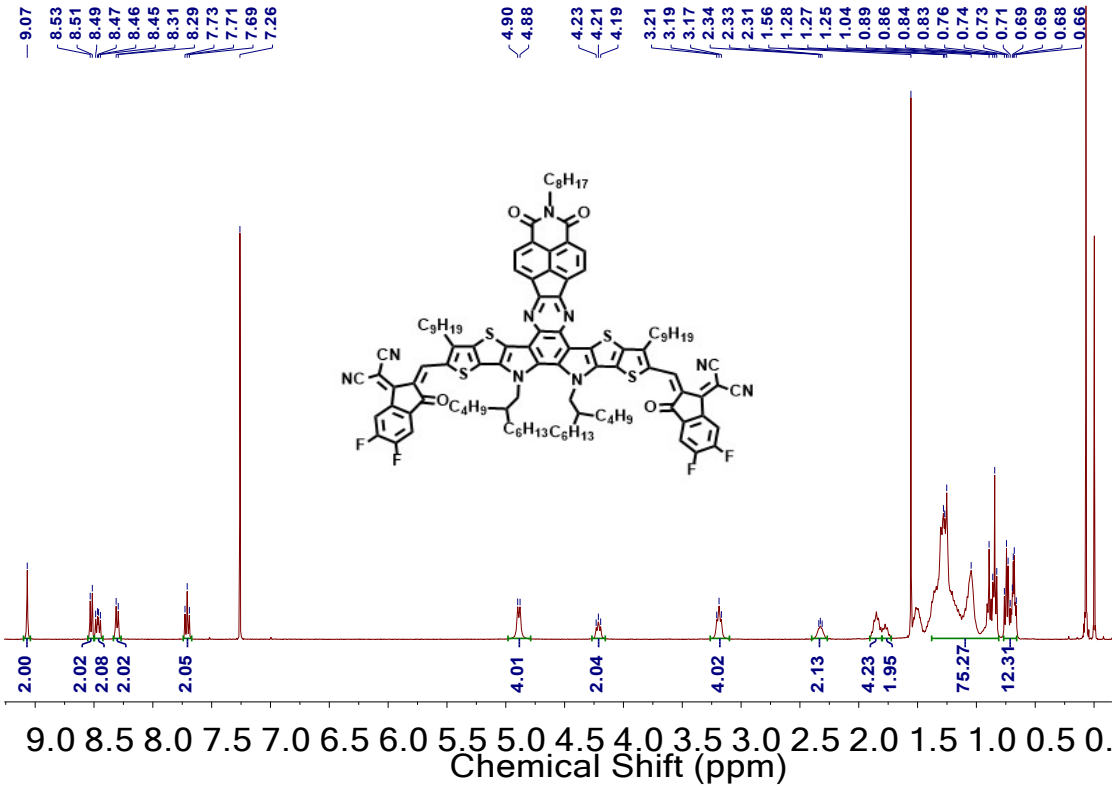


Figure S8. [1]H NMR spectrum of AQI4.

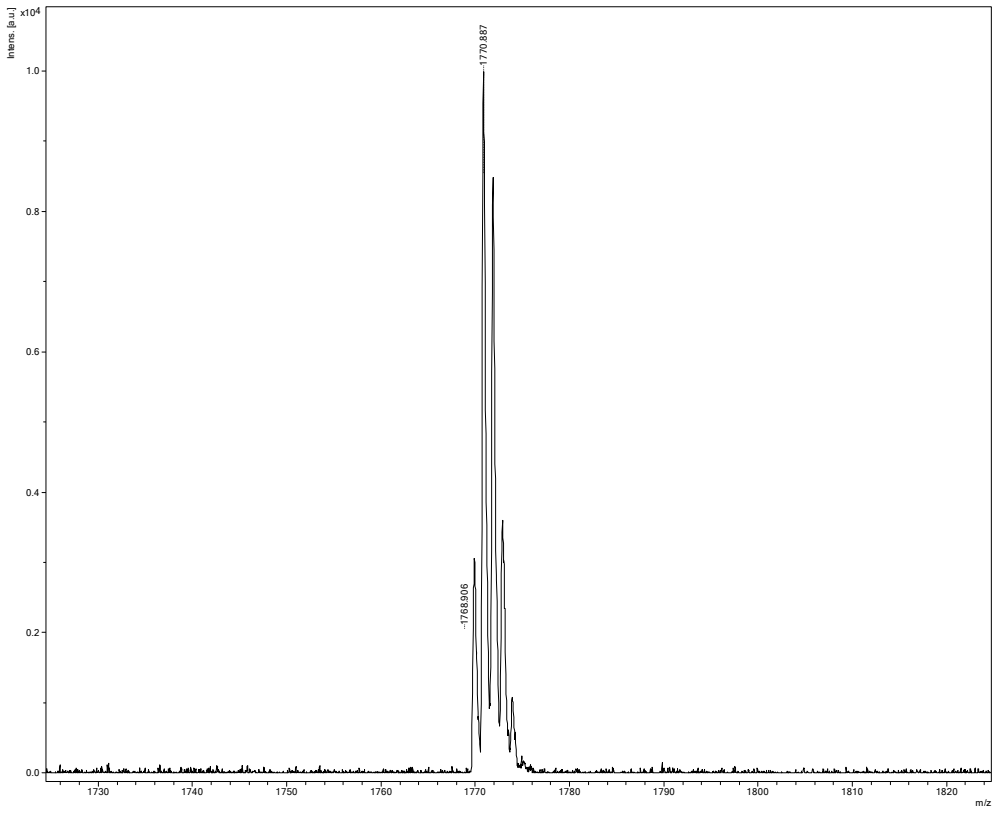


Figure S9. The mass spectrum (MALDI-TOF) of AQI2.

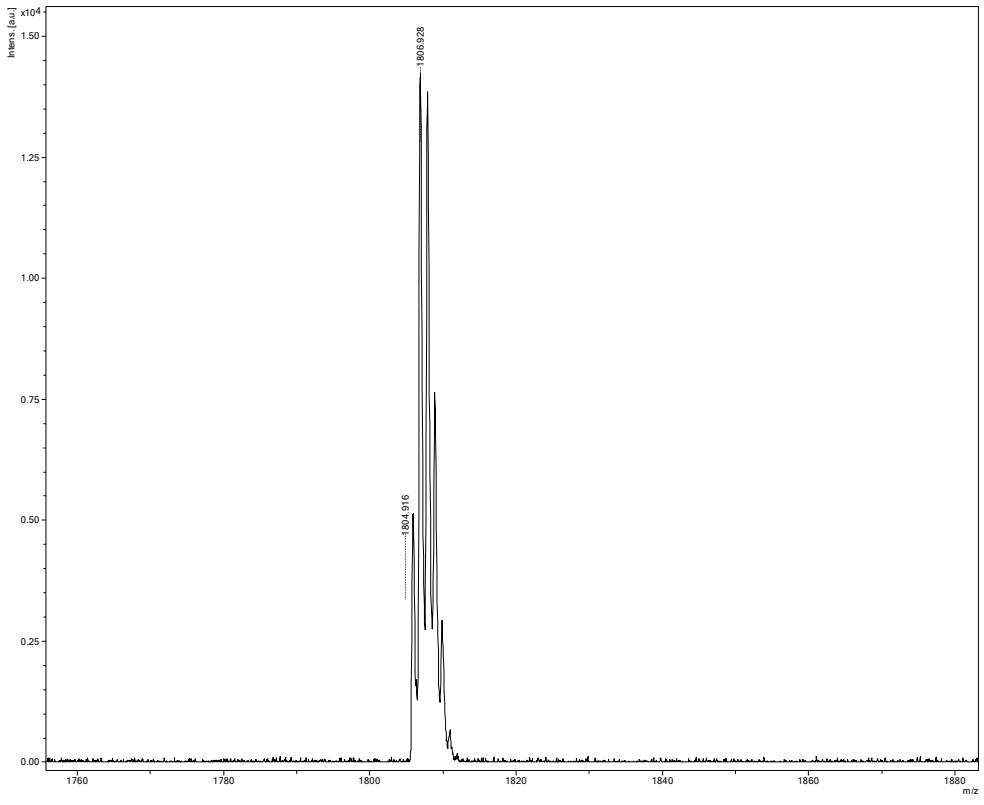


Figure S10. The mass spectrum (MALDI-TOF) of AQI4.

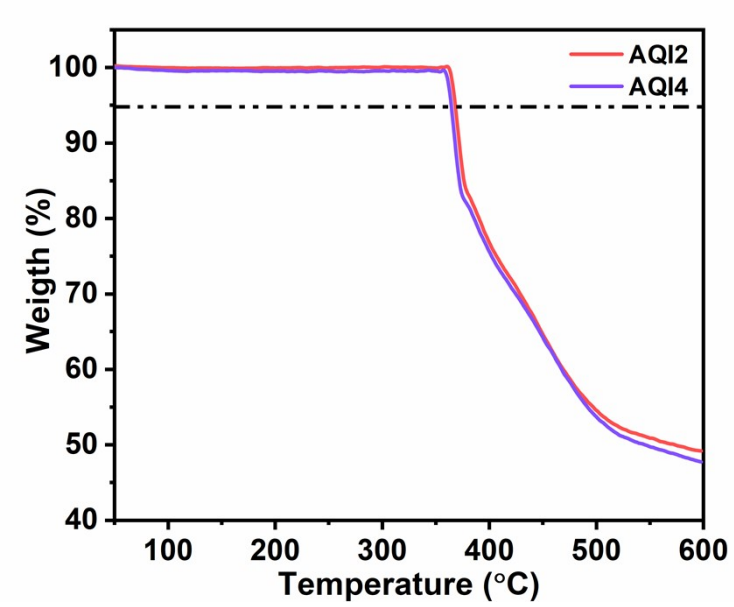
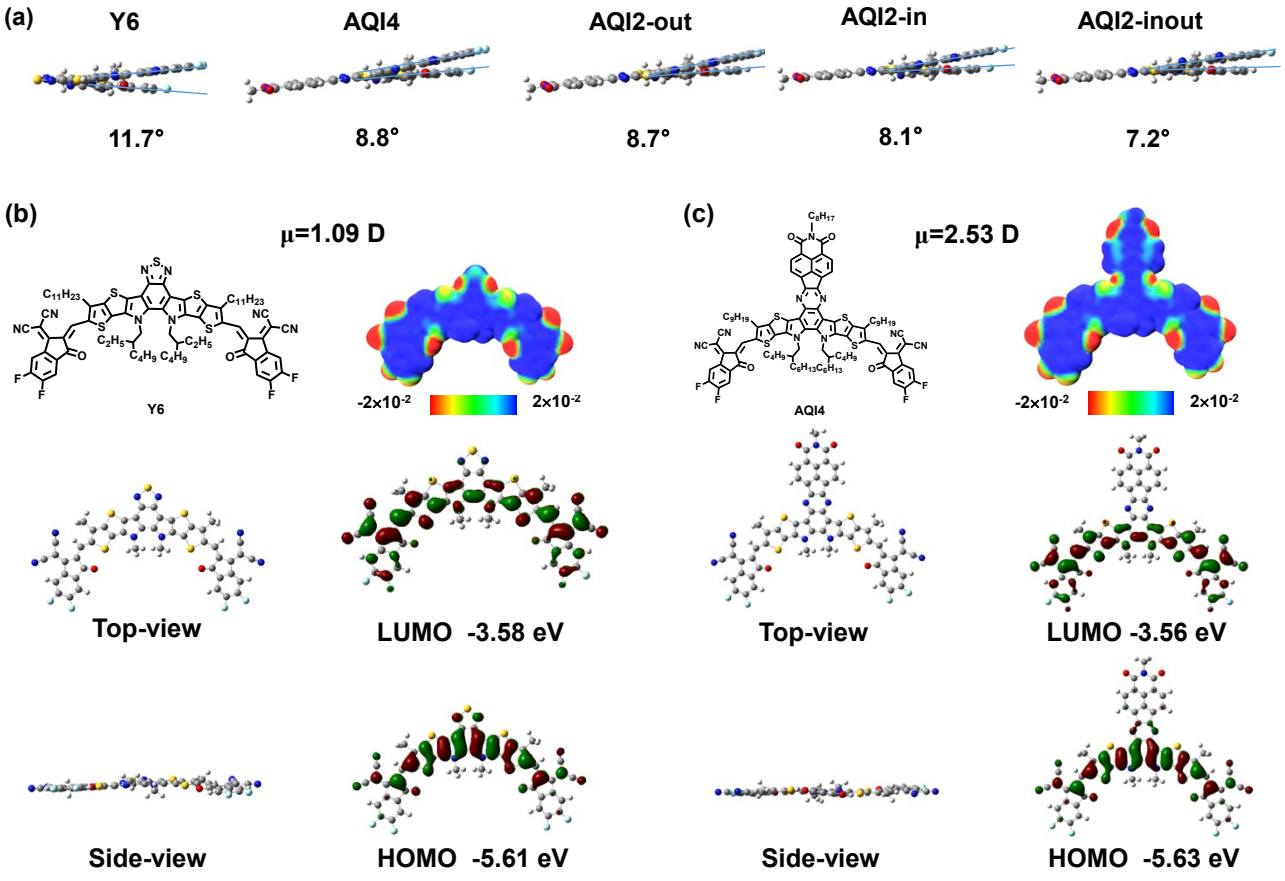


Figure S11. TGA of AQI2 and AQI4.



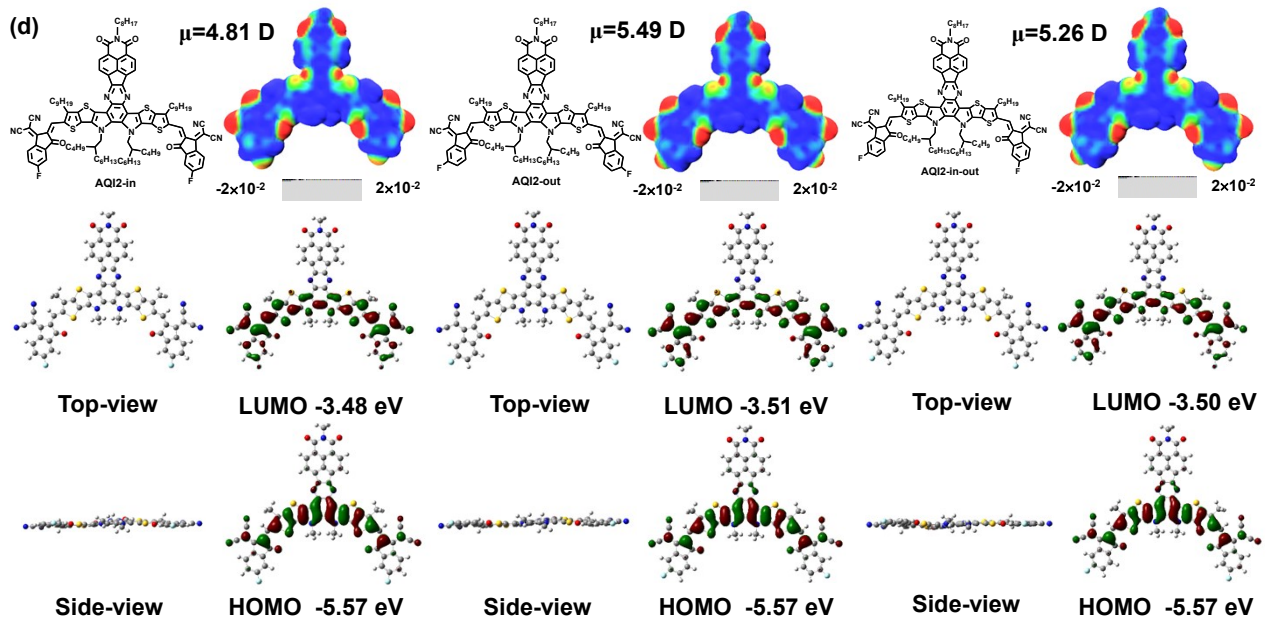


Figure S12. (a) The molecular torsion angle consisting of planes built from the two end groups of Y6, AQI4, and AQI2. The side view, top view, molecular orbital energy levels, and molecular electrostatic potential of (b) Y6, (c) AQI4, and (d) AQI2-in, AQI2-out, and AQI2-in-out.

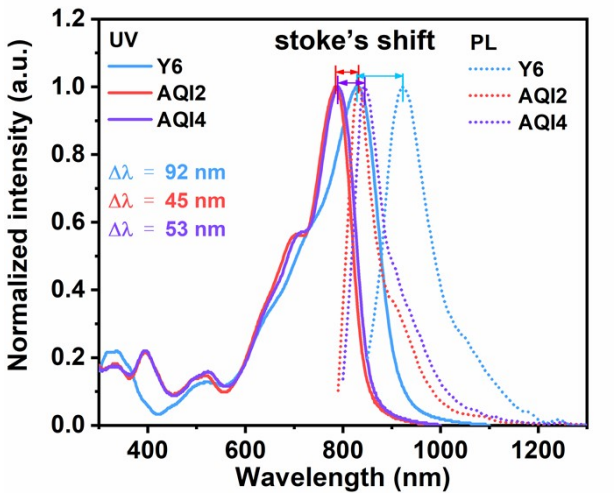


Figure S13. Stoke's shift of Y6, AQI2 and AQI4.

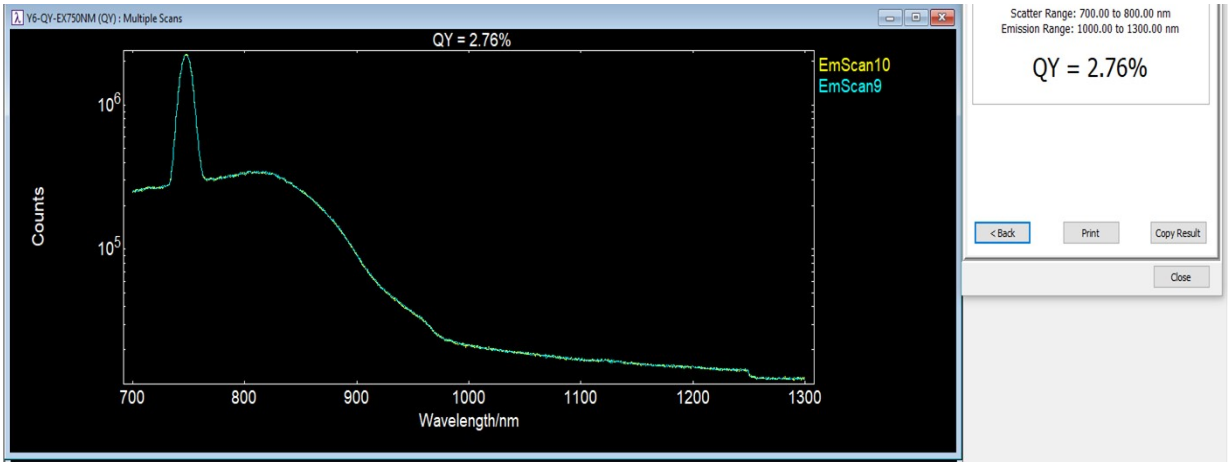


Figure S14. PLQY of Y6.

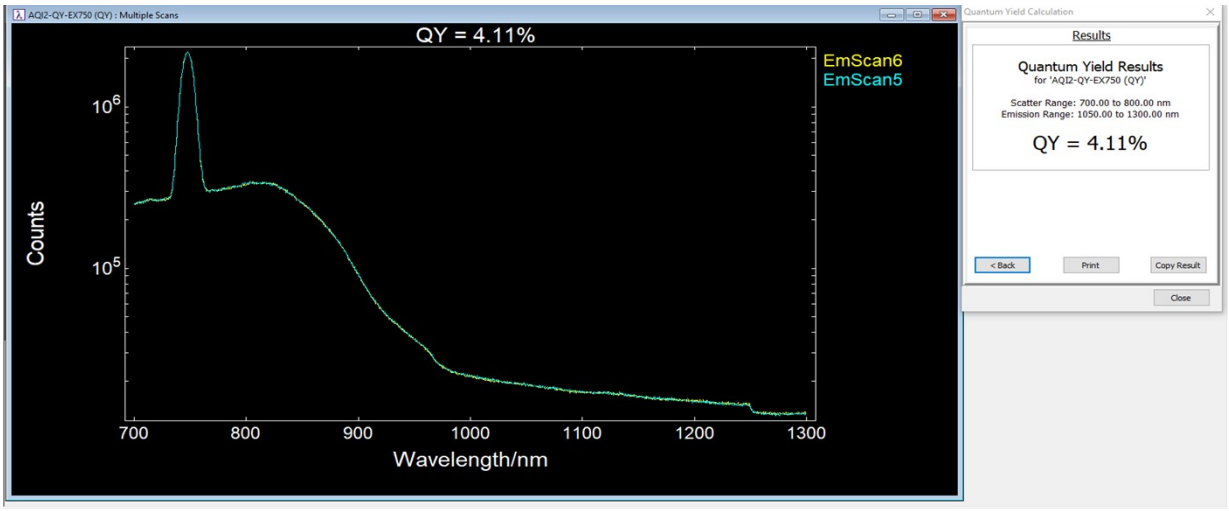


Figure S15. PLQY of AQI2.

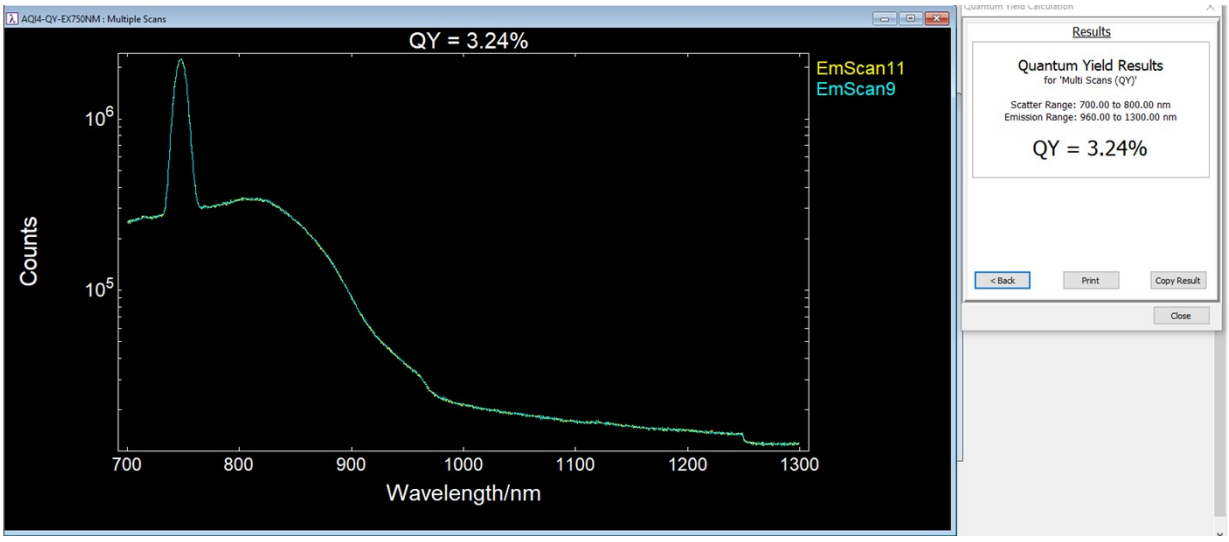


Figure S16. PLQY of AQI4.

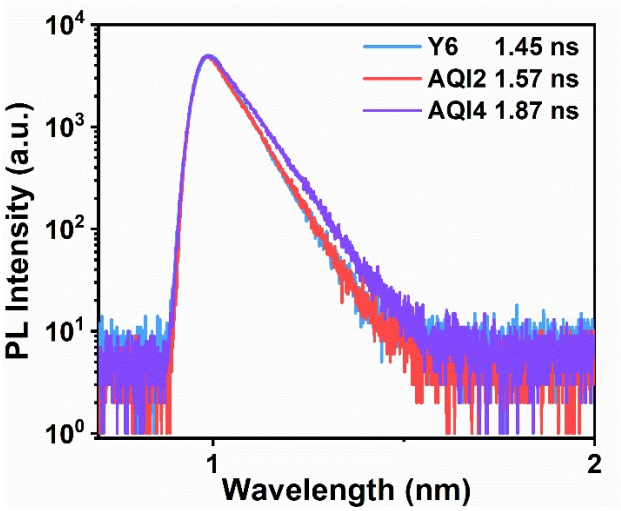


Figure S17. The time-resolved photoluminescence spectroscopy of Y6, AQI2, and AQI4 pure films.

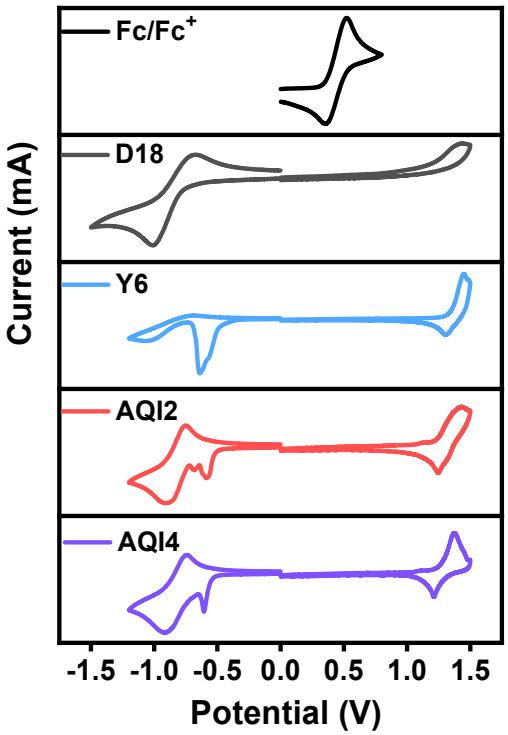


Figure S18. Cyclic voltammetry curves of D18, Y6, AQI2 and AQI4.

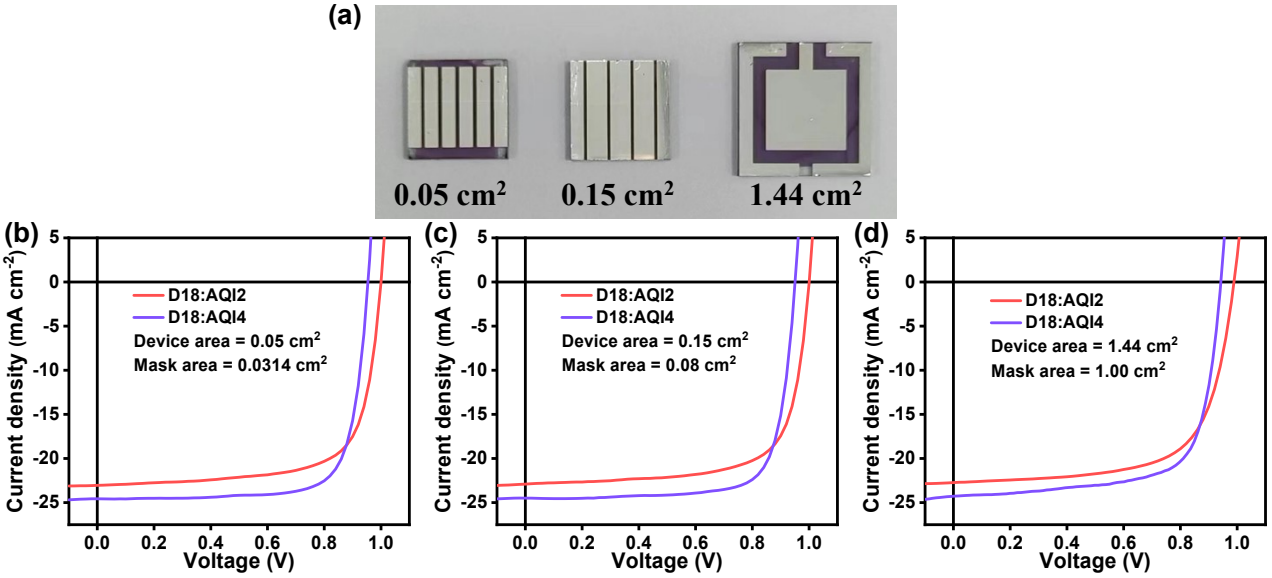


Figure S19. (a) The photos of devices with active areas of 0.05, 0.15 and 1.44 cm^{-2}, respectively. (b)-(c) *J-V* curves of D18:AQI2 and D18:AQI4 with different device areas and mask areas.

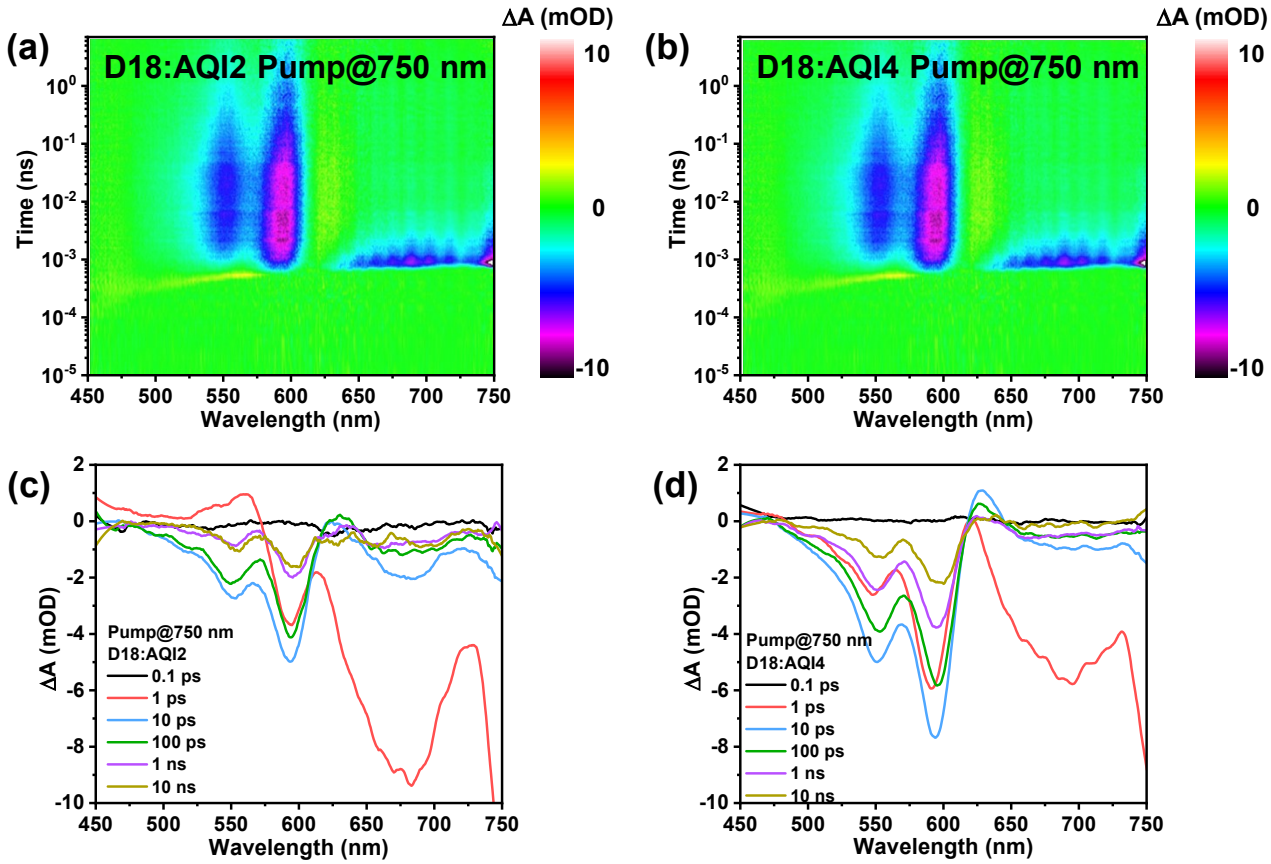


Figure S20. The TA image and the corresponding TA spectra of (a) and (c) D18:AQI2 film with

various decay times under 750 nm excitation. (b) and (d) D18:AQI4 film with various decay times under 750 nm excitation.

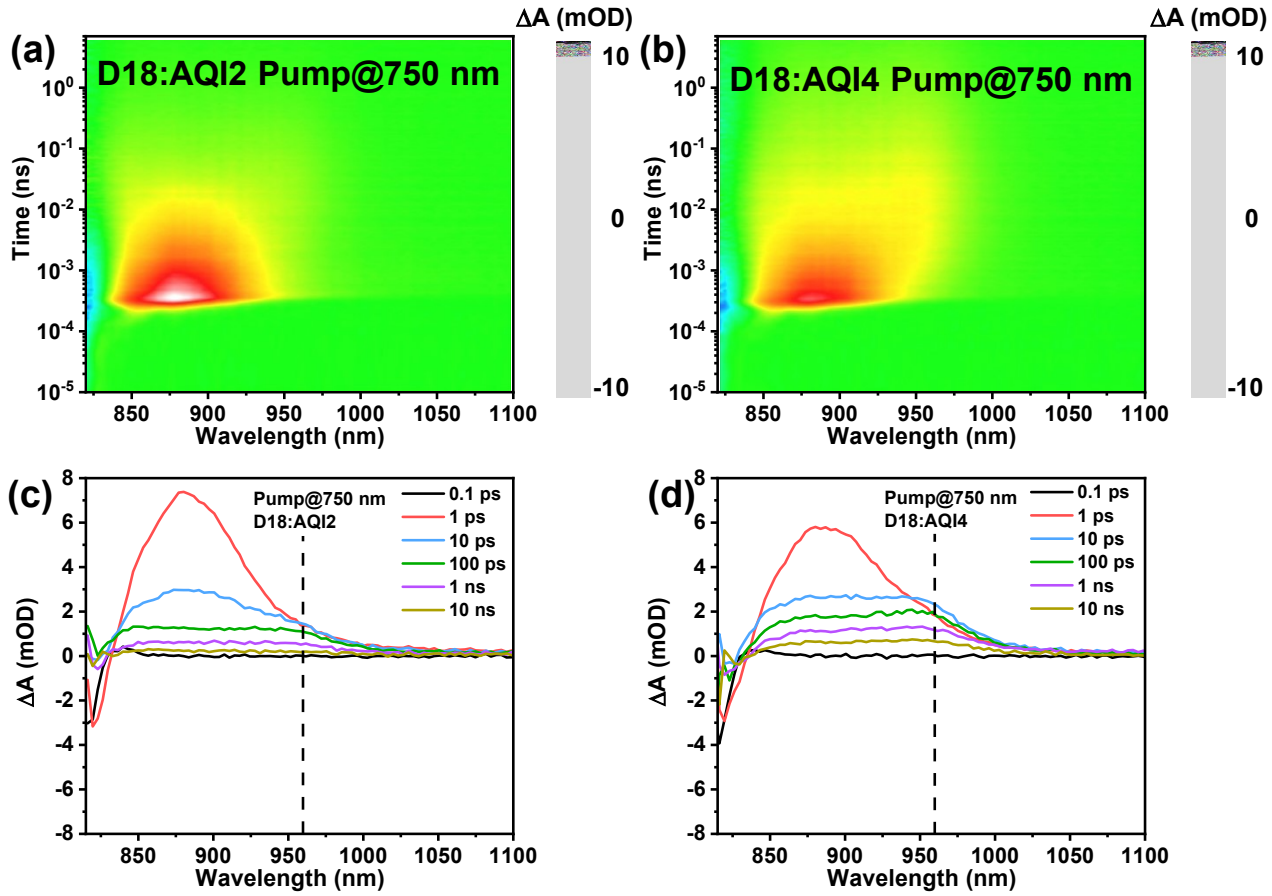


Figure S21. (a) and (c) The TA image and the corresponding TA spectra of D18:AQI2 film with various decay times under 750 nm excitation. (b) and (d) The TA image and the corresponding TA spectra of D18:AQI4 film with various decay times under 750 nm excitation.

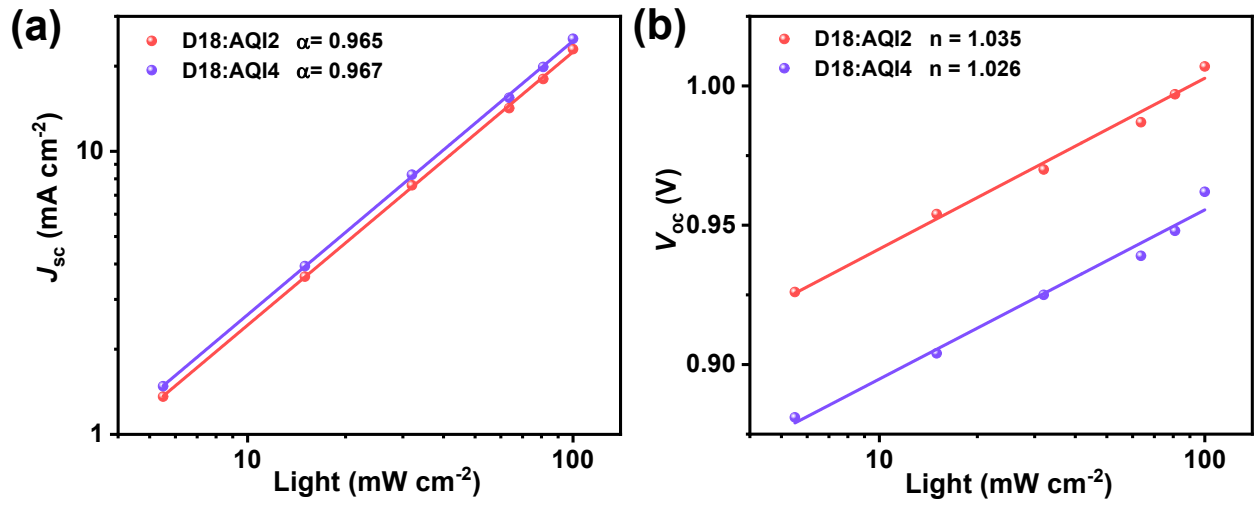


Figure S22. The dependence of the (a) J_{SC} and (b) V_{OC} on the light intensity for the optimum devices.

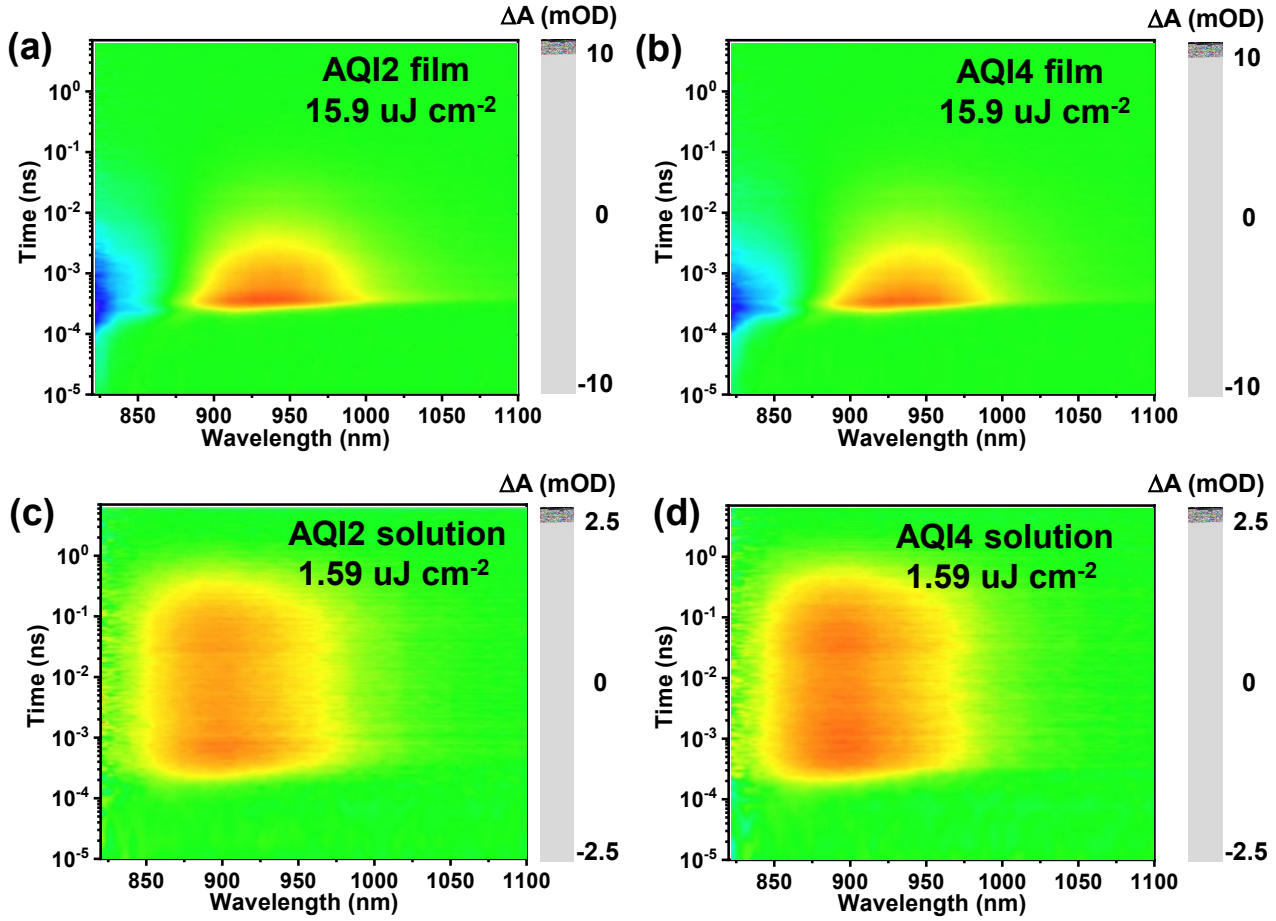


Figure S23. The TA image of (a) AQI2 film, (b) AQI4 film, (c) AQI2 solution, and (d) AQI4 solution under 750 nm excitation at different densities.

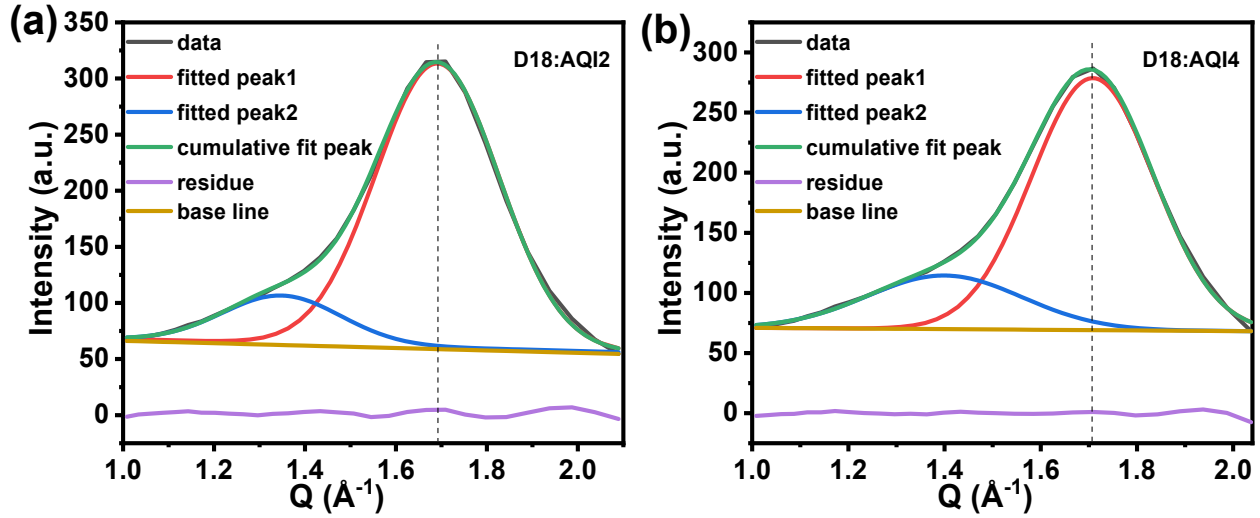


Figure S24. Blends' peak deconvolution of (010) peak in the OOP direction.

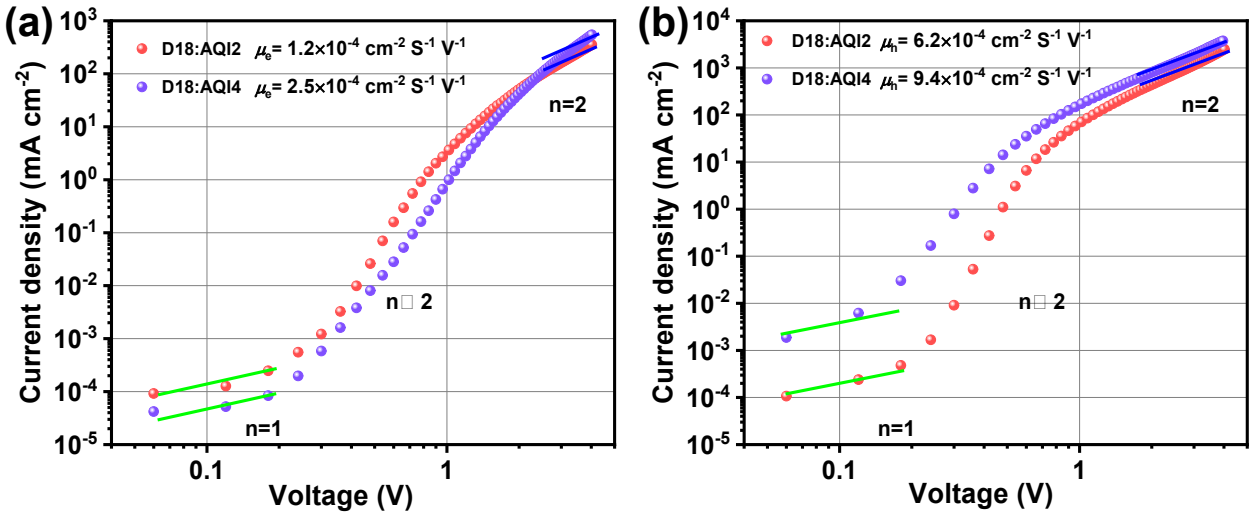


Figure S25. Space-charge limited current (SCLC) of (a) electron-only devices and (b) hole-only devices based on AQI2, and AQI4.

Supplementary Tables

Table S1. Photovoltaic performance of the reported binary OSCs with V_{OC}=0.8-0.9 V, PCE ＞18%; V_{OC}=0.9-1.0 V, PCE ＞16%, and so on until the V_{OC} reaches 1.3V.

Donor:Acceptor	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF	PCE	Ref.(SI)
PBDB-T:PN-Se	0.907	24.82	71.80	16.16	[2]
PBDB-T:PYT	0.910	23.07	77.00	16.05	[3]
PM6:PY-HD	0.937	24.05	72.80	16.41	[3]
PM6:PY-OD	0.943	23.95	73.20	16.53	[3]
PM6:Y6	0.853	26.79	80.20	18.33	[4]
PM6:BTP-eC9	0.852	27.60	78.30	18.40	[5]
PM6:M36	0.90	24.63	72.09	16.00	[6]
PM6:BTP-4F-P2EH	0.880	25.85	80.08	18.22	[7]
PM6:BTP-4F-C5-16	0.844	27.78	77.68	18.20	[8]

PM6:EH-HD-4F	0.840	27.50	79.30	18.38	[9]
PM6:CH6	0.875	26.62	78.40	18.33	[10]
PM6:YCF3	0.861	27.56	76.80	18.21	[11]
PM6:BTP-4F-P2EH	0.880	25.85	80.00	18.22	[12]
PM6:eC9	0.871	26.75	79.00	18.43	[13]
PM6:Y6-BO	0.840	27.90	76.90	18.00	[14]
PM6:AYT9Se11-Cl	0.843	27.40	78.43	18.12	[15]
PM6:L8-BO	0.883	26.94	81.3	19.4	[16]
PM6:BTP-4F-C5-16	0.834	27.65	80.2	18.5	[16]
PM6:L8-BO-F	0.934	23.42	76.9	16.82	[17]
PM6:Se46	0.879	26.22	80.10	18.46	[18]
PM6:BO-4Cl	0.871	27.26	76.4	18.14	[19]
PM6:eC9	0.861	27.88	80.39	19.31	[20]
PM6:CH-BBQ	0.881	26.25	78.9	18.19	[21]
PM6:CH-FC	0.886	26.31	77.66	18.1	[22]
PM6:CH-FB	0.889	26.38	77.86	18.26	[22]
PM6:CH6	0.875	26.62	78.4	18.33	[23]
PM6:CH8-2	0.928	24.24	74.9	16.84	[24]
PM6:CH21	0.873	26.57	78.13	18.12	[25]
PM6:CH22	0.884	26.74	80.62	19.06	[25]
PM6:CH23	0.876	26.64	80.45	18.77	[26]
PM6:TDY-α	0.864	26.9	78.0	18.1	[27]
PM6:BTP-S2	0.945	24.07	72.02	16.37	[28]
PM6:BTP1O-4Cl-C10	0.90	23.42	77.6	16.4	[29]

PM6:BTP1O-4Cl-C12	0.910	23.85	78.80	17.10	[29]
PM6:Y6-O	0.950	22.40	78.00	16.60	[30]
PM6:QX-THF	0.902	24.49	78.99	17.45	[31]
PM6:BTSi-4F	0.900	24.00	77.00	16.60	[32]
PM6:BTP-H2	0.932	25.33	78.50	18.50	[33]
PM6:NQF	0.921	25.79	73.96	17.57	[34]
PM6:Qx-1	0.911	26.10	75.50	17.90	[35]
PM6:Qx-2	0.934	26.50	73.70	18.20	[35]
PM6:MYT	0.917	24.29	74.00	16.44	[36]
PM6:DYT	0.942	24.89	74.00	17.29	[36]
PM6:TYT	0.964	25.07	75.00	18.15	[36]
PM6:BTP-γ-Ar3F	0.904	25.72	74.92	17.42	[37]
PM6:BTP-2O	0.965	22.00	75.60	16.10	[38]
PM6:BTP-O-S	0.912	24.50	77.50	17.30	[38]
PM6:L8-BO-F	0.934	23.42	76.90	16.82	[39]
PM6:BTP-ClBr	0.906	23.48	79.00	16.82	[40]
PM6:BTP-F	0.900	24.05	75.00	16.20	[41]
PM6:Y6-O2BO	0.963	21.50	78.10	16.20	[42]
PM6:QM1	0.910	25.23	74.00	17.05	[43]
PM6:QM2	0.910	25.42	70.50	16.36	[43]
PM6:Qx-Br	0.915	24.27	78.47	17.42	[44]
PM6:Qx-Cl	0.913	24.13	78.32	17.25	[44]
PM6:BTP-ICBCF3	0.925	25.90	74.00	17.80	[45]
PM6:Y6-1OBO	0.930	25.13	77.10	18.02	[46]

PM6:Y6-1O	0.900	25.51	73.50	16.81	[46]
PM6:LC301	0.911	24.21	78.10	17.21	[47]
PM6:PY-V-γ	0.912	24.80	75.80	17.10	[48]
PM6:PY-T-γ	0.929	24.10	71.90	16.10	[48]
PM6:2BTP-2F-T	0.911	25.50	78.28	18.19	[49]
PM6:PY-82	0.950	23.82	75.80	17.15	[46]
PM6:PYT-1S1Se	0.926	24.10	73.00	16.30	[50]
PM6:BTP-O-S	0.912	24.5	77.5	17.3	[51]
PM6:L15	0.93	25.95	77.26	18.72	[52]
PM6:DY-P2EH	0.905	24.03	78.58	17.09	[53]
PM6:PY-C11	0.937	24.92	78.6	18.35	[54]
PM6:PY-DT	0.952	24.00	77.6	17.73	[54]
PM6:PY-IT	0.945	26.37	76.48	19.06	[55]
PM6:O1-Br	1.04	20.00	74.40	15.50	[56]
PM6:BTP-S9	0.849	27.9	79.2	18.8	[57]
PM6:BTP-S11	0.878	27.1	79.3	18.9	[57]
PM6:BTP-S16	0.887	26.60	79.09	18.67	[58]
PM6:Tri-Y6-OD	0.916	25.3	77.8	18.03	[59]
PM6:DIBP3F-Se	0.917	25.92	76.1	18.09	[60]
PM6:Tet-0	0.914	23.95	76.0	16.63	[61]
PM6:Tet-1	0.919	24.53	76.8	17.32	[61]
PM6:Tet-3	0.921	24.21	75.9	16.92	[61]
PM6:OC8-4F	0.89	25.3	80.2	18.0	[62]
PM6:PQx3	0.934	25.21	76.72	18.06	[63]

PM6:A-C10ch	0.887	25.8	78.1	18.4	[64]
PM6:Qx-o-4F	0.928	23.64	75.46	16.55	[65]
PM6:Qx-p-4Cl	0.882	27.05	78.74	18.78	[65]
PM6:G-Trimer-C6C8	0.899	25.32	79.34	18.07	[66]
PM6:G-Trimer-C8C10	0.896	26.75	79.30	19.01	[66]
PM6:PZC2	0.936	24.61	74.93	17.30	[67]
PM6:M3	0.91	24.03	76.22	16.66	[68]
PM6:M36	0.91	23.93	78.40	17.0	[69]
PM6:EV-i	0.897	26.60	76.56	18.27	[70]
PM6:BTP-4F-P2EH	0.88	25.85	80.08	18.22	[71]
PBDB-TFCl:PY-IT	0.944	24.2	77.1	17.6	[72]
PM1:BTP-PhC6-C11	0.871	26.62	79.05	18.33	[73]
PM1:BTP-3Cl	0.844	27.8	76.7	18.0	[74]
PM1:L8-BO	0.893	26.20	79.13	18.51	[75]
PM1:BTP-2F2Cl	0.861	27.35	78.16	18.40	[75]
PM1:A-OSeF	0.880	27.2	77.3	18.5	[76]
D18:Y6	0.859	27.70	76.60	18.22	[77]
D18:N3	0.821	28.14	78.94	18.23	[78]
D18:L8-BO	0.918	26.86	77.25	19.05	[79]
D18:DYA-I	0.938	25.67	0.78	18.83	[80]
D18:DYA-IO	0.948	24.29	0.76	17.54	[80]
D18:DYA-O	0.961	23.32	0.73	16.45	[80]
D18:3-ClTh	0.856	27.80	78.90	18.80	[81]
D18:BS3TSe-4F	0.828	29.40	75.94	18.48	[82]

D18:AsymSSe-2F	0.840	27.44	79.46	18.31	[28]
D18:BTP-eC9-4F	0.868	27.7	79.8	19.3	[83]
D18:AQx-6	0.892	26.8	77.8	18.6	[84]
D18:AQx-8	0.913	24.7	75.7	17.1	[84]
D18:AQx-16	0.936	23.9	77.6	17.4	[85]
D18:AQx-18	0.938	25.0	77.8	18.2	[85]
D18:BTIC-CN	0.926	22.54	76.76	16.03	[86]
D18:FCC-Cl-4Ph	1.10	15.87	75.44	13.12	[73]
D18:DTY6	0.876	26.6	78.6	18.3	[87]
D18:DTC11	0.858	27.5	80.5	19.0	[87]
D18:CH8F	0.899	26.01	80.38	18.80	[88]
D18:Z9	0.894	25.9	80.2	18.6	[89]
D18:BTP-Cy-4F	0.937	24.86	79.51	18.52	[90]
D18:BTP-Th	0.891	26.77	79.71	19.02	[91]
D18-fu:L8-BO	0.886	26.0	79.8	18.4	[92]
HW-D18:L8-BO	0.91	26.48	80.65	19.65	[93]
HW-D18:Y6	0.85	28.10	79.01	19.00	[93]
HW-D18:BTP-eC9-4F	0.87	27.65	79.23	19.17	[93]
D18:Cl-BTA5	1.16	15.17	67.96	12.00	[94]
D18-Cl:BTP-4F-T3EH	0.873	26.84	77.85	18.25	[95]
D18-Cl:N3	0.859	27.85	75.70	18.13	[96]
D18-Cl:L8-BO	0.922	26.60	75.60	18.70	[34]
D18-Cl:PY-IT	0.970	23.6	75.7	17.3	[72]
D18-Cl:BTP-eC9	0.917	25.42	78.29	18.25	[97]

D18-Cl:L8-BO-X	0.893	26.78	79.6	19.04	[98]
PTQ10:*o*-BTP-C6Ph	0.924	22.80	76.20	16.00	[99]
PTQ10:T2EH	0.872	26.57	80.05	18.55	[100]
PTQ10:Cl-BTA33	1.13	14.39	74.52	12.16	[101]
PTQ10:4T-2F-2Cl	0.94	22.93	75.64	16.31	[102]
PB2:F-BTA3	1.22	13.50	0.69	11.40	[103]
PB2:FTCC-Br	1.05	17.90	78.80	14.80	[104]
PB2:FTCC-Br	1.05	17.66	77.91	14.46	[105]
PB3:FTCC-Br	1.06	17.95	79.07	15.01	[105]
PB4:FTCC-Br	1.09	17.90	76.14	14.79	[105]
PBQ6:m-THE	0.880	26.61	79.03	18.51	[104]
PQB-2:PY-IT	0.942	24.2	79.5	18.1	[106]
PBQx-H-TF:dBTICγ-EH	0.910	23.41	75.50	16.06	[107]
PBQx-H-TF:dBTIC-γV-BO	0.91	24.52	76.58	17.14	[108]
PBQx-TF:PY-IT	0.917	23.6	79.6	17.2	[106]
PBQx-TF:eC9-2Cl	0.879	27.20	80.40	19.20	[109]
PBQx-TCl:PYIT1	0.925	23.88	78.05	17.24	[110]
PBQx-TCl:PYIT2	0.938	24.61	79.66	18.39	[110]
PBQx-TCl:PYIT3	0.931	24.18	76.04	17.12	[110]
PBQx-TCl:BTA3	1.24	13.30	74.20	12.20	[111]
PNB-1:L8-BO	0.903	26.21	78.04	18.47	[112]
PNB-3:L8-BO	0.907	26.59	78.86	19.02	[112]
PNB-5:L8-BO	0.914	25.53	76.20	17.78	[112]
PF2:Y6-BO-4Cl	0.86	26.87	78	18.02	[113]

PF3:Y6-BO-4Cl	0.86	26.91	78	18.05	[113]
PF7:Y6-BO-4Cl	0.86	26.95	79	18.31	[113]
PF8:Y6-BO-4Cl	0.86	26.86	78	18.01	[113]
PF9:Y6-BO-4Cl	0.86	26.92	78	18.06	[113]
PBTz-F:L8-BO	0.896	26.71	77.59	18.57	[114]
PBTz-Cl:L8-BO	0.922	26.39	74.35	18.09	[114]
PTzBI-*d*F:BTP-TBr	0.845	27.50	78.80	18.30	[115]
PNTB-2T:N3	0.854	26.88	79.00	18.15	[116]
PFNT-Cl:N3	0.853	26.56	0.799	18.10	[117]
OPz11:Y6	0.865	27.02	78.71	18.42	[11]
PBTTz3Cl:L8-BO	0.888	26.60	77.81	18.38	[118]
PMT-CT-10:Y6	0.832	28.12	77.90	18.21	[34]
PL1:BTP-eC9-4F	0.904	25.91	69.39	16.25	[119]
PTF5:Y6-BO	0.84	28.0	77.2	18.2	[120]
PTTB:L8-BO	0.882	26.56	77.08	18.06	[121]
PQM-Cl:PY-IT	0.92	24.3	80.7	18.0	[122]
PTO2:m-DTC-Cl-2	1.05	18.40	73.10	14.10	[123]
PBDTS-TDZ:ITIC	1.10	17.78	65.4	12.80	[124]
TTC-Cl:HCl-BTA3	1.20	13.42	69.60	11.20	[125]
J52-Cl:BTA3	1.24	13.16	66.62	10.50	[126]
E18:BTA3-4F	1.30	11.22	68.98	10.03	[127]
D18:AQI2	1.00	23.03	71.55	16.48	This work
D18:AQI4	0.95	24.56	76.91	18.02	This work

Table S2. The photoelectric parameters of Y6, AQI2, AQI4.

NFAs	*λsol max* (nm)	*λfilm max* (nm)	*λfilm onest* (nm)	*Eopt g* (eV)	Stoke's shift (nm)	E_b (eV)	HOMO (eV)	LUMO (eV)
Y6	732	831	919	1.35	92	0.44	-5.69	-3.90
AQI2	730	785	847	1.45	45	0.37	-5.60	-3.78
AQI4	735	789	859	1.43	53	0.42	-5.66	-3.81

Table S3. The photovoltaic parameters of devices based on D18:AQI2 with different device optimization condition.

Active layer	D:A	Donor concentration（mg mL^{-1}）	SVA	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
D18:AQI4	1:1.5	4.2	w/o	0.956	23.76	74.47	16.91
	1:1.4	4.2	CF	0.950	24.47	76.40	17.76
	1:1.5	4.2	CF	0.965	24.74	75.80	18.10
	1:1.6	4.2	CF	0.941	24.54	74.41	17.18
	1:1.5	3.7	CF	0.942	24.36	74.48	17.09
	1:1.5	47	CF	0.940	24.29	71.47	16.32

Table S4. The photovoltaic parameters of devices based on D18:AQI4 with different device optimization condition.

Active layer	D:A	Donor concentration（mg mL^{-1}）	SVA	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
D18:AQI2	1:1.2	4.2	w/o	1.003	22.95	68.84	15.85
	1:1	4.2	CF	0.988	22.67	69.71	15.61
	1:1.2	4.2	CF	1.009	23.10	70.39	16.41
	1:1.4	4.2	CF	0.998	22.94	70.03	16.06

	1:1.2	3.7	CF	0.986	22.35	69.89	15.40
	1:1.2	47	CF	0.982	23.57	70.48	16.31

Table S5. The detailed photovoltaic parameters of the optimal devices based on D18:AQI2, and D18:AQI4 with different active area.

Active layer	Active area (cm^2)	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
D18:AQI2	0.05	1.00	23.03	71.55	16.48
	0.15	1.00	22.91	71.61	16.41
	1.44	0.989	22.73	67.25	15.11
D18:AQI4	0.05	0.954	24.56	76.91	18.02
	0.15	0.951	24.49	76.93	17.92
	1.44	0.943	24.27	71.17	16.29

Table S6. Summary of diffusion length (L_D) values for three acceptors.

Acceptors	K ($\times 10^{10}$ s^{-1})	γ ($\times 10^{-8}$ cm^3 S^{-1})	τ (ps)	D ($\times 10^{-3}$ cm^2 s^{-1})	L_D (nm)
AQI2	1.7	0.86	743	6.8	22.48
AQI4	1.3	1.38	994	11.0	33.17

Table S7. Detailed GIWAXS data of AQI2 and AQI4 pure films and D18:AQI2 and D18:AQI4 blend films.

Compound	OOP				IP			
	(010)				(100)			
	q ($Å^{-1}$)	d-spacing (Å)	FWHM ($Å^{-1}$)	CCL (Å)	q ($Å^{-1}$)	d-spacing (Å)	FWHM ($Å^{-1}$)	CCL (Å)

AQI2	1.71	3.67	0.40	14.61	0.34	18.48	0.23	25.41
AQI4	1.74	3.61	0.39	14.98	0.36	17.45	0.16	36.52
D18:AQI2	1.70	3.70	0.29	20.15	0.30	20.94	0.10	58.43
D18:AQI4	1.71	3.67	0.35	16.70	0.31	20.27	0.12	48.69

References

1. P. Zhou, L. Deng, Z. Han, X. Zhao, Z. Zhang and S. Huo, *RSC Adv.*, 2022, **12**, 13480-13486.
2. J. Du, K. Hu, J. Zhang, L. Meng, J. Yue, I. Angunawela, H. Yan, S. Qin, X. Kong, Z. Zhang, B. Guan, H. Ade and Y. Li, *Nat. Commun.*, 2021, **12**, 5264.
3. L. Zhan, S. Li, Y. Li, R. Sun, J. Min, Z. Bi, W. Ma, Z. Chen, G. Zhou, H. Zhu, M. Shi, L. Zuo and H. Chen, *Joule*, 2022, **6**, 662-675.
4. D. Li, F. Geng, T. Hao, Z. Chen, H. Wu, Z. Ma, Q. Xue, L. Lin, R. Huang, S. Leng, B. Hu, X. Liu, J. Wang, H. Zhu, M. Lv, L. Ding, M. Fahlman, Q. Bao and Y. Li, *Nano Energy*, 2022, **96**, 107133.
5. R. Sun, T. Wang, Y. Wu, M. Zhang, Y. Ma, Z. Xiao, G. Lu, L. Ding, Q. Zheng, C. J. Brabec, Y. Li and J. Min, *Adv. Funct. Mater.*, 2021, **31**, 2106846.
6. Y. Ma, D. Cai, S. Wan, P. Yin, P. Wang, W. Lin and Q. Zheng, *Natl. Sci. Rev.*, 2020, **7**, 1886-1895.
7. W. Song, K. Yu, E. Zhou, L. Xie, L. Hong, J. Ge, J. Zhang, X. Zhang, R. Peng and Z. Ge, *Adv. Funct. Mater.*, 2021, **31**, 2102694.
8. C. Yang, Q. An, H.-R. Bai, H.-F. Zhi, H. S. Ryu, A. Mahmood, X. Zhao, S. Zhang, H. Y. Woo and J.-L. Wang, *Angew. Chem. Int. Ed.*, 2021, **60**, 19241-19252.
9. S. Chen, L. Feng, T. Jia, J. Jing, Z. Hu, K. Zhang and F. Huang, *Sci. China Chem.*, 2021, **64**, 1192-1199.
10. H. Chen, H. Liang, Z. Guo, Y. Zhu, Z. Zhang, Z. Li, X. Cao, H. Wang, W. Feng, Y. Zou, L. Meng, X. Xu, B. Kan, C. Li, Z. Yao, X. Wan, Z. Ma and Y. Chen, *Angew. Chem. Int. Ed.*, 2022, **61**, e202209580.
11. Z. Liao, D. Hu, H. Tang, P. Huang, R. Singh, S. Chung, K. Cho, M. Kumar, L. Hou, Q. Chen, W. Yu, H. Chen, K. Yang, Z. Kan, F. Liu, Z. Xiao, G. Li and S. Lu, *J. Mater. Chem. A*, 2022, **10**, 7878-7887.
12. H. Ning, G. Zhang, H. Chen, Z. Wang, S. Ni, F. Lu, F. Liu, L. Dang, J. Liu, F. He and Q. Wu, *Chem. Mater.*, 2021, **33**, 1976-1982.
13. Z. Zheng, J. Wang, P. Bi, J. Ren, Y. Wang, Y. Yang, X. Liu, S. Zhang and J. Hou, *Joule*, 2022, **6**, 171-184.
14. C. Guo, D. Li, L. Wang, B. Du, Z.-X. Liu, Z. Shen, P. Wang, X. Zhang, J. Cai, S. Cheng, C. Yu, H. Wang, D. Liu, C.-Z. Li and T. Wang, *Adv. Energy Mater.*, 2021, **11**, 2102000.
15. X. Zhao, Q. An, H. Zhang, C. Yang, A. Mahmood, M. Jiang, M. H. Jee, B. Fu, S. Tian, H. Y. Woo, Y. Wang and J.-L. Wang, *Angew. Chem. Int. Ed.*, 2023, **62**, e202216340.
16. Z. Gan, L. Wang, J. Cai, C. Guo, C. Chen, D. Li, Y. Fu, B. Zhou, Y. Sun, C. Liu, J. Zhou, D. Liu, W. Li and T. Wang, *Nat. Commun.*, 2023, **14**, 6297.
17. Y. Cai, Y. Li, R. Wang, H. Wu, Z. Chen, J. Zhang, Z. Ma, X. Hao, Y. Zhao, C. Zhang, F. Huang and Y. Sun, *Adv. Mater.*, 2021, **33**, 2101733.
18. Z. Li, X. Li, J. Xue, J. Zhang, C. Zhu, J. Li, W. Ma, L. Meng and Y. Li, *ACS Energy Lett.*, 2023, **8**, 2488-2495.
19. W. Zhang, Y. Wu, R. Ma, H. Fan, X. Li, H. Yang, C. Cui and Y. Li, *Angew. Chem. Int. Ed.*, 2023, **62**, e202309713.
20. J. Fu, P. W. K. Fong, H. Liu, C.-S. Huang, X. Lu, S. Lu, M. Abdelsamie, T. Kodalle, C. M. Sutter-Fella, Y. Yang and G. Li, *Nat. Commun.*, 2023, **14**, 1760.

21. T. Duan, W. Feng, Y. Li, Z. Li, Z. Zhang, H. Liang, H. Chen, C. Zhong, S. Jeong, C. Yang, S. Chen, S. Lu, O. A. Rakitin, C. Li, X. Wan, B. Kan and Y. Chen, *Angew. Chem. Int. Ed.*, 2023, **62**, e202308832.
22. H. Liang, H. Chen, Y. Zou, Y. Zhang, Y. Guo, X. Cao, X. Bi, Z. Yao, X. Wan and Y. Chen, *Chem. Commun.*, 2023, **59**, 13367-13370.
23. H. Chen, H. Liang, Z. Guo, Y. Zhu, Z. Zhang, Z. Li, X. Cao, H. Wang, W. Feng, Y. Zou, L. Meng, X. Xu, B. Kan, C. Li, Z. Yao, X. Wan, Z. Ma and Y. Chen, *Angew. Chem. Int. Ed.*, 2022, **61**, e202209580.
24. Y. Xu, J. Wang, T. Zhang, Z. Chen, K. Xian, Z. Li, Y.-H. Luo, L. Ye, X. Hao, H. Yao and J. Hou, *Energy Environ. Sci.*, 2023, **16**, 5863-5870.
25. H. Liang, X. Bi, H. Chen, T. He, Y. Lin, Y. Zhang, K. Ma, W. Feng, Z. Ma and G. Long, *Nat. Commun.*, 2023, **14**, 4707.
26. H. Liang, H. Chen, P. Wang, Y. Zhu, Y. Zhang, W. Feng, K. Ma, Y. Lin, Z. Ma, G. Long, C. Li, B. Kan, Z. Yao, H. Zhang, X. Wan and Y. Chen, *Adv. Funct. Mater.*, 2023, **33**, 2301573.
27. Y. Bai, Z. Zhang, Q. Zhou, H. Geng, Q. Chen, S. Kim, R. Zhang, C. Zhang, B. Chang, S. Li, H. Fu, L. Xue, H. Wang, W. Li, W. Chen, M. Gao, L. Ye, Y. Zhou, Y. Ouyang, C. Zhang, F. Gao, C. Yang, Y. Li and Z.-G. Zhang, *Nat. Commun.*, 2023, **14**, 2926.
28. S. Li, L. Zhan, Y. Jin, G. Zhou, T.-K. Lau, R. Qin, M. Shi, C.-Z. Li, H. Zhu, X. Lu, F. Zhang and H. Chen, *Adv. Mater.*, 2020, **32**, 2001160.
29. Y. Chen, R. Ma, T. Liu, Y. Xiao, H. K. Kim, J. Zhang, C. Ma, H. Sun, F. Bai, X. Guo, K. S. Wong, X. Lu and H. Yan, *Adv. Energy Mater.*, 2021, **11**, 2003777.
30. J. Fu, H. Chen, P. Huang, Q. Yu, H. Tang, S. Chen, S. Jung, K. Sun, C. Yang, S. Lu, Z. Kan, Z. Xiao and G. Li, *Nano Energy*, 2021, **84**, 105862.
31. Z. Chen, Y. Huang, J. Gao, L. Zhang, Z. Ma, M. Liu, T. Emrick and Y. Liu, *ACS Energy Lett.*, 2022, **7**, 4052-4060.
32. S. Gong, Z. Yin, X. Zhang, X. He, W. Zhang, S. Yang and W. Song, *New J. Chem.*, 2022, **46**, 10611-10617.
33. C. He, Y. Pan, Y. Ouyang, Q. Shen, Y. Gao, K. Yan, J. Fang, Y. Chen, C.-Q. Ma, J. Min, C. Zhang, L. Zuo and H. Chen, *Energy Environ. Sci.*, 2022, **15**, 2537-2544.
34. G. Cai, Z. Chen, X. Xia, Y. Li, J. Wang, H. Liu, P. Sun, C. Li, R. Ma, Y. Zhou, W. Chi, J. Zhang, H. Zhu, J. Xu, H. Yan, X. Zhan and X. Lu, *Adv. Sci.*, 2022, **9**, 2200578.
35. Y. Shi, Y. Chang, K. Lu, Z. Chen, J. Zhang, Y. Yan, D. Qiu, Y. Liu, M. A. Adil, W. Ma, X. Hao, L. Zhu and Z. Wei, *Nat. Commun.*, 2022, **13**, 3256.
36. J.-W. Lee, C. Sun, T. N.-L. Phan, D. C. Lee, Z. Tan, H. Jeon, S. Cho, S.-K. Kwon, Y.-H. Kim and B. J. Kim, *Energy Environ. Sci.*, 2023, **16**, 3339-3349.
37. Z. Li, Z. a. Xu, S. Chen, J. Yao, H. Fu, M. Zhang, Y. Bai, H. Wang, Z. Liu and Z.-G. Zhang, *J. Mater. Chem. A*, 2023, **11**, 4539-4546.
38. J. Hai, Y. Song, L. Li, X. Liu, X. Shi, Z. Huang, G. Qian, Z. Lu, J. Yu, H. Hu and S. Chen, *Adv. Funct. Mater.*, 2023, **33**, 2213429.
39. Y. Cai, Y. Li, R. Wang, H. Wu, Z. Chen, J. Zhang, Z. Ma, X. Hao, Y. Zhao, C. Zhang, F. Huang and Y. Sun, *Adv. Mater.*, 2021, **33**, 2101733.
40. Z. Luo, R. Ma, Z. Chen, Y. Xiao, G. Zhang, T. Liu, R. Sun, Q. Zhan, Y. Zou, C. Zhong, Y. Chen, H. Sun, G. Chai, K. Chen, X. Guo, J. Min, X. Lu, C. Yang and H. Yan, *Adv. Energy Mater.*, 2020, **10**, 2002649.
41. Y. Qin, Y. Chang, X. Zhu, X. Gu, L. Guo, Y. Zhang, Q. Wang, J. Zhang, X. Zhang, X. Liu, K. Lu, E. Zhou, Z. Wei and X. Sun, *Nano Today*, 2021, **41**, 101289.
42. W. Huang, S. Zhao and J. Luo, *Chem. Mater.*, 2022, **34**, 5-28.
43. W. Liu, J. Yuan, C. Zhu, Q. Wei, S. Liang, H. Zhang, G. Zheng, Y. Hu, L. Meng, F. Gao, Y. Li and Y. Zou, *Sci. China Chem.*, 2022, **65**, 1374-1382.

44. D. Qiu, J. Zhang, K. Lu and Z. Wei, *Nano Res.*, 2023, **16**, 11630-11637.
45. Y. Shi, L. Zhu, Y. Yan, M. Xie, G. Liang, J. Qiao, J. Zhang, X. Hao, K. Lu and Z. Wei, *Adv. Energy Mater.*, 2023, **13**, 2300458.
46. J.-W. Lee, C. Sun, S.-W. Lee, G.-U. Kim, S. Li, C. Wang, T.-S. Kim, Y.-H. Kim and B. J. Kim, *Energy Environ. Sci.*, 2022, **15**, 4672-4685.
47. C. Li, G. Lu, H. S. Ryu, X. Sun, H. Y. Woo and Y. Sun, *ACS Appl. Mater. Interfaces*, 2022, **14**, 43207-43214.
48. R. Yang, J. Tian, W. Liu, Y. Wang, Z. Chen, T. P. Russell and Y. Liu, *Chem. Mater.*, 2022, **34**, 7293-7301.
49. X. Liu, Z. Zheng, J. Wang, Y. Wang, B. Xu, S. Zhang and J. Hou, *Adv. Mater.*, 2022, **34**, 2106453.
50. H. Fu, Q. Fan, W. Gao, J. Oh, Y. Li, F. Lin, F. Qi, C. Yang, T. J. Marks and A. K. Y. Jen, *Sci. China Chem.*, 2021, **65**, 309-317.
51. J. Hai, Y. Song, L. Li, X. Liu, X. Shi, Z. Huang, G. Qian, Z. Lu, J. Yu, H. Hu and S. Chen, *Adv. Funct. Mater.*, 2023, **33**, 2213429.
52. B. Liu, W. Xu, R. Ma, J. W. Lee, T. A. Dela Peña, W. Yang, B. Li, M. Li, J. Wu, Y. Wang, C. Zhang, J. Yang, J. Wang, S. Ning, Z. Wang, J. Li, H. Wang, G. Li, B. J. Kim, L. Niu, X. Guo and H. Sun, *Adv. Mater.*, 2023, **35**, 2308334.
53. M. Zhang, B. Chang, R. Zhang, S. Li, X. Liu, L. Zeng, Q. Chen, L. Wang, L. Yang, H. Wang, J. Liu, F. Gao and Z. G. Zhang, *Adv. Mater.*, 2023, **36**, 2308606.
54. J. Song, L. Ye, C. Liu, Y. Cai, C. Zhang, G. Yue, Y. Li, M. H. Jee, Y. Zhao, D. Wei, H. Y. Woo and Y. Sun, *Energy Environ. Sci.*, 2023, **16**, 5371-5380.
55. R. Zeng, L. Zhu, M. Zhang, W. Zhong, G. Zhou, J. Zhuang, T. Hao, Z. Zhou, L. Zhou, N. Hartmann, X. Xue, H. Jing, F. Han, Y. Bai, H. Wu, Z. Tang, Y. Zou, H. Zhu, C.-C. Chen, Y. Zhang and F. Liu, *Nat. Commun.*, 2023, **14**, 4148.
56. Z. Jia, Q. Ma, Z. Chen, L. Meng, N. Jain, I. Angunawela, S. Qin, X. Kong, X. Li, Y. M. Yang, H. Zhu, H. Ade, F. Gao and Y. Li, *Nat. Commun.*, 2023, **14**, 1236.
57. S. Li, C. He, T. Chen, J. Zheng, R. Sun, J. Fang, Y. Chen, Y. Pan, K. Yan, C.-Z. Li, M. Shi, L. Zuo, C.-Q. Ma, J. Min, Y. Liu and H. Chen, *Energy Environ. Sci.*, 2023, **16**, 2262-2273.
58. T. Chen, S. Li, Y. Li, Z. Chen, H. Wu, Y. Lin, Y. Gao, M. Wang, G. Ding, J. Min, Z. Ma, H. Zhu, L. Zuo and H. Chen, *Adv. Mater.*, 2023, **35**, 2300400.
59. C. Zhang, J. Song, J. Xue, S. Wang, Z. Ge, Y. Man, W. Ma and Y. Sun, *Angew. Chem. Int. Ed.*, 2023, **62**, e202308595.
60. J. Wu, Z. Ling, L. R. Franco, S. Y. Jeong, Z. Genene, J. Mena, S. Chen, C. Chen, C. M. Araujo, C. F. N. Marchiori, J. Kimpel, X. Chang, F. H. Isikgor, Q. Chen, H. Faber, Y. Han, F. Laquai, M. Zhang, H. Y. Woo, D. Yu, T. D. Anthopoulos and E. Wang, *Angew. Chem. Int. Ed.*, 2023, **62**, e202302888.
61. C. Zhang, J. Song, L. Ye, X. Li, M. H. Jee, H. Y. Woo and Y. Sun, *Angew. Chem. Int. Ed.*, 2023, **63**, e202316295.
62. Y. Xu, J. Wang, H. Yao, P. Bi, T. Zhang, J. Xu and J. Hou, *Chin. J. Chem.*, 2023, **41**, 1045-1050.
63. D. Qiu, H. Zhang, C. Tian, J. Zhang, L. Zhu, Z. Wei and K. Lu, *Adv. Mater.*, 2023, **35**, 2307398.
64. C. Xiao, X. Wang, T. Zhong, R. Zhou, X. Zheng, Y. Liu, T. Hu, Y. Luo, F. Sun, B. Xiao, Z. Liu, C. Yang and R. Yang, *Adv. Sci.*, 2023, **10**, 2206580.
65. M. Xie, Y. Shi, L. Zhu, J. Zhang, Q. Cheng, H. Zhang, Y. Yan, M. Zhu, H. Zhou, K. Lu and Z. Wei, *Energy Environ. Sci.*, 2023, **16**, 3543-3551.
66. C. Wang, X. Ma, Y.-f. Shen, D. Deng, H. Zhang, T. Wang, J. Zhang, J. Li, R. Wang, L. Zhang, Q. Cheng, Z. Zhang, H. Zhou, C. Tian and Z. Wei, *Joule*, 2023, **7**, 2386-2401.
67. Z. Zhang, Z. Li, B. Kan, T. Chen, Y. Zhang, P. Wang, Z. Yao, C. Li, B. Zhao, M. Li, T. Duan, X. Wan and Y. Chen, *Nano Energy*, 2023, **116**, 108766.

68. Y. Ma, M. Zhang, S. Wan, P. Yin, P. Wang, D. Cai, F. Liu and Q. Zheng, *Joule*, 2021, **5**, 197-209.
69. Y. Ma, R. Sun, Z. Chen, S. Zhang, D. Cai, S. Wan, W. Lin, S.-Q. Zhang, Q. Tu, W. Ma, J. Min, X. Hao and Q. Zheng, *Nano Energy*, 2023, **107**.
70. H. Zhuo, X. Li, J. Zhang, S. Qin, J. Guo, R. Zhou, X. Jiang, X. Wu, Z. Chen, J. Li, L. Meng and Y. Li, *Angew. Chem. Int. Ed.*, 2023, **62**, e202303551.
71. J. Zhang, F. Bai, I. Angunawela, X. Xu, S. Luo, C. Li, G. Chai, H. Yu, Y. Chen, H. Hu, Z. Ma, H. Ade and H. Yan, *Adv. Energy Mater.*, 2021, **11**, 2102596.
72. J. Wang, Y. Wang, K. Xian, J. Qiao, Z. Chen, P. Bi, T. Zhang, Z. Zheng, X. Hao, L. Ye, S. Zhang and J. Hou, *Adv. Mater.*, 2023, **36**, 2305424.
73. Z. Luo, Y. Gao, H. Lai, Y. Li, Z. Wu, Z. Chen, R. Sun, J. Ren, C. e. Zhang, F. He, H. Woo, J. Min and C. Yang, *Energy Environ. Sci.*, 2022, **15**, 4601-4611.
74. J. Wan, Y. Wu, R. Sun, J. Qiao, X. Hao and J. Min, *Energy Environ. Sci.*, 2022, **15**, 5192-5201.
75. R. Sun, Y. Wu, X. Yang, Y. Gao, Z. Chen, K. Li, J. Qiao, T. Wang, J. Guo, C. Liu, X. Hao, H. Zhu and J. Min, *Adv. Mater.*, 2022, **34**, 2110147.
76. C. Yang, Q. An, M. Jiang, X. Ma, A. Mahmood, H. Zhang, X. Zhao, H. F. Zhi, M. H. Jee, H. Y. Woo, X. Liao, D. Deng, Z. Wei and J. L. Wang, *Angew. Chem. Int. Ed.*, 2023, **62**, e202313016.
77. Z. Luo, T. Liu, R. Ma, Y. Xiao, L. Zhan, G. Zhang, H. Sun, F. Ni, G. Chai, J. Wang, C. Zhong, Y. Zou, X. Guo, X. Lu, H. Chen, H. Yan and C. Yang, *Adv. Mater.*, 2020, **32**, 2005942.
78. W. Song, Q. Ye, S. Yang, L. Xie, Y. Meng, Z. Chen, Q. Gu, D. Yang, J. Shi and Z. Ge, *Angew. Chem. Int. Ed.*, 2023, **62**, e202310034.
79. Y. Wei, Z. Chen, G. Lu, N. Yu, C. Li, J. Gao, X. Gu, X. Hao, G. Lu, Z. Tang, J. Zhang, Z. Wei, X. Zhang and H. Huang, *Adv. Mater.*, 2022, **34**, 2204718.
80. C. Sun, J. W. Lee, Z. Tan, T. N. L. Phan, D. Han, H. G. Lee, S. Lee, S. K. Kwon, B. J. Kim and Y. H. Kim, *Adv. Energy Mater.*, 2023, **13**, 2301283.
81. B. Fan, W. Gao, R. Zhang, W. Kaminsky, F. R. Lin, X. Xia, Q. Fan, Y. Li, Y. An, Y. Wu, M. Liu, X. Lu, W. J. Li, H.-L. Yip, F. Gao and A. K. Y. Jen, *J. Am. Chem. Soc.*, 2023, **145**, 5909-5919.
82. W. Gao, F. Qi, Z. Peng, F. R. Lin, K. Jiang, C. Zhong, W. Kaminsky, Z. Guan, C.-S. Lee, T. J. Marks, H. Ade and A. K.-Y. Jen, *Adv. Mater.*, 2022, **34**, 2202089.
83. H. Zhang, G. Ran, X. Cui, Y. Liu, Z. Yin, D. Li, X. Ma, W. Liu, H. Lu, R. Liu, L. Cai, W. Zhang, S. Guo, H. Li, J. Yu, Y. Lin, Y. Liu, G. Lu, Z. Ma, P. Cheng and Z. Bo, *Adv. Energy Mater.*, 2023, **13**, 2302063.
84. Y. Jiang, Y. Li, F. Liu, W. Wang, W. Su, W. Liu, S. Liu, W. Zhang, J. Hou, S. Xu, Y. Yi and X. Zhu, *Nat. Commun.*, 2023, **14**, 5079.
85. K. Liu, Y. Jiang, F. Liu, G. Ran, F. Huang, W. Wang, W. Zhang, C. Zhang, J. Hou and X. Zhu, *Adv. Mater.*, 2023, **35**, 2300363.
86. C.-Z. Sun, X. Lai, T. Rehman, H. Lai, C. Ke, X. Shen, Y. Zhu, L. Tian and F. He, *J. Mater. Chem. C*, 2022, **10**, 17174-17181.
87. Z. Zhong, S. Chen, J. Zhao, J. Xie, K. Zhang, T. Jia, C. Zhu, J. Jing, Y. Liang, L. Hong, S. Zhu, D. Ma and F. Huang, *Adv. Energy Mater.*, 2023, **13**, 2302273.
88. Z. Yao, X. Cao, X. Bi, T. He, Y. Li, X. Jia, H. Liang, Y. Guo, G. Long, B. Kan, C. Li, X. Wan and Y. Chen, *Angew. Chem. Int. Ed.*, 2023, **62**, e202312630.
89. F. Liu, Y. Jiang, R. Xu, W. Su, S. Wang, Y. Zhang, K. Liu, S. Xu, W. Zhang, Y. Yi, W. Ma and X. Zhu, *Angew. Chem. Int. Ed.*, 2023, **63**, e202313791.
90. M. Deng, X. Xu, Y. Duan, L. Yu, R. Li and Q. Peng, *Adv. Mater.*, 2023, **35**, 2210760.
91. H. Meng, W. Jing, X. Xu, L. Yu and Q. Peng, *Angew. Chem. Int. Ed.*, 2023, **62**, e202301958.

92. L. Chen, J. Yi, R. Ma, L. Ding, T. A. Dela Peña, H. Liu, J. Chen, C. Zhang, C. Zhao, W. Lu, Q. Wei, B. Zhao, H. Hu, J. Wu, Z. Ma, X. Lu, M. Li, G. Zhang, G. Li and H. Yan, *Adv. Mater.*, 2023, **35**, 2301231.
93. H. Lu, W. Liu, G. Ran, Z. Liang, H. Li, N. Wei, H. Wu, Z. Ma, Y. Liu, W. Zhang, X. Xu and Z. Bo, *Angew. Chem. Int. Ed.*, 2023, **62**, e202314420.
94. D. Li, N. Deng, Y. Fu, C. Guo, B. Zhou, L. Wang, J. Zhou, D. Liu, W. Li, K. Wang, Y. Sun and T. Wang, *Adv. Mater.*, 2023, **35**, 2208211.
95. A. Shang, S. Luo, J. Zhang, H. Zhao, X. Xia, M. Pan, C. Li, Y. Chen, J. Yi, X. Lu, W. Ma, H. Yan and H. Hu, *Sci. China Chem.*, 2022, **65**, 1758-1766.
96. J. Qin, L. Zhang, C. Zuo, Z. Xiao, Y. Yuan, S. Yang, F. Hao, M. Cheng, K. Sun, Q. Bao, Z. Bin, Z. Jin and L. Ding, *J. Semicond.*, 2021, **42**, 010501.
97. X. Xu, W. Jing, H. Meng, Y. Guo, L. Yu, R. Li and Q. Peng, *Adv. Mater.*, 2023, **35**, 2208997.
98. S. Luo, C. Li, J. Zhang, X. Zou, H. Zhao, K. Ding, H. Huang, J. Song, J. Yi, H. Yu, K. S. Wong, G. Zhang, H. Ade, W. Ma, H. Hu, Y. Sun and H. Yan, *Nat. Commun.*, 2023, **14**, 6964.
99. G. Chai, Y. Chang, J. Zhang, X. Xu, L. Yu, X. Zou, X. Li, Y. Chen, S. Luo, B. Liu, F. Bai, Z. Luo, H. Yu, J. Liang, T. Liu, K. S. Wong, H. Zhou, Q. Peng and H. Yan, *Energy Environ. Sci.*, 2021, **14**, 3469-3479.
100. X. Kong, J. Zhang, L. Meng, C. Sun, S. Qin, C. Zhu, J. Zhang, J. Li, Z. Wei and Y. Li, *CCS Chem.*, 2023, **5**, 841-850.
101. L. Qin, X. Li, C. Dong, J. Zhou, Q. Guo, A. Tang, Y. Zhong and E. Zhou, *Chem. Eng. J.*, 2023, **464**, 142743.
102. Y. Shao, R. Sun, W. Wang, X. Yang, C. Sun, Y. Li and J. Min, *Sci. China Chem.*, 2023, **66**, 1101-1110.
103. T. Zhang, C. An, Y. Cui, J. Zhang, P. Bi, C. Yang, S. Zhang and J. Hou, *Adv. Mater.*, 2021, **34**, 2105803.
104. X. Kong, C. Zhu, J. Zhang, L. Meng, S. Qin, J. Zhang, J. Li, Z. Wei and Y. Li, *Energy Environ. Sci.*, 2022, **15**, 2011-2020.
105. P. Bi, C. An, T. Zhang, Z. Chen, Y. Xu, Y. Cui, J. Wang, J. Li, Y. Wang, J. Ren, X. Hao, S. Zhang and J. Hou, *J. Mater. Chem. A*, 2023, **11**, 983-991.
106. T. Zhang, Y. Xu, H. Yao, J. Zhang, P. Bi, Z. Chen, J. Wang, Y. Cui, L. Ma, K. Xian, Z. Li, X. Hao, Z. Wei and J. Hou, *Energy Environ. Sci.*, 2023, **16**, 1581-1589.
107. H. Wang, C. Cao, H. Chen, H. Lai, C. Ke, Y. Zhu, H. Li and F. He, *Angew. Chem. Int. Ed.*, 2022, **61**, e202201844.
108. P. Tan, H. Chen, H. Wang, X. Lai, Y. Zhu, X. Shen, M. Pu, H. Lai, S. Zhang, W. Ma and F. He, *Adv. Funct. Mater.*, 2023, **34**, 2305608.
109. J. Wang, Y. Wang, P. Bi, Z. Chen, J. Qiao, J. Li, W. Wang, Z. Zheng, S. Zhang, X. Hao and J. Hou, *Adv. Mater.*, 2023, 2301583.
110. P. Bi, T. Zhang, Y. Cui, J. Wang, J. Qiao, K. Xian, X. W. Chua, Z. Chen, W. P. Goh, L. Ye, X. Hao, J. Hou and L. Yang, *Adv. Energy Mater.*, 2023, **13**, 2302252.
111. Y. Xu, Y. Cui, H. Yao, T. Zhang, J. Zhang, L. Ma, J. Wang, Z. Wei and J. Hou, *Adv. Mater.*, 2021, **33**, 2101090.
112. B. Pang, C. Liao, X. Xu, S. Peng, J. Xia, Y. Guo, Y. Xie, Y. Chen, C. Duan, H. Wu, R. Li and Q. Peng, *Adv. Mater.*, 2023, **35**, 2211871.
113. H. Bin, J. Li, A. Caiazzo, M. M. Wienk, Y. Li and R. A. J. Janssen, *ChemSusChem*, 2023, **16**, e202300006.
114. B. Pang, C. Liao, X. Xu, L. Yu, R. Li and Q. Peng, *Adv. Mater.*, 2023, **35**, 2300631.
115. B. Fan, W. Gao, X. Wu, X. Xia, Y. Wu, F. R. Lin, Q. Fan, X. Lu, W. J. Li, W. Ma and A. K. Jen, *Nat. Commun.*, 2022, **13**, 5946.
116. Q. Jiang, P. Han, H. Ning, J. Jiang, H. Chen, Y. Xiao, C.-R. Ye, J. Chen, M. Lin, F. He, X.-C. Huang and Q. Wu, *Nano Energy*, 2022, **101**, 107611.
117. J. Chen, D. Li, M. Su, Y. Xiao, H. Chen, M. Lin, X. Qiao, L. Dang, X. C. Huang, F. He and Q. Wu, *Angew. Chem. Int. Ed.*, 2023, **62**, e202215930.

118. Y. Shao, Y. Gao, R. Sun, M. Zhang and J. Min, *Adv. Mater.*, 2023, **35**, 2208750.
119. H. Lu, H. Wang, G. Ran, S. Li, J. Zhang, Y. Liu, W. Zhang, X. Xu and Z. Bo, *Adv. Funct. Mater.*, 2022, **32**, 2203193.
120. Z. U. Rehman, M. Haris, S. U. Ryu, M. Jahankhan, C. E. Song, H. K. Lee, S. K. Lee, W. S. Shin, T. Park and J. C. Lee, *Adv. Sci.*, 2023, **10**, 2302376.
121. L.-Y. Xu, Y. Gao, W. Wang, Y. Shao, M. Chen, X. Yang, Y. Fu, M. Zhang, X. Lu, R. Sun and J. Min, *Energy Environ. Sci.*, 2023, **16**, 3942-3950.
122. J. Wang, Y. Cui, Y. Xu, K. Xian, P. Bi, Z. Chen, K. Zhou, L. Ma, T. Zhang, Y. Yang, Y. Zu, H. Yao, X. Hao, L. Ye and J. Hou, *Adv. Mater.*, 2022, **34**, 2205009.
123. Z. Jia, Q. Ma, L. Meng, J. Zhang, S. Qin, Z. Chen, X. Li, J. Zhang, J. Li, Z. Zhang, Z. Wei, Y. Yang and Y. Li, *Adv. Funct. Mater.*, 2022, **32**, 2204720.
124. X. Xu, T. Yu, Z. Bi, W. Ma, Y. Li and Q. Peng, *Adv. Mater.*, 2018, **30**, 1703973.
125. P. Cong, X. Li, A. Tang, J. Wu, J. Chen, L. Chen and E. Zhou, *Chem. Commun.*, 2022, **58**, 13373-13376.
126. A. Tang, W. Song, B. Xiao, J. Guo, J. Min, Z. Ge, J. Zhang, Z. Wei and E. Zhou, *Chem. Mater.*, 2019, **31**, 3941-3947.
127. T. Dai, A. Tang, Z. He, M. Du, P. Lei, Q. Zeng, Z. Wang, Y. Wang, S. Lu, Y. Zhong and E. Zhou, *Energy Environ. Sci.*, 2023, **16**, 2199-2211.