C-Shaped *ortho*-benzodipyrrole Based Acceptors with Different Electronic Effect of Top Substituents for As-Cast Green-Solvent Processed High-Performance Organic Solar Cells

Yufei Gong[1,2†], Tianwei Zou[1,2†], Xiaojun Li[1,2*], Shucheng Qin[1,2], Guangpei Sun[1,2], TongLing Liang[2,3], Ruimin Zhou[4], Jianqi Zhang[2,5], Jinyuan Zhang[1,2], Lei Meng[1,2], Zhixiang Wei[2,5], Yongfang Li[1,2,6*]

1 Beijing National Laboratory for Molecular Sciences, CAS Key Laboratory of Organic Solids, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China.

2 School of Chemical Science, University of Chinese Academy of Sciences, Beijing 100049, China.

3 Center for Physicochemical Analysis and Measurement, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China.

4 College of Chemistry and Green Catalysis Center, Zhengzhou University, Zhengzhou 450001, China.

5 CAS Key Laboratory of Nanosystem and Hierarchical Fabrication, National Center for Nanoscience and Technology, Beijing 100190, China.

6 Laboratory of Advanced Optoelectronic Materials, College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou 215123, China.

Materials and Synthesis

Tributyl(6-(2-ethylhexyl)thieno[3,2-b]thiophen-2-yl)stannane, 5-bromomethyl-undecane and 2-(5,6-dichloro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile were sourced from Hyper Inc. 1,4-dibromo-2,3-difluoro-5,6-dinitrobenzene, 1,4-dibromo-2-fluoro-3-methyl-5,6-dinitrobenzene and 1,4-dibromo-2,3-dimethyl-5,6-dinitrobenzene were commercially available from Zhengzhou Alfa Chemical Ltd. The polymer donor PBQx-TCl was provided by Solarmer Materials Inc. Other chemical reagents and solvents were purchased from J&K Scientific and Nanjing Zhiyan Technology Ltd. Unless otherwise specified, all chemicals used directly without further purification.
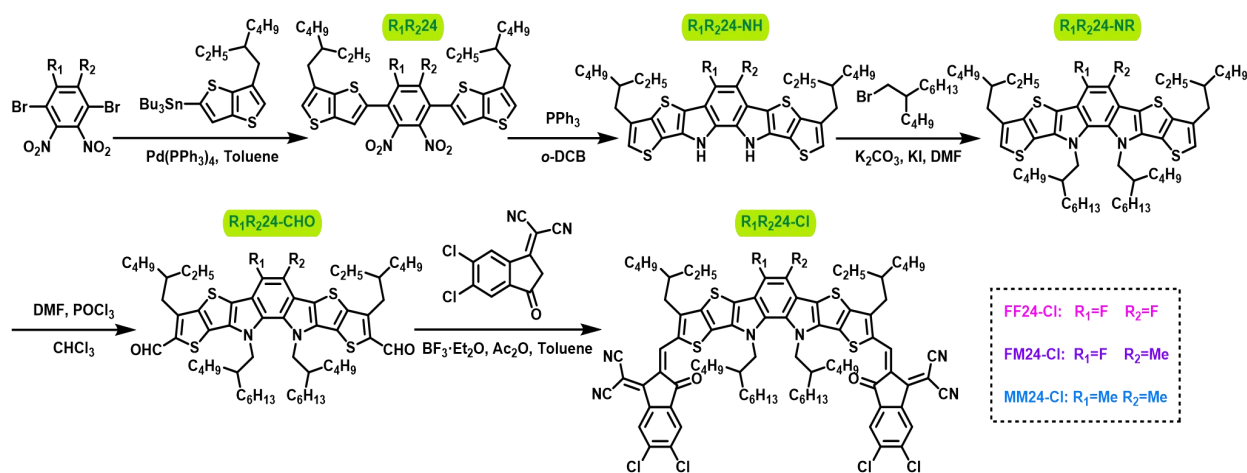
Scheme S1. Synthesis routes of FF24-Cl, FM24-Cl and MM24-Cl.

Synthesis of compound FF24

1,4-dibromo-2,3-difluoro-5,6-dinitrobenzene (723.8 mg, 2.0 mmol), tributyl(6-(2-ethylhexyl)thieno[3,2-b]thiophen-2-yl)stannane (2707.4 mg, 5.0 mmol) and tetrakis(triphenylphosphine)palladium (115.6 mg, 0.1 mmol) were dissolved in 25 mL of toluene. The reaction mixture was heated to 110 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether/dichloromethane (3/1, v/v) as the eluent to give a orange red solid (968.1 mg, 68.7% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 7.32 (s, 2H), 7.02 (s, 2H), 2.60 (d, J = 6.5 Hz, 4H), 1.71 (m, 2H), 1.28-1.20 (m, 16H), 0.86-0.78 (m, 12H). MALDI-FTICR-MS [M+H]$^+$ calcd. for (C$_{34}$H$_{39}$F$_2$N$_2$O$_4$S$_4$): 705.1755; found: 705.1762.

Synthesis of compound FM24

1,4-dibromo-2-fluoro-3-methyl-5,6-dinitrobenzene (715.8 mg, 2.0 mmol), tributyl(6-(2-ethylhexyl)thieno[3,2-b]thiophen-2-yl)stannane (2707.4 mg, 5.0 mmol) and tetrakis(triphenylphosphine)palladium (115.6 mg, 0.1 mmol) were dissolved in 25 mL of toluene. The reaction mixture was heated to 110 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether/dichloromethane (3/1, v/v) as the eluent to give a orange red solid (935.4 mg, 66.7% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 7.35 (s, 2H), 7.05 (s, 2H), 2.66 (m, 7H), 1.78 (m, 2H), 1.30-1.24 (m, 16H), 0.87-0.81 (m, 12H). MALDI-FTICR-MS [M+H]$^+$ calcd. for (C$_{35}$H$_{42}$FN$_2$O$_4$S$_4$): 701.2006; found: 701.2016.

Synthesis of compound MM24

1,4-dibromo-2,3-dimethyl-5,6-dinitrobenzene (707.9 mg, 2.0 mmol), tributyl(6-(2-ethylhexyl)thieno[3,2-b]thiophen-2-yl)stannane (2707.4 mg, 5.0 mmol) and tetrakis(triphenylphosphine)palladium (115.6 mg, 0.1 mmol) were dissolved in 25 mL of toluene. The reaction mixture was heated to 110 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether/dichloromethane (3/1, v/v) as the eluent to give a orange red solid (922.7 mg, 66.2% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 7.11 (s, 2H), 7.03 (s, 2H), 2.63 (m, 10H), 1.76 (m, 2H), 1.37-1.31 (m, 16H), 0.94-0.86 (m, 12H). MALDI-FTICR-MS [M+H]$^+$ calcd. for (C$_{36}$H$_{45}$N$_2$O$_4$S$_4$): 697.2257; found: 697.2267.

Synthesis of compound FF24-NH

Compound FF24 (704.9 mg, 1.0 mmol) and triphenylphosphine (2622.9 mg, 10.0 mmol) were dissolved in 5 mL of 1,2-dichlorobenzene. The reaction mixture was heated to 180 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was purified by column chromatography on silica gel using dichloromethane/ethyl acetate (4/1, v/v) as the eluent to give a dark brown solid (338.3 mg, 48.0% yield). MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{34}$H$_{38}$F$_2$N$_2$S$_4$): 640.1880; found: 640.1891.

Synthesis of compound FM24-NH

Compound FM24 (701.0 mg, 1.0 mmol) and triphenylphosphine (2622.9 mg, 10.0 mmol) were dissolved in 5 mL of 1,2-dichlorobenzene. The reaction mixture was heated to 180 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was purified by column chromatography on silica gel using dichloromethane/ethyl acetate (4/1, v/v) as the eluent to give a dark brown solid (362.1 mg, 56.8% yield). MALDI-FTICR-MS [M]$^+$ calcd. for ($C_{35}H_{41}FN_2S_4$): 636.2131; found: 636.2136.

Synthesis of compound MM24-NH

Compound MM24 (697.0 mg, 1.0 mmol) and triphenylphosphine (2622.9 mg, 10.0 mmol) were dissolved in 5 mL of 1,2-dichlorobenzene. The reaction mixture was heated to 180 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was purified by column chromatography on silica gel using dichloromethane/ethyl acetate (4/1, v/v) as the eluent to give a dark brown solid (350.7 mg, 50.3% yield). MALDI-FTICR-MS [M]$^+$ calcd. for ($C_{36}H_{44}N_2S_4$): 632.2382; found: 632.2391.

Synthesis of compound FF24-NR

Compound FF24-NH (640.9 mg, 1.0 mmol), 5-bromomethyl-undecane (996.9 mg, 4.0 mmol), potassium carbonate (1382.1 mg, 10.0 mmol) and potassium iodide (664.0 mg, 4.0 mmol) were dissolved in 25 mL of N,N-dimethylformamide. The reaction mixture was heated to 120 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether as the eluent to give a faint yellow solid (748.6 mg, 76.6% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 6.98 (s, 2H), 4.49 (d, J = 7.8 Hz, 4H), 2.72 (d, J = 7.3 Hz, 4H), 2.03 (m, 2H), 1.90 (m, 2H), 1.43-1.25 (m, 16H), 1.06-0.83 (m, 44H), 0.76-0.61 (m, 12H). MALDI-FTICR-MS [M]$^+$ calcd. for ($C_{58}H_{86}F_2N_2S_4$): 976.5636; found: 976.5643.

Synthesis of compound FM24-NR

Compound FM24-NH (637.0 mg, 1.0 mmol), 5-bromomethyl-undecane (996.9 mg, 4.0 mmol), potassium carbonate (1382.1 mg, 10.0 mmol) and potassium iodide (664.0 mg, 4.0 mmol) were dissolved in 25 mL of N,N-dimethylformamide. The reaction mixture was heated to 120 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether as the eluent to give a faint yellow solid (733.1 mg, 75.3% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 7.02 (s, 2H), 4.62 (d, J = 7.8 Hz, 4H), 2.79 (m, 7H), 2.16 (m, 2H), 2.02 (m, 2H), 1.49-1.39 (m, 16H), 1.16-0.93 (m, 44H), 0.86-0.71 (m, 12H). MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{59}$H$_{89}$FN$_2$S$_4$): 972.5887; found: 972.5890.

Synthesis of compound MM24-NR

Compound MM24-NH (633.0 mg, 1.0 mmol), 5-bromomethyl-undecane (996.9 mg, 4.0 mmol), potassium carbonate (1382.1 mg, 10.0 mmol) and potassium iodide (664.0 mg, 4.0 mmol) were dissolved in 25 mL of N,N-dimethylformamide. The reaction mixture was heated to 120 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether as the eluent to give a faint yellow solid (745.0 mg, 76.8% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 6.92 (s, 2H), 4.48 (d, J = 7.8 Hz, 4H), 2.74 (m, 10H), 2.01 (m, 2H), 1.91 (m, 2H), 1.40-1.26 (m, 16H), 1.04-0.80 (m, 44H), 0.76-0.60 (m, 12H). MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{60}$H$_{92}$N$_2$S$_4$): 968.6138; found: 968.6143.

Synthesis of compound FF24-CHO

Compound FF24-NR (195.5 mg, 0.2 mmol) and N,N-dimethylformamide (365.5 mg, 5.0 mmol) were dissolved in 10 mL of chloroform. After cooling to room temperature, phosphorus oxychloride (613.3 mg, 4.0 mmol) was added dropwise. The reaction mixture was heated to 60 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was poured into the saturated solution of sodium bicarbonate in water, extracted with chloroform for three times, concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether/dichloromethane (1/1, v/v) as the eluent to give a bright yellow solid (176.4 mg, 85.3% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 10.13 (s, 2H), 4.53 (d, J = 7.8 Hz, 4H), 3.09 (d, J = 7.5 Hz, 4H), 1.99 (m, 4H), 1.46-1.26 (m, 16H), 1.05-0.86 (m, 44H), 0.74-0.62 (m, 12H). MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{60}$H$_{86}$F$_2$N$_2$O$_2$S$_4$): 1032.5535; found: 1032.5542.

Synthesis of compound FM24-CHO

Compound FM24-NR (194.7 mg, 0.2 mmol) and N,N-dimethylformamide (365.5 mg, 5.0 mmol) were dissolved in 10 mL of chloroform. After cooling to room temperature, phosphorus oxychloride (613.3 mg, 4.0 mmol) was added dropwise. The reaction mixture was heated to 60 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was poured into the saturated solution of sodium bicarbonate in water, extracted with chloroform for three times, concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether/dichloromethane (1/1, v/v) as the eluent to give a bright yellow solid (187.8 mg, 91.2% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 10.12 (s, 2H), 4.56 (d, J = 7.8 Hz, 4H), 3.09 (d, J = 7.3 Hz, 4H), 2.69 (s, 3H), 2.00 (m, 4H), 1.47-1.30 (m, 16H), 1.09-0.83 (m, 44H), 0.75-0.62 (m, 12H). MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{61}$H$_{89}$FN$_2$O$_2$S$_4$): 1028.5785; found: 1028.5792.

Synthesis of compound MM24-CHO

Compound MM24-NR (193.9 mg, 0.2 mmol) and N,N-dimethylformamide (365.5 mg, 5.0 mmol) were dissolved in 10 mL of chloroform. After cooling to room temperature, phosphorus oxychloride (613.3 mg, 4.0 mmol) was added dropwise. The reaction mixture was heated to 60 °C and stirred for 12 h under argon atmosphere. After cooling to room temperature, the reaction mixture was poured into the saturated solution of sodium bicarbonate in water, extracted with chloroform for three times, concentrated by vacuum evaporation and purified by column chromatography on silica gel using petroleum ether/dichloromethane (1/1, v/v) as the eluent to give a bright yellow solid (167.1 mg, 81.5% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 10.10 (s, 2H), 4.51 (d, J = 7.8 Hz, 4H), 3.08 (d, J = 7.3 Hz, 4H), 2.72 (s, 6H), 1.98 (m, 4H), 1.45-1.24 (m, 16H), 1.10-0.76 (m, 44H), 0.75-0.61 (m, 12H). MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{62}$H$_{92}$N$_2$O$_2$S$_4$): 1024.6036; found: 1024.6043.

Synthesis of compound FF24-Cl

Compound FF24-NR (206.7 mg, 0.2 mmol), 2-(5,6-dichloro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (131.5 mg, 0.5 mmol), boron trifluoride diethyl etherate (354.8 mg, 2.5 mmol) and acetic anhydride (102.1 mg, 1.0 mmol) were dissolved in 5 mL of toluene. After stirring for 1 h, the reaction mixture was purified by column chromatography on silica gel using petroleum ether/dichloromethane (1/1, v/v) as the eluent to give a dark blue solid (233.5 mg, 76.6% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 9.16 (s, 2H), 8.79 (s, 2H), 7.96 (s, 2H), 4.68 (d, J = 7.2 Hz, 4H), 3.14 (d, J = 7.1 Hz, 4H), 2.10 (m, 2H), 1.97 (m, 2H), 1.50-1.30 (m, 16H), 1.18-0.85 (m, 44H), 0.76-0.66 (m, 12H). ^{13}C NMR (101 MHz, CDCl$_3$, δ/ppm): 186.10, 158.95, 153.55, 145.53, 139.62, 139.25, 139.10, 138.71, 136.76, 136.39, 136.08, 135.64, 134.46, 129.03, 126.94, 126.69, 125.02, 120.32, 115.12, 114.51, 69.07, 55.40, 41.60, 41.57, 38.83, 34.38, 32.68, 31.59, 30.40, 30.30, 29.71, 29.44, 28.76, 27.86, 27.80, 26.03, 25.47, 25.41, 22.99, 22.81, 22.52, 14.13, 14.07, 14.05, 13.74, 10.99. ^{19}F NMR (659 MHz, CDCl$_3$, δ/ppm): -154.55. MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{84}$H$_{90}$Cl$_4$F$_2$N$_6$O$_2$S$_4$): 1520.4725; found: 1520.4739.

Synthesis of compound FM24-Cl

Compound FM24-NR (205.9 mg, 0.2 mmol), 2-(5,6-dichloro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (131.5 mg, 0.5 mmol), boron trifluoride diethyl etherate (354.8 mg, 2.5 mmol) and acetic anhydride (102.1 mg, 1.0 mmol) were dissolved in 5 mL of toluene. After stirring for 1 h, the reaction mixture was purified by column chromatography on silica gel using petroleum ether/dichloromethane (1/1, v/v) as the eluent to give a dark blue solid (224.0 mg, 73.7% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 9.16 (s, 2H), 8.79 (s, 2H), 7.96 (s, 2H), 4.66 (d, J = 7.2 Hz, 4H), 3.16 (d, J = 7.1 Hz, 4H), 2.71 (s, 3H), 2.08 (m, 2H), 1.98 (m, 2H), 1.48-1.29 (m, 16H), 1.18-0.79 (m, 44H), 0.73-0.68 (m, 12H). ^{13}C NMR (101 MHz, CDCl$_3$, δ/ppm): 186.09, 159.06, 153.73, 153.55, 150.74, 148.34, 145.46, 145.08, 145.06, 139.42, 139.07, 138.98, 138.74, 138.52, 136.42, 136.32, 136.26, 136.21, 136.10, 134.32, 131.85, 131.76, 130.36, 130.31, 129.43, 127.12, 126.86, 124.92, 124.29, 124.22, 119.81, 119.76, 115.31, 115.28, 114.67, 114.44, 114.17, 108.62, 108.45, 68.49, 68.41, 55.32, 55.25, 41.64, 41.60, 38.81, 38.79, 34.37, 32.67, 32.65, 31.58, 30.47, 30.41, 30.36, 30.32, 29.43, 28.76, 28.73, 27.89, 27.86, 27.81, 27.80, 26.03, 25.48, 25.43, 25.39, 25.35, 23.00, 22.81, 22.80, 22.53, 14.07, 13.74, 11.24, 11.21, 11.00, 10.99. ^{19}F NMR (659 MHz, CDCl$_3$, δ/ppm): -133.56. MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{85}$H$_{93}$Cl$_4$FN$_6$O$_2$S$_4$): 1516.4976; found: 1516.4997.

Synthesis of compound MM24-Cl

Compound MM24-NR (205.1 mg, 0.2 mmol), 2-(5,6-dichloro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (131.5 mg, 0.5 mmol), boron trifluoride diethyl etherate (354.8 mg, 2.5 mmol) and acetic anhydride (102.1 mg, 1.0 mmol) were dissolved in 5 mL of toluene. After stirring for 1 h, the reaction mixture was purified by column chromatography on silica gel using petroleum ether/dichloromethane (1/1, v/v) as the eluent to give a dark blue solid (201.2 mg, 66.4% yield). ^{1}H NMR (400 MHz, CDCl$_3$, δ/ppm): 9.11 (s, 2H), 8.74 (s, 2H), 7.93 (s, 2H), 4.67 (d, J = 7.7 Hz, 4H), 3.13 (d, J = 7.6 Hz, 4H), 2.73 (s, 6H), 2.06 (m, 2H), 1.98 (m, 2H), 1.49-1.29 (m, 16H), 1.18-0.82 (m, 44H), 0.73-0.68 (m, 12H). ^{13}C NMR (101 MHz, CDCl$_3$, δ/ppm): 186.02, 159.07, 153.65, 144.99, 139.20, 138.87, 138.75, 138.47, 136.94, 136.10, 134.26, 131.83, 131.27, 126.74, 125.18, 124.80, 122.57, 119.29, 115.46, 114.82, 67.86, 55.10, 41.65, 41.61, 38.79, 34.34, 32.63, 31.59, 30.47, 30.39, 30.33, 29.44, 28.72, 27.90, 27.80, 26.03, 25.44, 25.33, 23.01, 22.79, 22.55, 15.96, 14.09, 13.75, 13.73, 11.00. ^{19}F NMR (659 MHz, CDCl$_3$, δ/ppm): no signal. MALDI-FTICR-MS [M]$^+$ calcd. for (C$_{86}$H$_{96}$Cl$_4$N$_6$O$_2$S$_4$): 1512.5226; found: 1512.5246.

Fabrication and Characterization

^{1}H, ^{13}C and ^{19}F nuclear magnetic resonance (NMR) spectroscopy were recorded on Bruker AV 400 and Bruker NEO 700 spectrometer in a CDCl$_3$ solution at 300 K. Chemical shifts (δ) are reported in parts per million (ppm) with tetramethylsilane (TMS) as a benchmark. Matrix-assisted laser desorption/ionization Fourier-transform ion cyclotron resonance mass spectrometry (MALDI-FTICR-MS) was collected on Bruker Solarix.

Thermogravimetric analysis (TGA) was performed by PerkinElmer TGA8000 under nitrogen atmosphere with a heating rate of 20 °C min^{-1} from 50 °C to 500 °C. Differential scanning calorimetry (DSC) was performed by DSC00881 under nitrogen atmosphere with a heating rate of 10 °C min^{-1} from 50 °C to 300 °C.

Ultraviolet-visible (UV-Vis) and temperature-dependent UV-Vis absorption spectra were obtained on HITACHI UH5700 and PerkinElmer Lambda 1050+, respectively. The SMAs were dissolved in *o*-xylene and spin-coated on quartz-plates to measure the absorption of the solution and film, respectively. Photoluminescence (PL) spectra was obtained on Horiba FluoroMax+. The excitation wavelength is 600 nm and the slit width is 3 nm.

Cyclic voltammetry (CV) was conducted by CHI660C, with glassy carbon as the working electrode, platinum wire as the counter electrode and Ag/AgCl as the reference electrode. The scan rate is 0.1 V s^{-1} and the supporting electrolyte is the acetonitrile solution of tetrabutylammonium hexafluorophosphate with a concentration of 0.1 mol L^{-1}. The solutions of the SMAs were dropped onto the working electrode to form thin films, and ferrocene/ferrocenium (Fc/Fc$^+$) redox couple was used as internal standard.

Unless otherwise specified, all calculations were conducted by ORCA (version 5.0.4)[1]. For simplifying the calculation, the outer and inner chains of the SMAs were replaced with methyl and isobutyl, respectively. The geometry optimization and energy calculation were performed under ωB97X-D3 function with def2-SVP basis-set. Multiwfn (version 3.8)[2] and VMD (version 1.9.4)[3] were used for analysis and visualization, respectively.

Single crystals of FF24-Cl, FM24-Cl and MM24-Cl were cultivated using a solvent diffusion method with methanol as a poor solvent and chloroform as a good solvent. According to standard procedure, X-ray single crystal data were collected on XtaLAB Synergy-R at 170 K protected by liquid nitrogen, analyzed using Mercury (version 4.0), and deposited at the Cambridge Crystallographic Data Center (CCDC).

The organic solar cells (OSCs) were fabricated with conventional device architecture of indium tin oxide (ITO) / poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) / Active Layer / N,N'-Bis{3-[3-(Dimethylamino)propylamino]propyl}perylene-3,4,9,10-tetracarboxylic diimide (PDINN) / Ag. The pre-patterned ITO glasses substrates (sheet resistance is 15 Ω sq^{-1}) were sonicated with deionized water (with and without detergent), acetone and isopropanol for 20 minutes in sequence. Before being used, they were dried in an oven and then treated by Ultraviolet-Ozone for 30 minutes. The PEDOT:PSS aqueous solution was filtered through a 0.45 mm filter and spin-coated on the pre-cleaned ITO glasses at 7000 rpm for 30 seconds, and then dried in air at 150 °C for 20 minutes. The PEDOT:PSS coated ITO substrates were transferred to the nitrogen-filled glove box for further processing. The recipe for the optimized device is as follows. The o-xylene was selected as the solvent and the total concentration of donor and acceptor is 9 mg mL^{-1}, with a weight ratio of 1:1.2. After stirring overnight for intensive mixing, the solution was spin-coated on the top of the PEDOT:PSS layer at 2500 rpm for 30 seconds. Then, the methanol solution of PDINN with a concentration of 1.0 mg mL^{-1} was deposited on the active layer at 3000 rpm for 30 seconds. Under a pressure of 10^{-4} Pa, silver was thermally evaporated to form the top electrode through a mask. The photovoltaic area of the device is 6.0 mm^2 (3 mm × 2 mm).

The current density-voltage (J-V) characteristic of OSCs were measured in the nitrogen-filled glove box equipped with Keysight B2901A, using Newport 94023A with 450 W xenon lamp and AM 1.5G filter as the light source. The light intensity was calibrated to 100 mW cm^{-2} by Enlitech SRC2020. The external quantum efficiency (EQE) of OSCs was measured by Enlitech QE-RT3011. The light intensity at each wavelength was calibrated by standard single-crystal silicon photovoltaic cells.

The hole and electron mobility were measured by using the method of space charge limited current (SCLC), and the device architectures were ITO/PEDOT:PSS/Active Layer/MoO$_3$/Ag and ITO/ZnO/Active Layer/PDINN/Al, respectively. They can be calculated according to the equation $J = 9\varepsilon_0\varepsilon_r\mu V^2/8d^3$. Where, J is the current density, V is the applied voltage minus the offset voltage, ε_0 is the vacuum permittivity, ε_r is the relative permittivity, μ is the carrier mobility, and d is the thickness of the active layer.

The exciton dissociation (η_{dis}) behavior was reflected by the curve of photocurrent density (J_{ph}) versus effective voltage (V_{eff}). J_{ph} is defined as $J_{ph} = J_{light} - J_{dark}$ and V_{eff} is defined as $V_{eff} = V_0 - V_{bias}$. Here, J_{light} and J_{dark} are the photocurrent density under standard light condition and dark condition respectively. The V_0 is the voltage when J_{ph} is zero, and V_{bias} is the applied bias voltage. The V_{eff} will affect the electric field in the device and influence the processes of exciton dissociation and charge collection. J_{ph} and V_{eff} initially shows a positive correlation. As V_{eff} increases to a certain extent, J_{ph} becomes saturated because all the excitons are dissociated into free charges and collected by the corresponding electrodes. In this work, the J_{ph} at $V_{eff} = 3.8$ V is selected as the saturation current density (J_{sat}), which represents the maximum free charge that can be collected. Therefore, the η_{dis} of these devices can be determined by the ratio of J_{ph} and J_{sat} under short-circuit condition.

The data of transient photocurrent (TPC), transient photovoltage (TPV) and photoinduced charge carrier extraction by linearly increasing voltage (Photo-CELIV) were obtained by the all-in-one characterization platform, Paios (Fluxim AG, Switzerland). In TPC testing, the light intensities were 10%, 17.8%, 31.6%, 56.2%, and 100% sunlight, respectively. The settling time was 100 µs, pulse length was 100 µs and the follow-up time was 200 µs. In the TPV testing, the light intensities were 0.10%, 0.23%, 0.53%, 1.23%, 2.83%, 6.52%, 15.0%, 34.6%, and 80.0% sunlight, respectively. The settling time was 30 ms, pulse length was 5 ms and the follow-up time was 30 µs. In the Photo-CLIVE testing, the delay time was set to 0 s, the light intensity was 100% sunlight, the pulse length was 100 µs, and the sweep ramp rate increased from 20 V ms^{-1} to 100 V ms^{-1}.

Grazing-incidence wide-angle X-ray scattering (GIWAXS) measurement was conducted at XEUSS small-angle X-ray scattering (SAXS) / wide-angle X-ray scattering (WAXS) equipment. The films to be tested were prepared by spin-coating all samples on silicon wafers. The packing distance (d) can be calculated using $d = 2\pi/q$, where q is the corresponding peak position, and crystalline coherence length (CCL) can be calculated using $CCL = 2\pi K/\Delta q$, where Δq was the full width at half maximum (FWHM) of the scattering peak, and Scherrer constant K was taken as 0.9.

Contact angle (CA) test was conducted through OCA 25 with water and diiodomethane. Atomic force microscope (AFM) and transmission electron microscopy (TEM) images were obtained by Bruker ICON2-SYS and JEOL JEM-1400, respectively. All samples were spin-coated on the ITO/PEDOT:PSS substrate. Some of them were peeled off by floating on the surface of deionized water, and then the floating films were picked up by copper mesh.
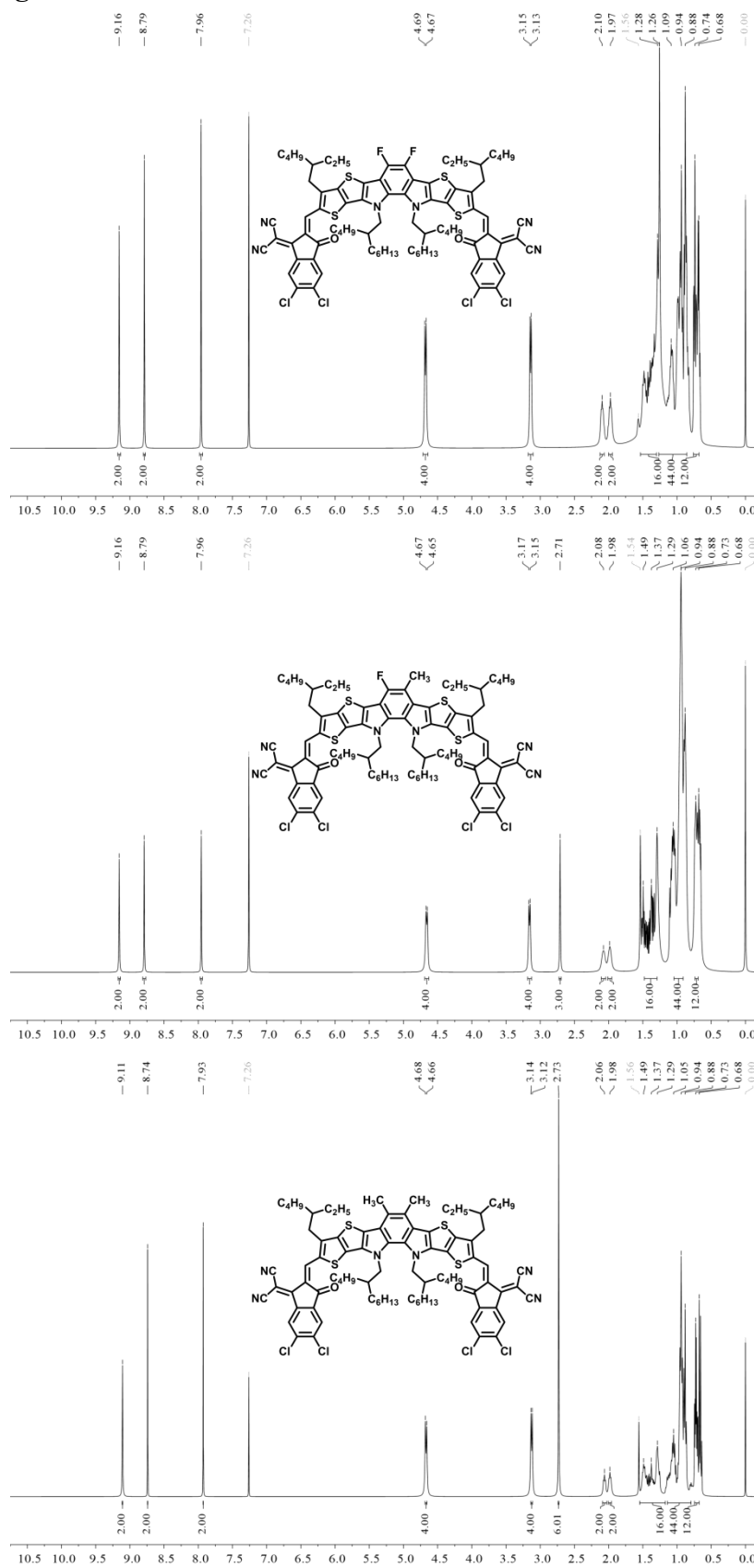
Supplementary Figures

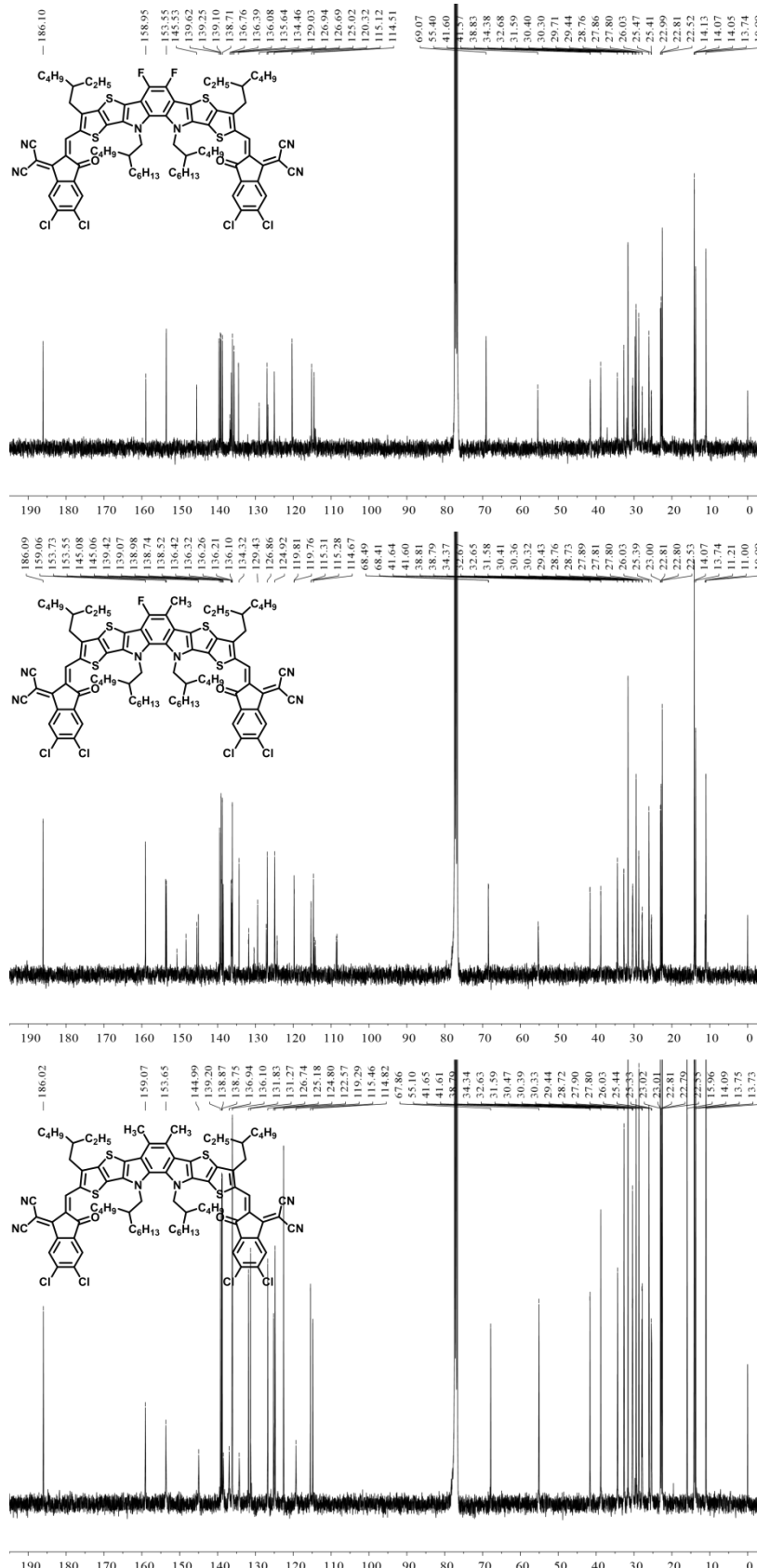**Figure S1.** ^{1}H NMR spectrum of FF24-Cl/FM24-Cl/MM24-Cl in CDCl$_3$.

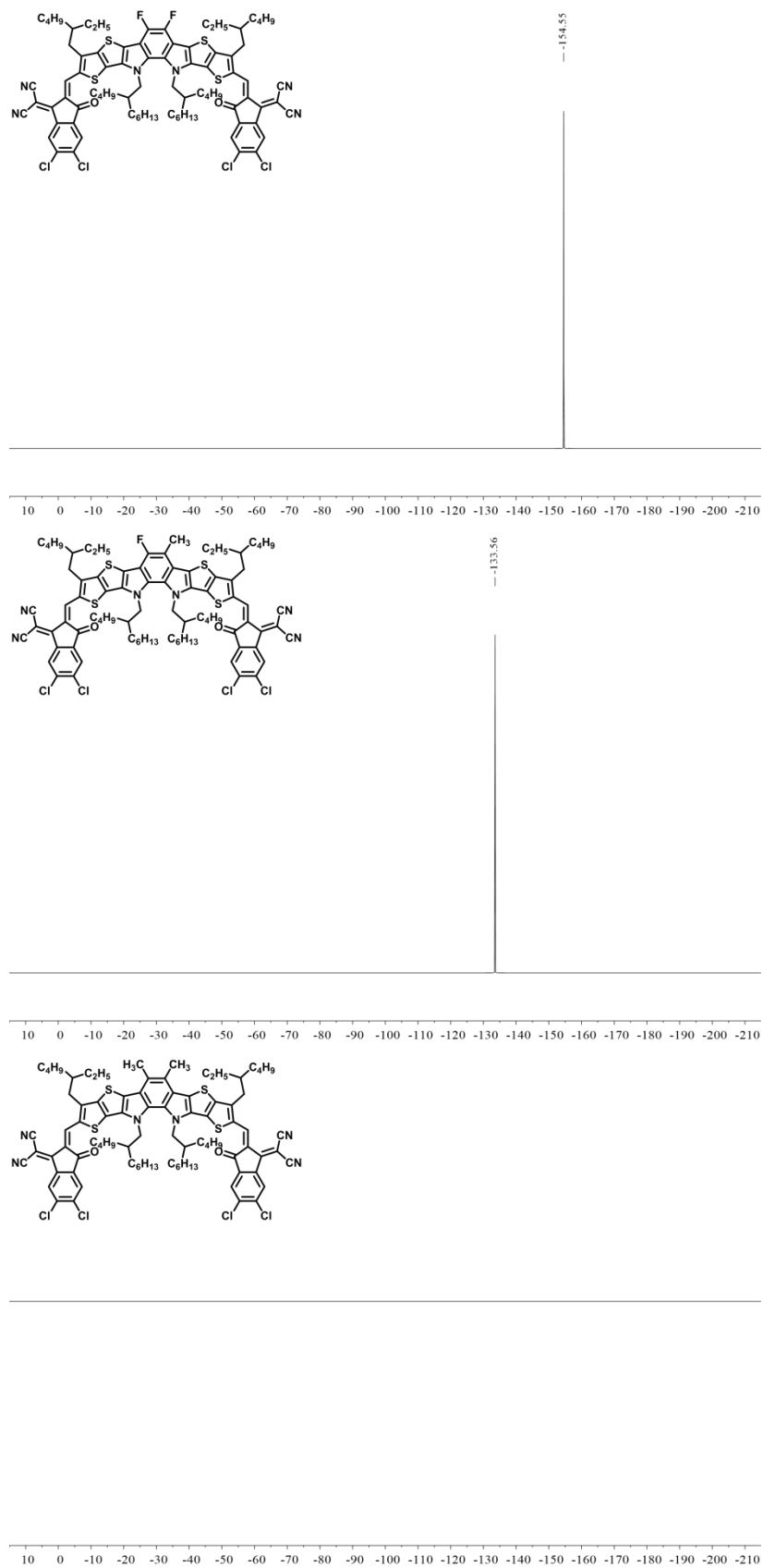**Figure S2.** ^{13}C NMR spectrum of FF24-Cl/FM24-Cl/MM24-Cl in CDCl$_3$.

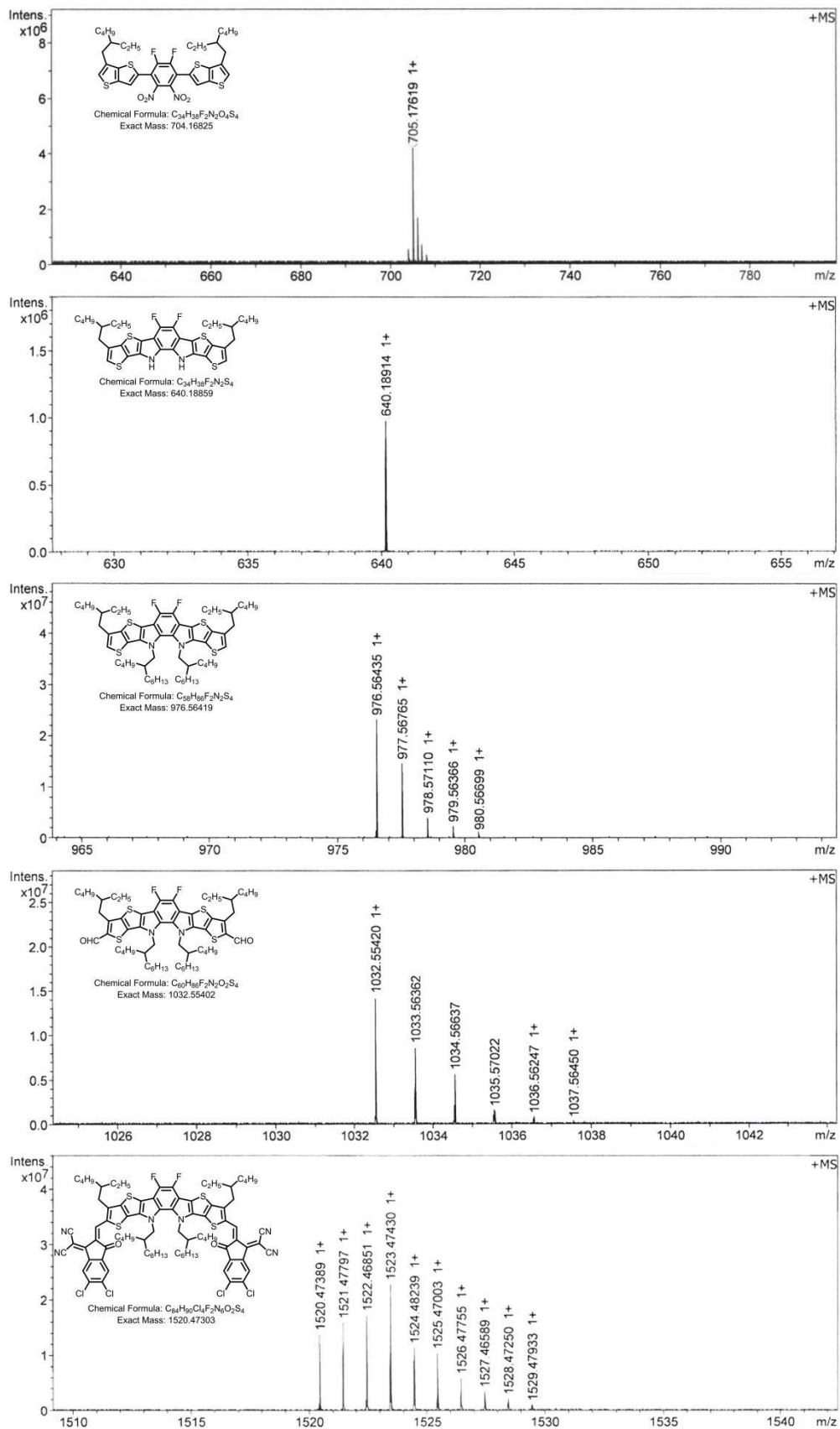**Figure S3.** ^{19}F NMR spectrum of FF24-Cl/FM24-Cl/MM24-Cl in CDCl$_3$.

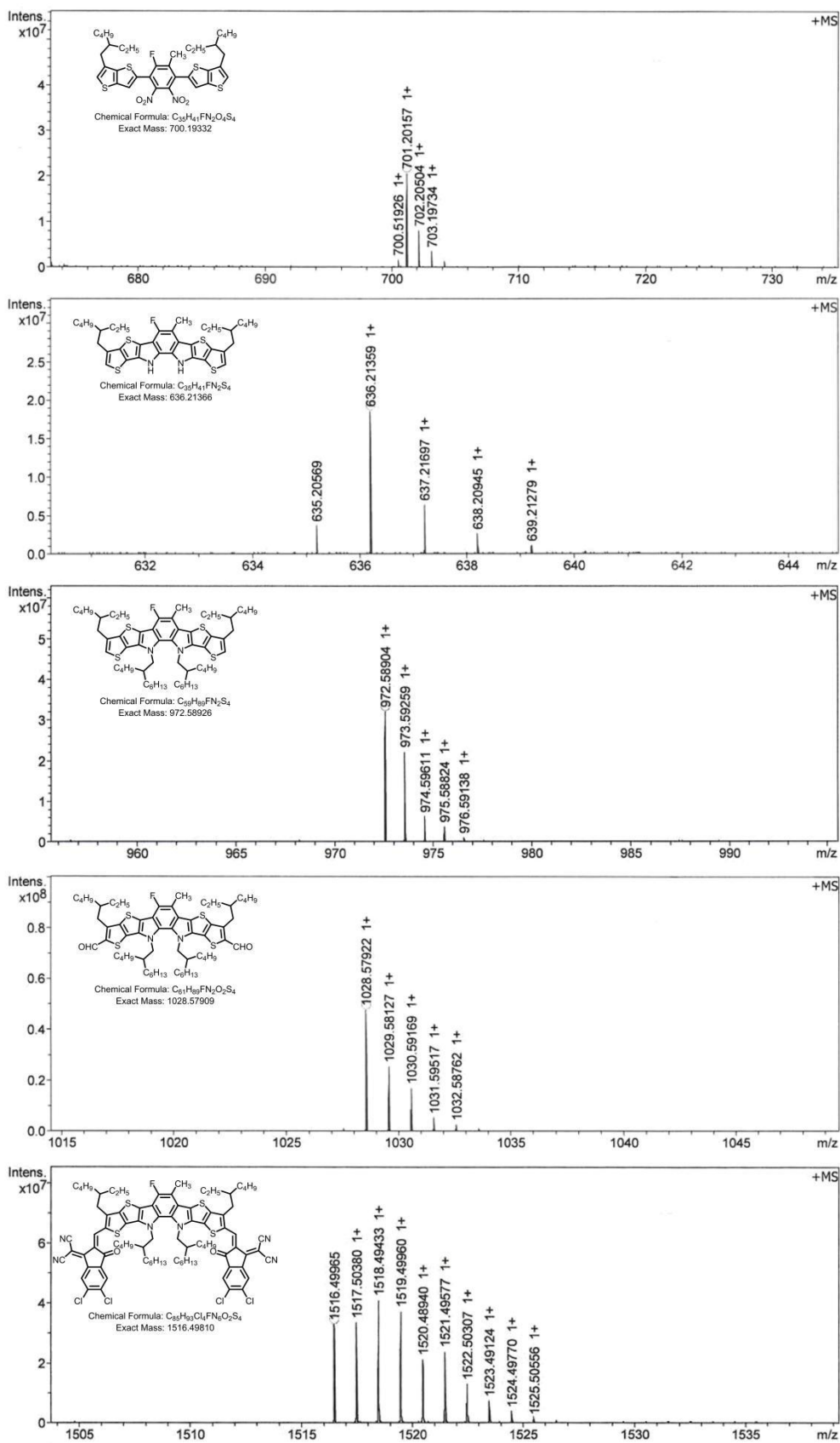**Figure S4.** MALDI-FTICR-MS spectrum of FF24-Cl and its intermediates.

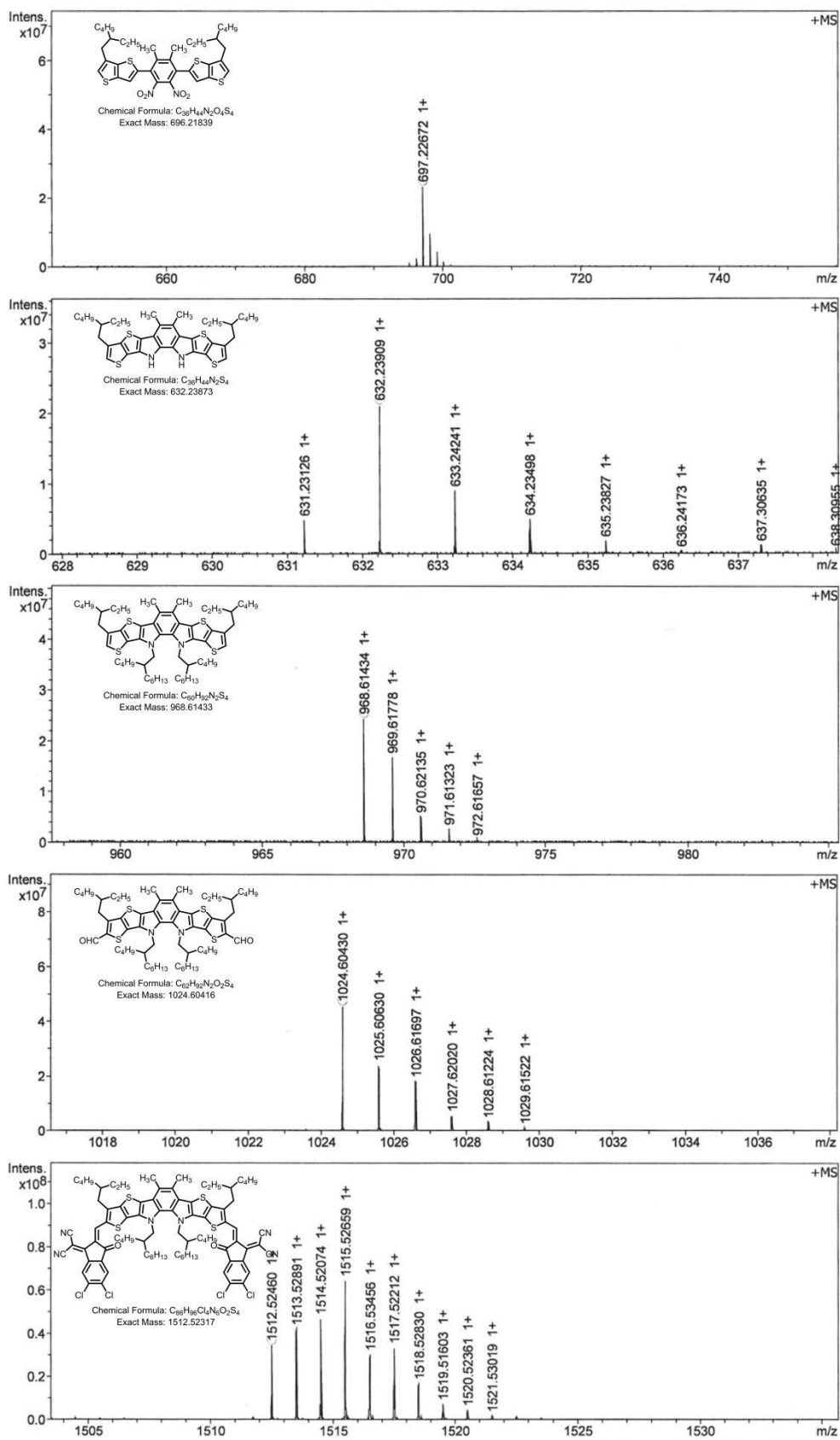**Figure S5.** MALDI-FTICR-MS spectrum of FM24-Cl and its intermediates.

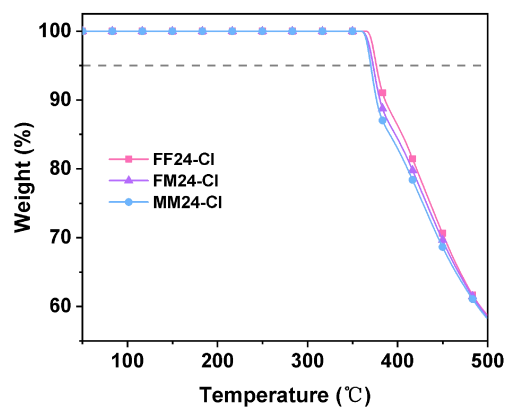**Figure S6.** MALDI-FTICR-MS spectrum of MM24-Cl and its intermediates.

Figure S7. TGA curves of FF24-Cl, FM24-Cl and MM24-Cl under nitrogen atmosphere with a heating rate of 20 °C min^{-1} from 50 °C to 500 °C.
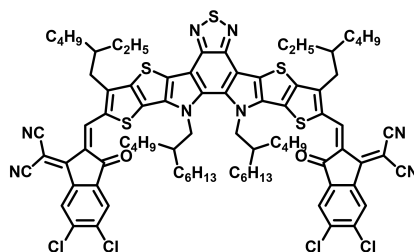

Figure S8. Chemical structure of BT24-Cl.
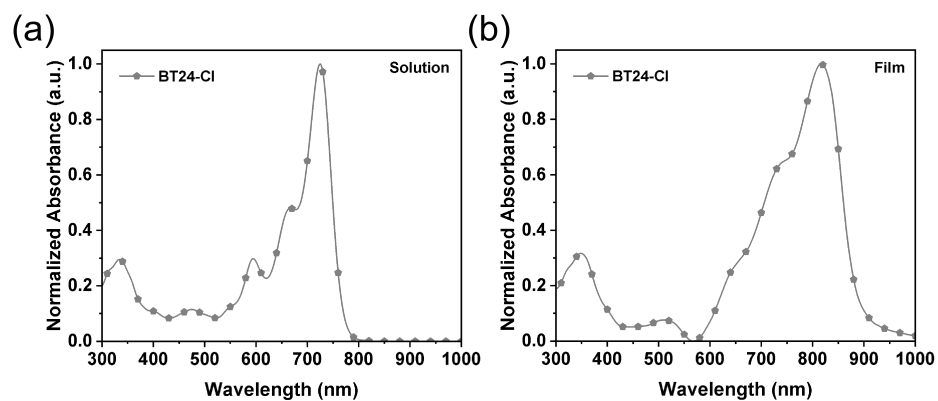
Figure S9. Normalized absorption spectra of BT24-Cl (a) in *o*-xylene solution and (b) in thin film.

Figure S10. Temperature-dependent UV-Vis absorption spectra of FF24-Cl, FM24-Cl and MM24-Cl in *o*-xylene solution.
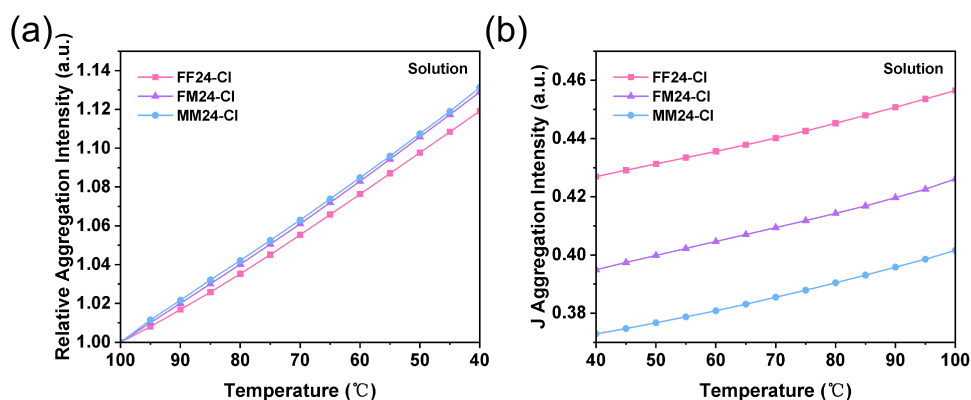
Figure S11. Temperature-dependent (a) relative aggregation intensity and (b) J aggregation intensity curves of FF24-Cl, FM24-Cl and MM24-Cl in *o*-xylene solution.
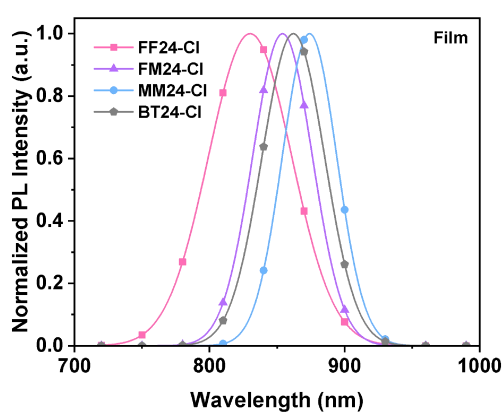
Figure S12. Normalized photoluminescence (PL) spectra of FF24-Cl, FM24-Cl, MM24-Cl and BT24-Cl in thin film.
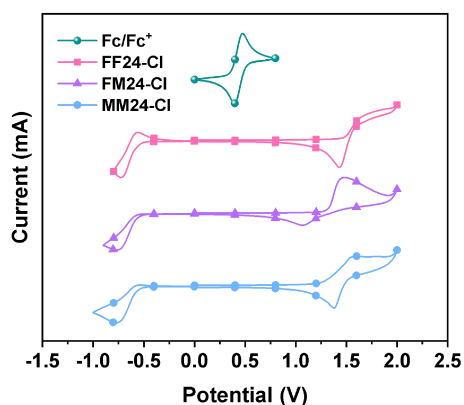
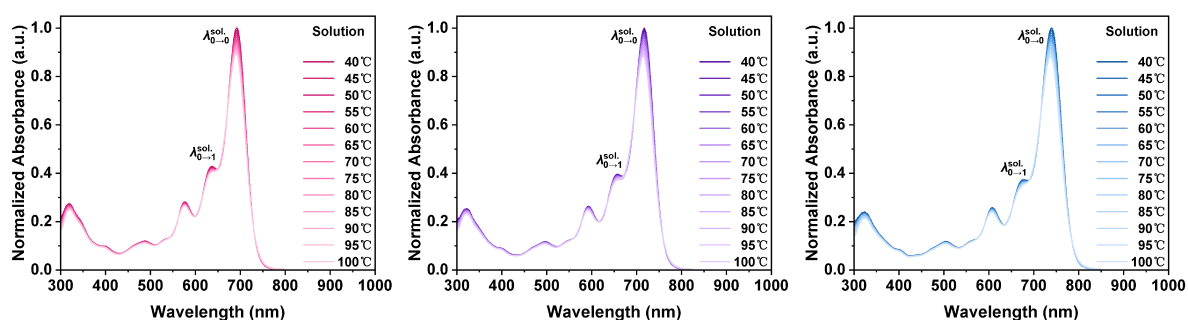**Figure S13.** CV curves of the FF24-Cl, the FM24-Cl and the MM24-Cl based film on Pt electrode in acetonitrile solution with Ag/AgCl as reference electrode.
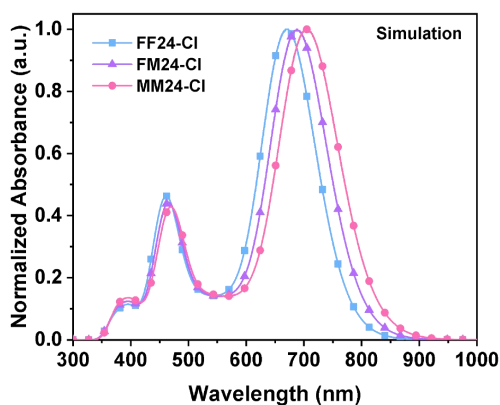
Figure S14. Simulated absorption spectra of FF24-Cl, FM24-Cl and MM24-Cl.
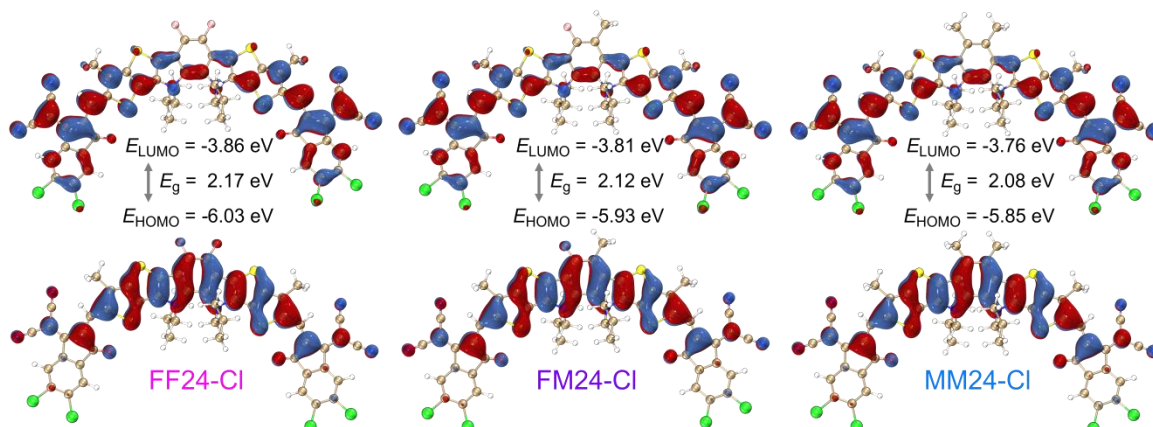
Figure S15. Calculated E_{HOMO} and E_{LUMO} of FF24-Cl, FM24-Cl and MM24-Cl.
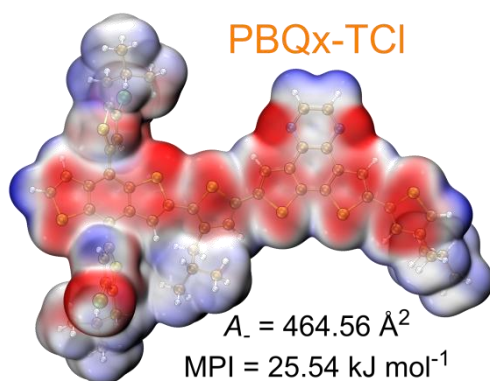
Figure S16. ESP diagram of a repeating unit of PBQX-TCl.
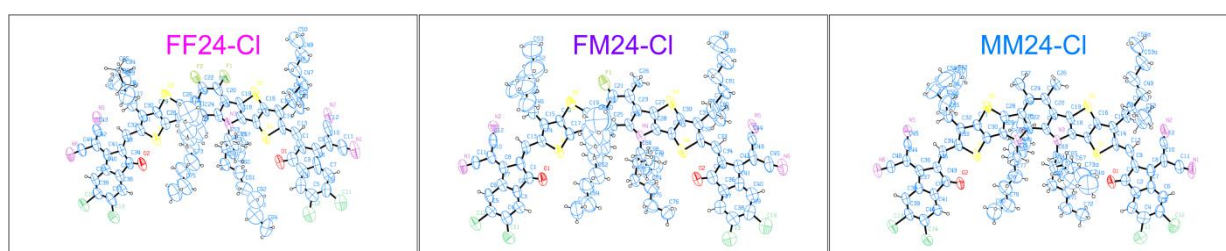
Figure S17. The Oak Ridge Thermal Ellipsoid Plot (ORTEP) view of FF24-Cl, FM24-Cl and MM24-Cl with atomic displacement parameters shown at the 30% probability level. Alerts justification: Numerous single crystals cultivated from various methods have been tested, but the data quality remains unsatisfactory, which is attributed to the disorder issues caused by the outer and inner alkyl chains, as well as the decomposition of single crystals during the testing process. Despite the extended exposure time, the single crystals did not exhibit discernible intensity at a resolution better than 0.90 Å, resulting in some alerts displayed in the CheckCIF report. In this work, we only focus on the molecular skeleton geometry and the single crystal packing pattern, without discussing the contributions of the outer and inner alky chains.

Figure S18. (a) Monomolecular geometry from the side view of FF24-Cl, FM24-Cl and MM24-Cl; (b) Single crystal packing diagrams from the side view of FF24-Cl, FM24-Cl and MM24-Cl (the diagrams are overlapped to clearly display the difference in π-π space).
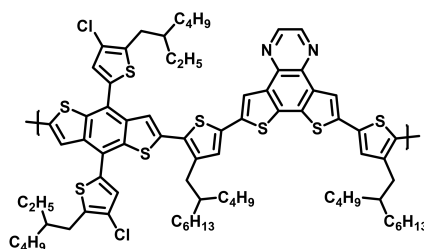
Figure S19. Chemical structure of PBQx-TCl.
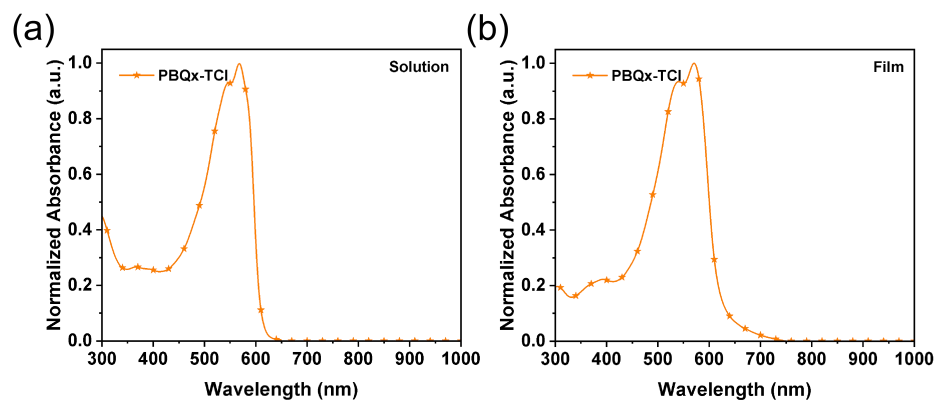
Figure S20. Normalized absorption spectra of PBQx-TCl (a) in *o*-xylene solution and (b) in thin film.
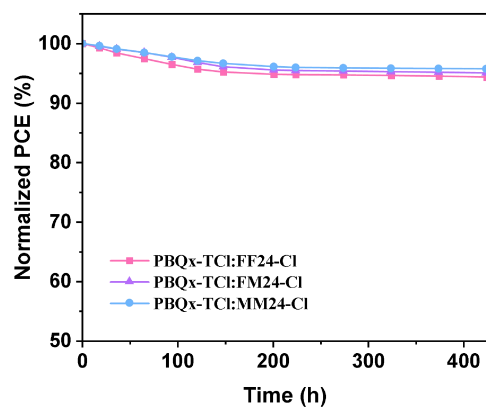
Figure S21. The durability of the OSC based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl under the irradiation of 100 mW cm^{-2}.
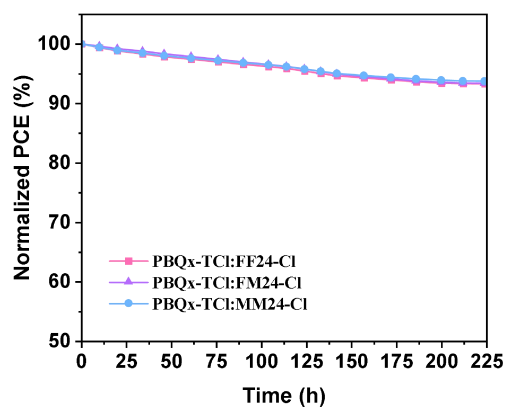
Figure S22. The durability of the OSC based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl under the heat of 65 °C.
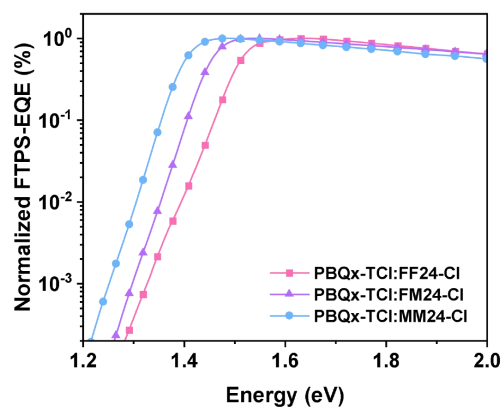
Figure S23. Normalized Fourier-transform photocurrent spectroscopy EQE (FTPS-EQE) spectra of the OSC based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl.
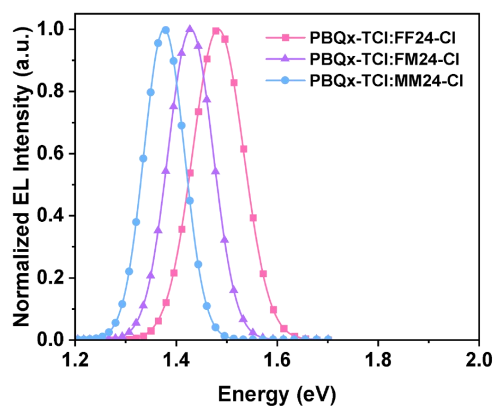
Figure S24. Normalized electroluminescence (EL) spectra of the OSC based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl.
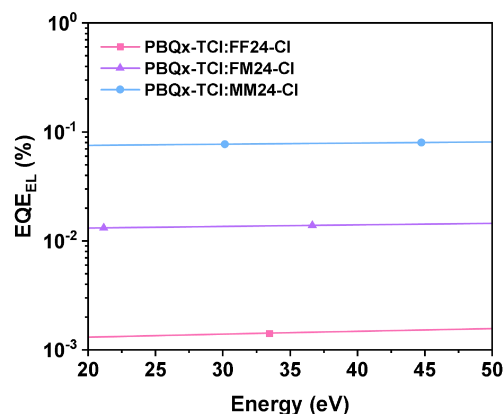
Figure S25. Electroluminescence quantum efficiency (EQE$_{EL}$) curve of the OSC based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl.
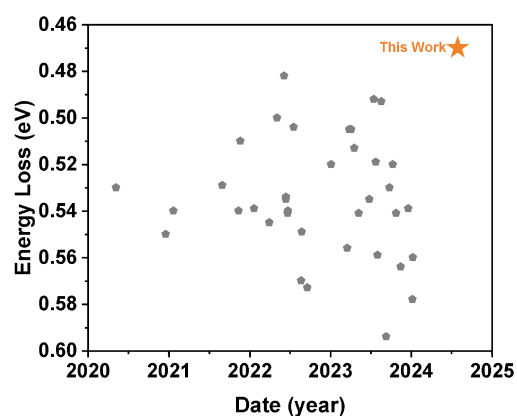
Figure S26. Statistical diagram of E_{loss} of OSCs with PCE exceeding 17%.
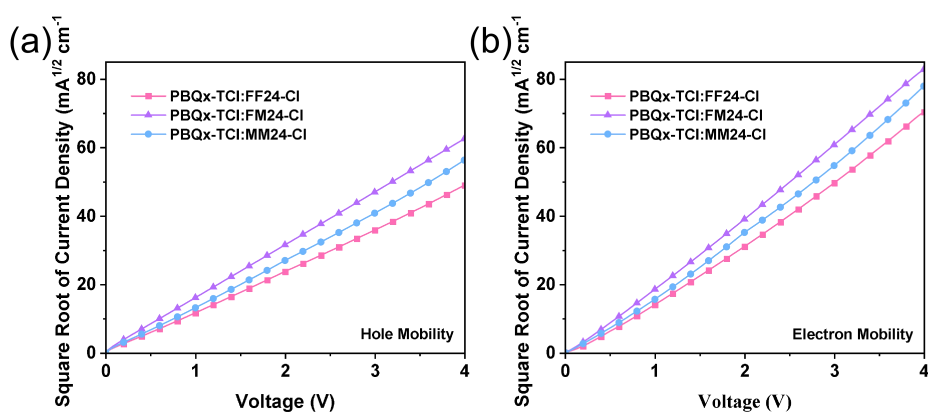
Figure S27. (a) Hole mobility and (b) Electron mobility of the OSC based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl.
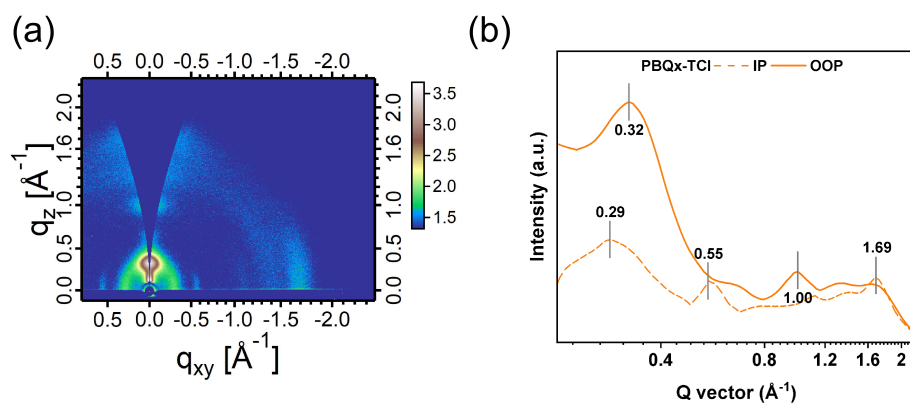
Figure S28. (a) Two-dimensional GIWAXS patterns and (b) the corresponding line-cuts in the IP (dashed lines) and OOP (solid lines) directions of neat film based on PBQx-TCl.

Supplementary Tables

Table S1 Thermal properties of FF24-Cl, FM24-Cl and MM24-Cl.

Acceptor	T_d [°C]	T_m [°C]	T_c [°C]
FF24-Cl	376	247	222
FM24-Cl	373	256	240
MM24-Cl	370	227	201

Table S2. Parameters for calculating the DED of FF24-Cl, FM24-Cl and MM24-Cl.

Acceptor	$E(\lambda_{0\to0}^{sol.})$ [eV]	$E(\lambda_{0\to0}^{film})$ [eV]	$E(\lambda_{0\to1}^{sol.})$ [eV]	E_w [eV]	S [a.u.]	SE_v [eV]	DED [a.u.]
FF24-Cl	1.789	1.585	1.949	0.407	0.427	0.068	5.95
FM24-Cl	1.729	1.529	1.887	0.401	0.395	0.062	6.43
MM24-Cl	1.675	1.485	1.834	0.381	0.373	0.059	6.45

Table S3. Quantitative calculation results of FF24-Cl, FM24-Cl and MM24-Cl.

Acceptor	λ_{max}^{cal} [nm]	E_g^{cal} [eV]	A_+ [Å^2]	MPI [kJ mol^{-1}]	S_r [a.u.]	D [Å]	E_{coul} [eV]	$\lambda_{S_0 \to S_1}$ [eV]	$\lambda_{S_1 \to S_0}$ [eV]
FF24-Cl	668	2.17	695.55	42.95	0.754	0.736	2.675	0.116	0.151
FM24-Cl	688	2.12	709.42	41.25	0.749	0.764	2.664	0.115	0.146
MM24-Cl	708	2.08	713.59	40.53	0.747	0.783	2.665	0.114	0.141

Table S4. Hole-electron analysis results of FF24-Cl.

Part	Holes [%]	Electrons [%]	Overlap [%]	ΔC [%]
1	14.80	24.45	19.02	9.65
2	27.14	20.31	23.47	-6.84
3	14.71	7.15	10.25	-7.56
4	27.14	20.31	23.47	-6.84
5	14.80	24.45	19.02	9.65

Table S5. Hole-electron analysis results of FM24-Cl.

Part	Holes [%]	Electrons [%]	Overlap [%]	ΔC [%]
1	14.42	23.85	18.55	9.43
2	26.82	19.92	23.12	-6.90
3	15.48	7.09	10.48	-8.39
4	27.19	20.98	23.88	-6.21
5	14.59	24.73	18.99	10.14

Table S6. Hole-electron analysis results of MM24-Cl.

Part	Holes [%]	Electrons [%]	Overlap [%]	ΔC [%]
1	14.25	24.13	18.54	9.88
2	26.86	20.65	23.54	-6.21
3	16.12	6.95	10.58	-9.17
4	26.86	20.65	23.54	-6.21
5	14.25	24.13	18.54	9.88

Table S7. Crystal data for FF24-Cl (CCDC 2333674), FM24-Cl (CCDC 2333675) and MM24-Cl (CCDC 2333676).

Acceptor	a [Å]	b [Å]	c [Å]	α [°]	β [°]	γ [°]	V [Å^3]	Z [a.u.]
FF24-Cl	24.2910(9)	23.9690(2)	33.8600(12)	90	107.264(4)	90	18825.9(19)	8
FM24-Cl	24.4208(12)	23.6514(17)	33.9562(15)	90	107.330(5)	90	18722.3(19)	8
MM24-Cl	24.5282(11)	23.8030(18)	34.0641(12)	90	107.624(4)	90	18954.7(18)	8

Table S8 Summary of the PCE of the OSCs with various device fabrication methods.

Fabrication Method	Publication Date	PCE [%]	Reference
without additive	2015-10-13	7.20	[4]
without additive	2015-10-29	8.52	[5]
without additive	2016-03-25	7.50	[6]
without additive	2016-09-07	10.16	[7]
without additive	2016-10-25	11.77	[8]
without additive	2016-10-26	10.88	[9]
without additive	2016-12-30	10.54	[10]
without additive	2017-04-10	8.26	[11]
without additive	2017-04-21	8.80	[12]
without additive	2017-05-31	10.46	[13]

without additive	2017-07-17	5.40	[14]
without additive	2017-08-04	3.49	[15]
without additive	2018-04-06	11.60	[16]
without additive	2018-05-16	13.24	[17]
without additive	2018-05-21	10.29	[18]
without additive	2018-06-21	12.20	[19]
without additive	2019-04-11	10.10	[20]
without additive	2019-05-06	10.14	[21]
without additive	2019-08-16	8.28	[22]
without additive	2019-12-10	6.24	[23]
without additive	2020-04-22	12.34	[24]
without additive	2020-08-03	7.66	[25]
without additive	2020-08-04	13.49	[26]
without additive	2020-09-29	12.63	[27]
without additive	2021-07-23	10.71	[28]
without additive	2021-11-02	9.87	[29]
without additive	2022-01-27	15.38	[30]
without additive	2022-07-20	16.03	[31]
without additive	2022-12-07	14.90	[32]
without additive	2023-03-25	16.90	[33]

without additive	2023-06-16	13.37	[34]
without additive	2023-06-26	17.54	[35]
without additive	2023-06-26	15.48	[36]
without additive	2023-07-24	17.13	[37]
without additive	2023-08-15	16.30	[38]
without additive	2023-08-27	15.40	[39]
without additive	2023-09-01	16.80	[40]
without additive	2023-09-05	16.20	[41]
without additive	2023-09-12	16.55	[42]
without additive	2023-09-29	16.80	[43]
without additive	2023-11-12	16.75	[44]
without additive	2023-11-15	16.33	[45]
without additive	2023-12-19	16.50	[46]
without additive	2024-01-02	14.40	[47]
without additive	2024-01-03	14.10	[48]
without additive	2024-01-09	16.10	[49]
without additive and thermal annealing	2015-01-07	7.52	[50]
without additive and thermal annealing	2015-04-20	6.71	[51]
without additive and thermal annealing	2016-11-09	9.95	[52]
without additive and thermal annealing	2017-03-16	8.99	[53]

without additive and thermal annealing	2017-03-21	5.37	[54]
without additive and thermal annealing	2017-05-03	8.59	[55]
without additive and thermal annealing	2017-05-09	8.19	[56]
without additive and thermal annealing	2017-07-17	9.13	[57]
without additive and thermal annealing	2017-07-21	8.90	[58]
without additive and thermal annealing	2017-07-24	9.60	[59]
without additive and thermal annealing	2017-07-27	8.12	[60]
without additive and thermal annealing	2017-08-17	6.36	[61]
without additive and thermal annealing	2017-08-25	9.03	[62]
without additive and thermal annealing	2017-08-31	6.05	[63]
without additive and thermal annealing	2017-09-11	10.50	[64]
without additive and thermal annealing	2017-09-28	10.12	[65]
without additive and thermal annealing	2017-11-07	6.26	[66]
without additive and thermal annealing	2017-11-20	7.25	[67]
without additive and thermal annealing	2017-12-06	12.80	[68]
without additive and thermal annealing	2017-12-10	11.00	[69]
without additive and thermal annealing	2017-12-13	11.90	[70]
without additive and thermal annealing	2018-01-17	10.45	[71]
without additive and thermal annealing	2018-02-05	5.30	[72]
without additive and thermal annealing	2018-02-12	9.60	[73]

without additive and thermal annealing	2018-02-21	10.41	[74]
without additive and thermal annealing	2018-02-22	7.23	[75]
without additive and thermal annealing	2018-03-23	10.55	[76]
without additive and thermal annealing	2018-06-21	11.64	[77]
without additive and thermal annealing	2018-06-22	11.14	[78]
without additive and thermal annealing	2019-01-21	7.72	[79]
without additive and thermal annealing	2019-03-06	5.02	[80]
without additive and thermal annealing	2019-06-01	9.09	[81]
without additive and thermal annealing	2021-04-08	10.40	[82]
without additive and thermal annealing	2022-01-27	11.68	[83]
without additive and thermal annealing	2022-03-26	15.20	[84]
without additive and thermal annealing	2022-07-13	15.41	[85]
without additive and thermal annealing	2022-12-13	15.99	[86]
without additive and thermal annealing	2023-05-24	16.50	[87]
without additive and thermal annealing	2023-07-09	17.20	[88]
without additive and thermal annealing	2023-10-10	17.03	[89]
without additive, thermal annealing and halogenated solvent	2016-02-03	2.31	[90]
without additive, thermal annealing and halogenated solvent	2017-01-23	11.90	[91]
without additive, thermal annealing and halogenated solvent	2017-12-06	12.80	[92]
without additive, thermal annealing and halogenated solvent	2018-02-20	9.55	[93]

without additive, thermal annealing and halogenated solvent	2019-11-25	7.99	[94]
without additive, thermal annealing and halogenated solvent	2022-12-09	15.00	[95]
without additive, thermal annealing and halogenated solvent	2023-10-31	16.50	[96]

Table S9 Photovoltaic performance parameters of the optimized OSCs with PBQx-TCl as the donor and BT24-Cl as the acceptor fabricated using non-halogenated *o*-xylene as the solvent without the treatment of additive and thermal annealing, under the irradiation of AM 1.5G, 100 mW cm^{-2}.

Device	V_{oc} [V]	J_{sc} [mA cm^{-2}]	J_{cal}[a] [mA cm^{-2}]	FF [%]	PCE[b] [%]
PBQx-TCl:BT24-Cl	0.877 (0.876±0.001)	24.08 (23.69±0.28)	23.38	72.87 (72.57±0.17)	15.39 (15.06±0.20)

a) The value was determined by integration of the EQE curve.

b) The average was calculated based on 20 individual devices.

Table S10 Photovoltaic performance parameters of these OSCs with the extra treatment of thermal annealing (100 °C, 10 min) under the irradiation of AM 1.5G, 100 mW cm^{-2}.

Device	V_{oc} [V]	J_{sc} [mA cm^{-2}]	J_{cal}[a] [mA cm^{-2}]	FF [%]	PCE[b] [%]
PBQx-TCl:FF24-Cl	0.884	23.29	22.58	74.89	15.42
	(0.884±0.001)	(23.00±0.13)		(74.73±0.12)	(15.20±0.09)
PBQx-TCl:FM24-Cl	0.904	25.76	25.00	78.77	18.34
	(0.904±0.001)	(25.43±0.23)		(78.65±0.17)	(18.08±0.17)
PBQx-TCl:MM24-Cl	0.911	25.20	24.49	75.93	17.43
	(0.910±0.001)	(24.86±0.16)		(75.73±0.12)	(17.13±0.12)

a) The values were determined by integration of the EQE curves.

b) The average were calculated based on 20 individual devices.

Table S11 Photovoltaic performance parameters of the optimized OSCs with PM6 or D18 as donor and FM24-Cl as acceptor fabricated using non-halogenated *o*-xylene as the solvent without the treatment of additive and thermal annealing, under the irradiation of AM 1.5G, 100 mW cm^{-2}.

Device	V_{oc} [V]	J_{sc} [mA cm^{-2}]	J_{cal}[a] [mA cm^{-2}]	FF [%]	PCE[b] [%]
PM6:FM24-Cl	0.915	24.28	23.54	72.23	16.04
	(0.913±0.001)	(23.95±0.17)		(72.01±0.39)	(15.75±0.16)
D18:FM24-Cl	0.925	24.07	23.33	76.49	17.03
	(0.924±0.002)	(23.68±0.15)		(76.41±0.34)	(16.71±0.16)

a) The values were determined by integration of the EQE curves.

b) The average were calculated based on 20 individual devices.

Table S12 Summary of E_{loss} of OSCs with PCE exceeding 17%.

Publication Date	E_{loss}	Reference
2020-05-07	0.530	[97]
2020-12-18	0.550	[98]
2021-01-22	0.540	[99]
2021-08-31	0.529	[100]
2021-11-13	0.540	[101]
2021-11-21	0.510	[102]
2022-01-22	0.539	[103]
2022-04-01	0.545	[104]
2022-05-06	0.500	[105]
2022-06-07	0.482	[106]
2022-06-15	0.535	[107]
2022-06-16	0.534	[108]
2022-06-23	0.541	[109]
2022-06-24	0.540	[110]
2022-07-20	0.504	[111]
2022-08-23	0.570	[112]
2022-08-26	0.549	[113]
2022-09-20	0.573	[114]

2023-01-06	0.520	[115]
2023-03-20	0.556	[116]
2023-03-29	0.505	[117]
2023-04-07	0.505	[118]
2023-04-21	0.513	[119]
2023-05-11	0.541	[120]
2023-06-28	0.535	[121]
2023-07-18	0.492	[122]
2023-07-27	0.519	[123]
2023-08-04	0.559	[124]
2023-08-21	0.493	[125]
2023-09-12	0.594	[126]
2023-09-27	0.530	[127]
2023-10-12	0.520	[128]
2023-10-27	0.541	[129]
2023-11-17	0.564	[130]
2023-12-21	0.539	[131]
2024-01-09	0.578	[132]
2024-01-11	0.560	[133]

Table S13. Carrier mobility of blend film based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl.

Film	μ_h [10^{-4} cm^2 V^{-1} s^{-1}]	μ_e [10^{-4} cm^2 V^{-1} s^{-1}]	μ_h/μ_e [a.u.]
PBQx-TCl:FF24-Cl	2.51	3.64	0.69
PBQx-TCl:FM24-Cl	4.09	5.07	0.81
PBQx-TCl:MM24-Cl	3.30	4.38	0.75

Table S14. GIWAXS experiment results of neat film based on FF24-Cl/FM24-Cl/MM24-Cl.

Film	IP direction (100)				OOP direction (010)			
	Peak [Å^{-1}]	Lattice Constant [Å]	FWHM [Å^{-1}]	CCL [Å]	Peak [Å^{-1}]	π-π Stacking Distances [Å]	FWHM [Å^{-1}]	CCL [Å]
FF24-Cl	0.43	14.63	0.11	52.36	1.78	3.53	0.39	14.53
FM24-Cl	0.43	14.63	0.11	51.38	1.77	3.55	0.38	15.08
MM24-Cl	0.43	14.63	0.11	50.57	1.76	3.58	0.37	15.38

Table S15. GIWAXS experiment results of blend film based on PBQx-TCl:FF24-Cl/FM24-Cl/MM24-Cl.

Film	IP direction (100)				OOP direction (010)			
	Peak [Å^{-1}]	Lattice Constant [Å]	FWHM [Å^{-1}]	CCL [Å]	Peak [Å^{-1}]	π-π Stacking Distances [Å]	FWHM [Å^{-1}]	CCL [Å]
PBQx-TCl: FF24-Cl	0.33	19.11	0.09	62.85	1.78	3.53	0.39	14.48
	0.44	14.12	0.08	68.15				
PBQx-TCl: FM24-Cl	0.33	19.11	0.09	61.67	1.76	3.57	0.38	15.02
	0.44	14.12	0.08	66.88				
PBQx-TCl: MM24-Cl	0.33	19.11	0.09	60.10	1.74	3.62	0.37	15.31

Table S16. CA test results of FF24-Cl, FM24-Cl and MM24-Cl.

Film	H_2O [°]	CH_2I_2 [°]	SE [mN m^{-1}]	χ [a.u.]
FF24-Cl	92.9	43.1	38.14	1.09 K
FM24-Cl	96.0	47.3	36.00	0.76 K
MM24-Cl	98.8	49.2	35.19	0.64 K
PBQx-TCl	108.9	65.3	26.32	\

References

[1] F. Neese, *WIREs Comput. Mol. Sci.*, **2018**, *8*, e1327.

[2] T. Lu and F. Chen, *J. Comput. Chem.*, **2012**, *33*, 580-592.

[3] W. Humphrey, A. Dalke and K. Schulten, *J. Mol. Graph.*, **1996**, *14*, 33-38.

[4] H. Lin, S. Chen, Z. Li, J. Lai, G. Yang, T. McAfee, K. Jiang, Y. Li, Y. Liu, H. Hu, J. Zhao, W. Ma, H. Ade and H. Yan, *Adv. Mater.*, **2015**, *27*, 7299-7304.

[5] Y. Hwang, H. Li, B. Courtright, S. Subramaniyan and S. Jenekhe, *Adv. Mater.*, **2016**, *28*, 124-131.

[6] Y. Lin, F. Zhao, Q. He, L. Huo, Y. Wu, T. Parker, W. Ma, Y. Sun, C. Wang, D. Zhu, A. Heeger, S. Marder and X. Zhan, *J. Am. Chem. Soc.*, **2016**, *138*, 4955-4961.

[7] Y. Qin, M. Uddin, Y. Chen, B. Jang, K. Zhao, Z. Zheng, R. Yu, T. Shin, H. Woo and J. Hou, *Adv. Mater.*, **2016**, *28*, 9416-9422.

[8] Y. Yang, Z. Zhang, H. Bin, S. Chen, L. Gao, L. Xue, C. Yang and Y. Li, *J. Am. Chem. Soc.*, **2016**, *138*, 15011-15018.

[9] Z. Li, K. Jiang, G. Yang, J. Lai, T. Ma, J. Zhao, W. Ma and H. Yan, *Nat. Commun.*, **2016**, *7*, 13094.

[10] K. Zhang, X. Liu, B. Xu, Y. Cui, M. Sun and J. Hou, *Chin. J. Polym. Sci.*, **2017**, *35*, 219-229.

[11] K. Kranthiraja, K. Gunasekar, H. Kim, A. Cho, N. Park, S. Kim, B. Kim, R. Nishikubo, A. Saeki, M. Song and S. Jin, *Adv. Mater.*, **2017**, *29*, 1700183.

[12] J. Zhang, K. Jiang, G. Yang, T. Ma, J. Liu, Z. Li, J. Lai, W. Ma and H. Yan, *Adv. Energy Mater.*, **2017**, *7*, 1602119.

[13] Y. Li, L. Zhong, B. Gautam, H. Bin, J. Lin, F. Wu, Z. Zhang, Z. Jiang, Z. Zhang, K. Gundogdu, Y. Li and L. Liao, *Energy Environ. Sci.*, **2017**, *10*, 1610-1620.

[14] L. Ye, Y. Xiong, S. Li, M. Ghasemi, N. Balar, J. Turner, A. Gadisa, J. Hou, B. O'Connor and H. Ade, *Adv. Funct. Mater.*, **2017**, *27*, 1702016.

[15] R. Pankow, N. Gobalasingham, J. Munteanu and B. Thompson, *J. Polym. Sci. Pol.*, **2017**, *55*, 3370-3380.

[16] W. Su, Q. Fan, X. Guo, J. Chen, Y. Wang, X. Wang, P. Dai, C. Ye, X. Bao, W. Ma, M. Zhang and Y. Li, *J. Mater. Chem. A*, **2018**, *6*, 7988-7996.

[17] W. Liu, J. Zhang, Z. Zhou, D. Zhang, Y. Zhang, S. Xu and X. Zhu, *Adv. Mater.*, **2018**, *30*,

1800403.

[18] H. Bin, J. Yao, Y. Yang, I. Angunawela, C. Sun, L. Gao, L. Ye, B. Qiu, L. Xue, C. Zhu, C. Yang, Z. Zhang, H. Ade and Y. Li, *Adv. Mater.*, **2018**, *30*, 1706361.

[19] Z. Luo, C. Sun, S. Chen, Z. Zhang, K. Wu, B. Qiu, C. Yang, Y. Li and C. Yang, *Adv. Energy Mater.*, **2018**, *8*, 1800856.

[20] N. Kolhe, D. Tran, H. Lee, D. Kuzuhara, N. Yoshimoto, T. Koganezawa and S. Jenekhe, *ACS Energy Lett.*, **2019**, *4*, 1162-1170.

[21] Y. Wang, B. Liu, C. Koh, X. Zhou, H. Sun, J. Yu, K. Yang, H. Wang, Q. Liao, H. Woo and X. Guo, *Adv. Energy Mater.*, **2019**, *9*, 1803976.

[22] H. Sun, B. Liu, C. Koh, Y. Zhang, J. Chen, Y. Wang, P. Chen, B. Tu, M. Su, H. Wang, Y. Tang, Y. Shi, H. Woo and X. Guo, *Adv. Funct. Mater.*, **2019**, *29*, 1903970.

[23] Y. Yin, Z. Zheng, M. Liu, S. Gao, F. Guo, G. Mola, J. Wang, L. Zhao and Y. Zhang, *Dyes Pigment.*, **2020**, *175*, 108119.

[24] Y. Zhang, Y. Shao, Z. Wei, L. Zhang, Y. Hu, L. Chen, S. Chen, Z. Yuan and Y. Chen, *ACS Appl. Mater. Interfaces*, **2020**, *12*, 20741-20749.

[25] Y. Yin, W. Zhang, Z. Zheng, Z. Ge, Y. Liu, F. Guo, S. Gao, L. Zhao and Y. Zhang, *J. Mater. Chem. C*, **2020**, *8*, 12516-12526.

[26] T. Chen, J. Yu, Y. Lin, S. Peng, S. Wu, Y. Su, V. Karapala, L. Hong, H. Yao, J. Hou and C. Hsu, *Sol. RRL*, **2020**, *4*, 2000357.

[27] F. Liu, D. Wang, J. Li, C. Xiao, Y. Wu, W. Li and G. Fu, *Chin. J. Polym. Sci.*, **2021**, *39*, 43-50.

[28] Y. Xin, J. Liang, Y. Zhong, T. Liang, C. Zhang and X. He, *Dyes Pigment.*, **2021**, *195*, 109661.

[29] Z. Zheng, E. He, J. Wang, Z. Qin, T. Niu, F. Guo, S. Gao, Z. Ma, L. Zhao, X. Lu, Q. Xue, Y. Cao, G. Mola and Y. Zhang, *J. Mater. Chem. A*, **2021**, *9*, 26105-26112.

[30] X. Wang, C. Feng, P. Liu, Z. He and Y. Cao, *Small*, **2022**, *18*, 2107106.

[31] C. Li, X. Gu, Z. Chen, X. Han, N. Yu, Y. Wei, J. Gao, H. Chen, M. Zhang, A. Wang, J. Zhang, Z. Wei, Q. Peng, Z. Tang, X. Hao, X. Zhang and H. Huang, *J. Am. Chem. Soc.*, **2022**, *144*, 14731-14739.

[32] D. Hu, H. Tang, S. Karuthedath, Q. Chen, S. Chen, J. Khan, H. Liu, Q. Yang, J. Gorenflot, C. Petoukhoff, T. Duan, X. Lu, F. Laquai and S. Lu, *Adv. Funct. Mater.*, **2023**, *33*, 2211873.

[33] 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**, *35*, 2301583.

[34] J. Panidi, E. Mazzolini, F. Eisner, Y. Fu, F. Furlan, Z. Qiao, M. Rimmele, Z. Li, X. Lu, J. Nelson, J. Durrant, M. Heeney and N. Gasparini, *ACS Energy Lett.*, **2023**, *8*, 3038-3047.

[35] L. Zhong, Z. Sun, S. Lee, S. Jeong, S. Jung, Y. Cho, J. Park, J. Park, S. Yoon and C. Yang, *Adv. Funct. Mater.*, **2023**, *33*, 2305450.

[36] Y. Su, Z. Ding, R. Zhang, W. Tang, W. Huang, Z. Wang, K. Zhao, X. Wang, S. Liu and Y. Li, *Sci. China Chem.*, **2023**, *66*, 2380-2388.

[37] X. Cao, J. Guo, Z. Li, X. Bi, H. Liang, Z. Xiao, Y. Guo, X. Jia, Z. Xu, K. Ma, Z. Yao, B. Kan, X. Wan, C. Li and Y. Chen, *ACS Energy Lett.*, **2023**, *8*, 3494-3503.

[38] R. Yu, R. Shi, Z. He, T. Zhang, S. Li, Q. Lv, S. Sha, C. Yang, J. Hou and Z. Tan, *Angew. Chem. Int. Edit.*, **2023**, *62*, e202308367.

[39] 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.

[40] M. Zhou, C. Liao, Y. Duan, X. Xu, L. Yu, R. Li and Q. Peng, *Adv. Mater.*, **2023**, *35*, 2208279.

[41] J. Zhao, S. Chung, H. Li, Z. Zhao, C. Zhu, J. Yin, K. Cho and Z. Kan, *Adv. Funct. Mater.*, **2023**, *33*, 2307355.

[42] W. Zhang, Y. Wu, R. Ma, H. Fan, X. Li, H. Yang, C. Cui and Y. Li, *Angew. Chem. Int. Edit.*, **2023**, *62*, e202309713.

[43] L. Cao, J. Zhang, X. Du, X. Li, Z. He, H. Lin, C. Zheng, G. Yang and S. Tao, *Sol. RRL*, **2023**, *7*, 2300665.

[44] Y. Ran, C. Liang, Z. Xu, W. Jing, X. Xu, Y. Duan, R. Li, L. Yu and Q. Peng, *Adv. Funct. Mater.*, **2024**, *34*, 2311512.

[45] M. Dong, S. Chen, L. Hong, J. Jing, Y. Bai, Y. Liang, C. Zhu, T. Shi, W. Zhong, L. Ying, K. Zhang and F. Huang, *Nano Energy*, **2024**, *119*, 109097.

[46] X. Wu, Y. Li, Y. Wang, M. Zhang, Q. Zhu, X. Huang, B. Li, Y. Xue, J. Qing and W. Cai, *Sol. RRL*, **2024**, *8*, 2300812.

[47] B. Du, M. Ma, P. Zhang, S. Wu, H. Bin and Y. Li, *ACS Appl. Mater. Interfaces*, **2024**, *16*, 2564-2572.

[48] W. Feng, T. Chen, Y. Li, T. Duan, X. Jiang, C. Zhong, Y. Zhang, J. Yu, G. Lu, X. Wan, B. Kan and Y. Chen, *Angew. Chem. Int. Edit.*, **2024**, *63*, e202316698.

[49] W. Liang, L. Chen, Z. Wang, Z. Peng, L. Zhu, C. Kwok, H. Yu, W. Xiong, T. Li, Z. Zhang, Y. Wang, Y. Liao, G. Zhang, H. Hu and Y. Chen, *Adv. Energy Mater.*, **2024**, *14*, 2303661.

[50] Y. Lin, J. Wang, Z. Zhang, H. Bai, Y. Li, D. Zhu and X. Zhan, *Adv. Mater.*, **2015**, *27*, 1170-1174.

[51] J. Jung, J. Jo, C. Chueh, F. Liu, W. Jo, T. Russell and A. Jen, *Adv. Mater.*, **2015**, *27*, 3310-3317.

[52] D. Baran, T. Kirchartz, S. Wheeler, S. Dimitrov, M. Abdelsamie, J. Gorman, R. Ashraf, S. Holliday, A. Wadsworth, N. Gasparini, P. Kaienburg, H. Yan, A. Amassian, C. Brabec, J. Durrant and I. McCulloch, *Energy Environ. Sci.*, **2016**, *9*, 3783-3793.

[53] B. Kan, H. Feng, X. Wan, F. Liu, X. Ke, Y. Wang, Y. Wang, H. Zhang, C. Li, J. Hou and Y. Chen, *J. Am. Chem. Soc.*, **2017**, *139*, 4929-4934.

[54] H. Bin, Y. Yang, Z. Zhang, L. Ye, M. Ghasemi, S. Chen, Y. Zhang, C. Zhang, C. Sun, L. Xue, C. Yang, H. Ade and Y. Li, *J. Am. Chem. Soc.*, **2017**, *139*, 5085-5094.

[55] Y. Guo, Y. Li, O. Awartani, H. Han, J. Zhao, H. Ade, H. Yan and D. Zhao, *Advanced Materials*, **2017**, *29*, 1700309.

[56] Z. Zhang, L. Feng, S. Xu, J. Yuan, Z. Zhang, H. Peng, Y. Li and Y. Zou, *J. Mater. Chem. A*, **2017**, *5*, 11286-11293.

[57] J. Wang, W. Wang, X. Wang, Y. Wu, Q. Zhang, C. Yan, W. Ma, W. You and X. Zhan, *Adv. Mater.*, **2017**, *29*, 1702125.

[58] Y. Firdaus, L. Maffei, F. Cruciani, M. Müller, S. Liu, S. Lopatin, N. Wehbe, G. Ndjawa, A. Amassian, F. Laquai and P. Beaujuge, *Adv. Energy Mater.*, **2017**, *7*, 1700834.

[59] B. Guo, W. Li, X. Guo, X. Meng, W. Ma, M. Zhang and Y. Li, *Adv. Mater.*, **2017**, *29*, 1702291.

[60] Y. Yi, H. Feng, M. Chang, H. Zhang, X. Wan, C. Li and Y. Chen, *J. Mater. Chem. A*, **2017**, *5*, 17204-17210.

[61] Z. Zhang, L. Feng, S. Xu, Y. Liu, H. Peng, Z. Zhang, Y. Li and Y. Zou, *Adv. Sci.*, **2017**, *4*, 1700152.

[62] Y. Ma, M. Zhang, Y. Yan, J. Xin, T. Wang, W. Ma, C. Tang and Q. Zheng, *Chem. Mat.*, **2017**, *29*, 7942-7952.

[63] L. Xue, Y. Yang, J. Xu, C. Zhang, H. Bin, Z. Zhang, B. Qiu, X. Li, C. Sun, L. Gao, J. Yao, X. Chen, Y. Yang, M. Xiao and Y. Li, *Adv. Mater.*, **2017**, *29*, 1703344.

[64] Y. Li, L. Zhong, J. Lin, F. Wu, H. Bin, Z. Zhang, L. Xu, Z. Jiang, Z. Zhang, F. Liu, T. Russell, Y. Li, L. Liao and S. Forrest, *Sol. RRL*, **2017**, *1*, 1700107.

[65] S. Feng, C. Zhang, Y. Liu, Z. Bi, Z. Zhang, X. Xu, W. Ma and Z. Bo, *Adv. Mater.*, **2017**, *29*, 1703527.

[66] J. Hamonnet, M. Nakano, K. Nakano, H. Sugino, K. Takimiya and K. Tajima, *Chem. Mat.*, **2017**,

29, 9618-9622.

[67] H. Wu, H. Fan, S. Xu, C. Zhang, S. Chen, C. Yang, D. Chen, F. Liu and X. Zhu, *Sol. RRL*, **2017**, *1*, 1700165.

[68] X. Xu, T. Yu, Z. Bi, W. Ma, Y. Li and Q. Peng, *Adv. Mater.*, **2018**, *30*, 1703973.

[69] B. Jia, S. Dai, Z. Ke, C. Yan, W. Ma and X. Zhan, *Chem. Mat.*, **2018**, *30*, 239-245.

[70] Q. Fan, Y. Wang, M. Zhang, B. Wu, X. Guo, Y. Jiang, W. Li, B. Guo, C. Ye, W. Su, J. Fang, X. Ou, F. Liu, Z. Wei, T. Sum, T. Russell and Y. Li, *Adv. Mater.*, **2018**, *30*, 1704546.

[71] Z. Zhang, J. Yu, X. Yin, Z. Hu, Y. Jiang, J. Sun, J. Zhou, F. Zhang, T. Russell, F. Liu and W. Tang, *Adv. Funct. Mater.*, **2018**, *28*, 1705095.

[72] Z. Liao, Y. Wang, Y. An, Y. Tan, X. Meng, F. Wu, L. Chen and Y. Chen, *Macromol. Rapid Commun.*, **2018**, *39*, 1700706.

[73] Z. Zhang and X. Zhu, *J. Mater. Chem. A*, **2018**, *6*, 4266-4270.

[74] C. Sun, F. Pan, H. Bin, J. Zhang, L. Xue, B. Qiu, Z. Wei, Z. Zhang and Y. Li, *Nat. Commun.*, **2018**, *9*, 743.

[75] D. Chen, J. Yao, L. Chen, J. Yin, R. Lv, B. Huang, S. Liu, Z. Zhang, C. Yang, Y. Chen and Y. Li, *Angew. Chem. Int. Edit.*, **2018**, *57*, 4580-4584.

[76] Z. Li, D. Yang, T. Zhang, J. Zhang, X. Zhao and X. Yang, *Small*, **2018**, *14*, 1704491.

[77] X. Li, J. Yao, I. Angunawela, C. Sun, L. Xue, A. Liebman-Pelaez, C. Zhu, C. Yang, Z. Zhang, H. Ade and Y. Li, *Adv. Energy Mater.*, **2018**, *8*, 1800815.

[78] X. Shi, X. Liao, K. Gao, L. Zuo, J. Chen, J. Zhao, F. Liu, Y. Chen and A. Jen, *Adv. Funct. Mater.*, **2018**, *28*, 1802324.

[79] L. Zhou, T. Lau, H. Peng, B. Qiu, L. Jiang, X. Lu, G. Zhang, J. Yuan, Y. Li and Y. Zou, *Sol. RRL*, **2019**, *3*, 1800340.

[80] T. Lee, Y. Eom, C. Song, I. Jung, D. Kim, S. Lee, W. Shin and E. Lim, *Adv. Energy Mater.*, **2019**, *9*, 1804021.

[81] M. Wu, L. Shi, Y. Hu, L. Chen, T. Hu, Y. Zhang, Z. Yuan and Y. Chen, *Chin. Chem. Lett.*, **2019**, *30*, 1161-1167.

[82] D. Yang, K. Yu, J. Xu, J. Zhang, J. Zhang, J. Gao, W. Song, D. Li, Z. Chen and Z. Ge, *J. Mater. Chem. A*, **2021**, *9*, 10427-10436.

[83] K. Hung, Y. Lin, Y. Xue, H. Yang, Y. Lai, J. Chang, C. Su, A. Su, C. Hsu, U. Jeng and Y. Cheng, *Adv. Energy Mater.*, **2022**, *12*, 2103702.

[84] L. Wang, T. Wang, J. Oh, Z. Yuan, C. Yang, Y. Hu, X. Zhao and Y. Chen, *Chem. Eng. J.*, **2022**, *442*, 136068.

[85] D. Zhang, Y. Li, M. Li, W. Zhong, T. Heumüller, N. Li, L. Ying, C. Brabec and F. Huang, *Adv. Funct. Mater.*, **2022**, *32*, 2205338.

[86] G. Zhang, D. Hu, H. Tang, H. Song, S. Duan, Z. Kan and S. Lu, *Sol. RRL*, **2023**, *7*, 2200994.

[87] L. Kong, Z. Zhang, N. Zhao, Z. Cai, J. Zhang, M. Luo, X. Wang, M. Chen, W. Zhang, L. Zhang, Z. Wei and J. Chen, *Adv. Energy Mater.*, **2023**, *13*, 2300763.

[88] C. Zhu, S. Chung, J. Zhao, Y. Sun, B. Zhao, Z. Zhao, S. Kim, K. Cho and Z. Kan, *Adv. Sci.*, **2023**, *10*, 2303150.

[89] H. Chen, X. Cao, P. Wang, F. Huang, Y. Zhang, H. Liang, X. Bi, T. He, W. Feng, Y. Guo, Z. Ma, G. Long, Z. Yao, B. Kan, C. Li, X. Wan and Y. Chen, *J. Mater. Chem. A*, **2023**, *11*, 25368-25376.

[90] K. Gao, L. Xiao, Y. Kan, L. Li, Y. Yan, H. Huang, J. Peng, Y. Cao and X. Peng, *Chinese Journal of Chemistry*, **2016**, *34*, 353-358.

[91] D. Liu, B. Yang, B. Jang, B. Xu, S. Zhang, C. He, H. Woo and J. Hou, *Energy Environ. Sci.*, **2017**, *10*, 546-551.

[92] X. Xu, T. Yu, Z. Bi, W. Ma, Y. Li and Q. Peng, *Adv. Mater.*, **2018**, *30*, 1703973.

[93] Y. An, X. Liao, L. Chen, J. Yin, Q. Ai, Q. Xie, B. Huang, F. Liu, A. Jen and Y. Chen, *Adv. Funct. Mater.*, **2018**, *28*, 1706517.

[94] M. Jeong, B. Lee, Y. Cho, J. Oh, S. Lee, J. Lee and C. Yang, *Small Methods*, **2020**, *4*, 1900696.

[95] N. Su, J. Chen, M. Peng, G. Li, R. Pankow, D. Zheng, J. Ding, A. Facchetti and T. Marks, *J. Energy Chem.*, **2023**, *79*, 321-329.

[96] S. Luo, C. Li, J. Zhang, X. Zou, H. Zhao, K. Ding, H. Huang, J. Song, J. Yi, H. Yu, K. Wong, G. Zhang, H. Ade, W. Ma, H. Hu, Y. Sun and H. Yan, *Nat. Commun.*, **2023**, *14*, 6964.

[97] Z. Luo, R. Ma, T. Liu, J. Yu, Y. Xiao, R. Sun, G. Xie, J. Yuan, Y. Chen, K. Chen, G. Chai, H. Sun, J. Min, J. Zhang, Y. Zou, C. Yang, X. Lu, F. Gao and H. Yan, *Joule*, **2020**, *4*, 1236-1247.

[98] W. Gao, H. Fu, Y. Li, F. Lin, R. Sun, Z. Wu, X. Wu, C. Zhong, J. Min, J. Luo, H. Woo, Z. Zhu and A. Jen, *Adv. Energy Mater.*, **2021**, *11*, 2003177.

[99] Y. Shi, J. Pan, J. Yu, J. Zhang, F. Gao, K. Lu and Z. Wei, *Sol. RRL*, **2021**, *5*, 2100008.

[100] X. Zhang, L. Qin, X. Liu, C. Zhang, J. Yu, Z. Xiao, N. Zheng, B. Wang, Y. Wei, Z. Xie, Y. Wu, Z. Wei, K. Wang, F. Gao, L. Ding and H. Huang, *Sol. RRL*, **2021**, *5*, 2100522.

[101] H. Wang, H. Lu, Y. Chen, G. Ran, A. Zhang, D. Li, N. Yu, Z. Zhang, Y. Liu, X. Xu, W. Zhang, Q. Bao, Z. Tang and Z. Bo, *Adv. Mater.*, **2022**, *34*, 2105483.

[102] Y. Pan, X. Zheng, J. Guo, Z. Chen, S. Li, C. He, S. Ye, X. Xia, S. Wang, X. Lu, H. Zhu, J. Min, L. Zuo, M. Shi and H. Chen, *Adv. Funct. Mater.*, **2022**, *32*, 2108614.

[103] C. He, Z. Bi, Z. Chen, J. Guo, X. Xia, X. Lu, J. Min, H. Zhu, W. Ma, L. Zuo and H. Chen, *Adv. Funct. Mater.*, **2022**, *32*, 2112511.

[104] B. Zheng, Y. Yue, J. Ni, R. Sun, J. Min, J. Wang, L. Jiang and L. Huo, *Sci. China Chem.*, **2022**, *65*, 964-972.

[105] H. Chen, Y. Zou, H. Liang, T. He, X. Xu, Y. Zhang, Z. Ma, J. Wang, M. Zhang, Q. Li, C. Li, G. Long, X. Wan, Z. Yao and Y. Chen, *Sci. China Chem.*, **2022**, *65*, 1362-1373.

[106] Y. Shi, Y. Chang, K. Lu, Z. Chen, J. Zhang, Y. Yan, D. Qiu, Y. Liu, M. Adil, W. Ma, X. Hao, L. Zhu and Z. Wei, *Nat. Commun.*, **2022**, *13*, 3256.

[107] 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.

[108] L. Zhang, Z. Zhang, D. Deng, H. Zhou, J. Zhang and Z. Wei, *Adv. Sci.*, **2022**, *9*, 2202513.

[109] 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.

[110] X. Yan, J. Wu, J. Lv, L. Zhang, R. Zhang, X. Guo and M. Zhang, *J. Mater. Chem. A*, **2022**, *10*, 15605-15613.

[111] J. Wang, H. Chen, X. Xu, Z. Ma, Z. Zhang, C. Li, Y. Yang, J. Wang, Y. Zhao, M. Zhang, X. Wan, Y. Lu and Y. Chen, *J. Mater. Chem. A*, **2022**, *10*, 16714-16721.

[112] J. Gao, N. Yu, Z. Chen, Y. Wei, C. Li, T. Liu, X. Gu, J. Zhang, Z. Wei, Z. Tang, X. Hao, F. Zhang, X. Zhang and H. Huang, *Adv. Sci.*, **2022**, *9*, 2203606.

[113] H. Lu, K. Chen, R. Bobba, J. Shi, M. Li, Y. Wang, J. Xue, P. Xue, X. Zheng, K. Thorn, I. Wagner, C. Lin, Y. Song, W. Ma, Z. Tang, Q. Meng, Q. Qiao, J. Hodgkiss and X. Zhan, *Adv. Mater.*, **2022**, *34*, 2205926.

[114] J. Zhang, S. Luo, H. Zhao, X. Xu, X. Zou, A. Shang, J. Liang, F. Bai, Y. Chen, K. Wong, Z. Ma, W. Ma, H. Hu, Y. Chen and H. Yan, *Angew. Chem. Int. Edit.*, **2022**, *61*, e202206930.

[115] Q. Fan, R. Ma, Z. Bi, X. Liao, B. Wu, S. Zhang, W. Su, J. Fang, C. Zhao, C. Yan, K. Chen, Y. Li, C. Gao, G. Li and W. Ma, *Adv. Funct. Mater.*, **2023**, *33*, 2211385.

[116] R. Xu, Y. Jiang, F. Liu, W. Su, W. Liu, S. Xu, H. Fan, C. Jiang, Q. Zong, W. Zhang and X. Zhu, *Chem. Eng. J.*, **2023**, *464*, 142507.

[117] Y. Gao, X. Yang, W. Wang, R. Sun, J. Cui, Y. Fu, K. Li, M. Zhang, C. Liu, H. Zhu, X. Lu and J. Min, *Adv. Mater.*, **2023**, *35*, 2300531.

[118] 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.

[119] Z. Li, C. Jiang, X. Chen, G. Song, X. Wan, B. Kan, T. Duan, E. Knyazeva, O. Rakitin and Y. Chen, *J. Mater. Chem. C*, **2023**, *11*, 6920-6927.

[120] Z. Chen, H. Yao, J. Wang, J. Zhang, T. Zhang, Z. Li, J. Qiao, S. Xiu, X. Hao and J. Hou, *Energy Environ. Sci.*, **2023**, *16*, 2637-2645.

[121] 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.

[122] Q. Fan, R. Ma, J. Yang, J. Gao, H. Bai, W. Su, Z. Liang, Y. Wu, L. Tang, Y. Li, Q. Wu, K. Wang, L. Yan, R. Zhang, F. Gao, G. Li and W. Ma, *Angew. Chem. Int. Edit.*, **2023**, *62*, e202308307.

[123] X. Si, Y. Huang, W. Shi, R. Wang, K. Ma, Y. Zhang, S. Wu, Z. Yao, C. Li, X. Wan and Y. Chen, *Adv. Funct. Mater.*, **2023**, *33*, 2306471.

[124] Y. Guo, Z. Chen, J. Ge, J. Zhu, J. Zhang, Y. Meng, Q. Ye, S. Wang, F. Chen, W. Ma and Z. Ge, *Adv. Funct. Mater.*, **2023**, *33*, 2305611.

[125] 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.

[126] L. Liu, Y. Yan, S. Zhao, T. Wang, W. Zhang, J. Zhang, X. Hao, Y. Zhang, X. Zhang and Z. Wei, *Small*, **2024**, *20*, 2305638.

[127] Y. Zhang, J. Deng, S. You, X. Huang, J. Liu, Y. Cheng, B. Huang, X. Chen, Z. Sun, C. Yang, Q. Cao, F. Wu and L. Chen, *Adv. Funct. Mater.*, **2024**, *34*, 2308151.

[128] C. Yang, Q. An, M. Jiang, X. Ma, A. Mahmood, H. Zhang, X. Zhao, H. Zhi, M. Jee, H. Woo, X. Liao, D. Deng, Z. Wei and J. Wang, *Angew. Chem. Int. Edit.*, **2023**, *62*, e202313016.

[129] W. Liu, X. Xu, S. He, R. Sun, Q. Chen, J. Min, Z. Zhang, J. Yuan, Y. Li and Y. Zou, *Macromolecules*, **2023**, *56*, 8623-8631.

[130] D. Sun, Z. Chen, J. Zhang, W. Song, J. Shi, J. Zhu, Y. Meng, F. Jin, S. Yang and Z. Ge, *Sci. China Chem.*, **2024**, *67*, 963-972.

[131] M. Xiao, Y. Meng, L. Tang, P. Li, L. Tang, W. Zhang, B. Hu, F. Yi, T. Jia, J. Cao, C. Xu, G. Lu, X. Hao, W. Ma and Q. Fan, *Adv. Funct. Mater.*, **2024**, *34*, 2311216.

[132] W. Liang, L. Chen, Z. Wang, Z. Peng, L. Zhu, C. Kwok, H. Yu, W. Xiong, T. Li, Z. Zhang, Y.

Wang, Y. Liao, G. Zhang, H. Hu and Y. Chen, *Adv. Energy Mater.*, **2024**, *14*, 2303661.

[133] Y. Zhang, W. Deng, C. Petoukhoff, X. Xia, Y. Lang, H. Xia, H. Tang, H. Chandran, S. Mahadevan, K. Liu, P. Fong, Y. Luo, J. Wu, S. Tsang, F. Laquai, H. Wu, X. Lu, Y. Yang and G. Li, *Joule*, **2024**, *8*, 509-526.