Supporting Information for

Quinone-Based Conjugated Polymer Cathodes Synthesized via Direct Arylation for High Performance Li-Organic Batteries

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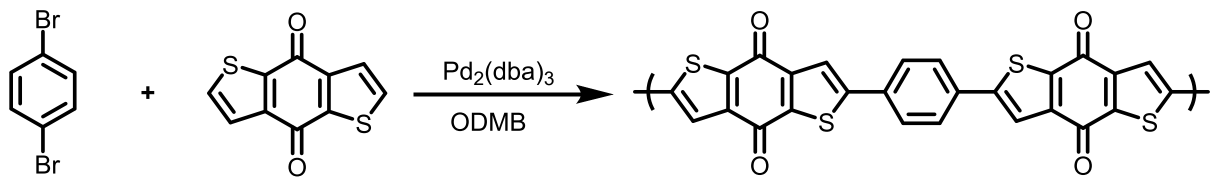
Experiment section

Materials:

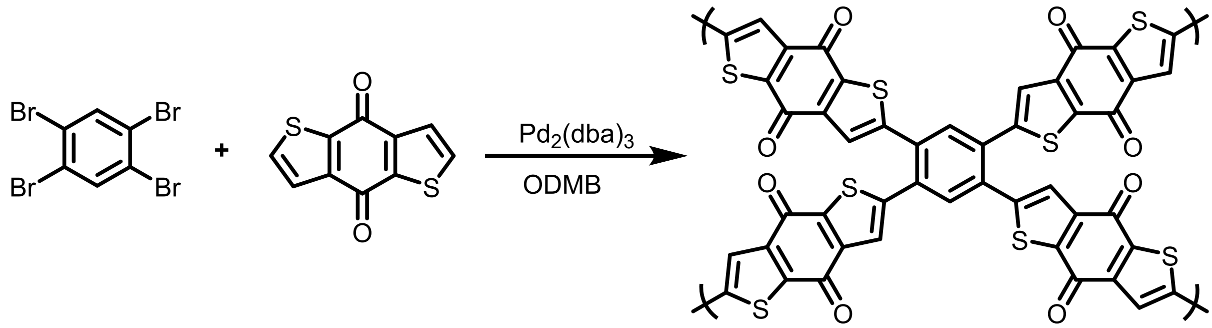
Anhydrous potassium carbonate (K_2CO_3), anhydrous 1,2- dimethylbenzene (ODMB), pivalic acid (PivOH), tris(dibenzylideneacetone)dipalladium ($Pd_2(dba)_3$), tris(2-methoxyphenyl)phosphine ($P(o\text{-}MeOPh)_3$), benzo[1,2-b:4,5-b']dithiophene-4,8-dione (BDTD), 1,4-dibromobenzene, 1,2,4,5-tetrabromobenzene, 1.0 M LiTFSI in dimethoxyethane (DME) and dioxolane (DOL) (1:1, v/v) , aluminum foil , Ketjen black and sodium alginate were purchased from Acros, Alfa, TCI, and Aldrich, and they are used directly without purification.

Synthesis of conjugated polymer PBDTD-1:Under a nitrogen atmosphere, BDTD (110.14 mg, 0.5 mmol), 1,4-dibromobenzene (117.95 mg, 0.5 mmol), anhydrous K_2CO_3 (180 mg, 1.30 mmol), PivOH (30.64 mg, 0.3 mmol) , $Pd_2(dba)_3$ (13.7 mg, 0.0015 mmol), $P(o\text{-}MeOPh)_3$ (10.6 mg, 0.03 mmol) and anhydrous o-xylene (5 mL) were added into a 100 mL two-necked flask. The reaction system was heated to 100 ^oC for 72 h, and then cooled to room temperature. After filtration, the obtained precipitate was washed three times with methanol, and water. Finally, the polymer was dried at 80 ^oC under vacuum for 24 h to obtain a deep-red powder. The average number molecular weight is 34,454 g/mol. The yield was 85.56%. Anal. Calcd. for $(C_{16}H_6O_2S_2)_n$ (%): C 65.29; H 2.05; O 10.87; S 21.78. Found: C 61.14; H 2.35; O 11.72; S 19.74.



Scheme S1. Synthetic route of the PBDTD-1.

Synthesis of conjugated polymer PBDTD-2: Under a nitrogen atmosphere, BDTD (220.27 mg, 1 mmol), 1,2,4,5-tetrabromobenzene (196.85 mg, 0.5 mmol), anhydrous K_2CO_3 (180 mg, 1.30 mmol), PivOH (30.64 mg, 0.3 mmol), $Pd_2(dba)_3$ (13.7 mg, 0.0015 mmol), $P(o\text{-}MeOPh)_3$ (10.6 mg, 0.03 mmol) and anhydrous o-xylene (5 mL) were added into a 100 mL two-necked flask. The reaction system was heated to 100 °C for 72 h, and then cooled to room temperature. After filtration, the obtained precipitate was washed three times with methanol, water, and dichloromethane. Finally, the polymer was dried at 80 °C under vacuum for 24 h to obtain an orange solid powder. The yield was 94.56%. Anal. Calcd. for $(C_{26}H_6O_4S_4)_n$ (%): C 61.16; H 1.18; O 12.53; S 25.12. Found: C 56.42; H 1.73; O 13.27; S 24.08.



Scheme S2. Synthetic route of the PBDTD-2.

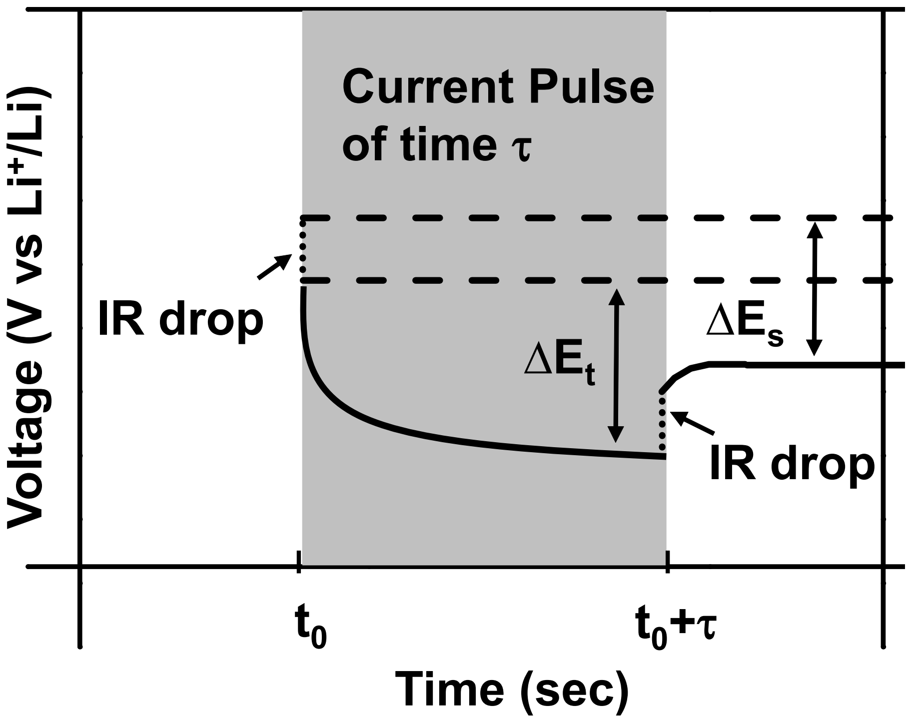
Characterizations: Infrared spectra of the polymers measured on a NICOLET 5397 AVATAR 360 FT-IR. The solid state ^{13}C CP/MAS NMR spectrum was performed on the JEOL RESONRNCE ECZ 400R NMR spectrometer with a MAS frequency of 12 kHz. The thermal stability of the polymers of PBDTD was performed by a thermogravimetric analysis instrument (Q1000DSC+LNCS+FACS Q600DT) with the temperature range from 20 to 800 °C under N_2 condition. Powder X-ray diffraction (PXRD) measurement was carried out on X-ray diffractometer (D/Max-3c). The elemental analysis measurement was carried out on the EURO EA30000 elemental analysis instrument. The morphology of the polymer was collected on the field emission scanning electron microscope (SEM) (SU8020, Hitachi). The UV-Vis adsorption spectra were recorded on a scanning UV-Vis spectrophotometer (UV-Lambda 950, PerkinElmer, US) using $BaSO_4$ as a reference sample. The specific surface area and pore size distributions were obtained by an ASAP 2420-4 (micromeritics) CO_2 adsorption analyzed at 195.6 K. The DFT simulation of the electronic structure of the polymer was measured at the B3LYP/6-31G (d) level by using Gauss 09. The density functional theory calculation of the binding energy was carried out by the projected enhanced wave (PAW) method.

Electrochemical performance measurements: The polymer cathodes were prepared by mixing active material (60 wt%) with Ketjen black (30 wt%) and sodium alginate binder (10 wt%) in a mortar using distilled water as the solvent. The obtained slurry was evenly coated on the aluminum foil current collector by a doctor blade, and then dried at 50 ^oC for 6 h and 110 ^oC for 12 h, respectively. The mass loading of the active material is in the range of 0.7-1 mg/cm^2. The CR2016 button cells were assembled using lithium foil as the anode, glass fiber membrane as the separator, and 1.0 M LiTFSI in dimethoxyethane (DME) / dioxolane (DOL) (1:1, v/v) as the electrolyte in a glove box with argon atmosphere (water and oxygen content < 1.0 ppm). The galvanostatic charge/discharge measurement was carried out on a NEWARE-BTS-5 V/10 mA battery tester. Cyclic voltammetry test was performed on an electrochemical workstation (CHI660E, Shanghai Chenhua).

The GITT results were calculated employing the following equation (1)[1]:
If the battery voltage has a linear relationship with τ 1/2, the above formula can be simplified as the following formula (2):

$$D=\frac{4}{\pi\tau}\left(\frac{M_B V_M}{M_B S}\right)^2\left(\frac{\Delta ES}{\tau\left({dE\tau}/{D\sqrt{\tau}}\right)}\right)^2 \quad (\pi \ll L^2/D) \qquad (1)$$

$$D=\frac{4}{\pi\tau}\left(\frac{V_m m_B}{S\ MB}\right)^2\left(\frac{\Delta Es}{\Delta E\tau}\right)^2 \qquad (2)$$



Where τ is the current pulse time (s), V_m is the molar volume of the polymer (cm^3 mol^{-1}), m_B is the mass of the polymer in the electrode (g), M_B is the molar mass of the polymer (g/mol), S is the contact surface area (cm^2) between the electrode and the electrolyte.

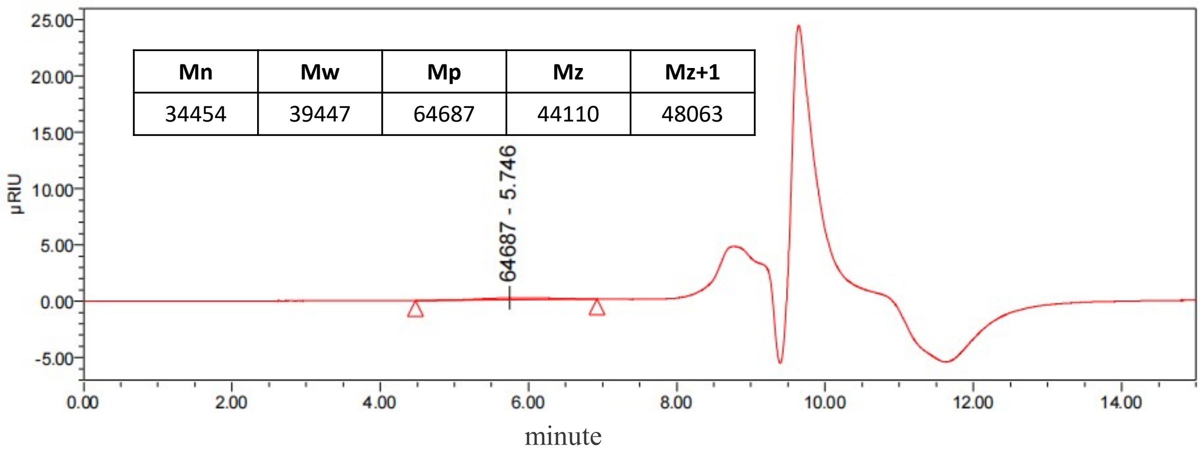


Fig. S1. The gel permeation chromatogram of PBDTD-1 for molecular weight.

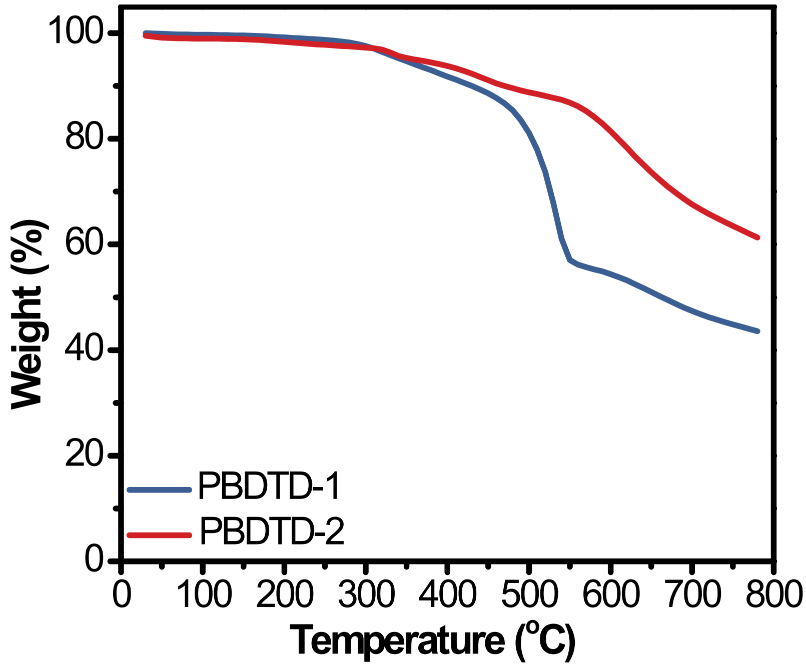


Fig. S2. The TGA curves of PBDTD-1 and PBDTD-2 under nitrogen atmosphere.

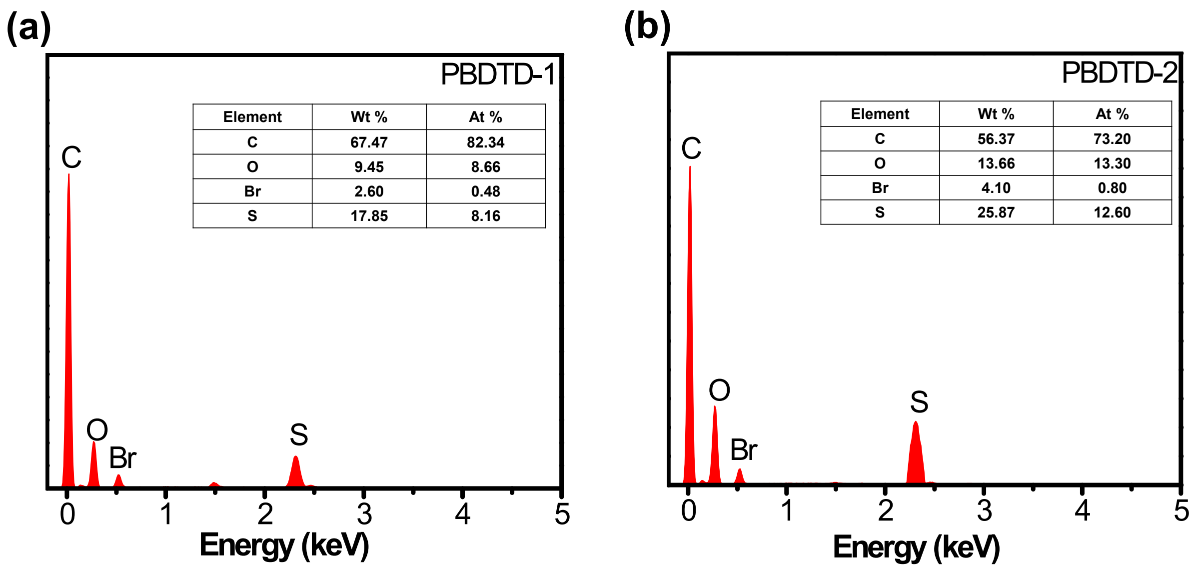


Fig. S3. The EDX spectra of the two polymers. (a) PBDTD-1, (b) PBDTD-2.

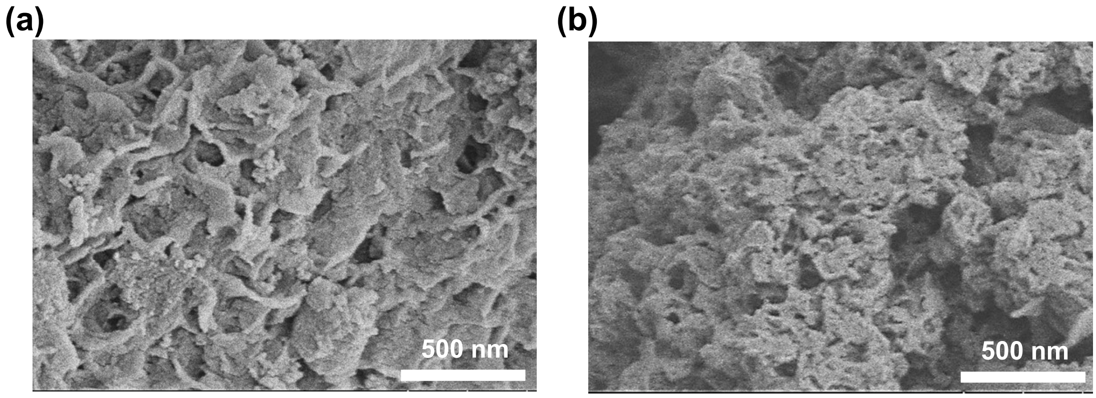


Fig. S4. SEM images of (a) PBDTD-1 and (b) PBDTD-2.

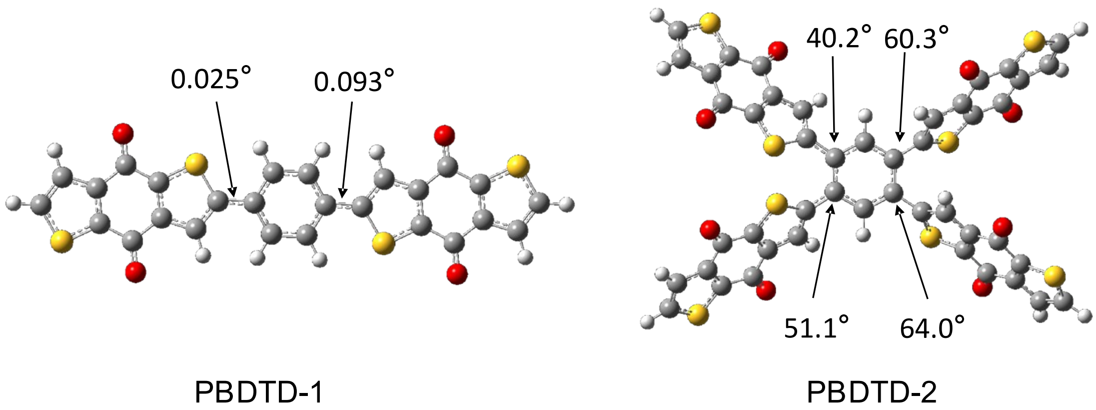


Fig. S5. DFT geometry optimizations and the dihedral angles of the simplified polymer fragments of the polymers.

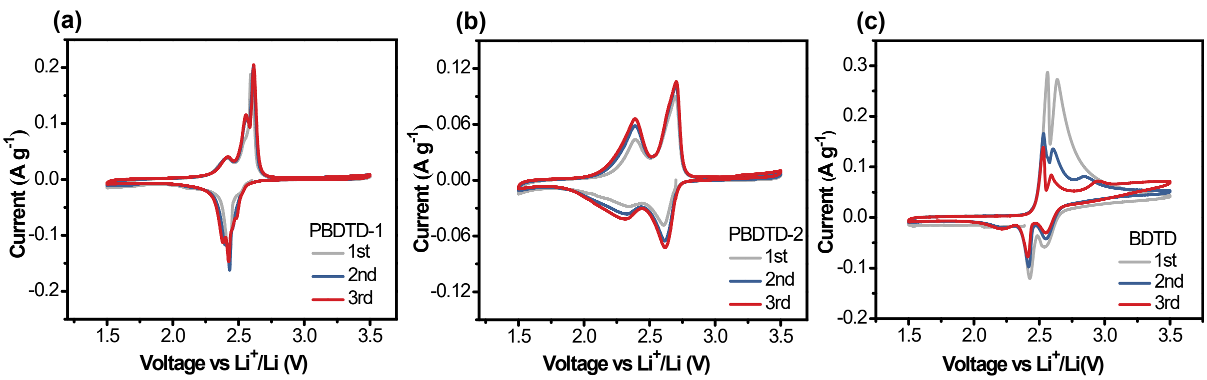


Fig. S6. The CV curves of (a) PBDTD-1, (b) PBDTD-2 and (c) BDTD at 0.1 mV s^{-1}.

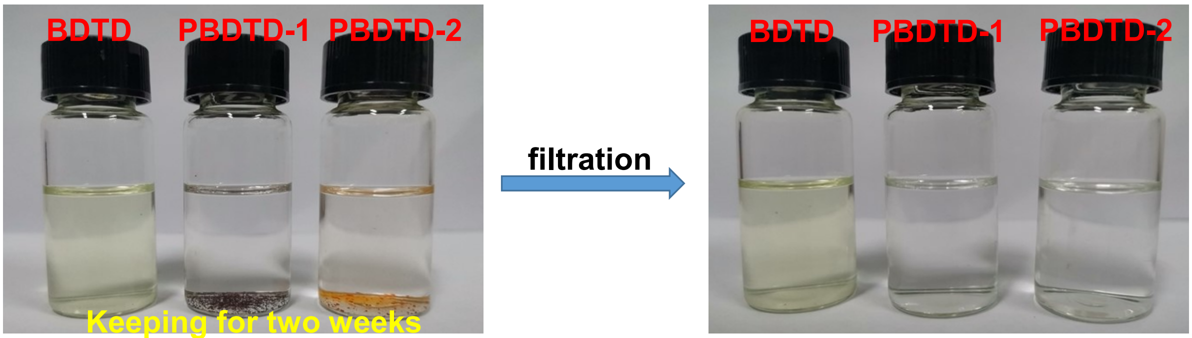


Fig. S7. The photographs of the BDTD and the two polymers in the electrolytes and the corresponding filtrates.

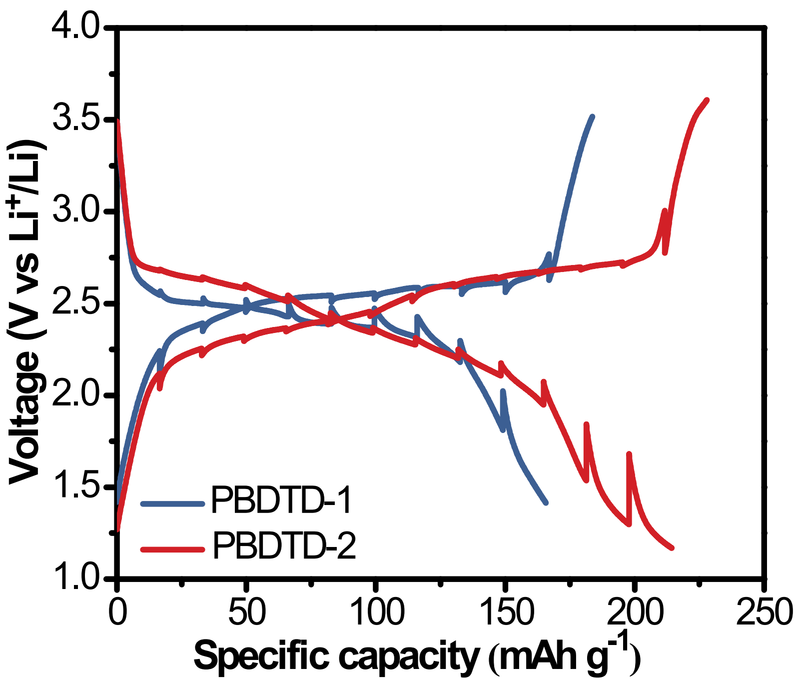


Fig. S8. The GITT response for PBDTD-1 and PBDTD-2.

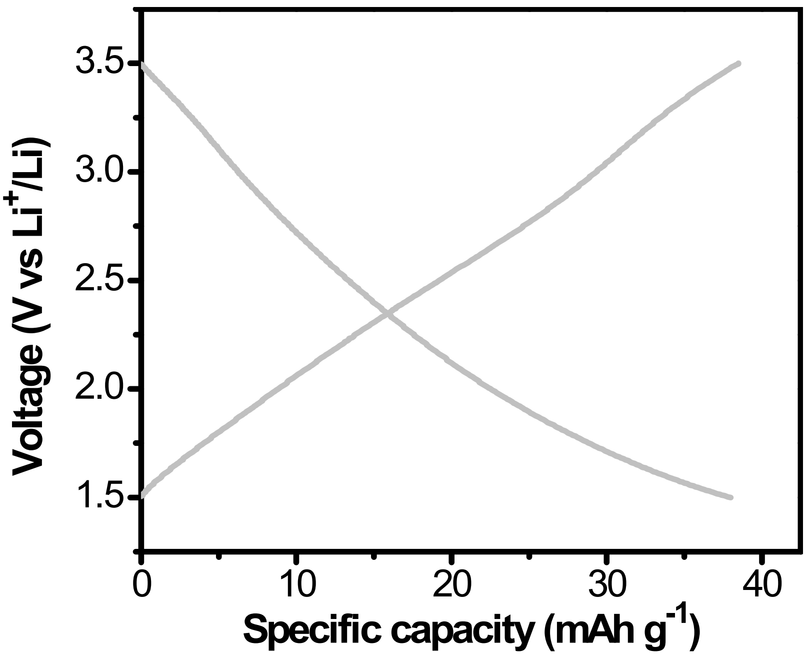


Fig. S9. The GCD curves of Ketjen black.

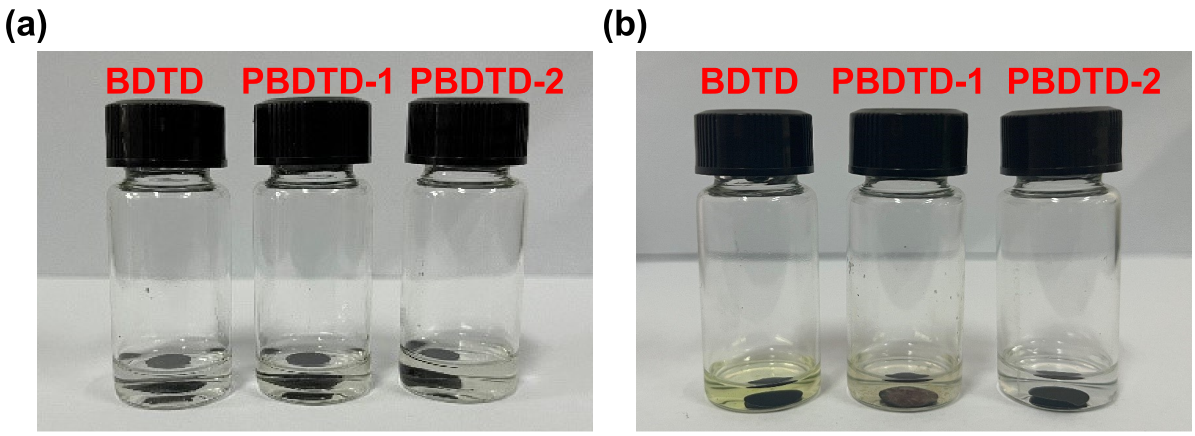


Fig. S10. Photographs of the electrodes immersed in the electrolytes at the pristine (a) and discharged (b) states, respectively.

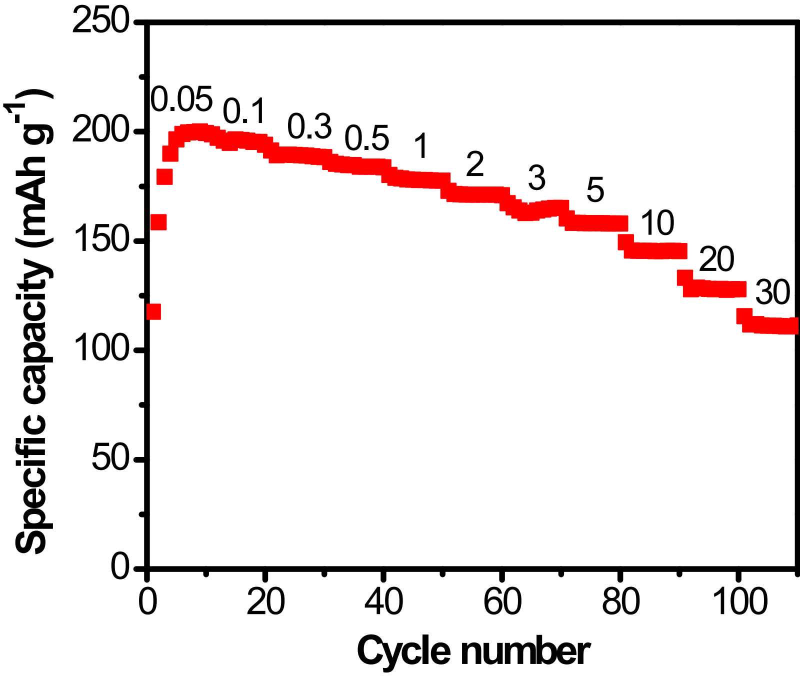


Fig. S11. The rate performance of PBDTD-2.

Table S1. The comparison of electrochemical performances of PBDTD-2 with previously reported organic carbonyl cathodes in alkali metal-ion batteries.

Sample	Specific capacity ($mAh\ g^{-1}$) (current)	Rate capacity ($mAh\ g^{-1}$) (current)	Cycle number (current)	Refs
PBDTD-2	**200 (0.05 A g^{-1})**	**111 (30 A g^{-1})**	**3000 (5 A g^{-1})**	**This work**
PBDAPA	132.7 (0.02 A g^{-1})	97.1 (0.2 A g^{-1})	100 (0.02 A g^{-1})	[2]
Li2PDHBQS	247 (0.05 A g^{-1})	123.5 (10 A g^{-1})	1500 (0.5 A g^{-1})	[3]
PBDTD	214.2 (0.25 A g^{-1})	125 (12.5 A g^{-1})	250 (1.25 A g^{-1})	[4]
PDPA-AQ	159 (0.02 A g^{-1})	50 (0.4 A g^{-1})	100 (0.02 A g^{-1})	[5]
PAQS	185 (0.05 A g^{-1})	151 (0.5 A g^{-1})	-	[6]
PT-CMPs	166.7 (0.05 A g^{-1})	93.1 (1 A g^{-1})	100 (0.05 A g^{-1})	[7]
P15AQS	250 (0.02 A g^{-1})	125 (1 A g^{-1})	200 (0.02 A g^{-1})	[8]
Na_2AQ26DS	138 (0.05 A g^{-1})	40 (2 A g^{-1})	900 (0.05 A g^{-1})	[9]
PMAQ-SWNT	190 (0.04A g^{-1})	120 (8 A g^{-1})	300 (0.2 A g^{-1})	[10]
PyAq	169 (0.02 A g^{-1})	142 (1 A g^{-1})	4000 (1 A g^{-1})	[11]
DTT	292 (0.03 A g^{-1})	220 (0.3 A g^{-1})	200 (0.03 A g^{-1})	[12]
PI10G	232.6 (0.02 A g^{-1})	159.8 (1 A g^{-1})	1000 (10 A g^{-1})	[13]
BAQB	212 (0.04 A g^{-1})	138 (2 A g^{-1})	100 (0.04 A g^{-1})	[14]
NTAQ	130.8 (0.05 A g^{-1})	62.4 (1.5 A g^{-1})	1000 (0.15 A g^{-1})	[15]
TAPT-NTCDA@CNT	155.5 (0.05 A g^{-1})	89.7 (2 A g^{-1})	500（1 A g^{-1})	[16]
PPTC	146.3 (0.05 A g^{-1})	87.2 (0.5 A g^{-1})	300 (0.5 A g^{-1})	[17]
PI-4	86.2 (0.025 A g^{-1})	42.1 (1 A g^{-1})	10000 (0.5 A g^{-1})	[18]
PAQS/CNT-2	184(0.02 A g^{-1})	79.1 (0.4 A g^{-1})	200 (0.04 A g^{-1})	[19]
PI50	165 (0.5 A g^{-1})	125 (2 A g^{-1})	9000 (2 A g^{-1})	[20]

C_6O_6	902 (0.02 A g^{-1})	382 (0.5 A g^{-1})	100 (0.05 A g^{-1})	21
AQ-COF@CNTs	144 (0.05 A g^{-1})	69 (10 A g^{-1})	3000 (0.25 A g^{-1})	22
PQ	197 (0.05 A g^{-1})	85 (5 A g^{-1})	500 (0.05 A g^{-1})	23
IEP-11-S5R5@ S10	83.7 (0.004 A g^{-1})	13.3 (1.5 A g^{-1})	1000 (0.15 A g^{-1})	24
PTCDI-DAQ	229 (0.1 A g^{-1})	46 (25 A g^{-1})	20000 (3 A g^{-1})	25
DAPQ-COF50	162 (0.5 A g^{-1})	94 (50 A g^{-1})	3000 (2 A g^{-1})	26
DTN	285.8 (0.05 A g^{-1})	153.6 (4 A g^{-1})	1000 (1.5 A g^{-1})	27
BA-PI	85 (0.025 A g^{-1})	9 (0.2 A g^{-1})	-	28
NP2	245 (0.05 A g^{-1})	188 (0.5 A g^{-1})	200 (0.5 A g^{-1})	29
IEP-11-E12	100 (0.15 A g^{-1})	47 (4.5 A g^{-1})	80000 (4.5 A g^{-1})	30
NDI-BQ	168 (0.04 A g^{-1})	-	200 (0.04 A g^{-1})	31
NC@AQ	202.1 (0.025 A g^{-1})	152.9 (0.5 A g^{-1})	1000 (0.05 A g^{-1}, 0.125 A g^{-1})	32
P500	165 (0.02 A g^{-1})	-	100 (0.02 A g^{-1})	33
PAQS2	177 (0.025 A g^{-1})	41 (0.8 A g^{-1})	1000 (0.5 A g^{-1},1 A g^{-1})	34
PAQnS	356 (0.1 A g^{-1})	180 (2 A g^{-1})	125 (0.05 A g^{-1})	35
PFNDI	24.6 (0.05 A g^{-1})	6.5 (2.5 A g^{-1})	500 (0.5 A g^{-1})	36
P14AQ/CNT	220 (0.05 A g^{-1})	25 (1.25 A g^{-1})	100 (0.05 A g^{-1})	37
PAQS	220(0.05 A g^{-1})	125 (1.25 A g^{-1})	1000 (1 A g^{-1})	38
DMAQ	168 (0.08 A g^{-1})	75 (2 A g^{-1})	80 (0.04 A g^{-1})	39
BQbTPL	152.9 (0.1 A g^{-1})	97.3 (1 A g^{-1})	1500 (1 A g^{-1})	40
BBQB	367 (0.04 A g^{-1})	171 (1.2 A g^{-1})	100 (0.04 A g^{-1})	41
PAQS@3D-C	219 (0.05 A g^{-1})	173 (2.5 A g^{-1})	500 (0.05 A g^{-1})	42

PPh-PTO	235 (0.1 A g^{-1})	94 (2 A g^{-1})	1400 (0.1 A g^{-1})	43
LiDHAQS	350 (0.1 A g^{-1})	236 (4 A g^{-1})	1200 (0.8 A g^{-1})	44
2D-PDI@CNT	104 (0.2 A g^{-1})	95 (2 A g^{-1})	8000 (0.5 A g^{-1})	45
PPTODB	185 (0.02 A g^{-1})	98 (1.5 A g^{-1})	150 (0.02 A g^{-1})	46
PT-2NO_2	301.3 (0.05 A g^{-1})	103.7 (1 A g^{-1})	500 (0.5 A g^{-1})	47
COF-TRO	268 (0.03 A g^{-1})	98 (0.6 A g^{-1})	100 (0.03 A g^{-1})	48
PIBN-G	271 (0.03 A g^{-1})	206.7 (1.5 A g^{-1})	300 (1.5 A g^{-1})	49
PAQS	129 (0.05 A g^{-1})	107 (1 A g^{-1})	200 (0.05 A g^{-1})	50
DAAQ-TFP COF	~60 (0.1 A g^{-1})	~30 (4 A g^{-1})	500 (0.2 A g^{-1})	51
PI-ECOF-1/rGO50	167 (0.014 A g^{-1})	90 (1.42 A g^{-1})	300 (0.014 A g^{-1})	52
PI@GS-2	165 (0.02 A g^{-1})	125 (4 A g^{-1})	5000 (1 A g^{-1})	53
PEPTO	249 (0.02 A g^{-1})	98 (1.5 A g^{-1})	1000 (0.8 A g^{-1})	54
PBDTDS	196 (0.2 A g^{-1})	-	4200 (2 A g^{-1})	55

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