

## Electronic Supplementary Information (ESI)

### A Nile red dye cathode with an asymmetric redox unit for lithium organic battery

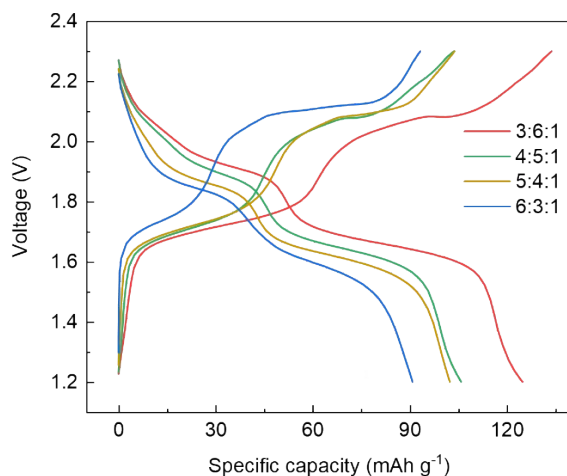
#### Experimental

**Materials:** All reagents were obtained from commercial sources and used without further purification. Nile Red (NR) were purchased from heowns and multi-walled carbon nanotubes (MWCNTs) were purchased from Shanghai Macklin Biochemical Co., Ltd. Polyvinylene difluoride (PVDF, Mw ~1,000,000) was purchased from Beijing Huawei Ruike Chemical Co., Ltd. The components of the electrolyte, including bis(trifluoromethylsulfonyl)amine lithium salt (LiTFSI), 1,3-dioxolane (DOL), and 1,2-dimethoxyethane (DME) were purchased from Dodochem.

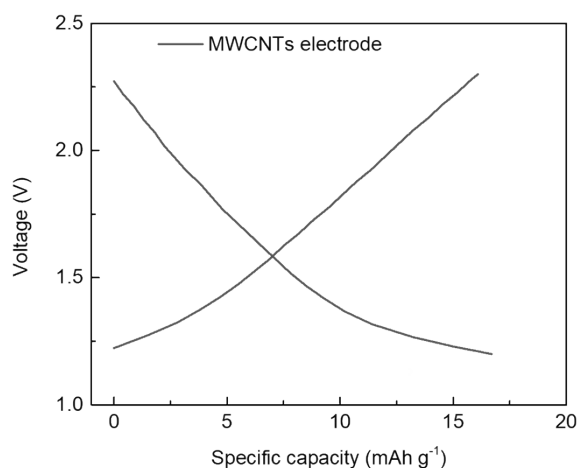
**Characterization:** Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) measurements were performed on an electrochemical workstation (CHI660E). Galvanostatic charge/discharge curves and rate capability tests were carried out on a Land test system (CT3001A, China). X-ray photoelectron spectroscopy (XPS) characterization was conducted on a Thermo Scientific Nexsa equipment. Electron spin resonance (ESR) spectra were recorded on a Bruker EMXplus-6/1 equipment. UV-vis spectra were recorded on a SIMADZU UV-3600i Plus spectrophotometer

**Cathode fabrication, coin cell assembly and battery test:** The conventional organic electrode is composed of a mixture of 30wt% electroactive materials (NR), 60wt% conductive carbon (MWCNTs), and 10wt% binder (PVDF). A series of electrodes with NR: MWCNTs: PVDF weight ratios as 4:5:1, 5:4:1, 6:3:1, and 0:6:1 were also fabricated and tested. The mixing process was conducted using a conventional mortar and pestle, during which NR, MWCNTs, and PVDF were added to the mortar in sequence and ground for 15 min. The mixture was poured into a tablet mold and punched into circular sheets with 12 mm in diameter. The electrodes were dried at 80 °C in a vacuum oven for over 8 h before assembling coin-type cells in

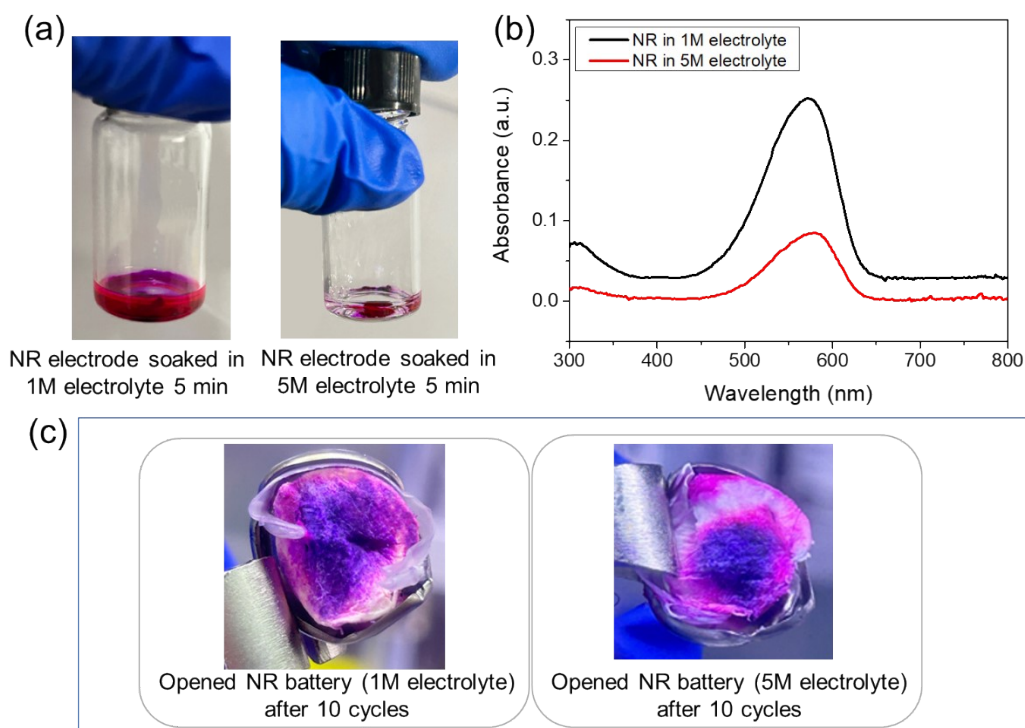
an argon-filled glovebox. The coin cells were composed of such electrodes as the cathode, lithium metal disc as the anode, glass fiber (GF) as the separator and 1.0 M (1M) or 5.0 M (5M) LiTFSI dissolved in DOL-DME (1:1, v:v) as the electrolyte.



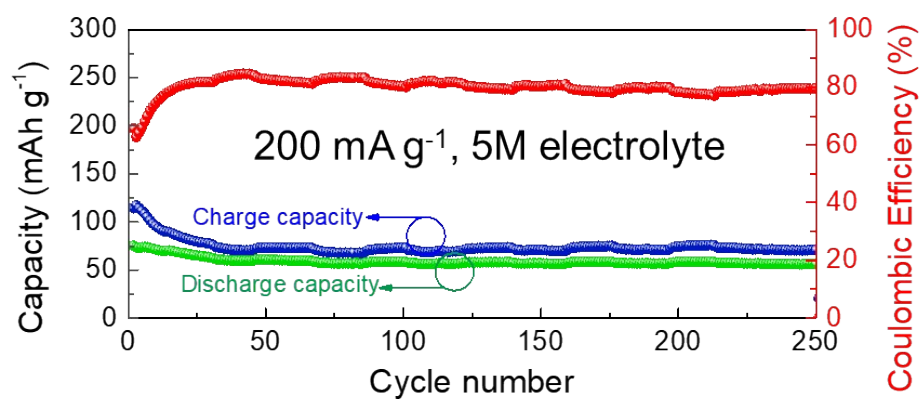
**Fig. S1.** Discharge/charge profiles of NR-based electrodes with the weight ratios of NR: MWCNTs: PVDF as 3:6:1, 4:5:1, 5:4:1, and 6:3:1, respectively.



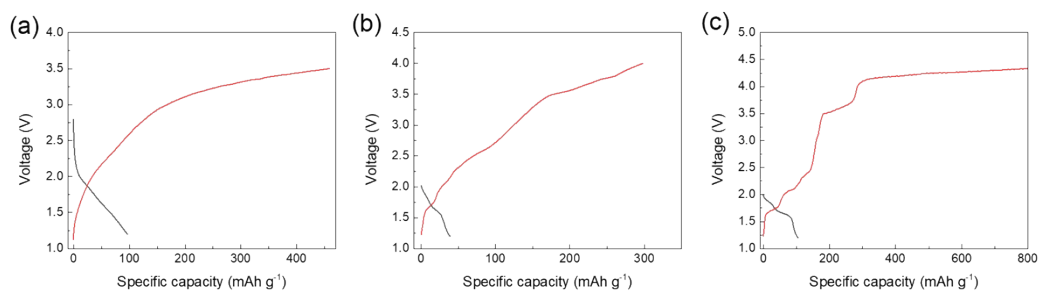
**Fig. S2.** Discharge/charge profile of MWCNTs electrode. The electrode weight ratio of NR: MWCNTs: PVDF is 0:6:1. For the relevant NR-based electrodes with a 60–30% MWCNTs weight content, the capacities contributions of MWCNTs are calculated only 5–10 mAh g<sup>-1</sup> out of their capacities of 90–125 mAh g<sup>-1</sup>.



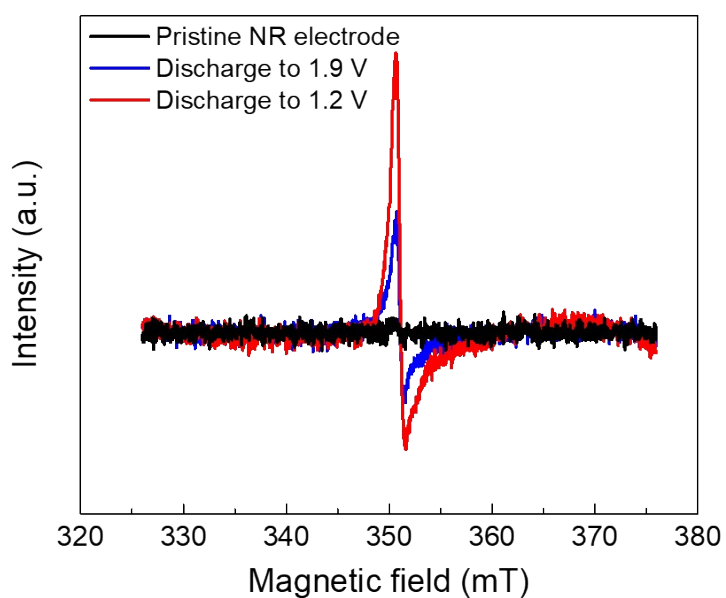
**Fig. S3.** (a) The images of NR electrode soaked in 1M/5M electrolyte. (b) UV-vis spectra of NR in 1M/5M electrolyte (tested using 1 mg NR soaked in 10 mL 1M/5M electrolyte 10 min). (c) The images of the opened NR batteries (1M and 5M electrolyte) after 10 cycles.



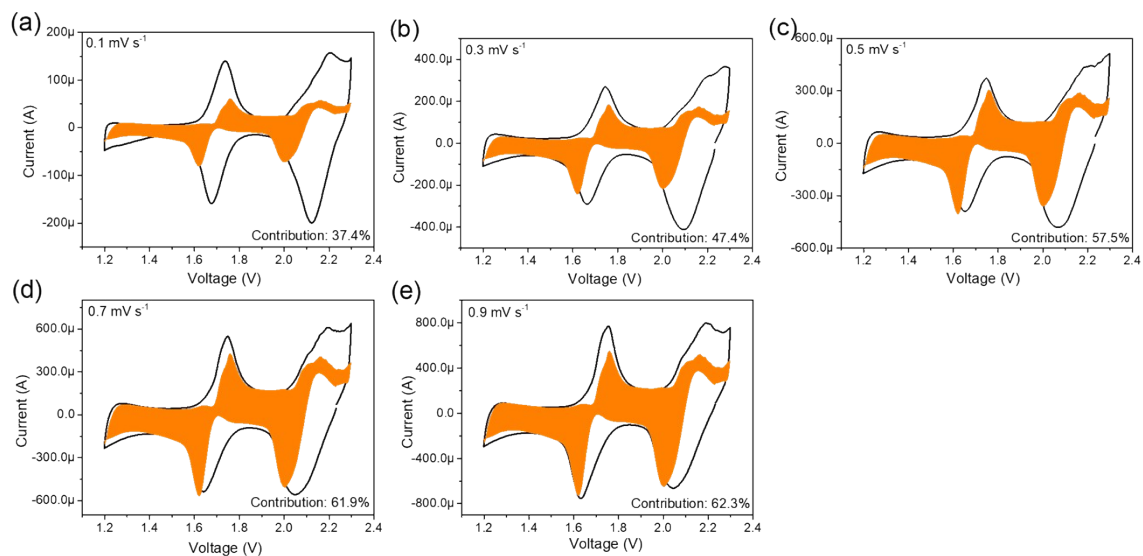
**Fig. S4.** Cycling performance of NR-based battery using 5M electrolyte at 200 mA g<sup>-1</sup>.



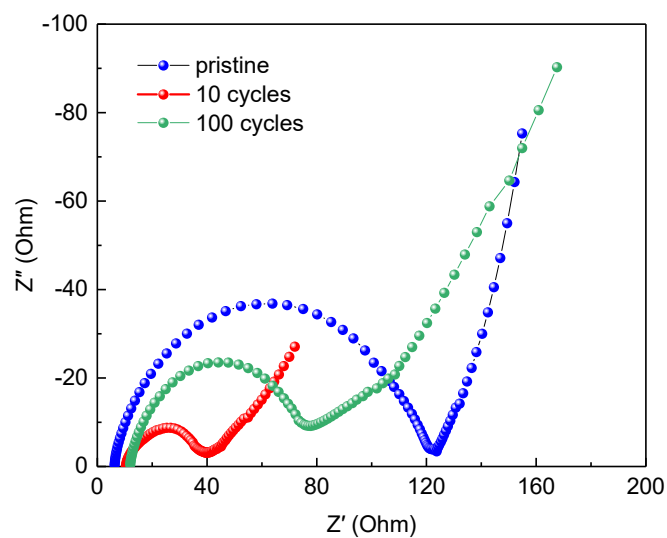
**Fig. S5.** Discharge/charge profiles of NR-based batteries under the voltage ranges of 1.2–3.5 V (a), 1.2–4.0 V (b), and 1.2–4.5 V (c).



**Fig. S6.** Electron spin resonance (ESR) spectra of the pristine and two different discharging states NR electrodes.



**Fig. S7.** CV curves and capacitive contributions of NR electrode at scan rates of (a) 0.1, (b) 0.3, (c) 0.5, (d) 0.7, and (e) 0.9  $\text{mV s}^{-1}$ .



**Fig. S8.** Nyquist plots of NR electrodes-pristine, NR electrodes after 10 cycles, and NR electrodes after 100 cycles.