Electronic Supplementary Material (ESI) for RSC Advances

Exceptional electrochemical performance of rechargeable Li-S batteries with polysulfide-containing electrolyte

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Experimental Details

Materials:

Dimethoxyethane (DME), dioxolane (DOL) and lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) certified to contain less than 20 ppm H_2O were obtained from Novolyte Technologies. Sulfur (99.5%), Li_2S (99.9%) and $LiNO_3$ (99.999%) were purchased from Alfa Aesar. All the chemicals were used as received.

Synthesis:

A reference electrolyte of 0.1 M LiTFSI + 0.2 M $LiNO_3$ in DOL/DME (1:1, v:v) was prepared first by dissolving proper amounts of LiTFSI and $LiNO_3$ in DOL/DME (1:1, v:v). To this solution, stoichiometric amounts of elemental sulfur and pristine Li_2S were added to form polysulfide-containing electrolytes of the required sulfur concentration ([S]) and average polysulfide chain length. The reaction and dissolution were complete after stirring for 6 h at 75 °C followed by 48 h at room temperature. Polysulfide-containing electrolytes prepared in this way were dark red and of moderate viscosity.

Sulfur cathodes were prepared by ball milling 50 wt% elemental sulfur, 40 wt% Super P carbon black, and 10 wt% PVDF binder in NMP solution at 300 rpm for 3 h to make the slurry, and then spreading the slurry on aluminium foil using a common doctor-blade coating method. After drying at 55 °C under vacuum overnight, the electrodes were cut into circular pieces of 1.13 cm^2 (12 mm diameter) with sulfur loading of about 0.6 mg cm^{-2} and incorporated into CR-2016 coin-type cells with a precisely controlled amount of either polysulfide-free or -containing electrolyte. All the electrolyte preparation and cell assembly steps were performed in an Ar-filled glove box with O_2 and H_2O less than 1 ppm.

Characterization:

The coin-type cells were galvanostatically cycled on battery testing systems (Neware BTS-5V1mA or Arbin BT-2000) at room temperature. The cutoff potentials for charge and discharge were set at 2.6 V and 1.6 V vs. Li^+/Li, respectively. Cyclic voltammetry (CV) scanning was carried out on a CHI660 system using coin-type cells and with a scanning rate of 0.1 mV s^{-1}.

Additional Figures

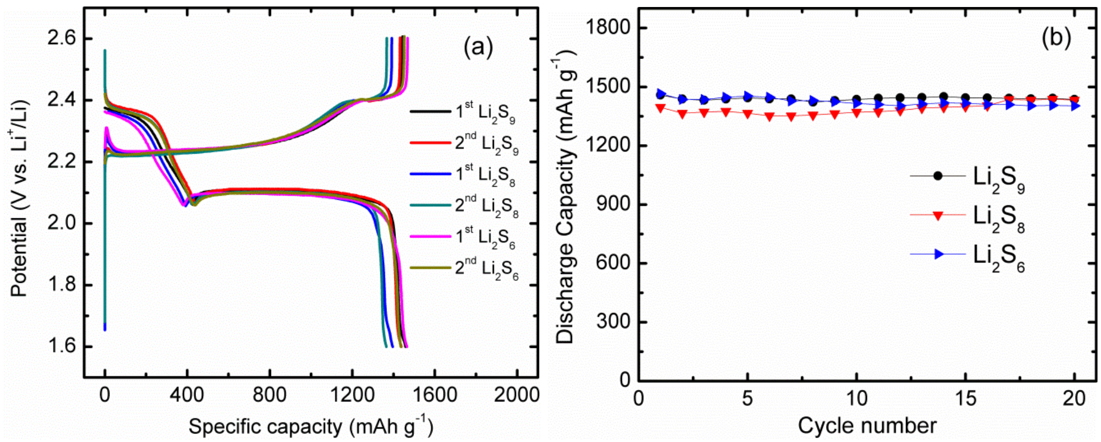

Figure S1. (a) Voltage profiles and (b) cycling performance at C/3 rate in 10 μL of polysulfide-containing electrolytes with different average polysulfide chain lengths and sulfur concentration [S]=2 M between 1.6 V and 2.6 V.

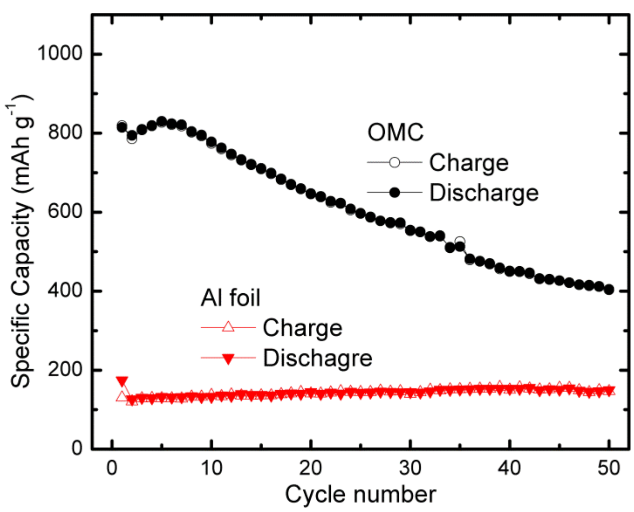

Figure S2. Cycling performance at a current density of 0.3 mA/cm^2 in 10 μL of polysulfide-containing electrolyte with sulfur concentration [S]=2 M charged/discharged between 1.6 V and 2.6 V using an Al foil and an ordered mesoporous carbon (OMC, as reported in the literiture[1]) electrode of 1.13 cm^2 as the positive electrodes.

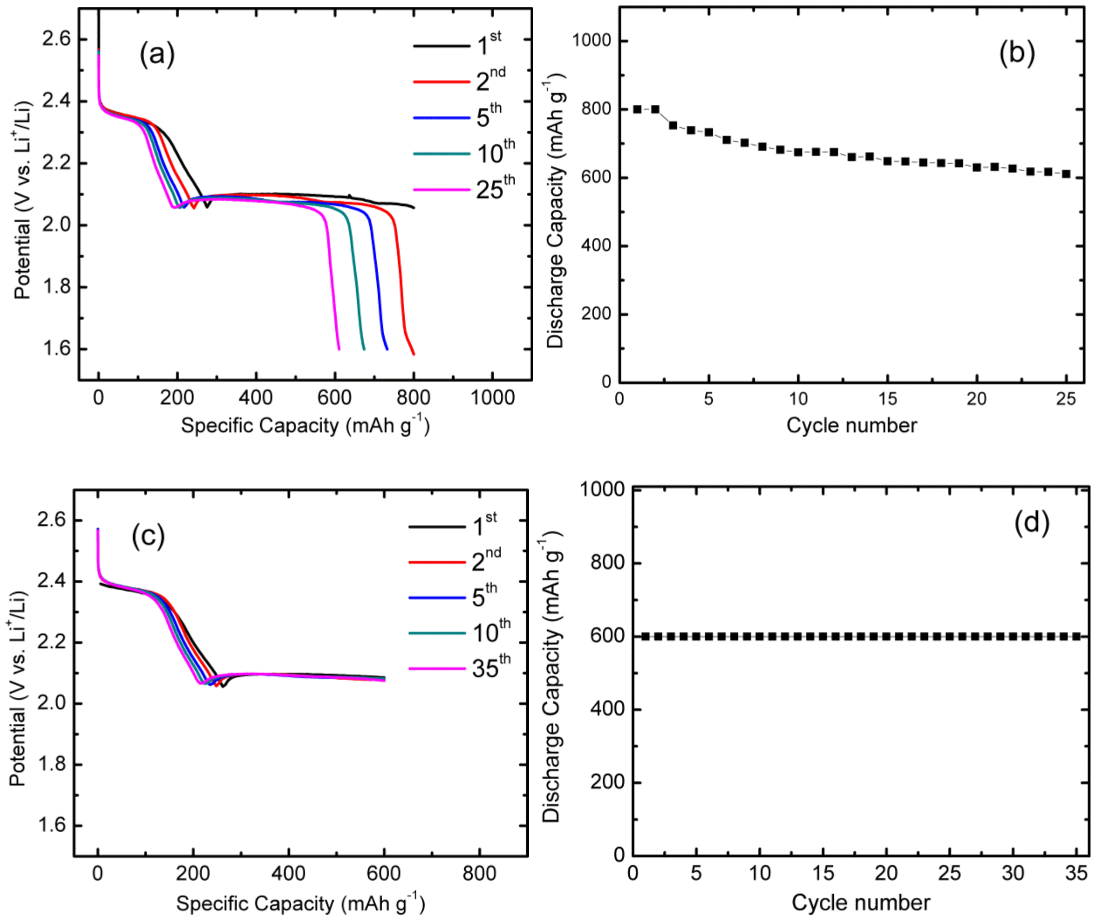

Figure S3. (a, c) Voltage profiles and (b, d) cycling performance at C/10 rate in 10 µL of polysulfide-free electrolyte discharged with a cut-off voltage at 1.6 V and a cut-off discharge capacity of (a, b) 800 mAh/g and (c, d) 600 mAh/g (cells stopped discharging at whichever condition was met first). It is clear that cycling performance was greatly improved when a cut-off discharge capacity of 600 mAh g^{-1} was selected.

References:

1. S. R. Chen, Y. P. Zhai, G. L. Xu, Y. X. Jiang, D. Y. Zhao, J. T. Li, L. Huang and S. G. Sun, *Electrochim. Acta*, 2011, **56**, 9549-9555.