Support Information

Superior Rate Capability of Sulfur Composite Cathode in Tris(trimethylsilyl)borate-Containing Functional Electrolyte

Lina Wang, Qinyu Li, Huijun Yang, Jun Yang, Yanna Nuli, Jiulin Wang[a*]

Shanghai Electrochemical Energy Devices Research Center, Department of Chemical Engieering, Shanghai Jiao Tong University, Shanghai 200240, China, email: wangjiulin@sjtu.edu.cn

Experimental section:

Sample preparation. A representative sulfur-based composite (S@pPAN) was used in this work as the cathode material. It was prepared by mixing 1g PAN and 8g sulfur homogeneously in the milling ball for 12 h with ethanol as a dispersant. Then the collected light yellow powder was dried and co-heated at 300 ^{o}C in N_2 atmosphere for 450 min, resulting in black powder S@pPAN composite.

Coin-cell assembly. The sulfur-based composite with 43 wt.%. S determined by elemental analysis (Vario EL III elemental analyzer, Elementar, Germany) was mixed with Super P and carbonyl-β-cyclodextrin (C-β-CD) which was prepared according to the Reference 9, using distilled water as the dispersant. The ratio of sulfur composite, Super P and C-β-CD in the mixture was 80:10:10. The slurries were stirred for 4 h and then uniformly coated onto aluminum foil substrate. After the water solvent was evaporated, the electrode film was cut to sheets with 14 mm in diameter and then dried at 70 ^{o}C under vacuum for 12 h, and the electrode loading was about 1 mg cm^{-2}. The basic electrolyte was 1 M $LiPF_6$/EC+DMC (1:1, v/v). The functional electrolytes were prepared by adding tris(trimethylsilyl) borate (TMSB) into the basic electrolyte. Finally, the CR2016-type coin cells were assembled with the above electrodes, pure lithium foil as counter electrode and Celgard 2400 as separator in an Ar-filled glove box (MBraun, Germany) containing less than 1 ppm water and O_2.

Electrochemical measurements. The galvanostatic charge and discharge tests were conducted on a LAND-CT 2001A Cell Test System (Wuhan, China), in a voltage range of 1.0-3.0 V (vs. Li/Li^{+}). The rate performance of Li-S battery was evaluated using the same equipment with a constant charge current density at 0.5C, and discharged at 0.5C, 1C, 2C, 3C, 4C, 5C, 6C, 7C, 10C respectively. The electrochemical impedance spectrascopy (EIS) of the cells were measured using AUTOLAB PGSTAT302N (Metrohm) at room temperature, in the frequency range from 0.1 MHz to 0.01 Hz. Cyclic voltammogram was measured with a three-electrode cell equipment using a CHI604A Electrochemical Workstation (Shanghai, China). The working electrode was sulfur-based electrode of 12 mm diameter, and lithium slice and lithium disc served as refer electrode (RE) and counter electrode (CE) respectively.

Physical characterizations. The ionic conductivity of the electrolyte solutions were measured at room temperature using a FE30 conductivity meter and an Inlab 710 conductivity measuring cell (Mettler Toledo, Switzerland), by inserting the Inlab 710 conductivity electrode into the electrolyte solution after calibration. The X-ray photoelectron spectroscopy (XPS) analysis was conducted to determine the electrode surface compositions, using the Kratos Axis UltraDLD spectrometer (Kratos Analytical-A Shimadzu Group Company) with monochromatic Al Ka radiation source (1486.6eV) under ultra-high vacuum. The analysis area was 700×300 um and analysis chamber pressure was less than 5×10^{-9} Torr. The obtained spectra were fitted using XPS peak software (version 4.1) using Gaussian-Gauss2 peak shapes and a Shirley function background correction. The Kratos charge neutralizer system was used on all specimens except the conductive samples. Before testing, the cycled sulfur-based cathode were washed with anhydrous DMC several times to remove residual salts, and then dried in vacuum oven for 2h at room temperature.

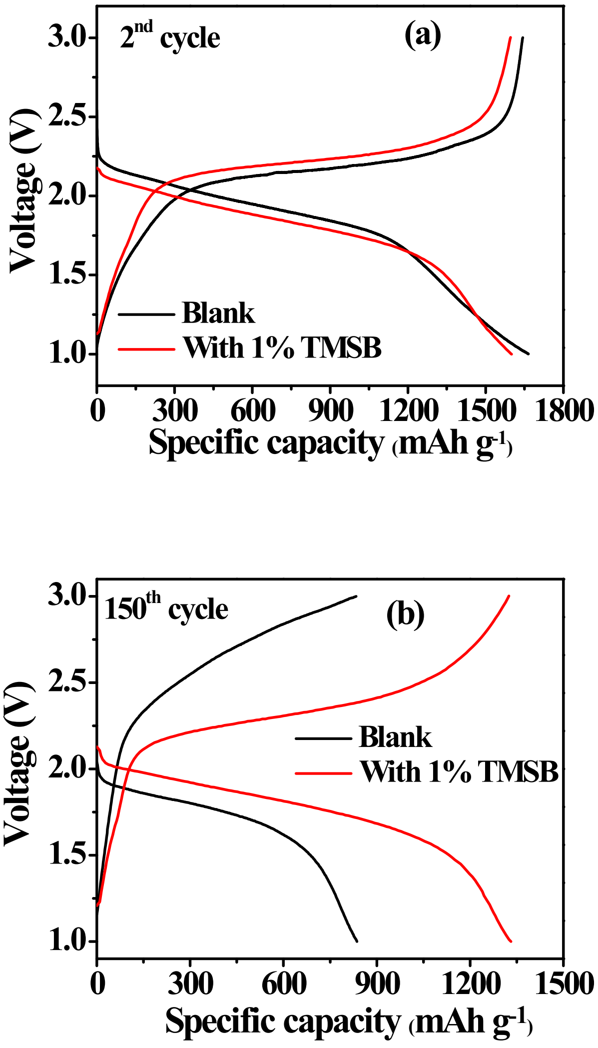

Fig. S1 Voltage profiles for the cells under 0.5 C (a) at the 2nd cycle, and (b) the 150th cycle.

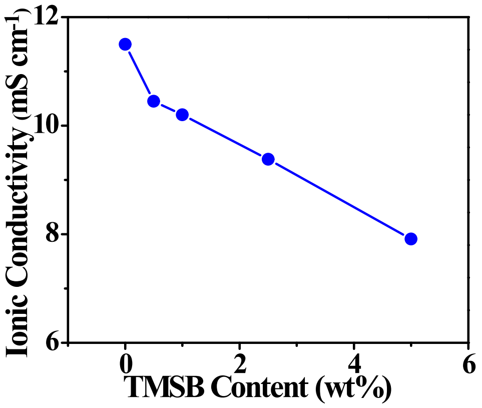

Fig. S2 Ionic conductivity of the electrolytes with different amounts of TMSB.

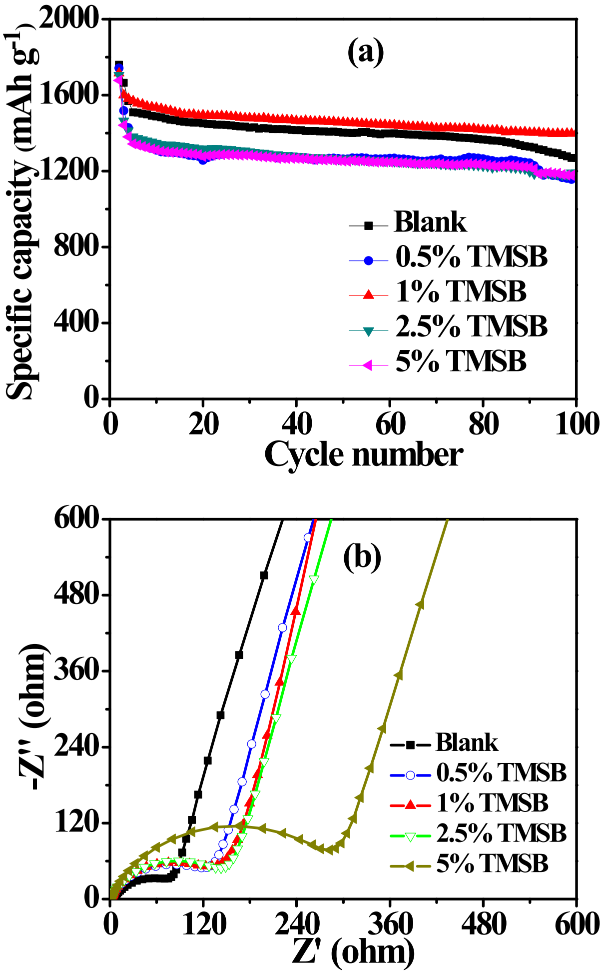

Fig. S3 (a) cyclic performance and (b) EIS of the S@pPAN cathodes with different amounts of TMSB.

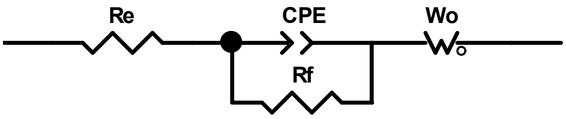

Fig. S4 Equivalent circuits for EIS fitting. Where R_e is the resistance of the electrolyte; CPE and R_f are capacitance and resistance of the interfacial film on the electrode; W_o represents the Warburg impedance related to the diffusion of Li^+ ion in the electrode materials.

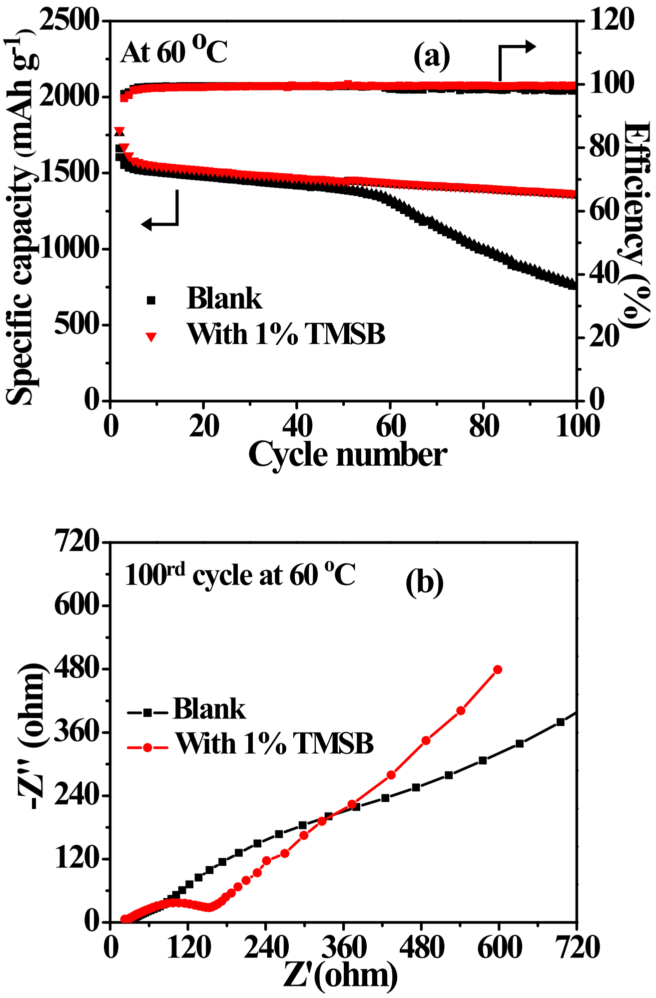

Fig. S5 (a) Cyclic performance and coulombic efficiency at 60 °C under 0.5 C. (b) EIS of the cells after 100 cycles at 60 °C.

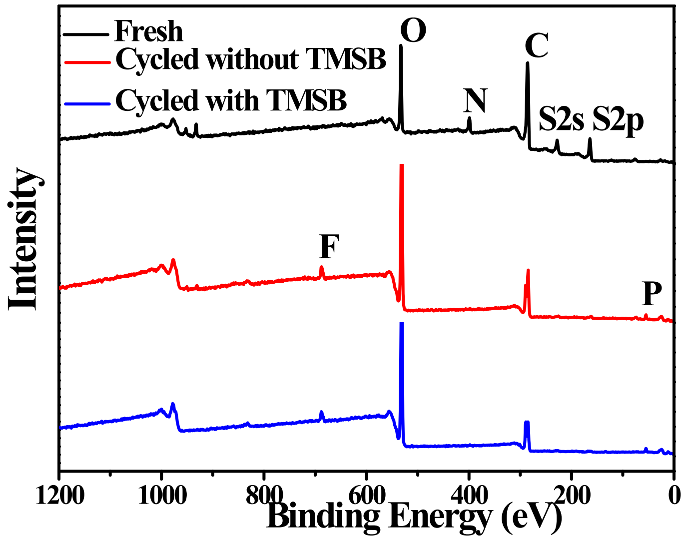

Fig. S6 XPS full-scale spectra of the S@pPAN cathodes.

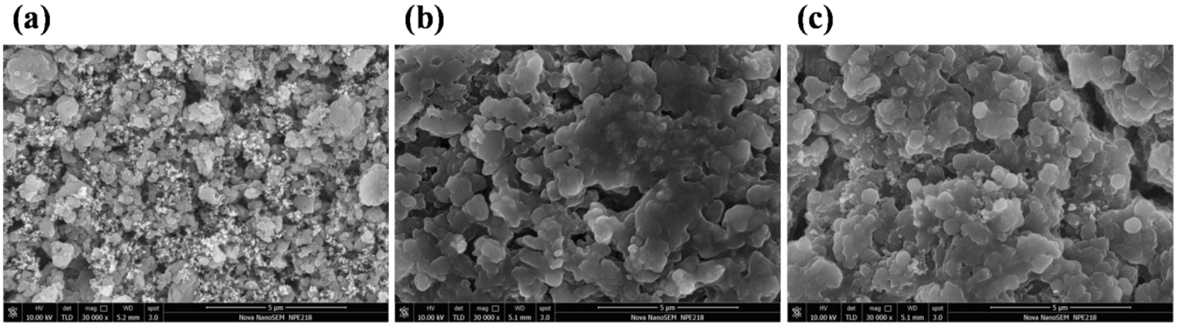

Fig. S7 SEM images of (a) fresh S@pPAN cathode, the S@pPAN cathodes at the 100th cycle in (b) blank electrolyte and (c) TMSB-containing electrolyte.