Supporting Information

Flexible Freestanding Sandwich-structured Sulfur Cathode with Superior Performance for Lithium-sulfur Batteries

*Jiangxuan Song, Zhaoxin Yu, Terrence Xu, Shuru Chen, Hiesang Sohn, Michael Regula, and Donghai Wang**

Experimental Section

Electrode fabrication

The flexible freestanding carbon membranes were prepared by roll press of Ketjen black carbon (Akzo Nobel Chemicals Inc.) and PTFE latex (PTFE, 60wt% in H_2O) composites with a mass ratio of 3:2. After drying at 80°C under vacuum, a large piece of flexible carbon membrane (e.g., 2 cm × 8 cm) was obtained. The average area mass density of carbon membrane is ~3 mg/cm^2. Ball-milled sulfur/Super P carbon black (7:3, weight ratio) composites were placed in the middle of two pieces of as-prepared carbon membranes, giving a flexible freestanding sandwich-structured sulfur cathode. The sulfur loading can be easily controlled by adjusting the dosage of middle composites and the electrodes with the sulfur loading of 1.5 mg-S/cm^2 and 4 mg-S/cm^2 were studied in this work, respectively.

The Super P-sulfur (C-S) cathodes were fabricated through conventional coating technique by following our previous reports.[S1, S2] Typically, sulfur and Super P together with PVDF binder in N-methyl-2-pyrrolidinone (NMP) were ball-milled to form homogeneous slurry. Then, the slurry was carefully coated on aluminum foil using doctor blade. Here, the weight ratio of sulfur and carbon was also controlled as

7:3 and the ratio of PVDF in whole electrode materials is 10wt%. The electrodes were dried at 60 °C for 10 h under vacuum. The C-S cathodes with sulfur loading are carefully controlled at ~1.5 mg-S/cm^2 as control samples.

Electrochemical tests:

The electrochemical performance of sandwich-structured sulfur cathodes was tested in a half-cell configuration using CR2016 type coin cells. The electrolyte was 1M Lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) dissolved in a mixture of 1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME) (1:1 v/v) with 0.2M $LiNO_3$ as additive. The separator was a microporous polypropylene membrane (25 μm thick, Celgard 2400). Cells were assembled in an argon-filled glove box and galvanostatically discharged and charged using a Landian battery tester at room temperature.

Characterization:

Scanning electron microscope (SEM) images and energy-dispersive X-ray spectroscopy (EDX) elemental scanning were taken on a NanoSEM 630. The samples were prepared by cycling the sandwich-structured cathode to the defined stage (the end of 1st discharge and the end of 1st charge, respectively). Then, they were disassembled in the argon-filled glove box and the corresponding sandwich-structured sulfur cathodes were carefully detached. Afterwards, these carbon membranes were dried under vacuum in glove box for SEM characterization.

The surface area and pore structure were characterized by nitrogen sorption using a Micrometrics ASAP 2020 physisorption analyzer. The surface area was

calculated by the Brunauer–Emmett–Teller (BET) method. The pore size distributions (Dp) were derived from the adsorption branches of isotherms using the Barrett–Joyner–Halenda (BJH) model. Electrochemical impedance spectroscopy (EIS) measurement was performed using Solartron electrochemical workstation within the frequency range of 100 kHz to 10 mHz at potentiostatic signal amplitudes of 0.1mV.

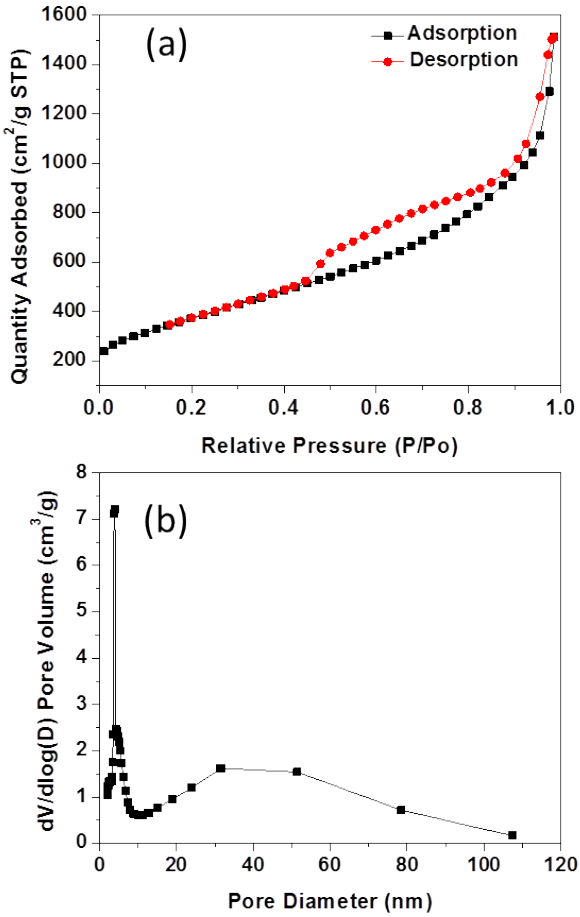

Figure S1. (a) N_2 sorption isotherms and (b) pore size distribution curve of Ketjen black carbon. The BET surface area and pore volume of Ketjen black carbon are 1326.84 m^2/g and 2.43 cm^3/g, respectively.

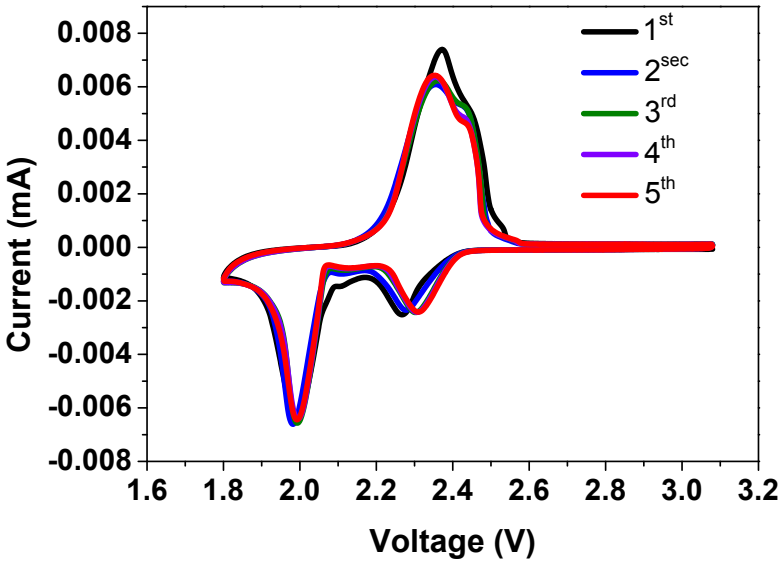

Figure S2. Cyclic voltammograms (CV) plots of sandwich-structured sulfur cathode for the first 5 cycles with a scan rate of 0.1mV/s.

Reference

S1. T. Xu, J. Song, M. L. Gordin, H. Sohn, Z. Yu, S. Chen and D. Wang, *ACS Appl. Mater. Interfaces*, 2013, **5**, 11355-11362.

S2. J. Song, T. Xu, M. L. Gordin, P. Zhu, D. Lv, Y. B. Jiang, Y. Chen, Y. Duan and D. Wang, *Adv. Funct. Mater.*, 2013, DOI: 10.1002/adfm.201302631.