# Defect-Rich Mo<sub>2</sub>S<sub>3</sub> loaded Wood-Derived Carbon Acts As A Spacer In Lithium-sulfur batteries: Forming Polysulfide Capture Net And Promoting Fast Lithium Flux

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## **Materials and Methods**

#### **Preparation of interlayer material**

Cut the wood in a direction perpendicular to the growth into small pieces with  $3 \times 3 \times 1$  cm. Then it was soaked in 0.5 M hydrochloric acid solution for 24 hours, followed by washed to neutral with deionized water. It was then heated to  $120^{\circ}$ C for 6 h in one solution contain a certain concentration of thioureone and ammonium molybdate. The wood chips were then polished on sandpaper to a size suitable for battery assembly (about 20 mm in diameter and 1 mm in height) using the high mechanical strength from the sublimated sulfur filling in its holes. The sublimated sulfur was then quickly evaporated at 200°C. Finally, the GO dispersion is filtered onto one side of the wood chips with a deposition mass of about 0.1% of the total spacer material. The pure GO@WC spacer material containing no sulfide was prepared by means of no sulfide source in the hydrothermal process.

## **Simulation and Calculation Methods**

Electric field distribution simulations were performed with COMSOL Multiphysics. The Nernst–Planck–Poisson equation was used to describe the charge and mass transport. A cylindrical geometry was used to represent the low-tortuosity channel in the WC. First-principle DFT calculations were performed with the PWmat code, the projector augmented wave potentials, and the generalized gradient approximation with Perdew–Burke–Ernzerh of functional. A plane wave basis set with a cutoff energy of 360 eV and a convergence criterion of  $10^{-5}$  eV and  $10^{-2}$  eV Å<sup>-1</sup> for energy and force during geometrical optimization were used in calculations. A vacuum space of 10 Å was used to avoid the interactions between periodic images. The Brillouin zones were sampled with  $2 \times 2 \times 1$  Monkhorst-Pack meshes. The Hubbard U (DFT+U) treatment was used for the Co transition metal with the U value of 4.0 eV.

## **Battery Assembly and Testing**

The symmetrical cell and the full cell with different interlayer materials were assembled. The battery model was 2032. The positive and negative electrodes of the symmetrical battery were both the lithium metal. And the negative electrode of the full battery was still lithium metal, except for the positive electrode was carbon black/sulfur composite, with a surface load of about 2 mg cm-<sup>2</sup>. The electrolyte is conventional ether

electrolyte (DME and DOL (v/v, 1:1)) with LiTFSI concentration of 1M, and without additives. Electrolyte dosage is 10-15  $\mu$ L per mg of active substance. Hubei Lambo charge-discharge test instrument was used to conduct charge-discharge test on the whole battery. The voltage range was 1.8-2.8 V, and the plating/stripping test of symmetrical battery was carried out in the constant current mode. CV and EIS tests were performed on CHI760e electrochemical workstation of Shanghai Chenhua company. EIS test was carried out at a frequency range of 0.1-106 Hz and a bias voltage of 10 mV.

## Characterization

The microstructure evolution was examined using a scanning electron microscope (SEM) (SUPRATM55, ZEISS, UK) equipped with an Energy Disperse Spectroscopy (EDS) at the accelerating voltage of 20 kV, and a transmission electron microscope (TEM, Tecnai<sup>TM</sup> G2 F30, FEI, USA) with an accelerating voltage of 200 kV. The phase compositions were identified by X-ray diffraction (D&A25ADVANCE, BRUKER, Germany) using Cu K $\alpha$  radiation ( $\lambda$ =0.15418 nm). X-ray photoelectron spectroscopy (XPS) (ESCALAB250Xi, Thermo Fisher) analysis was performed to identify chemical compounds of the samples. The Fourier transform infrared (FTIR) spectra were recorded on a (Thermo Fisher IS-50) spectrometer with KBr pellets.



Figure S1. SEM image of the WC (a) before and (b) after the activate of hydrochloric acid.



Figure S2. FT-IR of WC and NWC/Mo\_2S\_3 spacer



Figure S3. SEM images of NWC/Mo<sub>2</sub>S<sub>3</sub> synthesis in the solution contain the thioureone and ammonium molybdate with the ratio of 2:1, among them the concentration of the ammonium molybdate is (a) 2M, (b) 3M, (c) 4M, and (d) 5M.







Figure S5. XPS survey spectra of WC and NWC/Mo<sub>2</sub>S<sub>3</sub>.



Figure S6. High resolution XPS O1s peak of WC and NWC/Mo<sub>2</sub>S<sub>3</sub>.



Figure S7. Geometries of  $Mo_2S_3$ .



Figure S8. Simulation results of the electric field distribution of (a) WC and (b)  $NWC/Mo_2S_3$  under the discharge state.



Figure S9. The locally amplified voltage curve of the LiWCL symmetric battery under the current of (a) 2 mA cm<sup>-2</sup> and (b) 3 mA cm<sup>-2</sup>.



Figure S10. Simulation results of geometries of (a)  $Li_2S$  and (b)  $Li_2S_4$  on graphite carbon.



Figure S11. The pictures of the H-type cell with (a) WC and (b) NWC/Mo<sub>2</sub>S<sub>3</sub> spacers, the right part is Li2S6 in DEM and DOI electrolyte and the left is the blank DEM and DOI solution. (c) the corresponding UV–Vis spectra of the NWC/Mo<sub>2</sub>S<sub>3</sub> H-type cell after 48 h.



Figure S12. The pictures of (a) wood, (b) NWC/Mo<sub>2</sub>S<sub>3</sub> and (c) GO@ NWC/Mo<sub>2</sub>S<sub>3</sub>. SEM images of (d) WC and (e) GO@ NWC/Mo<sub>2</sub>S<sub>3</sub>.



Figure S13. (a) Cycling performance at 0.5C rate of the full cells equipped with  $GO@NWC@Mo_2S_3$  spacer and traditional PP separator. (b) rate capability test of the two kinds of cells.



Figure S14. Cycling performance at 0.5C rate of the full cells equipped with  $GO@NWC@Mo_2S_3$  spacer.