

Supporting Information

W₁₈O₄₉ Nanowires Composites as Novel Barrier Layers for Li-S Batteries based on High Loading of Commercial Micro-sized Sulfur

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1. Synthesis of W₁₈O₄₉ nanowires

W₁₈O₄₉ nanowires were prepared by a facile solvothermal route. In a typical procedure, 0.25 g of WCl₆ was dissolved in 50 mL of absolute ethanol. Then the solution was stirred, forming a transparent yellow solution. Subsequently, the resulting solution was transferred to a Teflon-lined stainless steel autoclave, sealed, and treated at 180 °C for 24 h. A blue precipitate was collected by centrifugation and purified with ethanol several times. Finally, the precipitate was dried in vacuum at 60 °C for 12 h.

2. Preparation of the W₁₈O₄₉ nanowire-modified glass fiber barrier layers.

The as-prepared W₁₈O₄₉ nanowires were mixed with polyvinylidene fluoride

(PVDF) binder (10% by weight) and Super P in a desired volume of N-methyl-2-pyrrolidinone to form a homogeneous slurry. The slurry was then coated onto the commercial glass fiber separator (GF/A) using doctor blade and dried at 60 °C for 12 h. Then $W_{18}O_{49}$ nanowires coated glass fiber was punched into 2 cm to form the barrier layers for polysulfides.

3. Electrode Fabrication and Electrochemical Measurements.

The sulfur cathode was prepared by conventional slurry coating. The slurry was prepared by mixing commercial micro-sized sulfur powder, Super P and PVDF at the ratio of 7:2:1 with appropriate volume of N-methylpyrrolidinone (NMP), then grinding the mixture for 5 h to be uniform. The slurry was then coated onto aluminum foil by a doctor blade and dried in a vacuum at 60 °C for 12h, followed by cutting into circular electrodes. The sulfur mass loading in cathode is ~3.0 mg (~1.9 mg/cm²). Then, 2032 type coin cells were assembled in an argon-filled glove box with H₂O and O₂ content below 1 ppm using Li metal foil as anode. The electrolyte used was a solution of 1M lithium bis(tri fluoromethanesulfonyl)imide (LiTFSI) in 1:1 (v/v) 1,3-dioxolane (DOL) and 1,2-dimethoxyethane(DME) containing LiNO₃ (1wt%). Galvanostatic charge/discharge cycling was studied with a multichannel battery testing system (LAND CT2001A). In a voltage window from 2.8 V to 1.5 V vs Li⁺/Li. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were tested with an electrochemical workstation (Autolab PGSTAT 302N) from 100 KHz to 10 mHz at the open-circuit voltage of the cells with the Li metal foil as both auxiliary and reference electrodes. Specific capacity values were calculated based on the mass of sulfur in the cathode.

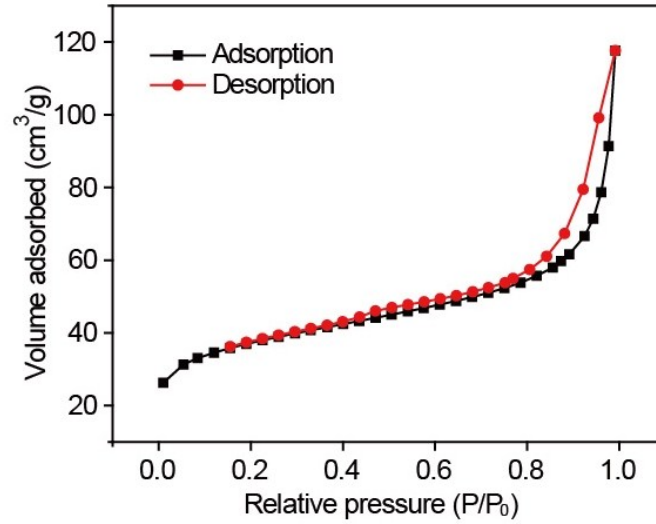


Figure S1. The N₂ adsorption-desorption isotherms of W₁₈O₄₉ nanowires.

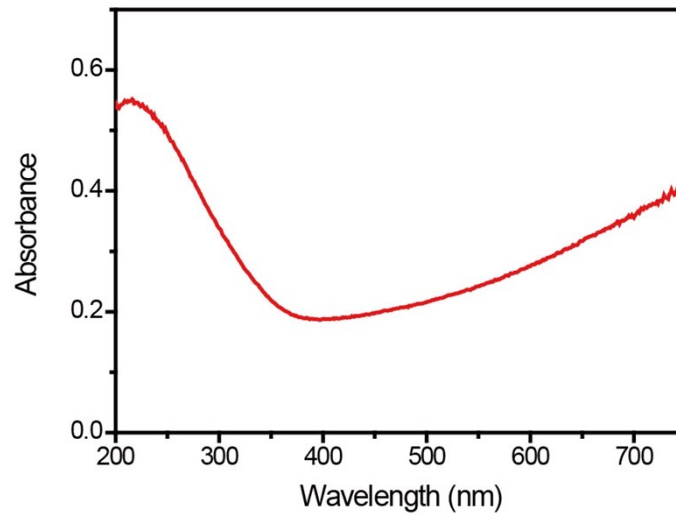


Figure S2. UV-vis absorption spectrum of W₁₈O₄₉ nanowires.

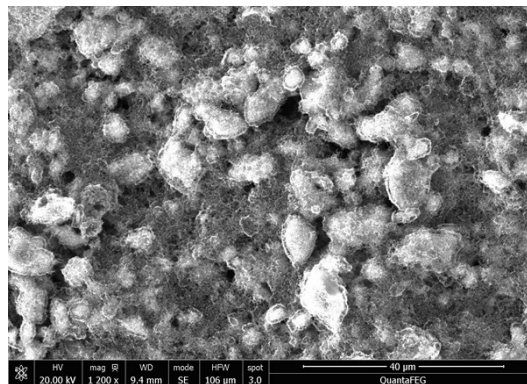


Figure S3. The SEM image of micro-sized sulfur cathode. The size of bulk sulfur without any processing is distributed in the range of 2-10 μm.

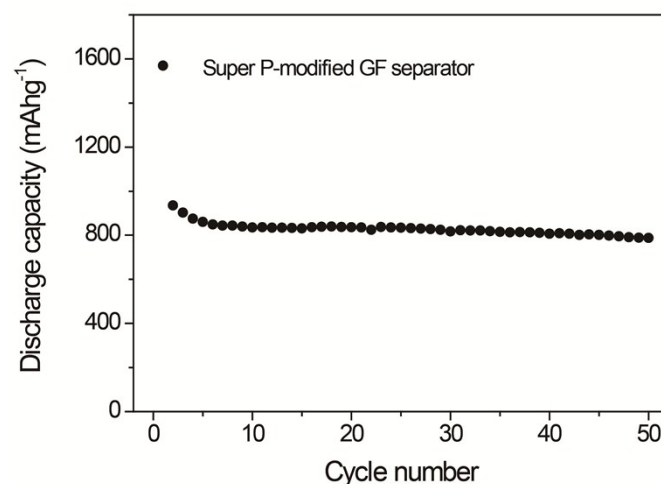


Figure S4. Cycling performance at the current density of 0.5 A g^{-1} with Super P-modified GF separator.

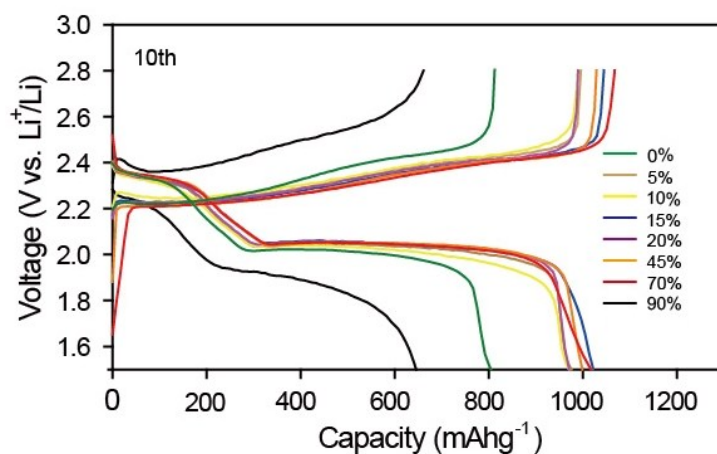


Figure S5. Galvanostatic charge-discharge profiles at the current density of 0.5 A/g with the $W_{18}O_{49}$ nanowires/Super P-modified GF separator at different mass ratios of $W_{18}O_{49}$ nanowire from 0%, 5%, 10%, 15%, 20%, 45% ,70% to 90%, respectively. At the mass ratio of $W_{18}O_{49}$ nanowire is 70%, the cell achieves the highest performance, because of the unique synergistic effect that combines excellent electronic conductivity, highly porous structure and strong polysulfides binding capacity, thus the best mass ratio of 70% is selected in the work.

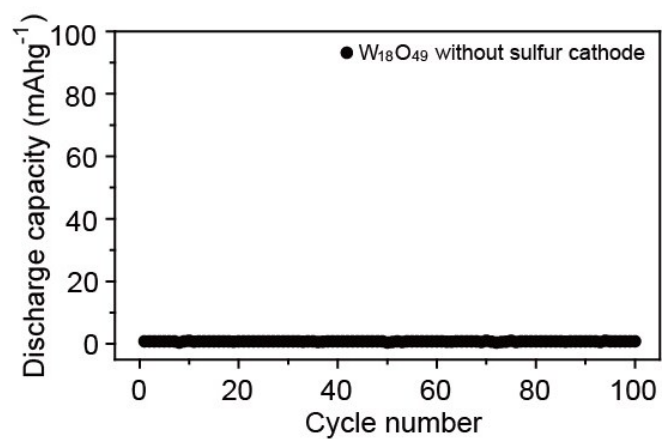


Figure S6. Cycling performance of $W_{18}O_{49}$ nanowire-modified GF separator without sulfur cathode at the current density of 0.5 A/g. The $W_{18}O_{49}$ nanowires-contributed capacity is negligible, implying that $W_{18}O_{49}$ nanowires are superior host or separator-modified materials in Li-S batteries rather than an additional lithium storage material in the operational voltage window.