Electronic Supporting Information

Rational design of coral ball-like MoS₂/N-doped carbon nanohybrids via atomic interface engineering for effective sodium/potassium storage

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Experimental Section

Materials. Hexaammonium heptamolybdate tetrahydrate ((NH₄)₆Mo₇O₂₄·4H₂O), thiourea and hexadecylpyridinium chloride monohydrate (CPC) were purchased from Sinopharm Chemical Reagent Co., Ltd. All chemical reagents were used directly without further purification.

*Synthesis of wool spherical-like MoS*₂/*N*-*C*. The hierarchical wool spherical-like MoS₂/N-C nanohybrids were prepared by a facile hydrothermal method. In a typical experiment, 0.14 mmol CPC and 0.8 mmol thiourea were dissolved in 5 ml deionized water under ultrasonic conditions. Meanwhile, 0.023 mmol (NH₄)₆Mo₇O₂₄·4H₂O was dissolved in 5 ml deionized water under ultrasonic conditions. Then the (NH₄)₆Mo₇O₂₄·4H₂O was dissolved in 5 ml deionized water under ultrasonic conditions. Then the (NH₄)₆Mo₇O₂₄ solution was added into the reaction system drop by drop to form a white translucent colloidal dispersion under vigorous stirring. Then, the suspension was transferred into a Teflon-lined stainless-steel autoclave and kept at 220 °C for 20 h. In this reaction, the sulfur to molybdenum ratio was 5: 1. After cooling to room temperature naturally, the precursor was centrifuged and washed with deionized water and ethanol several times, followed by drying at 80 °C overnight. Finally, the MoS₂/N-C nanohybrids were obtained by annealing at 800 °C for 2 h under argon flow.

For comparison, the MoS_2/N -C-4 and MoS_2/N -C-6 nanohybrids were prepared by adjusting the amount of thiourea to control the sulfur to molybdenum ratio of 4:1 and 6:1, respectively. In addition, the pristine MoS_2 sample was prepared under the same condition except without the addition of CPC. *Material Characterizations.* X-ray diffraction (XRD) patterns of the sample were recorded on a Rigaku smartlab 3kw X-Ray Diffraction. Raman spectra were carried out on Horiba LabRAM HR800 Raman spectrometer. Fourier transform infrared spectra (FTIR) were recorded with a Nicolet NEXUS 670 spectrophotometer. The morphologies of the samples were observed using a Carl Zeiss G300 FE-SEM System. The microstructures and lattice fringe were observed with a transmission electron microscope (TEM, JEOL H-7000) and high-resolution transmission electron microscope (HRTEM, JEOL JEM-2100). Elemental analysis was investigated using ESCALAB 250 X-ray photoelectron spectrometer (XPS, ThermoFisher Scientific). Thermogravimetric (TG) analysis was carried out by using the NETZSCH STA 449 C in the air with a heating rate of 10 °C min⁻¹. The specific surface area (SSA) was analyzed with the Brunauer-Emmett-Teller (BET) method by an accelerated surface area and porosimeter 2000 analyzer.

Electrochemical measurements. The electrochemical performances were tested in the 2032-type coin cells. The working electrode was prepared by mixing 70 wt% active materials, 15 wt% of acetylene black, and 15 wt% of carboxymethylcellulose sodium (CMC) as binder coated on a copper foil. The loading mass of the electrode was about 1.0 mg cm⁻². For sodium-ion batteries (SIBs), the electrolyte is 1.0 M NaClO₄ dissolved in a mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) (1: 1 vol%) containing 5 wt% fluoroethylene carbonate (FEC), while for potassium-ion batteries (PIBs), the electrolyte is 1.0 M KFSI in EC/DEC (1: 1 vol%). Fresh sodium and potassium metal disk were used as counter electrodes for SIBs and PIBs, respectively,

and glass fiber served as the separator. The whole cells assembly process was performed in a glove box filled with Ar. Cyclic voltammetry (CV) tests were carried out between 0.01 V and 3 V with a scan rate of 0.1 mV s⁻¹ using a CHI760E electrochemical working station. The galvanostatic charge-discharge tests were performed on a LAND CT2001A battery testing system between cutoff voltages of 3 V and 0.01 V at room temperature. Electrochemical impedance spectroscopy (EIS) patterns were collected at CHI760E between 100 kHz to 0.01 Hz with an amplitude of 5 mV.

For the Na-ion full cells, the cathode was assembled by mixing 80 wt% homemade $Na_3V_2(PO_4)_3/C$, 10 wt% acetylene black, and 10 wt% poly(vinylidene fluoride) (PVDF) as binder coated on an aluminum foil. The mass ratio of anode/cathode was about 1: 3.8. Meanwhile, the anode was electrochemically activated for five cycles at 0.1 A g⁻¹ before it was assembled in full cells.



Figure S1. (a) TEM and (b) HRTEM images of MoS₂/CPC precursor.



Figure S2. (a) TEM and (b) HRTEM images of pristine MoS₂ sample.



Figure S3. (a) XRD pattern and (b) Raman spectrum of pristine MoS_2 sample.



Figure S4. FTIR spectra of MoS₂/CPC precursor and MoS₂/NC nanohybrids.

In the FTIR spectrum of MoS_2/CPC precursor, the C-H stretching vibrations at 2,800-3,000 cm⁻¹, C=N group at 1645 cm⁻¹, and N-H mode at 1,420 cm⁻¹ indicate that the MoS_2 nanosheets were capped with CPC molecules.



Figure S5. Pore size distributions of MoS_2/CPC precursor and MoS_2/NC nanohybrids.



Figure S6. (a) XRD patterns and (b) Raman spectra of $MoS_2/NC-4$ and $MoS_2/NC-6$ nanohybrids; (c) TEM image of $MoS_2/NC-4$ nanohybrids and (d) TEM image of $MoS_2/NC-6$ nanohybrids.



Figure S7. For SIBs: Discharge-charge voltage profiles of MoS₂/CPC electrode at 0.1

A g^{-1} .



Figure S8. For SIBs: Cycling performance of MoS_2/CPC electrode at 1 A g^{-1} .



Figure S9. For SIBs: Cycling performance of pristine MoS_2 electrode at 1 A g^{-1} .



Figure S10. For SIBs: CV curves at the scan rate of 0.1 mV s⁻¹ of MoS₂/N-C and NVP/C.



Figure S11. For SIBs: CV curves of MoS_2/NC electrode with capacitive contribution

in the shaded region at different scan rates.



Figure S12. For SIBs: (a) CV curves of MoS_2/CPC electrode at different scan rates; (b) Log (*i*) vs. log (*v*) plots calculated from CV curves of MoS_2/CPC electrode; (c) Capacitive contribution ratio of MoS_2/CPC electrode at different rates; (d) The CV curve of MoS_2/CPC electrode with capacitive contribution in the shaded region at a scan rate of 1.0 mV s⁻¹.



Figure S13. For PIBs: CV curves of MoS₂/NC electrode with capacitive contribution

in the shaded region at different scan rates.



Figure S14. For PIBs: EIS spectrum of MoS₂/NC electrode.



Figure S15. For SIBs: (a, b) TEM and (c) SEM-EDX mapping images of $MoS_2/N-C$ electrode after 100 cycles at 1 A g⁻¹.



Figure S16. For PIBs: (a, b) TEM and (c) SEM-EDX mapping images of $MoS_2/N-C$

electrode after 100 cycles at 1 A g^{-1} .