Supporting Information

Rationally designing a $Ti_3C_2T_x/CNTs-Co_9S_8$ heterostructure as sulfur host with multi-functionality for high-performance lithium-sulfur batteries

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1. Characterizations

The samples' morphology and structure are analyzed using a scanning electron microscope (SEM, HITACHI SU 3500) and transmission electron microscope (TEM, Talos F200X). The element distribution of the samples is characterized by energy-dispersive X-ray spectroscopy (EDX) attached to TEM. The crystal structures of the samples are analyzed by X-ray diffraction (XRD, AXS D8 ADVANCE). The chemical compositions of the samples are characterized by X-ray photoelectron spectroscopy (XPS, K-ALPHA). The nitrogen adsorption-desorption isotherms are performed on the N₂ adsorption-desorption instrument (ASAP 2460, Micrometrics) to characterize the pore structures of the samples. The sulfur contents in the sulfur composite cathodes are determined by thermos-gravimetric analysis (TGA, STA449F3).

2. Electrochemical Measurements

The active material (*e.g.*, $Ti_3C_2T_x/CNTs-Co_9S_8/S$), CNTs conductive additive, and PVDF binder are mixed with a mass ratio of 7:2:1 and dispersed in N-methyl pyrrolidone (NMP) solvent to obtain the electrode slurry. Then the slurry is uniformly coated on the aluminum foil using a scraper and then vacuum dried at 60 °C for 12 h to obtain the working electrode. The sulfur mass loading of the electrode is 0.8 or 2.5 mg cm⁻². The coin cells (CR 2032) are assembled in an Ar-filled glovebox by using lithium foil as an anode, Celgard 2400 polypropylene membrane as a separator, and 1.0 M LiTFSI in DOL (1,3-dioxolane) and DME (dimethoxyethane) (1:1 vol%) with 1.0 wt% LiNO₃ additive as electrolyte. The added electrolyte to sulfur ratio is 20 μ L mg⁻¹ in the cells. The galvanostatic charge-discharge test is conducted between 1.5 and 2.8 V on the LAND CT2001A test system. The CV curves are tested by the Chenhua CHI760E electrochemical workstation. Electrochemical impedance spectra (EIS) are performed on the electrochemical workstation (GAMRY REFERENCE 3000).



Figure S1. (a) N₂ adsorption-desorption isotherms of the 3D porous $Ti_3C_2T_x/CNTs-Co_9S_8$ and $Ti_3C_2T_x$ MXene. (b) BJH pore size distributions of the 3D porous $Ti_3C_2T_x/CNTs-Co_9S_8$, the inset figure shows the specific distributions of pore size in the range of 0-10 nm.



Figure S2. TGA curves under the N₂ atmosphere of (a) the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ composite and (b) the $Ti_3C_2T_x/S$ composite.



Figure S3. The cross-sectional of the $Ti_3C_2T_x/CNTs\text{-}Co_9S_8/S$ cathode.



Figure S4. The optical pictures of the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ cathode before (left) and after 200 cycles (right).



Figure S5. (a-b) The post-mortem SEM image of the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ cathode after 200 cycles. (c) The cross-section SEM image of the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ cathode.



Figure S6. The post-mortem XPS spectra of the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ cathode after cycling: (a) Ti 2p spectrum and (b) S 2p spectrum.



Figure S7. (a) EIS spectra of the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ and $Ti_3C_2T_x/S$ cathodes. (b-c) EIS spectra of the $Ti_3C_2T_x/CNTs-Co_9S_8/S$ cathode at different voltages during the discharging and charging processes.