Electronic Supplementary Information

Lattice Expansion of MnO Induced by Sulphur Doping for Enhanced Aqueous Zinc-Ion Diffusion and Storage

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Material synthesis

1.1 MnO

First, 1225 mg of $Mn(CH_3COO)_2 \cdot 4H_2O$ and 300 mg of PVP were uniformly dissolved in a mixture of ethanol/water (125/125 mL), labeled as solution **A**. Then, 1350 mg of H₃BTC was uniformly dissolved in a mixture of ethanol/water (125/125 mL), labeled as solution **B**. Solution **B** was slowly added dropwise to solution **A** and stirred continuously for 30 min to obtain solution **C**. Solution **C** was left to stand for 24 h at 60 °C, centrifuged, washed, and dried to obtain Mn-BTC precursor. The Mn-BTC precursor was then heated to 700 °C in an argon atmosphere at a heating rate of 5 °C min⁻¹ for 3 h to obtain MnO material.

1.2 MnOS_{0.3}

After the synthesis of MnO, a certain mass of sulfur powder was weighed based on the molar ratio and mixed with the MnO powder. The mixture was ground for 30 min. Then, the mixed powder was placed in a tube furnace and heated to 400 °C in an argon atmosphere at a heating rate of 5 °C min⁻¹ for 2 hours to obtain MnOS_{0.3} material.

1.3 MnOS_{0.3}@rGO

During the synthesis of the Mn-BTC precursor, 100 mg of GO was added to solution **A** and stirred continuously for 30 min to obtain solution **C**. Solution **C** was left to stand for 24 h at 60 °C, centrifuged, washed, and dried to obtain Mn-BTC@GO precursor. The Mn-BTC@GO precursor was then heated to 700 °C in an argon

atmosphere at a heating rate of 5 °C min⁻¹ for 3 h to obtain MnO@rGO material. After the synthesis of MnO@rGO, a certain mass of sulfur powder was weighed based on the molar ratio and mixed with the MnO@rGO powder. The mixture was ground for 30 min. Then, the mixed powder was placed in a tube furnace and heated to 400 °C in an argon atmosphere at a heating rate of 5 °C min⁻¹ for 2 hours to obtain MnOS_{0.3}@rGO material.

Equipment used for testing and testing conditions:

Morphological and microstructural information of the samples were obtained using a field emission scanning microscope (FESEM, JSM-IT800, Japan), and transmission electron microscope (TEM, FEI Tecnai G2 F20, America). The elemental distributions were recorded using an energy-dispersive spectrometer (EDS) connected to a TEM (Hitachi HF5000, Japan). The crystal structure information of samples was obtained by X-ray diffractometer (XRD, Bruker D8A A25, Germany) with a Cu Ka source (λ =1.5418 Å). The chemical composition and electronic structure of the samples were analyzed by X-ray photoelectron spectroscopy (XSP, Thermo Scientific ESCALAB Xi+, America) with a monochromatic Al anode X-ray source. The elemental content of the samples was measured by an elemental analyzer (Elementar Unicube, Germany). LANHE CT3002A battery test systems (Wuhan, China). CHI 760E electrochemical workstation. Horiba LabRAM Evolution Laser (Raman), wavelength: 532nm (2.33eV) Acquisition time: 10s Accumulation: 1. The nitrogen adsorption-desorption isotherm curves of the samples were measured by Autosorb-iQ automatic specific surface and pore size distribution analyzer (Quantachrome, America). The Quenched Solid Density Functional Theory (QSDFT) mode was chosen to analyze the pore diameter, pore type, and pore volume of the samples. TG2 (GC301 METTLER TOLEDO).



Fig. S1 SEM and enlarged SEM images of MnO.



Fig. S2 Elemental mapping images of $MnOS_{0.3}$.



Fig. S3 XRD patterns of MnO, MnOS_{0.3}, and MnOS_{0.3}@rGO.



Fig. S4 Nitrogen sorption isotherms and pore size distribution of MnO, $MnOS_{0.3}$ and $MnOS_{0.3}@rGO$.



Fig. 5 TG curve of the precursor in O_2 atmosphere.



Fig. 6 (a) MnO CV curves. (Scan rate 0.1 mV s⁻¹) (b) MnOS_{0.3} CV curves. (Scan rate 0.1 mV s⁻¹) (c) MnOS_{0.3}@rGO CV curves. (Scan rate 0.1 mV s⁻¹)



Fig. 7 Cycling performance at a current density of 0.5 A g^{-1} for the MnOS_{0.3} electrodes.



Fig. 8 MnO curves measured at different scan rates from 0.2 to 0.8 mV s⁻¹; *b* value according to the relationship of log(i) and log(v) at different peaks; Pseudocapacitance contribution at different scanning speeds.



Fig. 9 MnOS_{0.3}@rGO curves measured at different scan rates from 0.2 to 0.8 mV s^{-1} ; *b* value according to the relationship of log(i) and log(v) at different peaks; Pseudocapacitance contribution at different scanning speeds.