Electronic Supplementary Information

Synthesis of Hierarchical ZnO/ZnCo₂O₄ Nanosheets with Mesostructures for

Lithium-Ion Anodes

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Synthesis of bi-component-active hierarchical ZnO/ZnCo₂O₄ nanosheets

All the chemicals used in this study are analytic-grade reagents and used after purchase without further purification. $0.329g Zn(CH_3COO)_2 \cdot 2H_2O$, $0.75g Co(CH_3COO)_2 \cdot 4H_2O$ and 0.45g hexamethylene tetramine is dissolved in 10 mL deionized water under vigorous stirring, and then, added 10 mL ethanolamine to the mixture under stirring for 1 hours at room temperature. Then the mixture is transferred into a 30 ml Teflon-sealed autoclave and maintained at 180 °C for 24 h. After the hydrothermally-treated solution is cooled to room temperature, the obtained products are centrifuged and washed with deionized water and ethanol at least 5 times separately, dried at 60 °C overnight and annealed at 400 °C for 120 min in air, resulting in the final product (ZZCO).

Synthesis of ZnCo2O4 nanosheets

 $0.329g Zn(CH_3COO)_2 \cdot 2H_2O$, $0.75g Co(CH_3COO)_2 \cdot 4H_2O$ and 0.45g hexamethylene tetramine is dissolved in 10 mL deionized water under vigorous stirring, and then, added 10 mL ethylene glycol to the mixture under stirring for 1 hours at room temperature. Then the mixture is transferred into a 30 ml Teflon-sealed autoclave and maintained at 210 °C for 24 h. After the hydrothermally-treated solution is cooled to room temperature, the obtained products are centrifuged and washed with deionized water and ethanol at least 5 times separately, dried at 60 °C overnight and annealed at 400 °C for 120 min in air, resulting in the final product (ZCO).

Materials Characterization

The obtained samples are characterized by X-ray diffraction (XRD, Rigaku D/Max III diffractometer with Cu K α -radiation, λ = 1.5418 Å), scanning electron microscopy (SEM, Nova Nano SEM 230), transmission electron microscopy (TEM, Tecnai G²F20, FEI), high-resolution TEM (HRTEM, Tecnai G²F20, FEI).

Electrochemical Measurements

For electrochemical tests, the working electrodes are prepared with active materials, acetylene black (AB), and polytetrafluoroethylene (PTFE) at the weight ratio of 75: 15: 10. The average weight of the electrodes is ~2 mg. In the test cells, lithium metal is used as the counter and reference electrode. The electrolyte is 1 M LiPF₆ dissolved in a 1: 1: 1 mixture of ethylene carbonate (EC), ethylene methyl carbonate (EMC) and dimethyl carbonate (DMC). The cells are assembled in a glove box filled with high-purity argon. The galvanostatic charge and discharge tests are performed with a battery tester LAND-CT2001A in the voltage range of 0.01–3.0 V at room temperature. Electrochemical impedance spectroscopy (EIS) is taken by using an IM6e electrochemical workstation at 25 °C with the frequency range from 10 kHz to 100 mHz and an AC signal of 5 mV in amplitude as the perturbation. The specific capacity is calculated according to the corresponding total weight of active materials in each electrode.



Figure S1. FESEM images of the as-prepared ZZCO materials.



Figure S2. XRD pattern of the as-prepared ZCO materials.



Figure S3. SEM images of the as-prepared ZCO materials.



Figure S4. N₂-sorption isotherms (a), pore-size distribution (b) of the as-prepared ZZCO materials.