Supporting Information

Flower-like W/WO₃ as Novel Cathode for Aqueous Zinc-Ion Batteries

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Experimental

Preparation of flower-like W/WO₃

The fabrication of flower-like W/WO₃ only includes a heat treatment process for pure tungsten powder (Xianglu Tungsten Co. Ltd., China). Typically, 1 g tungsten powder was directly calcined in muffle furnace to 550 °C with a heating rate of 5 °C min⁻¹ and then hold for 1 h. WO₃ was synthesized by heating tungsten powder in muffle furnace at 700 °C with a heating rate of 5 °C min⁻¹ and then hold for 1 h.

Characterization

The morphology of as-prepared materials was observed via scanning electron microscopy (SEM. Hitachi S4800) and the transmission electron microscopy (TEM, JEM-1400). And the phase structures and chemical composition of materials were investigated by X-ray diffraction (XRD, Rigaku D) and X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB).

Electrochemical performance

The electrochemical performance of the as-prepared materials was investigated by assembling the coin cells (CR2032) with zinc foil anode. And the cathodes were prepared via mixing the materials, conductive carbon and PVDF at the mass ratio of 8:1:1, and then coating the slurry on graphite paper. The average mass loading of the active materials was about 1.1 mg cm⁻². After drying under vacuum at 80 °Cs for 12 h, cutting into round electrode pieces with a diameter of 14 mm. The 2 M ZnSO₄ solution was served as electrolyte. The galvanostatic charge/discharge (GCD) measurement was performed to identify the rate performance and cycling stability (BTS, CT-4008,

Neware, China) with a potential range of 0.1-1.2 V. The cyclic voltammetry (CV) profiles and electrochemical impedance spectroscopy (EIS) (0.01 Hz to 10 kHz) of the materials were recorded on a CHI 660E workstation (China).

Details of the Calculation

Capacitance Contribution

According to the reported literatures, [1-2] the measure current (*i*) and scan rate (*v*) in CV curves have relationships with equation:

$$i = av^b \tag{1}$$

$$lg(i) = blg(v) + lg(a) \tag{2}$$

where *i* represents peak current, *a* denotes constant, *v* is the scan rate and *b* is equaled to the slope of the lg(v)-lg(i) plots. If *b* approximately equals to 0.5, suggesting the diffusion process is dominant, while a capacitive behavior will dominate when b is about 1. ^[3]

The capacitance contribution can be further measured according to equations (3) and (4):

$$i(v) = k_1 v + k_2 v^{1/2} \tag{3}$$

$$i(v)/v^{1/2} = k_1 v^{1/2} + k_2 \tag{4}$$

Where the capacitive contribution can be divided into capacitive (measured with k_1) and diffusion-controlled (measured with k_2). By determining values of both k_1 and k_2 , we can distinguish the fraction of the current from surface capacitance and diffusion limited. ^[4]

Zn²⁺ Diffusion Coefficient

The diffusion coefficient of Zn^{2+} was measured by using GITT and calculated based on the following equation: ^[5-7]

$$D = \frac{4L^2 \Delta E_S}{\pi \tau} (\Delta E_t)^2$$
(5)

Where t and τ represent the duration of current pulse (s) and relaxation time (s), respectively. L corresponds to Zn²⁺ diffusion length, which is equal to thickness of electrode. ΔE_s and ΔE_t are the steady-state voltage change (V) by the current pulse and voltage change (V) during the constant current pulse (eliminating the voltage changes after relaxation time).

Energy Density and Power Density

For coin cell, the energy density and power density were obtained by the following equations (6) (7):

$$E = \int_{0}^{M} \frac{V \times i}{m} dt \tag{6}$$

$$P = \frac{L}{1000 \times \Delta t} \tag{7}$$

where E (Wh kg⁻¹) is the energy density, P is the power density (kW kg⁻¹), V (V), i(mA), m (g) and $\triangle t$ (h) represent the working potential, discharging current, the mass loading of the cathode and the discharging time, respectively.



Figure S1 Quantitative analysis of the W/WO₃.



Figure S2 (a) (b) (c) SEM images of pure W.



Figure S3 EDS result of W/WO₃.



Figure S4 XPS survey of W/WO₃.



Figure S5 SEM image of W/WO₃ after cycling.

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