A Novel Aqueous Zinc-ion Battery Capable of Self-charging at Low Temperature

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Experimental section

1. Materials Synthesis

 $FeVO_4$ nH₂O was synthesized using a hydrothermal method. 20 milli liters (mL) of DI H₂O were used to dissolve 2 milli molar (2 mmol) FeCl₃, 2 mmol NH₄VO₃, and 4 mmol H₂C₂O₄ while stirring for 15 minutes. 3 mmol Hexamethylenetetramine was then added to the solution dropwise. The mixture was transferred to a Teflon-lined autoclave and heated at 150°C in the furnace for 6 hours after. The precipitate was collected and cleaned three times with DI H₂O. Then it was dried in air at 55-60°C. The as-obtained FeVO₄ nH₂O was then ground into a powder form.

2. Materials Characterizations

X-ray diffraction (XRD) was performed using Cu K α radiation (λ = 1.5405) on a Rigaku MiniFlex X-ray diffractometer at a scanning rate of 2 degree/min. FEI Quanta 3D FEG field emission scanning electron microscope (FESEM) was used to acquire scanning electron microscopy (SEM) images. An AXIS165 XPS spectrometer was employed to perform the X-ray Photoelectron Spectroscopy (XPS) measurements. Transmission electron microscopy (TEM) was conducted using a JEM-1400 TEM. Additionally, the thermogravimetric analysis (TGA) data were obtained by utilizing an SII STA7300 analyzer in a nitrogen atmosphere. The Brunauer-Emmett-Teller (BET) method was conducted to determine the surface area of the FeVO₄·H₂O powders.

3. Electrochemical Characterizations

2032-coin cells were used for the fabrication of full zinc-ion batteries. These cells were assembled using a base, anode, separator, electrolyte, cathode, spacer and spring under ambient condition. The working electrode (cathode film) was synthesized by mixing, scraping and rolled in thin films using 60% (by weight) active material (FeVO₄·nH₂O), 30% (by weight) conductive carbon, and 10% polytetrafluoroethylene (PTFE) binder. The electrolyte was an aqueous solution containing 1 m Zn (CF₃SO₃)₂ and 5.16 m LiTFSI. The anode was commercial zinc foil and the separator was glass fiber membrane. Battery analyzers (MTI corporation) was used for galvanostatic charge-discharge experiments. An electrochemical workstation (CHI 6504C) was used for cyclic voltammetry measurements. The battery was completely drained to 0.1 V at 0.2 A g-1 by the battery analyzer before the chemically self-charging process. The cell was then disassembled, and the discharged cathode was cleaned by DI water and then dipped in the electrolyte to let the cathode be oxidized by dissolved O₂ for various time periods. After certain hours, the cell was assembled again and retested for the open circuit potential and discharge capacity at 0.2 A g⁻¹. The self-charging process and measurement at 0°C was carried out in a

freezer with temperature control. The mass of active material (FeVO₄ \cdot nH₂O cathode) was used for calculating specific capacity.



Figure S1. SEM images of (a) Pristine FVO cathode film and (b) FVO cathode film after 100 charge-discharge cycles.



Figure S2. XRD pattern of FVO cathode film after 100 cycles.