

Supporting Information

Zn/V₂O₅ Aqueous Hybrid-Ion Battery with High Voltage Platform and Long Cycle Life

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Experimental Section

Synthesis of porous V₂O₅ microplates

In a typical experiment, NH₄VO₃ (0.1 mmol) and urea (2.5 mmol) were dissolved in deionized water (10 mL) under vigorous stirring at 80 °C. After stirring for 20 min, 50 mL acetonitrile was added to the above solution and vigorously stirred for 1 h at 80 °C, and then the color of the solution gradually turned into dark blue. After that, the solution was transferred into a 100 mL autoclave and heated at 180 °C for 12 h. After washing and drying, a brownish powder was obtained. Finally, the powder was calcined in air at 350 °C for 2 h with a heating rate of 5 °C min⁻¹ to obtain porous V₂O₅ microplates.

Characterization

In/Ex-situ X-ray diffraction (XRD) experiments during electrochemical measurement of the Zn/V₂O₅ aqueous hybrid-ion batteries were performed on a D8 Discover X-ray diffractometer with a nonmonochromated Cu K α X-ray source (2 θ range: 15–38°). An *in-situ* battery was designed with a Be window for X-ray penetration. The cathode material was prepared with 50 wt.% active material, 40 wt.% acetylene black, and 10 wt.% polytetrafluoroethylene (PTFE). Zn(CF₃SO₃)₂-LITFSI (1 m Zn(CF₃SO₃)₂ and 21 m LITFSI) was used as the electrolyte, and Celgard 2400 microporous membrane was used as the separator.

XRD measurements were performed to investigate crystallographic information using a D8 Discover X-ray diffractometer with a non-monochromated Cu K α X-ray source. Field-emission scanning electron microscopic (FESEM) images were collected using a JSM-7001F microscope at an acceleration voltage of 20 kV. Transmission electron microscopy (TEM) images and high-resolution TEM

(HRTEM) images were recorded by using a JEM-2100F STEM/EDS microscope. X-ray photoelectron (XPS) spectra were recorded on a Shimadzu Axis Ultra spectrometer with Mg K α =1253.6 eV excitation source.

Electrochemical characterization

The cathode was fabricated by pressing a mixture of 70 wt.% active material, 20 wt.% acetylene black, and 10 wt.% polyvinylidene fluoride onto a titanium foil and dried under vacuum at 70 °C for 12 h. Metallic Zn foil with a thickness of 0.25 mm was used as the anode. 1 m Zn(CF₃SO₃)₂ (1 M Zn(CF₃SO₃)₂ was dissolved in 1 Kg deionized water) or Zn(CF₃SO₃)₂-LITFSI (1 m Zn(CF₃SO₃)₂ and 21 m LITFSI) were used as the electrolytes. 2016-type coin cells were assembled in the air by sandwiching a filter paper (Whatman grade) filled with the electrolyte between the cathode and zinc foil anode. Galvanostatic charge/discharge cycling was studied over a potential range of 0.2–1.6 V *vs.* Zn²⁺/Zn with a multichannel battery testing system (LAND CT2001A). Cyclic voltammetry (CV) and electrochemical impedance spectra (EIS) were tested with electrochemical workstations (Autolab PGSTAT 302N). AC impedance spectra were tested with Autolab 302N Probe Station (Lake Shore, TTPX).

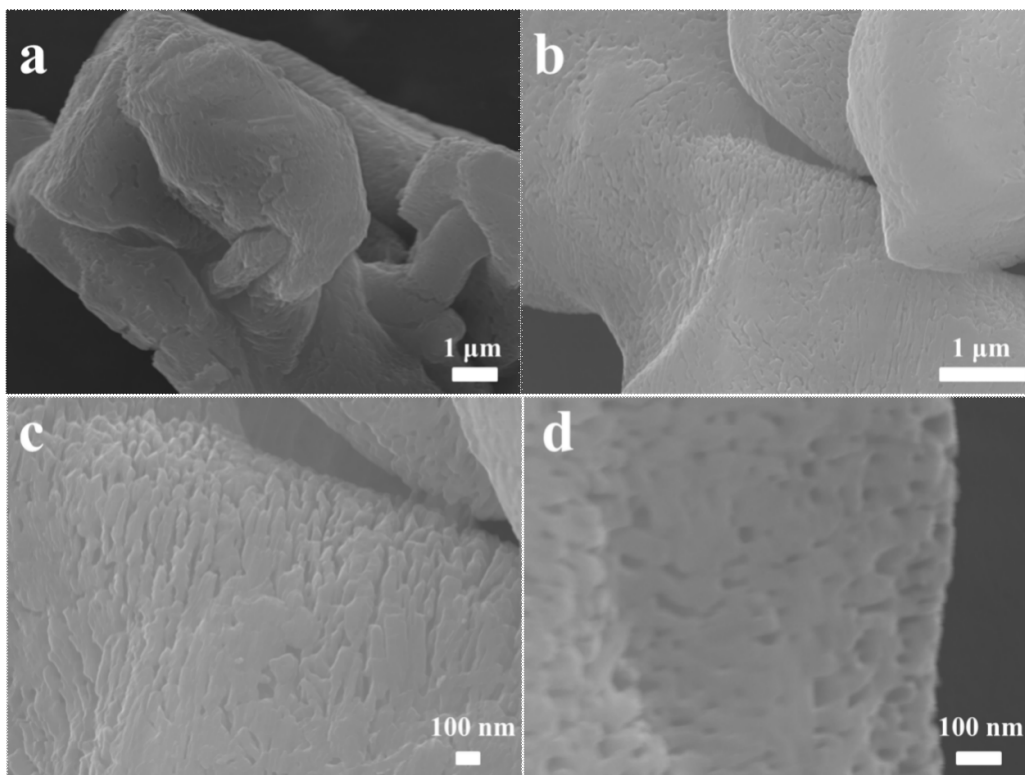


Figure S1. (a – d) The HRSEM images of porous V_2O_5 .

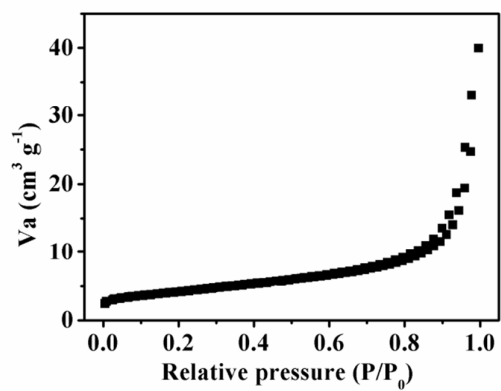


Figure S2. Nitrogen adsorption-desorption isotherm of 3D porous V_2O_5 .

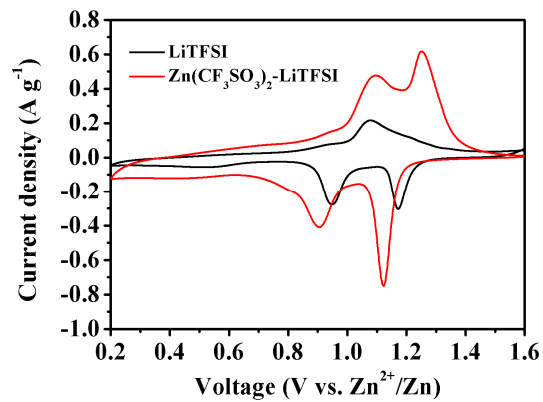


Figure S3. The CV curves of the V_2O_5 in LiTFSI and $Zn(CF_3SO_3)_2$ -LiTFSI at the scan rate of 0.1 mV s^{-1} .

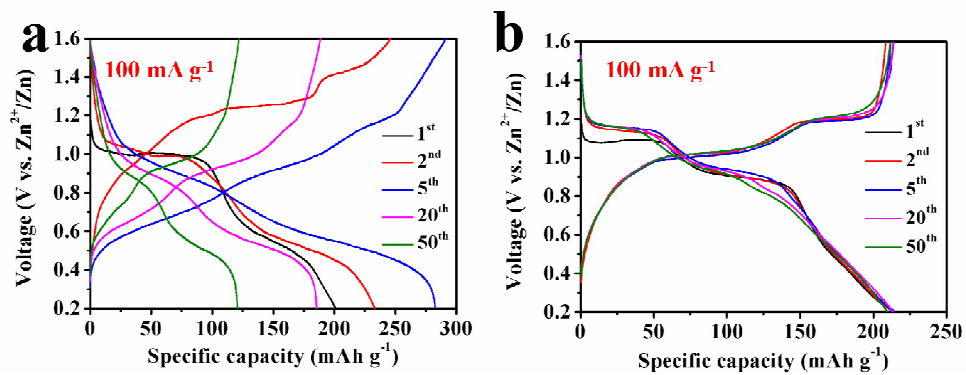


Figure S4. Charge/discharge curves at the different cycles of Zn/ V_2O_5 batteries with different electrolytes: (a) $Zn(CF_3SO_3)_2$, (b) $Zn(CF_3SO_3)_2$ -LiTFSI.

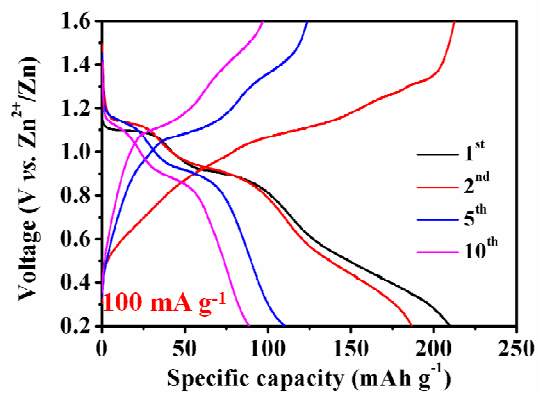


Figure S5. Charge/discharge curves of Zn/V₂O₅ batteries with 1 M LiTFSI electrolyte at different cycles.

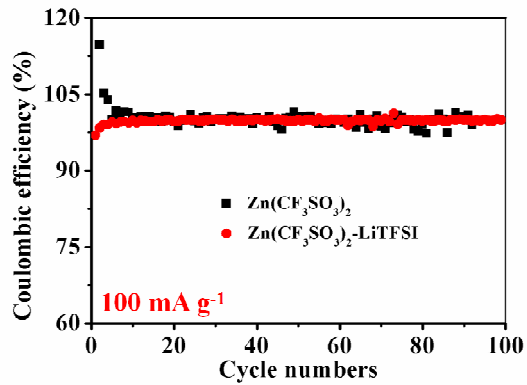


Figure S6. The coulombic efficiency of the V₂O₅ cathode in Zn(CF₃SO₃)₂ and Zn(CF₃SO₃)₂-LiTFSI.

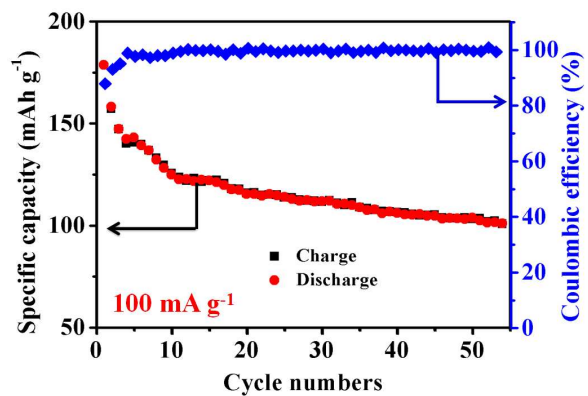


Figure S7. The cycling performance and coulombic efficiency of the V₂O₅ cathode in 21 m LiTFSI electrolyte.

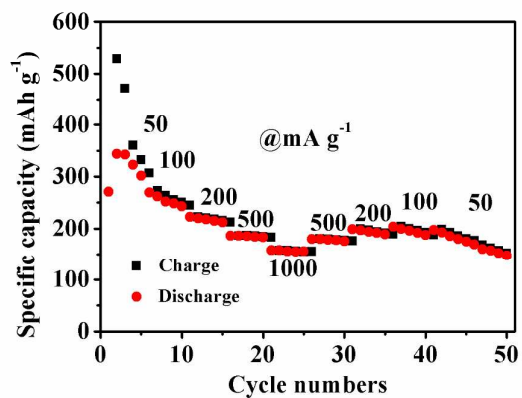


Figure S8. Rate performance of V₂O₅ in Zn(CF₃SO₃)₂ electrolyte.

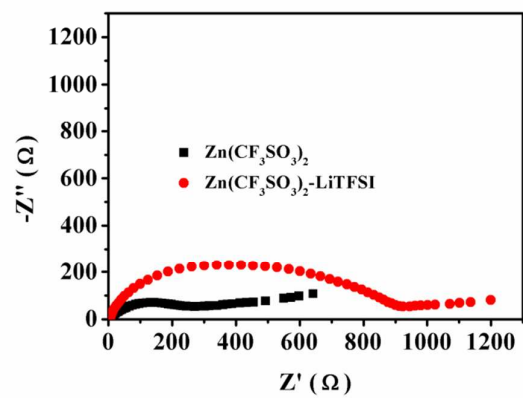


Figure S9. AC impedance spectra of the V_2O_5 electrode in the electrolytes of $Zn(CF_3SO_3)_2$ and $Zn(CF_3SO_3)_2$ -LiTFSI.