Supporting information

A high-capacity polyaniline-intercalated layered vanadium oxide for aqueous ammonium-ion battery

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Experimental

Materials Syntheses

Syntheses of polyaniline intercalated V_2O_5 (PVO): The PVO was prepared through one-step hydrothermal. In a typical procedure[1], 0.1814 g V_2O_5 powder were added to 30 mL Deionized (DI) water during violently stirring in room conditions. Then mixing 60 μL aniline to the above continuously stirred suspension dropwise. Subsequently, the pH of the mixture was adjusted to 3 by dropping 3 M HCl solution. After stirring at room temperature for 30 minutes, a dark green cloudy suspension was obtained. The above suspension was transferred to a 50 mL Teflon lined stainless steel autoclave and reacted at 120 ℃ for 24 hours. Next, the autoclave was taken out and cooled down at room temperature. After separating the dark green product by centrifugation (10000 r/min, 1 min), the product was washed with deionized water and ethanol three times each. The cleaned product was put into a 70 ℃ vacuum for 24 h, and finally a PVO powder was achieved.

Syntheses of water intercalated V_2O_5 (HVO)[2]: 1mmol V_2O_5 and 2 ml H_2O_2 (30%) were stirred and mixed in 25 mL deionized water for 1 hour. The resulting tawny transparent solution was then transferred to a 50 ml Teflon lined stainless steel autoclave and placed in an oven at 200 ℃ for 48 hours. After separating the brown product by centrifugation (10000 r/min,1 min), the product was washed with deionized water and ethanol three times each. The cleaned product was put into a 70 ℃ vacuum for 24 h, and finally a HVO powder was achieved.

Syntheses of V_2O_5 (VO): PVO powder was annealed in muffle furnace at 600 ℃ for 2 hours to obtain VO.

The organic solid of 3,4,9,10-Perylenetetracarboxylic diimide (PTCDI) was a commercially available pigment and was purchased from Alfa Aesar.

Materials characterization

The phase structure of as-prepared materials was conducted by X-Ray Diffraction (XRD, D8 Advance, Bruker AXS GmbH, Karlsruhe, Germany) with a Cu Kα X-ray source. The structure and surface morphology of the materials was demonstrated by scanning electron microscopy (SEM, JEOL-7100F microscope) and transmission electron microscopy (TEM) paired with energy dispersive spectrometry (EDS) elemental mapping (JEM-2100F STEM/EDS microscope). FTIR and Raman were used to research the chemical bonds of samples. Energy change during charging and discharging was studied via X-ray Photoelectron Spectroscopy (XPS, VG Multi Lab 2000, Thermo Fisher Scientific, Massachusetts, USA).

Electrochemical measurements

All of electrochemical measurements carried out in a three-electrode cell with 0.5 M $(NH_4)_2SO_4$ as the electrolyte, a platinum (Pt) as counter electrode, saturated calomel electrode (SCE) as reference electrode. And the working electrode was composed of the as-prepared samples powder, Super P and binder polyvinylidene fluoride (PVDF) in a certain proportion of 7:2:1. The electrode slurry was fully dispersed and coated on carbon paper, which was washed with deionized water and ethanol, with a mass loading of about 1.0–1.5 mg cm^{-2}. The galvanostatic charge/discharge (GCD) test and cyclic voltammetry (CV) tests were conducted via CHI760E. The electrochemical measurements of full-cell were performed in the electrolytic cell with 1M $(NH_4)_2SO_4$ as the electrolyte, PVO as cathode, PTCDI as anode. The voltage window was between 0.1V and 1.8 V. The full cell was designed as cathode-capacity limited, and the current density, specific capacity, and the energy density are all based on the active mass of cathode.

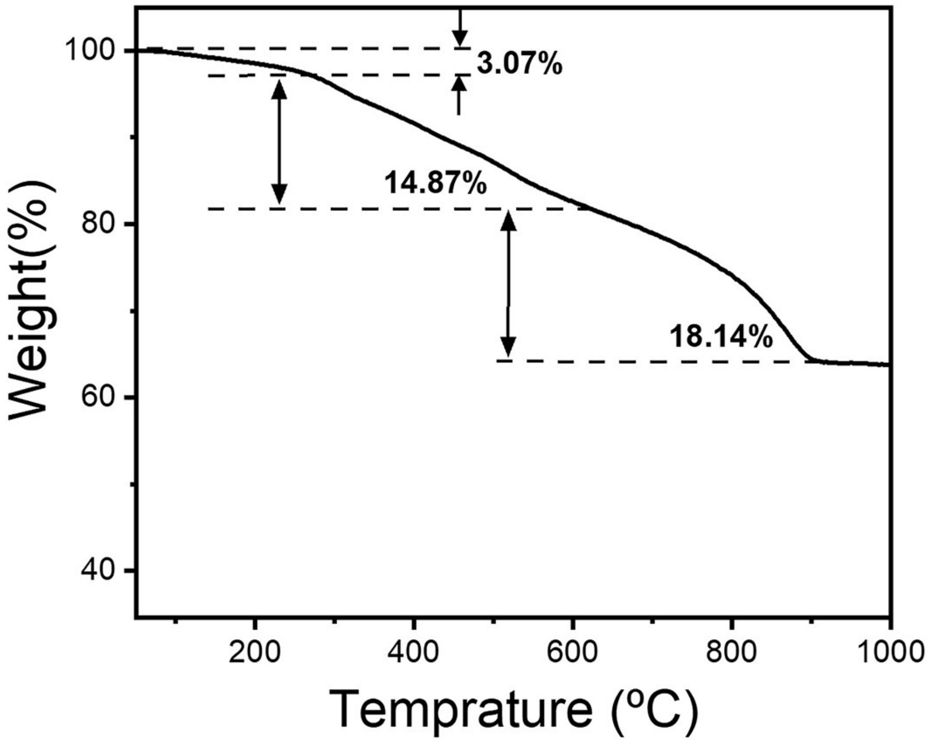

Fig S1. TG curve of PVO

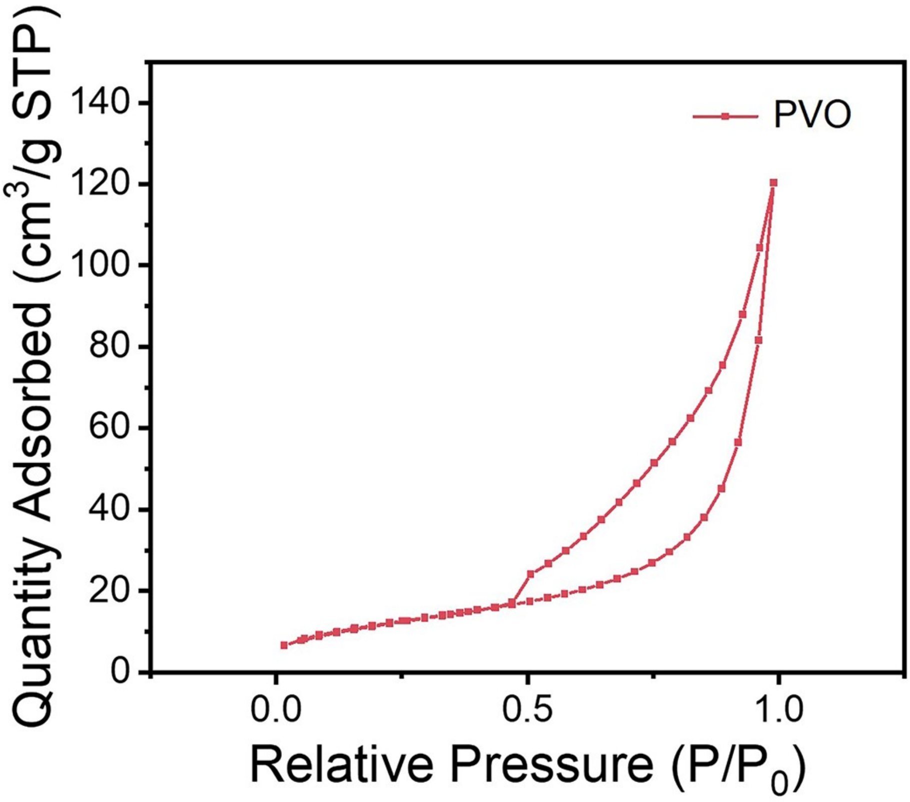

Fig S2. BET image of PVO

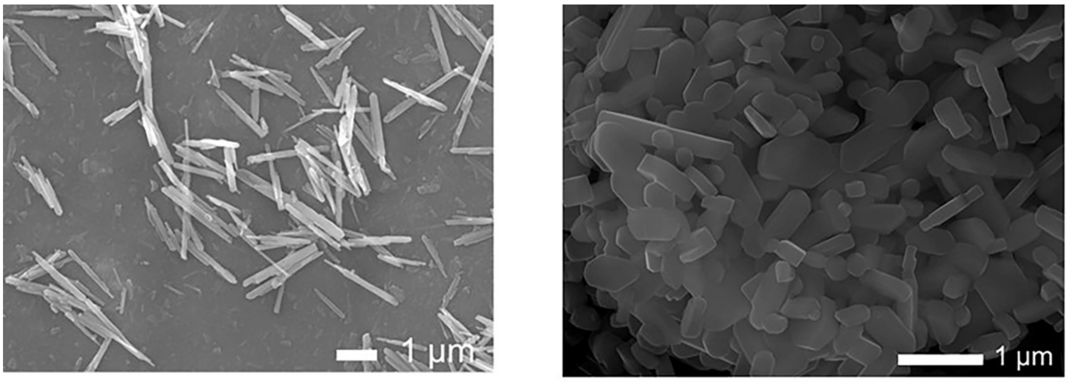

Fig S3. SEM image of HVO (a) and VO (b)

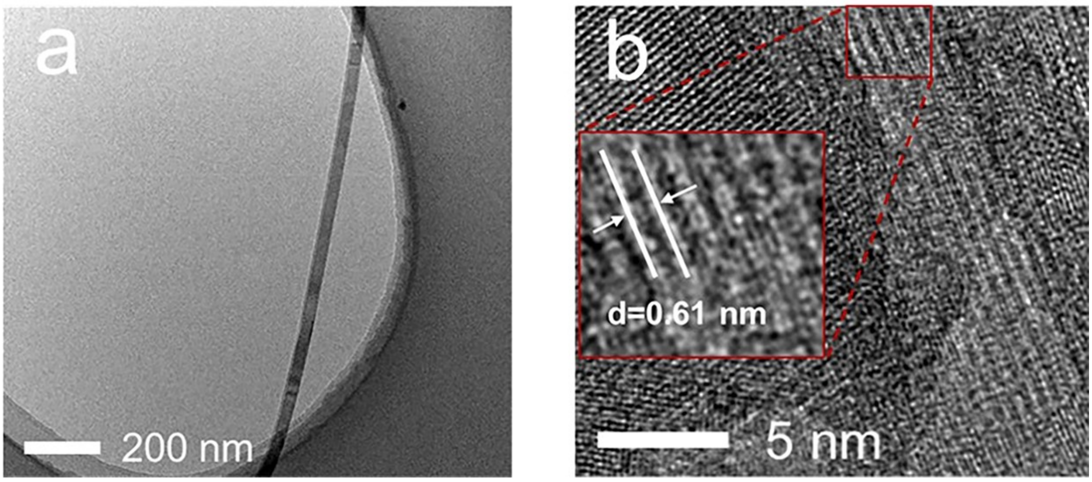

Fig S4. a) TEM pattern of HVO b) HRTEM pattern of HVO

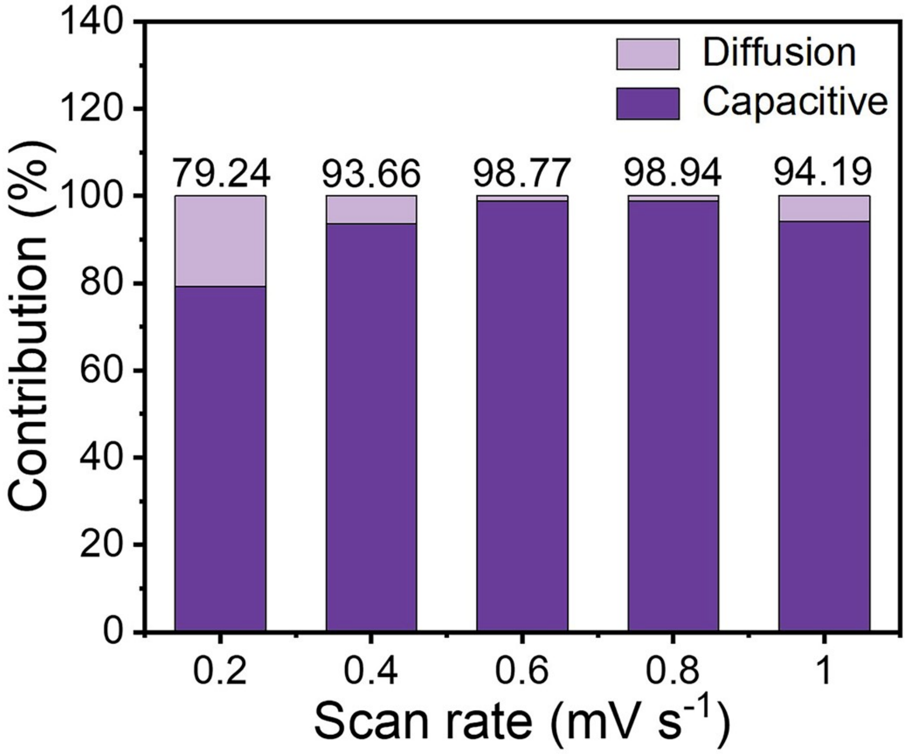

Fig S5. Capacitive contribusion of PVO cathode at different scan rate

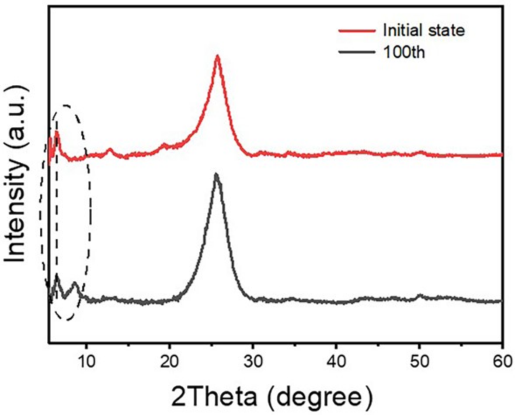

Fig S6 XRD of PVO electrodes after 100 cycles.

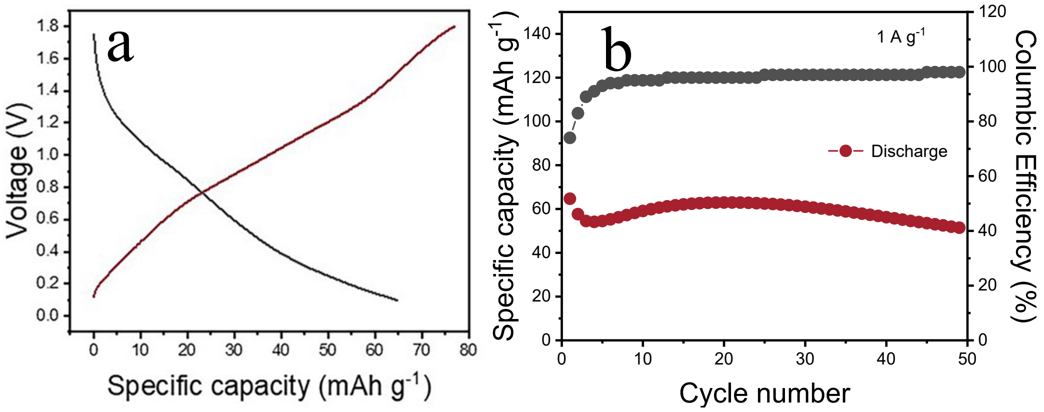

Figure S7. a) GCD of full cell. b) Cycle performance of full cell at the current density at 1A g^{-1}

According to the research of Ji[3] et al., PTCDI showed excellent ammonium storage performance. It displays the insertion/de-insertion capacity values are 127.9 and 158.9 mAh g^{-1} at a current density of 240 mAh g^{-1}. Therefore, we chose PTCDI as the anode, and the as-synthesized PVO as the cathode and 1M $(NH_4)_2SO_4$ as the electrolyte, carrying out electrochemical test in the electrolytic cell. Fig S7a is the GCD curve of the full cell, the discharge specific capacity is 65 mAh g-1 at 1A g-1. The as-assembled full cell also shows an excellent cycle performance, the capacity can be retained at 60.4 mAh g^{-1} after 100 cycles.

Reference

1. Chen, Y.; Yang, G.; Zhang, Z.; Yang, X.; Hou, W.; Zhu, J. J. *Nanoscale* **2010,** 2, (10), 2131-8.
2. Dong, S.; Shin, W.; Jiang, H.; Wu, X.; Li, Z.; Holoubek, J.; Stickle, W. F.; Key, B.; Liu, C.; Lu, J.; Greaney, P. A.; Zhang, X.; Ji, X. *Chem* **2019,** 5, (6), 1537-1551.
3. Wu, X.; Qi, Y.; Hong, J. J.; Li, Z.; Hernandez, A. S.; Ji, X. *Angew Chem Int Ed Engl* **2017,** 56, (42), 13026-13030.