

Supporting Information

Preparation of Heterogeneous VO_x/V₂C MXene@V₂O₅ via Van der Waals Self-Assembly for High-Performance Aqueous Zinc-Ion Batteries

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Experimental Section

1. Synthesis of Materials

V₂O₅ Nanosheets: V₂O₅ nanosheets were synthesized via a hydrothermal method. Briefly, 0.364 g of commercial V₂O₅ powder was dissolved in 30 mL of deionized water under stirring for 15 min. Subsequently, 15 mL of H₂O₂ was added, and the mixture was stirred for 30 min until a transparent light-yellow solution formed. The solution was transferred into a Teflon-lined autoclave and heated at 200 °C for 20 h. After cooling to room temperature, the resulting gel was collected and freeze-dried for 24 h to obtain V₂O₅ nanosheets. A dispersion with a concentration of 1 mg mL⁻¹ was prepared in N-methyl-2-pyrrolidone (NMP) for further use.

VO_x/V₂C Nanosheets: V₂CT_x MXene was first prepared by selectively etching the Al layer from V₂AlC MAX phase. In a typical procedure, 1.0 g of V₂AlC powder was slowly added to 20 mL of hydrofluoric acid (HF, 40 wt%) in an ice bath under stirring for 5 min. The mixture was then continuously stirred at 30 °C for 45 h. The resulting suspension was centrifuged and washed repeatedly with deionized water until the supernatant reached pH ≈ 6. Tetraethylammonium hydroxide (C₄H₁₃NO) was introduced as an intercalant, and the mixture was stirred at room temperature for 12 h, followed by ultrasonication for 1 h under N₂ atmosphere. After centrifugation at 3500 rpm, the sediment was washed

several times and re-dispersed in water. The obtained ink was further centrifuged at 9000 rpm for 30 min, and the collected solid was vacuum-dried for 8 h. Finally, the dried powder was annealed at 600 °C under a CO₂ atmosphere for 30 min to obtain VO_x/V₂C. A 1 mg mL⁻¹ dispersion in NMP was prepared for subsequent use.

VO_x/V₂C@V₂O₅ Hybrid: The VO_x/V₂C@V₂O₅ heterostructure was assembled via van der Waals interactions. Typically, the VO_x/V₂C-NMP dispersion was added dropwise into the V₂O₅-NMP dispersion at a mass ratio of 4:6 (VO_x/V₂C: V₂O₅). The mixture was sonicated for 1 h at room temperature to promote self-assembly. The final product was collected by vacuum filtration and dried overnight at 60 °C under vacuum.

2. Material characterization

The morphology and microstructure of the samples were examined by field-emission scanning electron microscopy (FE-SEM, Hitachi S-4800), scanning transmission electron microscopy (STEM, JEOL JEM-2800), and atomic force microscopy (AFM, Bruker Edge). Crystalline phases were identified by X-ray diffraction (XRD, Bruker D8 Discovery) using Cu K α radiation ($\lambda = 0.15406$ nm) in the 2θ range of 3-90° with a scan rate of 10° min⁻¹. Chemical states and surface composition were analyzed by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha). Raman spectra were recorded on a Horiba LabRAM HR Evolution spectrometer

with a 633 nm He-Ne laser. The wettability of the electrodes was evaluated using a contact-angle goniometer (POWREACH JC2000DM) by depositing a droplet of electrolyte onto the electrode surface.

3. Electrochemical measurements

Electrochemical tests were performed using an electrochemical workstation (DH7000, Jiangsu Donghua) and a multi-channel battery tester (CT-4008T-5V50mA-164, Neware). The cathode slurry was prepared by mixing the active material, Super P conductive carbon, and polyvinylidene fluoride (PVDF) binder in a weight ratio of 80:10:10 in N-methyl-2-pyrrolidone. The slurry was coated onto a stainless-steel mesh current collector and dried overnight at 60 °C under vacuum. The mass loading of active material was approximately 1.5 mg cm⁻². A glass fiber membrane (Whatman GF/D) was used as the separator, and 3 M Zn(CF₃SO₃)₂ aqueous solution was employed as the electrolyte. All measurements were carried out at room temperature.

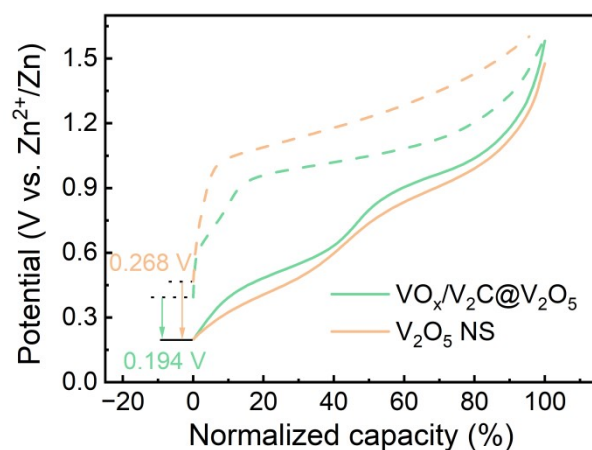


Figure S1 Overpotential of V_2O_5 NS and $VO_x/V_2C@V_2O_5$ at a current density of 3 A g^{-1} .

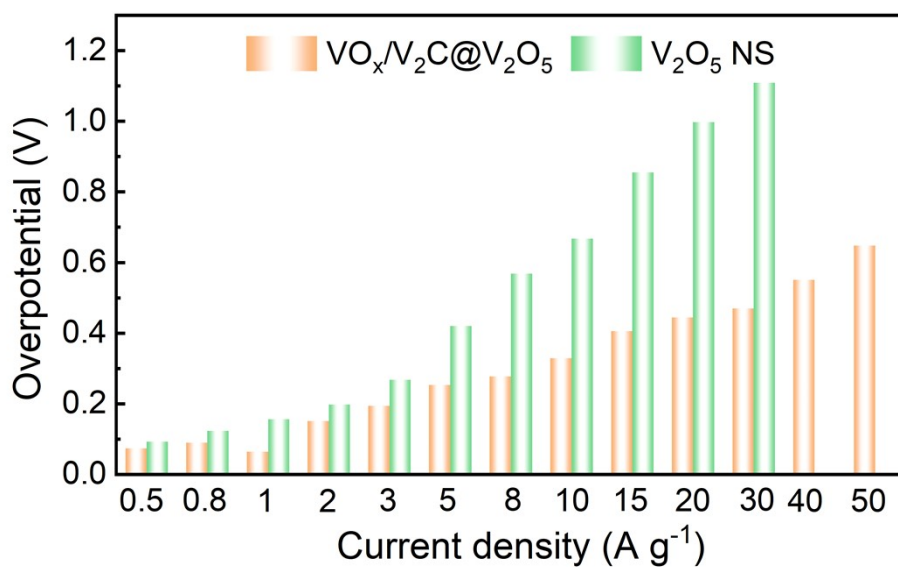


Figure S2 Summary of overpotentials of V_2O_5 NS and $VO_x/V_2C@V_2O_5$ at different current densities.