Supporting Information

In-situ electrochemically activated V₂O₃@MXene cathode for super high-rate and long-life Zn-ion battery

Yunlong Duan,^a Zhi Geng,^a Daohong Zhang,^{*a,b} Qiufan Wang^{*a,b}

^{*a*}Key Laboratory of Catalysis and Energy Materials Chemistry of Ministry of Education & Hubei Key Laboratory of Catalysis and Materials Science, Hubei R&D Center of Hyperbranched Polymers Synthesis and Applications, South-Central Minzu University, Wuhan 430074, China

^b Guangdong Provincial Laboratory of Chemistry and Fine Chemical Engineering Jieyang Center, Jieyang 515200, China

China. Email: zhangdh27@163.com (D. Zhang); ygdf@mail.scuec.edu.cn (Q. Wang)

Experimental section

Preparation method of monolayer MXene dispersion

First, 20 mL of 12 M HCl was added to a PTFE bottle, and then 1.6 g lithium fluoride (LiF, 99.98 %, Sigma-Aldrich) was added. The liquid was stirred at room temperature for 20 min to obtain HCl/LiF etching solution. Then 1 g MAX (Ti₃AlC₂) was slowly added to the etching solution and stirred at 40 °C for 24 h, the acid mixture product after the reaction was collected, centrifuged and repeatedly washed with 1 M HCl, and deionized water to make the pH>6. The supernatant was collected as a layered MXene product. In order to obtain a single-layer or multi-layer MXene dispersion, N₂ was introduced, and ultrasonic treatment was performed in an ice water bath for 3 times. Finally, collecting the supernatant as layered MXene product after centrifugation at 3500 rmp/min for 30min.

Preparation of V₂O₃@MXene Cathode Materials

First, the prepared MXene monolayer dispersion with a concentration of 10 mg/mL was diluted to 2.5 mg/mL and 1.25 mg/mL and added to the beaker. Add 0.1g of the prepared V_2O_3 to the beaker and stir magnetically for 24 h at room temperature. After standing for 4 h, collect the bottom precipitated product and dry it under vacuum

conditions to obtain V₂O₃@MXene.

Characterization

The phases of the synthesized samples and precursors were identified using X-ray diffraction (XRD). The surface morphology and internal structural characteristics of the product were studied using scanning electron microscopy (SEM) and transmission electron microscopy (TEM).

Electrochemical measurements

We used C2032 model button cells for electrochemical testing. First, the sample, carbon black (Super P) and polyvinylidene fluoride (PVDF) were mixed in N-methylpyrrolidone (NMP) at a mass ratio of 7:2:1. The slurry was evenly coated on the carbon cloth carrier fluid, dried under vacuum at 60° for 12 h, the V₂O₃@MXene cathode for the mass loading of active material in each electrode disc was around 0.8 mg cm⁻².and the electrochemical performance of the battery was tested through IVium and NEWARE systems.

Preparation of PVA-Zn(CF3SO3)2-gelatin gel electrolyte:

The gel electrolyte was obtained by dissolving 0.4 g Gelatin, 1.414 g $Zn(CF_3SO_3)_2$ and 0.877g PVA in 10 mL DI, and heating at 90 °C for 3 h.

Assembly of flexible and quasi-solid-state V_2O_3 (MXene rechargeable device

Flexible zinc as anode, V₂O₃@MXene as cathode, PVA- Zn(CF₃SO₃)₂-gelatin gel electrolyte and NKK separator are assembled, and three sets of devices are connected in series.

Computational details

(1) Capacitance contribution

The relationship formula between peak current (i) and sweep speed (v) follow the rule:

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

A b value of 1 suggests a capacitive process and 0.5 was indicative of diffusion controlled charge storage. The capacitive and diffusion contribution at various scan rates can be obtained via the equation:

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

where k_1 and k_2 are proportional to capacitance and diffusion contributions, respectively.

(2) Galvanostatic current intermittent titration technique (GITT)

The calculation method was according to the following equation:

$$D = \frac{4}{\pi \tau} \left(\frac{m_B V_M}{M_B A} \right)^2 \left(\frac{\Delta E_S}{\Delta E_r} \right)^2 \tag{3}$$

where τ was the duration time of the current pulse, m_B was the mass of the active material, M_B was the molecular weight (g/mol), V_M is its molar volume (cm³/mol), A was the total contacting area of electrode with electrolyte, ΔE_s was related to the change of steady-state voltage for the corresponding step.



Figure S1. XRD comparison of precursor VS_2 and V_2O_3 .



Figure S2. XRD small angle peaks of V₂O₃@MXene with different content ratios.



Figure S3. EPR signals of V₂O₃ and V₂O₃@MXene.



Figure S4. TGA curve of VS₂ samples.



Figure S5. Interplanar spacing of V₂O₃



Figure S6. Comparison of electrochemical properties of V₂O₃ obtained by different

calcination time in Ar.



Figure S7. Comparison of electrochemical properties of V₂O₃ obtained at different



Figure S8. Charge-discharge profiles of the V_2O_3 @MXene cathode at 0.2 A g⁻¹



(excluding the first lap long platform).



Figure S9. (a) The XRD patterns of V_2O_3 and (b) V_2O_3 @MXene after fifteen and thirty days

Figure S10. Comparison of electrochemical performance of V_2O_3 cathode under

different electrolyte.



Figure S12. EIS of (a) V₂O₃ and (b) V₂O₃@MXene cathode under different cycling

conditions.



Figure S13. (a) The CV curves. (b) b-values curves. (c) capacitive contribution of V_2O_3 cathode.