Supplementary Material

Two-dimension amorphous VOPO₄/graphene heterostructure for high-voltage aqueous Zn-Ion battery

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Experiment and methods

Material preparation

The synthesis of C-VOP was carried out through a solution-phase reaction. Specifically, 1.2 g of V_2O_5 (Sigma Aldrich) was combined with 6.65 mL of H_3PO_4 (Merck) and 28.8 mL of H_2O . The mixed reaction was then subjected to reflux conditions at 115 °C for 20 h. The resulting yellow-green precipitate was isolated through filtration and thoroughly washing with alternating H_2O /isopropanol solutions. The final C-VOP product was achieved after overnight drying at 60 °C.

For the preparation of A-VOP/G, a solvothermal approach was employed using C-VOP and graphene oxide as the precursor. Initially, 120 mg of C-VOP and 60 mg of graphene oxide were dispersed in 50 mL of isopropyl alcohol under ultrasonication. The resulting suspension was then transferred to an autoclave and maintained at 170 °C for 20 h. The solvent thermal process can lead to the formation of localized disorder in the crystal, which can affect the overall structure of the crystal. Finally, the rupture of the crystal may lead to the formation of an amorphous material. The A-VOP/G powder was subsequently collected and dried following the same procedure as described for C-VOP ¹. The preparation of A-VOP was similar to A-VOP/G procedure without the incorporation of graphene oxide.

Material characterization

Structural characteristics of C-VOP, A-VOP and A-VOP/G were systematically investigated using multiple analytical techniques. X-ray diffraction (XRD) patterns were acquired using a Rigaku-D/Max-3A diffractometer to examine crystalline phases. Morphological analysis was performed using scanning electron microscopy (SEM, SUPRA 55 SAPPHIRE) and transmission electron microscopy (TEM, JEM2100F) to evaluate the surface and structural features. Chemical composition and valence state analysis were conducted through X-ray photoelectron spectroscopy (XPS) measurements using a PerkinElmer PHI 1600 ESCA system with Al Kα radiation.

Electrochemical measurement

The electrode fabrication process involved thoroughly mixing the active material, acetylene black conductive agent, and polyvinylidene fluoride (PVDF) binder in a 8:1:1 weight ratio using N-methyl-2-pyrrolidone (NMP, Aldrich) as the solvent. The resulting homogeneous slurry was uniformly coated onto stainless steel mesh current collectors. After drying at 60 °C overnight, the cathodes wear achieved. The cells were configured with zinc foil anodes, 1 M Zn(CF₃SO₃)₂ + 21 M LiN(CF₃SO₂)₂ aqueous electrolyte, and glass fiber separators. Electrochemical impedance spectroscopy (EIS) measurement was performed with CHI 660E workstation (Chenhua, Shanghai) across a frequency range of 100 kHz to 10 mHz. Cyclic voltammetry (CV) analysis was conducted at scan rates ranging from 0.2 to 1.0 mV s⁻¹ to investigate the electrochemical kinetics. Galvanostatic charge-discharge (GCD) tests and long-term cycle stability was assessed using a LAND CT2001A battery testing system within a voltage window of 0.8-2.1 V.



Fig. S1. SEM image of C-VOP.



Fig. S2. (a) SEM and (b) TEM images of A-VOP and (c) the corresponding SAED image.



Fig. S3. SEM-EDS of A-VOP/G.



Fig. S4. Thermogravimetric analysis of A-VOP/G.



Fig. S5. XPS spectrum of A-VOP/G.



Fig. S6. XPS P 2p spectrum of A-VOP/G.



Fig. S7. XPS C 1s spectrum of A-VOP.



Fig. S8. Cycling performance of the pure graphene at 1 A g^{-1} .



Fig. S9. GCD curves of A-VOP and C-VOP.



Fig. S10. (a) SEM image and (b) XRD pattern of Zn anode after 100 cycles





Fig. S12. Log(i) versus log(v) plot of A-VOP and C-VOP.



Fig. S13. Capacitive and diffusion contribution A-VOP and C-VOP.



Fig. S14. GITT curve and diffusion coefficient of A-VOP.



Fig. S15. GITT curve and diffusion coefficient of C-VOP.

Sample	Discharge capacity	Voltage	Discharge	Ref.
	(mAh g ⁻¹)	window (V)	voltage (V)	
A-VOP/G	177.9 at 500 mA g^{-1} 73.6 at 5000 mA g^{-1}	0.8-2.1	1.50 V	This work
VOPO ₄	50 at 5000 mA $\rm g^{-1}$	0.8-2.1	1.56 V	2
PPy-VOPO ₄	65 at 100 mA g^{-1}	0.5-2.0	1.1 V	3
B-doped VOPO ₄	234.5 at 100 mA g^{-1}	0.2-1.9	0.8 V	4
Bilayer-VOP	313.6 at 100 mA g^{-1}	0.2-1.9	1.1 V	5
Zn _{0.56} VOPO ₄	100.6 at 200mA g^{-1}	0.5-1.9	1.46 V	6
KVOPO ₄	89.23 at 100 mA g^{-1}	0.8-2.0	~1.33 V	7

 Table S1. Summarize the electrochemical performance.

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