

Supporting Information for

**Interphase Reinforced Amorphous Vanadium Oxide Cathode Materials for Aqueous Zinc Ion Batteries**

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**S 1 Experiment Section**

**S 1.1 Synthesis of VO-PO@C**

**Polyethyleneimine nano-particles (PNPs)** were synthesized by a inverse emulsion method. In brief, 1.0 g polyethyleneimine (PEI, average Mw. 600, Aladdin, 99.8%) was dissolved in 4 mL deionized water. Subsequently, the solution was doped in a liquid paraffin (40 mL) contain 1.6 g Sorbitan monooleate (Span 80, Aladdin, AR) under 10000 rpm for 10 min. 5 g glutaraldehyde solution (Innochem, 25wt.% in water) was doped in the emulsion under 10000 rpm for 30 min. Afterward, the pink mixed emulsion at magnetic stirring for 8 h. The product was collected by centrifugal and wash with petroleum ether and ethanol thoroughly 3 times, respectively. The pink powder was dried at 80 °C for 12 h in air.

**Vanadate hybrid & polyethyleneimine nano-particles (VO@PNPs)** were synthesized by a chemical adsorption method. 5 g NaVO<sub>3</sub> (Aladdin, 99.8%) was dissolved in 70 mL deionized water at 80 °C for 15 min. Subsequently, the 40 mL suspension contain 2.0 g PNPs was added in the NaVO<sub>3</sub> solution, the pH is adjusted to 3 by HCl, and keep magnetic stirring for 30 min. The product was collected by centrifugal and wash with water and ethanol thoroughly 3 times, respectively. The yellow powder was dried at 80 °C for 12 h in air. And the, VO@CNP<sub>s</sub> obtained by annealing the yellow powder at 500 °C for 3 h under N<sub>2</sub>.

**Vanadate hybrid & polyethyleneimine nano-particles with phosphate interphase (PO-VO@PNPs)** were synthesized by same chemical adsorption method. 0.36 g phytic acid solution (Aladdin, 70 wt.% in water ) was added in 20 ml suspension contain 0.5 g VO@PNPs at

magnetic stirring for 30 min. The product was collected by centrifugal and wash with water and ethanol thoroughly 3 times, respectively. The yellow powder was dried at 80 °C for 12 h in air. And the, VO@CNPs obtained by annealing the yellow powder at 500 °C for 3 h under N<sub>2</sub>.

### **S 1.2 Material characterizations**

The XRD patterns of the sample were measured using a Bruker D8 ADVANCE X-ray diffractometer with Cu-K $\alpha$  radiation (maximum voltage = 50 kV and maximum current = 60 mA). The morphologies of the composite were observed via scanning electron microscopy (SEM, SU8010, Hitachi, Japan). The element distribution in the sample was scanned by energy dispersive spectroscopy (EDS). Transmission electron microscopy (TEM), Selected area electron diffraction (SADE) and high-resolution transmission electron microscopy (HRTEM, JEM-2100F (URP), JEOL, Japan) were performed to determine the overall morphologies and internal structure. Thermogravimetric analysis (TGA, DSC3+, TGA 4000, PerkinElmer, Netherlands) was employed to determine the thermal decomposition data of the complex. X-ray photoelectron spectroscopy (XPS, Escalab 250Xi, Thermo Fisher, Britain) was used to analyze the combination status of elements.

### **S 1.3 Electrochemical characterizations**

The V-based electrode was prepared by mixing V-based (70 wt.%), conductive carbon additives (Ketjen Black, 20 wt.%), and polyvinylidene fluoride (PVDF, 10 wt.%), and then the mixed slurry was cast onto Ti foil and dried in a vacuum oven at 80 °C for 8 h. Zinc foil ( $\Phi$  14 mm, thickness 30  $\mu$ m) and a glass fiber membrane (Whatman, GF/D) were used as the anode and separator, respectively, and zinc trifluoromethanesulfonate (Zn(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>, 3 mol in 1 L water, Aladdin, AR, 98.0%) solution was used as the electrolyte. A CR2032-type coin cell was assembled in air to evaluate the electrochemical performance on a LAND battery testing system (CT3001A). Cyclic voltammograms (CV) and electrochemical impedance spectroscopy (EIS) were performed on a CHI600E electrochemical workstation. All the tests were performed at room temperature (25 °C  $\pm$  2 °C). The calculation method of specific capacity was calculated by mass proportion of activate material in cathode electrode.

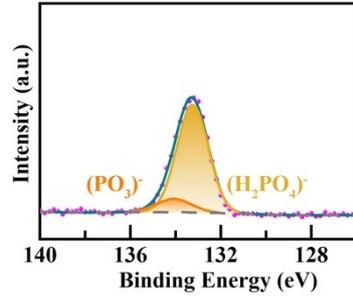


Fig. S1 The XPS peak of P<sub>2p</sub> element of VO-PO@CNPs

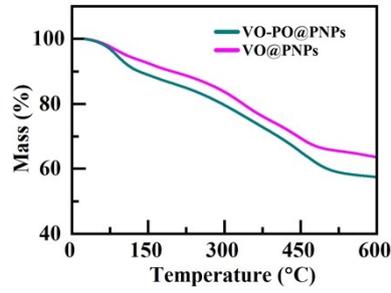


Fig. S2 The TGA of VO@PNPs and VO-PO@PNPs at N<sub>2</sub>.

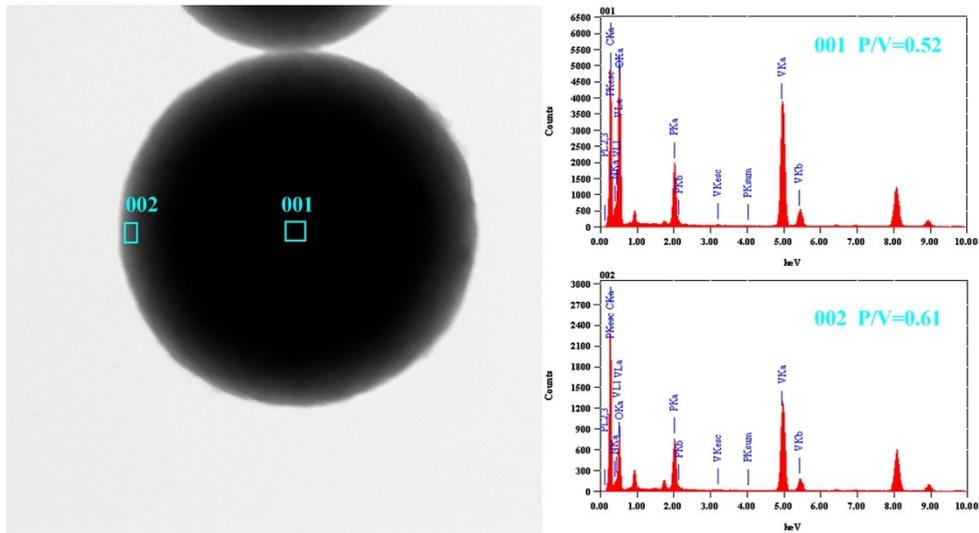
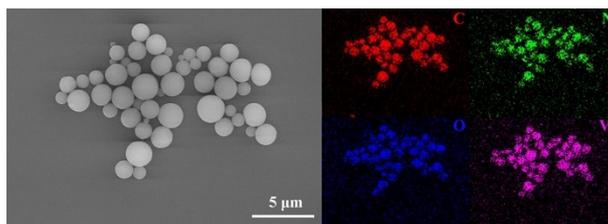
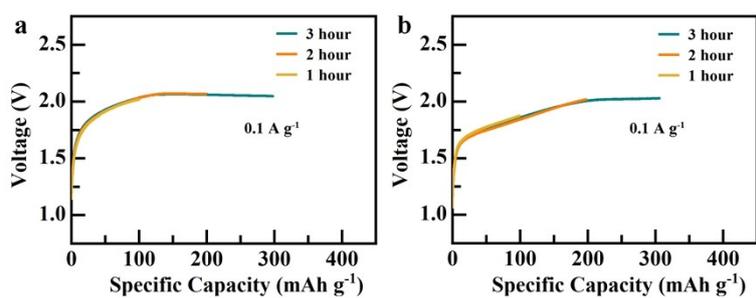


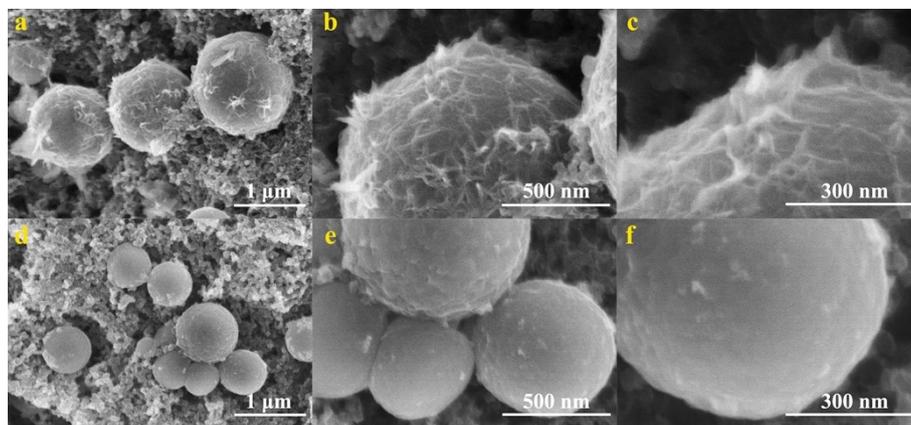
Fig. S3 Select energy dispersive spectroscopy of the VO-PO@CNPs



**Fig. S4** The SEM and elements mapping of VO@CNPs.



**Fig. S5** Activate profiles at different charge time, (a) VO@CNPs and (b) VO-PO@CNPs



**Fig. S6** SEM of VO@CNPs (a-c) and VO-PO@CNPs (d-f) after pre - charge at  $0.1 \text{ A g}^{-1}$  for 3 hours

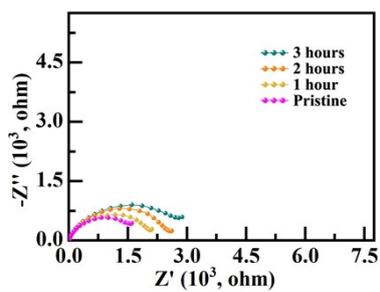


Fig. S7 EIS of VO@CNPs after pre - charge at  $0.1 \text{ A g}^{-1}$  for different time

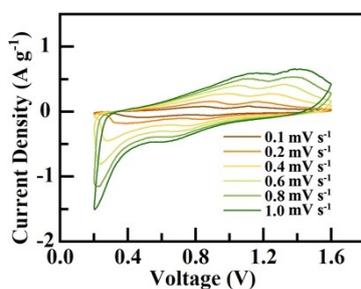


Fig. S8 CV profiles of VO-PO@CNPs at different scan rates.

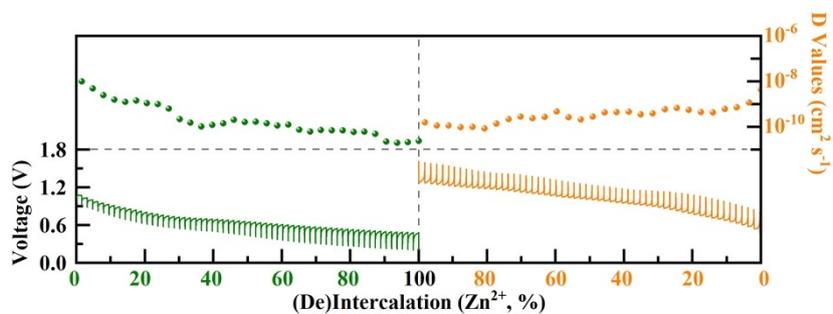
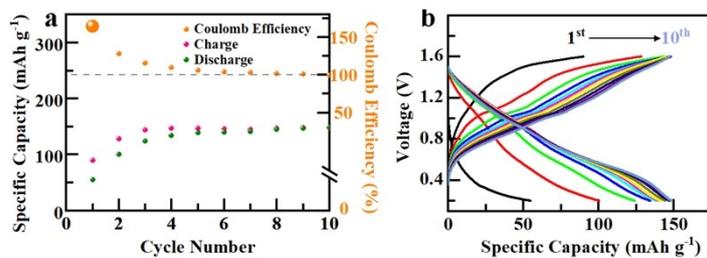
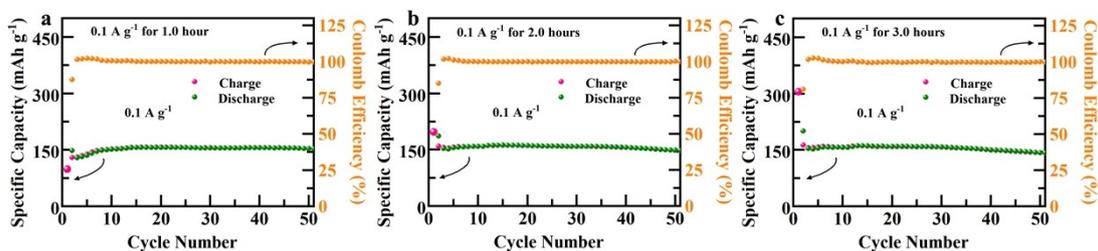


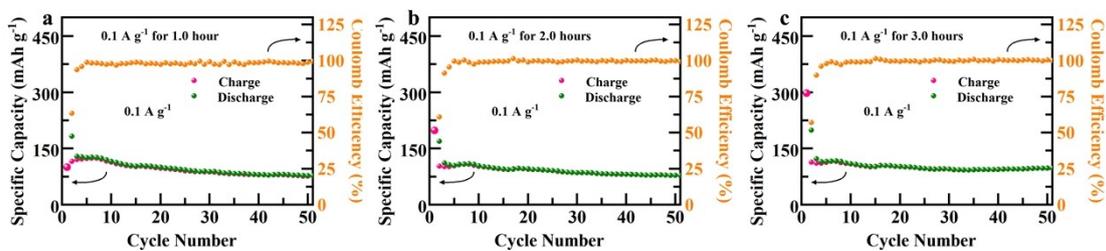
Fig. S9 GITT profiles and diffusion coefficient (D values) of VO@CNPs electrode



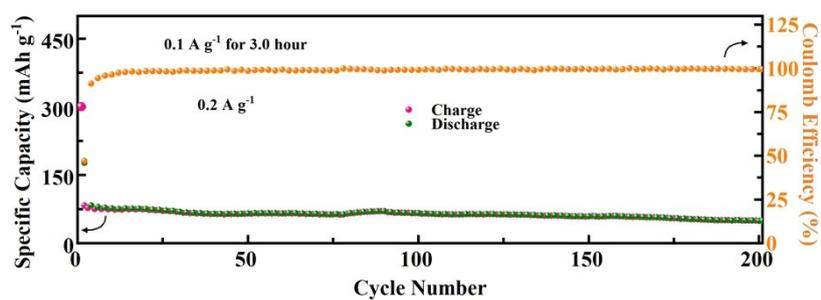
**Fig. S10** Cycle performance (a) and charge/discharge curves (b) of VO-PO@CNP<sub>s</sub> at 0.1 A g<sup>-1</sup> without pre-charge activation



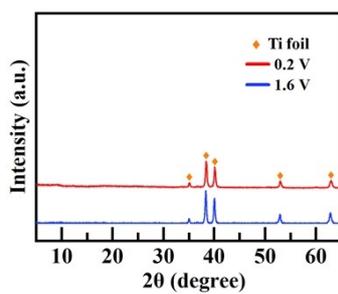
**Fig. S11** Cycle performance of VO-PO@CNP<sub>s</sub> at 0.1 A g<sup>-1</sup> after pre - charge at 0.1 A g<sup>-1</sup> for different time



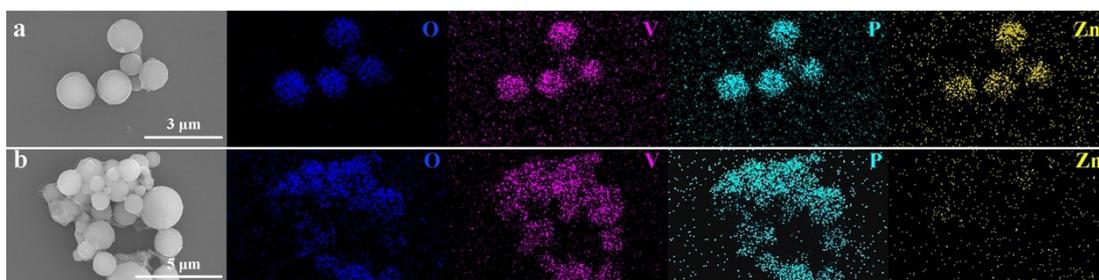
**Fig. S12** Cycle performance of VO@CNP<sub>s</sub> at 0.1 A g<sup>-1</sup> after pre - charge at 0.1 A g<sup>-1</sup> for different time



**Fig. S13** Cycle performance of VO@CNP<sub>s</sub> at 0.2 A g<sup>-1</sup> after pre - charge at 0.1 A g<sup>-1</sup> for 3 hours



**Fig. S14** In site XRD of VO@CNPs electrodes at fully charged / discharged under 10<sup>th</sup> cycle at 0.1 A g<sup>-1</sup>.



**Fig. S15** SEM images and elemental mapping of (a) discharged and (b) charged VO-PO@CNPs