Supporting information

An interfacial engineering strategy of VS_2/CNT 3D conductive network for stable and high-performance zinc-ion battery

Experimental Section

1. Materials preparation

Different amounts of carbon nanotubes (CNT) were added to the beaker, and 45 ml of deionized water was added. After ultrasonic treatment for two hours, a CNT ultrasonic dispersion solution was obtained. Then, 2 mM (0.234 g) NH_4VO_3 and 15 mM (1.127 g) C_2H_5NS (TAA) were placed in the beaker and stirred at room temperature for 10 minutes. After complete dissolution of NH_4VO_3, 2 mL of $NH_3 \cdot H_2O$ was slowly added to the beaker and stirred at room temperature for 20 minutes to ensure that $NH_3 \cdot H_2O$ fully participated in the reaction. Then, a 3×3 cm^2 piece of activated carbon cloth was cut as a current collector. The current collector and the solution were transferred to a lined autoclave, and a water heat reaction was carried out at 160 °C for 12 hours. After the reaction was completed, the product was collected, washed with deionized water, centrifuged 3~4 times, then washed with ethanol, centrifuged 2~3 times, and the carbon cloth covered with the materials was collected. It was placed in a 60°C vacuum drying box overnight to obtain the in-situ grown carbon cloth of VS_2/CNT. For comparison, a current collector carbon cloth without CNT was also prepared.

Materials characterization

The synthesized material was characterized using X-ray powder diffraction (XRD), X-ray photoelectron spectroscopy (XPS), Fourier transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), and transmission electron microscopy (TEM).

Electrochemical measurements

Electrochemical evaluation was performed by assembling CR2032-type coin cells, with zinc foil as the anode and fiberglass filter paper as the separator. The electrolyte consisted of a composite solution containing 80 μL of 2 M $Zn(CF_3SO_3)_2$ mixed with 2 M LiTFSI. Galvanostatic charge/discharge (GCD) tests were conducted on a Neware battery tester (BTS-4000) within a voltage window of 0~1.0 V versus Zn^{2+}/Zn. The galvanostatic intermittent titration technique (GITT) was carried out by applying a series of 5 minute galvanostatic discharge pulses at a current density of 100 mA h g^{-1}, followed by a 20 minute relaxation period. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) measurements were performed using a CHI760E electrochemical workstation.

Detail of calculation:

(1) Capacitance contribution

Calculation of the pseudocapacitance and diffusion-controlled contribution. In the CV test, the connection between the peak current i and the scan rate v is as follows:

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

$$\log(i) = b \times \log(v) + \log(a) \tag{2}$$

Where a and b are both variable parameters, and the variety of b values is 0.5 to 1, corresponding to diffusion control process and surface capacitance control process, respectively.

Moreover, the calculation formulas of capacitance control contribution k_1v and diffusion control contribution $k_2v^{0.5}$ are as follows:

$$i = k_1v + k_2v^{0.5} \tag{3}$$

(2) Galvanostatic current intermittent titration technique (GITT)

GITT is used to measure the diffusion coefficient of Zn^{2+}. The diffusion coefficient can be obtained according to the following equation:

$$D = \frac{4L^2}{\pi\tau}\left(\frac{\Delta Es}{\Delta E\tau}\right)^2 \tag{4}$$

Where $\tau(s)$ is the constant current pulse time. L corresponds to Zn^{2+} diffusion length, which is equal to thickness of the electrode. ΔE_s is the steady-state voltage change caused by current pulse. ΔE_τ is the voltage change during the constant current pulse.

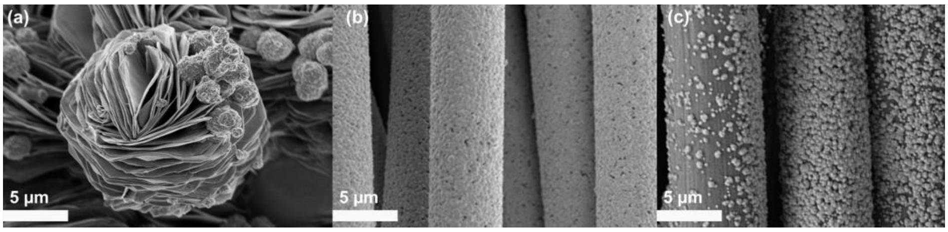

Figure S1. SEM images of (a) VS_2. (b) VS_2/CNT-0.01. (b) VS_2/CNT-0.04.

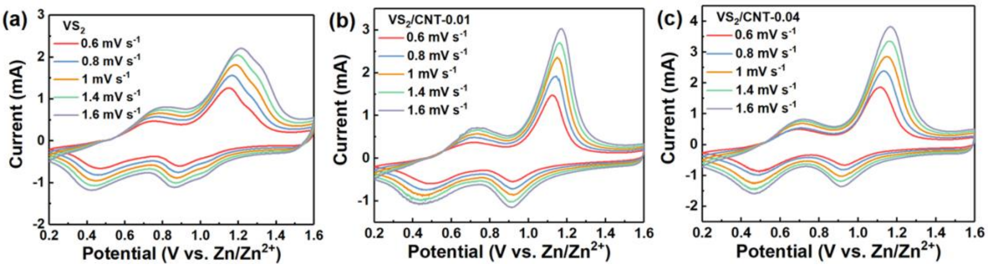

Figure S2. CV curves of (a) VS_2. (b) VS_2/CNT-0.01. (b) VS_2/CNT-0.04.

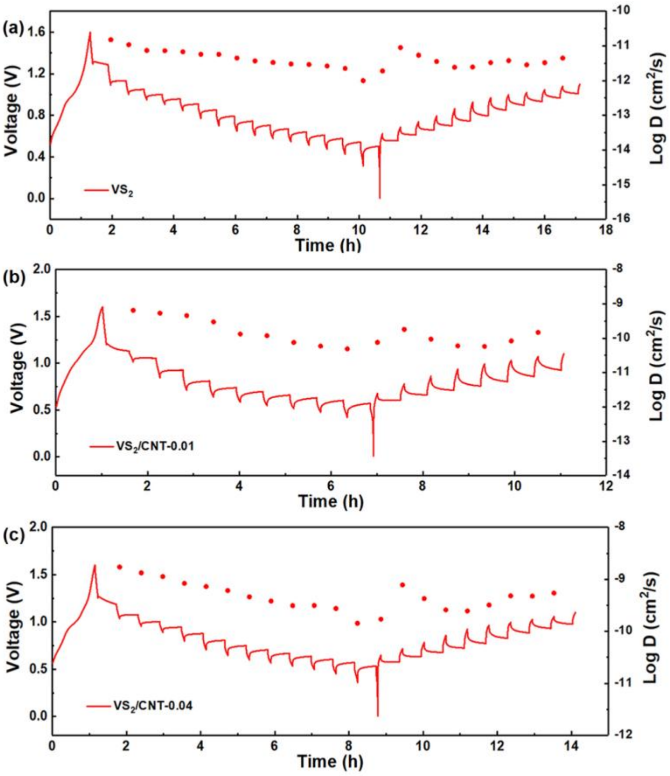

Figure S3. GITT curves and corresponding Zn^{2+} ion diffusion coefficient of (a) VS_2. (b) VS_2/CNT-0.01. (b) VS_2/CNT-0.04.

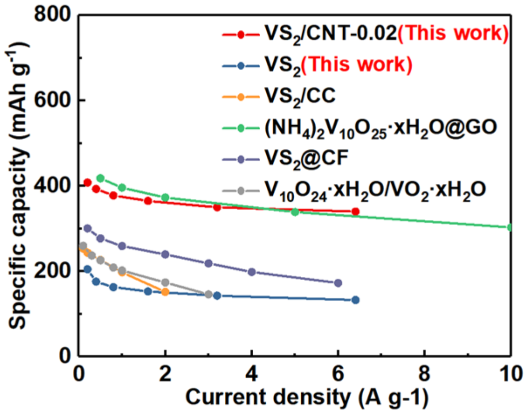

Figure S4. Comparison between VS, VS_2/CNT-0.02 and other materials.

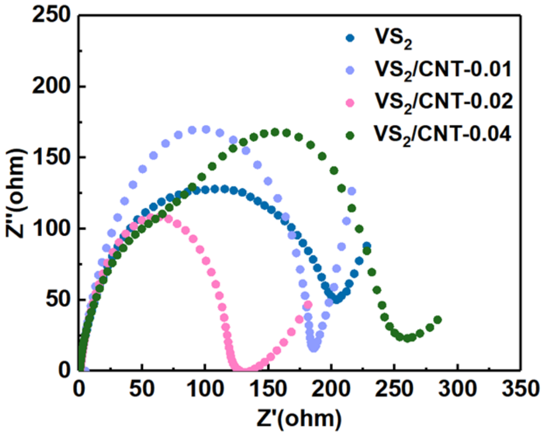

Figure S5. EIS of the samples in initial state.