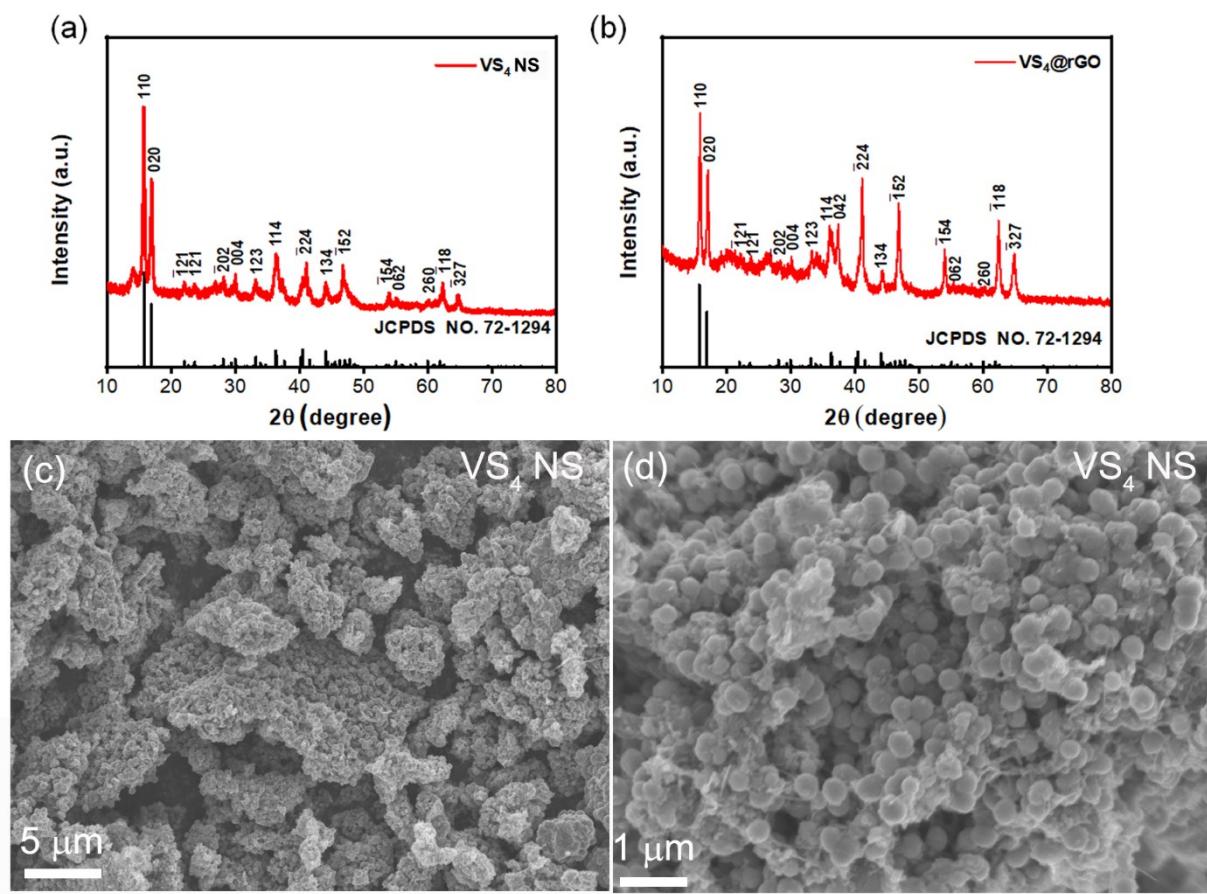


## Supporting Information

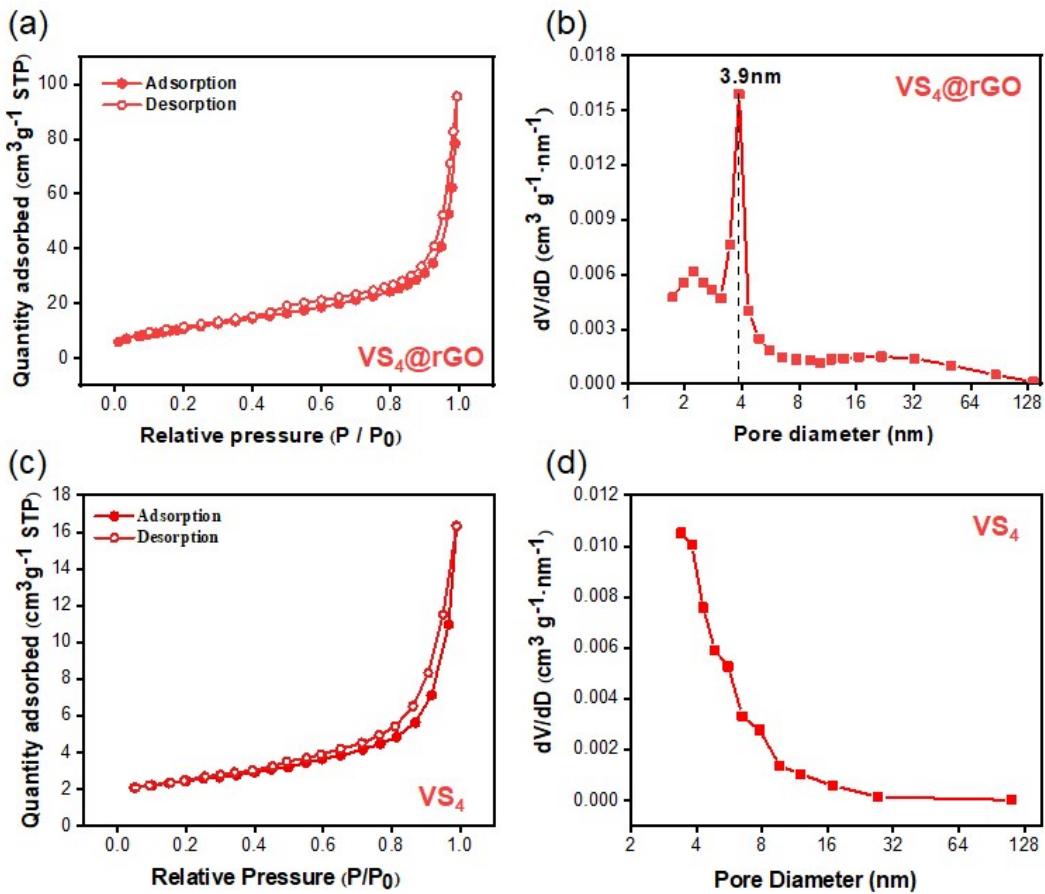
### Robust VS<sub>4</sub>@rGO Nanocomposite as a High-Capacity and Long-Life Cathode Material for Aqueous Zinc-Ion Batteries

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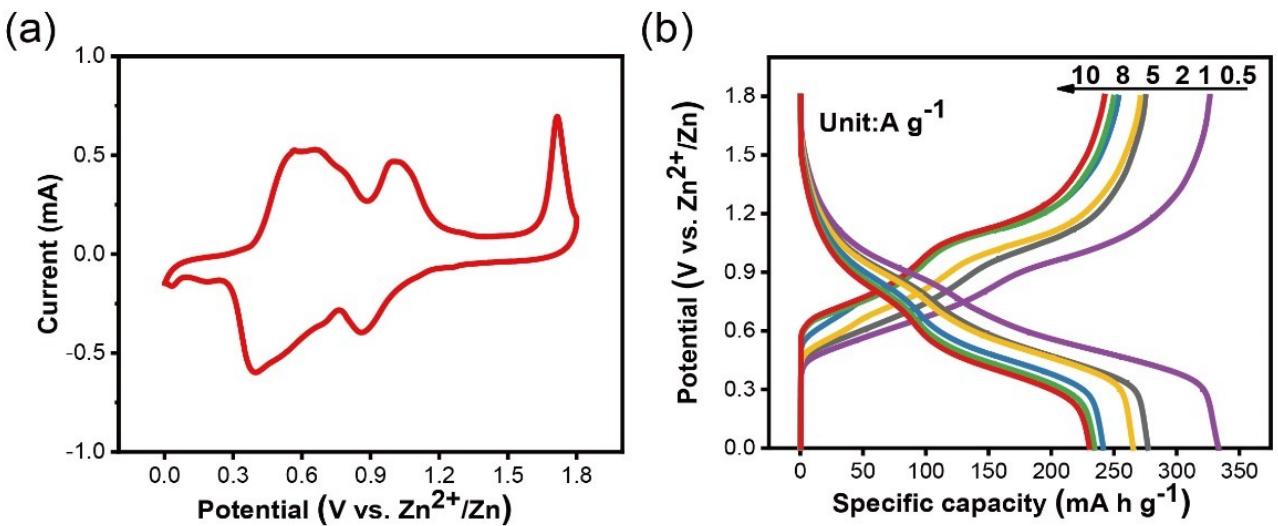
*Henan Key Laboratory of Diamond Optoelectronic Materials and Devices, Key Laboratory of Material Physics, Ministry of Education, School of Physics and Microelectronics, Zhengzhou University, Zhengzhou, 450052, China*



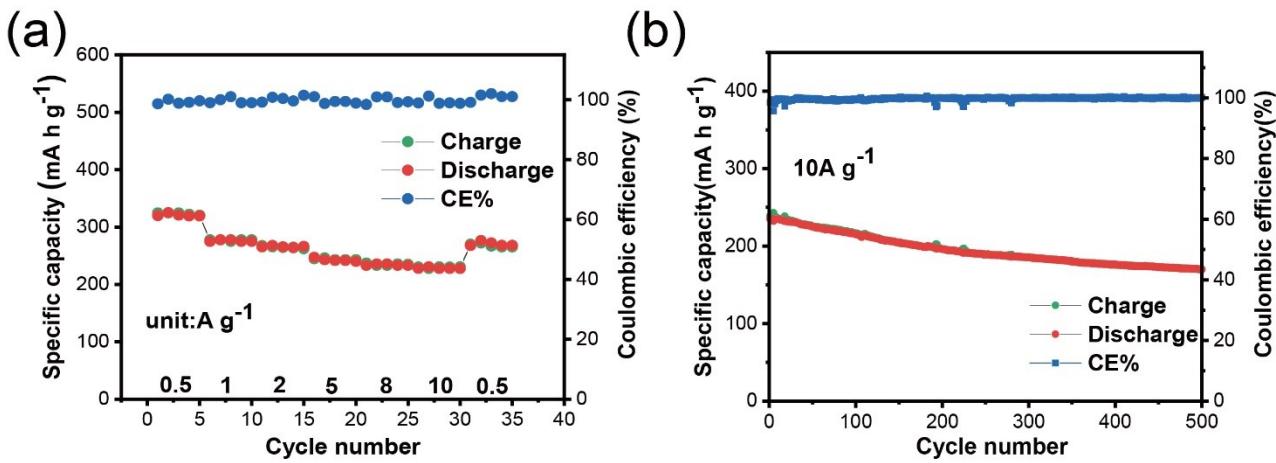
**Fig. S1** XRD pattern of the (a) VS<sub>4</sub> NS and (b) VS<sub>4</sub>@rGO sample. (c-d) Low-mag SEM images of VS<sub>4</sub> NS sample.



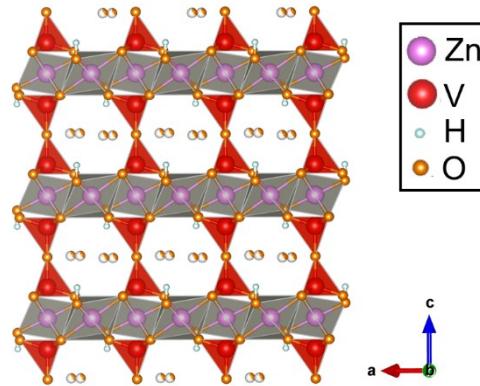
**Fig. S2** (a) Nitrogen adsorption-desorption isotherm and (b) pore size distribution of the  $\text{VS}_4@\text{rGO}$  composite. (c) Nitrogen adsorption-desorption isotherm and (d) pore size distribution of the  $\text{VS}_4$  NS sample.



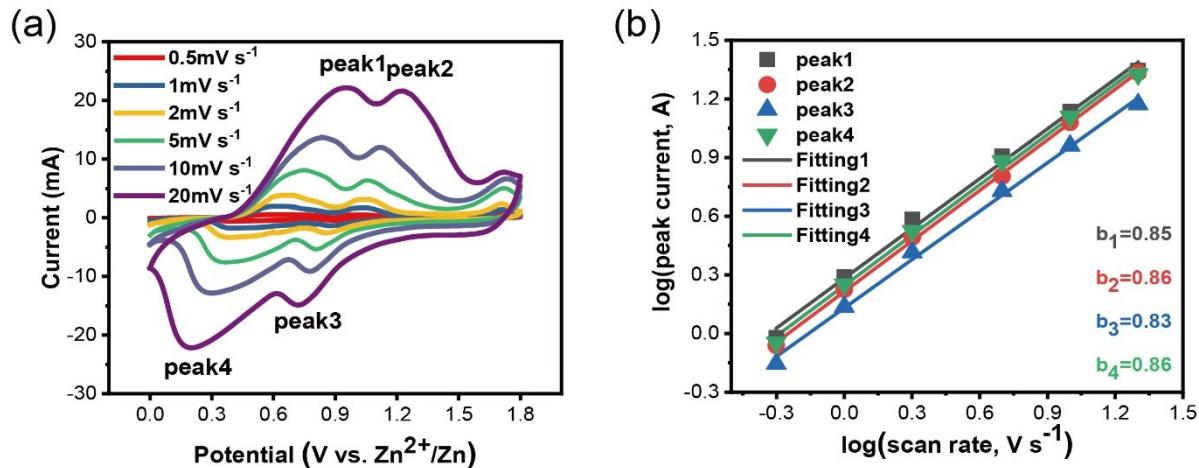
**Fig. S3** Electrochemical performance of  $\text{VS}_4$  NS. (a) CV curve at the scan rate of 0.5 mV s<sup>-1</sup>. (b) GDC curves at different current densities.



**Fig. S4** (a) Rate performance of the VS<sub>4</sub> NS electrode at current densities of 0.5, 1, 2, 5, 8 and 10  $\text{A g}^{-1}$ . (c) Cycle performance at the current density of 10  $\text{A g}^{-1}$ .



**Fig. S5** Schematic illustration of the crystal structure of Zn<sub>3</sub>(OH)<sub>2</sub>V<sub>2</sub>O<sub>7</sub>·2H<sub>2</sub>O viewed along (010) direction.



**Fig. S6** (a) CV curves of the VS<sub>4</sub>@rGO composite electrode at scan rates of 0.5, 1, 2, 5, 10 and 20  $\text{mV s}^{-1}$ . (b) log(i) versus log(v) plots at different redox peaks of the composite electrode.

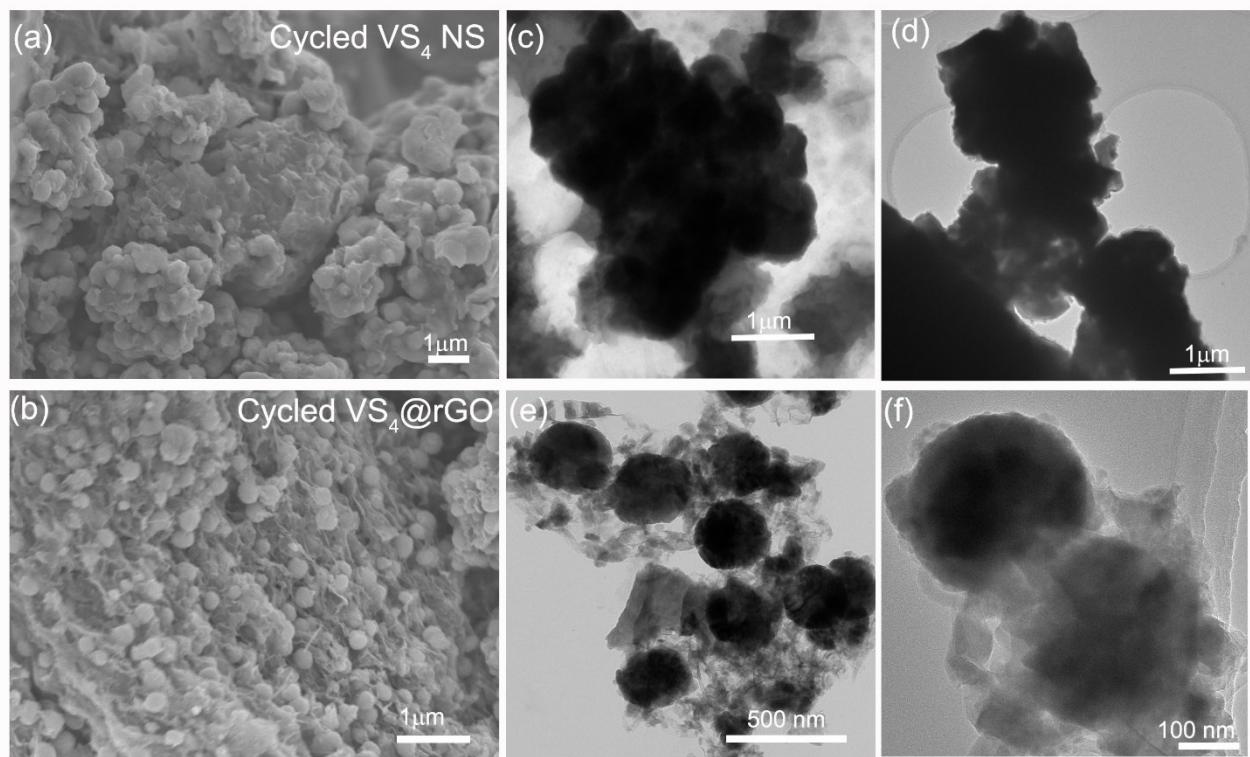


Fig. S7 (a) SEM and (c-d) TEM images of the VS<sub>4</sub> NS electrode after 1000 cycles at 10A g<sup>-1</sup>. (b) SEM and (e-f) TEM images of the VS<sub>4</sub>@rGO electrode after 1000 cycles at 10A g<sup>-1</sup>.

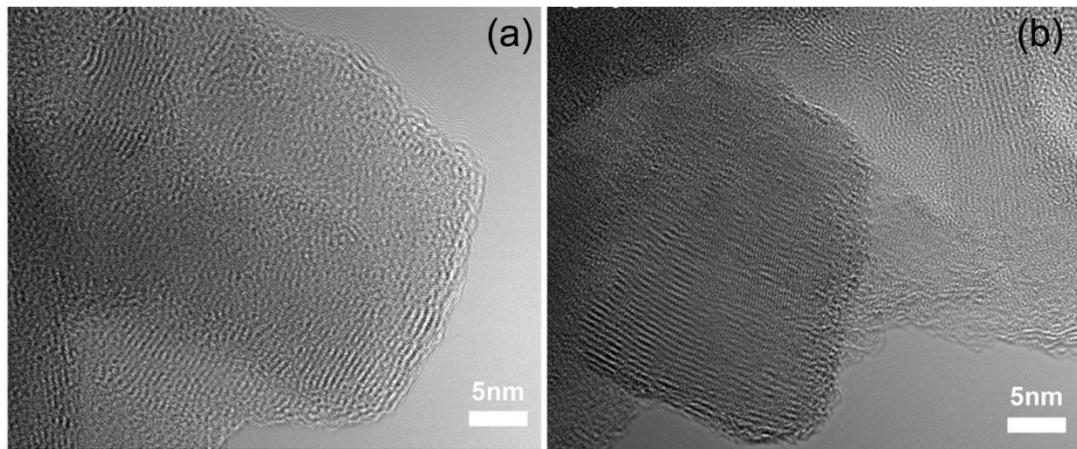
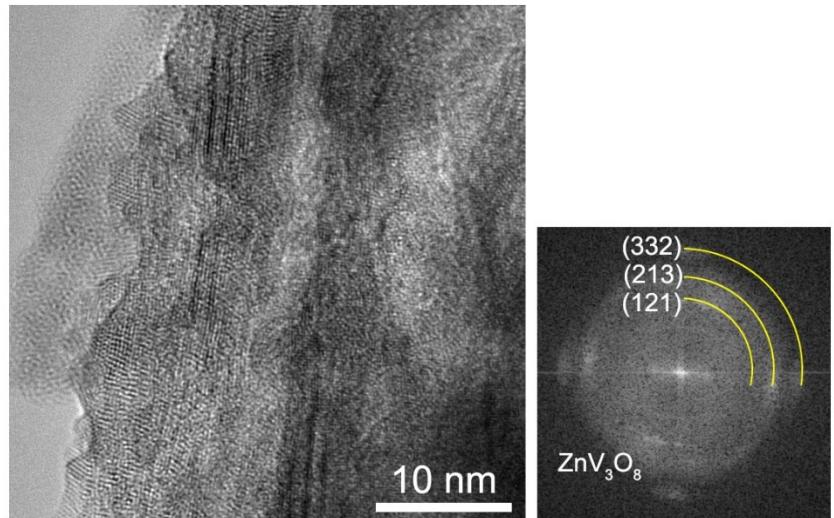


Fig. S8 HRTEM images of the cycled VS<sub>4</sub>@r-GO.



**Fig. S9** HRTEM image of the cycled VS<sub>4</sub>@rGO.

**Table S1.** Fitting results of the  $R_s$  and  $R_{ct}$  values in Nyquist curves (Fig. 3e)

Cycle Number	OCV ( <b>VS<sub>4</sub>@rGO</b> )	50 ( <b>VS<sub>4</sub>@rGO</b> )	OCV ( <b>VS<sub>4</sub> NS</b> )
$R_s$ ( $\Omega$ )	5.53	5.75	6.39
$R_{ct}$ ( $\Omega$ )	39.74	20.30	76.10

**Table S2** Comparison of operational voltage and discharge capacity of various cathode materials in aqueous Zn-ion batteries

Cathode material	Testing voltage (V)	Main operating voltage (V)	Discharge Capacity (mA h g <sup>-1</sup> )	Reference
VS <sub>4</sub> @rGO	0-1.8	0.89, 0.43	450@0.5A g <sup>-1</sup>	Our work
VS <sub>4</sub> @rGO	0.35-1.8	0.89, 0.54	225@0.5A g <sup>-1</sup>	1
VS <sub>4</sub>	0.2-1.6	0.89, 0.53	260@0.25A g <sup>-1</sup>	2
VS <sub>2</sub>	0.4-1.0	0.68, 0.6	178@0.1A g <sup>-1</sup>	3
rGO-VS <sub>2</sub>	0.2-1.8	0.92, 0.61	174@0.5A g <sup>-1</sup>	4
VS <sub>2</sub> @VOOH	0.4-1.0	0.68, 0.59	142.8@0.5A g <sup>-1</sup>	5
VS <sub>2</sub>	0.4-1.0	0.69, 0.58	168@0.5A g <sup>-1</sup>	6
VS <sub>2</sub>	0.4-1.0	0.75, 0.58	136.8@0.5A g <sup>-1</sup>	7
p-V <sub>2</sub> O <sub>5</sub>	0.3-1.5	0.91, 0.54	242@0.5 A g <sup>-1</sup>	8
V <sub>2</sub> O <sub>3</sub> /C	0.3-1.5	0.92, 0.56	272.4@0.5 A g <sup>-1</sup>	9
Zn <sub>3</sub> V <sub>2</sub> O <sub>7</sub> (OH) <sub>2</sub> ·2H <sub>2</sub> O	0.2-1.8	0.85, 0.58	122@0.5 A g <sup>-1</sup>	10
Na <sub>2</sub> V <sub>6</sub> O <sub>16</sub> ·2.14H <sub>2</sub> O	0.2-1.6	0.73, 0.45	346@0.5 A g <sup>-1</sup>	11
(NH <sub>4</sub> ) <sub>2</sub> V <sub>6</sub> O <sub>16</sub> ·1.5H <sub>2</sub> O	0.4-1.6	0.70, 0.49	307@0.5 A g <sup>-1</sup>	12

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