

## Support Information

### **Binder-Free Hierarchical VS<sub>2</sub> Electrodes for High-Performance Aqueous Zn Ion Batteries towards Commercial Level Mass Loading**

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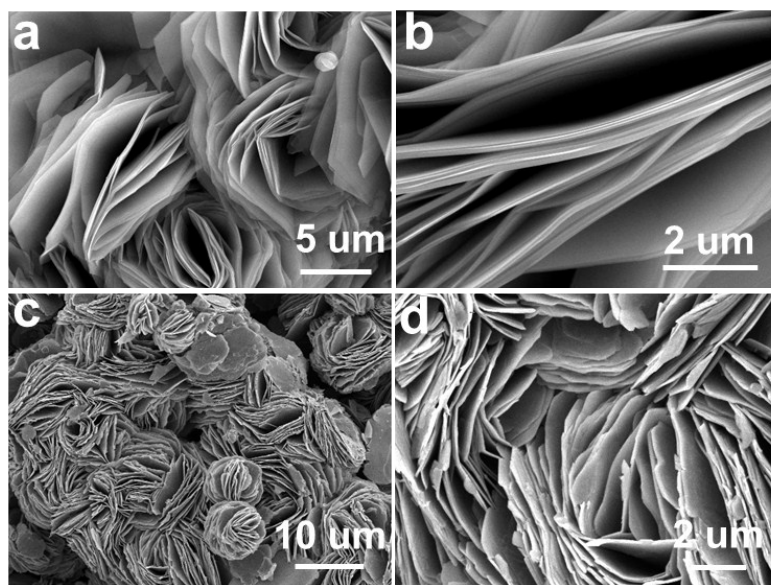


Figure S1. SEM images of (a, b)  $\text{VS}_2$ @SS electrodes, and (c, d)  $\text{VS}_2$  powder.

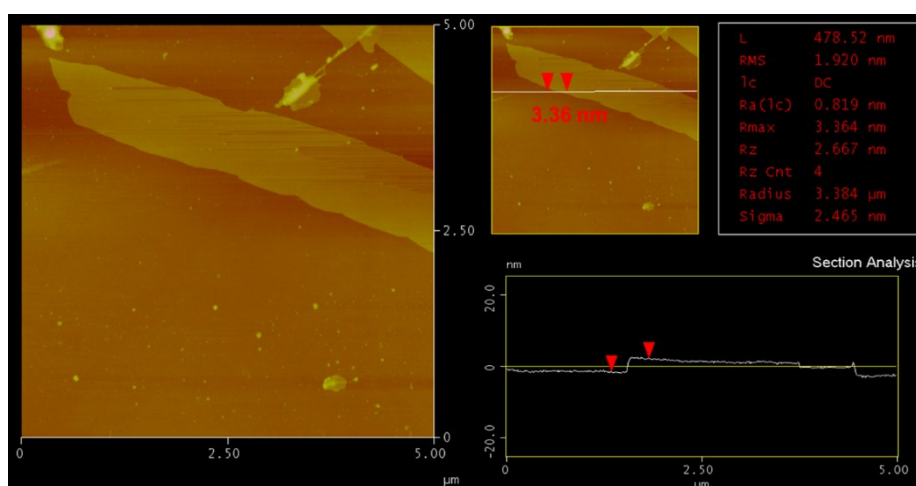


Figure S2. AFM images and the corresponding height profiles of individual  $\text{VS}_2$  nanosheets.

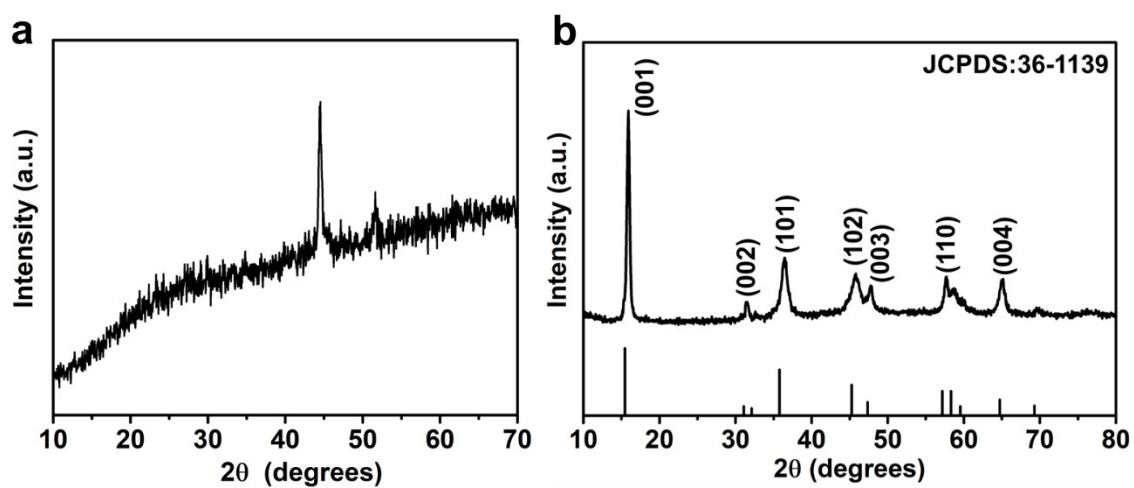


Figure S3. XRD patterns of (a) stainless steel mesh and (b)  $\text{VS}_2$  powder.

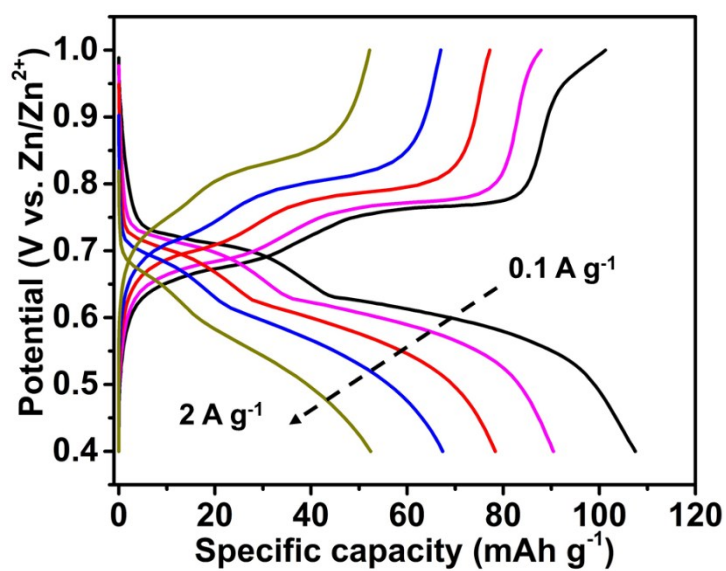


Figure S4. Galvanostatic discharge-charge curves of  $\text{VS}_2$  slurry-coated electrode.

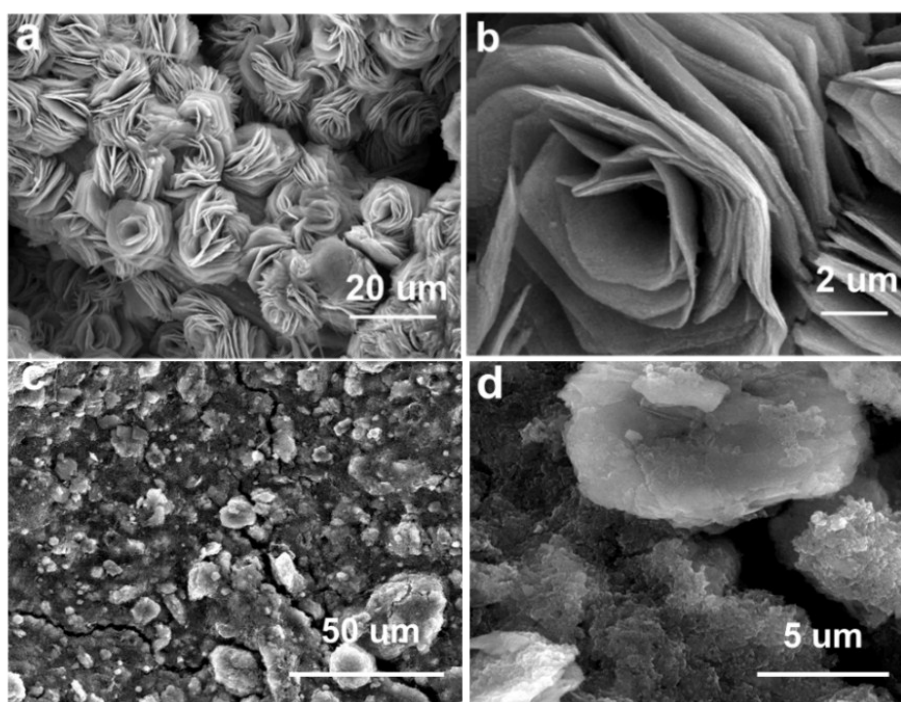
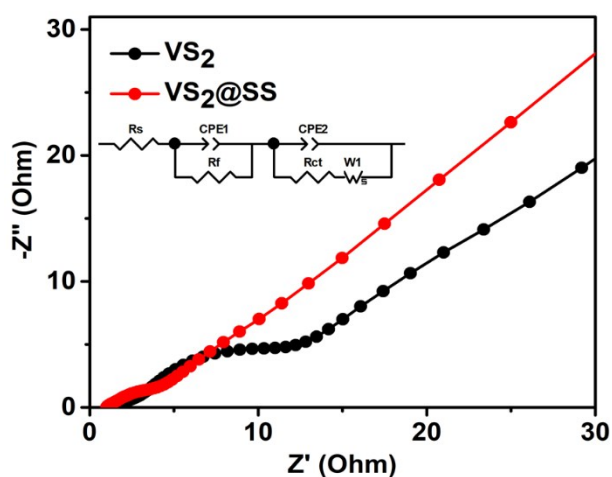


Figure S5. SEM images of (a, b) the  $\text{VS}_2@\text{SS}$  electrode after long-term cycling, and (c, d) the conventional  $\text{VS}_2$  slurry-coated electrode after cycling.



	$R_s$	$R_f$	$R_{ct}$
$\text{VS}_2@\text{SS}$ electrode	1.1	0.3	1.9
$\text{VS}_2$ slurry-coated electrode	1.8	1	6

Figure S6. Nyquist plots of  $\text{VS}_2@\text{SS}$  and  $\text{VS}_2$  slurry-coated electrodes. Inset is the equivalent circuit. The table is the comparison of two electrodes in the calculated values. This circuit is composed of  $R_s$  (the solution resistance),  $R_{ct}$  (resistance for charge transfer), CPE (constant-phase element) and W (Warburg diffusion process). The parallel circuit  $R_f$  CPE1 could be related either to the SEI layer or to the bulk resistance. Similar results have been reported in aqueous Li-ion batteries with  $\text{Li}_2\text{SO}_4$  and  $\text{LiNO}_3$  electrolyte or superconcentrated LiTFSI electrolyte<sup>1-3</sup>.

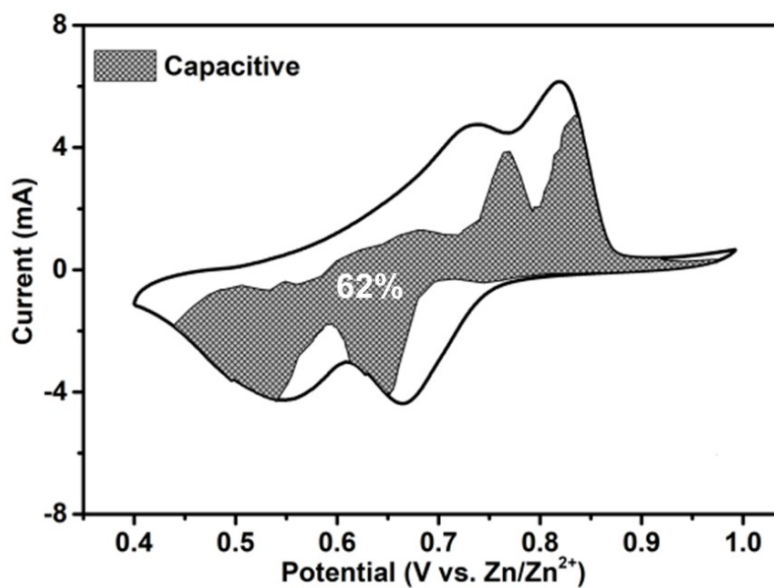


Figure S7. The pseudocapacitive component occupied 62% of total charge storage of  $\text{VS}_2@\text{SS}$  electrode at a scan rate of  $0.4 \text{ mV s}^{-1}$ .

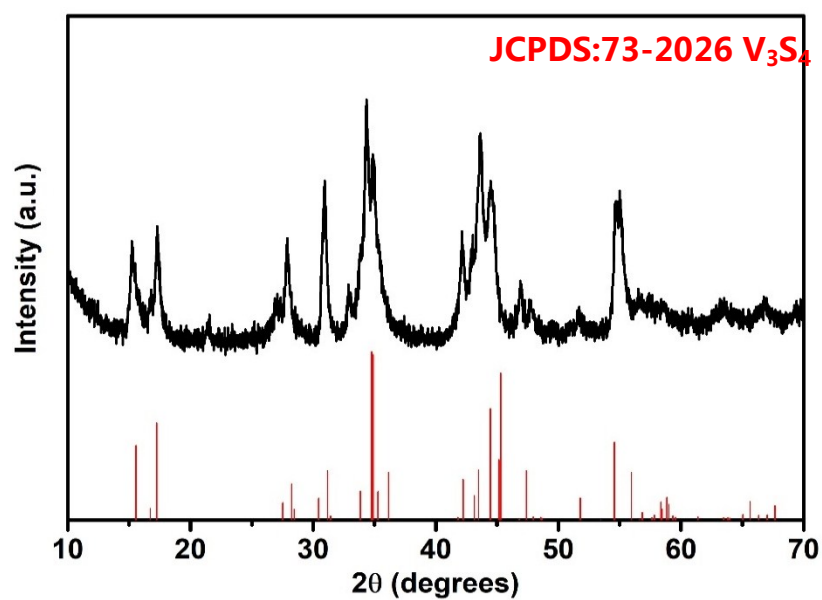
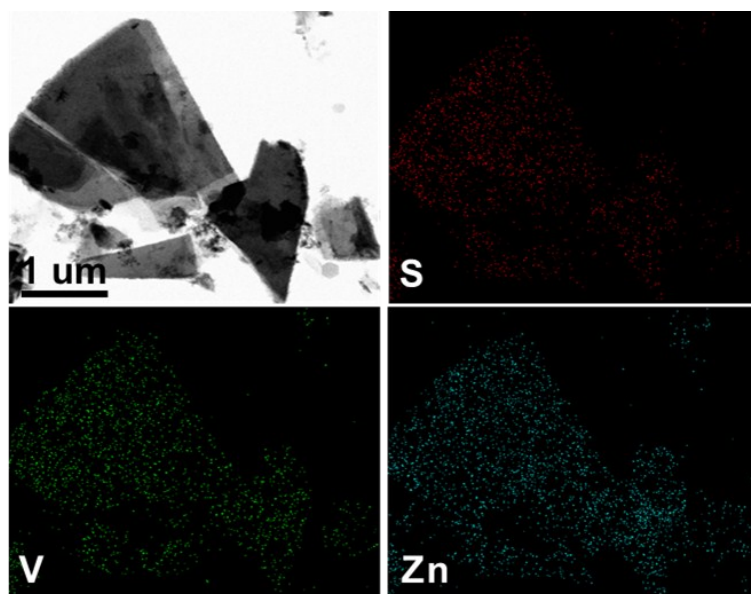


Figure S8. XRD pattern of the fully discharged  $\text{VS}_2@\text{SS}$  electrode.

Figure S9. STEM and the elemental mapping images of the fully discharged VS<sub>2</sub>@SS



electrode.

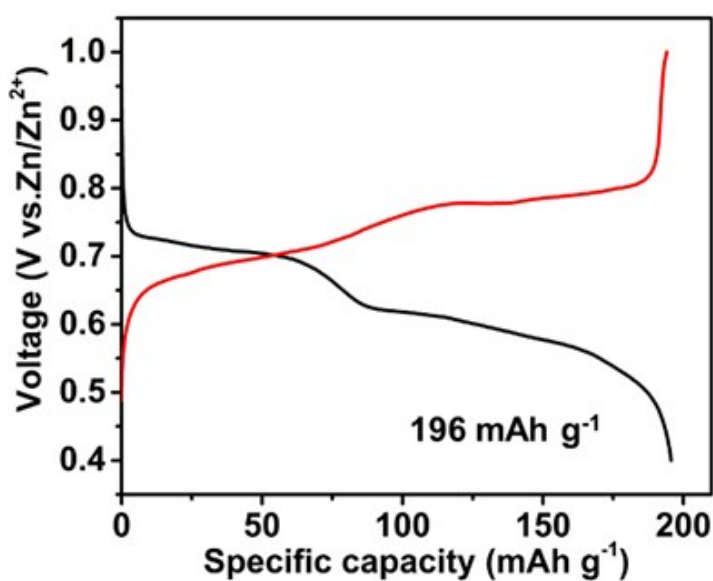


Figure S10. The charge and discharge curves of VS<sub>2</sub>@SS electrode at 50 mA g<sup>-1</sup>.

The intercalation number of Zn<sup>2+</sup> (x+y) is calculated according to the discharge curve at 50 mA g<sup>-1</sup> and based on the following equations:

$$x + y = \frac{nC_a M}{C_0}$$

(n is the number of electron exchanged, C<sub>a</sub> = 196 mAh g<sup>-1</sup>, M = 115 g mol<sup>-1</sup>, C<sub>0</sub> = 26.8 nm M<sup>-1</sup>)



<sup>1</sup>,  $F = N_A \cdot e = 96500 \text{ C mol}^{-1}$ ,  $1 \text{ Ah} = 3600 \text{ C}$ )

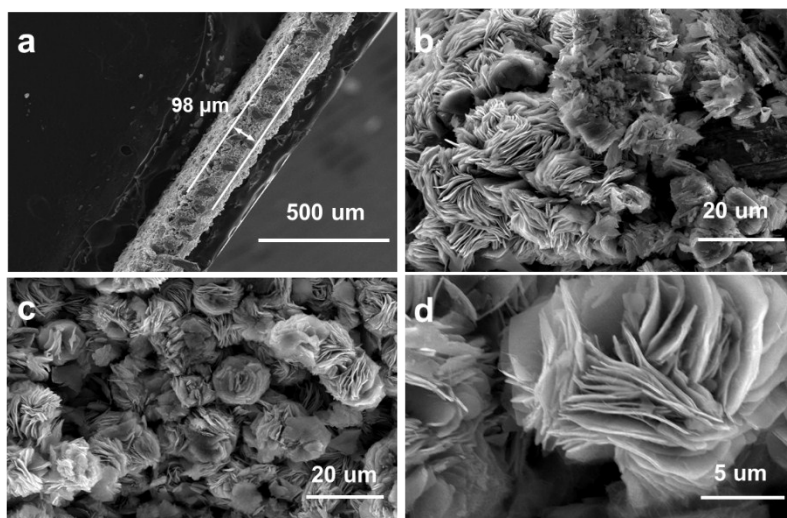


Figure S11. (a) and (b) The cross-section SEM images of VS<sub>2</sub>@SS electrode with a mass loading of about 13 mg cm<sup>-2</sup>. (c) and (d) SEM images of the electrode after cycling.

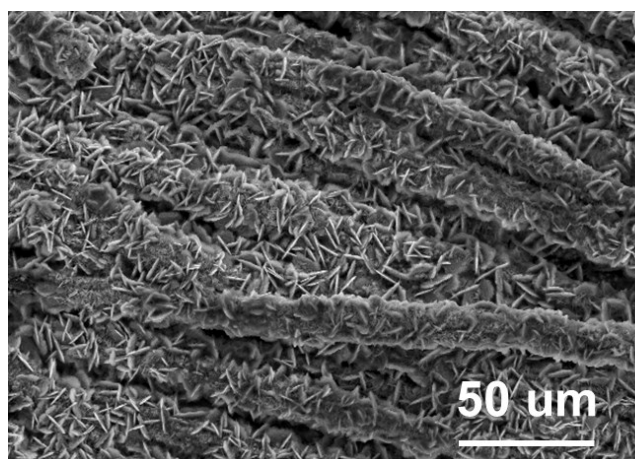


Figure S12. SEM image of the Zn electrodeposited carbon cloth.

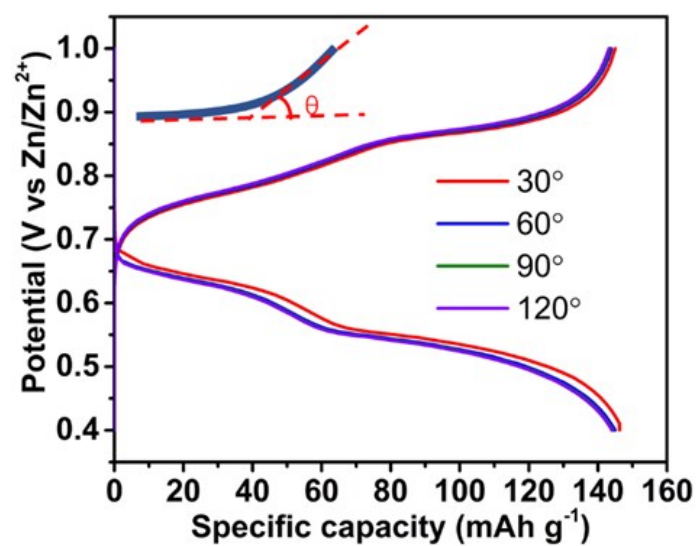


Figure S13. The galvanostatic charge/discharge curves of the flexible solid-state battery under various bending states at  $0.5 \text{ A g}^{-1}$ . Inset is the definition of the bending angle.

1. R. B. Shivashankaraiah, H. Manjunatha, K. C. Mahesh, G. S. Suresh and T. V. Venkatesha, *Journal of the Electrochemical Society*, 2012, **159**, A1074-A1082.
2. H. Manjunatha, K. C. Mahesh, G. S. Suresh and T. V. Venkatesha, *Electrochimica Acta*, 2011, **56**, 1439-1446.
3. L. M. Suo, D. Oh, Y. X. Lin, Z. Q. Zhuo, O. Borodin, T. Gao, F. Wang, A. Kushima, Z. Q. Wang, H. C. Kim, Y. Qi, W. L. Yang, F. Pan, J. Li, K. Xu and C. S. Wang, *Journal of the American Chemical Society*, 2017, **139**, 18670-18680.