Engineering Sulphur Vacancy in VS_2 as High Performing Zinc-ion Battery with High Cyclic Stability

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Computational Methods

The calculations were performed using density-functional theory (DFT) with the generalized gradient approximation (GGA) of PBE.[1] The projector augmented wave potentials [2] were employed as implemented in the vienna ab-initio simulation package (VASP).[3] The plane-wave cutoff energy of 450 eV was chosen. The convergence for energy and the Hellmann–Feynman force were chosen as 1×10^{-6} eV and 0.01 eV, respectively. For the structural optimization of layer VS_2 (001) with a $4 \times 2 \times 1$ supercell containing two S-V-S sandwich layers, the $3 \times 3 \times 1$ k-points sampling was employed. The spin polarization was considered throughout the calculations. To avoid the periodic interaction, a vacuum thickness of 15 Å for the sulfur vacancy and Zn adsorption studies were built. During the optimization process, the lattice constants for the supercell were fixed and all atoms were allowed to relax until the force on each atom was less than 0.001 eV $Å^{-1}$.

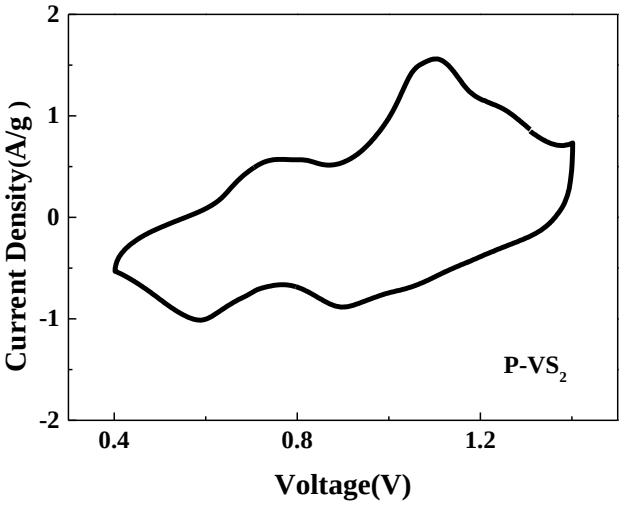

Figure S1: CV profile of P-VS_2

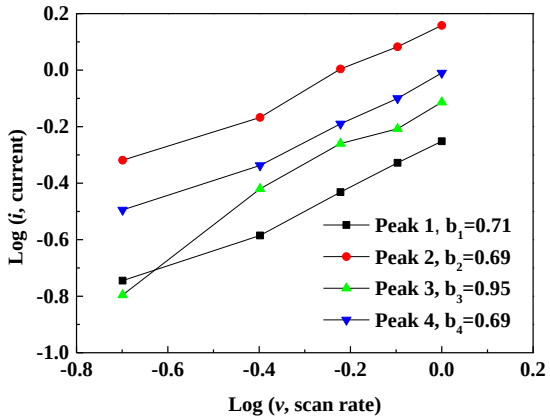

Figure S2: The corresponding plots of log (peak current) vs. log (scan rate) at each peak of P-VS_2

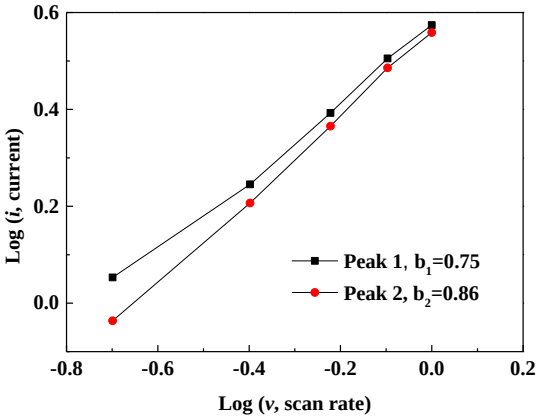

Figure S3: The corresponding plots of log (peak current) vs. log (scan rate) at each peak of D-VS_2

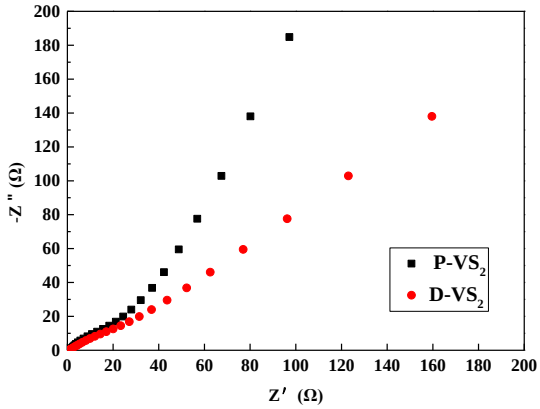

Figure S4: The Nyquist curve of the electrochemical impedance measurements (EIS) of P-VS_2 and D-VS_2 electrode.

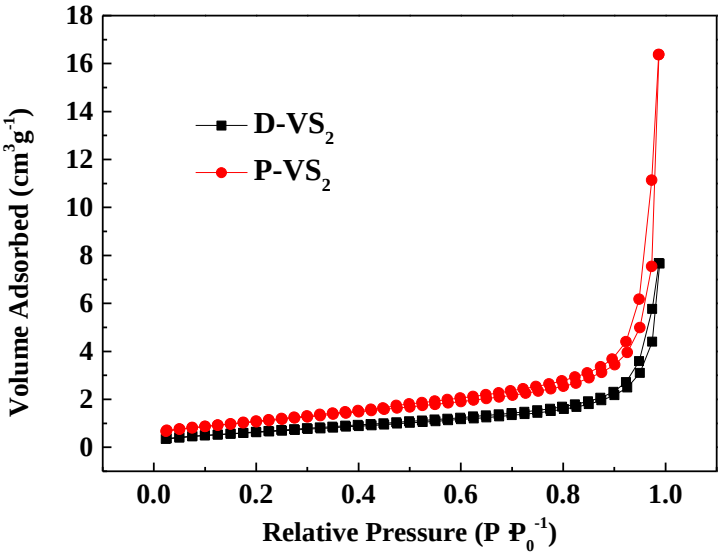

Figure S5: The Nitrogen adsorption–desorption isotherm of P-VS_2 and D-VS_2 electrode.

Table S1: The performance comparison of D-VS_2 with other metal dichalcogenides.

Type	Electrolyte	Cycle Stability(h)	Specific capacity (mAh g^{-1})	Ref.
D- VS_2 nanosheet	**$ZnSO_4$**	94% after 524h	**262**	**This work**
VS_2 nanosheet	**$ZnSO_4$**	98% after 109.5h	**159.1**	**4**
VS_2@SS nanosheet	**$ZnSO_4$**	90% after 336h	**187**	**5**
E-MoS_2 nanosheet	**$ZnSO_4$**	98.6% after 197.4h	**202.6**	**6**
MoS_{2-x} nanosheets	**$Zn(CF_3SO_3)_2$**	87.8% after 191.2h	**138.6**	**7**
MoS_2 nanosheets	**$Zn(CF_3SO_3)_2$**	98.1% after 95.2h	**168**	**8**
MoS_2−O nanosheets	**$Zn(CF_3SO_3)_2$**	-	**232**	**9**
Bi_2S_3 nanoparticles	**$ZnSO_4$**	100% after 161h	**161**	**10**
$Na_{0.14}TiS_2$	**$Zn(CF_3SO_3)_2$**	98% after 720h	**120**	**11**

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