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Supporting Information

Hierarchical Spheres Constructed by ultrathin VS_2 Nanosheets for Sodium-Ion Batteries

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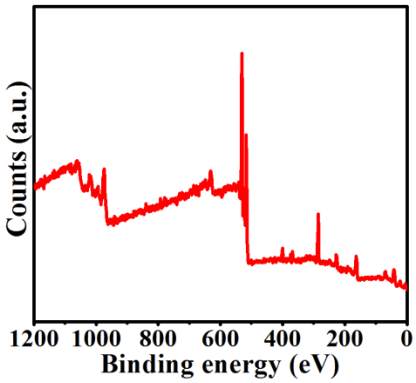

Figure S1. Full survey spectrum of hierarchical VS_2 spheres

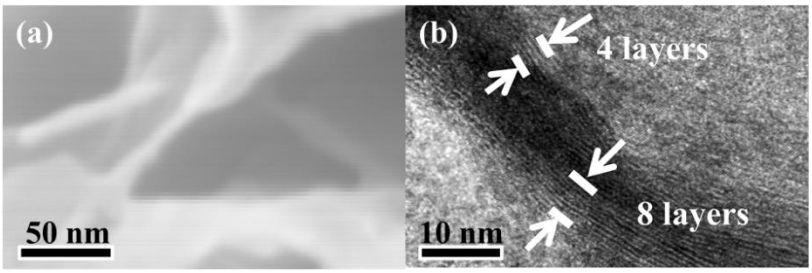

Figure S2. (a) SEM image and (b) HRTEM image of hierarchical VS_2 spheres.

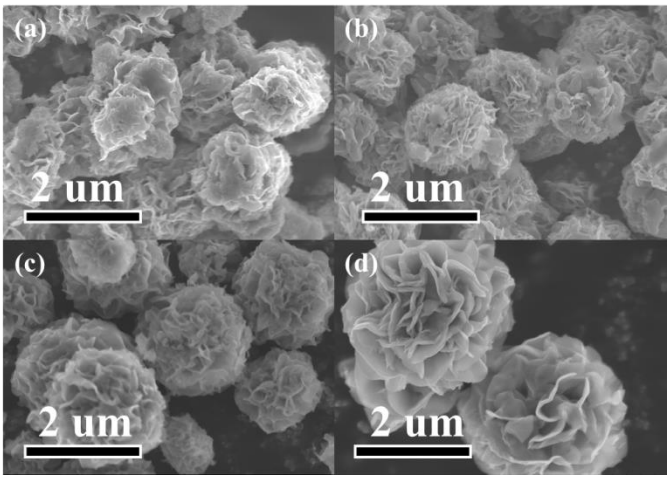

Figure S3. SEM images of hierarchical VS_2 spheres prepared in (a) 6, (b) 12, (c) 18 and (d) 24 h.

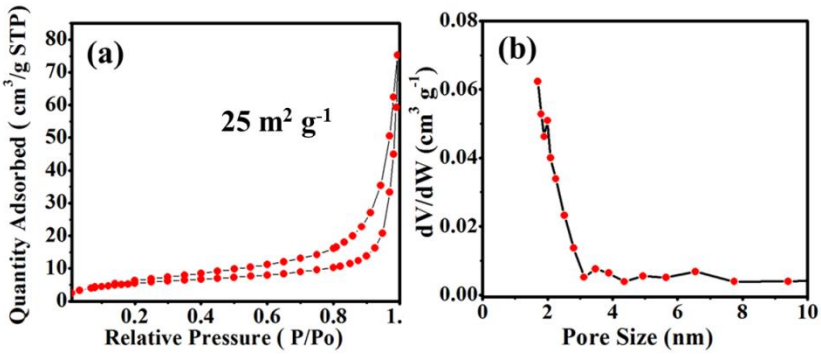

Figure S4. Nitrogen adsorption-desorption isotherms, and (b) pore size distribution of hierarchical VS_2 spheres.

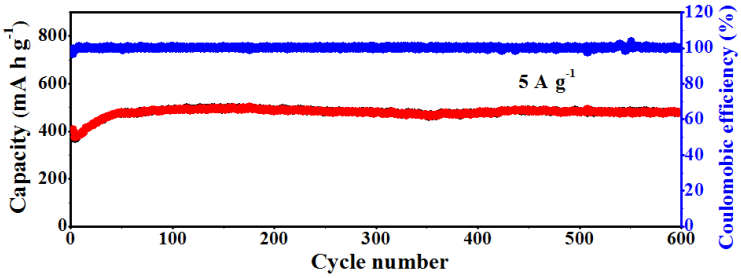

Figure S5 Cycling performance of hierarchical VS_2 spheres at 5 A g^{-1}.

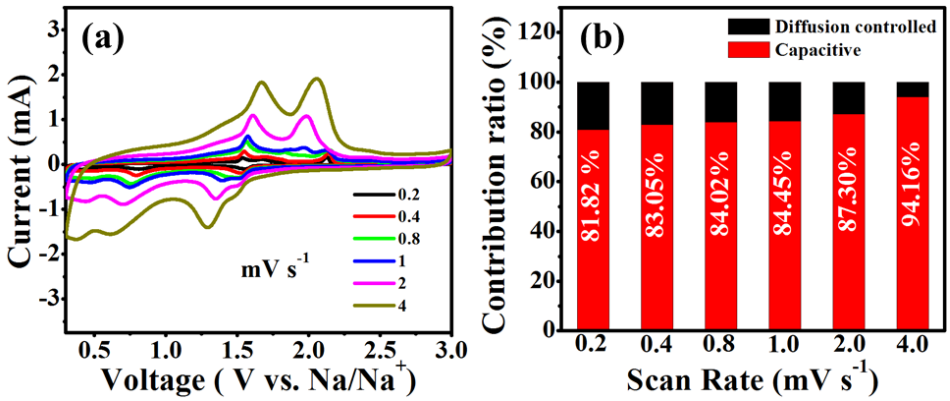

Figure S6 (a) CV curves at various rates from 0.2 to 4 mV s^{-1}, and (b) contribution ratio of the capacitive and diffusion-controlled at different corresponded scan rates for hierarchical VS_2 spheres.

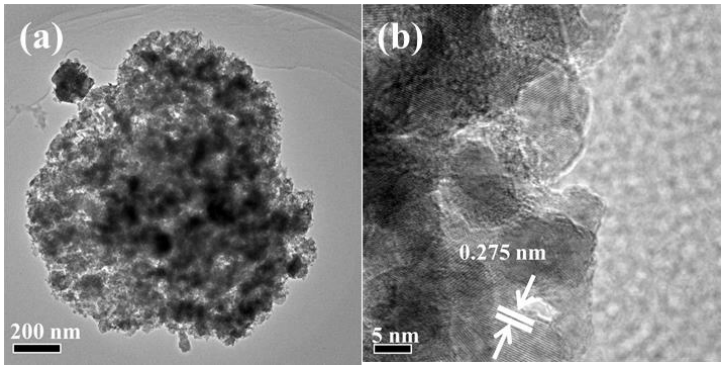

Figure S7 (a) TEM image and (b) HRTEM images of hierarchical VS_2 spheres after 100 cycles at 0.2 A g^{-1}.

Table S1. Electrochemical performance of VS_2-based anode materials for SIBs.

Materials	**Current density (A g^{-1})**	**Capacity (mA h g^{-1})**	**References**
Hierarchical VS_2 nanosheets	0.1 1	700 after 100 cycles 500 after 200 cycles	34
VOOH coated VS_2 flowers	0.2	330 after 150 cycles	35
Hierarchical flower-like VS_2	0.1	600 after 50 cycles	31
Layer by layer stacked VS_2	0.2 5	250 after 100 cycles 204 after 600 cycles	30
3D hierarchical VS_2 microrods	0.2	350 after 200 cycles	33
VS_2 single-crystal nanosheets	0.2	403 after 200 cycles	32
This work	**0.2** **2** **5**	**720 after 100 cycles** **565 after 1000 cycles** **479 after 600 cycles**	

Table S2. The fitted results of hierarchical VS_2 spheres based on the equivalent circuit model.

	Rs	Rf	Rct
Before cycling	6	0	137
After 20 cycles	3	20	26

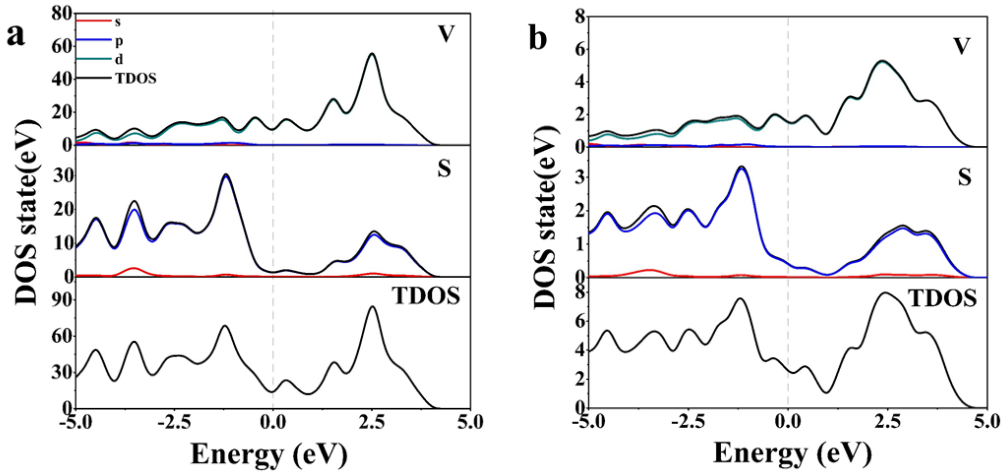

Figure S8 DOS and DOS for (a) H-monolayer and (b) H-bulk of VS_2, respectively.

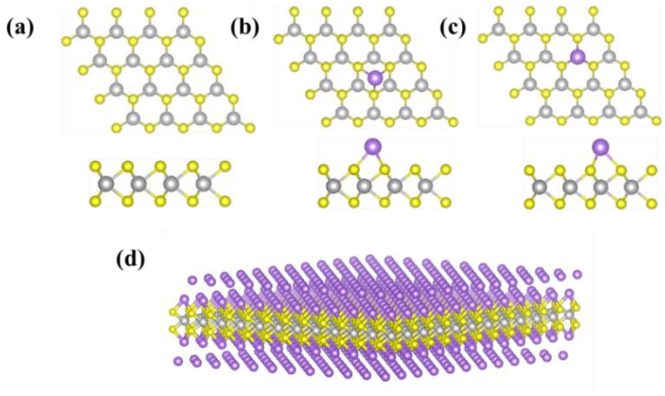

Figure S9. (a) Schematics of free H-VS_2 monolayer; (b,c) Na adsorption on H_{center} site and T site of V atom in top and side views, respectively; (d) The structure with two-layer Na atoms adsorbed on both surfaces of VS_2 monolayer (Na_4VS_2).

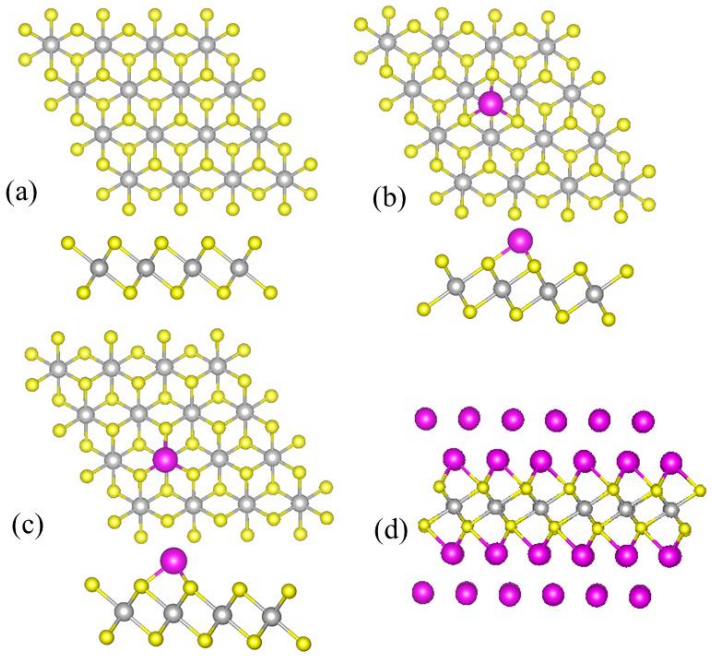

Figure S10. (a) Schematics of free T-VS_2 monolayer; (b,c) Na adsorption on H_{center} site and T site of V atom in top and side views, respectively; (d) The structure with two-layer Na atoms adsorbed on both surfaces of VS_2 monolayer (Na_4VS_2).

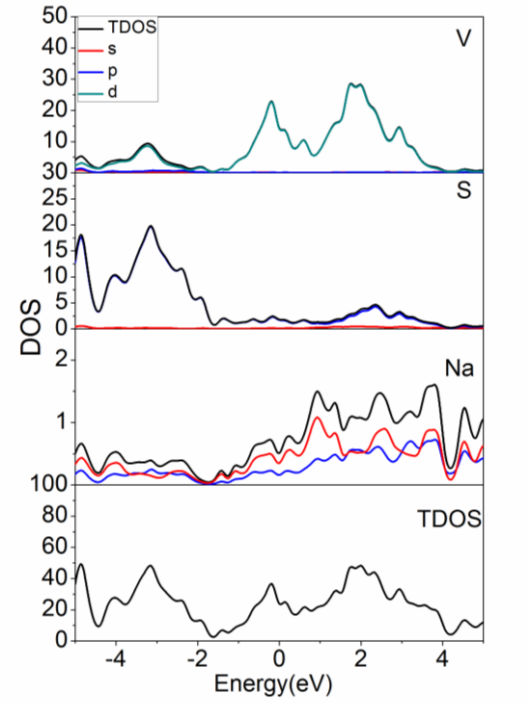

Figure S11. PDOS and DOS for T-Na_2VS_2 monolayer.

Table S3. The average energy changes per Na atom and the capacities of 1 Na, 32 Na, 48 Na, 64 Na, and 96 Na adsorbed on the VS_2 monolayer ($V_{16}S_{32}$) in H phase.

	E_b (eV)	**Capacity (m A h g^{-1})**
1Na (T site)	-1.91	14.56
1Na (H_{center} site)	-1.88	14.56
32 Na	-0.65	466
48 Na	-0.43	699
64 Na	-0.34	932
96Na	-0.23	1398

Table S4. The average energy changes per Na atom and the capacities of 1 Na, 18 Na, 27Na, and 36 Na adsorbed on the VS_2 monolayer (V_9S_{18}) in T phase.

	E_b (eV)	**Capacity (m A h g^{-1})**
1Na (T site)	-1.40	25.9
1Na (H_{center} site)	-1.38	25.9
18 Na	-0.64	466
27 Na	-0.45	699
36 Na	-0.32	932
54 Na	-0.22	1398