ARTICLE

Supporting Information

Delocalized Spin-Polarized Electrons Enhance the Performance of Ion Batteries

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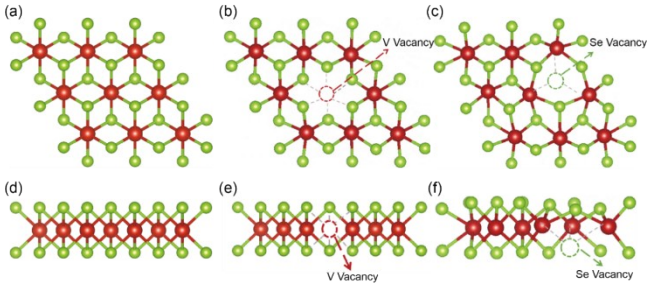

Figure S1. (a) (d) Top and side view of monolayer VSe_2. (b) (e) Top and side view of monolayer VSVV. (c) (f) Top and side view of monolayer VSSV.

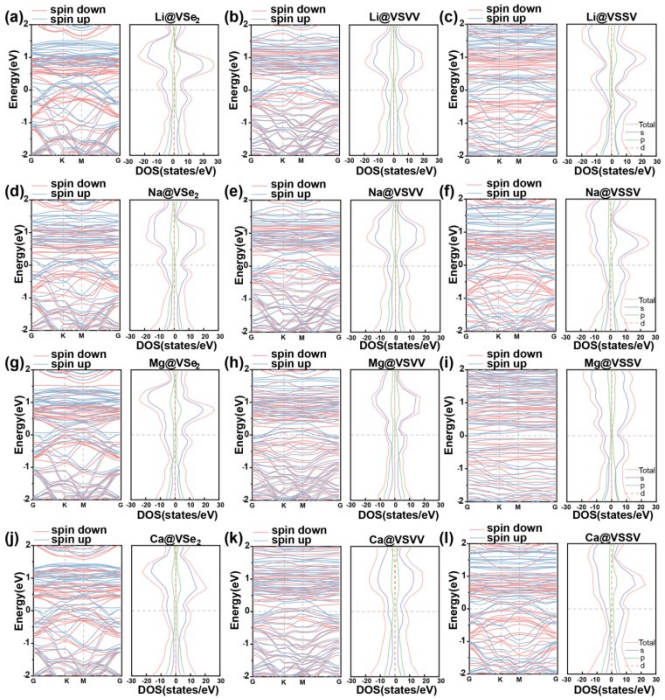

Figure S2. (a-c) Energy band and DOS of VSe_2, VSVV, and VSSV adsorbed with Li^+. (d-f) Energy band and DOS of VSe_2, VSVV, and VSSV (g-i) Energy band and DOS of VSe_2, VSVV, and VSSV adsorbed with Mg^{2+} (j-l) Energy band and DOS of VSe_2, VSVV, and VSSV adsorbed with Ca^{2+}.

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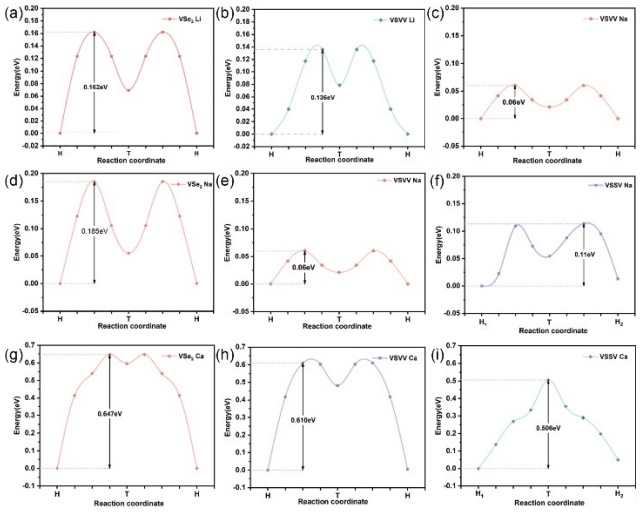

Figure S3. (a-c) Diffusion barriers for Li^+ ions in VSe_2, VSVV, and VSSV. (d-f) Diffusion barriers for Na^+ ions in VSe_2, VSVV, and VSSV. (g-i) Diffusion barriers for Ca^{2+} ions in VSe_2, VSVV, and VSSV.

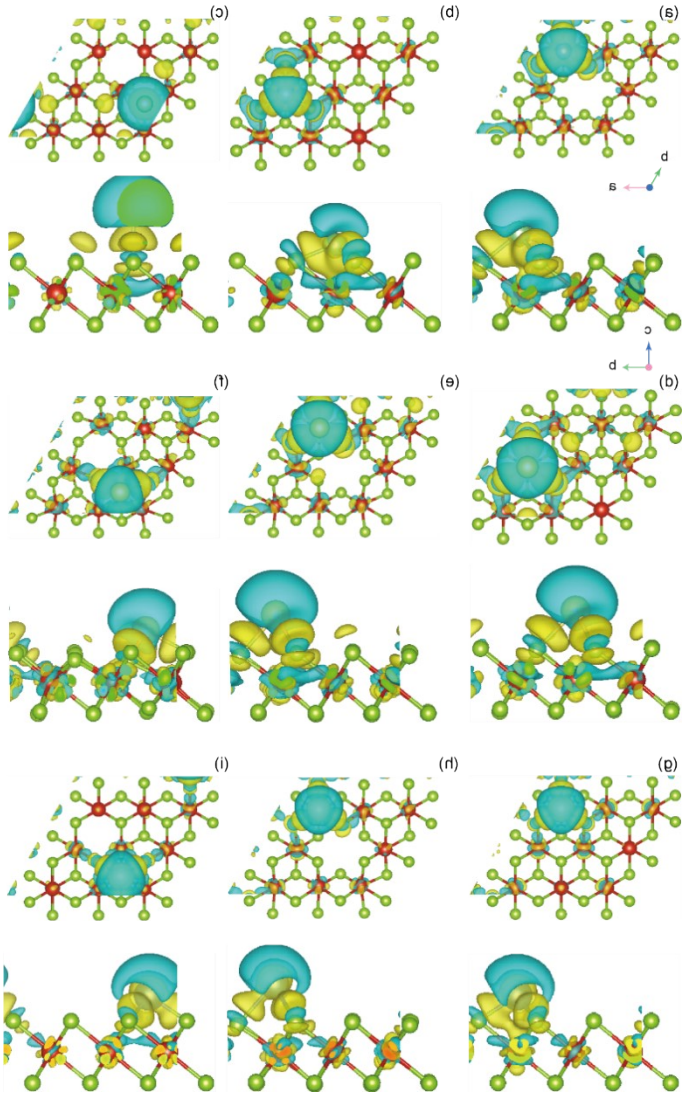

Figure S4. CDD of (a–o) Li^+, Na^+, and Ca^{2+} on VSe_2, VSVV, and VSSV. The blue and yellow regions represent the accumulation and depletion of charges. The upper level is the top view and the lower level is the side view. The isosurface level is set to 0.001 e/Å3

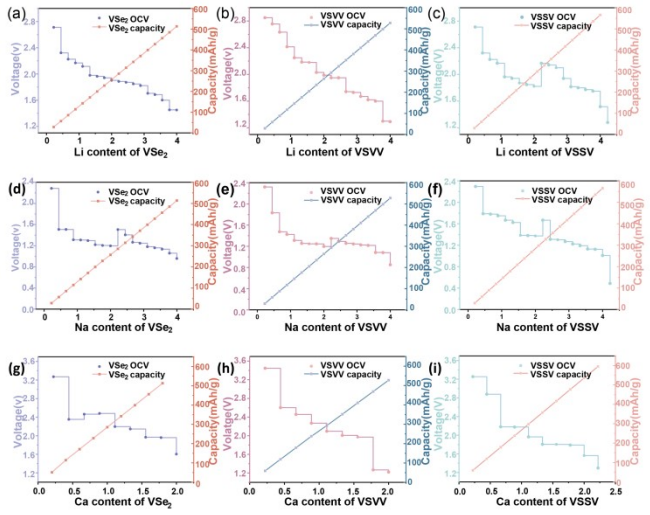

Figure S5. (a-c) OCV of Li^+ Adsorption on VSe_2, VSVV, and VSSV. (d-f) OCV of Na^+ Adsorption on VSe_2, VSVV, and VSSV. (g-i) OCV of Ca^{2+} Adsorption on VSe_2, VSVV, and VSSV.

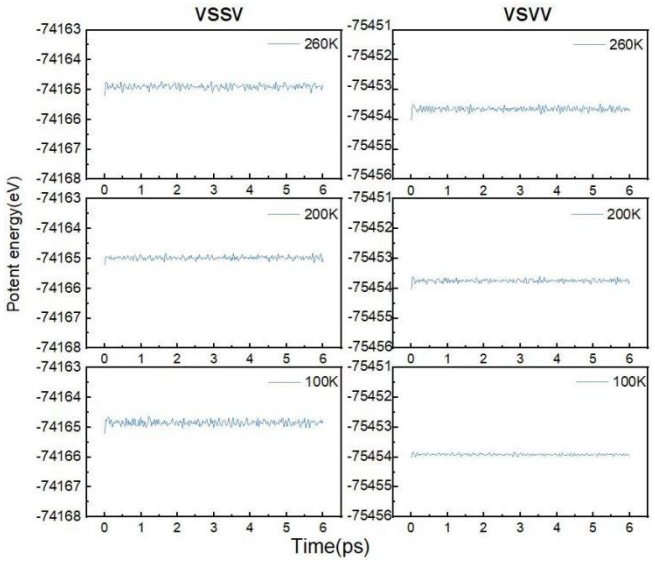

Figure S6. The structural stability of VSVV and VSSV evaluated by molecular dynamics simulations at 260 K, 200 K and 100 K.

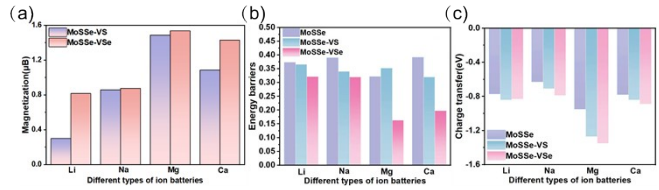

Figure S7.(a) Magnetic moments of S-vacancy MoSSe (MoSSe-VS) and Se-vacancy MoSSe (MoSSe-VSe); (b) Comparison of energy barriers for MoSSe-VS and MoSSe-VSee; (c) Comparison of charge transfer for MoSSe-VS and MoSSe-VSe.

Table S1. Stability of different metal cations adsorbed on the VSe_2, VSVV, and VSSV monolayer.

	Li	Na	Mg	Ca
VSe_2 H site	√	√	√	√
VSe_2 T site	√	√	√	√
VSe_2 T'site	×	×	×	×
VSe_2 B site	×	×	×	×
VSVV H site	√	√	√	√
VSVV T site	√	√	√	√
VSVV T' site	×	×	×	×
VSVV T'' site	×	×	×	×
VSVV B site	×	×	×	×
VSSV H site	√	√	√	√
VSSV T site	√	√	√	√
VSSV T' site	×	×	×	×
VSSV B site	×	×	×	×

Table S2. Adsorption energies of different metal cations at the H and T sites on the VSe_2, VSVV, and VSSV monolayer.

	Li	Na	Mg	Ca
VSe_2 H site	-2.859eV	-2.228eV	-2.08eV	-1.92eV
VSe_2 T site	-2.788eV	-2.222eV	-1.83eV	-1.32eV
VSVV H site	-2.85eV	-2.31eV	-2.02eV	-2.446eV
VSVV T site	-2.53eV	-2.68eV	-1.77eV	-1.83eV
VSSV H site	-2.59eV	-2.18eV	-2.14eV	-1.75eV
VSSV T site	-2.71eV	-2.29eV	-2.35eV	-2.253

Table S3. The energy barriers, charge transfer, capacity ratios, and OCV of different types of ion batteries adsorbed on three types of materials

Substrate material	Different types of ion batteries	Energy barriers (eV)	Charge transfer (eV)	Capacity (mAh/g)	Voltage (V)
VSe_2	Li	0.162	0.771	513.28	1.45
	Na	0.185	0.837	513.28	1.176
	Mg	**0.457**	**1.05**	**1026.57**	**1.295**
	Ca	0.647	1.189	513.28	1.6
VSVV	Li	0.136	0.863	527.59	1.292
	Na	0.12	0.97	527.59	0.85
	Mg	**0.253**	**1.25**	**1055.19**	**1.2**
	Ca	0.61	1.21	527.59	1.219
VSSV	Li	0.12	0.95	565.54	1.279
	Na	0.11	0.98	565.54	0.686
	Mg	**0.218**	**1.44**	**1131.1**	**0.898**
	Ca	0.506	1.28	593.52	1.297

Table S4. For VSe_2, VSVV, and VSSV batteries, after the introduction of vacancies, the percentage decreases in their ion diffusion energy barriers and OCV, as well as the percentage increases in their transferred charges and theoretical capacities.

Substrate material	Different types of ion batteries	The percentage of performance improvement compared with VSe_2			
		Energy barriers	Charge transfer	Capacity	Voltage
VSVV	Li	25.9%	12%	2.8%	10.8%
	Na	35%	15.8%	2.8%	27.7%
	Mg	**44.6%**	**19%**	**2.8%**	**7%**
	Ca	5.7%	1.7%	2.8%	23.8%
VSSV	Li	16%	23.2%	10%	11.8%
	Na	40%	17%	10%	41.7%
	Mg	**52.3%**	**37%**	**10%**	**30.6%**
	Ca	21.8%	7.6%	15.6%	18.9%

Table S5. Comparison of Key Performance Indicators Between VSe_2 with Selenium Vacancies (VSSV) and Reported Two-Dimensional Energy-Storage Materials.

2D Material	Open-Circuit Voltage (V)	Energy Barrier (eV)	Reference
Li^+@VSSV	0.95	0.12	-
$ScTe_2$ monolayer	1.04	0.32	[1]
1T-$ZrSe_2$ monolayer	0.76	0.22	[2]
Dumbbell silicene	0.38	0.19	[3]
$Li^+Mn_2NO_2$	2.82	0.28	[4]

Notes and references

[1] T. Dey, S. Chowdhury, S. G. Kang, P. Sen, B. C. Gupta and J. C. Mahato, Comput. Mater. Sci., 2025, 253.
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[3] D. D. Vargas, G. L. Cardoso, P. C. Piquini, R. Ahuja and R. J. Baierle, ACS Appl. Mater. Interfaces, 2022, 14, 47262–47271.
[4] P. Gong, X. Zhang, F. Liu, K. Yao and S. Zhu, Surfaces and Interfaces, 2022, 32.