Supporting Information

High-Stable Rare Earth YS_2 and ScS_2 Monolayers for Potassium-Ion Batteries: First-Principles Calculations

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Table S1. The total energies (eV) for 2H and 1T phased MS_2 monolayers, and the relative energy (ΔE, eV) between these two phases.

Material	Energy (eV)		ΔE (eV)	Stable phase
	2H	1T	$= E_{1T} - E_{2H}$	
ScS_2	–74.88	–75.18	–0.31	1T
YS_2	–76.82	–76.74	0.08	2H
TiS_2	–80.72	–82.30	–1.58	1T
ZrS_2	–86.46	–88.61	–2.15	1T
HfS_2	–92.078	–94.52	–2.44	1T
VS_2	–82.32	–82.25	0.07	2H
NbS_2	–90.08	–89.70	0.38	2H
TaS_2	–96.41	–96.10	0.31	2H
CrS_2	–82.23	–78.36	3.87	2H
MoS_2	–90.92	–87.54	3.38	2H
WS_2	–98.52	–94.98	3.54	2H
MnS_2	–77.55	–78.15	–0.60	1T
CoS_2	–65.46	–66.82	–1.37	1T
NiS_2	–58.20	–60.28	–2.08	1T
PdS_2	——	——	——	——
PtS_2	–57.95	–65.01	–7.06	1T

Table S2. The formation entropies (ΔH_f, eV) and single K adsorption energies (E_{ads}, eV) on MS_2.

Material	Formation energies ΔH_f (eV)	Adsorption energies (E_{ads} (eV)		
		T site	H site	B site
ScS_2	–4.29	–3.28	–3.35	——
$YS_{2(2H)}$	–4.62	–2.61	–2.67	——
$YS_{2(1T)}$	–4.56	–3.44	–3.51	——
TiS_2	–2.51	–2.01	–2.05	–2.01
ZrS_2	–4.65	–1.61	–1.68	–0.98
HfS_2	–4.75	–1.43	–1.49	–1.49
$VS_{2(2H)}$	–2.53	–2.33	–2.33	–1.86
$VS_{2(1T)}$	–2.52	–1.73	–1.74	–1.73
NbS_2	–3.17	–2.19	——	——
TaS_2	–3.15	–2.09	–2.10	–2.10
CrS_2	–2.06	–0.97	——	——
MoS_2	–2.60	–0.40	–0.36	–0.11
WS_2	–2.45	–0.16	–0.11	0.07
MnS_2	–1.95	——	–1.76	——
CoS_2	–0.99	——	–2.2	——
NiS_2	–1.34	——	–1.59	——
PdS_2	–0.85	–1.28	–1.28	——
PtS_2	–1.11	–0.98	–0.98	–0.90

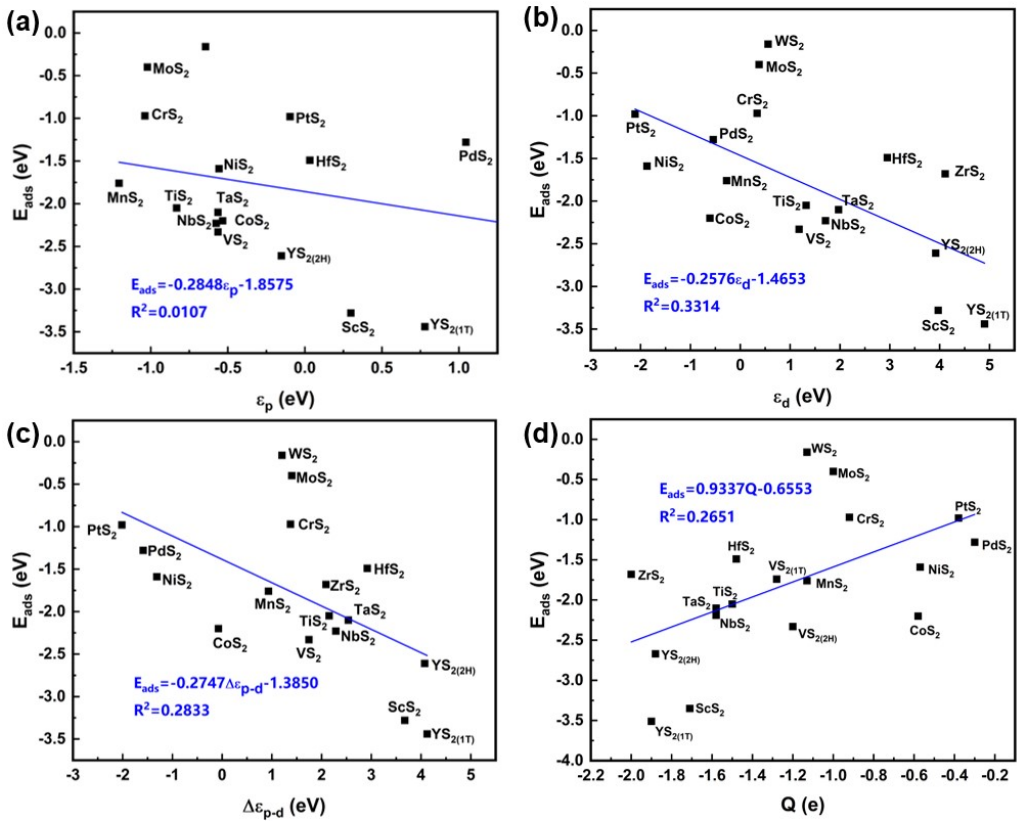

Fig. S1 Relationships between K adsorption energies (E_{ads}) and electronic characters of (a) *p*-band center, (b) *d* band center, (c) the difference between *d* band center and *p*-band center ($\Delta(d\text{-}p)$), and (d) Bader charges (Q).

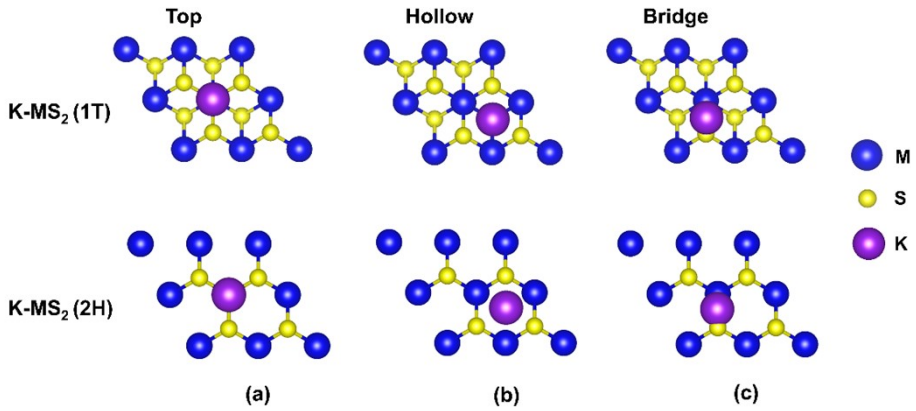

Fig. S2. The possible adsorption sites for K on the surface of 1T and 2H phased MS_2 monolayers.

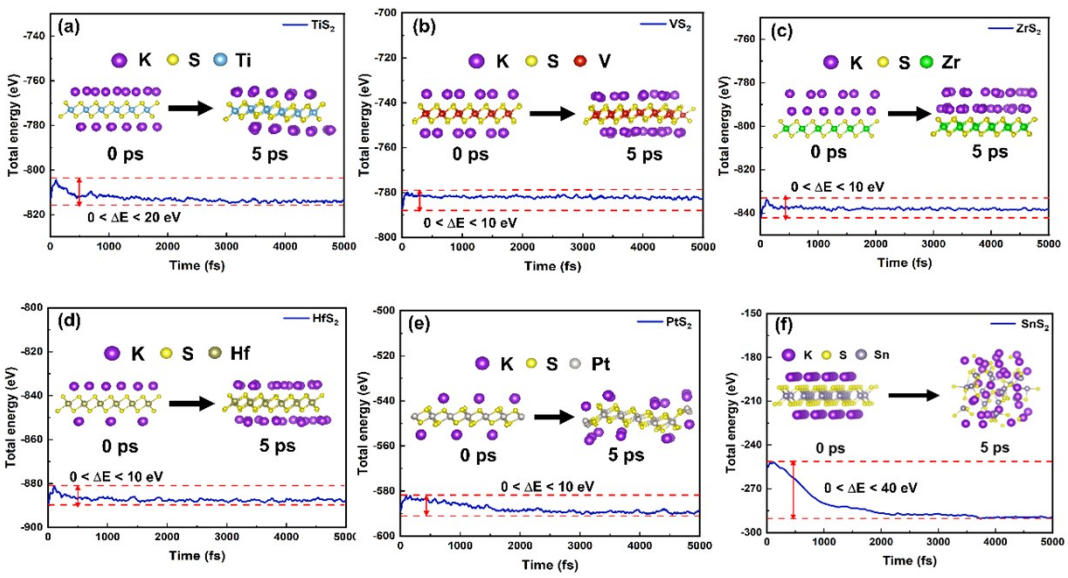

Fig. S3 Snapshots of trajectories for potassificated (a) TiS_2, (b) VS_2, (c) ZrS_2, (d) HfS_2 (e) PtS_2, and (f) SnS_2 nanosheets following 5 ps AIMD simulation at 300 K. It is noteworthy that the experimentally reported SnS_2 material was also selected to demonstrate the universality of using $E_a$ds and ΔH_f to distinguish between potashization mechanisms (intercalation vs. conversion). According to our definition, SnS_2 falls within the white region above the diagonal in Fig. 3a, suggesting that it undergoes a conversion mechanism. Moreover, our AIMD results confirm that the SnS_2 monolayer is significantly degraded at approximately 2.5 ps. This finding is fully consistent with experimental observations—electrochemical tests and X-ray diffraction (XRD) analysis have shown that SnS_2 transforms during the charging process, converting from SnS_2 to K_2S and Sn. During the first discharge, the characteristic diffraction peaks of SnS_2 completely vanish and are replaced by those of K_2S and Sn. [Synthesis of SnS_2 ultrathin nanosheets as anode materials for potassium ion batteries. Chemical Research in Chinese Universities, 2021, 37, 311-317].

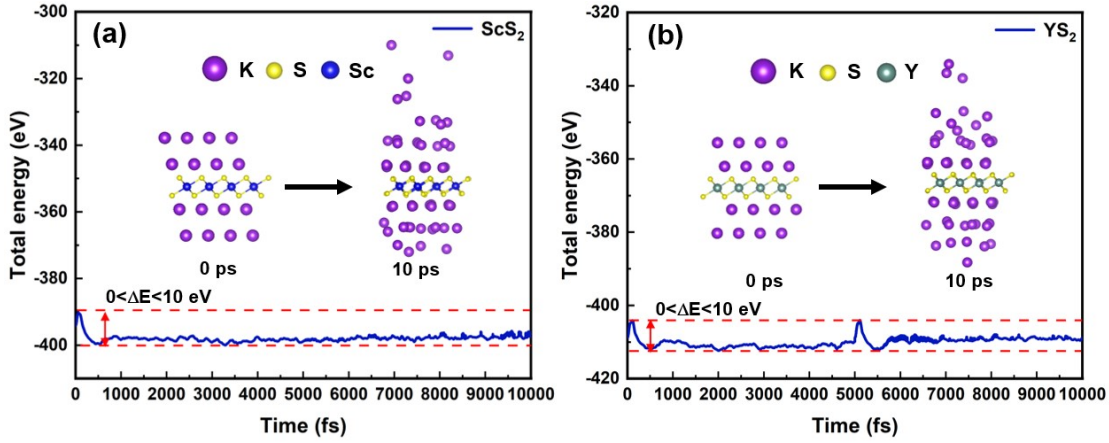

Fig. S4 Snapshots of trajectories for potassificated (a) ScS_2 and (b) YS_2 nanosheets following 10 ps AIMD simulation at 300 K.

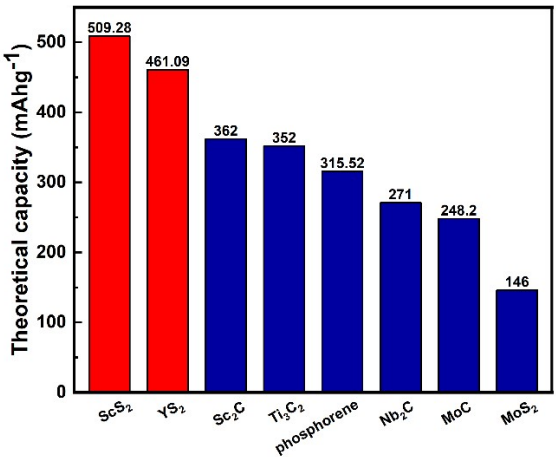

Fig. S5 Comparison of the theoretical capacities of various monolayer electrode materials for potassium-ion batteries.

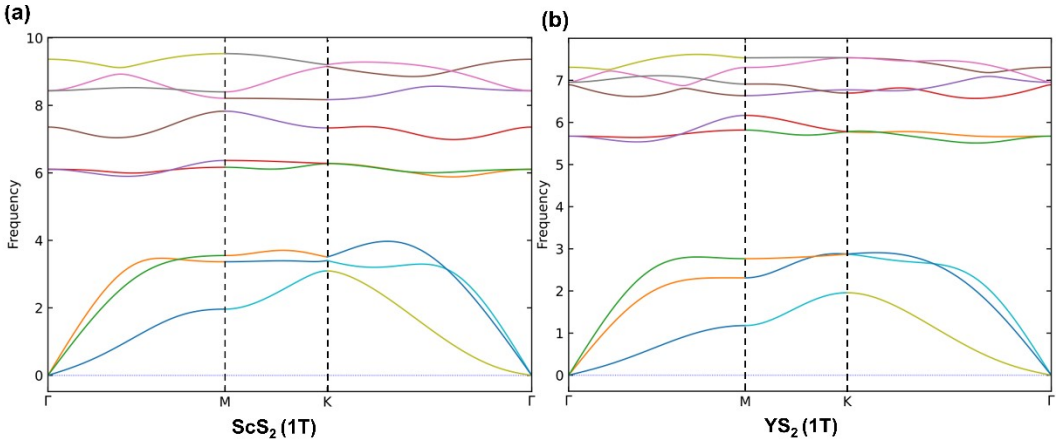

Fig. S6 Calculated phonon band structures of 1T phased ScS_2 and YS_2.