Supporting information for:

Comparative studies of hexagonal boron phosphide/V_2CS_2 heterostructure and its monolayers as metal-ion battery anodes

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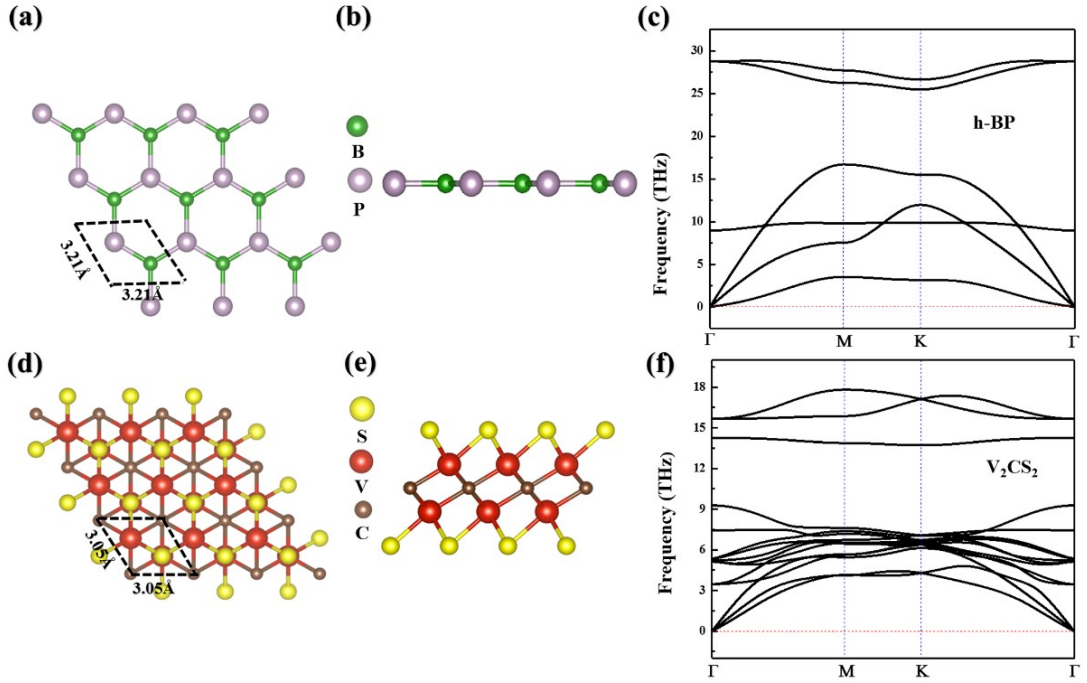

Figure S1. Top and side views of (a, b) hexagonal boron phosphide (h-BP) and (d, e) V_2CS_2, respectively. Phonon dispersion spectra of (c) h-BP monolayer and (f) V_2CS_2 monolayer, respectively.

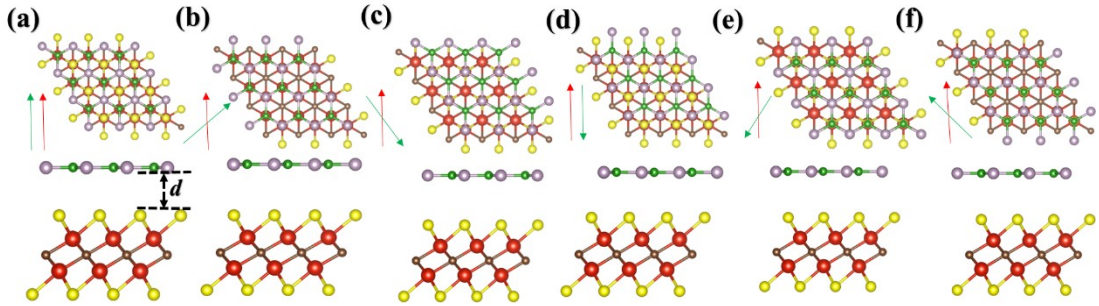

Figure S2. Top and side views of six stacking configurations of h-BP/V_2CS_2. The rotation angles of h-BP with respect to V_2CS_2 are set to (a) 0°, (b) 60°, (c) 120°, (d) 180°, (e) 240°, and (f) 300°, respectively. The d is the interface distance.

Table S1. The interface formation energy E_f and distance d of configurations of h-BP/V_2CS_2 are arranged from small to large.

	configuration					
	0°	180°	300°	240°	60°	120°
$E_{f(\mathrm{eV})}$	-0.13	-0.10	0.18	0.28	0.29	0.36
d (Å)	3.20	3.23	3.43	3.51	3.57	3.65

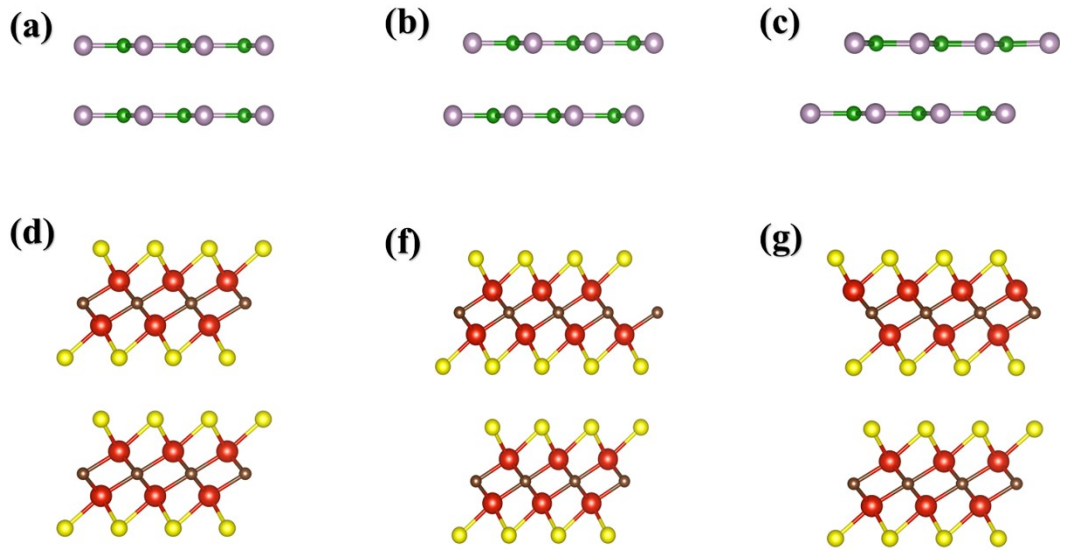

Figure S3. Three stacking configurations of Dh-BP (a, b, c) and DV_2CS_2 (d, f, g). The (b) and (d) are the easiest ways to combine h-BP and V_2CS_2 monolayers to double layers, respectively.

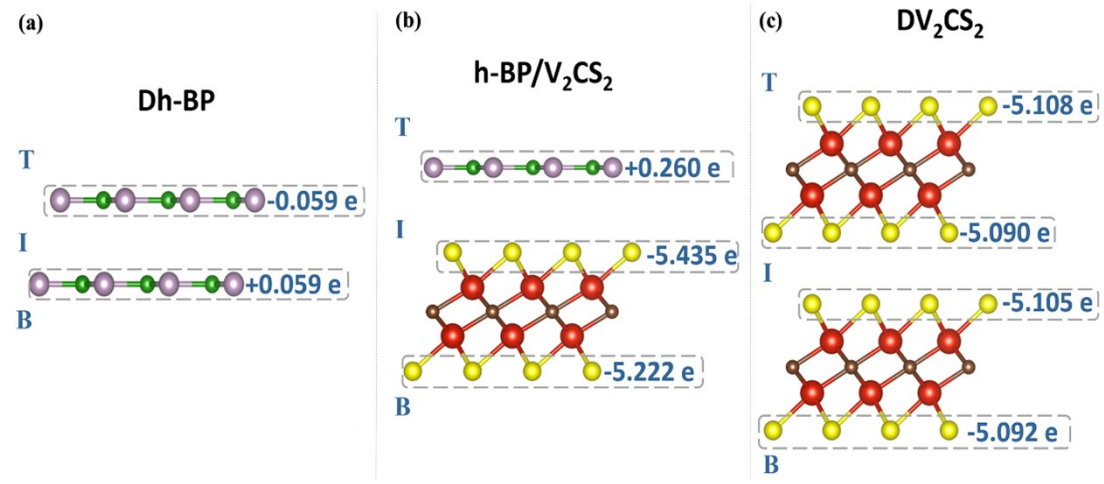

Figure S4. The accumulated charge of each surface layer atoms of (a) Dh-BP, (b) h-BP/V_2CS_2 and (3) DV_2CS_2 before adsorbing a metal atom.

Table S2. Comparison of adsorption energy of heterostructures adsorbing a metal atom on three surfaces. Bold number indicates the most negative adsorption energy on three surfaces in the same system.

site	T	I	B	reference
Li@MoS_2 /V_2CS_2	-0.521	**-1.920**	-1.652	[1]
Na@MoS_2/V_2CS_2	-0.355	**-1.715**	-1.394	[1]
Mg@MoS_2/V_2CS_2	1.157	**-1.025**	0.096	[1]
K@MoS_2/V_2CS_2	-0.790	**-1.955**	-1.733	[1]
Li@MoS_2/Ti_2CF_2	-0.32	**-1.68**	-1.01	[2]
Li@MoS_2/Ti_2CO_2	-0.31	**-2.22**	-2.01	[2]
Li@BlueP/Ti_2CS_2	-1.178	**-2.368**	-2.21	[3]
Li@VS_2/Ti_2CO_2	~-1.8	**~-2.6**	~-2.0	[4]
Na@VS_2/Ti_2CO_2	~-1.7	**~-2.3**	~-1.9	[4]
Mg@VS_2/Ti_2CO_2	~-0.6	**~-2.8**	~-1.1	[4]
Li@h-BP/V_2CS_2	-1.017	**-1.998**	-1.668	this work
Na@h-BP/V_2CS_2	-0.950	-1.545	**-1.575**	this work
Mg@h-BP/V_2CS_2	1.028	**-1.314**	-0.279	this work
Ca@h-BP/V_2CS_2	-0.410	**-2.642**	-2.003	this work

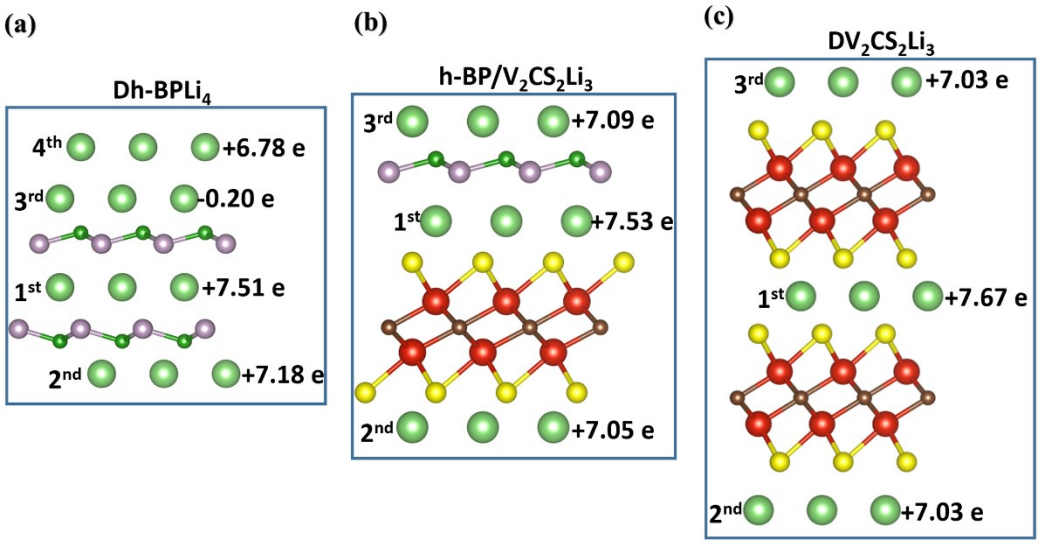

Figure S5. Number of electrons gained (-) or lost (+) per layer of Na atoms in the hosts that absorbs the most layers of Na atoms.

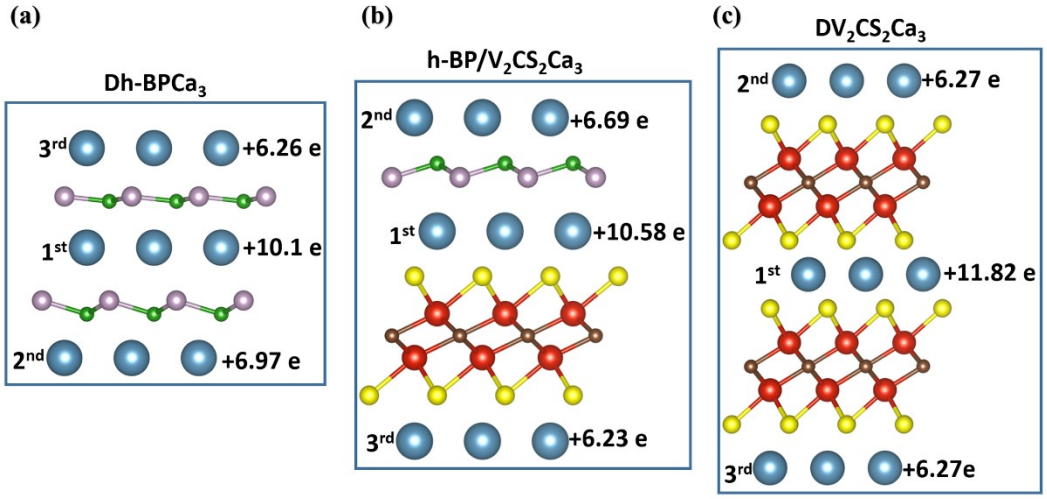

Figure S6. Number of electrons lost (+) per layer of Ca atoms in the hosts that absorbs the most layers of Ca atoms.

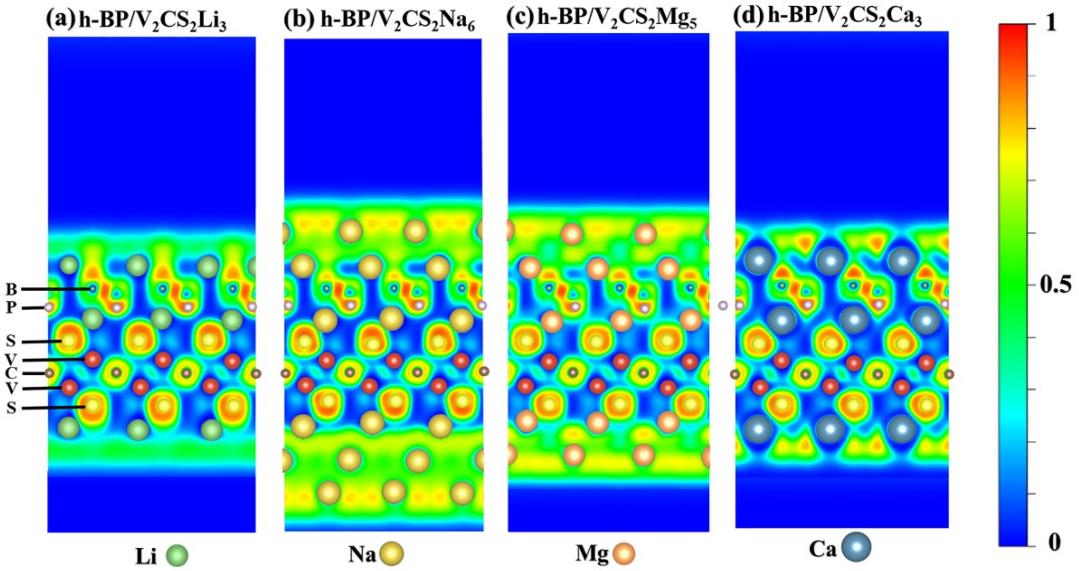

Figure S7. ELF plots of the (110) slices of h-BP/V_2CS_2 heterostructure loading (a) Li, (b) Na, (c) Mg and (d) Ca at maximum concentration.

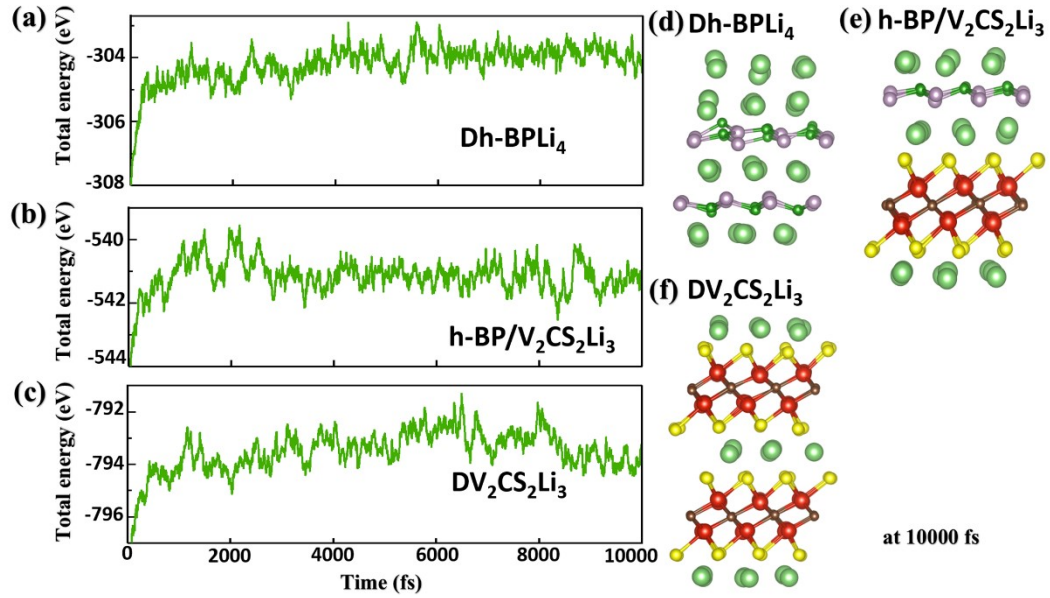

Figure S8. Total energy fluctuations within 10000 fs and snapshots at 10000 fs of (a, d) Dh-$BPLi_4$, (b, e) h-$BP/V_2CS_2Li_3$ and (c, f) $DV_2CS_2Li_3$ calculated by AIMD heating at 300 K.

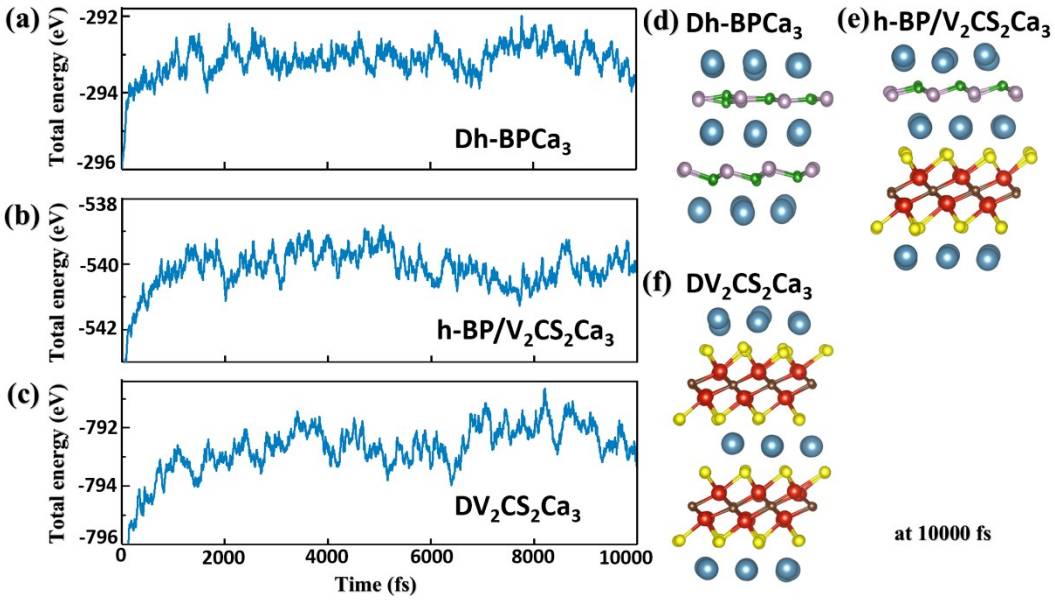

Figure S9. Total energy fluctuations within 10000 fs and snapshots at 10000 fs of (a, d) Dh-$BPCa_3$, (b, e) h-$BP/V_2CS_2Ca_3$ and (c, f) $DV_2CS_2Ca_3$ calculated by AIMD heating at 300 K.

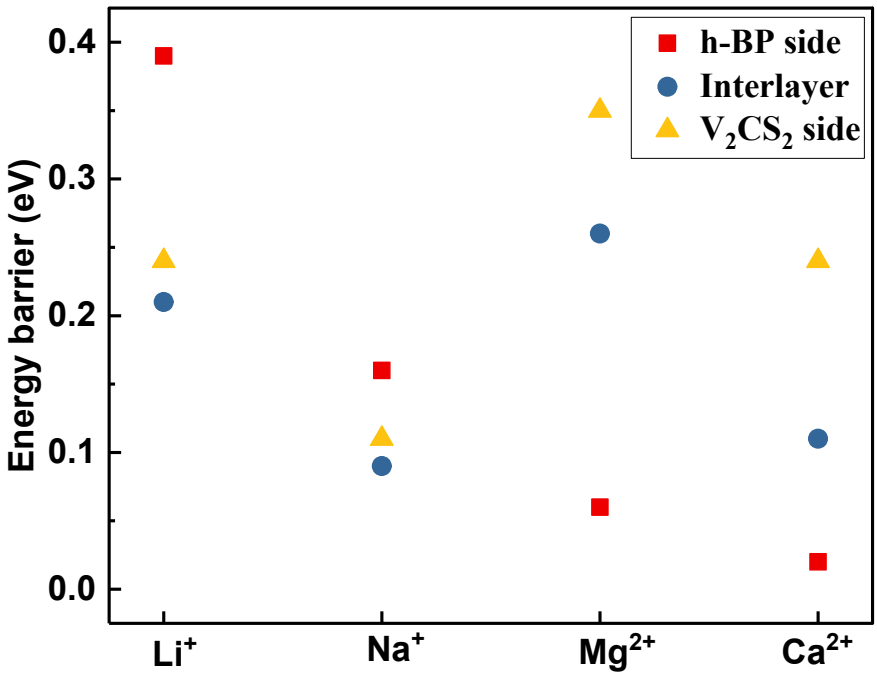

Figure S10. The energy barriers of a metal ion in the interlayer and both sides of the heterostructure.

1. X. Yuan, Z. Chen, B. Huang, Y. He and N. Zhou, *The Journal of Physical Chemistry C*, 2021, **125**, 10226-10234.
2. J. Li, Q. Peng, J. Zhou and Z. Sun, *The Journal of Physical Chemistry C*, 2019, **123**, 11493-11499.
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4. N. Li, Y. Li, X. Zhu, C. Huang and J. Fan, *Applied Surface Science*, 2021, **543**, 148772.