Supplementary Information

Theoretical Insights into the Intercalation Mechanisms of Li, Na and Mg ions in Metallic BN/VS_2 Heterostructure

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Table S1 Formation energy values of different materials

Type	layer	E_b (meV)
BN	Monolayer	-280.658
VS_2	Monolayer	-182.467
BN/VS_2	Heterostructure	-468.935

Table S2 Energy band values of different materials

Type	Special side (A)	Site	Energy (eV)
Monolayer BN	0.000000	Conduction band base	4.70373
	0.000000	Valence band top	0.03459
Monolayer VS_2	0.129002	Conduction band base	2.04720
	0.129002	Valence band top	1.29370
BN/VS_2	0.093666	Conduction band base	0.08304
	0.093666	Valence band top	0.01288

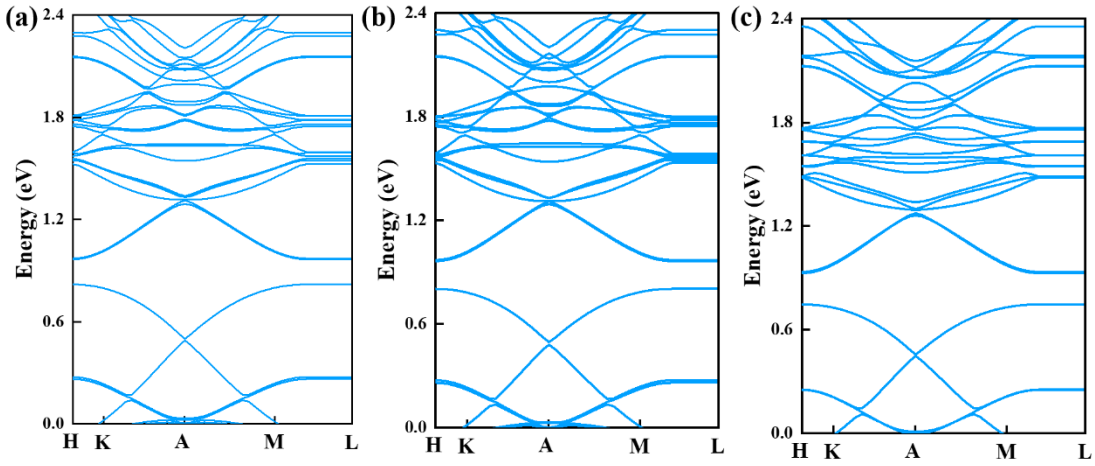

Fig. S1. The energy band of the intermediate sites in BN/VS_2 heterostructure adsorbing (a) Li, (b) Na, (c) Mg.

According to the previous calculation, the band gap of the BN/VS_2 heterostructure is 0.07 eV (Fig. 1e and Table S2), indicating the metallic property. Meanwhile, the energy bands of BN/VS_2 heterostructure adsorbed by Li, Na and Mg atoms in the interlayer were calculated (Fig. S1). The results indicated that their band gaps were all 0 eV. The roles of the adsorbent metal atoms were discussed in detail in the charge density (Fig. 3, 4, and Fig. S3). The outer layer of the VS_2 and the adsorbed metal atoms have some free electrons, and when the metal atom is adsorbed to the interlayer, the charge mainly was transferred to the VS_2 side, but a small part of the charge was transferred to the BN side. In this case, the charge of metal atoms not only have a high coincidence degree with V and S atoms in total energy region, but also partly overlaps with those of B and N atoms in low energy region. Due to the above reasons, the band gap of the intercalated heterostructure state is zero with the metallic properties.

Table S3 Adsorption energies of single Li atom on different sites of BN/VS_2 heterostructure

Adsorption site	Adsorption energy (eV)
a	−0.535
b	−0.862
c	−0.660
d	−2.352
e	−2.375
f	−2.376
g	−1.401
h	−2.112
i	−2.113

Table S4 Adsorption energies of single Na atom on different sites of BN/VS_2 heterostructure

Adsorption site	Adsorption energy (eV)
a	−1.755
b	−1.795
c	−1.844
d	−3.046
e	−3.020
f	−3.091
g	−2.754
h	−3.218
i	−3.234

Table S5 Adsorption energies of single Mg atom on different sites of BN/VS_2 heterostructure

Adsorption site	Adsorption energy (eV)
a	1.133
b	1.141
c	1.137
d	−1.999
e	−2.003
f	−2.006
g	0.196
h	−1.270
i	−1.311

Table S6 Adsorption energies of Li, Na and Mg atom on interlayer sites of different 2D anodes

Adsorbed atom	2D anodes	Interlayer (eV)	Refs.
Li	BN/VS_2	−2.38	This work
	VS_2/Graphene	−1.99	[1]
	VS_2/Blue phosphorene	−1.80	[2]
	MoS_2/Graphene	−1.74	[3]
	VS_2/Ti_2CO_2	−2.63	[4]
Na	BN/VS_2	−2.23	This work
	Phosphorene	−1.59	[5]
	MoS_2/Graphene	−1.20	[3]
	VS_2/Ti_2CO_2	−2.20	[4]
Mg	BN/VS_2	−2.0	This work
	MoS_2/Graphene	−1.89	[3]
	VS_2/Ti_2CO_2	−1.80	[4]
	monolayer VS_2	−0.58	[3]
	VO_2/VS_2	−4.38	[6]

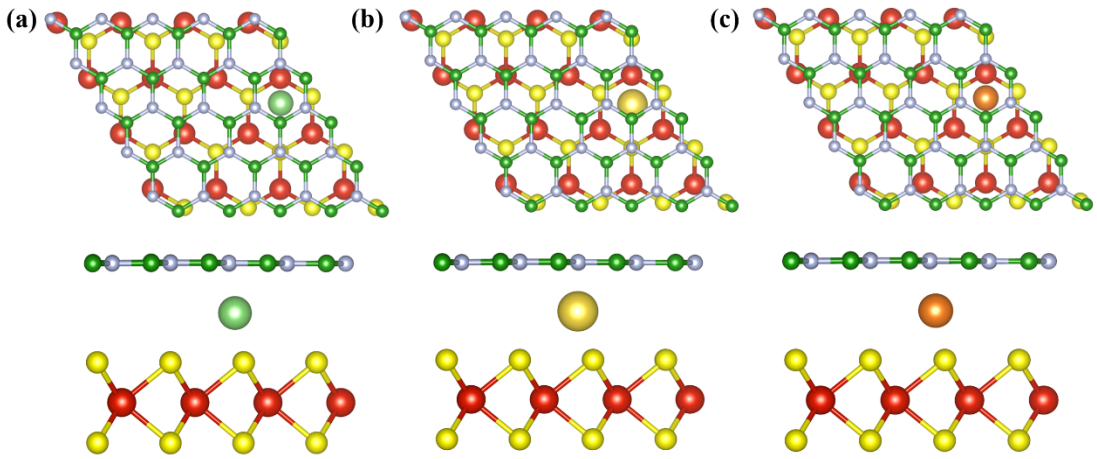

Fig. S2 (a-c) Top and side view of possible adsorption sites of Li, Na and Mg atoms on BN/VS_2 heterostructure.

Table S7 Adsorption energies of Li, Na and Mg atom on interlayer sites of different anode models

Adsorbed atom	4*4 BN & 3*3 VS_2	5*5 BN & 4*4 VS_2	Difference value
Li	−2.376 eV	−2.473 eV	0.097 eV
Na	−3.091 eV	−3.163 eV	0.072 eV
Mg	−2.006 eV	−2.147 eV	0.141 eV

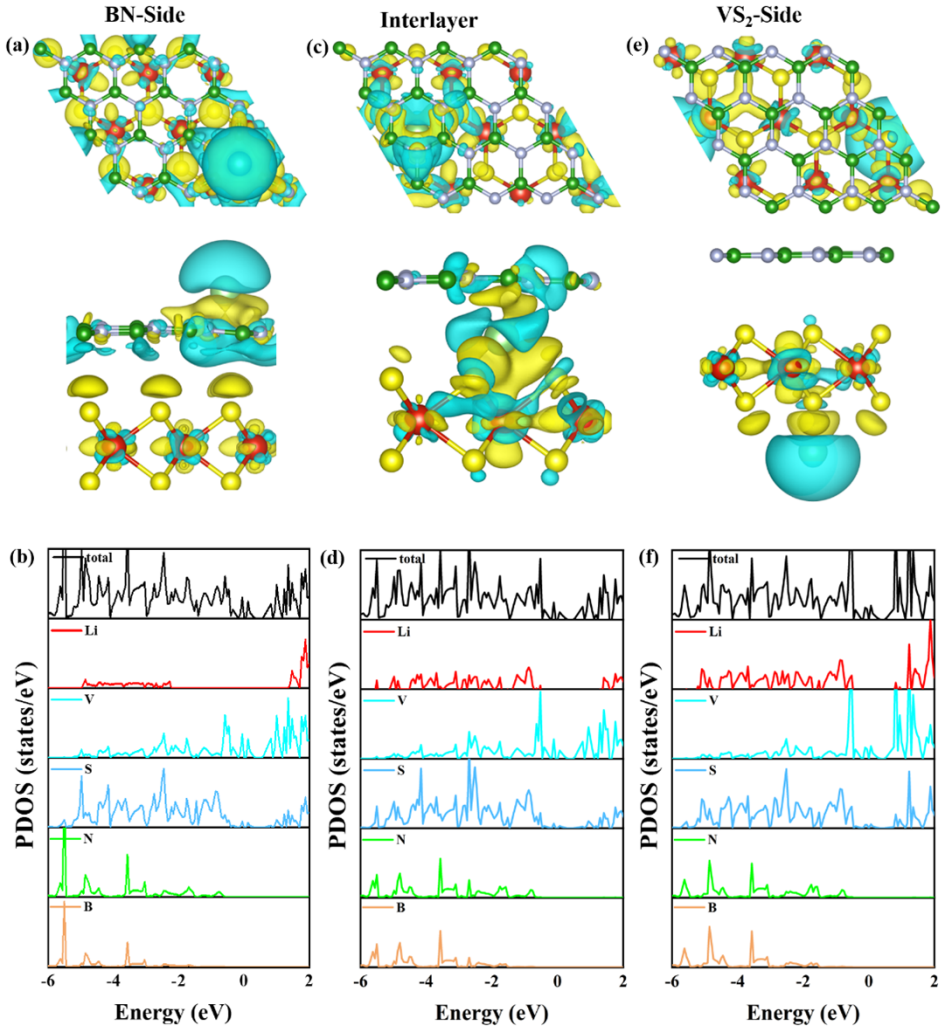

Fig. S3. Charge density isosurface configurations of Li adsorbed (a) Li/BN/VS_2, (c) BN/Li/VS_2, (e) BN/VS_2/Li. The isosurface level is set to 0.001 e $Å^{-3}$. Red, yellow, small green, grey, and big green balls represent V, S, B, N, and Li, respectively. Cyan and yellow regions indicate electron depletion and accumulation, respectively. The PDOS of (b) Li/BN/VS_2, (d) BN/Li/VS_2, (f) BN/VS_2/Li.

Table S8 Electron charge transfer from Li, Na and Mg to the BN/VS_2 heterostructures (unit: e− per atom).

Adsorbed atom	Amount of charge transferred
Li	0.8546
Na	0.7884
Mg	1.6033

Table S9 Average adsorption energy of monolayer and multilayer Li atoms on BN/VS_2 heterostructures.

Layer	**E_{ad} (eV)**
First-layer	−1.97
Second-layer	−1.62
Third-layer	−0.24
Fourth-layer	−0.17
Fifth-layer	0.04

Table S10 Average adsorption energy of monolayer and multilayer Na atoms on BN/VS_2 heterostructures.

layer	**E_{ad} (eV)**
First-layer	−2.630
Second-layer	−1.2905
Third-layer	−0.3097
Fourth-layer	−0.1420

Table 11 Average adsorption energy of monolayer and multilayer Mg atoms on BN/VS_2 heterostructures.

layer	**E_{ad} (eV)**
First-layer	−0.686
Second-layer	−0.228
Third-layer	0.129

Table S12 Diffusion energy barrier of atoms along a migration inside the VS_2 in the different 2D materials

Adsorbed atom	2D anodes	Interlayer (eV)	Refs.
Li	BN/VS_2	0.13	This work
	Monolayer VS_2	0.22	[7]
	BN/VS_2	0.54	[8]
Na	BN/VS_2	0.43	This work
	Monolayer VS_2	0.68	[9]
	VS_2/Ti_2CO_2	0.59	[10]
Mg	BN/VS_2	0.56	This work
	VS_2/Ti_2CS_2	0.86	[10]
	VS_2/Ti_2CO_2	1.01	[10]
	MoS_2	1.12	[11]

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