Supplementary information

Adsorption and Diffusion of Alkali Metals (Li, Na, and K) on Heteroatom-Doped Monolayer Titanium Disulfide

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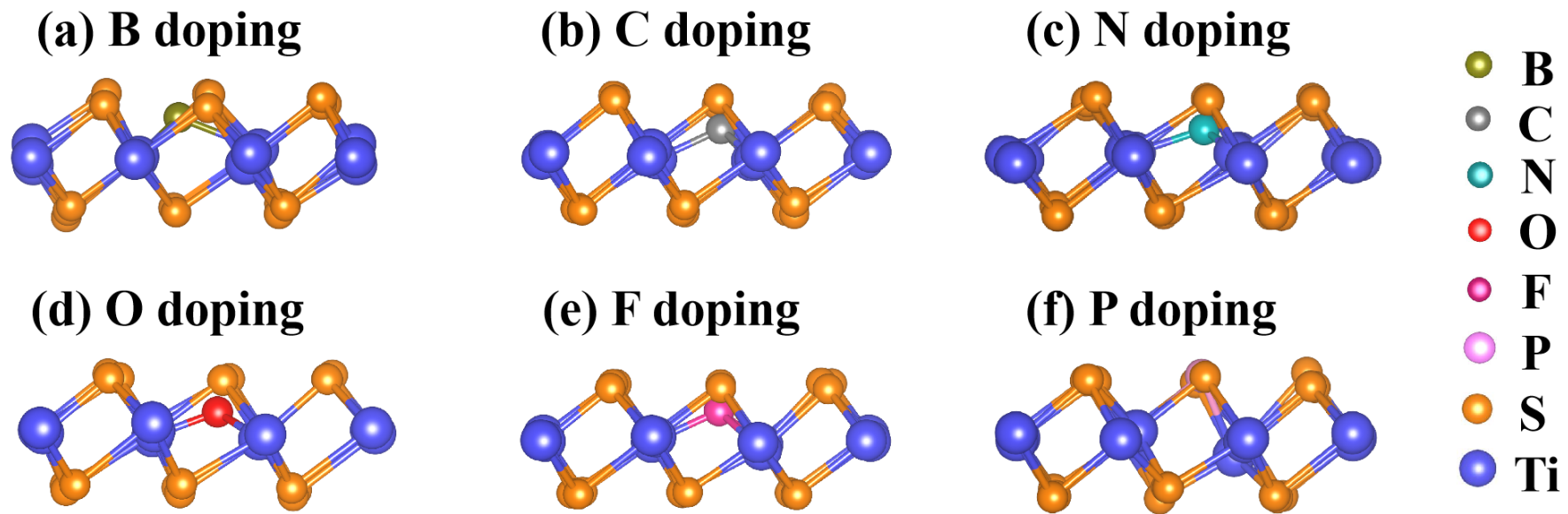
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Fig. S1 The total density of state (TDOS) of monolayer TiS_2 doped with heteroatoms (a) B, (b) C, (c) N, (d) O, (e) F, and (f) P. The Fermi energies in each case are set to 0 eV.
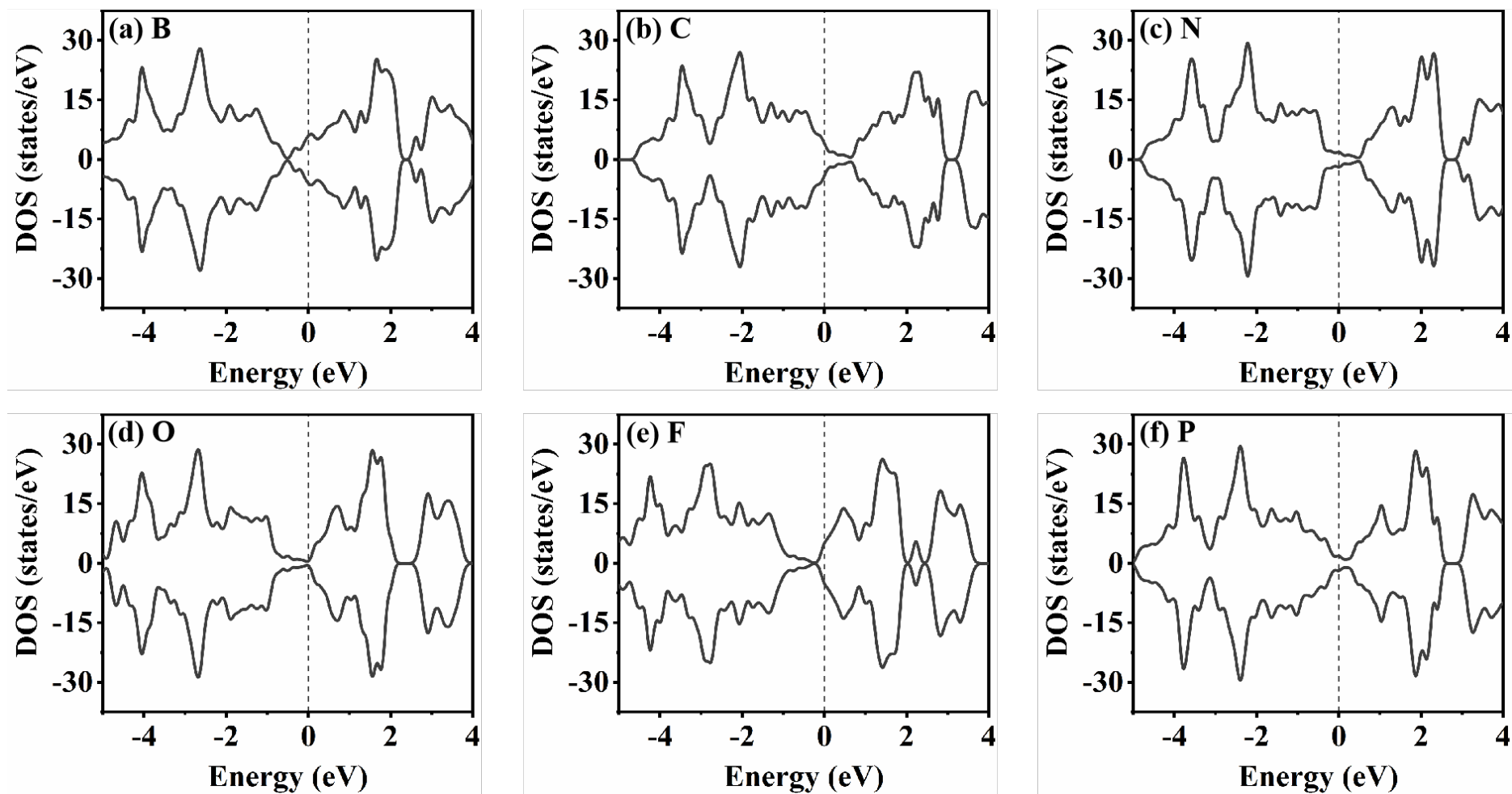
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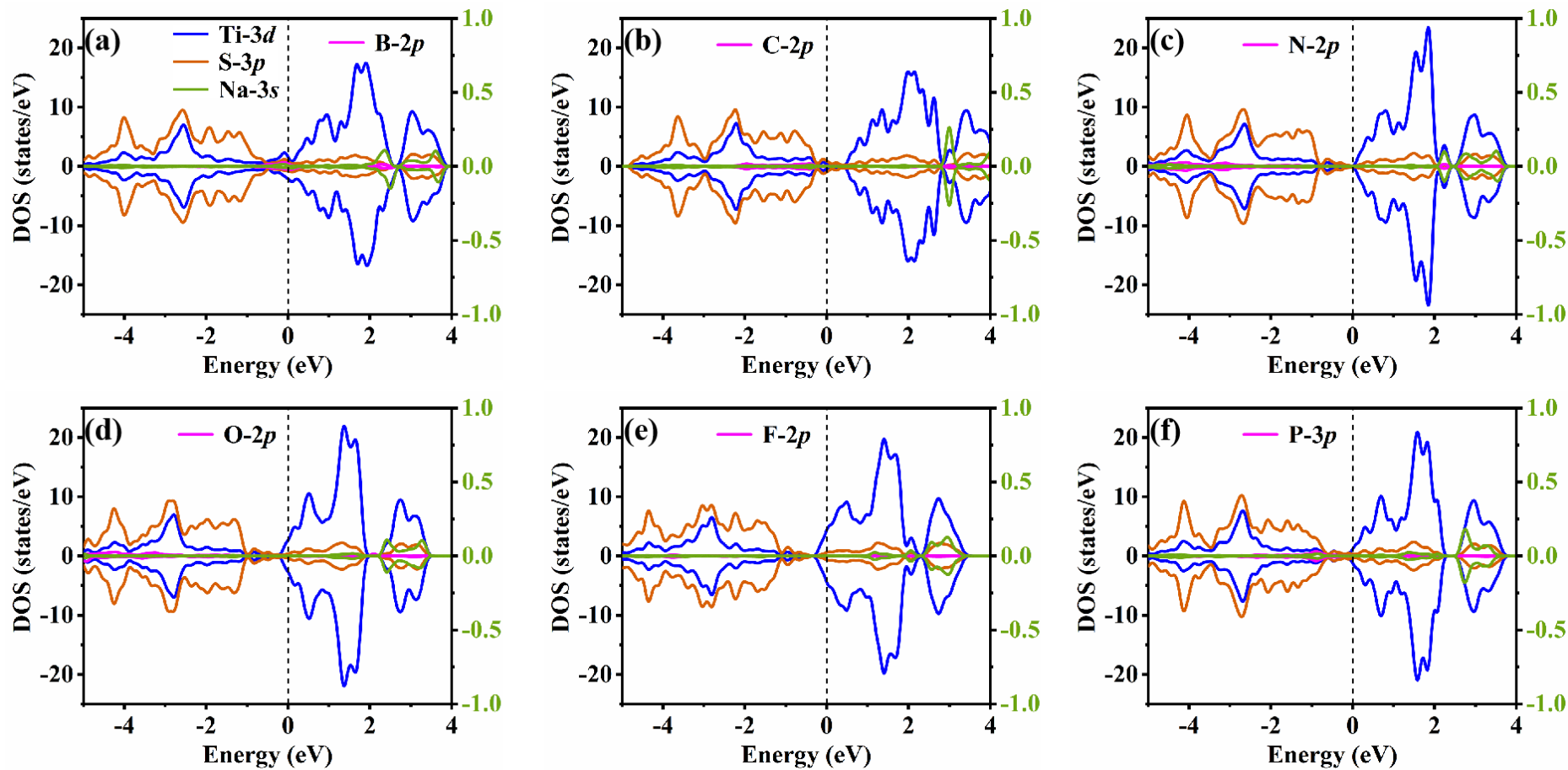
Fig. S3 The projected density of state (PDOS) of Na adsorbed on monolayer TiS_2 doped with heteroatoms (a) B, (b) C, (c) N, (d) O, (e) F, and (f) P. The Fermi energies in each case are set to 0 eV.

Fig. S4 The PDOS of K adsorbed on monolayer TiS$_2$ doped with heteroatoms (a) B, (b) C, (c) N, (d) O, (e) F, and (f) P. The Fermi energies in each case are set to 0 eV.

Fig. S5 The TDOS of (a) Li, (b) Na, and (c) K adsorbed on pristine monolayer TiS_2. The Fermi energies in each case are set to 0 eV.

Fig. S6 The TDOS of Li adsorbed on monolayer TiS$_2$ doped with heteroatoms (a) B, (b) C, (c) N, (d) O, (e) F, and (f) P. The Fermi energies in each case are set to 0 eV.

Fig. S7 The TDOS of Na adsorbed on monolayer TiS_2 doped with heteroatoms (a) B, (b) C, (c) N, (d) O, (e) F, and (f) P. The Fermi energies in each case are set to 0 eV.
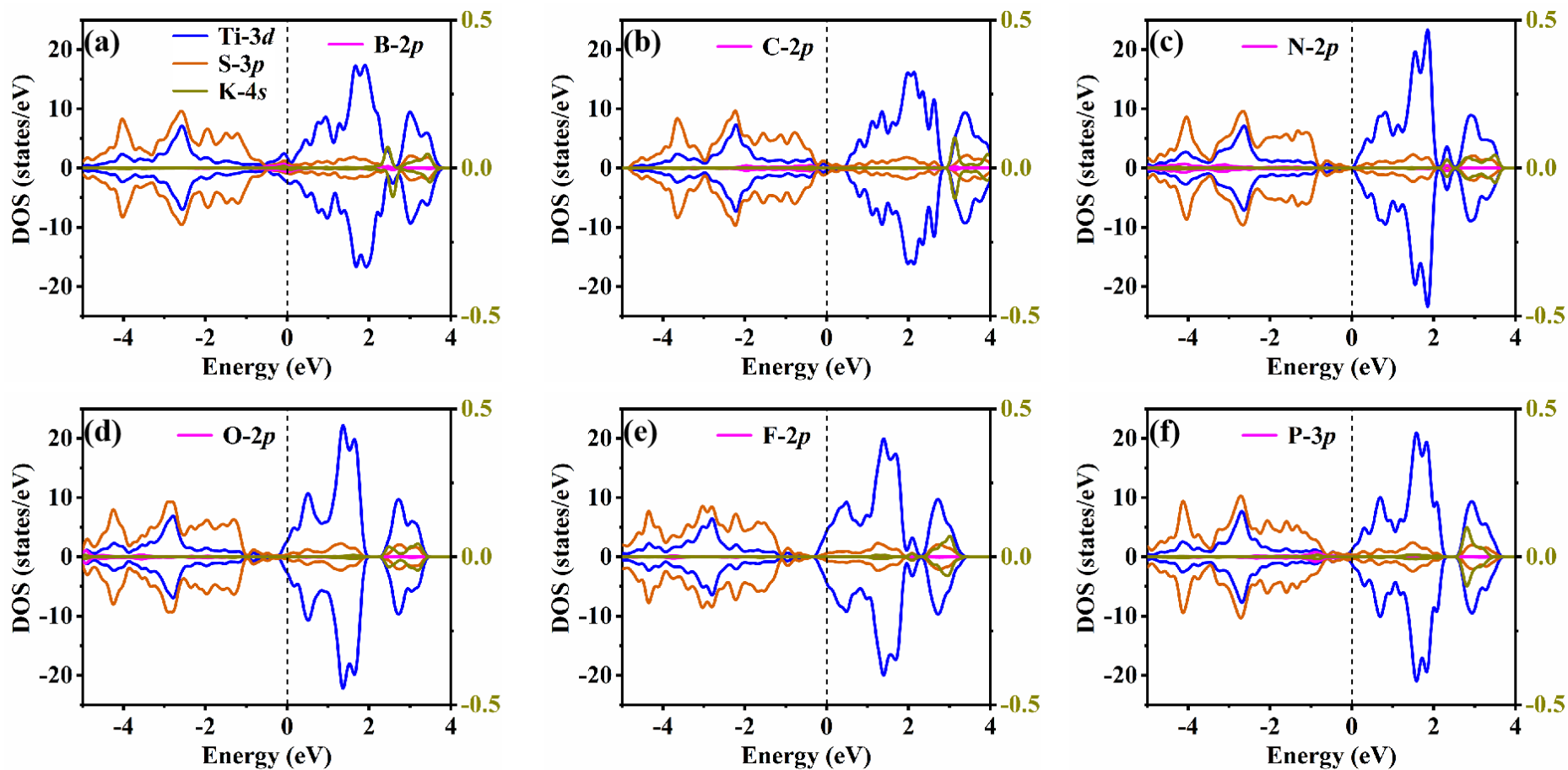
Fig. S8 The TDOS of K adsorbed on monolayer TiS_2 doped with heteroatoms (a) B, (b) C, (c) N, (d) O, (e) F, and (f) P. The Fermi energies in each case are set to 0 eV.
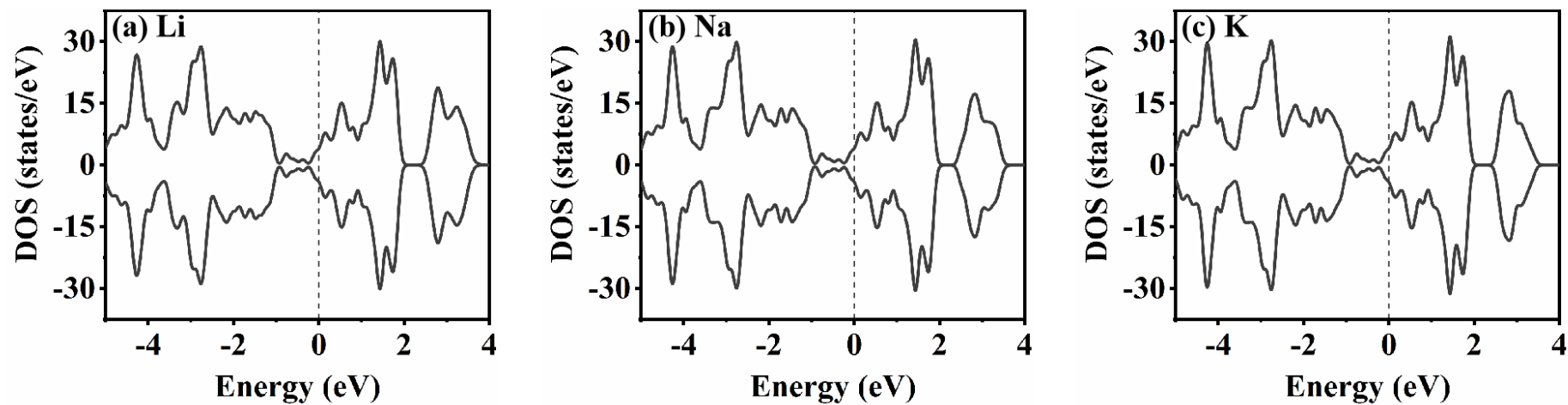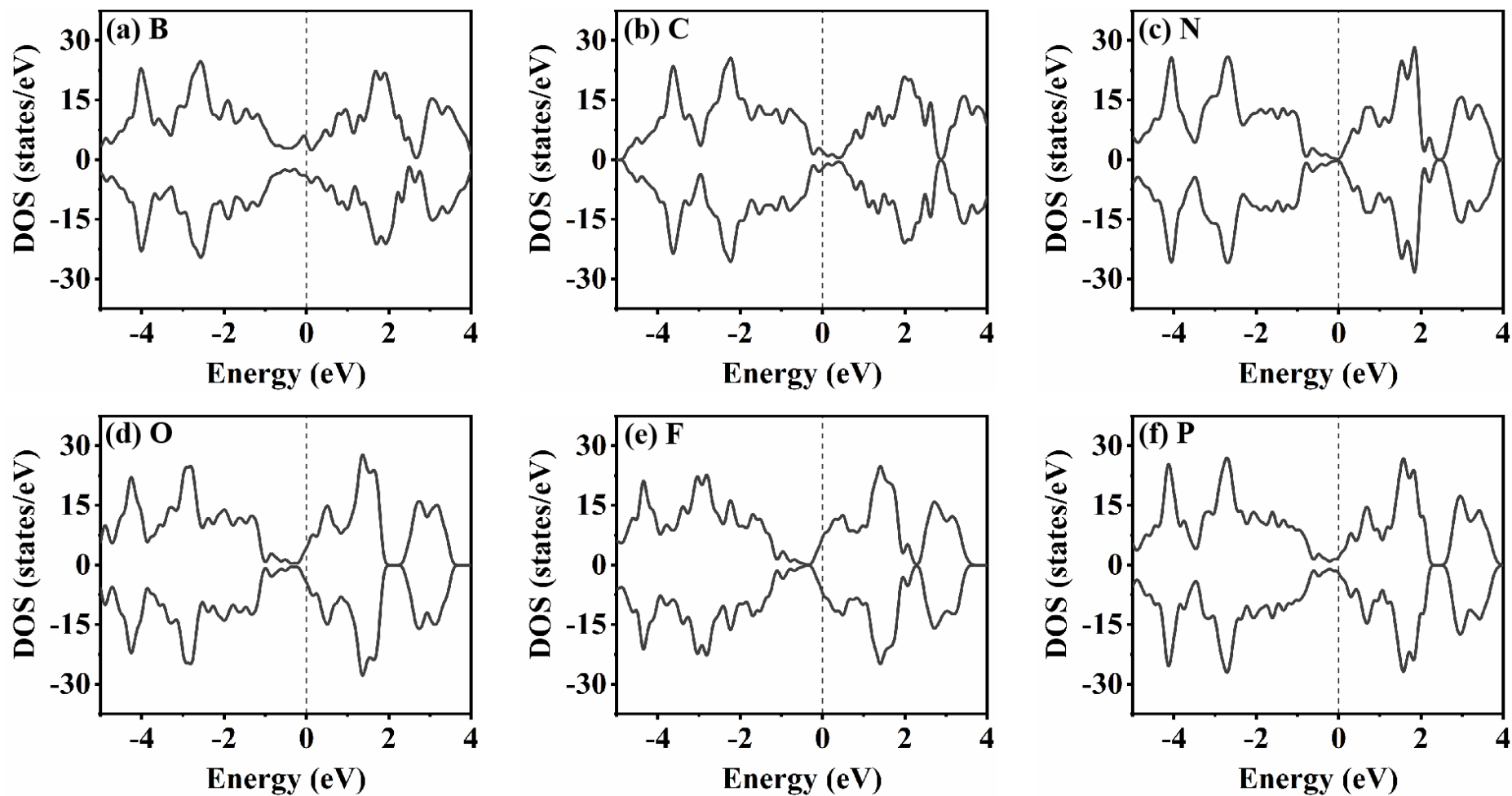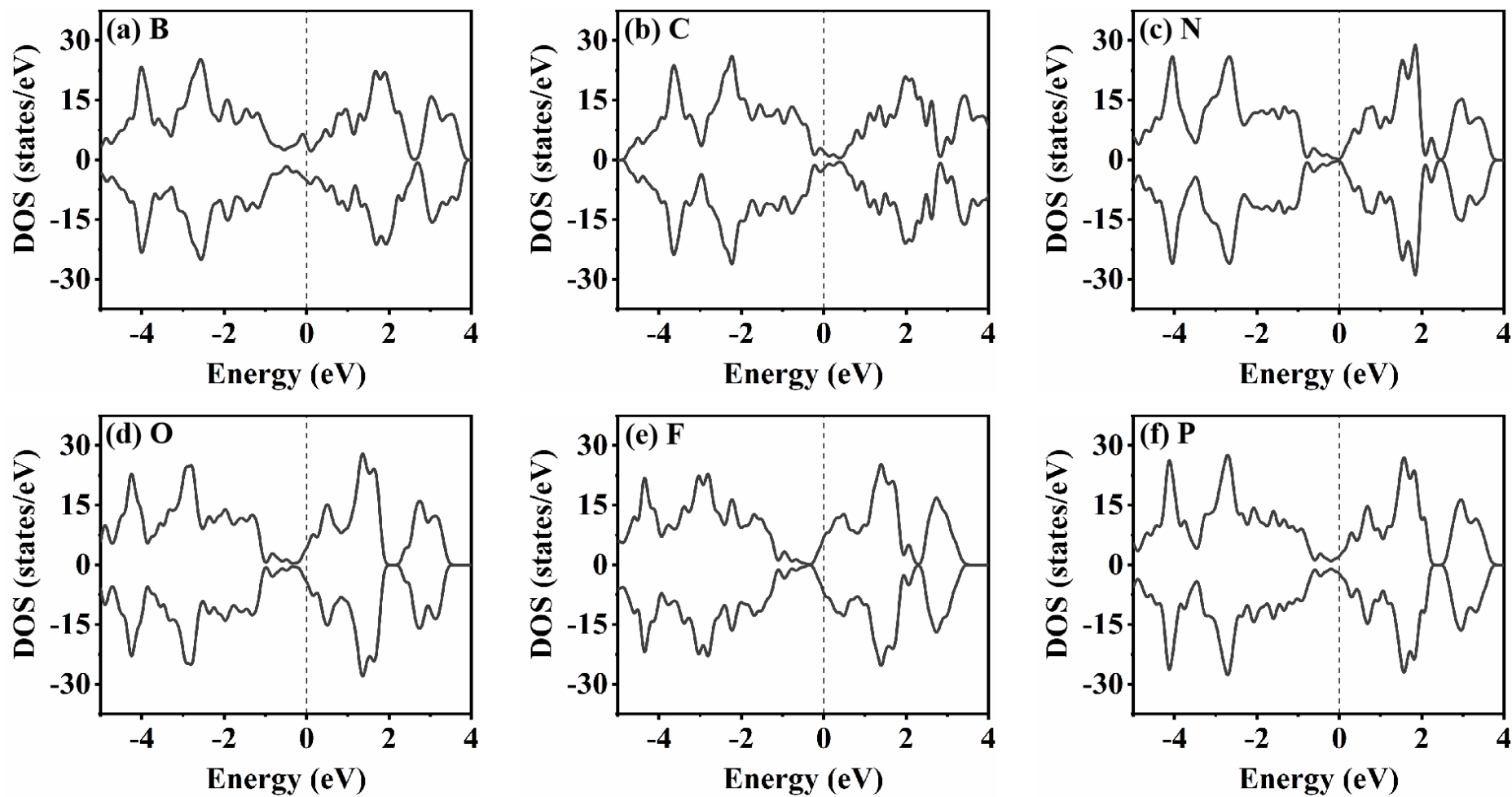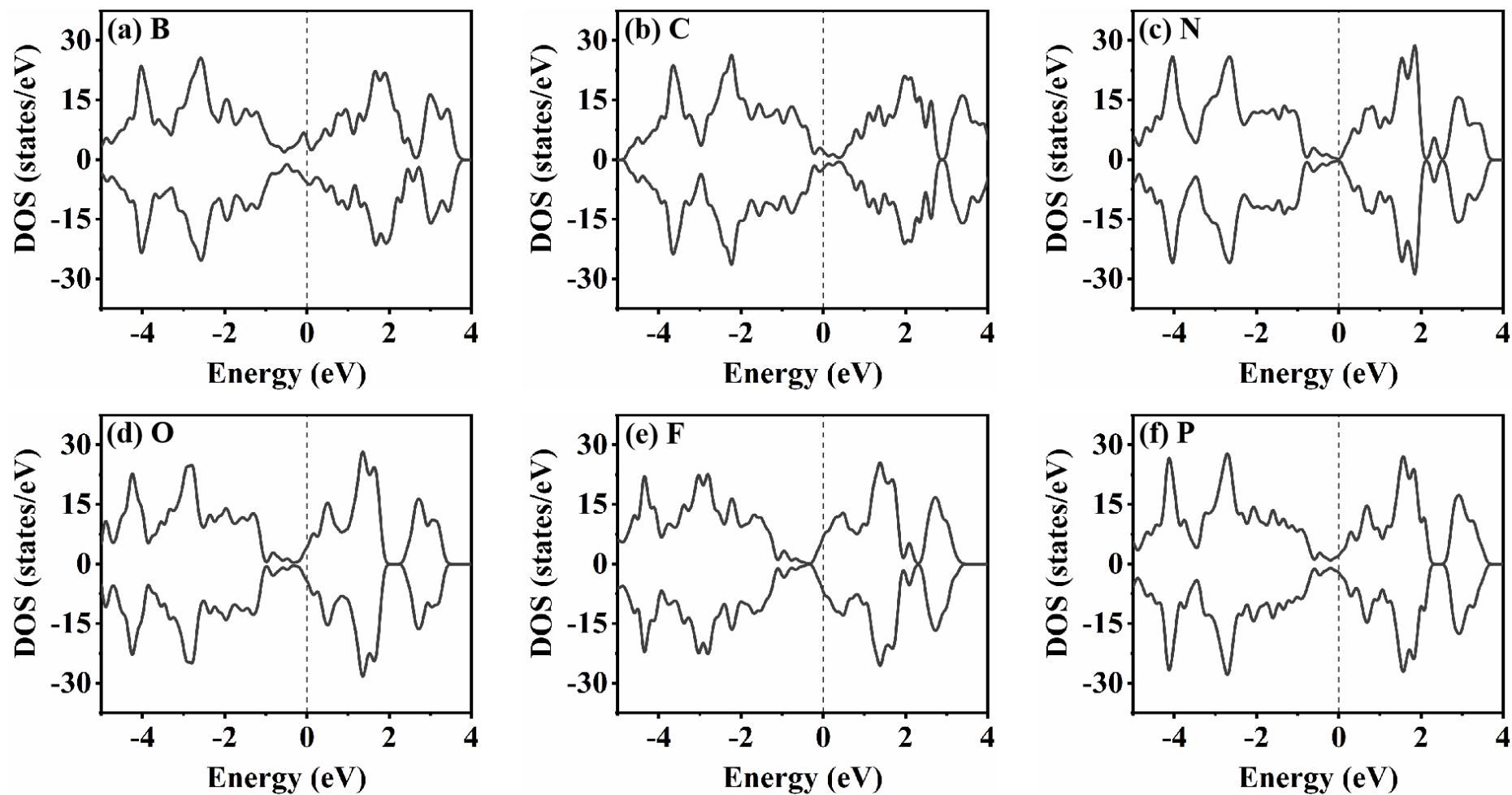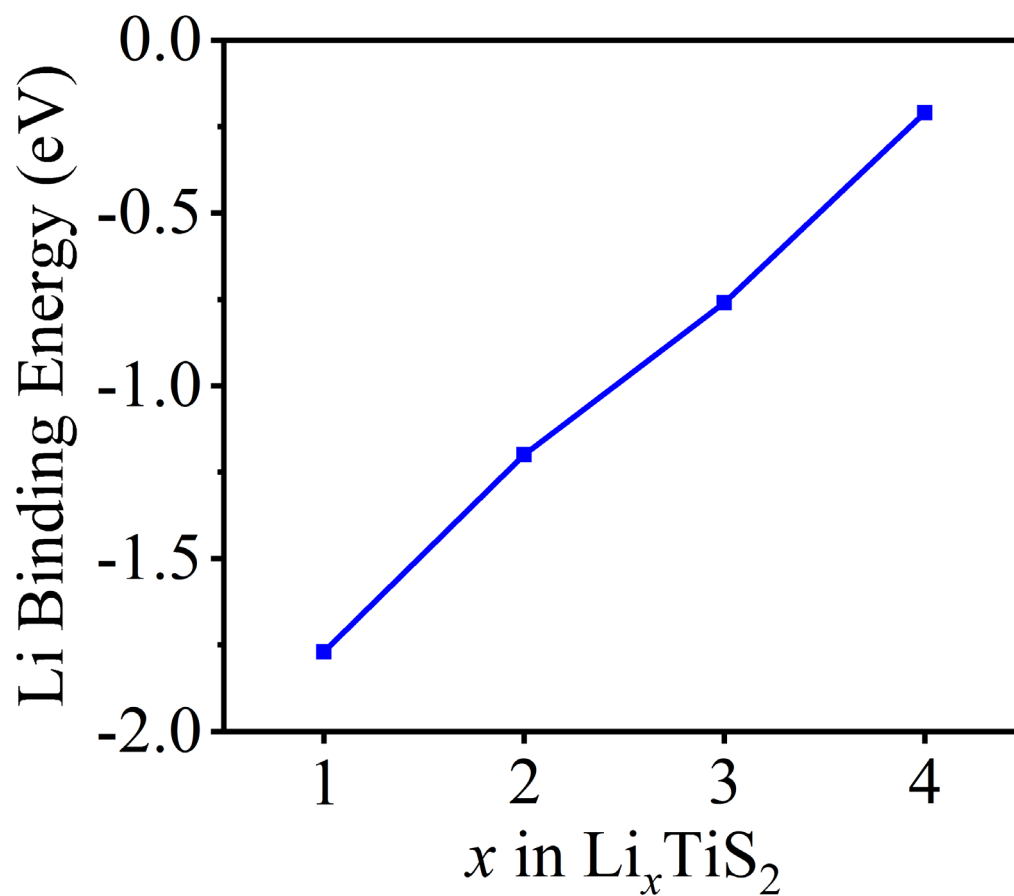
Fig. S9 The binding energy as a function of Li adsorption concentration.

Table S1 Calculated adsorption energies of Li, Na, and K adsorbed on different sites of the pristine and heteroatom-doped monolayer TiS_2.

Metals	Dopants	Sites							
		H_1	H_2	H_3	H_4	T_1	T_2	T_3	T_4
Li	None	–3.58	–3.58	–3.58	–3.58	–3.57	–3.57	–3.57	–3.57
	B	–3.77	–3.61	–3.64	–3.64	–3.59	–3.61	–3.61	–3.61
	C	–4.48	–4.11	–4.15	–4.15	–4.31	–4.13	–4.13	–4.13
	N	–4.15	–3.91	–3.94	–3.94	–4.02	–3.92	–3.93	–3.92
	O	–3.64	–3.50	–3.53	–3.53	–3.58	–3.49	–3.51	–3.49
	F	–3.49	–3.41	–3.44	–3.44	–3.44	–3.40	–3.41	–3.40
	P	–3.81	–3.75	–3.75	–3.75	–3.78	–3.75	–3.74	–3.75
Na	None	–2.96	–2.96	–2.96	–2.96	–2.95	–2.95	–2.95	–2.95
	B	–3.06	–2.99	–3.02	–3.02	–3.01	–2.99	–3.00	–2.99
	C	–3.83	–3.52	–3.55	–3.55	–3.78	–3.53	–3.53	–3.53
	N	–3.54	–3.30	–3.33	–3.33	–3.50	–3.31	–3.32	–3.31
	O	–3.05	–2.89	–2.91	–2.91	–3.03	–2.88	–2.90	–2.88
	F	–2.94	–2.79	–2.82	–2.82	–2.92	–2.78	–2.80	–2.78
	P	–3.19	–3.15	–3.15	–3.15	–3.17	–3.14	–3.14	–3.14
K	None	–3.22	–3.22	–3.22	–3.22	–3.21	–3.21	–3.21	–3.21
	B	–3.32	–3.24	–3.28	–3.27	–3.28	–3.23	–3.25	–3.24
	C	–4.10	–3.78	–3.80	–3.80	–4.08	–3.78	–3.78	–3.78
	N	–3.83	–3.56	–3.59	–3.58	–3.82	–3.56	–3.56	–3.57

O	–3.38	–3.14	–3.16	–3.16	–3.37	–3.13	–3.14	–3.13
F	–3.26	–3.04	–3.07	–3.08	–3.25	–3.03	–3.05	–3.03
P	–3.43	–3.41	–3.40	–3.40	–3.41	–3.40	–3.39	–3.40

Table S2 Calculated adsorption energies of Li, Na, and K adsorbed on different sites of the pristine and heteroatom-doped monolayer TiS$_2$. Note that the adsorption energy values are obtained by equation (2) expressing E_M with the energy of per atom in the bulk of crystal M (M = Li, Na, and K).

Metals	Dopants	Sites							
		H_1	H_2	H_3	H_4	T_1	T_2	T_3	T_4
Li	None	−1.88	−1.88	−1.88	−1.88	−1.87	−1.87	−1.87	−1.87
	B	−2.07	−1.91	−1.94	−1.94	−1.89	−1.91	−1.91	−1.91
	C	−2.78	−2.41	−2.45	−2.45	−2.61	−2.43	−2.43	−2.43
	N	−2.45	−2.21	−2.24	−2.24	−2.32	−2.22	−2.23	−2.22
	O	−1.94	−1.80	−1.83	−1.83	−1.88	−1.79	−1.81	−1.79
	F	−1.79	−1.71	−1.74	−1.74	−1.74	−1.70	−1.71	−1.70
	P	−2.11	−2.05	−2.05	−2.05	−2.08	−2.05	−2.04	−2.05
Na	None	−1.74	−1.74	−1.74	−1.74	−1.73	−1.73	−1.73	−1.73
	B	−1.84	−1.77	−1.80	−1.80	−1.79	−1.77	−1.78	−1.77
	C	−2.61	−2.30	−2.33	−2.33	−2.56	−2.31	−2.31	−2.31
	N	−2.32	−2.08	−2.11	−2.11	−2.28	−2.09	−2.10	−2.09
	O	−1.83	−1.67	−1.69	−1.69	−1.81	−1.66	−1.68	−1.66
	F	−1.72	−1.57	−1.60	−1.60	−1.70	−1.56	−1.58	−1.56
	P	−1.97	−1.93	−1.93	−1.93	−1.95	−1.92	−1.92	−1.92
K	None	−2.29	−2.29	−2.29	−2.29	−2.28	−2.28	−2.28	−2.28
	B	−2.39	−2.31	−2.35	−2.34	−2.35	−2.30	−2.32	−2.31

C	–3.17	–2.85	–2.87	–2.87	–3.15	–2.85	–2.85	–2.85
N	–2.90	–2.63	–2.66	–2.65	–2.89	–2.63	–2.63	–2.64
O	–2.45	–2.21	–2.23	–2.23	–2.44	–2.20	–2.21	–2.20
F	–2.33	–2.11	–2.14	–2.15	–2.32	–2.10	–2.12	–2.10
P	–2.50	–2.48	–2.47	–2.47	–2.48	–2.47	–2.46	–2.47

Table S3 The calculated formation energies (E_f) for the heteroatom-doped monolayer TiS_2 with and without the dipole correction, and their differences (ΔE) with and without the dipole correction.

Dopants	Dipole correction		No dipole correction		Differences	
	E_f (S-rich) (eV)	E_f (Ti-rich) (eV)	E_f (S-rich) (eV)	E_f (Ti-rich) (eV)	ΔE (S-rich) (eV)	ΔE (Ti-rich) (eV)
B	4.83	2.89	4.82	2.87	0.01	0.02
C	4.59	2.65	4.58	2.63	0.01	0.02
N	1.04	−0.90	1.03	−0.92	0.00	0.02
O	−2.34	−4.29	−2.36	−4.30	0.02	0.01
F	−2.72	−4.66	−2.73	−4.67	0.01	0.01
P	2.07	0.12	2.05	0.11	0.02	0.01

Table S4 The adsorption energies (E_d) of the most favorable configurations for Li, Na, and K adsorbed on the pristine and heteroatom-doped monolayer TiS_2 with and without the dipole correction, and their differences (ΔE) with and without the dipole correction.

Dopants	E_d (Dipole correction) (eV)			E_d (No dipole correction) (eV)			ΔE (eV)		
	Li	Na	K	Li	Na	K	Li	Na	K
None	−3.56	−2.90	−3.12	−3.58	−2.96	−3.22	0.02	0.06	0.10
B	−3.76	−3.03	−3.24	−3.77	−3.06	−3.32	0.01	0.03	0.08
C	−4.47	−3.80	−4.03	−4.48	−3.83	−4.10	0.01	0.03	0.07
N	−4.14	−3.50	−3.77	−4.15	−3.54	−3.83	0.01	0.04	0.06
O	−3.62	−3.02	−3.32	−3.64	−3.05	−3.38	0.02	0.03	0.06
F	−3.48	−2.89	−3.20	−3.49	−2.94	−3.26	0.01	0.05	0.06
P	−3.80	−3.14	−3.34	−3.81	−3.19	−3.43	0.01	0.05	0.09

Table S5 Comparison of Li/Na/K diffusion energy barriers of various 2D anode materials.

2D materials	Diffusion energy barriers (eV)			Reference
	Li	Na	K	
Blue phosphorene	0.16	0.11	0.09	[1,2]
Boron-Graphdiyne	0.36	0.28	0.12	[3]
Boron phosphide	0.36	0.22	0.16	[4]
Borophene	0.011	0.003	0.008	[5]
GeS	0.24	0.09	0.05	[6]
MoN_2	0.78	0.56	0.49	[7]
MoS_2	0.25	0.28		[8,9]
ReN_2		0.03	0.13	[10]
ReS_2	0.33	0.16		[11]
Si_2BN	0.48	0.32		[12]
Si_3C	0.47	0.34	0.18	[13]
Silicene	0.24	0.14		[14,15]
Ti_3C_2	0.07	0.10	0.10	[16]
Ti_2CP_2	0.32	0.29	0.19	[17]
Ti_2CSi_2	0.29	0.22	0.08	[17]
VS_2	0.22	0.09	0.06	[18,19]

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