Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

TiSe Monolayer as a superior anode for the applications of Li/Na/K-ion batteries

Mengke Wang[1], Shan Wang[2], Yunye Liang[1], Yiqun Xie[1], Xiang Ye[*1], Shoutian, Sun[*1]

[1]Department of Physics, Shanghai Normal University, Shanghai 200234, P. R. China

[2]School of Physics and Electronic Science, East China Normal University, Shanghai 200062, P. R. China

Email address: yexiang@shnu.edu.cn (Xiang Ye), stsun@shnu.edu.cn (Shoutian Sun)

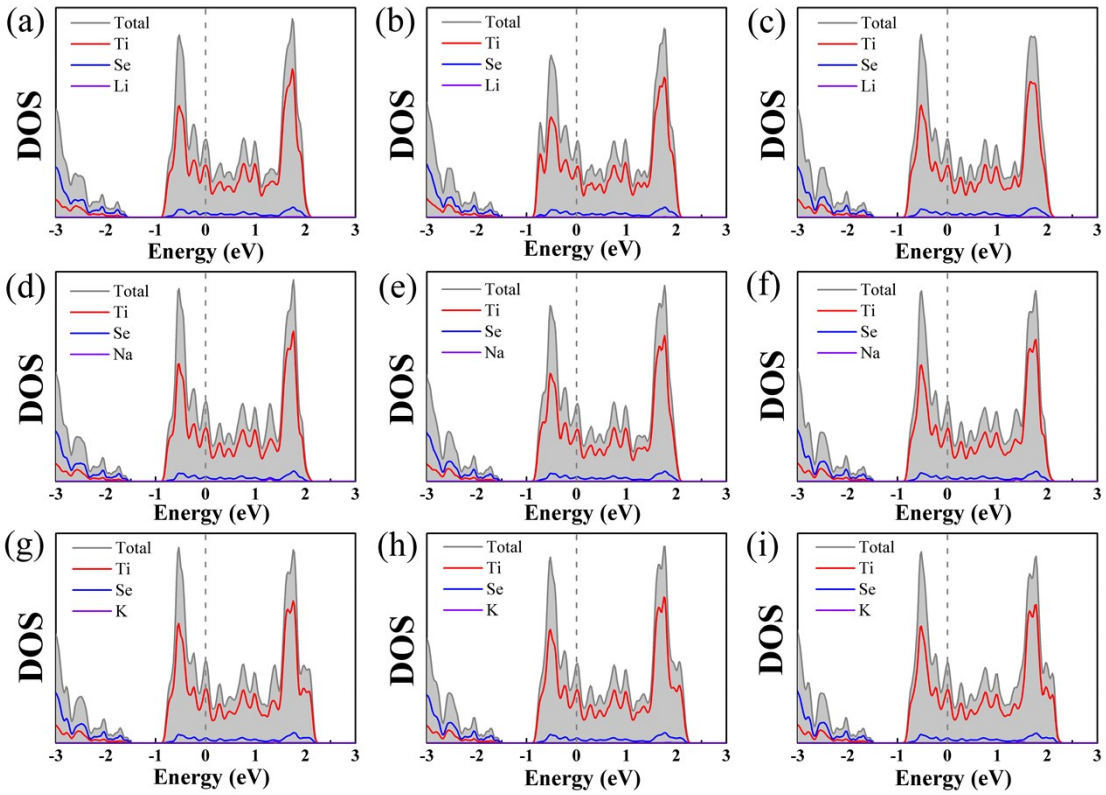

Fig. S1. Total and projected DOS of a single Li (a-c), Na (d-f), K (g-i) atom adsorbed onto TiSe for the BG, H, and T_{Ti} site, with fermi energy level set to 0 eV.

Table S1. The distances of alkali metal to the substrate at different adsorption sites.

Site	d (Å)		
	Li	Na	K
T_{Ti}	1.396	1.865	2.333
BG	1.281	1.943	2.397
H	0.671	1.227	1.823

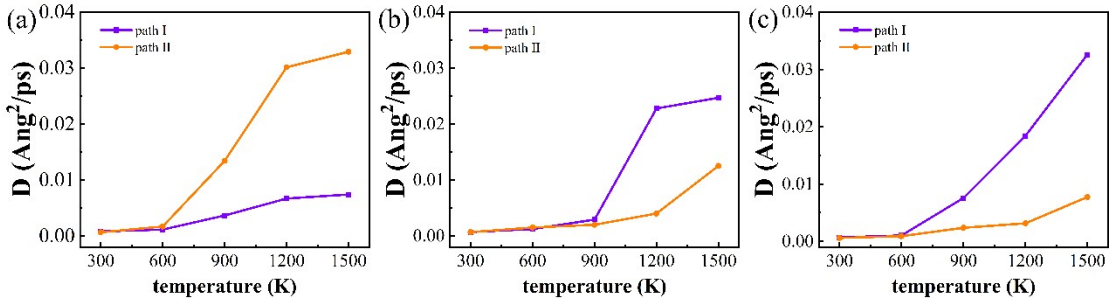

Fig. S2. Plots of the diffusion coefficient versus temperature for Li (a), Na (b), K (c) along paths I and II.

To investigate the diffusion of Li/Na/K in the substrate TiSe, AIMD simulation is carried out at different temperatures (300, 600, 900,1200, 1500 K). We have used a minimum Γ-centered 1 x 1 x 1 *k*-point grid. The time step is chosen to be 1 fs and the diffusion simulation is carried out for 10 ps. The diffusion coefficient is given by:

$$D = \lim_{t\to\infty}\left[\frac{1}{2dt}\left\langle\left[\mathbf{r}(t)\right]^2\right\rangle\right]$$

where t and d denote time and the dimension of the lattice. The average mean square displacement is calculated by:

$$\left\langle \left[\vec{r}(t) \right]^2 \right\rangle = \frac{1}{N} \sum_{i=1}^{N} \left\langle \left[\vec{r}_i(t+t_0) \right]^2 - \left[\vec{r}_i(t_0) \right]^2 \right\rangle$$

where N is the number of particles, and the square brackets indicate the average of the ensemble. $\vec{r}_i(t)$ is the coordinate of particle i at time t.

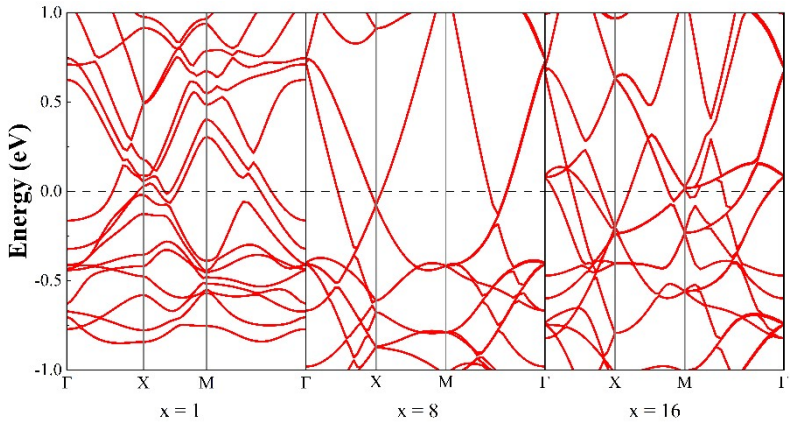

Fig. S3. The electronic band structure of the intermediate geometries $Li_x(TiSe)_4$ (x = 1, 8 and 16). The Fermi level is set to zero.

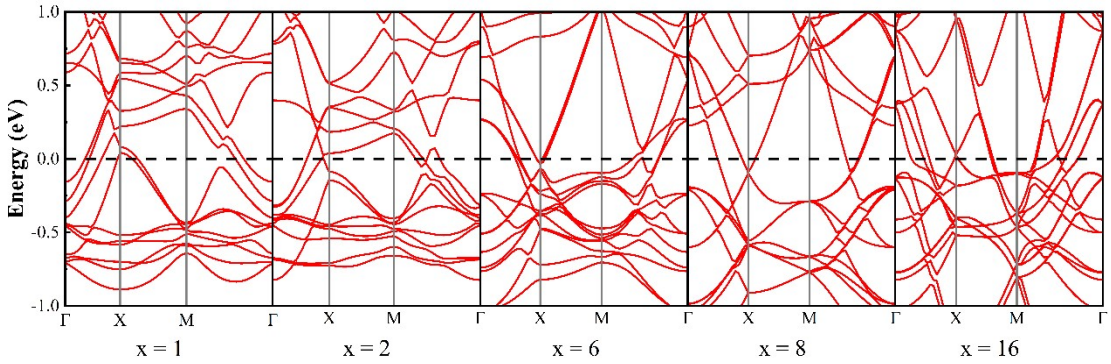

Fig. S4. The electronic band structure of the intermediate geometries $Na_x(TiSe)_4$ (x = 1, 2, 6, 8, and 16). The Fermi level is set to zero.

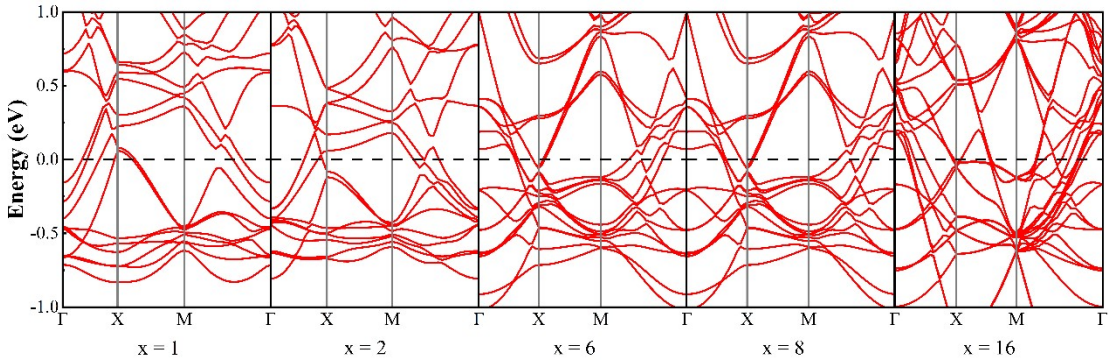

Fig. S5. The electronic band structure of the intermediate geometries $K_x(TiSe)_4$ (x = 1, 2, 6, 8, and 16). The Fermi level is set to zero.

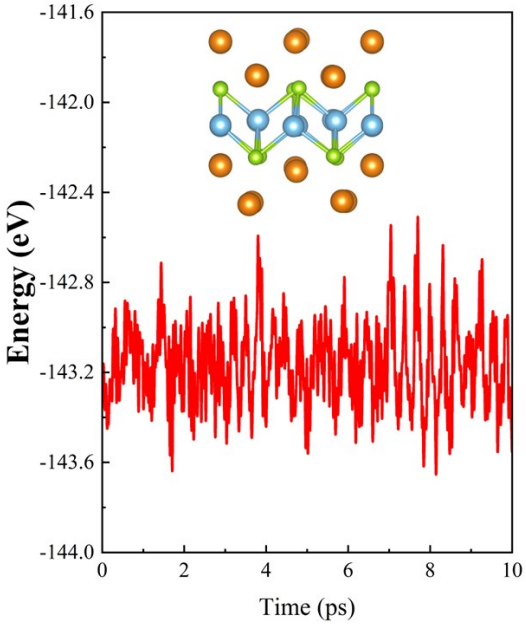

Fig. S6. AIMD simulation with 10 ps at 300 K for $Li_x(TiSe)_4$. The inset is the snapshot of last step.

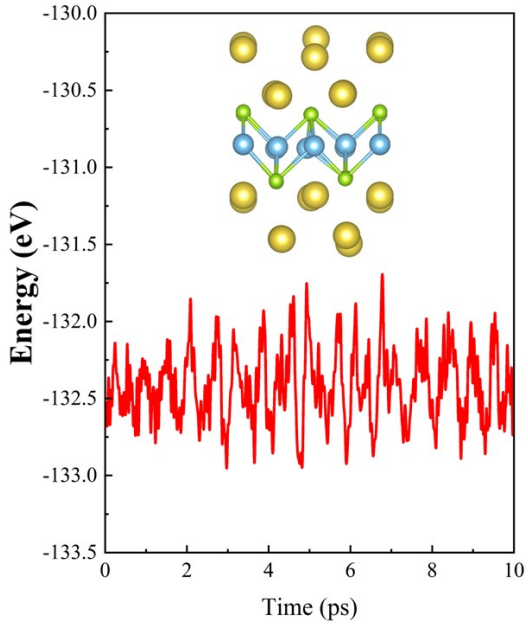

Fig. S7. AIMD simulation with 10 ps at 300 K for $Na_x(TiSe)_4$. The inset is the snapshot of last step.

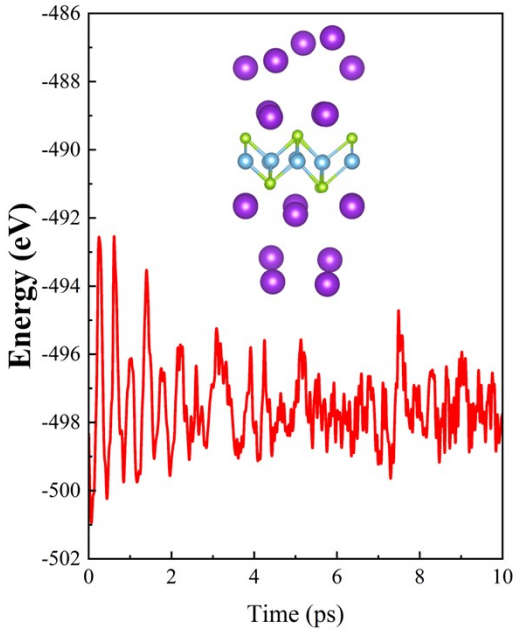

Fig. S8. AIMD simulation with 10 ps at 300 K for $K_x(TiSe)_4$. The inset is the snapshot of last step.

Table S2. The formation energy of alkaline ions added one by one to the TiSe monolayer.

N_{ions}	$E_{binding}$ (eV)		
	Li	**Na**	**K**
1	-1.54	-1.51	-1.97
2	-1.31	-1.26	-1.67
3	-1.25	-1.07	-1.38
4	-1.30	-1.05	-1.28
5	-1.31	-1.05	-1.18
6	-1.35	-1.07	-1.15
7	-1.37	-1.03	-1.10
8	-1.38	-1.01	-1.06
9	-1.11	-0.85	-0.95
10	-0.90	-0.72	-0.85
11	-0.80	-0.67	-0.79
12	-0.71	-0.63	-0.73
13	-0.68	-0.60	-0.68
14	-0.64	-0.57	-0.64
15	-0.60	-0.56	-0.61
16	-0.59	-0.56	-0.57