Supporting Information

Optimizing the Energy Storage Performance of Titanium Carbonitride MXenes for Potassium-Ion Batteries by Modulating Nitrogen Content

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Table S1 Space group and cohesive energies of the $Ti_3(C_{1-y}N_y)_2$ MXenes.

structure	Space group	Cohesive energies（eV）
$Ti_3(C_{0.875}N_{0.125})_2$	*P*3*m*1	-7.0760
	Pm	-7.0243
$Ti_3(C_{0.75}N_{0.25})_2$	*P*-3*m*1	-7.0277
	*C*2/*m*	-7.0277
$Ti_3(C_{0.625}N_{0.375})_2$	*Cm*	-6.9727
	*P*3*m*1	-6.9659
	*P*3*m*1	-6.9030
	*P*3*m*1	-6.9126
$Ti_3(C_{0.5}N_{0.5})_2$	*Cm*	-6.9126
	*C*2	-6.9159
	*P*2/*m*	-6.9159
	*P*21/*m*	-6.9158

Table S2 Independent elastic constants (C_{ij}), **Young's** modulus (Y^{2D}) in N m^{-1} and Poisson's ratio (V^{2D}) of $Ti_3(C_{1-y}N_y)_2$ MXenes.

	C_{11}	C_{22}	C_{12}	C_{66}	$Y^{2D}_{[01]}$	$Y^{2D}_{[10]}$	$V^{2D}_{[01]}$	$V^{2D}_{[10]}$
$Ti_3(C_{0.875}N_{0.125})_2$	247.513	247.513	49.425	99.044	237.644	237.644	0.200	0.200
$Ti_3(C_{0.75}N_{0.25})_2$	252.339	0.265	53.764	99.287	240.884	240.884	0.213	0.213
$Ti_3(C_{0.625}N_{0.375})_2$	239.795	239.795	61.690	89.053	223.925	223.925	0.257	0.257
$Ti_3(C_{0.5}N_{0.5})_2$	242.524	242.524	64.379	89.073	225.435	225.435	0.265	0.265

Table S3 Stable adsorption sites and adsorption energies of K on the $Ti_3(C_{1-y}N_y)_2$ MXenes surfaces.

K site	$Ti_3(C_{0.875}N_{0.125})_2$	$Ti_3(C_{0.75}N_{0.25})_2$	$Ti_3(C_{0.625}N_{0.375})$	$Ti_3(C_{0.5}N_{0.5})_2$
T_1	-1.3337	-1.3686	-1.3747	-1.4668
T_2	-1.3385	-1.3267	-1.3237	-1.4120
T_3	-1.3755	_____	-1.3706	_____
T_4	_____	_____	-1.4314	_____
T_5	_____	_____	-1.4271	_____
T_6	-1.3544	-1.3712	-1.3692	-1.4405
T_7	-1.3532	_____	-1.4104	_____
T_8	-1.3955	-1.3900	-1.3897	-1.4602
T_9	_____	_____	-1.4278	_____
H_1	-1.3544	-1.372	-1.3690	-1.4517
H_2	-1.3504	-1.3768	-1.3663	-1.4122
H_3	-1.3782	_____	-1.3767	_____
H_4	_____	_____	-1.4083	_____
H_5	_____	_____	-1.4060	_____
B_1	-1.3501	-1.3621	-1.3623	-1.4264
B_2	-1.3535	-1.3841	-1.3830	-1.4656
B_3	-1.3899	_____	_____	-1.4427

Table S4 Variation of lattice constants (*a*, *b* in Å) for the four K adsorbed $Ti_3(C_{1-y}N_y)_2$ configurations as a function of K concentration.

Material	K concentration	*a*	*b*	% change in *a*	% change in *b*	Average change in the *ab* plane
$Ti_3(C_{0.875}N_{0.125})_2$	$K_0Ti_3(C_{0.875}N_{0.125})_2$	12.335	12.335	———	———	———
	$K_1Ti_3(C_{0.875}N_{0.125})_2$	12.351	12.354	0.130	0.154	0.142
	$K_2Ti_3(C_{0.875}N_{0.125})_2$	12.358	12.363	0.186	0.227	0.207
	$K_3Ti_3(C_{0.875}N_{0.125})_2$	12.404	12.413	0.560	0.632	0.596
$Ti_3(C_{0.75}N_{0.25})_2$	$K_0Ti_3(C_{0.75}N_{0.25})_2$	12.297	12.297	———	———	———
	$K_1Ti_3(C_{0.75}N_{0.25})_2$	12.323	12.323	0.211	0.211	0.211
	$K_2Ti_3(C_{0.75}N_{0.25})_2$	12.329	12.330	0.260	0.268	0.264
	$K_3Ti_3(C_{0.75}N_{0.25})_2$	12.344	12.340	0.382	0.350	0.366
$Ti_3(C_{0.625}N_{0.375})_2$	$K_0Ti_3(C_{0.625}N_{0.375})_2$	12.265	12.265	———	———	———
	$K_1Ti_3(C_{0.625}N_{0.375})_2$	12.294	12.292	0.236	0.220	0.228
	$K_2Ti_3(C_{0.625}N_{0.375})_2$	12.302	12.303	0.302	0.310	0.306
	$K_3Ti_3(C_{0.625}N_{0.375})_2$	12.324	12.329	0.481	0.522	0.502
$Ti_3(C_{0.5}N_{0.5})_2$	$K_0Ti_3(C_{0.5}N_{0.5})_2$	12.242	12.223	———	———	———
	$K_1Ti_3(C_{0.5}N_{0.5})_2$	12.277	12.262	0.286	0.319	0.303
	$K_2Ti_3(C_{0.5}N_{0.5})_2$	12.258	12.248	0.131	0.205	0.168
	$K_3Ti_3(C_{0.5}N_{0.5})_2$	12.322	12.308	0.653	0.695	0.674

Table S5 The average adsorption energy (E_{ad}) of Potassium at different concentrations on $Ti_3(C_{1-y}N_y)_2$ MXenes.

Material	K concentration (*x*)	E_{ad} (eV)
$Ti_3(C_{0.875}N_{0.125})_2$	$K_1Ti_3(C_{0.875}N_{0.125})_2$	-0.744
	$K_2Ti_3(C_{0.875}N_{0.125})_2$	-0.368
	$K_3Ti_3(C_{0.875}N_{0.125})_2$	-0.245
$Ti_3(C_{0.75}N_{0.25})_2$	$K_1Ti_3(C_{0.75}N_{0.25})_2$	-0.737
	$K_2Ti_3(C_{0.75}N_{0.25})_2$	-0.427
	$K_3Ti_3(C_{0.75}N_{0.25})_2$	-0.313
$Ti_3(C_{0.625}N_{0.375})_2$	$K_1Ti_3(C_{0.625}N_{0.375})_2$	-0.771
	$K_2Ti_3(C_{0.625}N_{0.375})_2$	-0.434
	$K_3Ti_3(C_{0.625}N_{0.375})_2$	-0.272
$Ti_3(C_{0.5}N_{0.5})_2$	$K_1Ti_3(C_{0.5}N_{0.5})_2$	-0.791
	$K_2Ti_3(C_{0.5}N_{0.5})_2$	-0.390
	$K_3Ti_3(C_{0.5}N_{0.5})_2$	-0.307

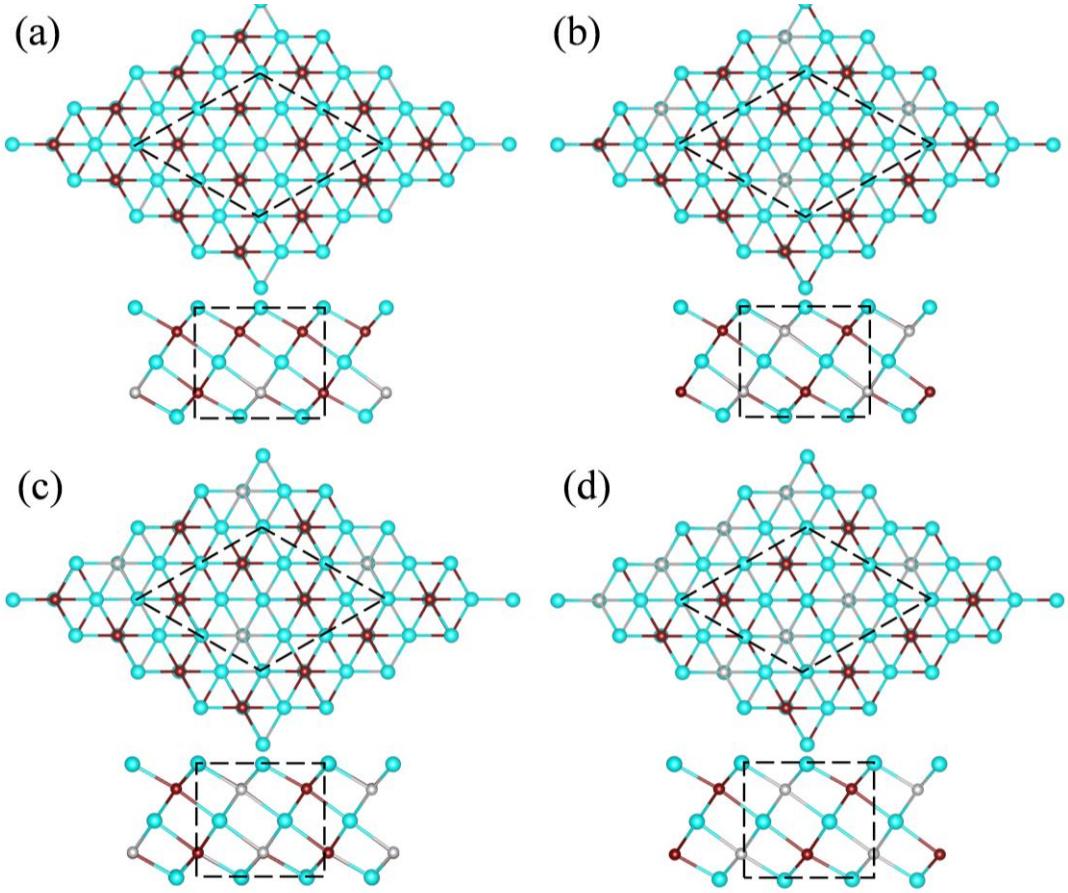

Fig. S1. Top and lateral views of the crystal structures for (a) $Ti_3(C_{0.875}N_{0.125})_2$, (b) $Ti_3(C_{0.75}N_{0.25})_2$, (c) $Ti_3(C_{0.625}N_{0.375})_2$, and (d) $Ti_3(C_{0.5}N_{0.5})_2$ MXenes.

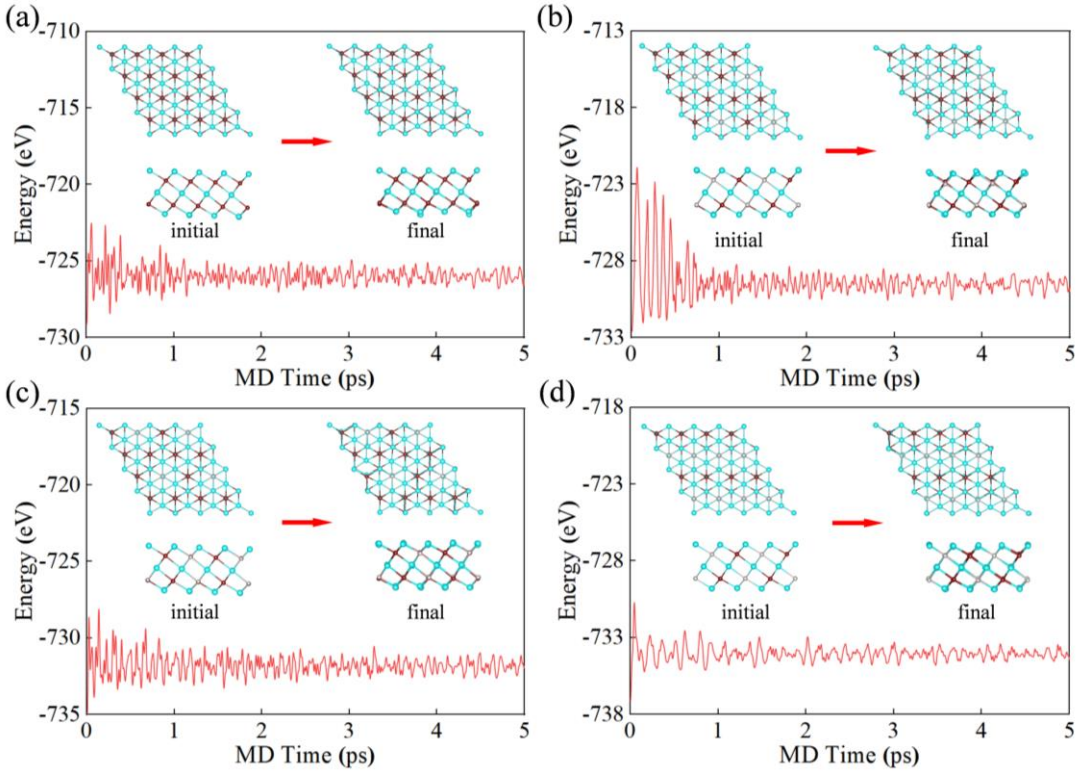

Fig. S2. The Ab-initio Molecular Dynamics simulations at 300K and 5ps for (a) $Ti_3(C_{0.875}N_{0.125})_2$, (b) $Ti_3(C_{0.75}N_{0.25})_2$, (c) $Ti_3(C_{0.625}N_{0.375})_2$, and (d) $Ti_3(C_{0.5}N_{0.5})_2$ MXenes, respectively.

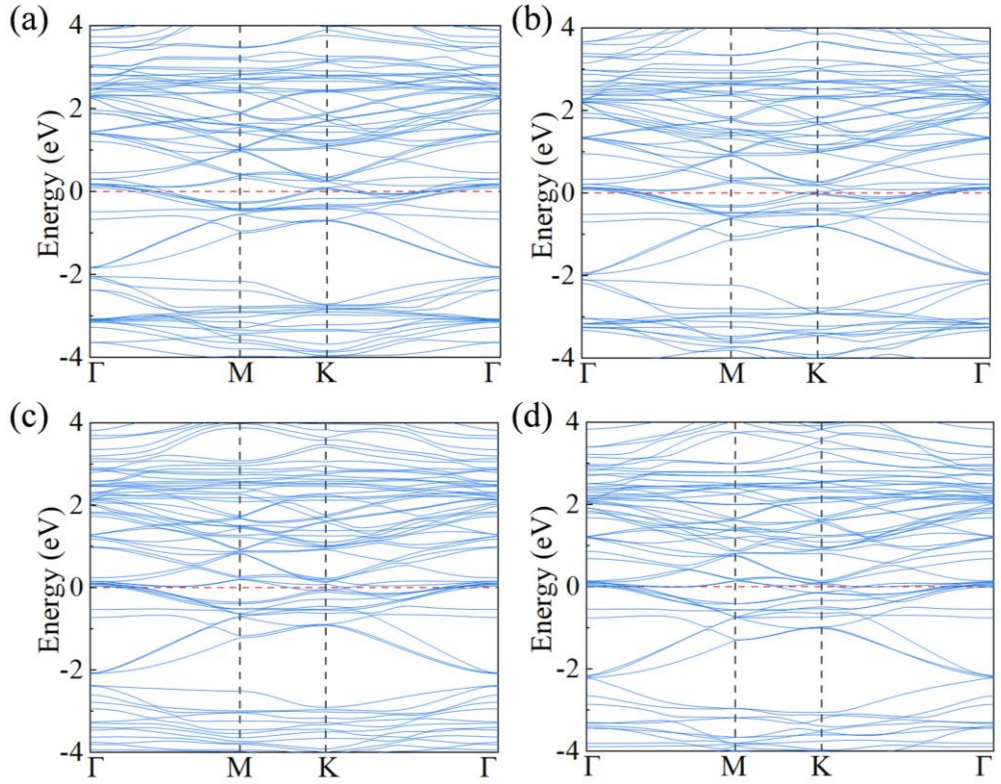

Fig. S3. The band structures of (a) $Ti_3(C_{0.875}N_{0.125})_2$, (b) $Ti_3(C_{0.75}N_{0.25})_2$, (c) $Ti_3(C_{0.625}N_{0.375})_2$, and (d) $Ti_3(C_{0.5}N_{0.5})_2$ MXenes.

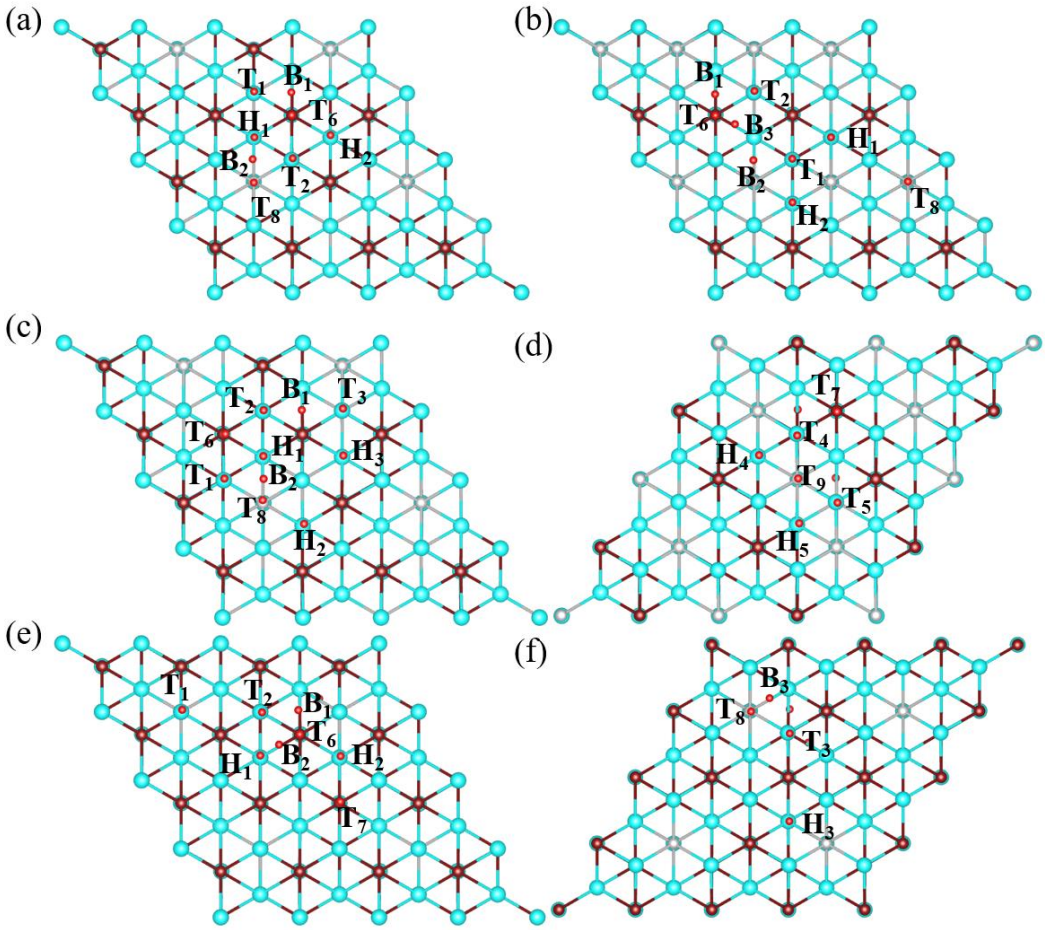

Fig. S4. Top views of K adsorbed on (a) $Ti_3(C_{0.75}N_{0.25})_2$ surfaces, (b) $Ti_3(C_{0.5}N_{0.5})_2$ surfaces; Top and bottom views of K adsorbed on (c-d) $Ti_3(C_{0.625}N_{0.375})$ surfaces and (e-f) $Ti_3(C_{0.875}N_{0.125})_2$, respectively.

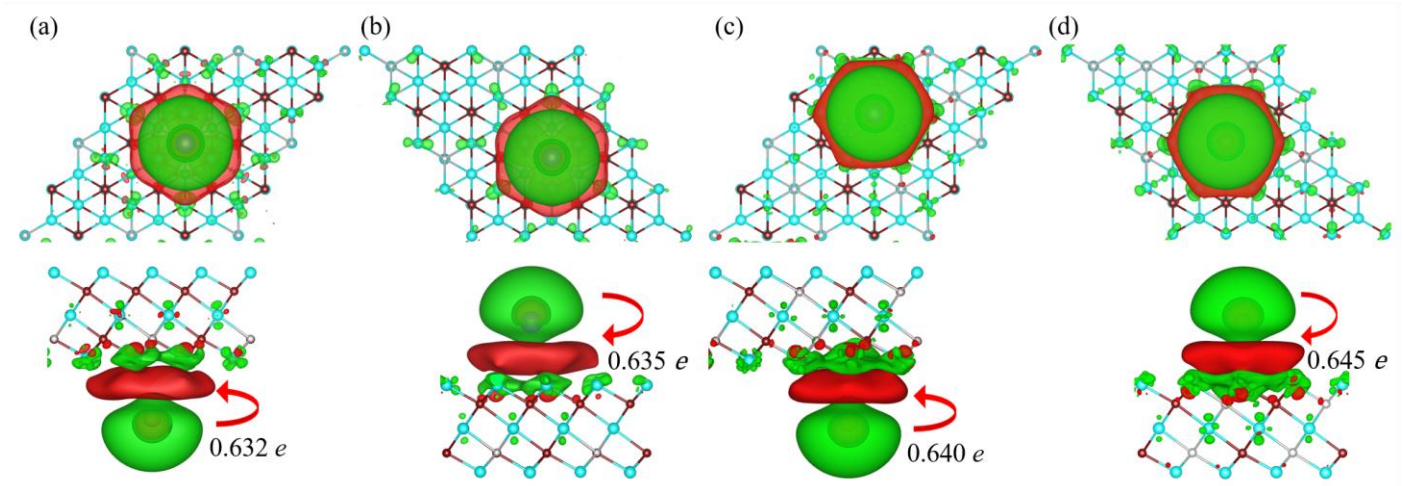

Fig. S5. The charge density difference of K adsorbed on (a) $Ti_3(C_{0.875}N_{0.125})_2$, (b) $Ti_3(C_{0.75}N_{0.25})_2$, (c) $Ti_3(C_{0.625}N_{0.375})_2$, and (d) $Ti_3(C_{0.5}N_{0.5})_2$ surfaces respectively. Red and green represent charge accumulation and consumption, respectively.

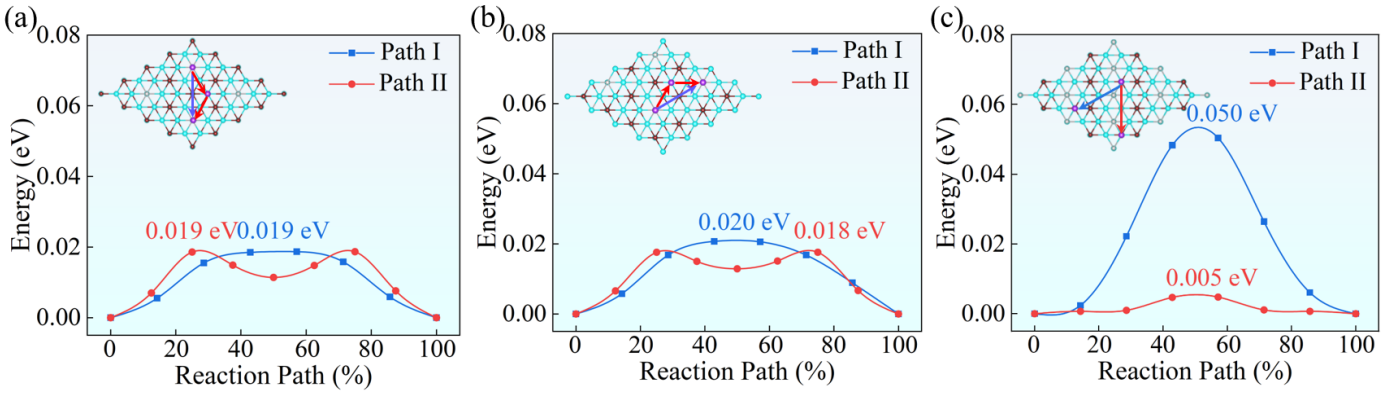

Fig. S6. The diffusion paths and the corresponding diffusion energy barriers of K-ion on (a) $Ti_3(C_{0.875}N_{0.125})_2$, (b) $Ti_3(C_{0.75}N_{0.25})_2$ and (c) $Ti_3(C_{0.625}N_{0.375})_2$ MXenes, respectively.

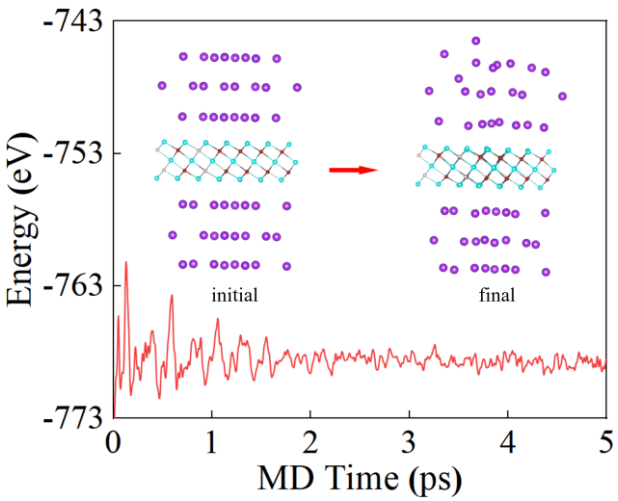

Fig. S7. Ab-initio Molecular Dynamics simulations of $K_3Ti_3(C_{0.5}N_{0.5})_2$ at 300 K over 5 ps.

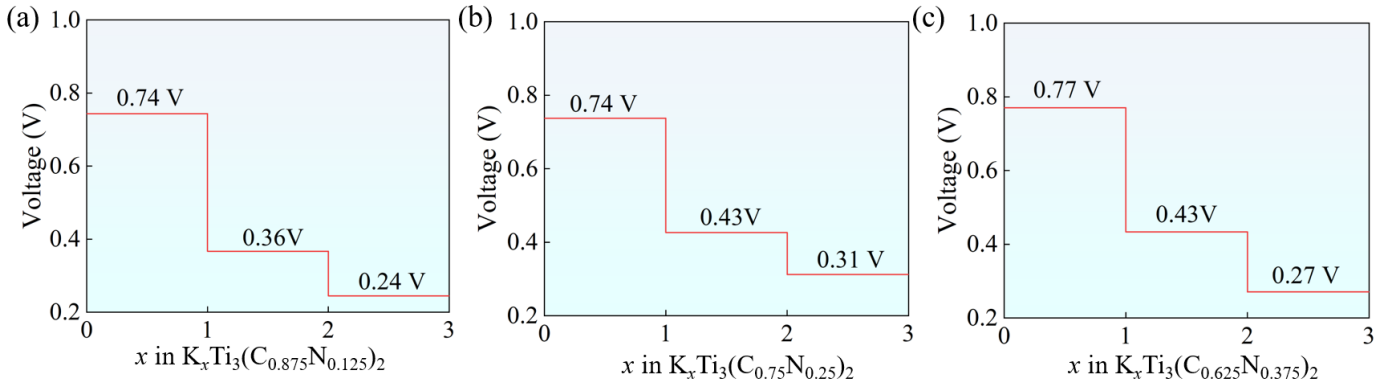

Fig. S8. Voltage platform of (a) $Ti_3(C_{0.875}N_{0.125})_2$, (b) $Ti_3(C_{0.75}N_{0.25})_2$ and (c) $Ti_3(C_{0.625}N_{0.375})_2$ MXenes, respectively. *x* represents concentration.

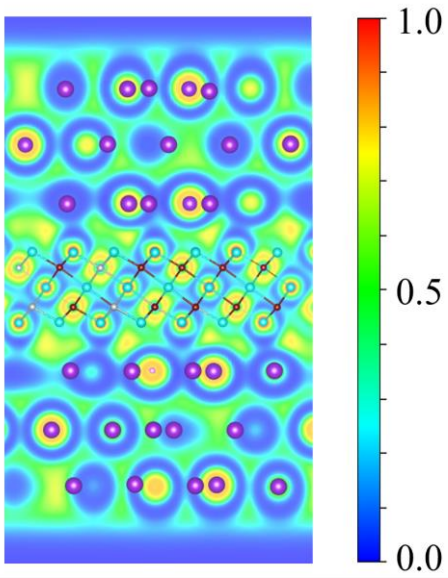

Fig. S9. Side view of electron localization function (ELF) map of $K_3Ti_3(C_{0.5}N_{0.5})_2$.

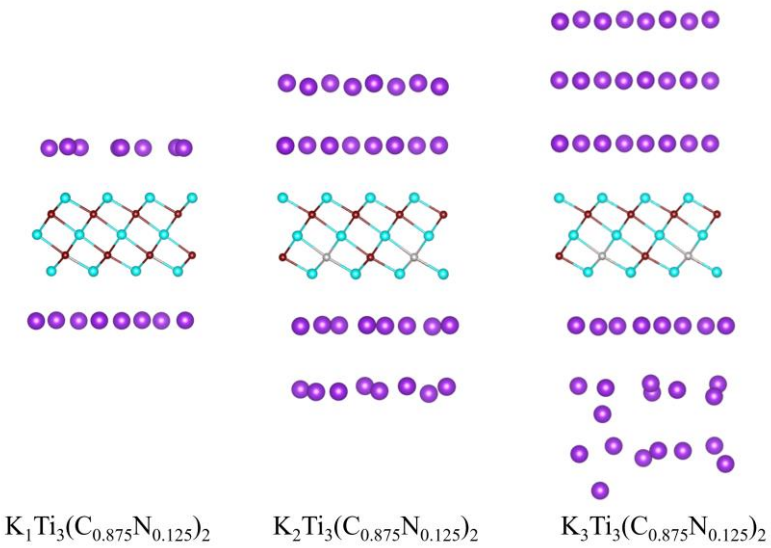

Fig. S10. Side view of the $K_xTi_3(C_{0.875}N_{0.125})_2$ structures with varying potassium concentrations (x = 1, 2, 3).

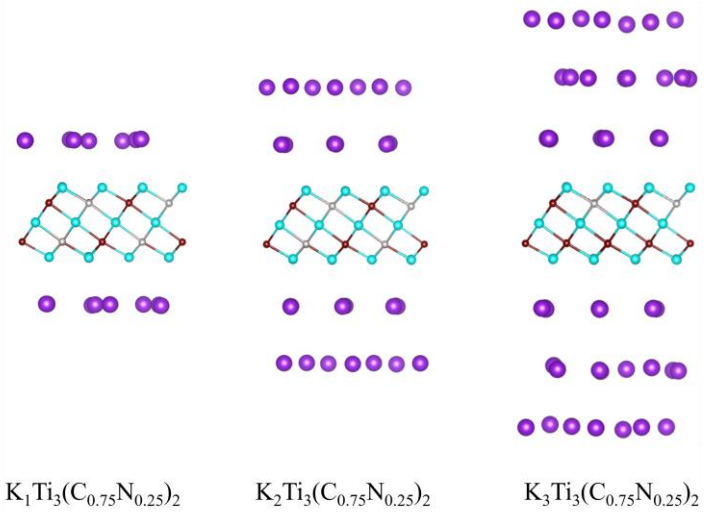

Fig. S11. Side view of the $K_xTi_3(C_{0.75}N_{0.25})_2$ structures with varying potassium concentrations (x = 1, 2, 3).

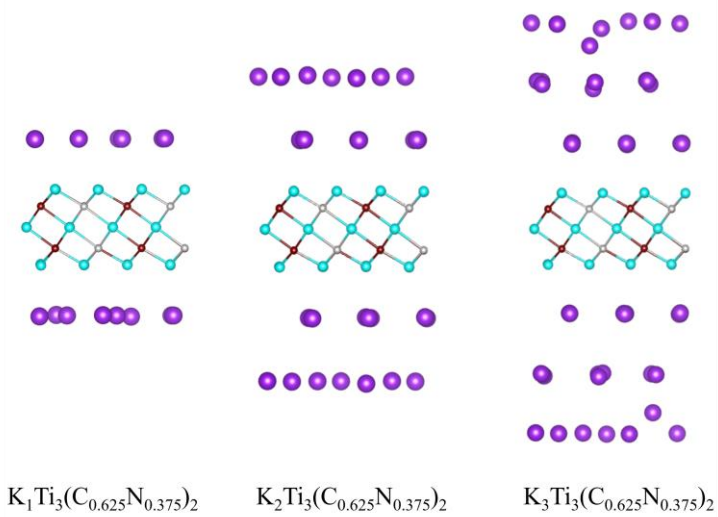

Fig. S12. Side view of the $K_xTi_3(C_{0.625}N_{0.375})_2$ structures with varying potassium concentrations (x = 1, 2, 3).

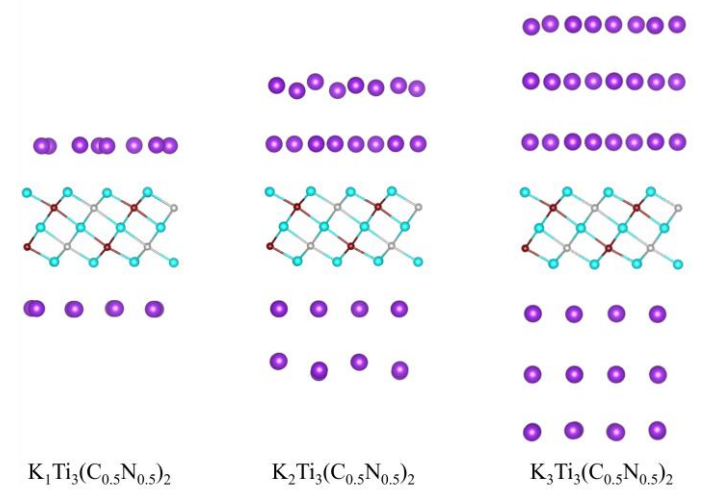

Fig. S13. Side view of the $K_xTi_3(C_{0.5}N_{0.5})_2$ structures with varying potassium concentrations (x = 1, 2, 3).