Electronic Supplementary Material (ESI) for Nanoscale.

A High-throughput Assessing of Adsorption Capacity and Li-ion Diffusion Dynamics in Mo Based Ordered Double-Transition-Metal MXenes as Anode Materials for Fast Charging LIBs

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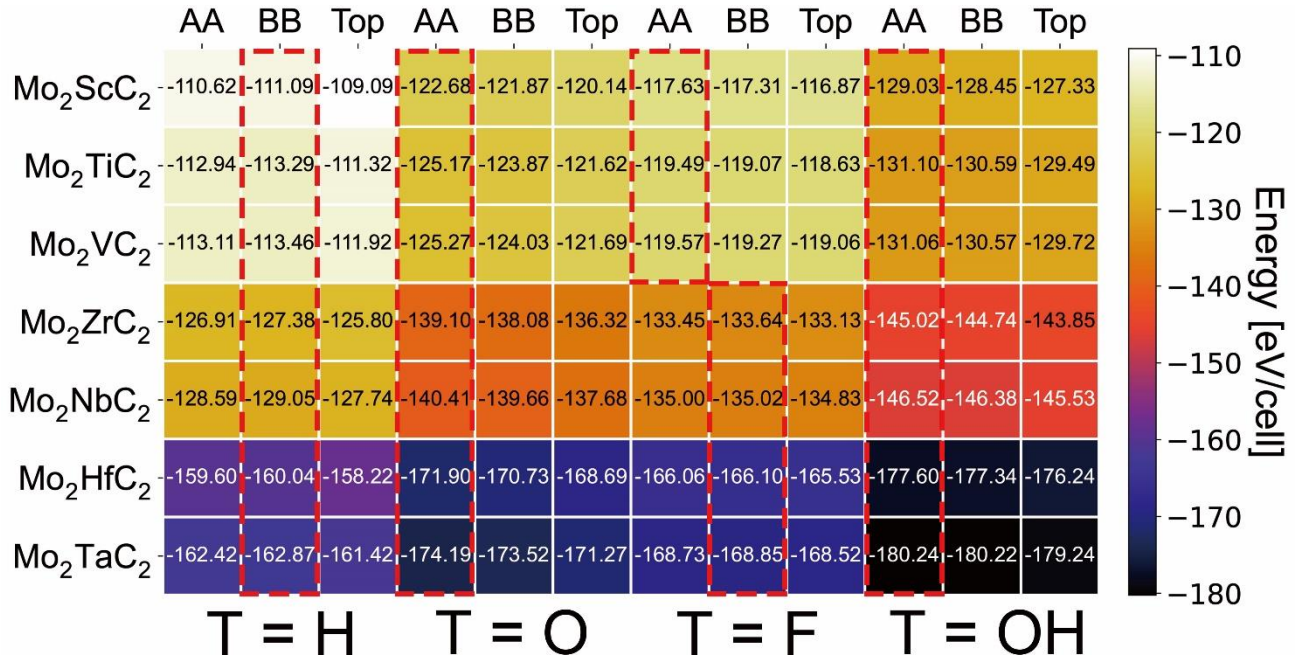


Figure S1. The total energies of Mo-based MXenes with surface functional groups T (H, O, F, and OH) at different adsorption sites. The most stable adsorption structures and associated total cell energies are highlighted in rectangular box by the red-bashed lines.

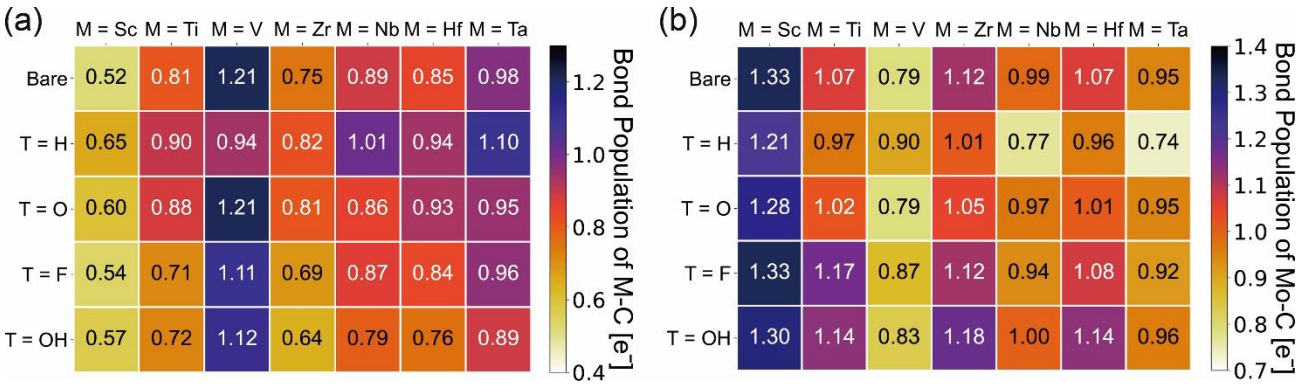


Figure S2. The bond population of (a) M-C and (b) Mo-C in Mo_2MC_2 and $Mo_2MC_2T_2$ (M = Sc, Ti, V, Zr, Nb, Hf, Ta, T = H, O, F, OH).

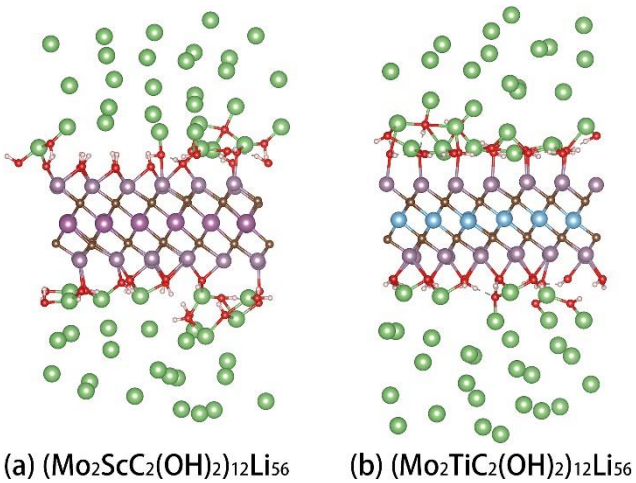


Figure S3. The snapshots of -OH functionalized MXenes placed in between two bulk Li metals in FPMD simulations at 200 K: (a) Mo_2ScC_2 and (b) Mo_2TiC_2.

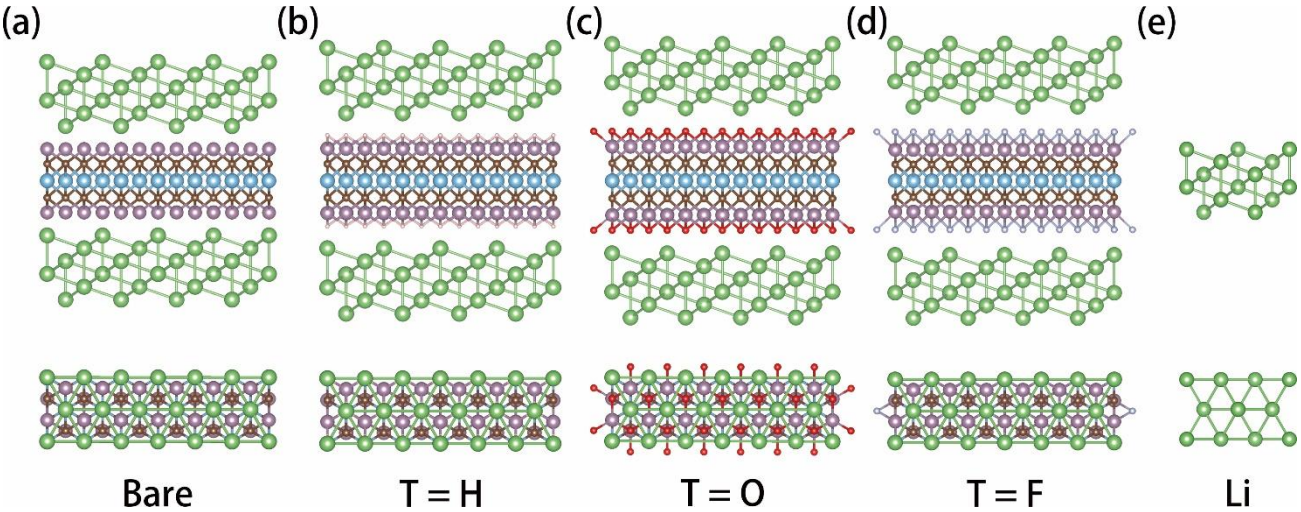


Figure S4. The top and side view of initial sandwich structures of (a) Mo_2MC_2, (b) $Mo_2MC_2H_2$, (c) $Mo_2MC_2O_2$, (d) $Mo_2MC_2F_2$ and the crystal-ographic plane (111) of bulk Li metal (BCC).

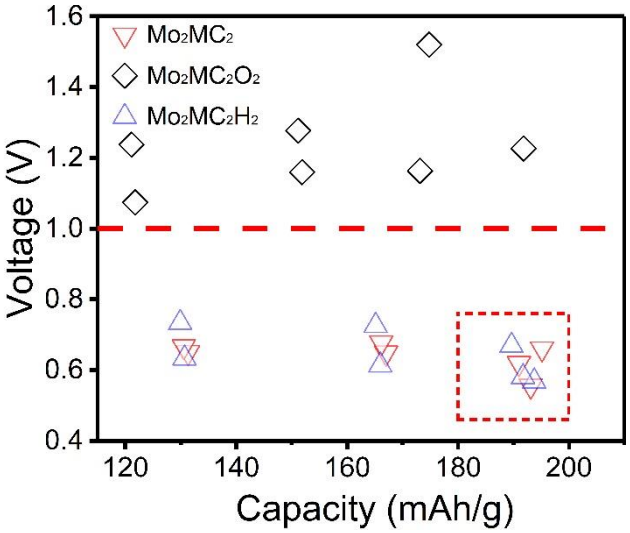


Figure S5. Average open circuit voltage and theoretical capacity of Li saturated adsorption structures of Mo_2MC_2 and $Mo_2MC_2T_2$ (M stands for transition metal, and T = H, and O). The red box area indicates an optimal active voltage window and with storage capacity greater than 180 mAh/g.

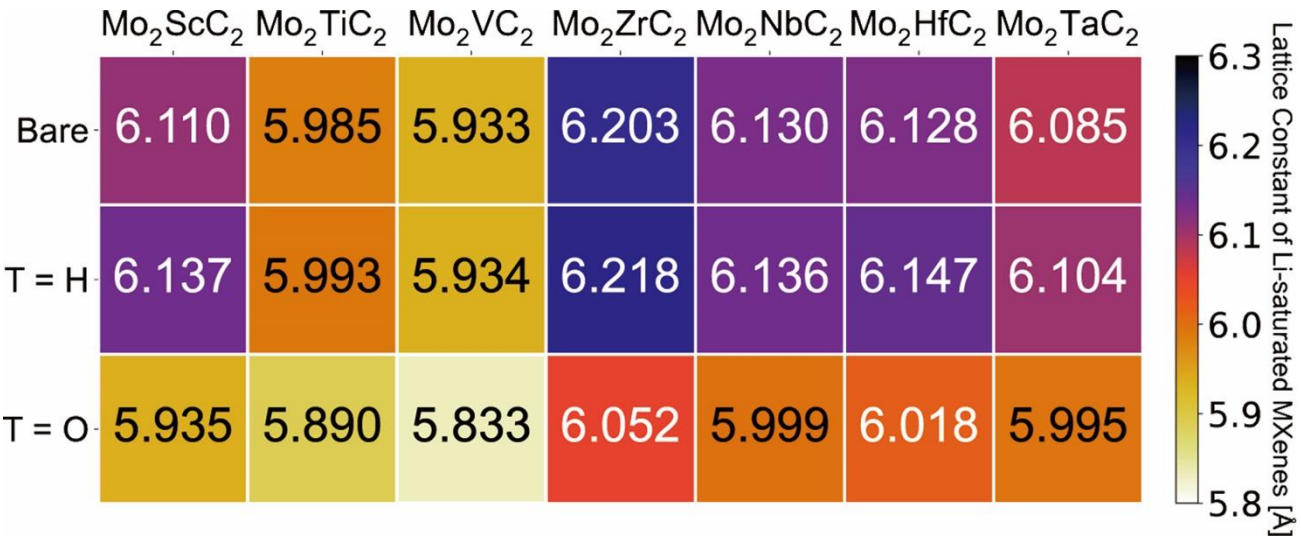
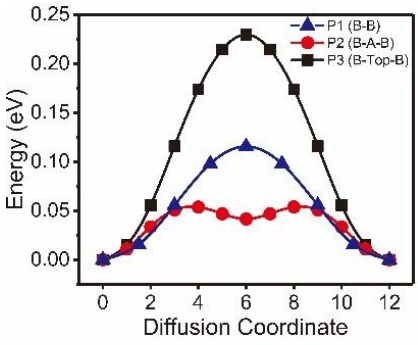
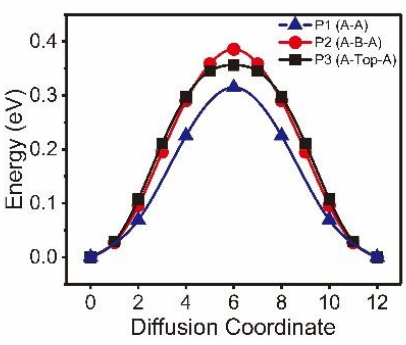


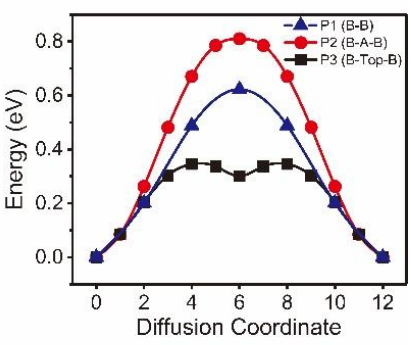
Figure S6. Planar lattice constants of 2×2×1 supercell model of Mo_2MC_2, $Mo_2MC_2H_2$ and $Mo_2MC_2O_2$ (M = Sc, Ti, V, Zr, Nb, Hf, Ta) with full Li adsorbed.



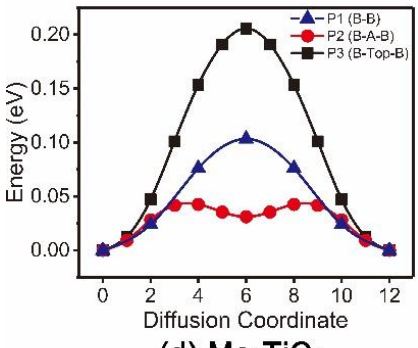
(a) Mo_2ScC_2



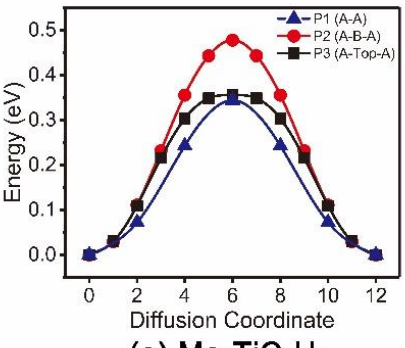
(b) $Mo_2ScC_2H_2$



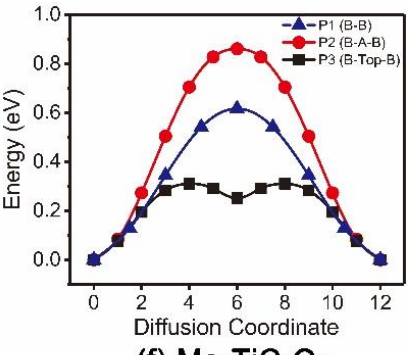
(c) $Mo_2ScC_2O_2$



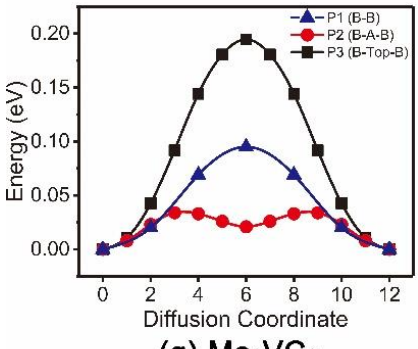
(d) Mo_2TiC_2



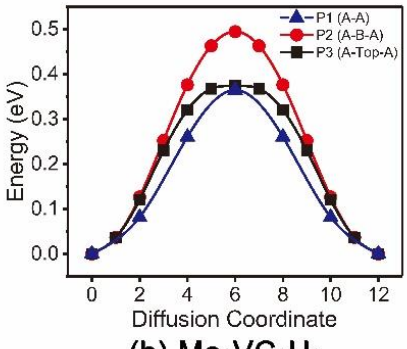
(e) $Mo_2TiC_2H_2$



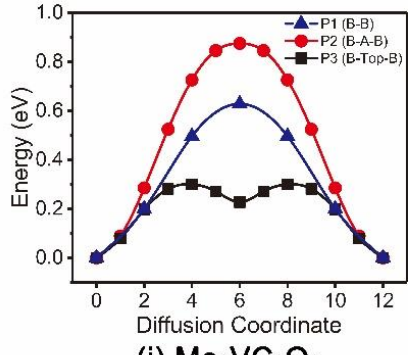
(f) $Mo_2TiC_2O_2$



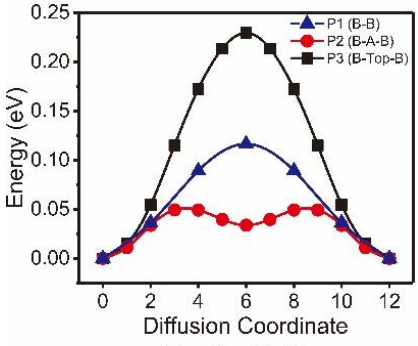
(g) Mo_2VC_2



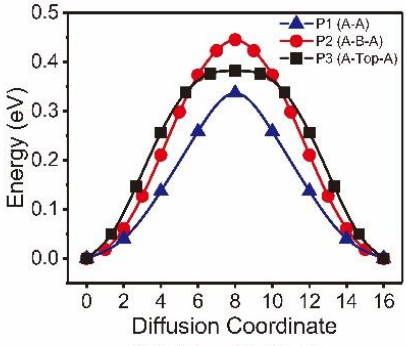
(h) $Mo_2VC_2H_2$



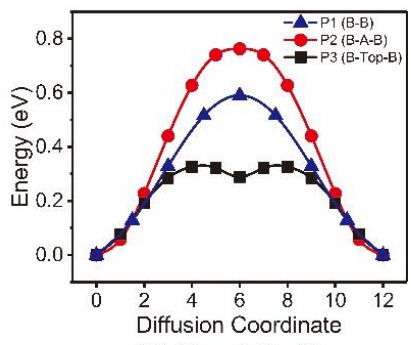
(i) $Mo_2VC_2O_2$



(j) Mo_2ZrC_2



(k) $Mo_2ZrC_2H_2$



(l) $Mo_2ZrC_2O_2$

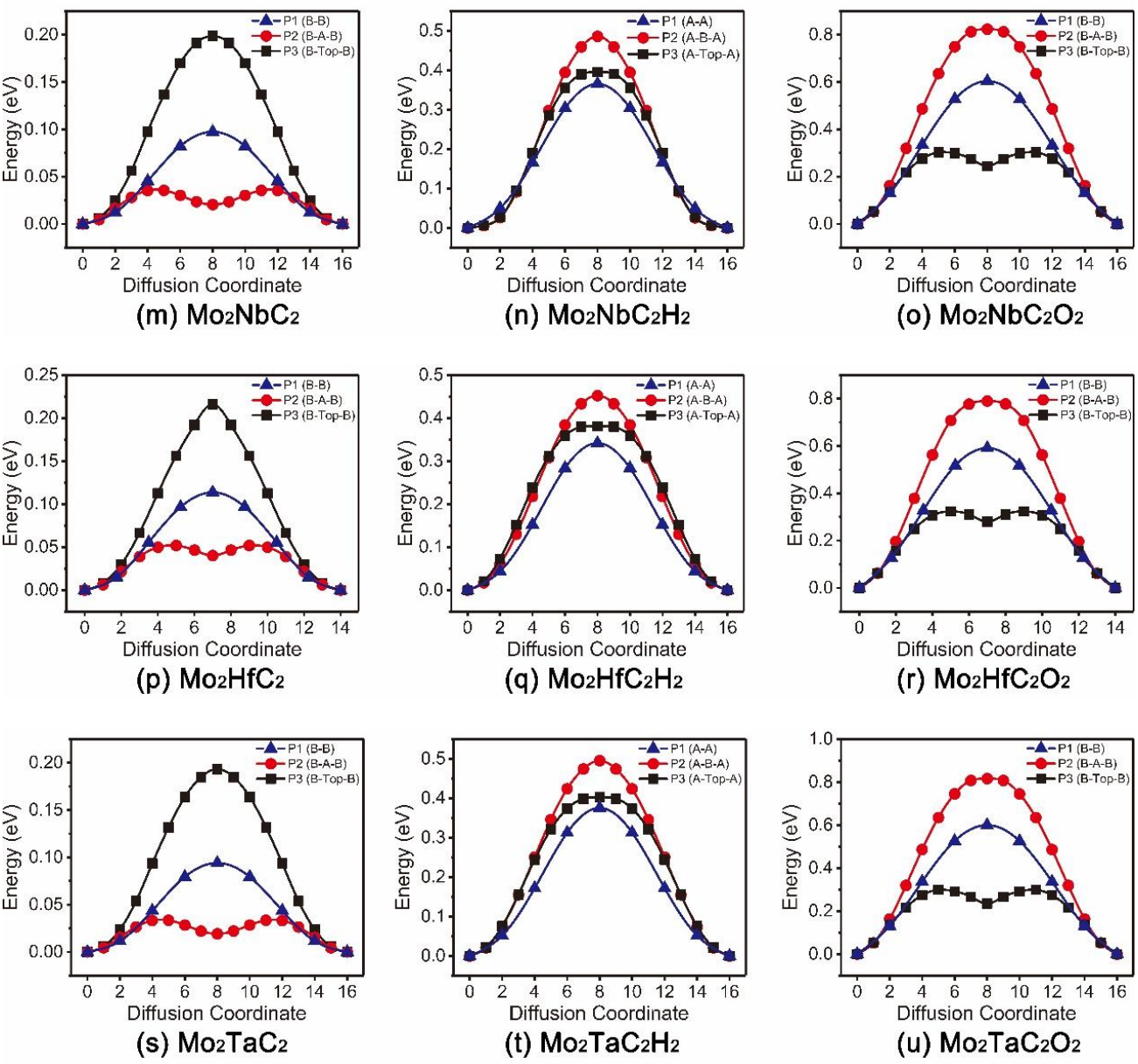


Figure S7. Diffusion energy profiles of Li-ion for Mo_2MC_2 and $Mo_2MC_2T_2$ (M = Sc, Ti, V, Zr, Nb, Hf, Ta, and T = H, O) MXenes, calculated using a 2×2×1 supercell model.

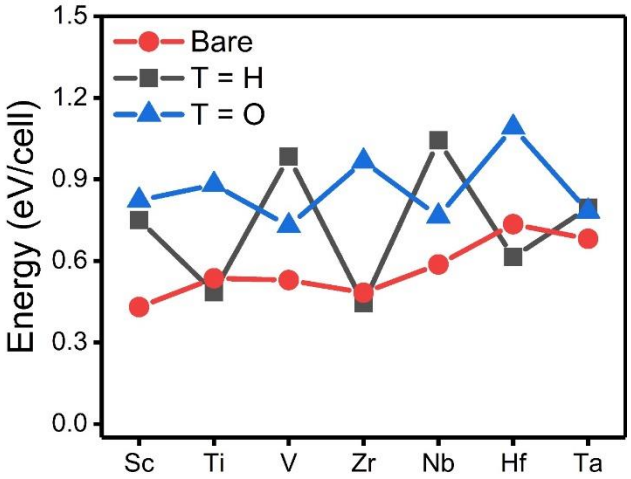


Figure S8. Formation energy of a Li vacancy in saturated adsorption structures $(Mo_2MC_2)_4Li_8$ and $(Mo_2MC_2T_2)_4Li_8$ (T = H, O).

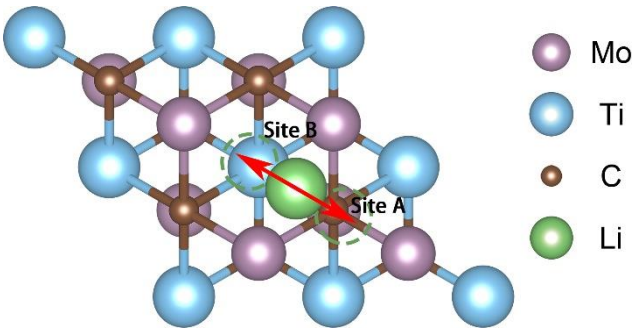


Figure S9. The phonon eigenvector of the soft mode at Γ-point for lithium atom at the saddle point. The direction of the vibration is the same as that of the diffusion.

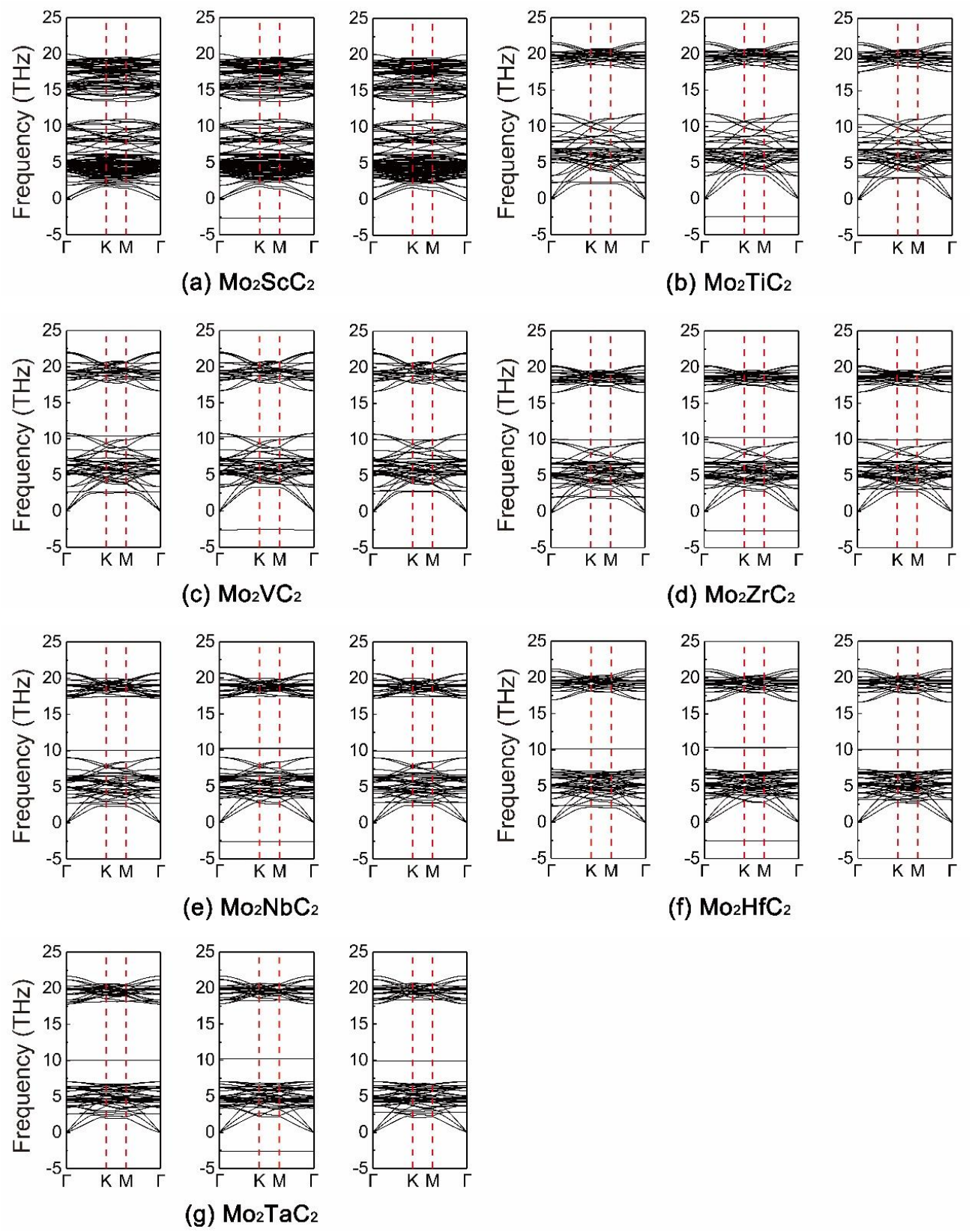


Figure S10. Calculated phonon band vibrational spectra of Li-$(Mo_2MC_2)_4$ (M = Sc, Ti, V, Zr, Nb, Hf, Ta) at the global energy minimum, the saddle-point, and metastable state on the diffusion pathways. Soft modes are displayed as the horizontal lines below 0 THz for all structure at saddle-point.

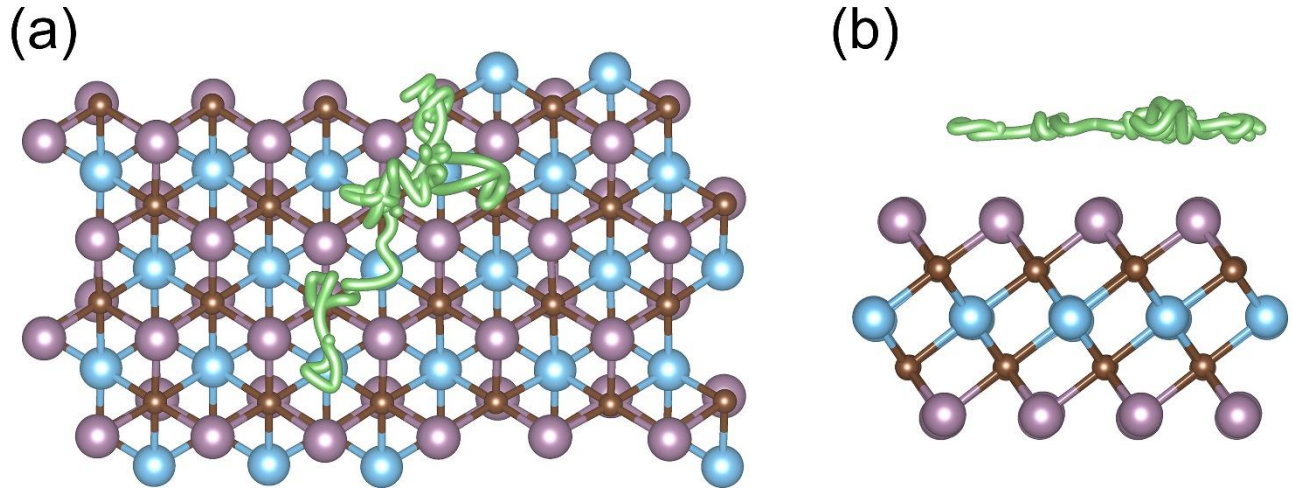


Figure S11. (a) Top view and (b) side view of the migration pathway of Li atom from FPMD simulation for Mo_2TiC_2 monolayer at 400K.

Table S1. The adsorption energies E_{ab} of Li atoms on Mo_2ScC_2 and $Mo_2ScC_2T_2$ with spin-polarized and non-spin-polarized calculations.

	Adsorption energy (eV/cell)	
	Spin-polarized	Non-spin-polarized
Mo_2ScC_2	-0.74	-0.74
$Mo_2ScC_2H_2$	-0.438	-0.440
$Mo_2ScC_2O_2$	-3.05	-3.05
$Mo_2ScC_2F_2$	-0.158	-0.159
$Mo_2ScC_2(OH)_2$	0.66	0.66

Table S2. The bond lengths of Mo-C and C-M bonds for embedded MXene layers in between two overlayers after FPMD simulations ($(Mo_2MC_2F_2)_{12}Li_{48}$).

	$d_{Mo\text{-}C}$ (Å)	$d_{C\text{-}M}$ (Å)	$d_{Mo\text{-}F}$(Å)
$Mo_2ScC_2F_2$	2.15	2.28	3.63
$Mo_2TiC_2F_2$	2.13	2.16	3.72
$Mo_2VC_2F_2$	2.09	2.07	3.86
$Mo_2ZrC_2F_2$	2.20	2.38	-
$Mo_2NbC_2F_2$	2.09	2.25	-
$Mo_2HfC_2F_2$	2.11	2.25	-
$Mo_2TaC_2F_2$	2.10	2.17	3.37

Table S3. The soft mode frequencies, diffusion barrier heights and corresponding diffusion coefficient obtained from TST+DFPT method, based on CI-NEB migration pathways for all intrinsic MXenes.

	Virtual frequency (THz)	Barrier height (eV)	Diffusion coefficient (m^2/s)
Mo_2ScC_2	-2.678	0.055	2.54×10^{-8}
Mo_2TiC_2	-2.466	0.043	3.14×10^{-8}
Mo_2VC_2	-2.582	0.034	4.25×10^{-8}
Mo_2ZrC_2	-2.639	0.049	2.46×10^{-8}
Mo_2NbC_2	-2.646	0.036	4.36×10^{-8}
Mo_2HfC_2	-2.508	0.052	2.60×10^{-8}
Mo_2TaC_2	-2.598	0.033	4.42×10^{-8}

Table S4. Comparison of diffusion coefficients of Li atoms calculated for Mo_2MC_2 (M = Sc, Ti, V) at 400K using TST+DFPT and FPMD simulations.

	Diffusion coefficient (m^2/s)		
	Mo_2ScC_2	Mo_2TiC_2	Mo_2VC_2
TST analysis	4.21×10^{-8}	4.35×10^{-8}	5.70×10^{-8}
FPMD	6.36×10^{-9}	2.70×10^{-9}	9.77×10^{-9}