

Tunable Li Diffusion in MXene Bilayers: Insights from First-Principles Calculations

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We investigate the properties of M_2CT_2 ($M = \text{Sc, Ti, V, Nb, Cr}$; $T = \text{O, F}$) MXenes using first-principles density functional theory. Our study explores monolayer and bilayer configurations, focusing on stability, lithium diffusion, selectivity, and cointercalation in Ti_2CO_2 bilayers. For monolayers, we identify the most stable functional group (T) adsorption sites. In bilayers, structural variations and interlayer spacing significantly influence lithium insertion and mobility. We analyze lithium diffusion barriers, rates, and selectivity, revealing that cointercalation (e.g., potassium) reduces the diffusion barrier from 0.36 eV to ~ 0.01 eV as interlayer spacing expands from 3.4 Å to 4.1 Å, enabling very low diffusion barrier for Li transport. Further spacing increases shift the diffusion barrier toward monolayer-like behavior, favoring surface migration. Our findings establish a computational framework for tuning lithium transport in MXene bilayers, offering insights into structure-diffusion interplay for energy storage applications.

References

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