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Reaction Paths, Transition States and Catalysis in $Li_4BN_3H_{10}$ from First Principles[1] DAVID FARRELL, CHRIS WOLVERTON, Dept of Materials Science and Engineering, Northwestern University — Thermodynamic analyses of the complex hydride $Li_4BN_3H_{10}$ (>10 wt.% H_2) predicted favorable hydrogen desorption reactions in the solid, whereas experiments found temperatures above melting were needed before appreciable H_2 desorption was observed, and the material released NH_3 at approximately the same temperature. More recent experimental studies successfully catalyzed H_2 desorption using $CoCl_2$ and $NiCl_2$, decreasing the H_2 release temperature. To elucidate the catalytic and decomposition mechanisms that resulted in the measured changes, we have applied *Ab Initio* Molecular Dynamics and Transition State Theory methods to find reaction pathways and determine the rate limiting steps in pristine and catalyzed $Li_4BN_3H_{10}$. We observed the formation of several important reaction intermediates, as well as free H_2 and NH_3 in the bulk liquid. Finally, We studied the formation of vacancies and interstitials that are promising candidates for rate-limiting steps in the desorption reactions and determined energy barriers for each reaction step.

[1]The computations were performed in part on the Kraken system at the National Institute for Computational Sciences and the Intrepid system at the Argonne Leadership Computing Facility

David Farrell
Dept of Materials Science and Engineering, Northwestern University

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