

Abstract Submitted
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Decomposition mechanisms in metal borohydrides and their ammoniates¹ EVAN WELCHMAN, TIMO THONHAUSER, Wake Forest University — We present results from *ab initio* calculations to explain the mechanisms that cause ammoniation to lower decomposition temperatures in metal borohydrides ($\mathcal{M}(\text{BH}_4)_x$, MBs) with \mathcal{M} of low electronegativity ($\chi_p < 1.6$), but raise decomposition temperatures of high- χ_p MBs ($\chi_p > 1.6$). Results include an examination of materials' dihydrogen bond networks, energy of formation calculations, and transition state searches for potential decomposition mechanisms. We find that ammoniation always causes thermodynamic destabilization, helping to explain lower decomposition temperatures for low- χ_p MBs. For high- χ_p MBs, we find that ammoniation blocks B_2H_6 formation, the preferred decomposition mechanism in these MBs. We describe two ways that B_2H_6 formation can be blocked, which are each valid in one of the ammoniated MBs ($\mathcal{M}(\text{BH}_4)_x \cdot y\text{NH}_3$, MBAs) studied. We also consider why these materials release either H_2 or NH_3 gas upon decomposition. We find that NH_3 is much more strongly coordinated with higher- χ_p metals, and that direct H_2 formation becomes easier in higher- χ_p materials, contributing to why higher- χ_p MBAs are more likely to produce H_2 upon decomposition.

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