

FIRST-PRINCIPLES PREDICTION OF HYDROGEN STORAGE ENERGETICS IN THE Li-B-N-H and Sr-Al-H SYSTEMS. [W.Sun](#)¹, C. Wolverton^{*1}, A. Akbarzadeh² and V. Ozolins², Northwestern University¹, Materials Science and Engineering, Evanston, IL 60201; Univ. of California at Los Angeles², Los Angeles, CA, c-wolverton@northwestern.edu

A significant bottleneck in the widespread introduction of hydrogen vehicles is finding a suitable onboard hydrogen storage technology. One possible solution which is eliciting a large world-wide research effort involves the use of a reversible solid-state material as the storage medium. However, current hydrides do not satisfy the stringent criteria of high volumetric and gravimetric capacity, coupled with suitable thermodynamic and kinetic properties for hydrogen insertion and release. Theoretical and computational materials science has been effective in predicting the performance of these materials before they are synthesized in laboratories, accelerating the search for more promising storage materials. In this talk, we describe recent efforts using first-principles density functional theory (DFT) based methods to elucidate the energetics of two hydrogen storage systems: Li-B-N-H and Sr-Al-H.

For the Li-B-N-H system, we present DFT energetics of a large number of phases in this system, including the recently-discovered Li₂BNH₆ compound, which has a high theoretical hydrogen storage density of 13.3 wt%. To predict the decomposition path of this compound (and others in this system) as a function of temperature, we use the DFT energies in a Gibbs free energy formalism combined with a linear programming approach. Our results can be used to determine whether further experimental study of this system is recommended.

For the Sr-Al-H system, we present DFT results of the energetic and decomposition reactions, specifically focused on the Sr(AlH₄)₂ compound. In this case, the crystal structure of Sr(AlH₄)₂ is currently unknown, and so we used DFT in a “database-searching” method to predict possible low-energy states of this material, as well as possible decomposition products, including Sr₃(AlH₆)₂, Sr₂AlH₇, SrAlH₅, SrAl₄, and SrH₂. The ground state crystal structure search for each of these crystals involved, on average, 50 candidate crystal structures for each phase from the Inorganic Crystal Structure Database, all computed using DFT. The decomposition path and its thermodynamics was then predicted in the same manner as the Li-B-N-H system.

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