

First-principles engineering of advanced hydrogen storage materials

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Hydrogen-fueled vehicles require a cost-effective, light-weight material that binds hydrogen strongly enough to be stable at ambient pressures and temperatures but weakly enough to liberate H₂ with minimal heat input. Since none of the simple metal hydrides satisfy all the requirements for a practical H₂ storage system, recent research efforts have turned to complex hydrides and advanced multicomponent material compositions. We will show how first-principles density-functional theory (DFT) calculations have become a valuable tool for understanding and predicting novel hydrogen storage materials. Recent studies in our group have used DFT calculations to (i) predict crystal structures of new solid-state hydrides, (ii) determine phase diagrams and thermodynamically favored reaction pathways in multinary hydrides, and (iii) study microscopic kinetics of hydrogen release reactions. We have developed theoretical methods for determining crystal structures and thermodynamic properties of novel complex hydrides, which allow accurate theoretical predictions of hydrogenation enthalpies without any experimental input. Using Li-Mg-N-H and Li-Mg-B-N-H as examples, we will demonstrate that phase diagrams and hydrogenation reactions in multicomponent systems can be determined entirely from the first principles. Finally, we will show recent DFT results that elucidate the kinetics of H₂ release and mass transport in the prototypical complex hydride, sodium alanate.