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First-Principles Studies of Phase Stability and Reaction Dynamics in Complex Metal Hydrides¹

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Complex metal hydrides are believed to be one of the promising materials for developing hydrogen storage systems that can operate under desirable conditions. At the same time, these are also a class of materials that exhibit intriguing properties. We have applied state-of-the-art computational techniques to study the structural, dynamic, and electronic properties of these materials. This talk will focus on the critical role played by the Ti catalyst in helping hydrogen cycling in the alanates, which remains a challenging topic for this hydrogen storage material. We have performed a series of calculations to address the hydrogen interaction on the aluminum surface in the presence of the Ti “dopant,” focusing on the effect of near-surface alloying on the Al(100) surface. It is found that Ti occupies subsurface sites near the Al surface. This subsurface Ti arrangement not only enhances H binding with the Al surface layer, but also improves H mobility on the surface. Based on existing experimental data and our preliminary results, we propose a model in which the catalyst does not enter the bulk, but facilitates hydrogen dissociation-recombination near the surface. In the dehydrogenation cycle, the catalyst kinetically facilitates the release and decomposition of AlH_3 from the solid-state alanate. In the hydrogenation cycle, the catalyst helps the adsorption of hydrogen and the formation of AlH_3 oligomers on Al surfaces. The implication of Ti as a catalyst for the hydrogenation reactions will be discussed.

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