Dehydrogenation Mechanism of Chemical Hydrogen Storage Materials: Catalytic Role of Metal

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Hydrogen is a promising choice of clean energy carrier for both transportation and stationary applications. Hydrogen storage materials via chemical approach have attracted intense interest nowadays. Varieties of promising materials, for instance metal-hydride and metal-N-H systems, have been developed in recent years with high hydrogen %wt and promising reversible hydrogen storage capacity. Molecular-level, mechanistic understanding of the hydrogenation and dehrogenation mechanisms of these hydrogen storage materials is critical for optimizing existing materials and rational development of novel hydrogen storage materials. In general, limited mechanistic information is available in experimental studies. Theory provides a powerful and insightful complementary approach to map out the detailed mechanism and understand the governing factors of the thermodynamics and kinetics of requirements of a particular hydrogenation/dehydrogenation reaction. In this presentation, we report our computational studies of the dehydrogenation mechanisms of several boron-based metal-N-H systems. We found that the various stages of H_2 release of these hydrogen storage materials are characterized by a common two-step mechanism: hydride transfer followed by H_2 release. The metal atom plays an important and crucial catalytic role in bringing two hydrogen atoms ($H^{\delta+}$ and $H^{\delta-}$) in close proximity and lowering the activation barrier of hydrogen release. A similar catalytic role is found for metal-doped hydrogen storage materials.