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Insights from ab initio studies of hydrogen storage systems

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Summary

The purpose of this talk is to provide an overview of the most recent theoretical studies undertaken by us in the field of hydrogen storage materials research. On selected examples, the application of our computational tool of choice, density functional theory, will be illustrated to show how ab initio calculations can be of use in the effort to reach a better understanding of hydrogen storage materials and to occasionally also guide the search for new promising approaches. Systems to be discussed include: Metal-organic frameworks, where we have studied hydrogen physisorption in three different types of iso-reticular MOFs, namely Zn-/Mg-/Ca-MOF16, decorated with either Li, Na, or K. Lithium amidoborane-ammonia borane (LiNH2BH3A.NH3BH3) which was synthesized recently and shows better dehydrogenation kinetics than LiNH2BH3 and NH3BH3 by themselves. Lithium ion diffusion in lithium nitride (Li3N), lithium imide (Li2NH), and lithium amide (LiNH2) studied by us using both ab initio molecular dynamics simulations and the nudged elastic band method. Adding potassium (in the form of KH) to Mg(NH2)2/LiH can drastically improve the hydrogen desorption. First-principles calculations suggest that the presence of potassium in the reacting system weakens the amide N-H and imide Li-N bonds, resulting in the observed enhanced reaction kinetics. Finally, catalysts play an important role in many hydrogen desorption processes. We found (through a combination of experiment and theory) that carbon nanostructures, in particular nanotubes and fullerenes, can be used as catalyzing agents for hydrogen uptake and release in complex metal hydrides (such as sodium alanate, NaAlH4) and provide a model which could explain the mechanism of the catalytic effect.