

## Atomic and Electronic Structure of Hydrogen-Related Centers in Hydrogen Storage Materials

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Hydrogen can serve as an energy carrier in a carbon-neutral system of energy production and use, but adequate hydrogen storage materials are still lacking in spite of many decades of investigations. In addition to being reversible and meeting stringent weight % and volume criteria, candidate materials must exhibit favorable kinetics for hydrogen uptake and release. The fundamental mechanisms of the (de)hydrogenation process have remained elusive to date. In this talk I will discuss first-principles studies that have allowed us to identify the mechanisms of hydrogen transport in non-metallic hosts.

First-principles calculations are playing an important role in developing a fundamental understanding of the physics and chemistry of hydrogen interactions with materials. Indeed, they have contributed to the identification of a number of centers that have been observed with  $\mu$ SR spectroscopy [1,2]. In the case of hydrogen storage materials, hydrogen-related defect centers must play a central role in kinetics. In order to accurately describe the mechanisms of hydrogen uptake and release that are at the core of the hydrogen storage process, it is necessary to consider the addition or removal of individual hydrogen atoms. We have performed density functional calculations to model addition or removal of hydrogen, corresponding to the formation of hydrogen interstitials or vacancies. While the concepts discussed here are general, I will illustrate them with detailed results for sodium alanate, a viable hydrogen storage material [3]. We find that hydrogen-related point defects are the dominant defect species involved in (de)hydrogenation of sodium alanate. These defects are positively or negatively charged, and hence their formation energies are Fermi-level dependent – an important feature that has not been recognized in past studies. This dependence enables us to explain why small amounts of transition-metal additives drastically alter the kinetics of dehydrogenation. Some of the defects induce large lattice relaxations, which will be discussed in detail.

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