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Role of point defects and additives in kinetics of hydrogen storage materials[1]

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First-principles computational studies of hydrogen interactions with storage materials can provide direct insight into the processes of H uptake and release, and may help in developing guidelines for designing storage media with improved storage capacity and kinetics. One important conclusion is that the defects involved in kinetics of semiconducting or insulating H-storage materials are charged, and hence their formation energy is Fermi-level dependent and can be affected by the presence of impurities that change the Fermi level [1,2]. This provides an explanation for the role played by transition-metal impurities in the kinetics of $NaAlH_4$ and related materials. Desorption of H and decomposition of $NaAlH_4$ requires not only mass transport of H but also of Al and/or Na. This process is mediated by native defects. We have investigated the structure, stability, and migration enthalpy of native defects based on density functional theory. The results allow us to estimate diffusion activation energies for the defects that may be involved in mass transport. Most of the relevant defects exist in charge states other than neutral, and consideration of these charge states is essential for a proper description of kinetics. We propose specific new mechanisms to explain the observed activation energies and their dependence on the presence of impurities. We have also expanded our studies to materials other than $NaAlH_4$. In the case of $LiBH_4$and $Li_4BN_3H_{10}$we have found that the calculations have predictive power in terms of identifying which impurities will actually enhance kinetics. Other complex hydrides that we are currently investigating include Li_2NH and $LiNH_2$.

[1] A. Peles and C. G. Van de Walle, Phys. Rev. B 76, 214101 (2007).
[2] C. G. Van de Walle, A. Peles, A. Janotti, and G. B. Wilson-Short, Physica B 404, 793 (2009).

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