

A first-principles molecular dynamics study of hydrogen diffusion in select fuel-cell materials

Brandon Wood and Nicola Marzari

Department of Materials Science and Engineering, Massachusetts Institute of Technology

Cambridge, MA, USA

The emergent worldwide energy crisis has prompted a recent push toward the realization of the "hydrogen economy", yet key technological hurdles to the large-scale adoption of fuel-cell technology remain. Key among the materials challenges are the discovery of more efficient solid electrolytes and candidates for lightweight, reversible hydrogen storage in the solid state. However, progress in this regard has been somewhat curtailed by a fundamental lack of understanding of the detailed atomistic behavior and dynamic electronic structure of diffusive hydrogen in such materials.

We present results of first-principles molecular dynamics on CsHSO_4 , an anhydrous solid-state electrolyte material with superprotonic character; and NaAlH_4 , a complex metal hydride with promising hydrogen storage applications. The dynamic nature of the chemical interactions between ionic species in these materials makes a first-principles approach particularly attractive.

For CsHSO_4 , we discuss the key transport mechanisms involved, including cooperative proton diffusion and jump behavior, and suggest structural and chemical motivations for the anomalously high protonic conductivity. We also examine the hydrogen bond network topology in order to extract configurations particularly conducive to jump events. In the case of NaAlH_4 , we present initial results of simulations of ion transport in the presence of defects, as motivated by suggestions in the literature that the presence of vacancies are key contributors to hydrogen absorption and desorption.