Hydrogen-Rich Materials in Frustrated and Confined Chemical Environments

Vitalie Stavila, PhD
Sandia National Laboratories
7011 East Avenue, Livermore, CA, 94551, USA
Email: vnstavi@sandia.gov

Abstract

Materials in frustrated and confined chemical environments display unique chemical and physical properties, with altered nanointerfaces, charge-transfer states, and reactivity. Particularly, the confinement of hydrogen-rich materials significantly influences the pathways of chemical reactions and impacts the steps that limit the reactions rates. This presentation will focus on three key areas, each illustrating the combination of innovative synthesis, computational modeling, and state-of-the-art characterization tools, all aimed at probing and comprehending the behavior of hydrogen-rich materials and molecules within frustrated and constrained chemical environments:

1) The application of machine learning to predict interstitial metal hydrides, combining hydriding and non-hydriding elements to achieve tunable thermodynamics and unusually high hydrogen-to-metal ratio (e.g. DOI: 10.1039/D3TA02323K) for hydrogen storage and superconductivity.
2) The stabilization of "molecular" metal hydrides within functionalized pores of Metal-Organic Frameworks (MOFs) and Covalent-Triazine Frameworks (CTFs), representing the ultimate level of hydride confinement (e.g. DOI: 10.1002/anie.202107507).
3) The development of surface-anchored and MOF-confined catalysts for the reversible dehydrogenation of Liquid Organic Hydrogen Carriers (LOHCs) through homocoupling reactions in polyalcohols (e.g. DOI: 10.1021/acsaem.3c00462).

These distinctive chemical environments lead to changes in thermodynamic stability, accelerate hydrogen release and uptake rates, and significantly reduce the pressure and temperature requirements for various chemical processes. Collectively, these findings mark substantial progress in our understanding of materials in frustrated and confined spaces, yet they also underscore that there is much more to discover regarding the reactivity and dynamics of small molecules and clusters within highly-functionalized environments. (DOI: 10.1038/s41557-022-01056-2).

Dr. Vitalie Stavila is a Principal Investigator in the the Hydrogen Advanced Materials Research Consortium (HyMARC, www.hymarc.org) at Sandia National Laboratories in Livermore, California, United States. His research focuses on the fundamental science and applications of inorganic and organometallic compounds. Current interests include materials-based hydrogen storage, near-ambient superconductivity, transport phenomena through nanointerfaces, and heterogeneous catalysis.