Predictive Insight On High-Density Hydrogen Storage In Metal-Organic Frameworks

Romit Chakraborty,[†] Yuto Yabuuchi,[‡,‡] Henry Jiang,[‡] Kurtis M. Carsch,[‡] Justin J. Talbot,[‡] Hiroyasu Furukawa,[‡] Jeffrey R. Long,[‡] and Martin Head-Gordon [*,¶,‡]

[†]*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley California*

[‡]*Department of Chemistry, University of California, Berkeley*

[¶]*Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley California*

E-mail: romit@berkeley.edu,mhg@cchem.berkeley.edu

Abstract

A detailed chemical understanding of H_2 interactions within the nanoporous crystalline structure of metal-organic frameworks (MOFs) is critical for the design of new sorbent materials. With state-of-the-art density functional approximations, we present predictions of H_2 adsorption equilibria for four MOFs: MOF-5, Ni_2-m-dobdc, Cu(I)MFU-4l, and V-btdd. Energy decomposition analysis offers a finer understanding H_2-binding with the delineation of the interaction energy into chemically intuitive contributions that arise from permanent electrostatics, dispersion, polarization, and charge transfer. Placing particular emphasis on multiple hydrogen ligation at open metal sites in MOFs, we show it not only has a multiplicative effect on the usable capacity of materials but also results in more effective usage of available binding sites. With predictions of feasible ligation of three equivalents of H_2 per open metal site in metal-exchanged MFU-4l and four equivalents per metal site in Ca-porphyrin, a motif that can be incorporated in MOFs, a path toward high-density hydrogen storage is envisioned through the lens of density functional simulations.