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Predictive DFT+U Methods for Small Molecule Binding in MOF-74 GREGORY MANN, Dept. of Chemistry, UC Berkeley, KYUHO LEE, Department of Chemical & Biomolecular Engineering, UC Berkeley; Molecular Foundry, Lawrence Berkeley National Lab, MATTEO COCOCCIONI, Institute of Materials, École polytechnique fédérale de Lausanne, BEREND SMIT, Departments of Chemistry and Chemical & Biomolecular Engineering, UC Berkeley, JEFFREY NEATON, Molecular Foundry, Lawrence Berkeley National Lab; Dept. of Physics, UC Berkeley — In order to use density functional theory (DFT) to reliably treat small molecule binding at open metal sites in metal-organic frameworks (MOFs), electron correlation effects associated with the localized d-states present at the metal centers must be accounted for. Incorporation of a Hubbard U-like term can be an approximate but computationally efficient means, yielding excellent agreement with experiment provided an appropriate value for the parameter U is chosen. To predict adsorption energetics for as-yet unsynthesized MOFs, we would need to select U using a systematic, physically motivated approach rather than the ad hoc methods typically employed. Here, we use an *ab initio*linear response approach to calculate U. We show that U values determined with this method reproduce previous results for the binding of carbon dioxide in Co-MOF-74 and Cu-MOF-74, and we discuss the method's application to other 3d metals in theMOF-74 framework; our preliminary results suggest that a wide range of U's above a critical value will produce accurate binding energies. Finally, we present U values calculated for Co2+ ions in other systems, probing the environment dependence of this parameter. This work supported by DOE, and computational resources provided by NERSC.

Gregory Mann
Dept. of Chemistry, UC Berkeley

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