Bridging the gap between quantum chemistry and classical simulations for CO$_2$ capture

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During the formation of some metal organic frameworks, the metal centers coordinate with both linker and solvent molecules. Subsequent activation removes the solvent molecules and creates open metal sites, which have strong affinity for CO$_2$. At present the interactions of CO$_2$ with these open metal sites are so poorly understood that commonly used force fields underestimate the adsorption of CO$_2$ in Mg-MOF-74 at carbon capture conditions by as much as two orders of magnitude. Here we present a systematic procedure to develop a force field for Mg-MOF-74 and related materials using high-level quantum chemical calculations.$^1$ This transferable force field accurately takes into account the chemistry of the open metal sites.

I will also present the results of a recent quantum chemical study on the mechanism of carbon dioxide adsorption in an alkylamine functionalized metal-organic framework.$^2$ Our calculations demonstrate that in these materials a 2 amine: 2 CO$_2$ chemistry occurs with a binding energy that is lower compared to the 2:1 chemistry that is observed in most materials. We explain this behavior in the form of a hydrogen-bonded complex, involving 2 amines and 2 CO$_2$ molecules.
