

Abstract Submitted
for the MAR11 Meeting of
The American Physical Society

First-Principles Calculations of the Role of Dispersive Interactions in CO₂ binding in metal-organic frameworks ROBERTA POLONI, University of California, Berkeley; Molecular Foundry, LBNL; University of California, Davis, JOSHUA HOWE, University of California, Berkeley, JEFFREY B. NEATON, Molecular Foundry, LBNL, GIULIA GALLI, University of California, Davis, BEREND SMIT, University of California, Berkeley and LBNL — Metal-organic frameworks (MOFs) have attracted much attention over the past 20 years for their possible applications in gas storage. In this study, we provide computational insight into what makes a MOF structure optimum for CO₂ capture. We present a density functional theory-based study of the electronic and structural properties of recently synthesized frameworks M'₃[(M₄Cl)₃(BTT)₈]₂, with M'=extraframework cation and M=Ca [1]. We study the interactions between CO₂ and different binding sites, and predict an unexpected favored binding site at the organic linker. We explore how binding energies are affected by the ordering and type of the extraframework cations. Finally, we address the role of dispersion forces by employing a recent non-local van der Waals functional [2], and compare with a DFT+D approach [3].

[1] M. Dinca et al., *J. Am. Chem. Soc.* 128, 16876 (2006)

[2] M. Dion et al., *Phys. Rev. Lett.* 92, 246401 (2004)

[3] A. Tkatchenko et al., *Phys. Rev Lett.* 102, 073005 (2009)

Roberta Poloni
University of California, Berkeley; Molecular Foundry, LBNL;
University of California, Davis

Date submitted: 30 Dec 2010

Electronic form version 1.4