

# Computational study of spin crossover MOFs for carbon capture

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Even today fossil fuels dominate the global energy production and the resulting increase of carbon dioxide emission is at the core of the greenhouse effect. To effectively prevent carbon dioxide from entering the atmosphere, metal-organic frameworks (MOFs) have been widely investigated as selective CO<sub>2</sub> adsorbents from flue gases [1]. We propose to computationally design a novel family of MOFs whose high affinity for CO<sub>2</sub> can be modified under external stimulus potentially leading to an efficient capture and release process. Recently, J.R Long et al. employed CO gas molecules to demonstrate the guest-induced cooperative spin transition mechanism in open-metal sites MOFs [2]. Our idea is rather to use temperature to trigger a spin crossover transition from low spin to high spin on the open-metal sites upon CO<sub>2</sub> uptake. Due to the occupation of antibonding molecular orbitals in the high spin state, a significant lowering of the CO<sub>2</sub> binding energy is expected [3]. Specifically, we study the well characterized Hofmann-like clathrate Fe(pz)Pt(CN)<sub>4</sub> (pz=pyrazine) and its porous extension Fe(bpac)[M(CN)<sub>4</sub>] (bpac=bis(4-pyridyl)acetylene, M=Fe,Pt,Ni). In this proof-of-concept work, we employ density functional theory to demonstrate the novelty and implications of this strategy by computing the magnetization curve and the thermodynamics of adsorption of CO<sub>2</sub> before and after the transition.

[1] Zhou H.C., Long J.R. & Yaghi O.M. *Chem. Rev.* **112**, 673-674 (2012).

[2] Long J.R. et al. *Nature* **550**, 96 (2017).

[3] Poloni R., Lee K., Berger R.F., Smit B. Neaton J.B. *J. Phys. Chem. Lett.* **5**, 861-865 (2014).