Spin-crossover molecular-based materials for efficient carbon capture and release. The role of anthropogenic CO_2 emissions as a major contributor to climate change is nowadays beyond discussion and a proposed way to mitigate these effects in the medium term is to employ carbon capture and sequestration or recycling. CO_2 capture using porous compounds relies in the selective adsorption of CO_2 from a flue gas. By increasing temperature or pressure CO_2 is desorbed and the cost of the process is related to the working capacity, i.e. the difference between the amount of adsorbed CO_2 at adsorption and desorption conditions. Here we propose to design **novel materials that can more efficiently adsorb and desorb CO_2.** We propose to design materials whose affinity for CO_2 can be modified upon a change in their electronic structure, here **spin-crossover**, allowing for a larger working capacity. Spin-crossover may occur in transition-metal complexes (d^4-d^7 electrons) where the competition between the electron pairing energy and the ligand field splitting yields two possible electronic configurations (low-spin and high-spin) that can be inter-converted upon the application of external perturbations (temperature, pressure, etc).

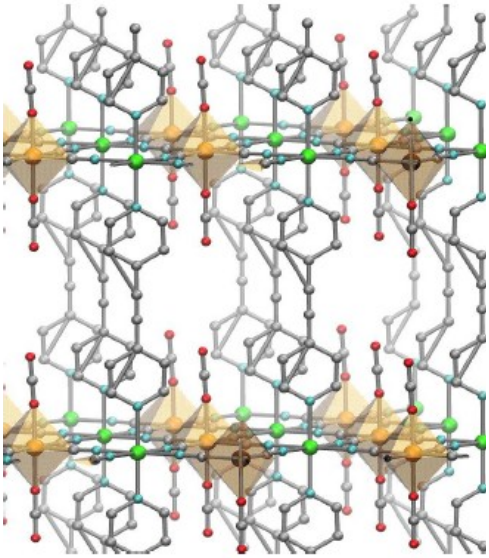

Figure 1: *Crystal structure of $M(bpac)M'(CN)_4$ with CO_2 molecules adsorbed at the apical sites of the 4-fold coordinated metal M.*

We will study several **Hofmann-type clathrates** (see Figure 1 for an example) exhibiting promising features for an efficient carbon capture and release, e.g open-metal sites, porosity occurrence of spin transition with an abrupt transition and small hysteresis, and a transition temperature close to room temperature.

The PhD candidate will combine calculations (SIMaP) and experiments (ILL). The mechanism of CO_2 binding will be studied by neutron diffraction using the instrument suite of the diffraction group at the ILL, and by neutron vibrational spectroscopy and QENS. Samples synthesis and characterization will be done at IMDEA, Madrid. Density-functional theory and the GW approach will be employed to study the electronic structure of these metal-organic frameworks and to characterize the spin transition of a few selected candidates.