

**MS40-1-3 Insights on Selective Gas Sorption Behavior in a Family of MOF-like Materials from Operando Crystallographic Studies with DFT Calculations**  
**#MS40-1-3**

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**Abstract**

Design and development of new solid gas-sorbent materials, having adjustable structures to accommodate high densities of adsorbed gas solvent molecules, continues to be of major interest. This is driven by universal needs for molecular sorption and gas detection, enhanced oil and gas recovery, various gas storage and separation applications, as well as the need for increasingly efficient, inexpensive gas sorbents to address climate change through carbon dioxide reduction (CDR) [1]. In this context, metal organic frameworks (MOFs) and similar materials show promise. These comprise metal ions or clusters linked into highly porous 3D networks by coordinated organic ligands. Their behaviour can include an ability to exhibit hysteresis in reversible structural transitions between low- and high-porosity states during adsorption/desorption cycles. One group of microporous compounds possessing such MOF-like structures is the pillared cyanonickelate ("PICNIC") family of Hofmann compounds, based on square planar sheets of Ni(CN)<sub>2</sub> bridged by organic ligands terminated by N atoms at each end [2]. Since the network is not inherently rigid against shearing of Ni(CN)<sub>2</sub> planes against to each other, some PICNIC materials are flexible, although not all. Experimental results based on X-ray and neutron diffraction (XRD and ND), as well as small-angle scattering (SAXS and SANS) will be presented for several generic members of the PICNIC family, including operando studies under dual gas flow conditions (CO<sub>2</sub> with N<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>) and supercritical CO<sub>2</sub> conditions [3-5]. Significant insights have been gained regarding the different structural and microstructural gas sorption responses for different members of the PICNIC family by comparing these experimental results to the predictions of density functional theory (DFT) calculations [6].

**References**

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