Designing Metal-Organic Frameworks for Efficient Gas Adsorption Using Force Field-Based Molecular Simulations

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Metal-Organic Frameworks (MOFs) are promising materials for addressing critical environmental and health challenges, such as clean water access and sustainable energy, due to their high porosity, tunable chemistry, and structural diversity. Despite extensive research, their large-scale application remains limited by challenges in controlling structure–property relationships from the molecular to macroscopic level.

In this talk I will focus on the development and modelling of new nano-scale MOF materials based on the structures of zinc- triazolate and zinc- triazolate- oxalate complexes for several high-need applications such as gas storage and separations. To achieve this, computational methods (combining force field based grand canonical Monte Carlo (GCMC) simulations and density functional theory (DFT) calculations) have been used to improve structure-property predictions and to calculate the adsorption isotherms of investigated gases with and without the presence of water. The crystal structures of α-CALF-20 and its polymorph derivative were fully geometry-optimized (atomic positions and cell parameters relaxed) at the periodic DFT level without imposing any constraints in terms of topology/geometry. Force-field parameters and derived atomic partial charges of the MOFs have been validated by an excellent agreement between the CO_2 and N_2 adsorption isotherm simulated for α-CALF-20 with the corresponding available experimental data in the pressure domain of 0–1 bar. In addition, we have calculated gas adsorption isotherms for β-CALF-20. We observed that investigated MOFs show good adsorption performance for CO_2 capture.

In addition, the zinc triazolate framework such as MAF-66 shows strong adsorption performance for SO_2 and H_2S gases. The research also shows that modifying this MOF into ZTF MOF enhances its capture abilities for these gases The nature of the interactions between investigated gases with the pore surface cavities was examined at the microscopic level. More details can be found in Ref. 1-4. The GCMC simulations were performed using RASPA code on PARADOX IV supercomputing facilities [5].

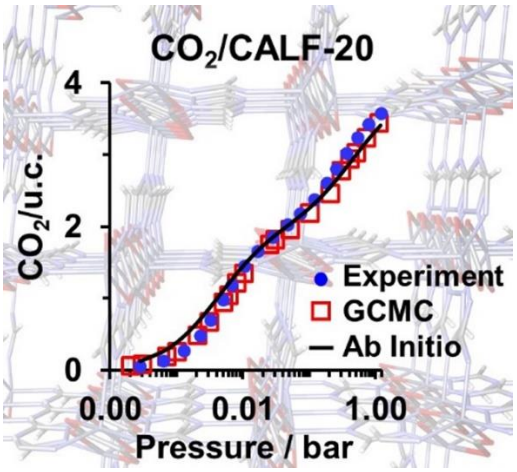

Figure 1 – *Chemically Accurate Prediction of CO_2 Adsorption Thermodynamics in Metal-Organic Framework CALF-20*

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[5] PARADOX IV supercomputing facility at the Scientific Computing Laboratory of the Institute of Physics Belgrade.