The steady rising level of atmospheric carbon dioxide resulting from anthropogenic emissions is now widely accepted as the main contributor to the worsening of the global climate change. These emissions are projected to continue to increase in the future due to the economy that is heavily relied on fossil fuels. Although the transition of the existing carbon-based infrastructure to cleaner alternatives or renewable energy would be ideal to curb the CO$_2$ emission, such a change requires drastic modifications to the current energy infrastructure. Thus, carbon capture and sequestration technologies that efficiently capture CO$_2$ from existing emission sources will play a vital role. Fossil fuel power plants are attractive candidates for a possible CO$_2$ capture regulation. Unfortunately, the current CO$_2$ capture technologies using amine solvent are energy intensive, expensive, and environmentally unfriendly. Therefore, development of new, advanced adsorbent materials for CO$_2$ capture presents a possible solution to this problem. A special class of metal-organic framework adsorbent named elastic layered metal-organic framework (or ELM) with step-like CO$_2$ adsorption isotherm offers a clear advantage in CO$_2$ capture application compared with other adsorbents with Langmuir-like adsorption isotherm.

Molecular simulations and experiments were therefore carried out to study the adsorption phenomena observed for ELM adsorbents and evaluate the potential application for CO$_2$ capture at conditions relevant to flue gas in coal-fired and syngas in integrated gasification combined cycle power plants. Simulation for the first time has provided atomistic insights into the mechanism leading to the expansion of the material and CO$_2$ isosteric heat of adsorption profiles. The adsorption induced expansion was shown to depend on the CO$_2$ orientation transition and the interplay of the CO$_2$-framework configuration to maximize CO$_2$-CO$_2$ interaction energy. Predicted CO$_2$ selectivity is higher than those typically observed with other microporous materials that have been proposed for use as carbon capture adsorbents. Ideal adsorbed solution theory (IAST) estimates of CO$_2$ selectivity are generally comparable to that of molecular simulation.

Finally, structure-function relationships in this series of ELM adsorbent will be discussed to accelerate the targeted synthesis of new ELM material and aid the development of reverse engineering an ELM structure for CO$_2$ capture application.