Simulation of Xe Adsorption into UiO-66 Crystal by Grand Canonical Monte Carlo Method

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Abstract

In nuclear power plants, Xe/Kr gases generated from the fission of nuclear materials can be leaked during high level waste disposal or due to physical failures in fuel. Therefore, selective adsorption technology of radioactive gases is important for accident prevention or waste management.

Because of high cost of Xe gas, it is not financially effective to test many list of MOFs and select model with better results. Therefore, it is very cost effective and saving time by filtering the models by computer simulation and verify its performance by actual experiments.

In this study, The MOF-66 model was simulated and experimented with N2 gas to check the similarity between simulation and experiment. After than, another simulation tried to predict adsorption amount of Xe gas.

Grand Canonical Monte Carlo (GCMC) Simulation

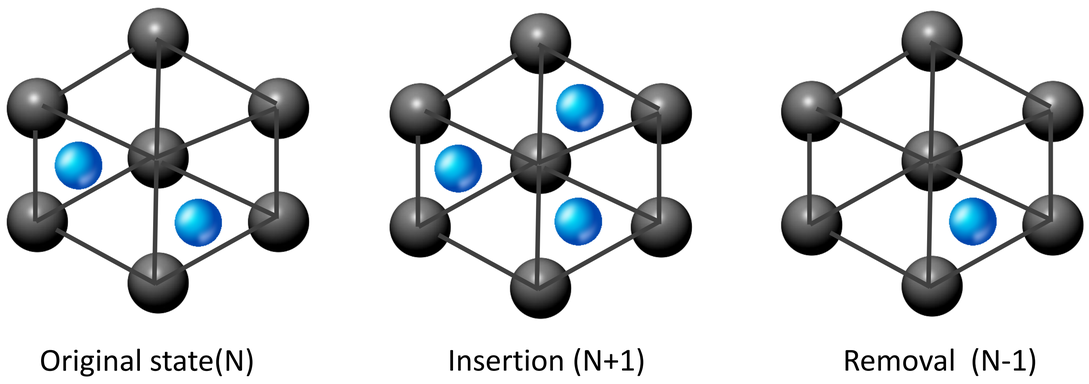

Fig1. Stages of GCMC simulation in MOFs

Grand Canonical Monte Carlo(GCMC) simulations have probabilities for the addition, removal, and movement of particles in given system. And then, use those probabilities to determine behaviors of the particle through Monte Carlo simulations.

Each Probability can be calculated as following, while $\Lambda = \sqrt{\frac{h^2}{2\pi m k_b T}}$ and $\beta = \frac{1}{k_b T}$ [1].

$$\text{Insertion} : P(N \to N+1) = \min\left[1, \frac{V}{\Lambda^3 (N+1)} e^{\beta(\mu - \Delta U)}\right]$$

$$\text{Removal} : P(N \to N-1) = \min[1, \frac{\Lambda^3 N}{V} e^{-\beta(\mu - \Delta U)}]$$

ΔU is calculated by sum of Lennard-Jones potential, which is the interaction energy between inserting particle and other particles already exist in system. And μ can be calculated by partial pressure of target gas with Gibbs free energy.

$$U_{ij} = 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]$$

$$\mu = \mu^0 + k_b T \ln\left(\frac{p}{p_0}\right)$$

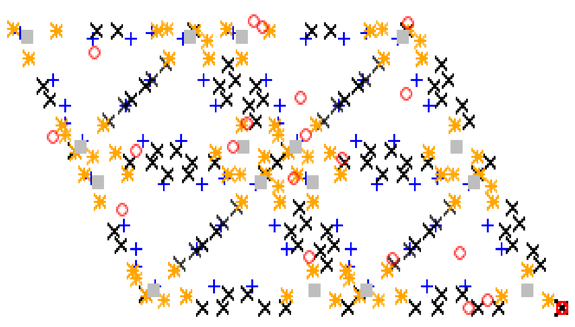

Fig2. 2D example plot of UiO-66 simulation structure.

The simulation model is consisted by UiO-66 MOF structure. This model is consisted of 8 unit cells.

The gas target particles are take place in the MOF simulation system with red colored dots in the Figure 2.

Result and Conclusion

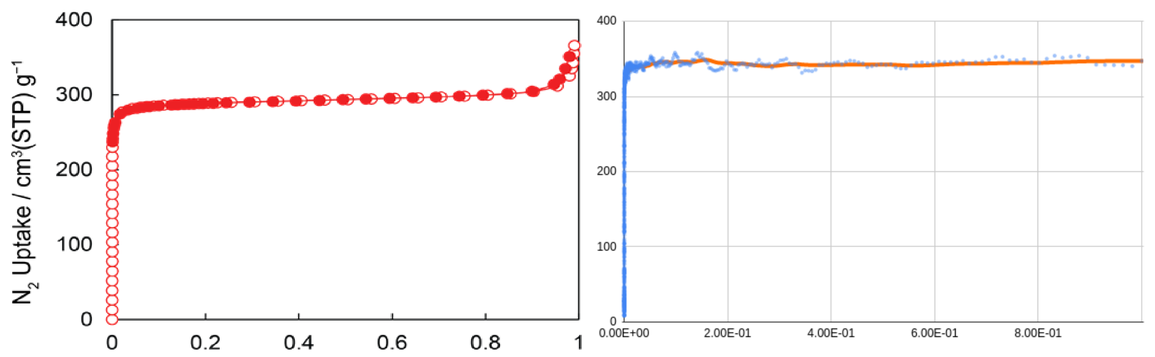

Fig3. Comparison between experimental result and simulation result of N2 gas uptake

From the experimental result, intake of N2 gas increases sharply at a very small partial pressure ratio, then stays almost constant level, and have another sharp increase when the partial pressure ratio approaches at 1.

At simulation result, it is same when very small partial pressure and constant level of graph. But it didn't find clear increase when approaching partial pressure ratio at 1.

With the logscale of x axis of partial pressure axis, there are partial pressure range of increasement of gas uptake. And this range is expanded as temperature increases.

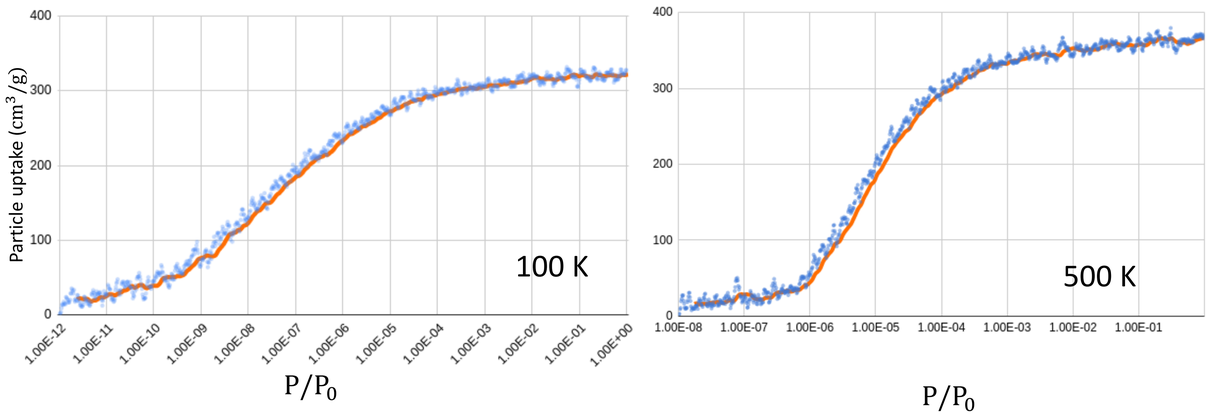

Fig4. Increasement range changes depend on temperatures, with logscale.

The final objective of this study is to develop materials with high-selective of Xe/Kr gas. With GCMC simulation of N2 gas absorption , it shows consistency with experimental result in Fig 3. similarity in range of partial pressure without around 1. Therefore, simulation result with Xe's physical parameter can provide prediction of Xe gas adsorption except partial pressure range around 1.

In conclusion, the GCMC simulation code made consistence result with N2 gas. The simulation gave considerable prediction except partial pressure around 1. Applying parameters of Xe gas to this code, the Xe absorption simulation is predicted. Since we don't have experimental result of adsorption of Xe gas in UiO-66, we will conduct experiment to confirm adsorption performance of MOF and compare simulation results

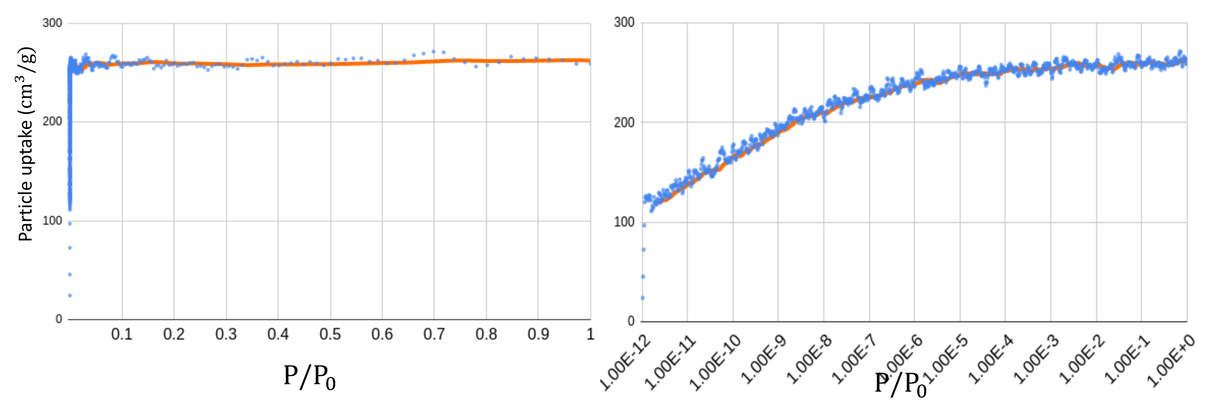

Fig5. Simulation results of Xe adsorption in 300 K

Reference

[1] Y.R. Tao , G.H. Zhang , H.J. Xu (2022) Grand canonical Monte Carlo (GCMC) study on adsorption performance of metal organic frameworks (MOFs) for carbon capture

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