Gibbs Adsorption Simulator

A tool to calculate the adsorption of a gas molecule onto a metal organic framework using Gibbs ensemble

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This tool uses RASPA for its backend simulation. RASPA is a molecular simulation software for performing adsorption and diffusion calculations in nanoporous materials. It implements state-of-art algorithms for Molecular Dynamics and Monte Carlo for various ensembles. In this simulation, we used Monte Carlo with the Gibbs ensemble.

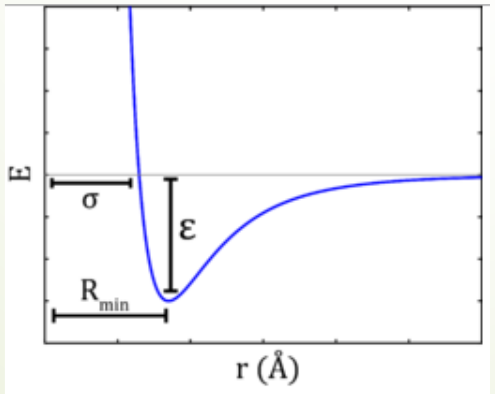
Every simulation is performed with initialization and production cycles of 5000 and 10000 respectively.

- Initialization cycles warm up the simulation. The data is collected during production cycles.

- Transferrable Potential for Phase Equilibria (TraPPE) forcefield is used to define the molecular interactions for helium, nitrogen, hydrogen, oxygen, carbon dioxide and methane.

Mol	Def	Reference
CH_4	TraPPE	*J. Phys. Chem. B,* 1998, 102,2569-2577
He	TraPPE	*J. Am. Chem. Soc.,*1992, 114, 10024-10035
H_2	TraPPE	Vapor-Liquid Equilibria of Mixtures Containing Alkanes, Carbon Dioxide, and Nitrogen; AIChEJ., 2001, 47, 1676-1682
N_2	TraPPE	
CO_2	TraPPE	

Lennard-Jones Potential



en.wikibooks.org/wiki/Molecular_Simulation/The_Lennard-Jones_Potential

- For the MOFs, forcefields parameterized by the developers of RASPA were used.

Zhiwei Q., Nanyi W., Jianwen ., Jian Z. Design of amine functionalized metal-organic frameworks for CO2 separation, *Chem. Commun.*, 2016,52, 974-977

Dubbledam D., Walton K. S., Ellis D. E., Snurr R. Q.,Exceptional Negative Thermal Expansion in IsoreticularMetal–Organic Frameworks, Angew. Chem. Int. Ed. 2007, 46, 4496 –4499

MOF	Flexibility	Void Fraction (%)	Forcefield
IRMOF-1	Rigid	82.62	Dubbledam2007FlexibleIRMOF-1
IRMOF-16	Rigid	92.65	Dubbledam2007FlexibleIRMOF-16

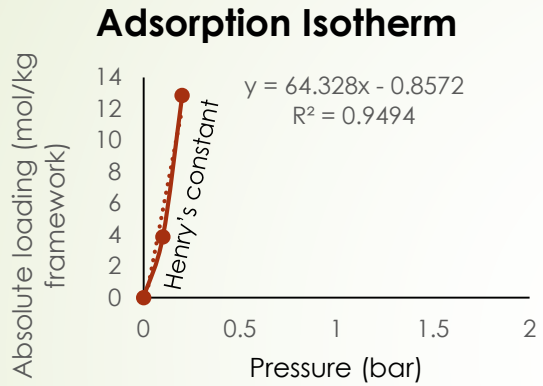
- Lorentz-Berthelot's model was used as the general mixing rule for the Lennard-Jones potentials

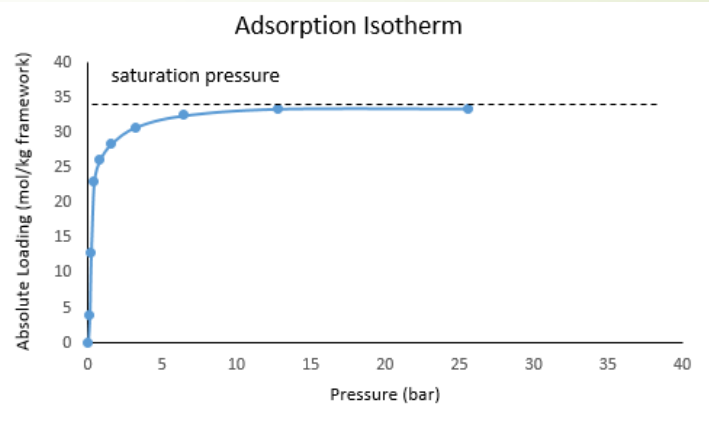
$$\sigma_{ab} = \frac{\sigma_a + \sigma_b}{2} \qquad \varepsilon_{ab} = \sqrt{\varepsilon_a \varepsilon_b}$$

- The tool outputs the absolute adsorption which is given in mol/kg framework. Absolute loading is the total number of molecule adsorbed onto the material.
- The main difference between Gibbs adsorption and the adsorption using grand canonical ensemble (μVT) is that Gibbs adsorption uses two boxes for the simulation, the framework is contained in one box and the gas molecule is contained in the other box. The simulation computes the adsorption using the forcefield and not a fugacity coefficient or an equation of state. The volume of the gas box changes but the one for the framework remains constant.

Calculation of Adsorption Isotherm

- The adsorption isotherm is shown by plotting a graph of the absolute adsorption against pressure. The simulation would need to be run a number of times, and each time at the same temperature and different pressure.





The graph of the pressures against the absolute adsorption is used to calculate Henry's constant which is given by the slope of the graph at the lower pressures (left panel). The saturation loading is the maximum amount adsorbed at high pressure (right panel).

The simulation was carried out using IRMOF-1 and methane at a temperature of 150 K and different pressures.