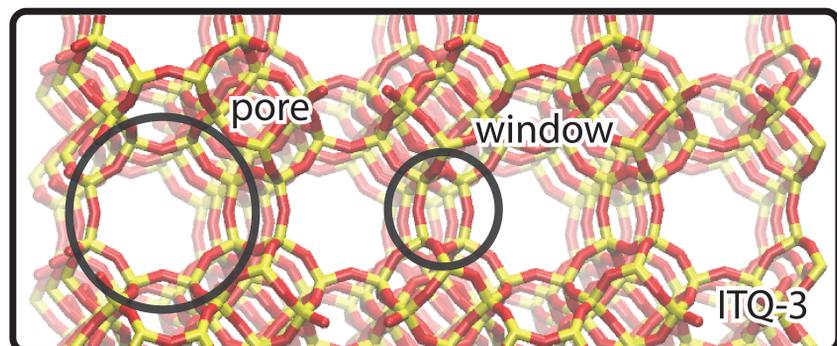


Introduction

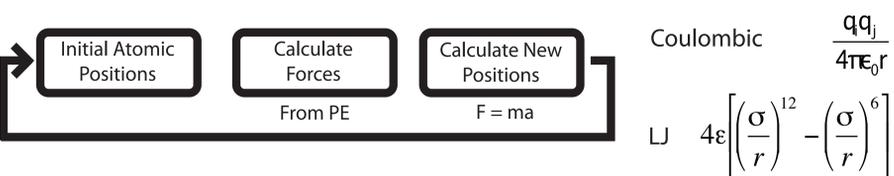
Zeolites are crystalline structures made of oxygen, silicon, and sometimes aluminium (or other metallic cations). The zeolite framework has features called pores, channels, and windows. In these channels smaller molecules (such as CO₂ or N₂) can interact with the zeolite walls and be adsorbed.



My research involves studying how gaseous molecules are adsorbed, explore, and interact with the walls of the zeolite and the other adsorbed molecules. Specifically I will be studying the effects of pore loading and the molar fraction of N₂ to CO₂ in ITQ-3 on the diffusion coefficients of the adsorbed species.

Simulation of Motion

A molecular dynamics (MD) simulation is used to determine the positions and energies of atoms over time. This type of simulation uses Newton's equations of motion iteratively over a short timestep (a femtosecond) to determine the position of each atom. Forces are calculated based on Lennard-Jones (LJ) and coulombic potential energy equations.



Analyzing Motion

By saving the positions of molecules at the end of each timestep, calculations can be performed on their movement. We can get diffusion coefficients which are quantitative descriptions of particle movement. Self Diffusion (Ds) is the area explored by a molecule in a period of time.

$$D_s(c) = \lim_{t \rightarrow \infty} \frac{1}{6t} \left\langle \frac{1}{N} \sum_{i=1}^N \left\| \vec{r}_i(t) - \vec{r}_i(0) \right\|^2 \right\rangle$$

Acknowledgements

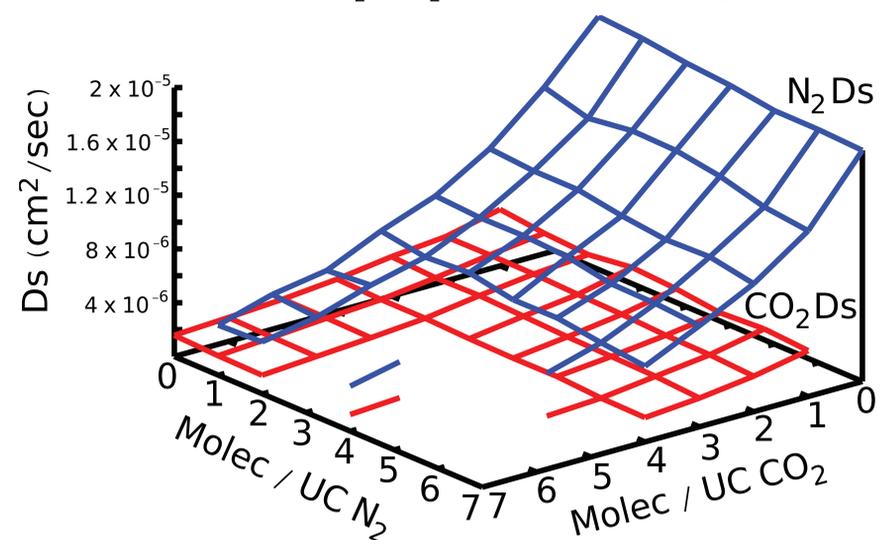
Thanks to Felix Amankona-Diawuo, Disan Davis, and Jayme Dahlin for their intellectual support and HHMI, NSF, and Carleton College for funding.

Analysis of Diffusion and Potential Energies of Carbon Dioxide and Nitrogen in Zeolites

David Selassie and Dani Kohen
Carleton College

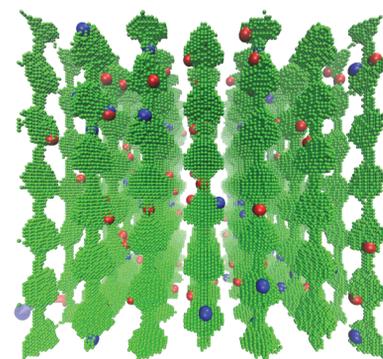
Molar Fraction Effects

By running MD simulations with varying CO₂ and N₂ concentrations, we have discovered that only CO₂ concentration has a significant effect on either CO₂ or N₂ diffusion rates in ITQ-3.



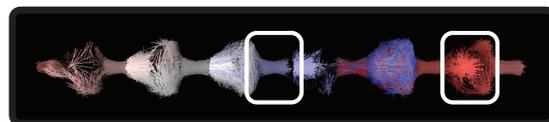
Examining Favored Sites

Previously, we had run MD simulations and recorded the frequency at which molecules visited each position in the UC and determined the most favored sites in the UC.



Green spheres show space available for adsorbate molecules for reference. Red spheres show the most visited sites for CO₂ and blue spheres show the most visited sites for N₂.

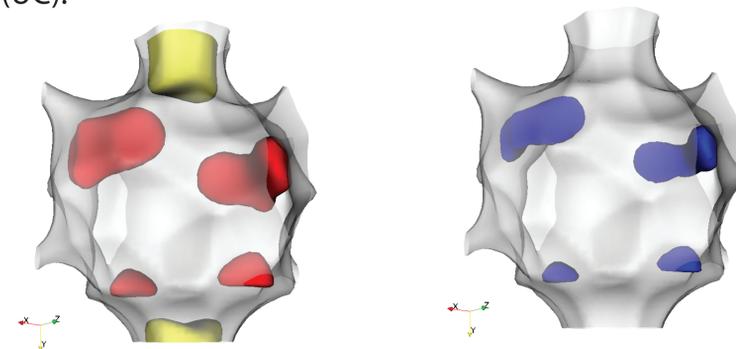
In order to better understand diffusion, we ran extended simulations with only one molecule of CO₂ and displayed its position over time:



We noticed that the CO₂ molecule spent an unexpectedly large amount of time in the narrow windows that connect ITQ-3's spherical pores and also that when the CO₂ molecule was in the pores, it did not spend an equal amount of time on all surfaces of the pore.

Potential Energy Well Comparison

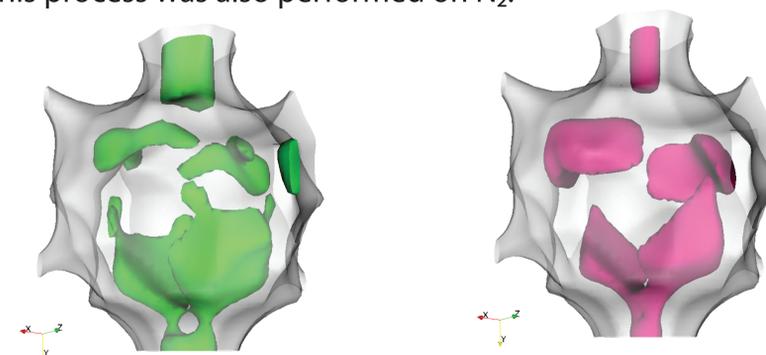
In order to investigate the preference of CO₂ to certain sites in ITQ-3's unit cell, we visualized the PE interactions that carbon and oxygen atoms in CO₂ would have with the zeolite framework if they were placed at every position in the unit cell (UC).



Within the white volume are favorable carbon sites (as a reference to ITQ-3's pores). Within the yellow volumes are the most favorable carbon sites, within the red volumes are the most favorable oxygen sites, and within the blue volumes are the most favorable nitrogen atom sites.

Rotational Configurations

In order to find the most favorable position for the whole CO₂ molecule, we placed a molecule of CO₂ at every position in the unit cell then recorded the PE for 1000 random rotational configurations at that position which provides the average and minimum PE for a CO₂ molecule at that position. This process was also performed on N₂.



Within the green volumes are the favorable minimum configuration (PE < -3 x 10³ K) sites for the center of mass of a CO₂ molecule. Within the pink volumes are the most favorable minimum configuration sites (PE < -1.5 x 10³ K) for the center of mass of an N₂ molecule.

There is a strong correlation between the depth of the PE wells for CO₂ and the amount of time that a typical CO₂ molecule spends in that place during our favored sites simulations. Surprisingly the deepest PE wells are in the y and z-axis windows which were expected to be repulsive due to sterics. N₂ has similar PE wells except that the depth of these wells is only half of the CO₂ wells. ITQ-3's selectivity towards CO₂ is attributed only to the increased depth of the CO₂ potential energy wells.