Because of the threat of global warming, it is important that new means are established to deal with the increased production of greenhouse gases, such as carbon dioxide (CO₂). If a material that can be used to separate CO₂ from other gases is found, CO₂ can be removed at the source and stored elsewhere. Zeolites are one promising type of material to perform this task.

Zeolites are made of silicon, oxygen, and sometimes aluminum, assembled in a crystalline structure. This structure has pores inside which work well for trapping small molecules (such as CO₂). When these molecules become trapped on the surfaces of the zeolite, it is called adsorption. To quantitatively describe adsorption, we use graphs called isotherms:

From isotherms like this, it is clear that some zeolites preferentially adsorb CO₂ to N₂, both separately and as a mixture. This analysis also seems to apply when considering the varied adsorption of CO₂ on different zeolite structures. From the preliminary data that I have collected, there is a clear trend in the self-diffusion of CO₂ on both materials.

In addition to looking at the overall materials, I have examined the self-diffusion trends varying in the x, y, and z directions of both materials, relating back to channel size and the various channel types mentioned in the background.

The largest pores in silicate are in the x and y-directions and are associated with the higher diffusion values. For ITQ-3, the y and z-directions have the largest pores, but only the only the y-direction has high diffusion because the z channels are barely larger than the CO₂ molecules themselves.

I also hope to study corrected/transport diffusion but currently do not have enough runs to converge to accurate results. I am also beginning to extend this research to look at the third material, ITQ-7, to further extend our understanding of the dynamics involved in CO₂ adsorption in zeolites.

**Background**

**Introduction**

Dani’s group has been studying zeolites using computer simulation. We want to discover what makes some zeolites such great adsorbers of CO₂. Work started by comparing the adsorption of CO₂ on three similar zeolites. My goal is to study the way that gas molecules move through the materials as a means of verifying that our equilibrium description is accurate and to learn more about their behavior.

**Simulation Dynamics**

**Method**

Molecular Dynamics (MD) is a computational method that simulates the motions of particles over time. We use MD to record the trajectories of adsorbates through zeolite crystals in hopes of better understanding how particle movement affects adsorption. Using trajectories recorded from MD, we can calculate measurements in a few ways:

- **Self-diffusion (Dᵢ)** is the area that one particle explores in a given amount of time:
  \[
  Dᵢ(t) = \lim_{t \to \infty} \frac{1}{t} \sum_{i=1}^{N} \left( \frac{1}{t} \sum_{j=1}^{t} \left( \mathbf{r}(t) - \mathbf{r}(0) \right) \right)
  \]

- **Transport diffusion**, is a property of the group of particles, and it is the proportionality constant between the flux and the concentration gradient. Transport diffusion can be calculated computationally using corrected diffusion (Dᵢ), which is found using the average of many individual trajectories.

**Validation**

The gray blob represents the places that the CH₄ has explored over time, and the red and gold framework is the zeolite silica. To know that our equilibrium results are accurate, we can visually verify the movement of particles through the zeolite as well as compare calculated diffusion values to previously collected data.

**Preliminary Dynamics Results**

We can compare the diffusion of CH₄ to that of CO₂ using these diffusion coefficients and we can see how that relates to adsorption by comparing the isotherms for CH₄ and CO₂. The diffusion of CO₂ is slower than that of CH₄ because of greater interactions between CO₂ and the zeolite. Greater interactions with the zeolite cause higher adsorption, and thus CO₂’s slower diffusion correlates with higher adsorption. However, we can also see that the diffusion of CO₂ is significant enough to suggest that the molecules are moving well within the pores, which gives us confidence in our equilibrium data.

**Acknowledgements**

Thank you to Meghan Thurlow and Eric Feise for their guidance and assistance. Thank you to Carleton College for funding and resources.