Do Coulombic Interactions Dominate the Rate of Diffusion of CO₂ and N₂ Within All-Silica Zeolites?

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Introduction

The level of CO₂ in our atmosphere has been increasing in recent years and has been linked to Global warming. As the level of CO₂ keeps increasing it is becoming more important to reduce the amount of CO₂ in our atmosphere. One way of doing this is by separating it from other gases in the atmosphere then sequestering it somewhere safe. One set of materials that are being studied to be used in the separation process are called zeolites. Zeolites are crystalline materials composed of oxygen, silicon, and sometimes aluminum (or other metallic cations). These materials have pores and channels that allow smaller molecules, such as CO₂ and N₂ to interact with the zeolite framework and be adsorbed. The Kohen group has been studying three different Zeolites: silicalite, itq3 and itq7, all of which have the chemical composition of SiO₂.

Methods and Models Continued

We used Grand Canonical Monte Carlo (GCMC) and Molecular Dynamics (MD) to equilibrate and run our simulations. GCMC is an action based algorithm that uses random numbers to let the system arrive to thermal equilibrium. In GCMC the molecules have four different "action" types, rotation, translation, insertion and deletion. During the simulation a random molecule's configuration is selected. The computer then performs a randomly chosen "action". The computer uses the potential energies at the initial and final position along with the Boltzmann equation to determine if the "action" is accepted.

Adsorption Simulation

The adsorption of N₂ and CO₂ gas in the zeolites was examined to check how well the zeolites were at selectively adsorbing CO₂. Previous adsorption calculations have shown that all three materials selectively adsorb CO₂ over N₂. Adsorption of N₂ is similar for all three materials, but CO₂ adsorption shows differences between the three materials. ITQ adsorbs the most CO₂ which is a feature of its unique structure compared to the other two zeolites studied. A previous study was done to investigate the origin of the differences between the N₂ and CO₂ adsorption and it removed the coulombic interactions from the simulation. The removal of the coulombic interactions had a large negative effect on the amount of CO₂ adsorbed in the three materials. This means that the coulombic interactions played an important role in allowing the zeolites to adsorb CO₂ molecules.

Diffusion Simulation

It is necessary to examine the diffusion of the adsorbates inside the adsorbents to be able to better understand the CO₂. If the molecules did not diffuse quickly enough it would not be practical to use zeolites as filters. With the data from MD, it is possible to track the movement of the adsorbates over the course of the simulation. From this we can tell how far the adsorbates are moving in a time interval and then determine how fast the molecules diffuse. The self diffusion coefficient is the area that a molecule explores in a given time. We calculate the molecules self diffusion coefficient using Einstein's equation of diffusion. If the movement of the molecule is truly random then the diffusion coefficient should not change over time. The three zeolites studied have large enough diffusion coefficients to make them practical for separation and storage of CO₂. The diffusion for the two gases decreased as the number of molecules in the zeolite was increased. This is because of the larger step strain on the molecules as it becomes more crowded inside of the zeolite.

Future Plans

To finish collecting data for both CO₂ and N₂ gas diffusion without coulombic charges in all three zeolite materials. With the diffusion data I have collected I would like to use the probability for each position in the cell for the molecules. Also, I hope to collect diffusion data for mixed gas simulations.

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