INTRODUCTION

Due to the connection between increasing atmospheric levels of the greenhouse gas CO₂ and global warming, it is of the utmost importance that new technologies for sequestration of CO₂ are developed. The separation of CO₂ from a mixture of gaseous species requires a filter that strongly selects CO₂ relative to other non-hazardous atmospheric gases, such as N₂. Zeolite crystals act as molecular sponges that soak up CO₂ in a network of “pores” during a process called adsorption. Zeolites are crystals that are composed of aluminosilicates in a wide variety of crystalline structures (Figure 1). In order to quantitatively describe the adsorption process, adsorption isotherms are used (Figure 2).

Behavior of CO₂ within Zeolites I:
Aluminum Substituted Zeolites
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BACKGROUND RESEARCH

Changing the Al/Si ratio within a zeolite crystal has been shown to alter adsorption characteristics of the zeolite (1). We are investigating the impact of Al substitution in the three zeolites shown in Figure 1. Computational techniques allow for observation of adsorption processes on an atomic scale, which may be highly useful to engineers seeking to synthesize zeolites for CO₂ sequestration. Adsorption characteristics shown in Figure 2 were obtained using Grand Canonical Monte Carlo (GCMC) simulations. GCMC simulates equilibrium and makes use of random number generation and the Boltzmann distribution to test the most probable locations of a CO₂ molecule in the zeolite framework. Results show that ITQ-3 is best at adsorbing CO₂. However, results from “fake” GCMC simulations, where all coulombic interactions are absent, indicate that the adsorption ability of ITQ-3 relies on coulombic interactions (Figure 3).

ALUMINUM SUBSTITUTED ZEOLITES CONTINUED...

We wrote a new function in the unmodified code that calculates PE directly based on the LJ potential and the position of a CO₂ molecule relative to the zeolite crystal (Figures 5 and 6). It is also necessary to modify the code to remove interpolation of forces. Forces are used in Molecular Dynamics (MD) simulations and are calculated using classical mechanics. Similar to potential energy, pre-calculated force values are stored with corresponding grid locations. Preliminary results from modified code that calculates forces directly based on the position of a CO₂ molecule relative to the zeolite crystal are shown in Figure 7. The relationship between the interpolated and directly calculated forces is encouraging; however, these data await rigorous evaluation to confirm their validity.

FUTURE RESEARCH

Now that we have produced a modified code that bypasses the PE interpolation scheme, we will proceed with the following research goals:

- Validate interpolation bypass in forces
- Create modified xyz input files
- Choose Al positions for Silicalite and create a separate code that will replace some Si by Al

REFERENCES


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