Abstract Submitted
for the 4CF09 Meeting of
The American Physical Society

Reactive Inorganic Membranes for CO_2/N_2 separations: Ab-initio Density Functional Theory Calculations[1] M. OSTWAL, J.D. WAY, M. LUSK, Colorado School of Mines — The selectivity (CO_2/N_2) of mesoporous silica membranes can be enhanced by surface modification using APTS (3-aminopropyl-triethoxy silane). The hypothesized transport mechanism in such materialsis the reaction of CO_2 with surface amine groups to form a carbamate species and subsequent surface "hopping" of CO_2. DFT calculations were performed in order to elucidate the mechanism of CO_2 transport in APTS modified membranes, to compute the CO_2 diffusivity through the membrane, and to calculate its binding energy on an amine strand. The computed binding energy for docking one CO_2 molecule to an amine was calculated to be 15.5 kcal/mol (0.67 eV). The activation/barrier energy for a CO_2 molecule to hop from one amine strand (in form of carbamate) to another computed using Transition State Theory (TST) was 7.2 kcal/mol (0.31 eV) and compares well with our experimental data ($\sim$ 8kcal/mol; 0.35 eV). In the configuration studied, CO_2 hops from one strand to another in a zigzag fashion due to thermal motion of the strands; a strand with the CO_2 molecule undulates and eventually moves so that the CO_2 can be attracted by an adjacent strand. The CO_2 diffusivity calculated using the computed activation energy ranged from 1.1 X $10^{-11}m^2$/sec (@ 25 C) to 5.7 X $10^{-10}m^2$/sec (@100 C).

[1]We gratefully acknowledge the financial support from Department of Energy Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences and Biosciences division under Grant DE-FG03-93ER14363.

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Date submitted: 29 Sep 2009 Electronic form version 1.4