192f Computer Simulations of Adsorption and Transport of a Quaternary Mixture Including Hydrogen in Zsm-5 and Silicalite

Martha C . Mitchell, Marco Gallo-Estrada, Venkata Krishna K. Upadhyayula, and Tina M. Nenoff

Inorganic molecular sieve materials such as zeolites can be fabricated into thin film membranes. These membranes are effective for separating H_2 from CO_2,CO, and CH_4at high temperatures,major product gases of natural gas combustion. Current hydrogen separation membranes based on organic materials and have functional groups that destabilize at high temperatures. The destabilization process severely affects the structural integrity of these membranes. In contrast, zeolites are crystalline framework materials with a thermally stable inorganic matrix that allow them to operate effectively at high temperatures. Furthermore these materials have crystallographically well defined that can be tuned to the molecular dimensions of light gases and are capable of size exclusion separations, a distinctive feature not found in organic membranes. ZSM-5, and its siliceous analog, silicalite, have been identified as potential candidates for the separation of light gases for industrial applications, including separating hydrogen from the product gases of steam reformation of natural gas.

Computer modeling is an essential part of the directed design of zeolite membranes. In this work we evaluated the adsorption and transport behavior of light gases in ZSM-5 and silicalite. We have performed Molecular Dynamics (MD) and grand canonical Monte Carlo (GCMC) simulations in silicalite and ZSM-5 of a quaternary mixture that mimics the product of steam reformation. Initially, MD simulations were conducted with the same bulk composition of 76.2% H_2, 3.4% CH_4, 6.8% CO and 13.6% CO_2 used by Nenoff et al. (Sandia National Laboratories) in their experimental studies. GCMC simulations were performed in both silicalite and ZSM-5 to determine the pore composition of these gases. MD simulations were performed again based on the pore composition to evaluate the transport behavior of these gases in pores of ZSM-5 and silicalite and the self diffusivity coefficients were obtained.