TITLE: CO_2, Separation Using Zeolite Membranes

PIs: Richard D. Noble
John L. Falconer

STUDENTS: Joseph C. Poshusta, Christopher J. Gump, Tracy Q. Gardner, Ana Flores, Joseph Tamburini, Megan Fortado

INSTITUTION: University of Colorado
Department of Chemical Engineering
Campus Box 424
Boulder, Colorado 80309
(303) 492-6100 phone
(303) 492-4341 fax
nobler@colorado.edu

GRANT NO.: DE-FG2698-FT40123

PERIOD OF PERFORMANCE: July 1, 1998 – June 30, 2001 Date: June, 2000

ABSTRACT

Zeolite MFI and SAPO-34 membranes were prepared on the inside surface of porous alumina tubes by hydrothermal synthesis, and single gas and binary mixture permeances were measured to characterize the membranes' performance. A mathematical diffusion model was developed to determine the relative quantities of zeolite and non-zeolite pores in different membranes by modeling the permeation data of CO_2. This model expresses the total flux through the membrane as the sum of surface diffusion through zeolite pores and viscous flow and Knudsen diffusion through non-zeolite pores. As predicted by the model, the permeance of CO_2 decreased with increasing pressure at constant pressure drop for membranes with few non-zeolite pores, but the permeance increased through viscous flow pores and was constant through pores allowing Knudsen diffusion. Membranes having more non-zeolite pores had lower CO_2/CH_4 selectivities.

The SAPO-34 membranes were characterized for light gas separation applications, and the separation mechanisms were identified. Light gas mixtures with significant differences in size (H_2/CH_4, H_2/N_2) were separated by differences in diffusivity, and the selectivities were relatively unaffected by temperature. Similar sized molecules were separated by competitive adsorption, which decreased with temperature, and the highest CO_2/CH_4 selectivity was 80 at 270 K. For a mixture where the larger molecule was more strongly adsorbing (CO_2/H_2) the competitive adsorption and differences in diffusivity mechanisms combined competitively, and the selectivity was low. In a mixture where the smallest molecule was also the more strongly adsorbing one (CO_2/CH_4), the mechanisms combined cooperatively, and the selectivity was high.

The effects of humidity on gas permeation were studied with SAPO-34 membranes of different qualities. Membranes with high CO_2/CH_4 selectivities (greater than 20) were stable in water vapor under controlled conditions, but degradation was seen for some membranes. The degradation opened non-SAPO-34 pores that were larger than SAPO-34 pores as shown by the i-C_4H_{10} permeance, CO_2/CH_4 selectivity, and CO_2 flux pressure dependence. In SAPO-34 pores, water adsorbed strongly and appeared to completely block the pores. In non-SAPO-34 pores, water apparently adsorbed and increased the gas permeances. Thus the effect of water on gas permeation is a useful indicator of the membrane quality.

ARTICLES, PRESENTATIONS, AND STUDENT SUPPORT

Journal Articles (peer reviewed)

- Poshusta, J.C., Tuan, V.A., Falconer, J.L., Noble, R.D., "Synthesis and Permeation Properties of SAPO-34 Tubular Membranes", *Ind. Eng. Chem. Res.*, 37 3924-3929 (1998).

- Poshusta, J.C., Noble, R.D., Falconer, J.L., "Temperature and pressure effects on CO_2 and CH_4 permeation through MFI zeolite membranes", *J. Membr. Sci.*, 160 115-125 (1999).

- Poshusta, J.C., Tuan, V.A., Pape, E.A., Noble, R.D., Falconer, J.L., "Separation of light gas mixtures using SAPO-34 membranes, *AIChE J.*, 46, 779-789, (2000)

- Poshusta, J.C., Noble, R.D., Falconer, J.L., "Characterization of SAPO-34 Membranes by Water Adsorption", in preparation

Conference Presentations

- Poshusta, J. C., T. A. Vu, J. L. Falconer, R. D. Noble, Effects of Temperature, Pressure, and Humidity on Light Gas Permeation through SAPO-34 Membranes, *AIChE Annual Meeting*, Dallas, Oct. 31 - Nov. 5, 1999.

- Poshusta, J. C., E. A. Pape, T. A. Vu, J. L. Falconer, and R. D. Noble, Separation of Light Gas Mixtures Through SAPO-34 Membranes, poster presentation at the *International Conference on Membranes*, Toronto, Canada, June 12-18, 1999.

- Poshusta, J. C., E. A. Pape, V. A. Tuan, J. L. Falconer, and R. D. Noble, Temperature and Pressure Dependence of Light Gas Mixtures through SAPO-34 Membranes, *Materials Research Society*, San Francisco, April 5-9, 1999.

- Poshusta, J. C., T. A. Vu, J. L. Falconer, R. D. Noble, Synthesis and Gas Permeation Properties of Tubular Zeolite Membranes, *Proceedings of the International Workshop on Zeolitic Membranes and Films*, Gifu, Japan, (1998) 9-12.

- Pape, E.A., J.C. Poshusta, V.A. Tuan, R.D. Noble, J.L. Falconer, Characterization of SAPO-34 Membrane on a Tubular Alumina Support, *AIChE Annual Meeting*, Miami, Nov. 1998.

- Poshusta, J. C., J. L. Falconer, R. D. Noble, Temperature and Pressure Effects in CO_2/CH_4 Permeation in Zeolite Membranes, *Proceedings of the Fifth International Conference on Inorganic Membranes*, Nagoya, Japan, (1998) 132-134.

- Poshusta, J. C., J. L. Falconer, R. D. Noble, Modeling of Single Gas Permeance in Zeolite Membranes. *Proceedings of the 9th Annual Meeting of the North American Membrane Society*, May 31 - June 4, 1997.

Students supported under this grant

- Joseph Poshusta, graduate (Ph.D.) student in chemical engineering, University of Colorado
- Christopher Gump, graduate (Ph.D.) student in chemical engineering, University of Colorado
- Tracy Gardner, graduate (Ph.D.) student in chemical engineering, University of Colorado
- Ana Flores, undergraduate in chemical engineering, University of Colorado
- Joseph Tamburini, undergraduate in chemical engineering, University of Colorado
- Megan Fortado, undergraduate in chemical engineering, University of Colorado