Materials Science & Engineering Doctoral Defense

Synthesis and Characterization of Microporous Inorganic Membranes for Propylene/Propane Separation

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abstract

Membrane-based gas separation is promising for efficient propylene/propane (C_3H_6/C_3H_8) separation with low energy consumption and minimum environment impact. Two microporous inorganic membrane candidates, MFI-type zeolite membrane and carbon molecular sieve membrane (CMS) have demonstrated excellent thermal and chemical stability. Application of these membranes into C_3H_6/C_3H_8 separation has not been well investigated. This dissertation presents fundamental studies on membrane synthesis, characterization and C_3H_6/C_3H_8 separation properties of MFI zeolite membrane and CMS membrane.

MFI zeolite membranes were synthesized on α-alumina supports by secondary growth method. Novel positron annihilation spectroscopy techniques were used to non-destructively characterize the pore structure of these membranes. A bimodal pore structure consisting of intracrystalline zeolitic micropores of ~0.6 nm in diameter and irregular intercrystalline micropores of 1.4 to 1.8 nm in size was revealed for the membranes. The template-free synthesized membrane exhibited a high permeance but a low selectivity in C_3H_6/C_3H_8 mixture separation.

CMS membranes were synthesized by coating/pyrolysis method on mesoporous γ-alumina support. Such supports allow coating of thin, high-quality polymer films and subsequent CMS membranes with no infiltration into support pores. The CMS membranes show strong molecular sieving effect, offering a high C_3H_6/C_3H_8 mixture selectivity of ~30. The gas transport and separation properties of CMS membrane are membrane thickness dependent. This can be explained by the thickness dependent chain mobility of the polymer film resulting in final carbon membrane of reduced pore size with different effects on transport of gas of different sizes.

CMS membranes demonstrate excellent C_3H_6/C_3H_8 separation performance over a wide range of feed pressure, composition and operation temperature. No plasticization was observed at a feed pressure up to 100 psi. CMS membrane experienced a decline in permeance, and an increase in selectivity over time under on-stream C_3H_6/C_3H_8 separation. This aging behavior is due to the reduction in effective pore size and porosity caused by oxygen chemisorption and physical aging of the membrane structure.

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