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Fluorine-induced microporous silica membranes: Dramatic improvement in hydrothermal stability and pore size controllability for highly permeable propylene/propane separation

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Abstract

A molecular sieving membrane was fabricated using triethoxyfluorosilane (TEFS), which contains Si-F bonds and is categorized as a pendant-type alkoxysilane. The hydrothermal stability and hydrocarbon (C_3H_6, C_3H_8) permeation properties were evaluated for TEFS membranes. When a fluorine-induced silica membrane had a Si-F bond in the amorphous structure, the reaction of steam and Si-F groups during steam treatment formed Si-OH groups, which slightly decreased the gas permeance. Even though gas permeance slightly decreased under a steam atmosphere, a TEFS membrane calcined at 350 ^{o}C had networks that were looser and more uniform than those of a conventional SiO_2. In addition, the formation of adsorption sites (Si-OH groups) under steam treatment enhanced both interactions with the π-bonds (C=C double bond) of C_3H_6 and the C_3H_6/C_3H_8 permeation properties (C_3H_6 permeance: 2.2 x 10^{-7} mol m^{-2} s^{-1} Pa^{-1}, C_3H_6/C_3H_8 permeance ratio: 42 at 35 ^{o}C). The hydrothermal stability was dramatically enhanced by calcination temperatures as high as 750 ^{o}C due to the presence of fewer Si-OH and Si-F bonds in the amorphous structure, although the network pore size of a TEFS membrane was the same whether it was calcined at 750 ^{o}C or at 350 ^{o}C.

Keywords: Amorphous silica; Fluorine; Hydrothermal stability; Affinity control; Gas separation