

Separation of Hydrogen from Mixtures with CO₂ and CH₄ by Indigenous Alumina and Mullite Supported Silica Membranes

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Abstract

Hydrogen is considered as the green energy carrier of the future. Steam reforming is the most popular process for producing hydrogen from hydrocarbon feed stocks such as methane. In order to get pure hydrogen, it is necessary to remove carbon dioxide and methane from the reformer gas by a suitable technique. The present work is concerned with the separation of hydrogen from hydrogen-carbon dioxide and hydrogen-methane gas mixtures by indigenously developed inorganic membranes. Silica membranes with alumina and mullite supports have been developed to separate hydrogen from hydrogen-carbon dioxide and hydrogen-methane gas mixtures. The basic pressing technique was used to achieve alumina and mullite supports for membranes. Then the silica membranes have been developed on the support by sol gel dip coating method. Characterization of membranes was done under scanning electron microscope and surface area, pore volume and the average pore size were measured using BET surface area analyzer. The pore size of alumina supported silica membrane was found to be in the range of 1.54 to 2.69 nm in the water and TEOS molar ratio range of 8 to 12 for single to quadruple coated membranes. The mullite supported silica membrane pore size was in the range of 1.51 to 2.42 nm in the water and TEOS molar ratio range of 8 to 12 for single to triple coated membranes. The membranes were found to be porous having good mechanical strength as well as high hydrogen permeance and selectivity. The feed mixtures of H₂ and CO₂, H₂ and CH₄ were passed separately across the silica membranes to study the membrane permeance and selectivity. The effect of pressure and temperature were studied and the process conditions were optimized. The most favourable condition for hydrogen separation from hydrogen-carbon dioxide gas mixture for alumina supported silica membranes was established at 450 kPa pressure and 600 °C temperature. Under this condition, the flux and permeance of hydrogen were 6.77 mol /m²s and 1.48 x 10⁻⁵ mol s / kg m, respectively and the H₂/CO₂ selectivity was 8.43. For the mullite supported membrane, under the optimum condition (500 kPa pressure and 650°C temperature), the flux and permeance of hydrogen

were $6.64 \text{ mol /m}^2\text{s}$ and $1.32 \times 10^{-5} \text{ mol s / kg m}$, respectively and the H_2/CO_2 selectivity was 8.08.

The separation of hydrogen from hydrogen-methane gas mixture at a pressure of 450 kPa and a temperature of $600 \text{ }^\circ\text{C}$, the flux and permeance of hydrogen were obtained as $7.8 \text{ mol /m}^2\text{s}$ and $1.71 \times 10^{-5} \text{ mol s / kg m}$ respectively and the H_2/CH_4 selectivity is 8.05 for alumina supported silica membranes. Whereas for mullite membranes at a pressure of 500 kPa and a temperature of $650 \text{ }^\circ\text{C}$, the flux and permeance of hydrogen obtained are $6.9264 \text{ mol /m}^2\text{s}$ and $1.377 \times 10^{-5} \text{ mol s / kg m}$, respectively and the H_2/CH_4 selectivity is 8.00. The dusty gas model was used for mathematical analysis of hydrogen separation from hydrogen-carbon dioxide and hydrogen-methane gas mixtures.

Key words: *Hydrogen separation, Mullite, Silica membrane, Sol-gel synthesis, Dusty gas model, Permeance, Hydrogen flux, Hydrogen selectivity*