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H2 Permeation Properties of a Catalytic Membrane Reactor in Methane Steam Reforming Reaction

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Abstract : Cylindrical alumina microfiltration membrane (GMITM Corporation, inside diameter=9 mm, outside diameter=13 mm, length= 50 mm) with an average pore size of 0.5 micrometer and porosity of about 0.35 was used as the support for membrane reactor. This support was soaked in boehmite sols, and the mean particle size was adjusted in the range of 50 to 500 nm by carefully controlling hydrolysis time, and calcined at 650 °C for two hours. This process was repeated with different boehmite solutions in order to achieve an intermediate layer with an average pore size of about 50 nm. The resulting substrate was then coated with a thin and dense layer of silica by counter current chemical vapour deposition (CVD) method. A boehmite sol with 10 wt.% of nickel which was prepared by a standard procedure was used to make the catalytic layer. BET, SEM, and XRD analysis were used to characterize this layer. The catalytic membrane reactor was placed in an experimental setup to evaluate the permeation and hydrogen separation performance for a steam reforming reaction. The setup consisted of a tubular module in which the membrane was fixed, and the reforming reaction occurred at the inner side of the membrane. Methane stream, diluted with nitrogen, and deionized water with a steam to carbon (S/C) ratio of 3.0 entered the reactor after the reactor was heated up to 500 °C with a specified rate of 2 °C/ min and the catalytic layer was reduced at presence of hydrogen for 2.5 hours. Nitrogen flow was used as sweep gas through the outer side of the reactor. Any liquid produced was trapped and separated at reactor exit by a cold trap, and the produced gases were analyzed by an on-line gas chromatograph (Agilent 7890A) to measure total CH₄ conversion and H₂ permeation. BET analysis indicated uniform size distribution for catalyst with average pore size of 280 nm and average surface area of 275 m².g⁻¹. Single-component permeation tests were carried out for hydrogen, methane, and carbon dioxide at temperature range of 500-800 °C, and the results showed almost the same permeance and hydrogen selectivity values for hydrogen as the composite membrane without catalytic layer. Performance of the catalytic membrane was evaluated by applying membranes as a membrane reactor for methane steam reforming reaction at gas hourly space velocity (GHSV) of 10,000 h^{−1} and 2 bar. CH₄ conversion increased from 50% to 85% with increasing reaction temperature from 600 °C to 750 °C, which is sufficiently above equilibrium curve at reaction conditions, but slightly lower than membrane reactor with packed nickel catalytic bed because of its higher surface area compared to the catalytic layer.

Keywords : catalytic membrane, hydrogen, methane steam reforming, permeance
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