

SEPARATION OF HYDROGEN AND CARBON DIOXIDE USING A NOVEL MEMBRANE REACTOR IN ADVANCED FOSSIL ENERGY CONVERSION PROCESSES

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ABSTRACT

Inorganic membrane reactors offer the possibility of combining reaction and separation in a single operation at high temperatures to overcome the equilibrium limitations experienced in conventional reactor configurations. Such attractive features can be advantageously utilized in a number of potential commercial opportunities, which include dehydrogenation, hydrogenation, oxidative dehydrogenation, oxidation and catalytic decomposition reactions. However, to be cost effective, significant technological advances and improvements will be required to solve several key issues which include: (a) permselective thin solid film, (b) thermal, chemical and mechanical stability of the film at high temperatures, and (c) reactor engineering and module development in relation to the development of effective seals at high temperature and high pressure.

In this project, we are working on the development and application of palladium and palladium-silver alloy thin-film composite membranes in membrane reactor-separator configuration for simultaneous production and separation of hydrogen and carbon dioxide at high temperature. From our research on Pd-composite membrane, we have demonstrated that the new membrane has significantly higher hydrogen flux with very high perm-selectivity than any of the membranes commercially available. The steam reforming of methane by equilibrium shift in Pd-composite membrane reactor is being studied to demonstrate the potential application this new development.

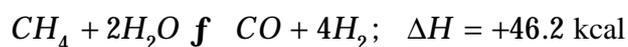
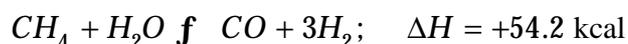
To have a better understanding of the membrane reactor, we developed a two-dimensional pseudo-homogeneous reactor model for steam reforming of methane by equilibrium shift in a tubular membrane reactor. Radial diffusion was taken into account for concentration gradient in the radial direction due to H_2 -permeation through the membrane. For H_2 -permeation, Sieverts

law was used in the solution of the reactor model equations with the index n (dependent on the type and thickness of the membrane film) as a parameter.

$$N_H = \frac{Q_H}{h} (P_H^n - p_H^n)$$

where N_H is the hydrogen flux, Q_H is the permeability coefficient and h is the effective membrane thickness (Pd-film). Hydrogen partial pressures on the feed side and separation side are given by P_H and p_H , respectively.

The steam reforming of methane can be represented by the following equations:



With appropriate reaction rate expressions, a set of partial differential equations was derived using the continuity equation for the reaction system. The equations were solved by finite difference methods. The solution of the model equations is complicated by the coupled reactions. To overcome the numerical difficulties, several alternative schemes were implemented in the solution algorithm to get converged solution. The solution would be straightforward if only the third reaction is considered. Since the concentration of carbon monoxide (water-gas shift reaction) may be significant in many operating conditions, and a good model should be able to predict it, one needs to consider the system of reactions as presented.

In this presentation, modeling work on the steam reforming of methane in the membrane reactor along with perm-selectivity data on hydrogen through pd-composite membrane for a number of mixed gases would be presented.

List of Published Journal Articles, Completed Presentations and Students Receiving Supports from the Grant:

Journal Articles:

1. Khan, M.H., and Ilias, S., "Steam Reforming of Methane by Equilibrium Shift in a Pd-Composite Membrane Reactor," In Preparation for Sep. Sci. Technol., (2001).

Presentations:

1. Carter, S., Khan, M.H., and Ilias, S., "Development of H₂-selective Metal-Ceramic Composite Membranes for High Temperature Gas Separations and Membrane Reactors," US DOE HBCU/OMI Annual Symposium, Pittsburgh, Pennsylvania, June 8-9, 2000.

Students Receiving Supports:

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