PREPARATION AND CHARACTERIZATION OF COMPOSITE MEMBRANE FOR HIGH
TEMPERATURE GAS SEPARATION

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Abstract

A palladium-ceramic composite membrane was developed by electroless deposition of palladium on microporous ceramic substrate. By measuring the permeation characteristics of pure and mixed gases, it was found that the new membrane has a very high permeability and selectivity for hydrogen. During this reporting period, a cocurrent flow model for a membrane module was developed to study the performance of palladium-ceramic composite membranes for hydrogen separation in terms of product purity under various operating conditions. The model neglects mass transfer resistance but it was found that the difference between the predicted and actual hydrogen composition was less than 12%.

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Executive Summary

To develop a new class of hydrogen permselective inorganic membrane, electroless plating was used to deposit a thin palladium film on microporous ceramic substrate. The palladium film on the microporous ceramic substrate acts as selective separation barrier for hydrogen separation. This palladium-ceramic composite membrane was tested and characterized for hydrogen separation. It was found that the new membrane has a very high permselectivity for hydrogen and permeability increases with increasing operating temperature. Thus the membrane has potential for applications at elevated temperature for hydrogen separation and also for use in membrane reactors for production and separation of hydrogen from dehydrogenation reactions. During this reporting period, a mathematical model was developed to study the separation of hydrogen in membrane module using the palladium-ceramic composite membrane under cocurrent flow configuration. The model neglects mass transfer resistance but it was found that the difference between the predicted and actual hydrogen composition was less than 12%. Given the simplicity of the model, it appears that the model is adequate for the present work.

Introduction

To develop a new class of permselective inorganic membranes, we have identified electroless plating as potential a route to deposit a thin metal film on porous substrate. Electroless plating is a controlled autocatalytic deposition of continuous film on the surface of a substrate by the interactions of a metal salt and a chemical reducing agent. This method can give thin films of metals, alloys and composites on both conducting and nonconducting surfaces. The objective of this project is to develop thin film palladium membranes for separation of hydrogen in high temperature applications. We plan to use electroless plating to deposit thin palladium films on microporous ceramic and silver substrates. We plan to characterize the membrane in terms of permeability and selectivity for gas separation. To accomplish the research objective, the project requires three tasks:

I. Development of a Process for Composite Membrane Formation

The work will involve the selection of methods for thin metal/metal-alloy film deposition on inorganic and ceramic microporous substrates. To assure reproducible film, optimization of the electroless deposition will be required. This step will involve determination of the optimum configuration of the plating bath, which consists of studying the effect of parameters like pH, temperatures, concentrations, type of reducing agent and sensitizing solution on the rate of deposition of the film.

II. Characterization of Fabricated Composite Membrane

The work for this task will start with the fabrication of composite membrane. The membrane will be characterized by EDAX and SEM analysis. The measurements will also include permeability and steady state diffusion of pure and mixed gases for hydrogen, and the effect of thermal cycling on the stability of the membrane film.

III. Development of Theoretical Model For Hydrogen Gas Separation

The work for this task will be to find or develop a suitable model to describe the permeation of hydrogen gas through a thin palladium film on a porous support. Models developed in the literature deal only with the permeation of gases through a metal membrane. These models may or may not be applicable to our composite case. The applicability of the model will depend on the role of the porous support in permeation of gases. It is well known that the permeability of gases through porous ceramics increases at higher temperatures. However, the behavior of hydrogen permeation through a palladium film is much more complex. It is believed that the hydrogen molecule dissociates into hydrogen atom at one end and reassociates at the other end. A theoretical model will be developed to facilitate prediction and interpretation of data obtained in the permeation experiments. Permeation through the porous substrate will be analyzed in terms of Knudsen and binary gas diffusion as well as

surface diffusion. In case of Pd/substrate composites, a model will be developed which accounts for the kinetics of hydrogen diffusion.

Results and Discussion

This report covers the quarter ending May 31, 1997. During this quarter, we completed the analysis of cocurrent flow configuration for separation of hydrogen using our high-selective palladium-ceramic composite membrane. When compared with the experimental data on product purity, the result shows that model under predicts the reject composition and over predicts the product purity. These results indicate that the gas phase mass transfer resistance restricts hydrogen diffusion. In our simple model, the mass transfer resistance is ignored. The difference between the predicted and actual hydrogen composition is less than 12%. Given the simplicity of the model, it can be concluded that the model accurately predicted the cocurrent membrane module performance.

During the next quarter we plan to complete the counter-current flow model and with this we hope to complete the project.

Conclusions

A cocurrent flow model is developed to analyze the separation of hydrogen using high perm-selective palladium ceramic composite membrane. The model ignores the mass transfer resistance but is useful in predicting the performance of the membrane module for hydrogen separation in terms of product purity for a given set of operating conditions.