PROJECT facts

U.S. Department of Energy
Office of Fossil Energy
National Energy Technology Laboratory

Carbon Sequestration

04/2008

A New Concept for the Fabrication of Hydrogen Selective Silica Membranes

Background

As stated in the National Research Council report on Novel Approaches to Carbon Management, a novel membrane is needed that can achieve the separation of carbon dioxide (CO_2) and hydrogen (H_2) at a high temperature and pressure. Extensive efforts over the last several decades have explored high temperature H_2-selective membranes made of silicon dioxide (SiO_2) and other oxides, palladium (Pd) and other metals or alloys and, more recently, various zeolites and non-aluminosilicate molecular sieves. Although promising separation results have been reported for many of them, these technologies, they all suffer from high production costs for membrane fabrication and from long term stability problems. This project revisits the objective of high temperature H_2-selective membranes with a fresh look. It explores a new concept for the fabrication of ultrathin, hydrothermally stable, molecular sieve, H_2-selective membranes.

Description

Researchers will investigate a new method for making extremely thin, high-temperature, hydrogen-selective silica membranes with high H_2 selectivity, flux, and stability in a water gas shift reactor environment. The proposed membrane will rely on the use of layered microporous silicates that have very limited pore openings perpendicular to the layers. The largest pore openings that are perpendicular to the layers are ideal for H_2 molecular sieving membranes. The new membrane fabrication method consists of synthesis of layered silicates, preparation of thin plate-like particles from these layered silicates, and deposition of the particles using layer-by-layer assembly followed by calcination. If successfully developed, the membranes could be used for economical production of H_2 from coal-supplied synthesis gas, while simultaneously producing a concentrated carbon dioxide (CO_2) stream for capture and storage.

The project will include investigation and development of membrane synthesis techniques, membrane synthesis, and evaluation of membrane separation performance and stability. Specifically, membranes will be tested for H_2 separation from CO_2 at high temperatures and pressure and for thermal stability at high temperature and pressure in the presence of water vapor.

Primary Project Goal

The primary goal of the proposed research is to develop a new economic and easy to scale up method for the fabrication of hydrogen selective silica membranes with high hydrogen selectivity, flux, and stability in a water gas shift reactor environment.

CONTACTS

Sean Plasynski
Sequestration Technology Manager
National Energy Technology Laboratory
626 Cochrans Mill Road
P.O. Box 10940
Pittsburgh, PA 15236
412-386-4867
sean.plasynski@netl.doe.gov

David Lang
Project Manager
National Energy Technology Laboratory
626 Cochrans Mill Road
P.O. Box 10940
Pittsburgh, PA 15236-0940
412-386-4881
david.lang@netl.doe.gov

Michael Tsapatsis
Principal Investigator
University of Minnesota
Department of Chemical Engineering & Materials Science
151 Amundson Hall
421 Washington Avenue, SE
Minneapolis, MN 55455
612-626-0920
tsapatsi@cems.umn.edu

PARTNERS

University of Minnesota

Department of Chemical Engineering and Materials Science

PERFORMANCE PERIOD

8/1/2004 to 4/30/2008

COST

Total Project Value

$267,393

DOE/Non-DOE Share

$267,393 / $ 0

ADDRESS

National Energy Technology Laboratory

1450 Queen Avenue SW
Albany, OR 97321-2198
541-967-5892

2175 University Avenue South
Suite 201
Fairbanks, AK 99709
907-452-2559

3610 Collins Ferry Road
P.O. Box 880
Morgantown, WV 26507-0880
304-285-4764

626 Cochrans Mill Road
P.O. Box 10940
Pittsburgh, PA 15236-0940
412-386-4687

One West Third Street,
Suite 1400
Tulsa, OK 74103-3519
918-699-2000

CUSTOMER SERVICE

1-800-553-7681

WEBSITE

www.netl.doe.gov

Objectives

The specific objective is to demonstrate the fabrication of a 100 nm or thinner supported film of SiO_2 using a technique that can be easily and economically scaled up and to show that this membrane can meet the following requirements:

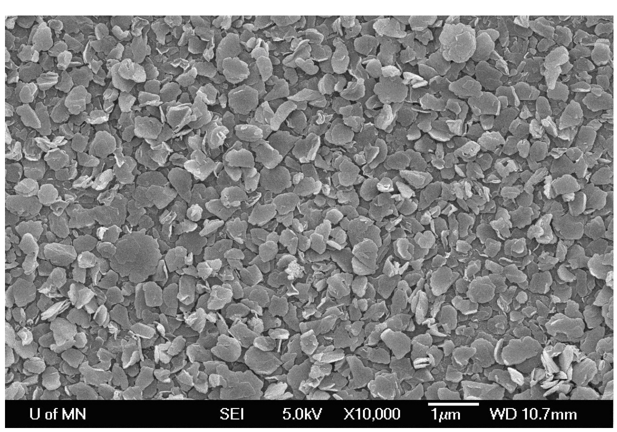
- Hydrogen permeance in excess of 10^{-7} mol/m^2-s-Pa with a H_2/CO_2 selectivity in excess of 100 at temperatures in the range of 500-700 °C, a pressure of 20 bar, and a stream composition representative of feed to a water gas shift reactor.
- Maintain stable membrane performance at the above values and conditions in the presence of steam (25 percent H_2O) for at least one month.

Benefits

Fossil fuels provide over 80 percent of the world's energy today and are expected to continue their dominance throughout the foreseeable future. Innovations in technologies that could lead to practical and cost-effective means for either reducing emissions from fossil-fueled power plants or removing CO_2 from the atmosphere could have far-reaching benefits for the economy of the United States. This project will investigate a novel alternative to current technology for the capture and sequestration of CO_2 that could result in a process for the economic production of H_2 from coal-supplied synthesis gas while simultaneously producing a concentrated CO_2 stream for sequestration. This approach has the potential to show a significant improvement in performance and cost compared to currently available technologies.

Accomplishments

- Plate-like small pore layered silicate crystals have been synthesized and calcined without significant agglomeration.
- A deposition process that leads to uniform, high-coverage (~100 percent) coating of the layered silicate particles on porous alpha-alumina supports has been developed.
- Multi-layered coatings on alpha alumina oxide discs have been obtained by repeating the deposition process.
- A permeation system has been designed and constructed to test the coated discs.
- Deposition method was extended for the film fabrication on zirconia coated stainless steel (SS) tubes.
- Permeation setup for testing the coated SS tubes was installed.
- Both coated discs and SS tubes were fabricated; permeation tests were conducted for both. For coated discs, demonstrated activated gas transport and hydrogen selectivity above one achievable by Knudsen permeation mechanism.
- Initiated investigations of the exfoliation of MCM-22(P) to single layers in order to improve the deposition procedure, reduce the membrane thickness, and/or increase the silicate particle overlap.



Coating of one layer of MCM-22 on γ-alumina coated α-alumina support