



A NEW CONCEPT FOR THE FABRICATION OF HYDROGEN SELECTIVE SILICA MEMBRANES

Background

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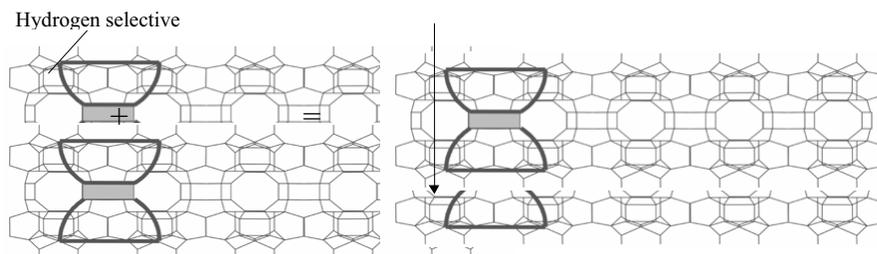
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As stated in the NRC report on Novel Approaches to Carbon Management, there is a need for a novel membrane that can achieve the separation of CO_2 and H_2 at a high temperature and pressure. Extensive efforts over the last several decades have explored high temperature H_2 -selective membranes made of SiO_2 and other oxides; 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 these technologies, they all suffer from high production costs for membrane fabrication and 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.

The concept is based on the use of crystalline layered silicates that are often encountered as by-products in the synthesis of, and in some cases are "precursors" to, high silica zeolites. Several of the currently known high silica layered materials are made up of microporous layers that may contain pores running within the layers but with no open microporosity perpendicular to the layers. The largest pore openings in the direction perpendicular to the layers are 6-membered-ring openings, i.e. rings of 6 interconnected SiO_4 tetrahedra. These ultra-small pore openings are ideal for H_2 molecular sieving membranes.



Condensation of silicate layers leads to a zeolite by elimination of water and formation of Si-O-Si linkages between the layers. In this view, large pores are evident however in the direction indicated by the arrow transport is limited by pores that will allow permeation of hydrogen but not of carbon dioxide. The dangling bonds at the end of the layers represent Si-OH while the lines represent Si-O-Si.



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COST

Total Project Value
\$237,393

DOE/Non-DOE Share
\$237,393/\$0

The fabrication method consists of the synthesis of ordered 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. The membranes will be tested for H₂ separation from CO₂ at high temperature and pressure and tested for thermal stability at high temperature and pressure in the presence of water vapor.

Primary Project Goal

The primary goal of this project is to develop a new, economic, easily scaled up method for the fabrication of a hydrogen selective silica membrane that has high hydrogen selectivity and flux and is stable at the environmental conditions existing in a water gas shift reactor.

Objectives

The specific objective is to demonstrate the fabrication of a 100 nanometer (nm) or thinner supported film of SiO₂ 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/m²-s-Pa with a H₂/CO₂ 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% H₂O) for at least 1 month.

Benefits

Fossil fuels provide over 80% 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 CO₂ from the atmosphere could have far-reaching benefits for the economy of the United States. This proposal represents a novel alternative to current technology for the capture and sequestration of CO₂ that could result in a process for the economic production of H₂ from coal supplied synthesis gas while simultaneously producing a concentrated CO₂ stream for sequestration. This approach has the potential to show a significant improvement in performance and cost compared to currently available technologies.