II.D.2 Scale Up of Hydrogen Transport Membranes for IGCC and FutureGen Plants

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Subcontractor:
Eastman Chemical Co., Kingsport, TN

Project Start Date: October 1, 2005
Project End Date: June 2012; American Recovery and Reinvestment Act (ARRA) funded extension through September 2015

Fiscal Year (FY) 2011 Objectives

- Demonstrate a cost-effective H_2/CO_2 membrane separation system.
- Scale up membrane manufacturing.
- Construct, install, and operate subscale engineering prototype.
- Down-select engineering, procurement, and construction firm and conduct pre-front end engineering design for a 4-10 ton per day pre-commercial scale up.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Production section of the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan:

(K) Durability

(Q) Testing and Analysis

(R) Cost

Technical Targets

Eltron's hydrogen membrane system has demonstrated high flux rates of >99.99% pure hydrogen at expected temperatures and differential operating pressures. This project is currently focused on scaling up these membranes while meeting the following 2015 DOE technical targets for dense metallic membranes:

- Module Cost: $500/ft^2 of membrane
- Durability: >43,800 hours
- Hydrogen Recovery: 90%

FY 2011 Accomplishments

- Membrane manufacturing was scaled up to produce 5 ft-long tubular membranes. These tubular membranes are ½ inch outside diameter and have a wall thickness of 500 microns.
- Two foot-long membranes were tested at Eltron Research & Development and demonstrated 70% hydrogen recovery.
- A subscale engineering prototype pilot reactor was designed, constructed, and installed at Eastman Chemical Co.

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Introduction

The overall objective of this project is to scale up the hydrogen transport membrane technology system for energy efficient carbon capture and hydrogen separation from industrial sources thereby enabling early technology commercialization by reducing time, technology risk, and cost. The goal of the project is to scale up Eltron's dense hydrogen transport membranes which can extract and purify hydrogen to very high levels from coal-derived water-gas shift mixtures, while minimizing the pressure drop of CO_2 in order to lower capital and compression costs for CO_2 sequestration. Dense hydrogen separation membranes are being developed to be compatible with high-temperature water-gas shift reactor conditions for water-gas shift reactors placed downstream from coal gasifiers in integrated gasification combined cycle (IGCC) type systems. Hydrogen separation membranes must be compatible with high-temperature water-gas shift reactor temperatures (approximately 320-440°C) and with mixtures of water-gas shift components containing hydrogen, steam, CO_2 and CO as well as residual impurities which escape upstream warm-gas clean up systems and the beds of water-gas shift catalyst. The hydrogen separation membranes must also function at pressures near that of the coal gasifiers, 450-1,000 psi.

Approach

Eltron is addressing all key issues to successfully scale up this technology. Bench scale testing is being conducted to demonstrate flux, hydrogen recovery, and durability. In addition, process modeling and techno-economic analyses are being developed. Eltron is scaling up this technology in three steps. Reactors nominally sized for 12 lbs/day and 250 lbs/day hydrogen separation will be designed, constructed, and operated. Eltron was awarded an ARRA project to further scale up this technology to a 4-10 ton per day pre-commercial module scale. The approach for the ARRA portion of the project is to continue membrane development, conduct pre-front end engineering design and front end engineering design engineering packages for the selected pre-commercial module site; and to design, construct, and operate the pre-commercial module and conduct appropriate engineering analyses.

Results

Figure 1 shows a general schematic of Eltron's membrane system. The membrane is composed of three dense metal alloy layers. The center layer is 500 microns thick and is a low-cost metal alloy with high hydrogen permeability. A thin catalyst layer (<1 micron) is deposited on both the inside and outside surface of the membrane. High pressure shifted syngas is fed to the outside surface of the membrane. Pure hydrogen is collected on the inside of the tube. This membrane was designed to operate at water-gas shift conditions and retain CO_2 at high pressures to minimize compression costs for CO_2 capture and sequestration.

In FY 2011, Eltron scaled up the manufacturing of these tubular membranes to 5 ft-long tubes. These tubes are ½ inch outside diameter tubes with a wall thickness of 500 microns. Eltron deposits metal alloy catalysts on the inside and outside surface of the tubes. A scanning electron microscope image of a deposited catalyst layer is shown in Figure 2. The deposited catalyst is uniform, dense, and well-adhered to the bulk metal surface.

Two foot pieces of membrane were tested at Eltron at 340°C and a differential pressure of 400 psig in a simulated water-gas shift feed stream. Figure 3 shows the observed hydrogen flux as a function of time. A stable flux rate of 28 scfh/ft^2 was observed for 16 hours. This flux rate was lower than expected due to the effect of concentration polarization inherent in low-flow rate bench-scale reactors. In this particular test the membrane was able to recovery 70% of the hydrogen in the feed stream.

Finally, Eltron designed and constructed the first reactor in our three step scale up plan. Figure 4 shows the constructed sub-scale engineering prototype. This reactor was designed to separate up to 12 lbs/day of hydrogen from a gasified coal feed stream at Eastman Chemical Co. site in Kingsport, TN.

On the ARRA portion of the project Eltron has down-selected URS as the engineering, procurement, and construction firm for the pre-commercial module. URS and Eltron are evaluating three potential sites for operating the pre-commercial module.

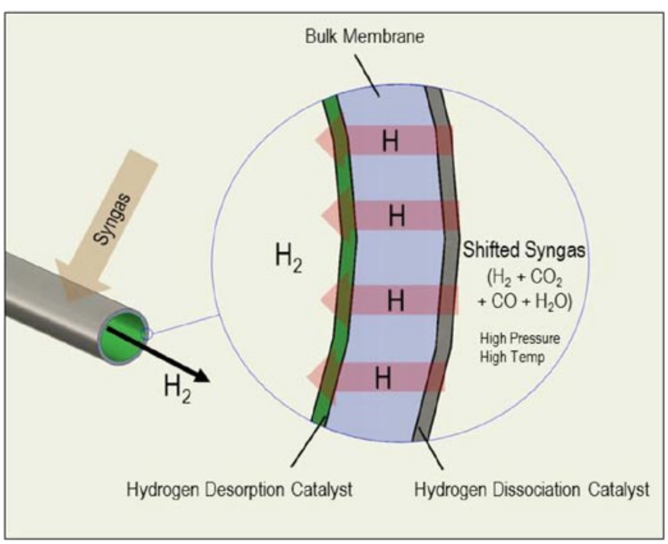

FIGURE 1. Schematic of Eltron's Dense Metal Tubular Membrane System

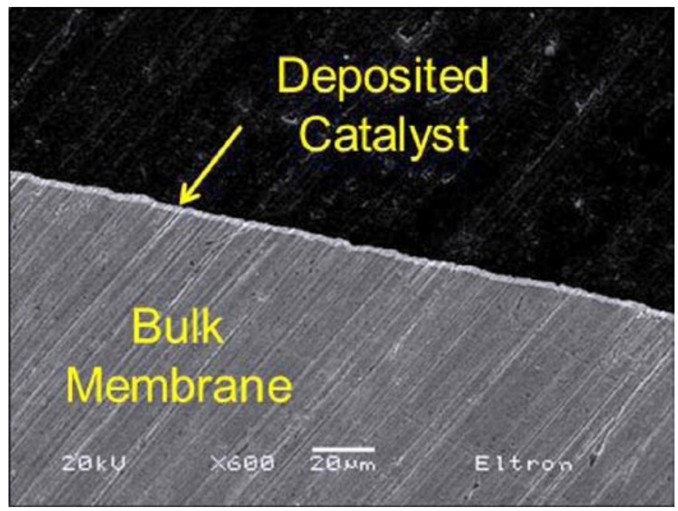

FIGURE 2. Scanning Electron Microscope Image of Deposited Catalyst

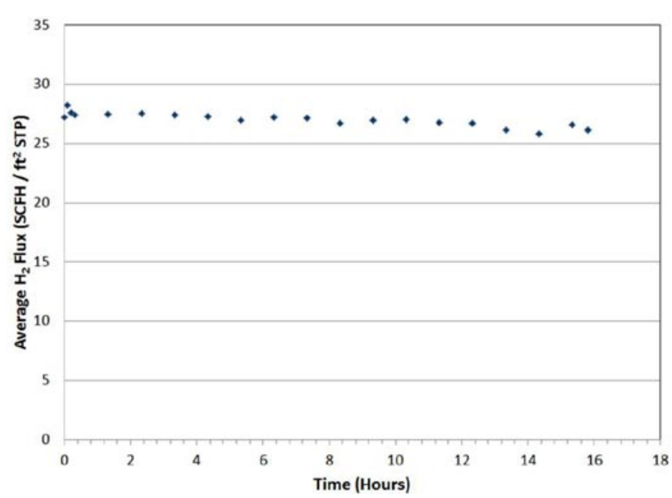

FIGURE 3. Hydrogen Flux vs. Time Recorded for a Two Foot-Long Membrane Tested at 340°C and 400 psig Differential Pressure

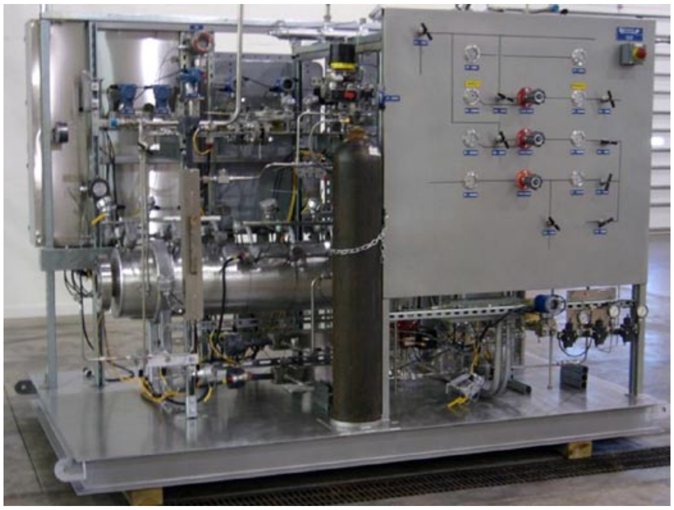

FIGURE 4. Completed Sub-Scale Engineering Prototype

Conclusions and Future Directions

In FY 2011 Eltron scaled up manufacturing of our tubular membrane and successfully tested membranes up to two feet long under expected operating conditions. In addition, Eltron designed, constructed, and installed the first scale-up reactor. During the next FY Eltron will operate the sub-scale engineering prototype, update our economic models with the results, and begin the design process for the next scale up step. Front end engineering design will be conducted on the down-selected site for the pre-commercial module.

Patents Issued

1. Mundschau, M.; Xie, X.; Evenson, C.; Grimmer, P.; Wright, H. Hydrogen Separation Process. US Patent 7,947,116. May 24, 2011.

FY 2011 Publications/Presentations

1. David Anderson. Carl Evenson. John Faull. Doug Jack. Scale-up of Membranes for Separation of Hydrogen from Syngas for Carbon Capture. 2010 AIChE Annual Meeting. November 9, 2010.

2. Michelle Livingston. e*t. al.* Advanced Membrane System for Hydrogen Separation and Carbon Dioxide (CO_2) Capture. 2011 North American Membrane Society Meeting. June 8, 2011.