# IV.A.11 Ceramic Membrane Reactor Systems for Converting Natural Gas to Hydrogen and Synthesis Gas (ITM Syngas)

Christopher M. Chen Air Products and Chemicals, Inc. 7201 Hamilton Boulevard Allentown, PA 18195 Phone: (610) 481-3315; Fax: (610) 706-6586; E-mail: chencm@airproducts.com

DOE Technology Development Manager: Arlene Anderson Phone: (202) 586-3818; Fax: (202) 586-9811; E-mail: Arlene.Anderson@ee.doe.gov

DOE Project Officer: Venkat Venkataraman Phone: (304) 285-4105; Fax: (304) 285-4403; E-mail: venkat.venkataraman@netl.doe.gov

Contract Number: DE-FC26-97FT96052

Subcontractors: Ceramatec, Inc.,Salt Lake City, UT Chevron, Richmond, CA Eltron Research Inc., Boulder, CO SOFCo EFS, Alliance, OH Pennsylvania State University, University Park, PA University of Alaska Fairbanks, Fairbanks, AK University of Pennsylvania, Philadelphia, PA

Start Date: October 1, 1997 Projected End Date: February 28, 2010

## Objectives

- Research, develop and demonstrate the Ion Transport Membrane (ITM) Syngas ceramic membrane reactor system for the low-cost conversion of natural gas to hydrogen and synthesis gas.
- Scale-up the ITM Syngas reactor technology through three levels of pilot-scale testing and precommercial demonstration.
- Obtain the technical, engineering, operating and economic data necessary for the final step to full commercialization of the ITM Syngas technology.

## **Technical Barriers**

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

- A. Fuel Processor Capital Costs
- E. Carbon Dioxide Emissions
- T. Oxygen Separation Technology

This project also addresses DOE Fossil Energy objectives to develop lower cost methods to produce hydrogen from natural gas.

## **Technical Targets**

ITMs are a revolutionary platform technology for producing hydrogen and synthesis gas for applications in power generation, transportation fuels, and chemicals. The ITM Syngas process provides a lower cost method for converting natural gas to hydrogen and synthesis gas by combining oxygen separation and natural gas partial oxidation in a single-step ceramic membrane reactor, with the potential for capital cost savings of over 30 percent.

The ITM Syngas process is amenable for capturing 95 percent of the carbon dioxide generated, since nearly all the carbon dioxide is in the high pressure syngas, rather than diluted in low pressure flue gas, as would be the case for conventional steam methane reforming. The ITM Syngas process addresses the DOE targets for reducing fuel processor capital costs, reducing carbon dioxide emissions, and improving oxygen separation technology.

#### Approach

ITM ceramic membranes are fabricated from nonporous, multi-component, metallic oxides and operate at high temperatures with exceptionally high oxygen flux and selectivity. Oxygen ions from low-pressure air permeate the ceramic membrane and are consumed through chemical reactions, creating a chemical driving force that pulls oxygen ions across the membrane at high rates. The oxygen reacts with natural gas in a partial oxidation process to produce a hydrogen and carbon monoxide mixture (synthesis gas) (see Figure 1).

During the current Phase 2 of the development project, the ITM Syngas technology is being scaled up from a 24,000 SCFD process development unit (PDU) to a 1 million SCFD subscale engineering prototype (SEP). The PDU operates at an equivalent of 24,000 SCFD of synthesis gas capacity and tests pilot-scale planar membranes under commercial process conditions. The SEP will be built to demonstrate the ITM Syngas technology using full-size membranes in sub-scale modules at up to an equivalent of 1 million SCFD of synthesis gas capacity.

Pilot-scale membrane modules are being fabricated for testing in the PDU and fabrication of the membrane reactor modules will be scaled-up in a production development facility (PDF) to supply the requirements

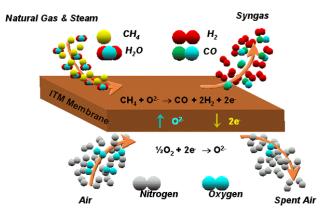


Figure 1. Conceptual ITM Syngas Process

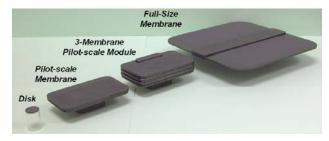


Figure 2. Over 300-Fold Increase in Membrane Area Since 1999

of the SEP (see Figure 2). Membrane reactors will be designed for the ITM Syngas process at the SEP and commercial scales. Membrane materials, catalysts, and seals will be tested at the laboratory scale under process conditions to obtain statistical performance and lifetime data. Advanced ITM Syngas processes will be developed, and the economics of operation at the commercial-plant scale will be evaluated.

#### Accomplishments

• Membrane modules with a full-size ceramic membrane and ceramic manifold components were manufactured using a proprietary all-ceramic joining method. The all-ceramic joints were formed with no increase in the leak rate of the module. The same all-ceramic joining process was used to successfully

join multiple sintered ceramic components with the same seal area and the number of joints as those in the membrane modules to be tested in the SEP. The all-ceramic joining process is a key enabling technology, since it allows the module to be made of the same ceramic composition throughout. By matching the expansion behavior of module components, stresses in the module are reduced during operation.

- Testing of ceramic-to-metal seals for the SEP was initiated. Seals performed well in initial high temperature tests with leak rates that were at or below target.
- As part of the planned scale-up from a lab-scale tapecaster, several batches of porous layer precursor tape were cast on a larger industrial tapecaster and were successfully used to make sintered PDU wafers.
- High activity methane reforming catalysts were developed and tested on dense-porous multilayer wafers at both atmospheric and full process pressures.
- Slow crack growth testing of ceramic samples in air at 750°C was completed. The tests showed that the ITM Syngas ceramic material does not exhibit any significant slow crack growth at these conditions.
- Changes to the PDU gas feed system materials of construction and the use of gas contaminant removal beds have eliminated contaminants from the system piping that had previously been detected on the surfaces of the membranes.
- The 24,000 SCFD PDU reactor vessel shell was modified to test full-size planar membranes. The modified reactor vessel successfully underwent shakedown at elevated temperature and pressure.
- A conceptual distributed-scale hydrogen process (1,500 kg/day hydrogen, 500 units/year) was developed, with a projected cost of under \$1.50/kg hydrogen for hydrogen production (reforming plus purification).
- Commercial ITM Syngas reactor control concepts were developed. The control concepts are expected to offer improved steady-state operability of the ITM Syngas reactor, as well as increased ability for process turndown and reduced potential for carbon formation.