II.A.5 Novel Catalytic Fuel Reforming

Patricia Irving (Primary Contact), Quentin Ming, and Jeffrey Harrison
InnovaTek Inc.
350 Hills Street, Suite 104
Richland, WA 99352
Phone: (509) 375-1093; Fax: (509) 375-5183; E-mail: irving@tekkie.com

DOE Technology Development Manager: Mark Paster
Phone: (202) 586-2821; Fax: (202) 586-9811; E-mail: Mark.Paster@ee.doe.gov

Objectives

The ultimate goal of this research project is to develop technology that will produce pure hydrogen from natural gas and liquid fuels using catalytic steam reforming and membrane hydrogen separation.

Phase IV is intended to demonstrate the InnovaGen™ fuel processing technology with multiple fuel types. The objectives to meet this goal are to:

- Achieve cost and efficiency targets for hydrogen production through microchannel design and advanced thermal management.
- Develop and test second-generation components.
- Integrate components and demonstrate multi-kW system with both liquid and gaseous fuels.

Phase IV includes considerable design, fabrication, and testing efforts. The result of these efforts will be prototypes that successfully demonstrate a fuel processing system at specified operating conditions.

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
- B. Operation and Maintenance (O&M)
- C. Feedstock and Water Issues
- E. Control & Safety

Approach

Develop a commercially viable product by:

- Integrating novel microstructured components (including reactor, heat exchanger, fuel injector) to increase thermal and chemical efficiency and reduce coking.
- Developing a sulfur-tolerant catalyst for multiple fossil and renewable fuels, using an advanced hydrogen-permeable membrane for hydrogen purification.
- Conducting iterative testing of system components that are progressively integrated.

Accomplishments

- Microstructured steam reformer, heat exchangers, and fuel injector were designed and integrated for high-efficiency operation.

- Proprietary catalyst and support foils with sulfur tolerance and high space velocity were developed for microchannel reactor.
- Hydrogen purification rate was doubled using a method to enhance a Pd-alloy membrane.
- Moveable test platform with integrated system components, controls, and insulation was designed and constructed.
- Fuel processor control system with data acquisition and safety algorithms was developed.
- Natural gas and diesel fuel reforming sufficient for multi-kW fuel cells were successfully demonstrated.
- Thermal efficiency of 65% for natural gas reforming was achieved.
- Capital cost for fuel processor is estimated to be within the DOE 2010 target of $1.50 per kg hydrogen.

Future Directions

This represents the final year of our contract. Further development of this technology is needed before a commercial system is viable. The following future work is recommended.

- Implement additional strategies to increase system efficiency and thermal integration.
- Scale up and develop casing for hydrogen purification module.
- Fully integrate with at least one fuel cell model.
- Conduct thorough reliability testing.
- Enhance controls for automated operation, off-site monitoring, data mining, self-diagnostics.
- Develop documentation – drawings, bills of material, manufacturing routers.

Introduction

Generation of hydrogen using primary fuel sources from existing production and distribution networks – i.e. natural gas, gasoline, diesel or jet fuels – will help accelerate commercial use of fuel cells. Fossil fuel-powered fuel cells or refueling stations using fossil fuels to produce hydrogen can form the bridge to a future when renewable resources are converted to hydrogen for fuel cells. When compared to compressed hydrogen, reformed hydrocarbon fuels offer a significant cost advantage in the delivery of power. The high energy density of these fuels will also contribute to increased run times per unit of fuel consumed, and size and weight reductions associated with fuel storage.

The InnovaGen™ fuel processor being developed by InnovaTek reforms multiple fuel types, including natural gas, gasoline, and diesel, to produce pure hydrogen. The fuel processor integrates microreactor and microchannel heat exchanger technology with advanced sulfur-tolerant catalysts and membrane technology for hydrogen purification. The ultimate goal of this cooperative project is the demonstration of an integrated bench-top prototype that can produce 12-60 liters per minute of hydrogen. The fuel processor being developed will provide a pure output stream of hydrogen that can be used without further purification for electrical generation by a 1-5 kW proton exchange membrane (PEM) fuel cell.

Approach

The objective of this research project is to develop fuel processing technology that makes it possible for fuel cells to replace internal combustion engines as the power source for electrical generators and auxiliary power units in the 1-5 kW range. This will require that the fuel processor developed will produce pure hydrogen from fuels using cost-competitive, highly efficient technology.

The design and optimization of a fuel processing system is complex because of the number of required components and functions (Figure 1). Our approach has been to design, fabricate, and test the individual components, then integrate the components and test the system with several fuel types and under a range of conditions. After analyzing test results from the components and the system, the process begins again to develop the next-generation system with design changes to improve the system's efficiency and cost.

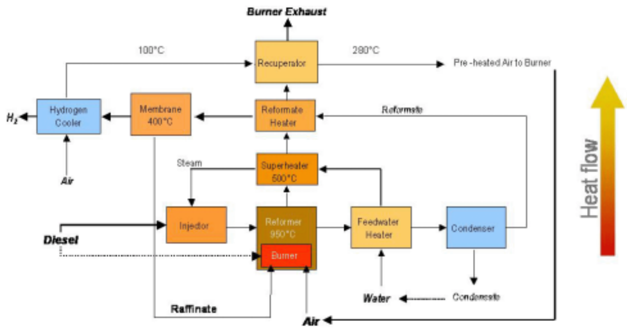

Figure 1. Process and Flow Diagram of the InnovaGen™ Fuel Processor

In FY 2004, we completed a demonstration of the second-generation integrated system.

The following objectives (according to key subsystems) were identified to achieve our goals.

- Conduct overall system modeling, analysis and design to provide component design parameters and system configuration to optimize efficiency.
- Develop a microchannel steam reformer with an integrated cross-flowing heat exchanger that is highly efficient and manufacturable.
- Develop a combustor that burns raffinate (membrane off-gas stream) to supply heat to the cross-flowing channels of the endothermic steam reformer.
- Produce a sulfur-tolerant catalyst for steam reforming of gaseous and liquid hydrocarbon fuels that has a high conversion rate and high space velocity as well as a support structure compatible with a microchannel reactor.
- Improve the fuel injector that atomizes liquid fuels and completely mixes steam with fuel without coking.
- Develop a palladium alloy membrane-based hydrogen purifier that has a high permeation rate without leaks.
- Develop an efficient thermal management system that uses tightly integrated microchannel heat exchangers.
- Implement operational and safety considerations, including failure means and effects analysis (FMEA) and Hazard and Operability Study (HAZOP) procedures.
- Produce full documentation, including drawings and product data management.

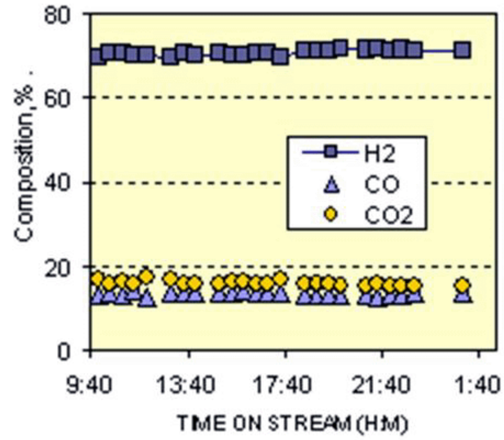

Figure 2. Product Composition from Steam Reforming of Ultra-low Sulfur Diesel Fuel

- Improve physical layout and balance of plant, including pumping, insulation, control devices, and packaging.
- Develop an advanced sensor and control system, including hardware and software design for metering and flow measurement, automation of operation, diagnostics, and data logging.

Results

Component Development and Testing

Steam Reforming Catalyst

A method was developed for placing InnovaTek's proprietary steam reforming catalyst on a foil support structure that is compatible with our microchannel reactor design. The catalyst was tested in a single microchannel reactor using either methane or diesel as the feed fuel for the reforming process. Results indicated that the catalyst was efficient for both fuel types, reforming methane at a high space velocity of 237,000 per hour and ultra-low-sulfur diesel (5 ppm) at 165,300 per hour (Figure 2).

Microchannel Reactor with Integrated Fuel Injector, Burner, and Heat Exchangers

A microchannel reactor was designed that was based on subunits consisting of chemically etched, diffusion-bonded shims that are stacked to achieve a scaled-up device. Each subunit consists of five shim types, and our reactor was composed of 20 stacked 5-shim subunits. A fuel injector and burner were

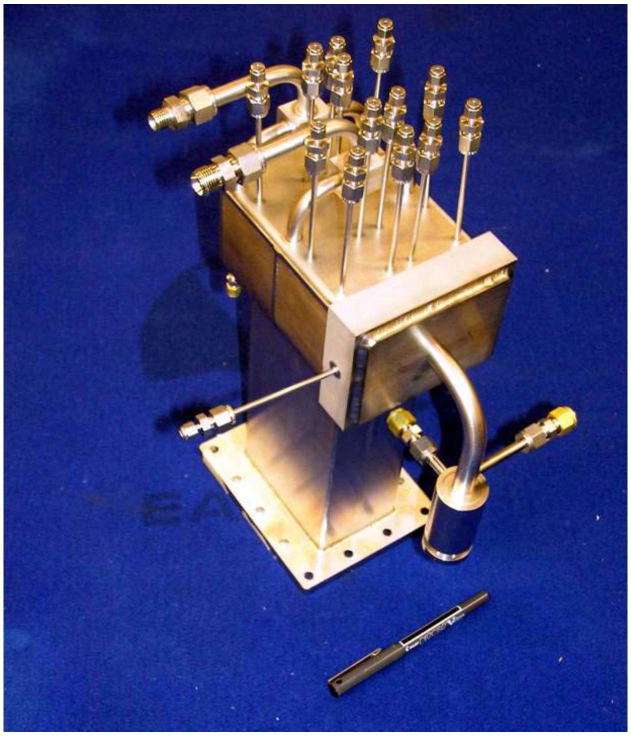

Figure 3. Integrated Components of the InnovaGen™ Fuel Processor Including Microchannel Steam Reformer, Fuel Injector, and Burner (A number of temperature and pressure sensors are also integrated in the reformer.)

manifolded to the reactor (Figure 3). The fuel injector provided atomization of the liquid fuel and thorough mixing of fuel and steam at the entry to the reactor. The burner combusted raffinate (off-gas from the membrane) to provide heat to the cross-flowing channels for the endothermic steam reforming reaction. Three different heat exchangers provided the correct temperatures for various zones within the system. This reactor is capable of producing as much as 50 liters per minute of hydrogen in the reformate stream.

Membrane-Based Hydrogen Purifier

A method was developed for treating commercially available Pd-alloy foils to reduce the thickness from 25 micrometers to 12.5 and 9 micrometers. These thin foils were then supported and sealed into a membrane subunit plate that was tested with reformate to determine efficiency for hydrogen purification. Test results indicated that the hydrogen permeation rate was proportional to the membrane thickness and the partial pressure of

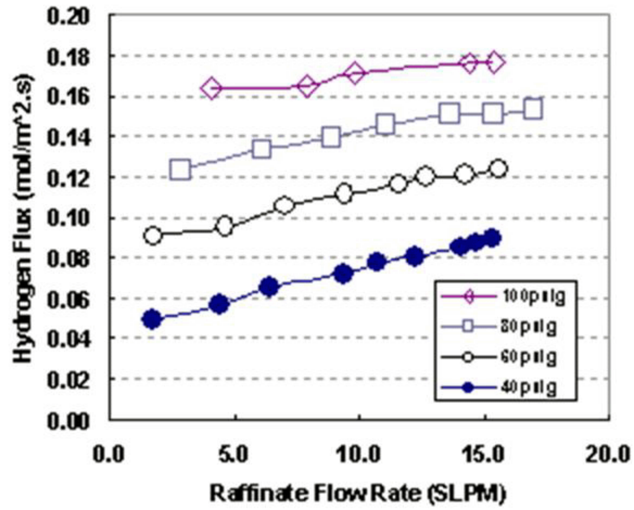

Figure 4. Hydrogen Flux versus Raffinate Flow Rate from a Feed Stream of Simulated Reformate at Four Different Pressures through a 12.5 µM Pd-Alloy Membrane

reformate (Figure 4). A scaled-up membrane module is prepared by stacking subunit plates and manifolding them.

System Integration

Initially, the reformer components were integrated and tested without the membrane purifier to examine performance of this subsystem. The components were connected with pumps, valves, tubing, and temperature and pressure sensors and housed in a mobile cabinet with an electronic control module (Figure 5). For the final system demonstration, the membrane module will be included and all components will be integrated according to the simplified schematic in Figure 1. The final prototype demonstration is scheduled for September 2004.

Prototype Operation and Testing

We performed a series of tests of the initial prototype using either methane or diesel fuel and examined performance under various conditions of pressure, temperature, and fuel flow rate. With methane, the reformer was operated at fuel flow rates to produce hydrogen sufficient for 1.0- and 2.5-kWe PEM fuel cells. With diesel, the system was operated at a rate sufficient for a 1.0-kWe PEM fuel cell. During these tests, increased understanding was obtained for catalyst, fuel injector, microchannel

Figure 5. Mobile Demonstration Platform That Houses All System Components, Pumps, Valves, and Control System Functions of the InnovaGen™ Fuel Processor

reactor, and condenser operations and performance in an integrated system. The use of a microchannel reactor greatly improved system efficiency and space velocity, which allowed a more compact system size. In addition, although the catalytic steam reforming channels and heat exchanger channels were micro-sized, there was very little pressure drop (<5 psi) in the reformer. Another finding was that the fuel injector is a critical component that can help eliminate coking, one of the most serious problems associated with reforming heavy hydrocarbon fuels.

Conclusions

- Microstructured components, especially an integrated system of catalytic and heat exchange microchannels, produce a compact, thermodynamically efficient fuel processor design.
- Catalytic microchannel reactors can be designed to have very low pressure drop when an appropriate method for catalyst loading is used.
- Performance tests of InnovaTek's proprietary steam reforming catalyst, conducted in a 100-W test bed and a 1-kW system, indicate that the catalyst can be used for multiple fuel types without the need for prior sulfur removal, although periodic regeneration and higher temperatures are required at higher sulfur concentrations.
- Reformate can be purified to 99.995% hydrogen through the use of supported ultra-thin Pd-alloy films that are sealed in a support framework. These modules are much smaller than the alternative water gas shift and preferential oxidation reactors.
- Coking, the primary reason for failure of reformers for complex hydrocarbon fuels, can be avoided through the use of a fuel injection system that thoroughly atomizes and mixes fuel and steam into a homogeneous mixture.

FY 2004 Publications/Presentations

1. Patricia Irving, Quentin Ming, Andrew Lee, and Trevor Moeller, "Hydrogen Production Using Novel Micro-Technology for Fuel Processing", In: Proceedings of the 15th Annual U.S. Hydrogen Conference, Los Angeles, April 27-30, 2004.

2. Jeffrey Harrison, Andrew Lee, Quentin Ming, and Y. Peng, "Demonstration of the InnovaGen™ Diesel and Logistical Fuel Processor", In: Proceedings of the 15th Annual U.S. Hydrogen Conference, Los Angeles, April 27-30, 2004.

3. Patricia Irving, Trevor Moeller, and Quentin Ming, "Hydrogen Production from Heavy Hydrocarbons Using a Fuel Processor with Micro-Structured Components", In: Proceedings of the 2003 Fuel Cell Seminar, Miami Beach, November 3-6, 2003.

4. Quentin Ming, Andrew Lee, Redwood Stephens, and Tony Dickman, "Development of a Diesel Fuel Processor for PEM Fuel Cells", In: Proceedings of the 2003 Fuel Cell Seminar, Miami Beach, November 3-6, 2003.

5. Patricia Irving, Jeffrey Harrison, Quentin Ming, Andrew Lee, Trevor Moeller, "The InnovaGen™ Diesel Fuel Processor", In: Proceedings of the 4th Department of Defense Logistical Fuel Reforming Conference, Philadelphia, October 22-23, 2003.

6. Patricia Irving, Quentin Ming, and Trevor Moeller, "Hydrogen Production Using the InnovaGen™ Fuel Processor", Presented at the NW Hydrogen Conference, Seattle, June 16, 2003.