

VII.F.6 Fuel Processors for PEM Fuel Cells

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Objectives

- Demonstrate high-performance desulfurizer, catalyst, microreactor and microcombustor/microvaporizer concepts that will enable production of compact fuel processors for proton exchange membrane (PEM) fuel cells.
- Design, fabricate and evaluate a 1-kW_e fuel-flexible (including EPA Phase II reformulated gasoline) fuel processor (during first 36 months).
- Design, fabricate and evaluate a fuel-flexible (including EPA Phase II reformulated gasoline) fuel processor capable of producing up to 10 kW_e hydrogen.

Technical Barriers

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

- A. Durability
- B. Cost
- J. Startup Time/Transient Operation

Technical Targets

The Fuel Cell team conducted a review of on-board fuel processing for transportation applications and, in August 2004, decided to discontinue on-board fuel processing research and development. Specific criteria for the on-board fuel processing decision are shown below along with the status of our project. Following the decision to discontinue on-board fuel processing research, we were encouraged to investigate the use of our systems and materials to convert natural gas into hydrogen-rich gas for stationary PEM fuel cells.

Attribute	Units	2004 Demo Criteria	Current Status
Energy efficiency	%	78	70
Power density	W/L	700	500*
Specific power	W/kg	700	500*

* Estimated from values for breadboard components.

Approach

Tasks devised to accomplish the project objectives include:

- Design and model components and systems that would meet power density targets.
- Develop high-capacity sorbents capable of removing sulfur to target levels.
- Develop better-performing autothermal reforming (ATR), water-gas-shift (WGS) and preferential oxidation (PrOx) catalysts and associated microreactors.
- Fabricate highly efficient microcombustors/microvaporizers.
- Fabricate microreactors and systems.
- Evaluate components and fuel processing systems.
- Estimate cost for fuel processor.

Accomplishments

- Demonstrated adsorber prototype containing high-capacity, reversible sulfur sorbents that significantly out-performed other available sorbents. The capacity exceeded $5 \text{ mg}_{\text{sulfur}}/\text{gr}_{\text{sorbent}}$ for gasoline and $10 \text{ mg}_{\text{sulfur}}/\text{gr}_{\text{sorbent}}$ for diesel.
- Demonstrated prototype fuel and water microcombustors/microvaporizers. The prototype incorporated a 50- μm flash channel and catalytic tailgas-surrogate burner and achieved the target vaporization rate of 8 mL/min.
- Demonstrated durable ATR, WGS and PrOx prototype microreactors containing metal foam supported catalysts. These catalysts significantly out-performed available commercial catalysts.
- Assembled and tested microreactor-based iso-octane fuel processors. Achieved an efficiency of 70% with 100- W_e prototype modules.
- Designed and constructed thermally integrated natural gas fuel processor.

Future Directions

- During the remainder of the project, we will continue developing the microreactor-based fuel processors, explore their effectiveness in processing natural gas, and evaluate a newly constructed thermally integrated natural gas fuel processor.

Introduction

Fuel cells are being developed to power cleaner, more fuel-efficient automobiles and for small distributed power requirements. The fuel cell technology preferred for these applications is PEM fuel cells operating with H_2 from liquid fuels like gasoline, diesel fuel, and propane, and from natural gas. A key challenge is the lack of sufficiently small and inexpensive fuel processors. Improving the

performance and cost of the fuel processor will require the development of better-performing catalysts, new reactor designs and better integration of the various fuel processing components.

Approach

Prototype gasoline fuel processors have been produced and evaluated against the Department of Energy technical targets. Significant improvements

over the present state-of-the-art were achieved by integrating low-cost microreactor systems, high-activity catalysts, π -complexation sorbents, and high-efficiency microcombustors/microvaporizers being developed at the University of Michigan. The microreactor system allows (1) more efficient thermal coupling of the fuel processor operations, thereby minimizing heat exchanger requirements; (2) improved catalyst performance due to optimal reactor temperature profiles and increased heat and mass transport rates; and (3) better cold-start and transient responses.

The project was accomplished in 3 phases. The Phase I effort focused on demonstrating compact desulfurizer, microreactor and microcombustor/microvaporizer components, and developing low-cost methods for the production of microchannel systems. These components provided the basis for design and fabrication of an integrated gasoline fuel processor during Phase II. During the final phase, work focused on evaluating performance of the integrated fuel processors. Following the decision of the Fuel Cell team, we investigated the use of our systems and catalysts to convert natural gas into hydrogen-rich gas for stationary PEM fuel cells.

Results

High-performance components described in earlier reports were integrated into a breadboard fuel processor and evaluated for the conversion of iso-octane into hydrogen-rich gas. The vaporizer/combustor consisted of a 50- μm flash channel mated with a catalytic tailgas-surrogate burner and achieved the target vaporization rate of 8 mL/min. The microreactor components incorporated FeCrAlY metal foam (80 ppi) supported Ni/Ce_{0.75}Zr_{0.25}O₂ (ATR), Au/CeO₂ (WGS) and Pt/Al₂O₃ sol-gel derived (PrOx) catalysts. A schematic of the 100-W_e breadboard fuel processor module is illustrated in Figure 1. Liquid H₂O (0.69 ml/min) and iso-octane (0.4 ml/min) were delivered using high-pressure liquid chromatography pumps and vaporized in heated lines with air (1 l/min). Nominal operating temperatures for the ATR, high-temperature shift (HTS), low-temperature shift (LTS) and PrOx microreactors were 650, 340, 290 and 185°C, respectively. The effluent streams were analyzed

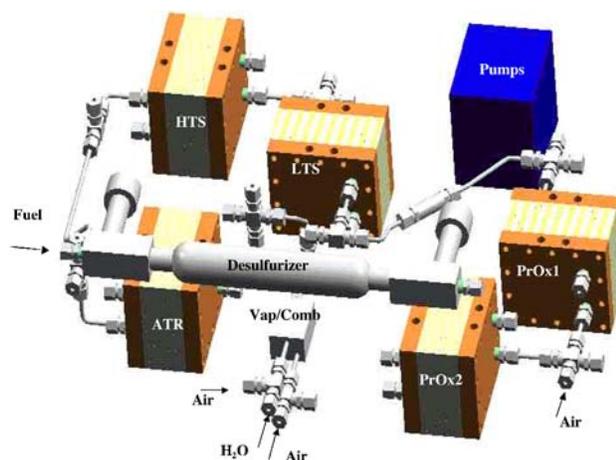


Figure 1. Schematic of Microreactor-Based Iso-Octane Fuel Processor

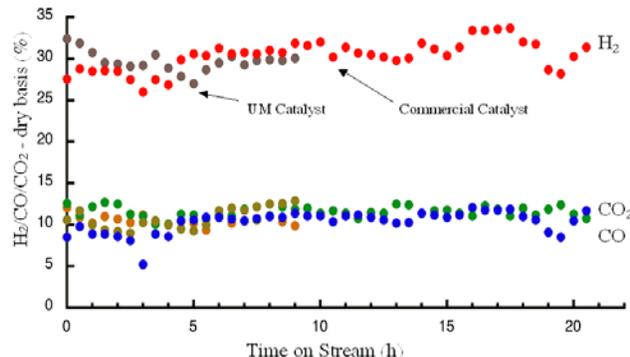


Figure 2. Performance of the ATR Microreactor Component of the Iso-Octane Fuel Processor. Liquid H₂O (0.69 ml/min) and iso-octane (0.4 ml/min) were delivered using HPLC pumps and vaporized in heated lines with air (1 l/min). The nominal operating temperature for the ATR microreactor was 650°C.

using a gas chromatograph and a non-dispersive infrared CO analyzer.

To illustrate the performance of individual components in the fuel processor, the microreactors were engaged sequentially starting with the ATR. Figure 2 compares the performance of systems incorporating the Ni/Ce_{0.75}Zr_{0.25}O₂ catalyst developed at the University of Michigan and a commercial ATR catalyst. The performance was comparable for the two catalysts; however, the pressure drop became excessive for the Ni/Ce_{0.75}Zr_{0.25}O₂-containing microreactor after approximately 5 hours on stream. Subsequently, the

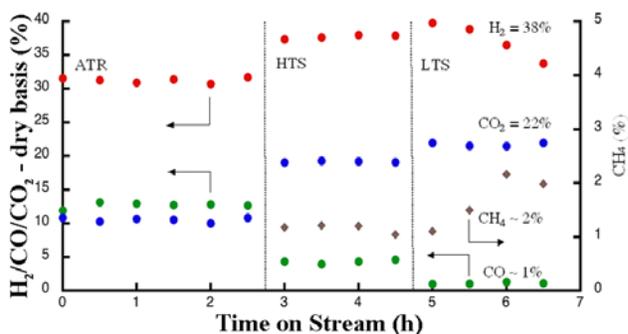


Figure 3. Performance of the ATR, HTS and LTS Microreactor Components of the Iso-Octane Fuel Processor. Liquid H₂O (0.69 ml/min) and iso-octane (0.4 ml/min) were delivered using HPLC pumps and vaporized in heated lines with air (1 l/min). The nominal operating temperature for the ATR, HTS and LTS microreactors were 650, 340 and 290°C, respectively.

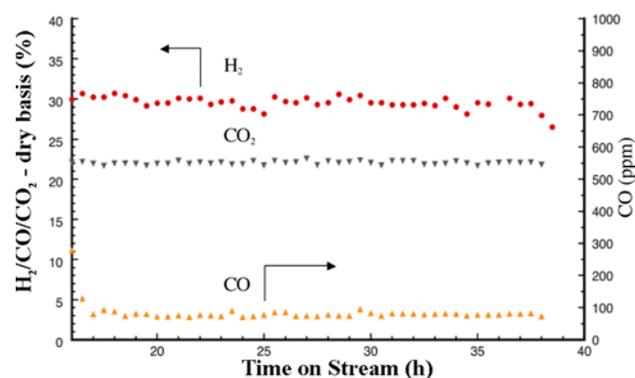


Figure 4. Performance of the Iso-Octane Fuel Processor. Liquid H₂O (0.69 ml/min) and iso-octane (0.4 ml/min) were delivered using HPLC pumps and vaporized in heated lines with air (1 l/min). The nominal operating temperature for the ATR, HTS, LTS and PrOx microreactors were 650, 340, 290 and 185°C, respectively.

commercial ATR catalyst was used. The CO content was reduced to <1% after the two-stage WGS microreactors were engaged (Figure 3). This level of performance exceeded the design. Engaging the single-stage PrOx microreactor enabled the production of hydrogen-rich gas with approximately 80 ppm CO (Figure 4). The two-stage PrOx produced H₂-rich gas with less than 10 ppm CO.

The breadboard fuel processor functioned steadily for nearly 40 hours. Table 1 compares the performance of the system and microreactor components after 6 and 40 hours on stream. The initial performance was promising with the system producing 142 W of H₂ at an efficiency of 68%. After 40 hours on stream, the performance degraded and the system produced 107 W of H₂ at an efficiency of 51%. This degradation in performance was associated with methane slip through the ATR.

We developed a simple natural gas fuel processor concept that essentially consists of a counter-flow heat exchanger into which a series of fuel processing catalyst beds were placed. A premixed reactant stream flows inside the heat exchanger. Since the heat recovered from the fuel processing reactions is not sufficient to bring the reactants to the desired temperature prior to the reforming section, additional heat must be supplied using an anode tail gas combustor (Figure 5). To control the temperature profile, the vapor fraction of the premixed stream can be changed before it enters the fuel processor using the tail gas combustor. This enables the fuel processor exhaust to remain at a nearly constant temperature while temperatures of the internal beds are controlled. This system has been constructed and will be evaluated during the balance of the project.

Table 1. System Performance

Hours on Stream	ATR		HTS		LTS		PrOx	
	6 hr	40 hr	6 hr	40 hr	6 hr	40 hr	6 hr	40 hr
H ₂	30%	22%	38%	30%	40%	32%	38%	30%
CO	12%	12%	4%	4%	1%	1%	80 ppm	80 ppm
CO ₂	11%	11%	19%	19%	22%	22%	22%	22%

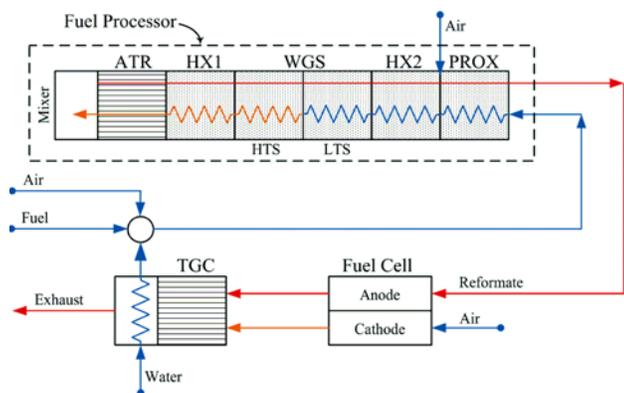


Figure 5. Schematic Diagram of Thermally Integrated, Natural Gas Fuel Processor

Conclusions

Significant progress was made over the last year. Fully integrated prototype devices were demonstrated. Efforts for the balance of the project will focus on using these devices, materials and know-how to convert natural gas into hydrogen-rich gas for stationary PEM fuel cells.

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