VII.F.9 Forecourt Fuel Processing: Micro-Channel Steam Reforming of Natural Gas for Distributed Hydrogen Production

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Objectives

Develop thermally efficient/low-capital-cost system for distributed hydrogen production from natural gas.

- Adapt the micro-channel steam reformer technology, initially developed for on-board reforming of gasoline, to distributed steam reforming of natural gas.
- Demonstrate at small scale (5 kg/day) a compact, efficient, integrated hydrogen production system.
- Provide basis for initiating evaluation of reduced-cost manufacture of micro-channel hardware for 1500 kg/day system.
- Advance technology through interactions.

Technical Barriers

This project addresses the following technical barrier 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

Technical Targets

This project addresses the technical target for hydrogen production energy efficiency from Table 3.1.2 of the Multi-Year Research, Development and Demonstration Plan as shown below:

	2005 Target	2010 Target
Production Energy Efficiency ^a	69%	80%

^adefined as (LHV H_2 produced)/(LHV fuel consumed + any other power inputs)

Approach

- Adapt on-board gasoline reformer to forecourt natural gas application. Design factors that change include:
 - Pressure: 200 psig vs. <50 psig
 - Less critical factors startup time, startup energy, weight, volume
 - More critical factors thermal efficiency, system durability

- Retain micro-channel design concept:
 - Rapid heat and mass transport
 - Small quantity of highly active catalyst
 - Efficient heat transfer enabling tight thermal integration
 - Cost reduction through reduction in overall system size and materials
- Design for 2-pass reformer system:
 - Increased temperature change on combustion stream passing through the reformer reduces air flow and improves efficiency
 - Reacting reformate stream is recuperated between reforming stages
- Fabricate/demonstrate compact system at 5 kg/day capacity.

Accomplishments

- Mass and energy balance simulations completed for two system flowsheets, one using pressure-swing adsorption (PSA) for H₂ purification and one using a palladium membrane.
- Iterated on flowsheet calculations using actual hardware designs to arrive at efficient flowsheets consistent with expected hardware performance.
- Target efficiencies for the two flowsheets calculated.
- Designed micro-channel components in the steam reforming system.
- Fabrication of individual components completed.
- Reformer system assembly and test preparations underway.
- Numerous interactions underway.

Future Directions

- Remainder of FY 2005: Test unit to demonstrate thermal efficiency.
- In FY 2006:
 - Evaluate scale-up to 1500 kg/day system.
 - Explore fabrication techniques and/or design changes to reduce cost for producing laminated microchannel structures.
 - Estimate fabrication cost of 1500 kg/day systems when produced in a quantity of 100s/year.
 - Decision point at end of FY 2006: Down-select research for distributed hydrogen production from natural gas.

Introduction

This project is applying the rapid heat and mass transfer attainable in micro-channels to the development of a highly compact and efficient steam reformer-based hydrogen production system. Once deployed, the steam reformer will convert natural gas to hydrogen at the refueling station to refuel hydrogen fuel cell vehicles. The current year's effort is focused on demonstrating a high thermal efficiency for the production of hydrogen from natural gas by designing, building and operating a small-scale (5 kg/day) prototype system. The prototype system is based on a steam reformer, a water-gas shift reactor and a palladium membrane. Within the steam reformer, the methane reacts with steam in the reaction:

$$CH_4 + H_2O \rightarrow CO + 3H_2$$

Additional hydrogen is then produced via the water gas shift reaction:

$$CO + H_2O \rightarrow CO_2 + H_2$$

A copper-palladium membrane is then used to separate high-purity hydrogen from the gas mixture.

The remaining gas, which includes residual H_2 , CO, CH_4 , unreacted H_2O and CO_2 , is then utilized as fuel to maintain the reforming reaction and generate the required steam.

This project builds upon prior work aimed at reforming gasoline on-board vehicles. Many of the components developed for that system have application in the current effort, including reactors, vaporizers and recuperators designed for low pressure drop which minimize the parasitic power consumption for air movement. Additional information on prior development of the technology is provided in references 1, 2 and 3.

Approach

The micro-channel steam reformer under development for on-board reforming has been re-designed for the stationary vehicle refueling application. The stationary application results in a greater emphasis on efficiency and durability with less emphasis on start-up time, system size and weight. The current system will utilize a conventional (iron-chrome based) high-temperature shift reactor and a copper-palladium membrane separator to produce high-purity hydrogen. A prototype system is being designed and tested at a 5 kg/day scale. The system is now designed to operate at 200 psi in order to provide a pressure driving force for purification. The steam reforming reactor itself has retained the low-pressure-drop "panel" design due to its advantages with respect to minimizing parasitic power consumption for air movement. In order to increase efficiency, two rows of panels, one high temperature, one low temperature, each with a dedicated reformate recuperator, are being used. The combustor uses a micro-channel mixer for fuel mixing but now includes a catalytic element to assure complete methane utilization. The catalyst also provides the initial ignition in place of the spark-ignition system used previously. The intent of the current year's effort is to operate the small-scale system to demonstrate high thermal efficiency for the production of hydrogen from natural gas.

<u>Results</u>

Reforming Flowsheet Simulations

Flowsheets were simulated for two flowsheet designs, one using a palladium membrane separator and one using a PSA unit. Calculations were performed using the ChemCad software package for mass and energy balances and using in-house models for micro-channel component thermal performance. The flowsheets were optimized in order to achieve ~80% thermal efficiency (2010 goal) in reasonablysized hardware. Simulations used methane as a surrogate for natural gas in order to be consistent with the planned test operation.

The membrane-based system assumed a hydrogen recovery of up to 90%. This level of recovery has been observed in a related system, and the membrane for the test system has been designed to be capable of this level of recovery. The steam:carbon ratio was selected at 2.8:1 to assure that equilibrium carbon formation is not predicted at any point in the system. The point of greatest concern is near the outlet of the membrane retentate where the hydrogen content of the stream has been largely depleted. Simulations examining the inclusion of water gas shift (WGS) demonstrated that overall system efficiency could be improved by 1.3% if a high-temperature shift bed is included, with the added benefit that the reforming temperature could be reduced from 865°C to 800°C, which will favorably impact system durability. As a result, an Fe-Cr based catalyst, WGS bed was included between the reforming system and the membrane separator. Low-temperature shift showed no advantage with respect to thermal efficiency.

In order to provide a fair comparison between the membrane and PSA systems, the efficiency of the membrane system was downgraded to account for the energy cost to recompress the hydrogen to 180 psig from a near-ambient-pressure permeate stream. Including compression reduces the overall efficiency by 2.7%. The flow logic of the membrane-based flowsheet is illustrated in Figure 1.



Figure 1. Flow Logic for the Membrane-Based Flowsheet and Experimental Test System

The PSA flowsheet examined is similar except that PSA is used for the separation and purification of the hydrogen. Unlike the membrane system, the PSA-based system retains the hydrogen outlet at a pressure near the feed pressure (assumed to be 180 psi for a 200-psi feed pressure), which is a potential efficiency advantage. However, the PSA system must cool the reformate stream to near ambient temperature, dropping out much of the water, before the stream can undergo separation. As a result, the retained gas routed to the combustor is cool, which is a disadvantage with respect to efficiency. The combustor for the PSA system would need to be carefully designed to avoid coking at the fuel inlet since the steam and most of the hydrogen have been removed from this stream. For modeling purposes, it was assumed that a PSA could meet the required product purity with up to 90% recovery of hydrogen in an economic size. However, specific sizing of a PSA unit was not performed.

The PSA flowsheet provided a predicted efficiency of 80%. The membrane-based flowsheet provided an efficiency of 81.0% without hydrogen compression and 78.4% after accounting for compression of hydrogen to 180 psig for comparison to the PSA efficiency. Both results are close to the 2010 target of 80%.

5 kg/day Prototype System

The design and fabrication of individual components is complete, and the assembly of the



Figure 2. Steam Reforming Micro-Channel Reactor Panels for 5 kg/day System (When installed, these panels have a combined thermal duty of 1.8 kW with 1.2 in. H₂O pressure drop with a total weight of 1220 g. These reactors are fabricated in Inconel 625.)



Figure 3. Air Recuperator for 5 kg/day System (This heat exchanger provides 90% thermal effectiveness with a pressure drop that is 3.0 in. H_2O on the hot side and 1.9 in. H_2O on the cold side. Effective air recuperation with low pressure drop (and hence low parasitic power for air movement) is a critical factor in achieving efficient operation.)

prototype system is very nearly complete. Selected individual components are shown in Figures 2, 3, and 4 with the assembled test system in Figure 5. As of this writing, test data has not yet been collected.

Scale-up to 1500 kg/day

The raw material costs for a 1500 kg/day system were estimated based on the construction of the



Figure 4. Micro-Channel Water Vaporizer for 5 kg/day System (This heat exchanger produces superheated steam from water feed in a single step with very low air-side pressure drop.)



Figure 5. Assembled 5 kg/day System (The membrane separator and WGS reactor are not included in this photo. The system includes ¹/₂-inch of insulation inside the shell but will be insulated externally as well.)



Figure 6. President Bush and Energy Secretary Sam Bodman are Briefed on the Bradley Fuel Cell APU System (Micro-channel components developed under EERE funding were an enabling factor for development of this prototype system.)

5 kg/day prototype. The high-value materials include the rhodium metal for the reforming catalyst (25,000 @ 63.60/gram), the Inconel for the reactors and recuperators (23,570 @ 17.00/lb), and the stainless steel (2,870 @ 2.31/lb). Based on this, it is clear that the key to capital cost feasibility will be the costs added during fabrication rather than the starting raw materials. A conceptual layout for the system was developed which fits within a 4 ft x 5 ft x 6 ft footprint.

Interactions

Micro-channel heat exchanger and reactor technology developed under DOE Office of Energy Efficiency and Renewable Energy (EERE) funding has resulted in a number of interactions. These include an interaction with Battelle Memorial Institute/United Defense Limited Partnership/U.S. Army National Automotive Center in which a fuel cell auxiliary power unit (APU) was built for the Bradley Fighting Vehicle (Figure 6). Other interactions include providing lightweight cryogenic heat exchangers to a DOE/NA-22 national security project, a vaporizer loaned to Catalytica to support development of their steam reforming system, and a vaporizer loaned to Idatech for evaluation. In addition, technology developed with DOE-EERE funding has been applied to the construction of a highly efficient prototype economizer for a 3 MMBTU/hr boiler system and is currently being applied to Air Force work involving fuel processing for a solid oxide fuel cell-based power system.

Conclusions

Flowsheet simulation calculations along with heat exchanger and reactor design calculations have provided two flowsheets which approximately meet the 2010 target efficiency of 80%. A prototype system has been designed and built and is being readied for testing. Based on the small-scale prototype, the feasibility of the system with respect to capital cost will be largely dependent on costs added during fabrication rather than raw material costs. The technology developed under EERE funding is finding applications in other areas.

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