

II.A.7 Low-Cost Hydrogen Distributed Production System Development

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Subcontractors:

Süd Chemie, Inc., Louisville, Kentucky
Naval Research Laboratory, Washington, D.C.

Start Date: July 1, 2005

Projected End Date: September 30, 2008

value of fuel cell-grade hydrogen out/lower heating value of fuel in + electricity in): 70%

- Distributed hydrogen from natural gas cost: \$2.50/kg (production capital cost component: \$0.47/kg)
- Distributed hydrogen from ethanol cost: \$3.60/kg (production capital cost component: \$0.50/kg)

Accomplishments

- Designed, fabricated and tested improved boiler system.
- Tested new steam methane reformer (SMR) and water-gas-shift (WGS) catalysts, including 1,000-hour durability testing.
- Best catalysts field-tested for over 2,500 hours each in two HGM-2000s.
- Built two PSA test rigs to characterize adsorbents.
- Screened and down-selected improved adsorbents that increased pressure swing adsorption (PSA) recovery.
- Began initial design of major HGM-10,000 components.
- Obtained tax-free ethanol permit and ordered various grades of ethanol for SMR tests.

Objectives

- Design, build and test a steam methane reformer system that will achieve the DOE cost and efficiency targets for 2015.
- Demonstrate the efficacy of a low-cost renewable hydrogen generation system based on distributed production of hydrogen from ethanol.

Technical Barriers

This project addresses the following technical barriers from the Production section (3.1.4.2.1) of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- (A) Fuel Processor Capital Costs
- (B) Fuel Processing Manufacturing
- (C) Operation and Maintenance (O&M)
- (D) Feedstock Issues
- (E) Carbon Dioxide Emissions
- (F) Control and Safety

Technical Targets

- Total production system (steam methane reformer + pressure swing adsorption) efficiency (lower heating

Introduction

Achieving the DOE cost targets will require improved efficiency and also larger hydrogen capacity compared to the current HGM-2000 system (2,000 scfh or 113 kg/day capacity). We are also reducing catalyst cost while improving SMR, shift and PSA performance and increasing feedstock diversity.

Approach

We are using the existing HGM-2000 as a test bed to develop improved SMR and PSA components, including improved catalysts and PSA adsorbents. The best components are then tested in full-scale field operation. Those improvements that pass these real-world tests will then be incorporated into the design of the HGM-10,000 system.

We will build and test the key components of the HGM-10,000 at our plant. After component testing, the final system will be assembled and tested in the field. Since the output of this HGM-10,000 is within a factor of three of distributed generation system (1,500 kg/day) used in setting the DOE technical targets, full-scale field

testing of the HGM-10,000 should provide the DOE with a reliable measure of potential distributed hydrogen costs.

We originally planned a ground-up design of the HGM-10,000, optimizing all major components. To cut costs with reduced DOE funding, however, we now plan to use many components from the HGM-2000 in the HGM-10,000. For example, we will use the exact diameter and length reformer tubes in the large SMR, rather than conducting optimization studies with diameter and length as parameters. Another design, build, test iteration may be necessary later to meet DOE targets.

Results

Results to date are reported in five areas:

- Improvements in HGM-2000 system components
- Advances in SMR and WGS catalysts
- Advances in PSA systems
- Initial design of the HGM-10,000
- Initial planning for the ethanol reforming task

Improved HGM-2000 System Components

We added a recuperator that transferred 2.1 kW of heat (at 2,000 scfh output) from the reformato to the incoming natural gas. We also built and tested two generations of boilers for the HGM-2000 to further improve heat recovery.

These improvements (along with improved PSA recovery and catalyst improvements described below) increased the HGM-2000 output from its design capacity of 2,000 scfh to over 2,400 scfh, as shown in Figure 1.

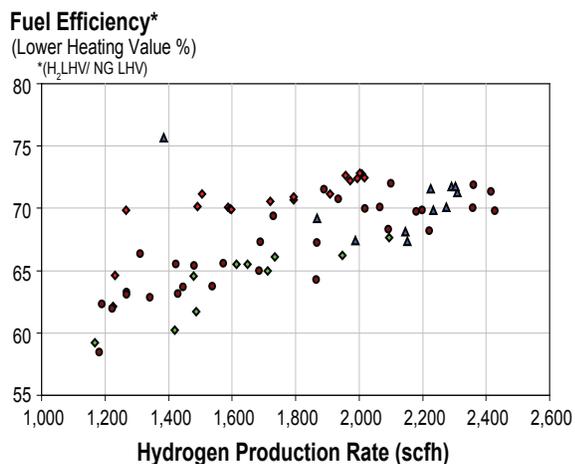


FIGURE 1. Fuel efficiency versus hydrogen output from four different HGM-2000 systems.

The SMR and PSA fuel efficiency (lower heating value of hydrogen/lower heating value of natural gas in) reached 72% for these hydrogen production levels from four different HGM-2000 systems. Note that each datum point in Figure 1 represents the efficiency and hydrogen production level averaged over a 20 to 30 minute time interval for full-scale, automated equipment operating 24 hours per day. Adding the electrical consumption to the denominator of the efficiency calculation reduces *system* efficiency to 68%, just below the DOE target of 69% system efficiency (including electricity) for 2005 and 70% by 2010.

Advances in SMR and WGS Catalysts

We reduced SMR catalyst costs by decreasing the active material loading. Süd Chemie prepared and we conducted a series of screening tests with the original active material loading, and also samples with half, one-quarter, and one-eighth of the original level. These catalysts were tested in single SMR reactor tubes and then aged for approximately 1,000 hours at simulated SMR conditions.

The activities of these aged catalysts are plotted in Figure 2, showing that catalysts with both one-half and one-quarter of the original active material have sufficient activity (above 5 SLPM/meter of tube length of conversion required in the 500 to 900°C range). We conclude that the active material can be reduced by a factor of four without adversely affecting SMR operation, cutting catalyst cost by 35%.

Four new WGS catalysts were also evaluated. Two pairs of SMR and WGS catalyst formulations that demonstrated promising performance in the short term testing were further subjected to a 1000-hour aging test.

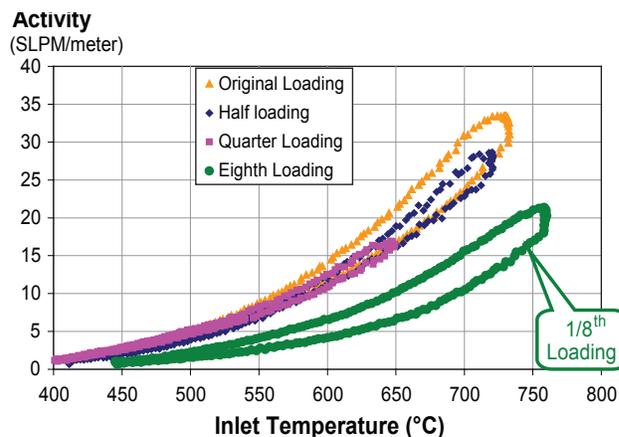


FIGURE 2. Activity of aged SMR catalyst samples with original active material loading, one-half, one-quarter and one-eighth of the original loading.

PSA Advances

The PSA development activities included: adsorbent screening, improved PSA cycles, and improved PSA computer simulations.

We developed two test stands to conduct PSA adsorbent screening in the laboratory: an isotherm characterization unit (ICU) and an adsorption test stand (ATS). These devices measure the adsorption capacity and mass transfer characteristics of adsorbent materials. They were used to screen different activated carbon adsorbents and have identified one material that exhibits an improvement in the adsorption capacity and mass transfer properties.

Alternative molecular sieve adsorbents have also been examined. We completed mass transfer characterization of one promising molecular sieve zeolite on the ATS, but have not yet completed ICU testing.

We have changed the PSA operating cycles from two to three pressure equalization cycles, increasing hydrogen recovery approximately 3 to 5%. We expect that the combination of three equalization cycles with the improved adsorbent materials described above will push the hydrogen recovery number into the high 80 percent range.

We have further expanded, improved and validated our ability to simulate PSA performance with a complex in-house PSA computer simulator. We can now evaluate various PSA operating parameters mathematically and optimize for maximum PSA performance without the need to run time-consuming experiments. We have validated our in-house PSA simulator against data sets obtained from ATS experiments in the micro-reactor room to the HGM-2000 scale. We can now match PSA performance numbers for product flow, purity and recovery to within 10% of the experimental values.

Initial HGM-10,000 Work

We have completed preliminary designs for these system components:

- Burner
- Steam generator
- Condenser
- Air preheater
- Reactor

During 2006 we procured a low pressure drop burner for the HGM-10,000 system. By reducing pressure drop, we will be able to significantly cut the operating cost of the system air blower.

Initial Ethanol Work

We have obtained a tax-free permit from the U.S. Department of the Treasury to conduct initial experiments reforming ethanol. We have also made arrangements through the Energy and Environmental Research Center (EERC) at the University of North Dakota to obtain samples of ethanol from various stages of the Chippewa Valley Ethanol Plant. We will be exploring whether lower grades of ethanol are sufficient for making pure hydrogen.

Conclusions and Future Directions

Based on initial work improving and field-testing the HGM-2000 and the initial designs of the HGM-10,000, we conclude that the DOE 2010 cost target of \$2.50/kg for fuel cell-grade compressed hydrogen is potentially feasible.

Future work will include:

- Continue field testing of the HGM-2000 to establish reliability under real world operating conditions at full output capacity.
- Continue the evaluation and laboratory testing of advanced SMR and WGS catalysts and PSA adsorbents to improve efficiency and PSA hydrogen recovery.
- Complete the design of key HGM-10,000 components.
- Build and field-test the HGM-10,000 and demonstrate 1,000 hours operation at full capacity in the field.
- Demonstrate material costs for the HGM-10,000 system less than \$350,000.
- Conduct 1,000-hour laboratory tests of ethanol reforming catalysts.

FY 2006 Publications/Presentations

1. Presentation to the 2006 DOE Annual Peer Review meeting, May 18, 2006.