II.A.1 Autothermal Cyclic Reforming Based Hydrogen Generating and Dispensing System

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Praxair, Tonawanda, NY
University of California, Irvine, CA

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Objectives

- Develop a reformer-based hydrogen generating system capable of delivering 40 kg/day of hydrogen.
- Produce fuel cell vehicle grade hydrogen (99.99+% purity; < 1 ppm CO).
- Achieve 69% hydrogen generation efficiency (lower heating value basis).

Technical Barriers

This project addresses the following technical barriers from the Production section (3.1.4.2) of the DOE Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(A) Fuel Processor Capital Costs
(C) Operation and Maintenance (O&M)
(D) Feedstock Issues
(F) Control and Safety
(L) Durability
(M) Impurities

Technical Targets

Parameter	Units	2005 Target	Current System Status
Production Energy Efficiency	%	69	71

Accomplishments

- Operated the high-pressure reformer (150 psig) and pressure swing adsorption (PSA) unit at University of California, Irvine (UCI) for extended periods.
- Generated 99.999% pure hydrogen.

Introduction

The hydrogen generation system uses GE's proprietary autothermal cyclic reforming (ACR) technology to convert natural gas to a hydrogen-rich gas that is purified downstream. The hydrogen generation system is designed for vehicle refueling. The generating system includes Praxair's PSA unit to purify the hydrogen. The basics of the ACR process and its advantages over the conventional reforming processes have been discussed in the 2003 annual report [1].

Approach

The project is broken down into three phases:

- Phase I - Conceptual Design and Analysis
- Phase II - Sub-System Development
- Phase III - System Design, Fabrication, and Operation

During Phase I, a conceptual design of the entire ACR-based refueling system was developed. The system design, process flow diagrams and component design for the ACR reactors and other components were completed. An analysis was performed to determine the competitiveness of the design relative to alternative concepts.

In Phase II, the major task was experimental evaluation of small-scale ACR and PSA units.

A low-pressure pilot-scale reformer was operated and then the experimental data was used to design a high-pressure pilot-scale reformer. A bench-scale PSA unit has also been operated and then this data was used to design a pilot-scale PSA unit.

During Phase III, the high-pressure pilot-scale ACR and PSA units were fabricated, installed and operated at UCI. The ACR and PSA units were integrated and generated a 99.999% hydrogen stream.

Results

Shakedown of Pilot-Scale System

The pilot-scale system consists of the ACR unit, the shift reactor, and the PSA unit. A picture of the pilot-scale system is shown in Figure 1. After installation at UCI, the ACR unit was pressure tested and the safety systems were verified. A standard operating procedure including a safe shut-down procedure was developed and validated. In preparation for integration with the PSA unit, the reformer unit was operated in stages. This required first operating one ACR reactor, then two ACR reactors simultaneously, and then integration of the two ACR reactors with the shift reactor.

The shakedown of the PSA unit was performed at UCI. The beds were loaded and the cyclic operation of the pressurized PSA unit was verified. All safety systems were checked for functionality.

Pilot-Scale System Operation

The pilot-scale system uses a computer-based dynamic process control system to stabilize the cyclic process. The control system was first tested using a theoretical process model, which accounted for the chemical reaction kinetics in the catalyst bed. The control system was then experimentally validated in the pilot-scale system and was able to successfully stabilize the process for extended periods of time.

FIGURE 1. High pressure autothermal cyclic reforming (ACR) and PSA units. Installed at University of California, Irvine (UCI).

The pilot-scale ACR reactors were operated at 150 psig and the validation culminated in eight overnight runs with steady temperatures in both reactors and methane concentrations $< 5\%$ at the outlet of the reformer. In one of the runs, the reformer was operated for 60 hours continuously with only two shut-downs for a few minutes. The shift reactor was operated for 60 hours continuously with carbon monoxide levels at the outlet of the shift of less than 2% (dry concentration). Figure 2 shows the ACR reactor temperature during this 60-hour run. Figure 3 shows the typical gas concentrations at the outlet of the combined reformer and shift unit during the 60-hour run.

The ACR unit was integrated with Praxair's PSA unit and generated 99.999% pure hydrogen. This gas was suitable for filling of a fuel cell vehicle. The hydrogen compressor, storage, and dispenser systems were designed but not installed. However, the hydrogen

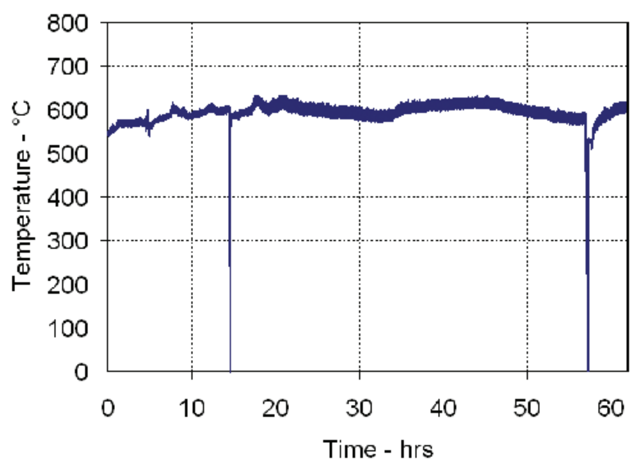

FIGURE 2. ACR reactor: 60 hours continuous operation. Shut downs: 1) Operator accidental shut down, and 2) Defective water flow meter was fixed.

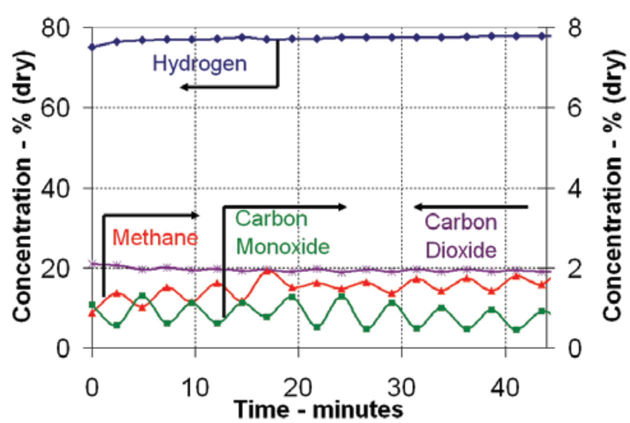

FIGURE 3. Gas concentrations at the outlet of the combined reformer and shift unit during cyclic operation.

gas was utilized in a stationary proton exchange membrane (PEM) fuel cell. The fuel cell was operated by UCI in a coordinated California Energy Commission project and generated 5 kWe.

The experimental data from the pilot-scale system was fed to the process model of the system and the projected efficiency was 71% on a lower heating value (LHV) basis. This efficiency was calculated by dividing the LHV of the hydrogen produced by the sum of the LHV of the methane fed and the electricity required to operate the system.

Conclusions and Future Directions

- The pilot-scale ACR and PSA units were successfully installed and integrated.
- The reformer and shift reactors generated 40 kg/day of hydrogen and were operated for 60 hours continuously with only two shut-downs for a few minutes.
- The ACR and PSA units generated 99.999% pure hydrogen.
- The project was completed in March of 2006.

References

1. Kumar R., Barge S., Kulkarni P., Moorefield C., Zamansky V., Smolarek J., Manning M., Baksh S., and Schwartz J., Autothermal Cyclic Reforming Based Hydrogen Generating And Dispensing System, DOE Annual Report 2003.

FY 2006 Publications/Presentations

1. Kumar R, "Autothermal Cyclic Reforming and H_2 Refueling System," DOE Project Review, Arlington, VA, May 2006.

Special Recognitions & Awards/Patents Issued

1. Patent #6,878,362 was issued to GE.