II.G.1 Laboratory-Scale High Temperature Electrolysis System

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Start Date: January 23, 2003
Projected End Date: 2009 (completion of pilot-scale experiments)

Objectives

- Develop energy-efficient, high-temperature, solid-oxide electrolysis cells (SOECs) for hydrogen production from steam.
- Develop and test integrated SOEC stacks operating in the electrolysis mode.
- Develop optimized plant configuration for coupling to Generation IV Reactor.
- Combine components in an integrated laboratory-scale (ILS) experiment.
- Scale-up to a 200 kW pilot plant and a 1 MW engineering demonstration facility.

Technical Barriers

This project addresses the following technical barriers of the Nuclear Hydrogen Initiative:

- Nuclear reactor and central hydrogen production facility costs
- The need for high-temperature, corrosion resistant materials, particularly in steam-oxygen and steam-hydrogen environments
- Oxygen separation (and handling) technology

Technical Targets

- No greenhouse gas releases from industrial scale H_2 production
- Energy efficiency: 50% (lower heating value of H_2 produced/total thermal output of reactor)
- Cost of hydrogen: $2.50/kg – centralized production only
- Life of cells: 40,000 hours in continuous operation

Accomplishments

- Ceramatec fabricated and Idaho National Laboratory (INL) tested a 22-cell stack at hydrogen production rates greater than 100 normal liters per hour for 196 hours (August 3, 2005).
- Completed review article on *Sealing Technologies Applicable to Solid Oxide Electrolysis Cells* (INL Nov. 30, 2005).
- Finalized integrated ILS performance, space and power requirements (INL February 1, 2006).
- Operated 25-cell stack at 177 normal liters per hour (average) for 1,000 hours (INL/Ceramatec February 16, 2006).
- Began testing of initial dual stack ILS module at Ceramatec Facility (2 x 60 cells). Initial output 1.2 Nm^3 per hour (June 28, 2006).

Introduction

A research project is under way at the Idaho National Laboratory and collaborating laboratories to assess the performance of solid-oxide cells operating in the steam electrolysis mode for hydrogen production over a temperature range of 800 to 900°C. The research project includes both experimental and modeling activities. Experimental results were obtained from 22-cell and 25-cell planar electrolysis stacks, fabricated by Ceramatec, Inc. The electrolysis cells are electrolyte-supported, with scandia-stabilized zirconia electrolytes (~140 µm thick), nickel-cermet steam/hydrogen electrodes, and manganite oxygen electrodes. The metallic interconnect plates are fabricated from ferritic stainless steel. Hydrogen production rates up to 205 normal liters per hour were demonstrated. The 22-cell stack was tested for 196 hours in July and August 2005 and the 25-cell stack was tested for 1,000 hours in January and February 2006. A range of static and dynamic operating parameters were explored.

A three-dimensional computational fluid dynamics (CFD) model was also created to model high-temperature steam electrolysis in a planar solid oxide electrolysis cell (SOEC). A solid-oxide fuel cell (SOFC) model adds the electrochemical reactions and loss mechanisms and computation of the electric field throughout the cell. The FLUENT SOFC user-defined subroutine was modified for this work to allow for operation in the SOEC mode. Model results are shown to compare favorably with the experimental results obtained from the 10-cell, 22-cell and 25-cell stacks tested at INL.

Approach

An updated schematic of the stack-testing apparatus used for these tests is presented in Figure 1. The use of nitrogen as a carrier gas allows us to independently vary both the partial pressures and the flow rates of the steam and hydrogen gases while continuing to operate at atmospheric pressure. The flow rates of nitrogen, hydrogen and air are established by means of precision mass-flow controllers.

Hydrogen is included in the inlet flow as a reducing gas in order to help prevent oxidation of the nickel-zirconia cermet electrode material. Since the nitrogen and hydrogen flow rates are fixed by the mass flow controllers, and the steam partial pressure is fixed by the bath temperature, the complete gas composition is precisely known.

A close-up photograph of the 25-cell solid-oxide electrolysis stack, fabricated by Ceramatec, Inc. for this test, is shown in Figure 2. The stack is designed to operate in cross flow, with the steam/hydrogen gas mixture entering the inlet manifold on the right of Figure 2(a), and exiting through the outlet manifold, visible on the left in Figure 2(b). Air flow enters at the rear though an air inlet manifold (not visible in Figure 2) and exits at the front directly into the furnace. The power lead attachment tabs, integral with the upper and lower interconnect plates are also visible in Figure 2(b).

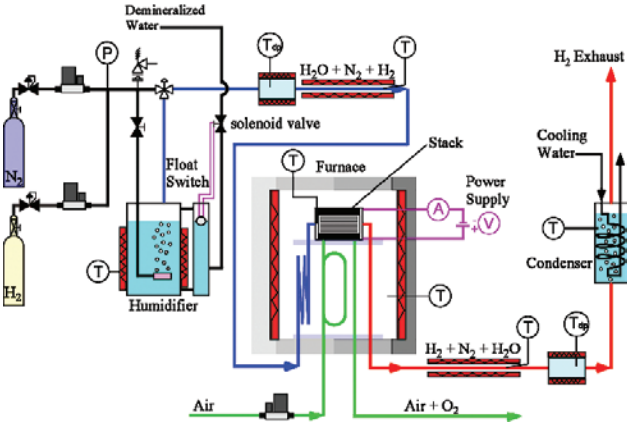

FIGURE 1. Schematic of Experimental Apparatus for Electrolysis Stack Testing

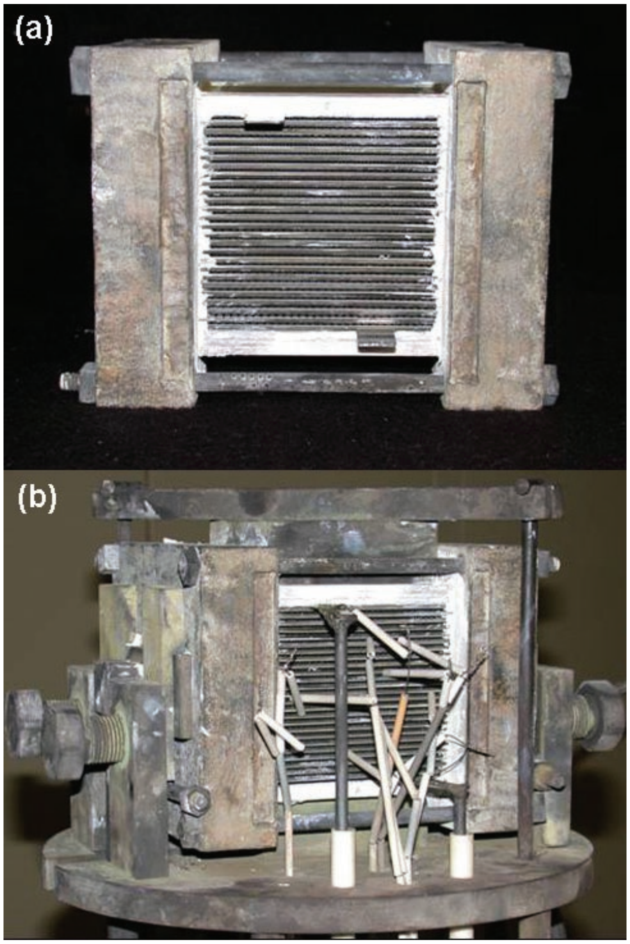

FIGURE 2. 25-Cell Planar Stack; (a) Front View of Stack Prior to Installation, (b) Instrumented Stack on Test Stand

Additional instrumentation is also visible in Figure 2(b). These include four intermediate voltage taps and five miniature thermocouples inserted into the air-side flow channels to monitor internal stack temperatures. These were inconel-sheathed 0.020-inch outside diameter, mineral-insulated, type-K, ungrounded thermocouples.

Results

Hydrogen production rates measured during the 1,000-hour test are presented in Figure 3. Two data sets are used, one based on stack current and the second based on the change in dewpoint between the inlet and outlet.

The stack was operated for the initial 100 hours at 800°C, after which the temperature was increased to 830°C. The increase in hydrogen production is due to a decrease in electrolyte resistance. The mean hydrogen production rate during the 1,000-hour test was 177 NL/hr, a factor of five larger than the highest reported hydrogen production rate using the sulfur-iodine (S-I) process.

Electrode Development

The overall goal of the Electrode Materials Development task at the Argonne National Laboratory (ANL) is to develop SOEC oxygen electrodes and steam electrodes that will lead to improved performance, reduced cost, and improved durability. The majority of voltage efficiency loss in an electrolyte-supported high-temperature steam electrolysis cell, such as those produced by Ceramatec, arises from ohmic losses due to the high resistance of the thick electrolyte layer (~150 µm) and activation losses due to the sluggish kinetics of the oxygen evolution reaction. These electrolyte and oxygen electrode losses are comparable and constitute 80% of the total cell (anode/electrolyte/cathode) voltage efficiency loss.

Durability of the SOEC system is an important requirement to lower the amortized cost of the hydrogen production system. For high efficiency operation, the SOEC hydrogen electrode inlet must operate with high steam content to reduce the need for recycling of the product hydrogen. In addition, the hydrogen electrode must be stable under high steam concentrations at the cell inlet and high hydrogen concentrations at the outlet. The currently-used SOEC hydrogen electrode is a cermet made of nickel, for electronic conductivity and catalytic activity, and yttria- or scandia-stabilized zirconia (YSZ or ScSZ) to provide oxygen ion conductivity. Past high-temperature steam electrolysis work has shown that the polarization of a nickel-based electrode is high at high steam concentrations, which has been attributed to surface oxidation of the nickel and loss of catalytic activity. Furthermore, mechanical deterioration of the electrode is observed as the nickel undergoes repeated oxidation-reduction cycles that may be encountered under varying cell operating conditions.

Numerical Modeling

INL and ANL researchers are analyzing the electrochemical and thermal-fluid behavior of SOECs for high temperature steam electrolysis using CFD techniques. The major challenges facing commercialization of steam electrolysis technology are related to efficiency, cost, and durability of the SOECs. The goal of this effort is to guide the design and optimization of performance for high temperature electrolysis (HTE) systems.

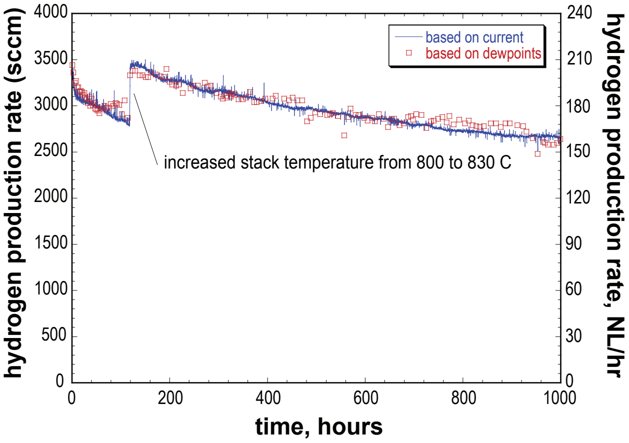

FIGURE 3. Hydrogen Production Rates Measured During 1,000-hour Test

An SOEC module developed by FLUENT Inc. as part of their general CFD code was used for the SOEC analysis by INL. ANL has developed an independent SOEC model that combines the governing electrochemical mechanisms based on first principals to the heat transfer and fluid dynamics in the operation of SOECs. The ANL model was embedded into the commercial STAR-CD CFD software, and is being used for the analysis of SOECs by ANL.

The FY 2006 analysis performed by ANL and reported here covered the influence of electrochemical properties, SOEC component resistances and their contributing factors, SOEC size and inlet flow conditions, and SOEC flow configurations on the efficiency and expected durability of these systems.

Analysis performed at INL evaluated the effects of variations in operating temperature, gas flow rate, and contact resistance. For these variations, mean per-cell resistance values decrease with increasing current density, consistent with experimental data. Predicted mean outlet hydrogen and steam concentrations vary linearly with current density, as expected.

Conclusions

- Experimental results from a 25-cell stack, 64 cm^2 active area, fabricated by Ceramatec:
  - Hydrogen production rates in excess of 160 NL/hr were maintained with a 25-cell solid-oxide electrolysis stack for 1,000 hours over

  the time period from January 5 to February 17, 2005.
  - The stack endurance test was terminated due to completion of the milestone and not due to any problem with the stack itself.
  - Stack performance as measured by the per-cell area specific resistance was good, beginning at 1.60 ohm-cm^2 and finishing at 2.15 ohm-cm^2.
- The ILS experiment will produce ~6 N m^3 of H_2, while incorporating, at reduced scale, all of the components of a commercial plant.
- INL, in collaboration with FLUENT and ANL, using STAR-CD, developed electrochemical CFD models for electrolysis that predict product flow, current density, temperature and ASR distributions within the SOEC.
- Increasing the inlet mass flux while going to larger cells can be a compromise to overcome increasing thermal and current density gradients while increasing the cell size. This approach could be beneficial for the economics of the SOECs.
- The presence of excess hydrogen at the SOEC inlet to avoid Ni degradation can result in a sizeable decrease in the process efficiency.
- A parallel-flow geometry for SOEC operation (if such a thing be achieved without sealing problems) yields smaller temperature gradients and current density gradients across the cell, which is favorable for the durability of the cells.
- Contact resistances can significantly influence the total cell resistance and cell temperatures over a large range of operating potentials. Thus it is important to identify and avoid SOEC stack conditions leading to such high resistances due to poor contacts.

Future Directions

A schematic of the 15 kW High Temperature Electrolysis ILS experiment is provided in Figure 4. Recuperating heat exchangers in the schematic are shown with cross-hatching and a bypass line, indicating that these components may not be present in the initial ILS deployment. However, the facility will be designed such that later insertion of heat recuperation equipment will be simplified. A drawing of a four-stack module is shown in Figure 5 and the 6-foot by 8-foot skid for the overall test is shown in Figure 6. The ILS experiment will use three 4-stack modules and produce about 6 N m^3 of H_2 per hour.

FY 2006 Publications/Presentations

1. Hawkes, G. L., O'Brien, J. E., Stoots, C. M., Herring, J. S., Shahnam, M., "Thermal and Electrochemical

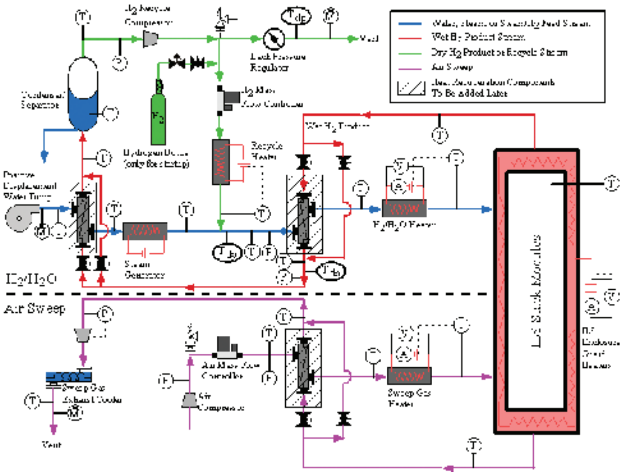

FIGURE 4. Schematic of High Temperature Electrolysis Integrated Laboratory-Scale Experiment

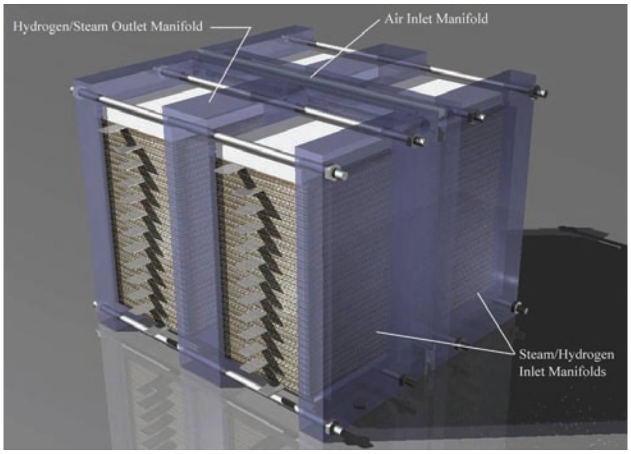

FIGURE 5. Integrated Laboratory-Scale Four-Stack Module

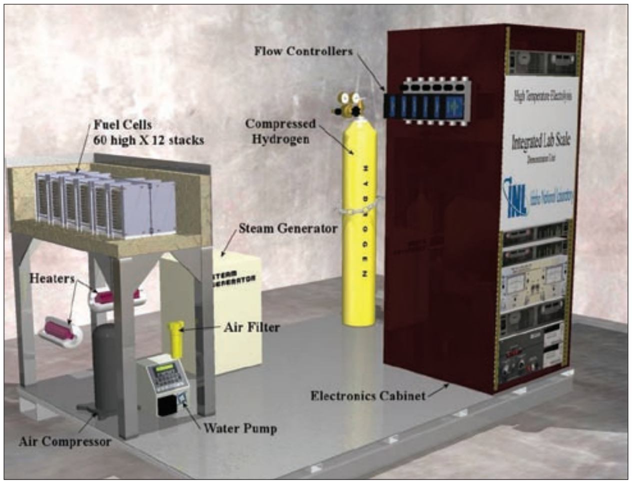

FIGURE 6. Integrated Laboratory-Scale System Mounted on 6 x 8-foot Skid

Three Dimensional CFD Model of a Planar Solid Oxide Electrolysis Cell," *Proceedings, 2005 ASME Heat Transfer Conference*, July 17-22, 2005, San Francisco.

2. O'Brien, J. E., Stoots, C. M., Herring, J. S., Lessing, P. A., Hartvigsen, J. J., and Elangovan, S., "Performance Measurements of Solid-Oxide Electrolysis Cells for Hydrogen Production from Nuclear Energy," *Journal of Fuel Cell Science and Technology*, Vol. 2, August 2005, pp. 156-163.

3. Hawkes, G. L., O'Brien, J. E., Stoots, C. M., Herring, J. S., Shahnam, M., "CFD Model of a Planar Solid Oxide Electrolysis Cell for Hydrogen Production from Nuclear Energy," to be presented at the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics NURETH-11, Popes Palace Conference Center, Avignon, France, October 2-6, 2005.

4. O'Brien, J. E., Herring, J. S., Stoots, C. M., Lessing, P. A., "High-Temperature Electrolysis for Hydrogen Production From Nuclear Energy," to be presented at the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics NURETH-11, Popes Palace Conference Center, Avignon, France, October 2-6, 2005.

5. Herring, J. Stephen, O'Brien, James E., Stoots, Carl M., Hawkes, G. L., Lessing, Paul, Windes, William, Wendt, Daniel, McKellar, Michael, Sohal, Manohar and Hartvigsen, Joseph J Hartvigsen, "Progress in High Temperature Electrolysis for Hydrogen Production," *Third Information Exchange Meeting on Nuclear Production of Hydrogen*, Oarai Research Establishment, JAEA, Japan October 5-7, 2005

6. Herring, J. S., O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "High Temperature Electrolysis for Hydrogen Production Using Nuclear Energy" Paper #501, GLOBAL 2005, Paper #501, Tsukuba, Japan, October 9–13, 2005.

7. Stoots, C. M., O'Brien, J. E., McKellar, M. G., Hawkes, G. L., and Herring, J. S., "Engineering Process Model for High-Temperature Steam Electrolysis System Performance Evaluation," AIChE 2005 Annual Meeting, Cincinnati, Ohio, October 30–November 4, 2005.

8. O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "Comparison of a One-Dimensional Model of a High-Temperature Solid-Oxide Electrolysis Stack with CFD and Experimental Results," presented at the 2005 ASME International Mechanical Engineering Congress and Exposition, November 5–11, 2005, Orlando, FL.

9. Herring, J. S., O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "Progress in High-Temperature Electrolysis for Hydrogen Production using Planar SOFC Technology," accepted for publication in the *International Journal of Hydrogen Energy,* 2006.

10. O'Brien, J. E., Stoots, C. M., Herring, J. S., and Hartvigsen, J. J.,"Hydrogen Production Performance of a 10-Cell Planar Solid-Oxide Electrolysis Stack," *Journal of Fuel Cell Science and Technology*, May, 2006.

Presentations

Steve Herring

1. HTE presentation to Northern Lights delegation, INL, June 23, 2005.

2. Hydrogen Tutorial to World Nuclear University students, INL, July 14, 2005.

3. Seminar University of Nevada, Reno, October 4, 2005.

4. HTE presentation to Alberta Research Council, INL, December 8, 2005.

5. Presentation to Jerry Paul, DOE-NNSA (National Nuclear Security Administration) at INL, February 28, 2006.

6. Briefing to Japan Science and Technology Agency delegation, INL, March 1, 2006.

7. Seminar University of Cincinnati and Ohio State University, April 4, 2006.

8. Presentation to EPRI visitor, INL, April 12, 2006.

9. Briefing to Japanese Ministry of Education Ministry of Education, Culture, Sports, Science and Technology (MEXT) delegation visit to INL, May 9, 2006.

10. American Nuclear Society Teacher's Workshop, Reno, June 3, 2006.

Jim O'Brien

1. Seminar on High Temperature Electrolysis, Michigan Technological University, Houghton, Michigan, March 2, 2006.

2. Seminar, NASA Glenn Research Center, Cleveland, Ohio, March 6, 2006.