

IV.G Hydrogen from Nuclear Energy

IV.G.1 High-Temperature Electrolysis

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Subcontractor:
Ceramatec, Inc., Salt Lake City, UT

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 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:

- The need for high-temperature, corrosion resistant materials, particularly in steam-oxygen and steam-hydrogen environments
- Nuclear reactor and central hydrogen production facility costs

Approach

- Develop energy-efficient, high-temperature SOECs for hydrogen production from steam
- Optimize energy efficiency, cost and durability
- Optimize electrolyte materials (e.g., yttria-stabilized zirconia, scandia-stabilized zirconia, sealants)
- Investigate alternate cell configurations (e.g., electrode-supported or tubular)
- Develop and test integrated SOEC stacks operating in the electrolysis mode with an aim toward scale-up to a 200 kW pilot plant and a 1 MW engineering demonstration facility
- Increase SOEC stack durability and sealing with regard to thermal cycles
- Improve material durability in a hydrogen/oxygen/steam environment
- Perform a progression of electrolysis stack testing activities at increasing scales and complexities

- Develop computational fluid dynamics (CFD) capability for SOEC
- Utilize advanced systems modeling codes (e.g., HYSYS, ASPEN)
- Perform cost and safety analyses

Accomplishments

- The high-temperature electrolysis (HTE) system configuration study was completed, identifying the options and technical issues for HTE systems, including electrolyzer cell and module configurations and high-temperature steam distribution. An assessment of the engineering materials issues and requirements for construction of an HTE system was completed to provide input to the high-temperature materials testing program. An experiment plan for the development of high-temperature electrolysis components and systems was completed to provide input to the longer term scaling of HTE systems.
- An assessment of the high-temperature reactor and HTE process interface requirements was completed.
- Completed engineering analyses for high-temperature electrolysis scaling demonstration experiments, including steam distribution systems and electrolyzer cell and module configurations.
- Completed a series of button cell experiments to examine cell operational characteristics under a range of conditions.
- Completed initial testing of a 10 cell stack in preparation for hydrogen production testing.
- Completed final design for laboratory-scale HTE experiments
- Developed engineering process model for HTE system performance evaluation, including the development of CFD and electrochemical modeling for planar geometry electrolyzer cells, including mass and thermal transport.
- Demonstrated high-temperature electrolysis stack testing at a production rate of 50 normal liters (NL)/hr of hydrogen.
- Produced 15 button cells using plasma deposition on nickel aluminide substrate (now being tested).

Future Directions

FY 2005

- Demonstrate HTE stack testing at a production rate of 100 NL/hr of hydrogen.
- Complete conceptual design documentation for HTE pilot-scale experiments.

FY 2006

- Complete design of HTE integrated laboratory-scale experiments.
- Construct stack/module arrays for integrated laboratory-scale experiments.

FY 2007

- Begin HTE integrated lab-scale experimental operations.
- Complete HTE cell testing.
- Conduct HTE stack/module tests.
- Perform initial testing on candidate pilot scale module tests.
- Complete preliminary pilot scale experiment design.

FY 2008

- Pilot-scale experiment final design.
- Complete HTE integrated lab-scale experimental operations.
- Implement cell/module technology improvements.

Introduction

A research project is under way at the Idaho National Laboratory (INL) 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 a ten-cell planar electrolysis stack, fabricated by Ceramtec, Inc. The electrolysis cells are electrolyte-supported, with scandia-stabilized zirconia electrolytes (~140 μm thick), nickel-cermet steam/hydrogen electrodes, and manganite air-side electrodes. The metallic interconnect plates are fabricated from ferritic stainless steel. The experiments were performed over a range of steam inlet mole fractions (0.1 – 0.6), gas flow rates (1,000 – 4,000 sccm), and current densities (0 to 0.38 A/cm^2). Hydrogen production rates up to 90 NL/hr were demonstrated. Stack performance is shown to be dependent on inlet steam flow rate. A three-dimensional CFD model was also created to model high-temperature steam electrolysis in a planar SOEC. The model represents a single cell as it would exist in the experimental electrolysis stack. Mass, momentum, energy, and species conservation and transport are provided via the core features of the commercial CFD code FLUENT. 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 provide detailed profiles of temperature, Nernst potential, operating potential, anode-side gas composition, cathode-side gas composition, current density and hydrogen production over a range of stack operating conditions. Mean model results are shown to compare favorably with the experimental results obtained from the ten-cell stack tested at INL.

Approach

A schematic of the stack-testing apparatus is presented in Figure 1. Primary system components include gas supply cylinders, gas mass-flow controllers, a humidifier, dew point measurement

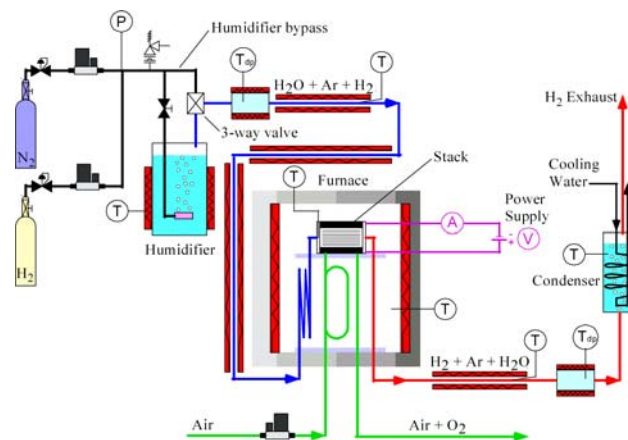


Figure 1. Schematic of Experimental Apparatus for Electrolysis Stack Testing

stations, temperature and pressure measurement, high-temperature furnace, and a solid oxide electrolysis stack. Nitrogen was used as an inert carrier gas. The use of 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 (Hastings Model HFC-302, with Hastings Model 400 electronics). Air flow to the stack is supplied by the laboratory shop air system, after passing through a two-stage extractor/dryer unit.

Downstream of the mass-flow controller, nitrogen is mixed with a smaller flow of hydrogen gas. Hydrogen is included in the inlet flow as a reducing gas in order to prevent oxidation of the nickel cermet electrode material. The nitrogen/hydrogen gas mixture is mixed with steam by means of a heated humidifier. The humidifier consists of a heated stainless-steel vessel containing demineralized/deionized water through which the nitrogen/hydrogen flow is bubbled. The dew point temperature of the steam/hydrogen/nitrogen gas mixture exiting the humidifier is monitored continuously using a precision dew point sensor (Vaisala Model HMP247). The humidifier is fitted with a clamp heater and is externally insulated. The humidifier temperature is maintained at a constant set point value using feedback control. Since the vapor pressure of the water and the resulting partial pressure of the steam exiting the humidifier are

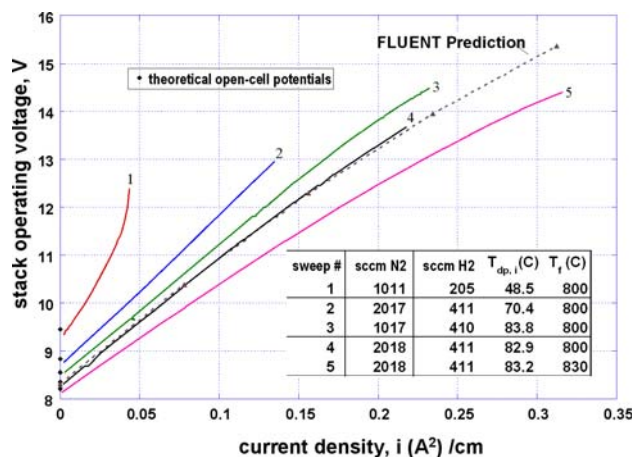


Figure 2. Stack Operating Potential as a Function of Current Density

determined by the water bath temperature, the water vapor mass flow rate is directly proportional to the carrier gas flow rate for a specified bath temperature. Also, 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 at all times. All gas lines located downstream of the humidifier are heat-traced in order to prevent steam condensation. Gas line temperatures are monitored by thermocouples and controlled by means of variable transformers.

Results

Results of several representative sweeps are shown in Figure 2 in the form of operating voltage versus current density. Test conditions for each of the five experimental sweeps shown are tabulated in the figure. Four of the sweeps were obtained at a furnace temperature of 800°C and one at 830°C. Inlet dew point values were varied as shown. Theoretical open-cell potential values are also shown in the figure for each sweep using a single data point at zero current density. Note that the measured open-cell potentials are in excellent agreement with these theoretical values for each sweep. In addition to the experimental curves, a FLUENT prediction is also shown. This prediction was obtained for the conditions of sweep 4, with the gap contact resistance values empirically adjusted to yield an overall area-specific resistance value that matches the experimentally observed value. Additional

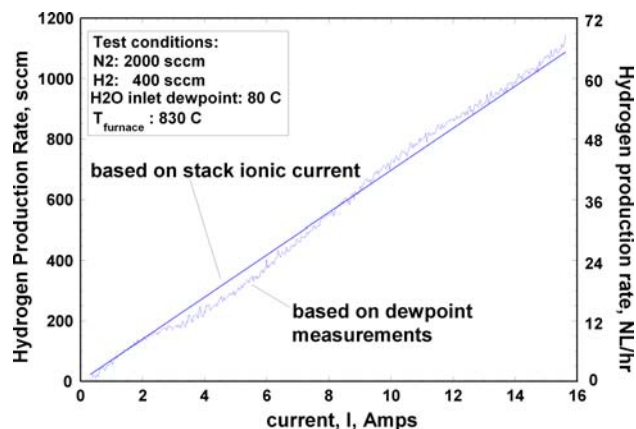


Figure 3. Hydrogen Production Rates During DC Potential Sweep

FLUENT predictions of local temperatures and current densities presented later were obtained using the same procedure.

Hydrogen production rates can be calculated directly from the stack electric current and independently from the measured inlet and outlet dew points. A representative plot of hydrogen production during a DC potential sweep is shown in Figure 3. The left-hand vertical scale is in sccm and the right-hand vertical axis is in NL/hr. The current-based hydrogen production rate is simply a straight line since hydrogen production is directly proportional to the current. The dew point-based measurement shows some scatter associated with the instantaneous measured inlet and outlet dew point values. Agreement between the two measurements is generally very good. Hydrogen production rates as high as 90 NL/hr were achieved with this stack.

Conclusions

- High-temperature nuclear reactors have the potential for substantially increasing the efficiency of hydrogen production from water splitting, with no consumption of fossil fuels, no production of greenhouse gases, and no other forms of air pollution.
- High-temperature electrolytic water splitting supported by nuclear process heat and electricity has the potential to produce hydrogen with overall system efficiencies near those of the thermochemical processes, but without the fossil

- fuel consumption and greenhouse gas emissions associated with hydrocarbon processes.
- A high-temperature advanced nuclear reactor coupled with a high-efficiency high-temperature electrolyzer could achieve a competitive thermal-to-hydrogen conversion efficiency of 45 to 55%.

FY 2005 Presentations

Steve Herring

1. Georgia Institute of Technology, January 20, 2005
2. University of Wisconsin, February 24, 2005
3. University of California at Berkeley, March 14, 2005
4. MIT, April 4, 2005
5. University of Idaho, April 29, 2005
6. ANS Teacher's Workshop, San Diego, June 4, 2005

Jim O'Brien

1. University of Cincinnati and Ohio State University, April 19, 2005

FY 2005 Publications

1. Hawkes, G. L., O'Brien, J. E., Stoots, C. M., Herring, J. S., Shahnam, M., "Thermal and Electrochemical Three Dimensional CFD Model of a Planar Solid Oxide Electrolysis Cell," to be presented at the 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," to be published in *Journal of Fuel Cell Science and Technology*, August 2005.
3. 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," submitted for presentation at the 2005 AIChE Spring Annual Meeting, April 10 – 14, 2005, Atlanta, GA.
4. 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," submitted for presentation at the ASME 3rd International Conference on Fuel Cell Science, Engineering, and Technology, May 23 – 25, 2005, Ypsilanti, MI.
5. 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.
6. 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.
7. Herring, J. S., O'Brien, J. E., Stoots, C. M., Lessing, P. A., Hartvigsen, J. J., and Elangovan, S., "Hydrogen Production through High-Temperature Electrolysis," chapter in book edited by Dr. Masao Hori, "Nuclear Production of Hydrogen - Technologies and Perspectives for Global Deployment, International Nuclear Societies Council, *Current Issues in Nuclear Energy*, 2004.