II.F Electrolysis

II.F.1 High-Temperature Solid-Oxide Electrolyzer System

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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.
- Aim toward scale-up to a 500-kW pilot plant and a 5-MW engineering demonstration facility.

Technical Barriers

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

- V. High- and Ultra-High-Temperature Thermochemical Technology
- X. Policy and Public Acceptance

Approach

- Develop energy-efficient, high-temperature SOECs for hydrogen production from steam.
  - Reduce ohmic losses to improve energy efficiency.
  - Minimize electrolyte thickness.
  - Utilize high-performance electrolyte materials [e.g., scandia-stabilized zirconia (ScSZ), lanthanum strontium gallium magnesium oxide (LSGM)].
  - Investigate alternate cell configurations (e.g., electrode-supported).
  - Conduct single-cell performance characterization testing.
- Develop and test integrated SOEC stacks operating in the electrolysis mode with an aim toward scale-up to a 500-kW pilot plant and a 5-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.

  - Perform cost and safety analyses.

Accomplishments

- Established button cell for single-cell tests.
  - Measured open-cell potentials were shown to agree well with theoretical predictions during system heatup and testing.
  - Observed values of area-specific resistance (ASR) ranged from about 0.35 to 1.0, depending on test conditions and the specific cell being tested. Degradation of ASR associated with thermal cycling of the cells was noted.
  - The effects of steam starvation on ASR in the electrolysis mode were evident at higher current densities.
  - Measured dewpoint-change values obtained during the steady-state tests were shown to agree very closely with predictions based on cell current.
  - Thermal efficiency values based on measured hydrogen production were in agreement with values based on cell operating voltage.
  - Both efficiencies approach their respective theoretical reversible limits at low current density.
  - In general, cell performance was shown to be continuous from the fuel-cell mode to the electrolysis mode of operation.
- Initial stack testing has begun.

Future Directions

- Continue button-cell testing to identify optimum cell materials.
- Continue stack testing to evaluate and optimize electrical conductivity, seals, and thermal cycling performance.
- Expand scale of stack testing, ultimately to a 500-kW pilot plant and a 5-MW engineering demonstration facility, to address integration issues such as hydrogen and oxygen storage and handling, heat recuperation, facility maintenance, and interface issues.

Introduction

A research project is being conducted by the Idaho National Engineering and Environmental Laboratory (INEEL) and Ceramatec, Inc., to test the high-temperature, electrolytic production of hydrogen from steam using a solid-oxide cell. The high-temperature heat and the electrical power would be supplied simultaneously by a high-temperature nuclear reactor. Alternatively, in somewhat smaller applications, the heat might be supplied through solar concentrators and the electricity through wind or other renewables. Operation at high temperature reduces the electrical energy requirement for electrolysis and also increases the thermal efficiency of the power-generating cycle. The high-temperature electrolysis process will utilize heat from a specialized secondary loop carrying a steam/hydrogen mixture. It is expected that, through the combination of high-efficiency electrical generation and high-temperature electrolysis, the process will achieve an overall thermal conversion efficiency of 40 to 50% while avoiding the challenging chemistry and corrosion issues associated with thermochemical processes.

Approach

This project is a collaborative effort between the INEEL and Ceramatec of Salt Lake City. The effort includes a logical progression of research and testing activities from small-scale bench testing to a large-scale demonstration. Three scales of testing are to be performed: button-cell, stack, and integrated laboratory/demonstration scale. Button-cell testing is intended to address cell material development, optimization, and performance characterization. Stack-scale (multiple cells) testing addresses inter-cell electrical contact, cell and manifold sealing issues, gas flow distribution, and overall stack performance characterization. Finally, integrated laboratory/demonstration-scale testing will address production issues including energy management; utility requirements; and product purification, handling, and storage.

Electrode and electrolyte materials research typically occurs at the single- (button) cell scale. New materials are still being identified (mostly supported by fuel-cell research funding) to combine high ionic conductivity with lower-temperature operation. Basic cell designs include electrolyte-supported and electrode-supported configurations, with further possibilities for multi-layer cells with graded-porosity electrodes. Fabrication methods include traditional ceramic fabrication techniques such as tape casting and screen printing, in addition to advanced thermal spray techniques.

Stack configurations introduce a host of new research issues, including geometric configuration (e.g., planar or tubular), interconnect materials, seals, manifolding, gas flow distribution, and electrical connections. Stacks may also require the use of conductive interface layers.

The proposed integrated laboratory scale of testing would include 8 stacks of 250 cells each, a total of 2000 cells. At this scale, integral effects similar to those that would be encountered in large-scale facilities will exist. For example, the integrated lab-scale experiment will not rely upon a furnace for preheating and maintaining the stack temperature. In-line gas heaters and/or recuperative heat exchangers will be used, as will be the case in larger-scale facilities. Engineering demonstration scales will concentrate on material handling (hydrogen and oxygen) issues, product purification, product storage, and system maintenance issues. Instrumentation development will occur at all levels of testing.

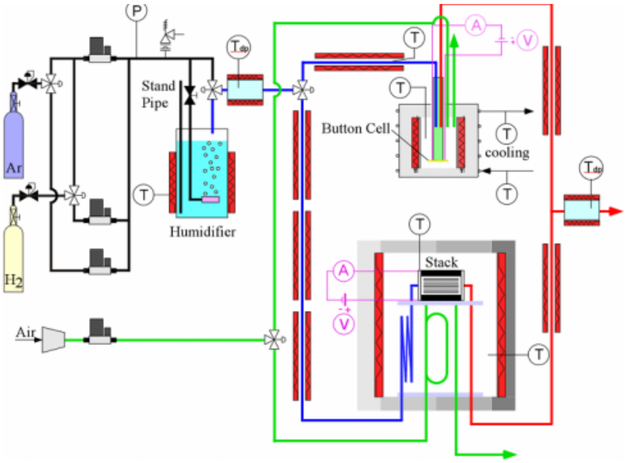

Figure 1. Testing Apparatus for Button Cells and Stacks

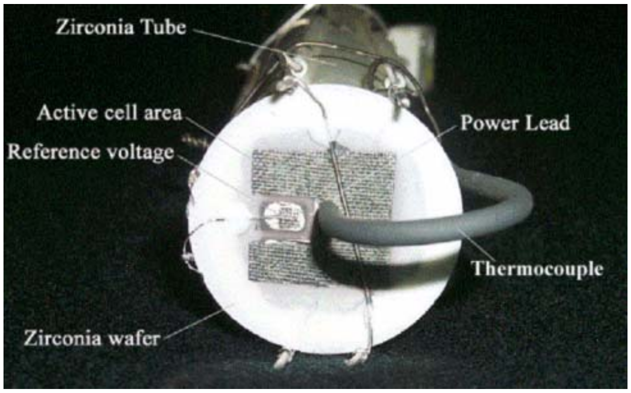

Figure 2. Button Cell Ready For Testing

Results

The INEEL SOEC test loop for button-cell and stack testing is shown schematically in Figure 1. Argon is being used as a carrier gas to control the steam-hydrogen ratio being fed to the button cell or stack in the furnace. The dewpoint temperatures of the feed stream and the exiting stream are directly measured to determine the amount of hydrogen produced by the button cell or stack. For safety reasons, the oxygen is diluted with air before being exhausted.

The first cells to be tested were single cells based on existing materials and fabrication technology developed at Ceramatec for production of solid-oxide fuel cells. Figure 2 is a button cell ready for testing. These cells use a relatively thick (~150 μm)

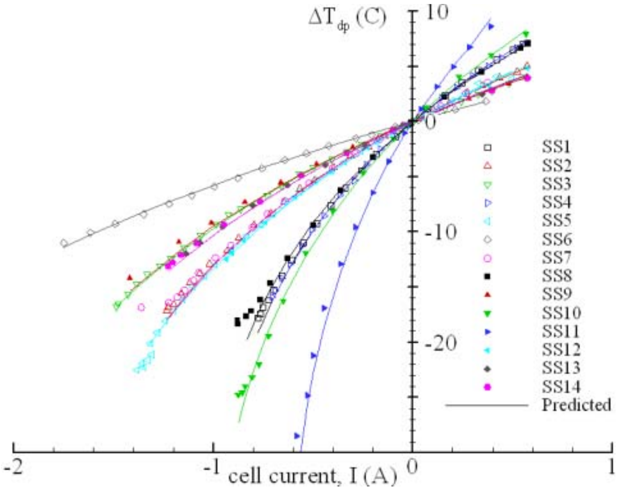

Figure 3. Dewpoint Changes for Various Cell Currents

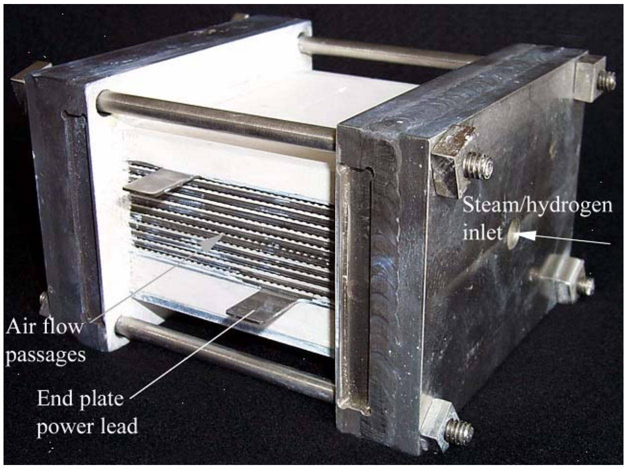

Figure 4. Ten-Cell Electrolysis Stack

electrolyte of yttria- or scandia-stabilized zirconia, with nickel-zirconia cermet anodes and strontium-doped lanthanum manganite cathodes. Additional custom cells with lanthanum gallate electrolyte have been developed and tested. Results to date have shown an area-specific resistance (ASR) as low as 0.35 Ω-cm^2 at 850°C. Dewpoint measurements correlated well with expected hydrogen production rates based on measurements of electrical current supplied to the cell (Figure 3).

The INEEL also has developed an electrolysis cell stack testing capability. A photograph of a 10-cell solid-oxide electrolysis stack in preparation for testing is shown in Figure 4. This stack has a per-cell active area of 64 cm^2 and should be capable of hydrogen production in excess of 60 L/h. It is designed to operate. in cross flow, with the steam/hydrogen gas mixture entering the inlet manifold on the right in the photograph and exiting through the outlet manifold, visible on the left in the photograph. Air flow enters at the front through an air inlet manifold (not shown) and exits at the rear directly into the furnace. The power lead attachment tabs, integral with the upper and lower interconnect plates, are also visible in the photograph. The interconnect plate is fabricated from ferritic stainless steel (alloy 441). It includes 50 perforated flow channels across its width to provide full-coverage gas flow distribution for the hydrogen/steam mix on the cathode side and the oxygen flow on the anode side. The interconnect plates also serve as electrical current distributors. The electrolyte in this case is scandia-stabilized zirconia. Electrodes are

Figure 5. Ten-Cell Electrolysis Mounted for Testing at INEEL

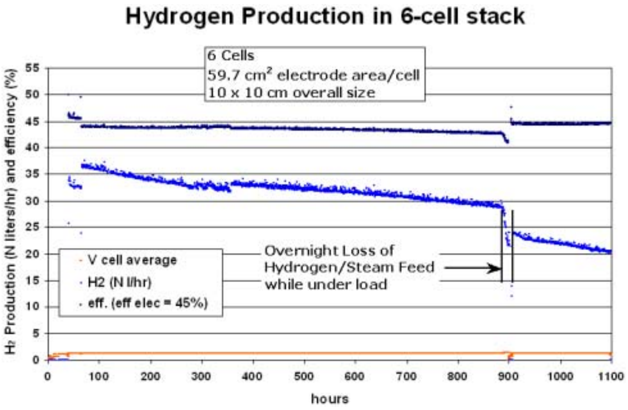

Figure 6. 1100-Hour Test of Six-Cell Stack at Ceramatec Facility, Oct.-Nov. 2003

strontium-doped lanthanum manganite and nickel-zirconia cermet.

A photograph of the stack, mounted on its inconel test fixture and resting on the furnace base, is shown in Figure 5. The power leads are inconel rods insulated with alumina tubing. The hydrogen/steam and air inlet tubes are coiled to provide additional length for heat transfer upstream of the stack.

Preliminary electrolysis stack testing has been completed at the Ceramatec facility in Salt Lake City. Results of the most recent electrolysis stack tests are shown in Figure 6 This stack included six cells, each having an electrode area of 59.7 cm^2. In electrolysis mode, with a stack current of 13 A, the stack produced about 32 normal liters of H_2 per hour for about 800 hours during the 1100-hour test. The current density for this test was 218 mA/cm^2.

The critical parameters for a 300-$MW_{hydrogen}$ commercial electrolysis plant have been determined based on these experimental results. A preliminary design for modular electrolyzer units, to be used in conjunction with a high-temperature heat source, has also been developed.

Conclusions

High-temperature electrolysis using solid-oxide technology appears to be a viable means for producing hydrogen using nuclear energy. Laboratory-scale experiments over the last year have shown that this technology can produce hydrogen at close to the theoretical parameters.

A conceptual design of an electrolytic plant to be attached to a 600-MWth reactor has been developed, suggesting that the plant would be of moderate size and that the parameters of cells would be reasonable.

A rail-transportable, modular unit has been described such that the modules could be manufactured in a factory and installed at the reactor site. The modules would each produce about 0.18 kg (2000 normal liters) of hydrogen per second and require an electrical input of about 20 MWe.

FY 2004 Publications/Presentations

1. Herring, J., O'Brien, J., Stoots, C., Lessing, P., Anderson, R., Hartvigsen, J., and Elangovan, S., "Hydrogen Production through High-Temperature Electrolysis," Chapter prepared for book on *Nuclear Production of Hydrogen*, INSC Series, published by ANS, 2004.

2. Elangovan, S. and Hartvigsen, J., (Ceramatec, Inc.) and O'Brien, J., Stoots, C., Herring, J., and Lessing, P., "Operation and Analysis of Solid Oxide Fuel Cells in Steam Electrolysis Mode," 6th European SOFC Forum, Lucerne, Switzerland, June 28 - July 2, 2004.

3. O'Brien, J., Stoots, C., Herring, J., and Lessing, P., "Performance Characterization of Solid-Oxide Electrolysis Cells for Hydrogen Production," ASME 2nd International Conference on Fuel Cell Science, Engineering, and Technology, Rochester, NY, June 14-16, 2004.

4. Herring, J., O'Brien, J., Stoots, C., and Lessing, P., "Hydrogen Production from Nuclear Energy via High Temperature Electrolysis," 2004 International Congress on Advances in Nuclear Power Plants (ICAPP '04), paper 4322, Pittsburgh, PA, June 13-17, 2004.

5. O'Brien, J., Stoots, C., Herring, J., Lessing, P., "Performance Measurements of Solid-Oxide Electrolysis Cells for Hydrogen Production From Nuclear Energy," 12th International Conference on Nuclear Engineering ICONE12, Arlington, VA, April 25-29, 2004.

6. Herring, S., O'Brien, J., Stoots, C., Lessing, P., Anderson, R., Hartvigsen, J., Elangovan, S., "Hydrogen Production through High-Temperature Electrolysis Using Nuclear Energy," AIChE Spring 2004 Meeting, New Orleans, LA, April, 2004.
7. Herring, S., Lessing, P., O'Brien, J., Stoots, C., Hartvigsen, J., Elangovan, S., "Hydrogen Production through High-Temperature Electrolysis in a Solid Oxide Cell," Second Information Exchange Meeting on Nuclear Production of Hydrogen, Argonne National Laboratory, IL, October 2 and 3, 2003.
8. Herring, S., Anderson, R., O'Brien, J., Lessing, P., Stoots, C., "Development of a High Temperature Solid Oxide Electrolyser System," presented at the 2003 Hydrogen and Fuel Cells Merit Review Meting, Berkeley, CA, May 20, 2003.
9. O'Brien, J., Herring, J., Lessing, P., and Stoots, C., "High Temperature Steam Electrolysis from Advanced Nuclear Reactors Using Solid Oxide Fuel Cells," presented at the First International Conference on Fuel Cell Science, Engineering and Technology, Rochester, NY, April 21-23, 2003.