

High-Temperature Solid Oxide Electrolyser System

J. Stephen Herring (Primary Contact), James O'Brien, Carl Stoots, Paul Lessing and Ray Anderson

Idaho National Engineering and Environmental Laboratory (INEEL)

P. O. Box 1625 MS 3860

Idaho Falls, Idaho 83415-3860

Phone: (208) 526-9497; Fax: (208) 526-2930; E-mail: sth@inel.gov

DOE Technology Development Manager: Matthew Kauffman

Phone: (202) 586-5824; Fax: (202) 586-9811; E-mail: Nancy.Garland@ee.doe.gov

Subcontractor: Joseph Hartvigsen, S. Elangovan and Robert Lashway

Ceramatec, Inc., Salt Lake City, Utah

Objectives

- Develop and test energy-efficient, regenerative, solid oxide electrolyser cells (SOECs) for hydrogen production from steam using a high temperature heat source
 - Reduce ohmic losses to improve energy efficiency
 - Increase SOEC durability and sealing with regard to thermal cycles
 - Minimize electrolyte thickness
 - Improve material durability in a hydrogen/oxygen/steam environment
- Demonstrate cell performance and associated equipment operation in a flexible test loop
- Optimize stack configuration for incorporation into a full-scale plant
- Test integrated SOEC stacks operating in the electrolysis mode
- Specify and test hydrogen-permeation-resistant materials for high-temperature heat exchangers

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 R,D&D Plan:

- Q. Cost
- R. System Efficiency
- U. Electricity Costs
- W. High Temperature Materials
- X. Policy and Public Acceptance

Approach

- Develop solid oxide electrolyte, electrode, and catalyst materials optimized for high-temperature steam electrolysis
- Develop a flexible test loop at INEEL for performance testing of single electrolysis cells and cell stacks and for hydrogen permeation testing
- Test single solid oxide electrolysis cells for performance over a range of operating temperatures, gas compositions, and current densities both at Ceramatec and at INEEL
 - Initial testing will be performed on baseline cells, fabricated using existing Ceramatec solid oxide fuel cell (SOFC) technology (Ytria-stabilized zirconia electrolyte-supported cells)

- Subsequent testing will be performed on cells developed with different electrolyte compositions, electrode-supported cells, various catalysts, etc.
- Develop a high performance electrolysis stack configuration based on the results of the single-cell tests, including high-performance bi-polar plates
- Perform electrolysis testing in realistic cell stacks over a range of operating conditions and system pressures
- Perform high-temperature hydrogen permeation testing of candidate heat exchanger materials

Accomplishments

- Test loop designed, passed safety approvals and placed in operation January 21, 2003
- Single "button" cells tested for performance in electrolysis and fuel cell modes at Ceramatec and at INEEL
- Initial parameters incorporated in conceptual full-scale plant design
- Planning for multi-cell stack testing begun
- Numerical analysis by Argonne National Laboratory under way

Future Directions

- Evaluate alternative electrolytes and catalysts in single-cell testing
- Develop multi-cell stacks and stack testing capability
- Conduct test of multi-cell stacks
- Analyze operation of cells and overall plant with numerical techniques

Introduction

Of the many methods proposed for hydrogen production, water-splitting is considered ideal because it avoids CO₂ emissions. Two types of water-splitting technologies have been studied: thermochemical and electrolysis. Chemical-reaction-based water-splitting processes are hindered by the extremely corrosive process environment (for example, in the iodine-sulfur process, H₂SO₄ is produced and then dissociated at 850°C). Conventional low-temperature electrolysis of steam results in an unsatisfactory thermal efficiency.

This work is an experimental research project being conducted via a collaboration between the INEEL and Ceramatec, Inc. of Salt Lake City, Utah, to test the high-temperature, electrolytic production of hydrogen from steam using a reversible, solid oxide cell. The research team is designing and testing solid oxide cells for operation in the electrolysis mode, and evaluating materials for the high-temperature heat

exchanger, condenser, and other components necessary for producing hydrogen using a high-temperature process heat source plus electrical power. This high-temperature process heat and the electrical power requirement could be supplied simultaneously by a high-temperature nuclear reactor. 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 with this combination of a high-temperature reactor and high-temperature electrolysis, the process will achieve a thermal conversion efficiency of 40 to 50% while avoiding the challenging chemistry and corrosion issues associated with the thermochemical processes. Planar solid oxide cell technology is being utilized because it has the best potential for high efficiency due to minimized voltage and current losses. INEEL and Ceramatec scientists have been developing planar solid oxide cell technology for the past 20 years.

Approach

Solid oxide cells will be developed and tested for operation in the electrolysis mode. Initial testing will document the performance of single “button” cells, such as the cell shown in Figure 1. Subsequent testing will document the performance of multiple-cell stacks operating in the electrolysis mode. Testing will be performed both at Ceramatec and at INEEL. A logical progression is being followed in the test project. 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. These cells use a relatively thick (~175 μm) 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. During the second phase of this project, multiple-cell stacks will be developed using the most promising materials identified through the single-cell testing.

In order to carry out the initial phase of the test project at INEEL, a new test loop has been developed for testing of single solid oxide electrolysis cells. A photograph of the setup is shown in Figure 2. It will be modified later for testing of multiple-cell stacks. The test loop is designed for cell operation in the 700-1000°C temperature range. The following parameters are measured and controlled during testing: cell temperature, anode-side gas flow rate and composition (Ar , H_2 , H_2O), inlet and outlet dewpoint temperature, applied cell voltage (± 0.1 V



Figure 1. Details of Solid-Oxide Button Cell Fabricated by Ceramatec (TC = thermocouple, YSZ = yttria-stabilized zirconia)

differential from open-cell voltage), cell current, area-specific resistance, and hydrogen production rate. This facility is more heavily instrumented (52 channels of instrumentation) than the test facility used at Ceramatec for single-cell testing.

The scope of corrosion / permeation work to be performed in the second and third years of this project will focus on testing of various candidate high-temperature heat exchanger materials over a wide range of temperatures and partial pressures. Effects of permeation barrier treatments such as calorizing, oxide layers, and/or coatings will also be assessed. Candidate materials include Inconel and Hastelloy alloys, as well as ceramics. Permeation test materials will be characterized before and after testing using Scanning Electron Microscopy and Auger Electron Spectroscopy. Rates of permeation will be measured using techniques such as hydrogen permeation current, ultrasonics, and direct measurement of hydrogen leakage by capture of escaping hydrogen (using a containment envelope and carrier gas) and subsequent on-line gas analysis.

Results

Initial testing has been performed using single electrolyte-supported (thick) button cells fabricated by Ceramatec. Results of several representative “button” cell tests are presented in Figures 3 – 6. Figure 3 shows the open-cell potential measured across the reference electrodes of a button cell during heat-up in the INEEL test furnace. The measured values are compared to a prediction obtained from the Nernst equation, which quantifies the ideal open-



Figure 2. High-Temperature Single-Cell Steam Electrolysis Test Setup at INEEL

circuit potential from thermodynamic principles as a function of cell temperature, gas composition, and system pressure. Agreement between the measured values and the predictions is excellent, instilling confidence in the experiment design. The INEEL test loop is instrumented with precision dewpoint sensors located upstream and downstream of the electrolysis cell, allowing for direct measurement of steam consumption or production in electrolysis or fuel-cell modes, respectively. Figure 4 provides a plot of dewpoint change as a function of electric current for a single-cell test performed at INEEL in both electrolysis and fuel-cell modes. The figure shows both the measurements and a prediction, revealing good agreement. Figure 5 displays the results of a direct current (DC) potential sweep test

performed on a scandia-stabilized zirconia-electrolyte cell at INEEL in which a power-supply voltage is applied to the cell in order to force operation over a range of voltages. The results shown in the figure demonstrate reversible operation between the fuel-cell and electrolysis modes of operation. The slope of the potential sweep is highest for large negative current densities where the effects of steam starvation begin to have an effect. A similar figure representing a DC potential sweep test performed at Ceramtec on a lanthanum gallate-electrolyte cell is presented in Figure 6. The figure

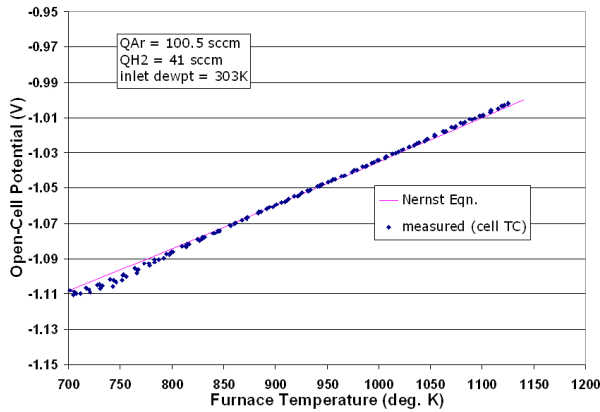


Figure 3. Open-Cell Electrical Potential Measured Across a Button Cell as a Function of Cell Temperature During Heat-up

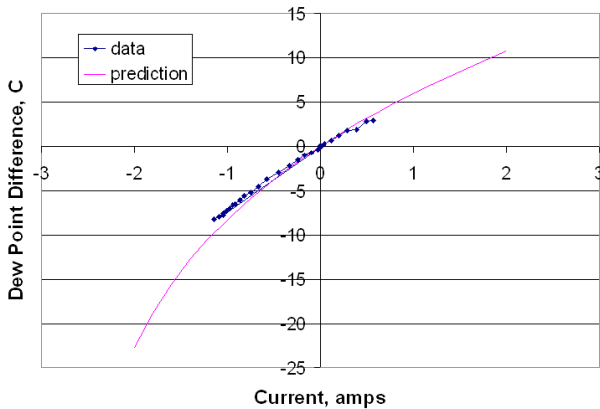


Figure 4. Dew Point Change as a Function of Cell Electric Current, Measurements And Theory

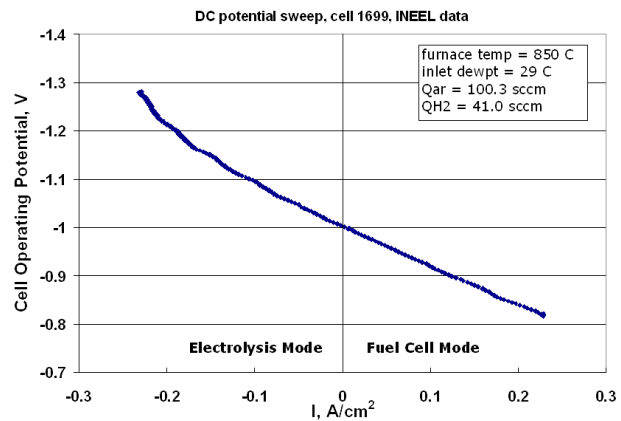


Figure 5. DC Potential Sweep Showing Reversible Operation of a Scandia-Stabilized Zirconia Button Cell Tested at INEEL in Both Fuel-Cell and Electrolysis Modes at 850°C

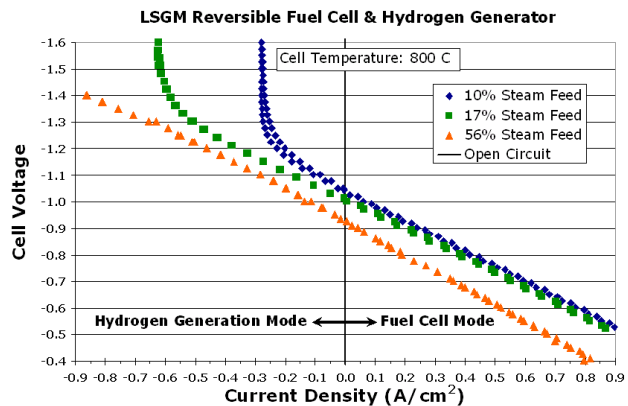


Figure 6. DC Potential Sweep Showing Reversible Operation of a Lanthanum-Gallate Button Cell Tested at Ceramtec in Both Fuel-Cell and Electrolysis Modes at 800°C (LSGM = lanthanum strontium gallium magnesium oxide)

again demonstrates reversible operation between the fuel-cell and electrolysis modes of operation. It also clearly demonstrates the effects of steam starvation in the electrolysis mode at low steam concentrations. This cell exhibited excellent performance with an area-specific resistance (ASR) of $0.57 \Omega\text{-cm}^2$, indicating low ohmic losses.

Initial results of these single-cell tests are very promising in terms of demonstrating high efficiency high-temperature steam electrolysis using solid oxide cells. Subsequent testing will be focused on testing cells fabricated from alternate materials optimized for the electrolysis application. Long-duration performance tests will also be performed. Stack configurations will be developed and tested.

Conclusions

- High-temperature steam electrolysis using a high-temperature heat source is a viable near-term strategy for large-scale hydrogen production.
- Solid oxide cells represent a logical choice for high-temperature steam electrolysis due to their high operating temperature and high efficiency.
- INEEL and Ceramatec have initiated a research project that is showing promising initial results on the thermoelectric efficiency issues related to the high-temperature electrolysis process.

FY 2003 Publications/Presentations

1. O'Brien, J. E., Herring, J. S., Lessing, P. A., and Stoots, C. M., "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