II.J.7 Test of High Temperature Electrolysis ILS Half Module

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Objectives

- Reduce risk for the Integrated Laboratory Scale (ILS) demonstration at INL
- Test "Half-ILS Module" at Ceramatec
  - Demonstrate hydrogen production rate greater than 1 Nm^3/hr
  - Validate 2 x 60 cell stacks configuration planed for full ILS module
  - Show that short stack performance is retained in a tall stack
  - Assess system issues with tall stack operation
- Exercise component production capacity at 100 cells/month
  - Deliver first full ILS module to INL

Technical Barriers

This project addresses the following technical barriers from the Production section (3.1) of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

H2 Generation by Water Electrolysis

(G) Capital Cost

(H) System Efficiency

(I) Grid Electricity Emissions

(J) Renewable Integration

Contribution to Achievement of DOE NHI Milestones

The Half-ILS module test, full ILS module delivery and test report conducted under this project were specific high level milestones in the high temperature electrolysis plan under the DOE Nuclear Hydrogen Initiative:

- Fabricate and operate an ILS half module
- Half-ILS Test Report
- First full ILS module fabrication and delivery

Accomplishments

- Constructed a 2 x 60 cell solid oxide electrolysis cell (SOEC) half-ILS module
- Operated the half-ILS module for over 2,000 hours
  - Doubled the 1,000 hour test life demonstrated earlier in CY 2006
- Demonstrated production rate of 1.25 Nm^3/hr hydrogen at start of life
  - A six-fold increase in production over previous stack tests
  - Eclipsing the planned production rate for the full thermochemical ILS
- Operated at 1.3 V/cell
  - an efficiency increase of 40% over conventional water electrolysis
- Demonstrated flexibility of co-electrolysis
  - directly converting CO_2 and steam to synthesis gas
- Delivered first full 4 x 60 cell SOEC module to INL

Introduction

The Department of Energy Office of Nuclear Energy is carrying out the Nuclear Hydrogen Initiative to develop methods through which nuclear energy can be used to split water into hydrogen and oxygen without the release of greenhouse gases. There are two types of processes for such hydrogen production, thermochemical cycles, using heat and chemical reactions, and electrolysis, where electricity and heat are used. This report documents the progress in the

development of High Temperature Electrolysis, in which steam at a temperature in the range of 800-850° C is split into hydrogen and oxygen using a solid oxide electrolytic cell. The specific test that was carried out June-September, 2006 used two stacks of 60 cells each in the configuration planned for the Integrated Laboratory Scale (ILS) experiment during FY 2007. The ILS will contain three modules of four stacks each, the first of which was delivered to the INL in March of 2007. The Pre-ILS test, which was fabricated and operated at Ceramatec in Salt Lake City, consisted of one half of one ILS module.

The primary goal of these tests has been to demonstrate the production of hydrogen in significant quantities using the ILS configuration. A secondary outcome was the demonstration of concurrent electrolysis (co-electrolysis) of steam and carbon dioxide to produce carbon monoxide and hydrogen, a mixture known as "synthesis gas." When synthesis gas is introduced over a catalyst at the proper temperature and pressure, a variety of hydrocarbons can be produced. Depending on temperature, pressure and catalyst, these hydrocarbons can range from methane to alcohols to diesel and jet fuels to wax. The technology for the use of synthesis gas is well-developed and has been used commercially. Virtually all of the hydrogen produced and consumed in the current multi-billion dollar version of the hydrogen economy is produced from natural gas by catalytic steam reforming, which emits CO_2. Much of this hydrogen is used in upgrading and refining of operations for transportation fuels. In contrast, high temperature electrolysis process is the most efficient means of hydrogen production, and rather than producing CO_2, it can recycle it back into transportation fuels. All that is required is an abundant supply of CO_2-free electric power.

Approach

High temperature steam electrolysis (HTSE) was accomplished by use of a solid oxide electrolysis cell (SOEC). An SOEC is virtually identical to a solid oxide fuel cell (SOFC). While Ceramatec has less than five years experience in the HTSE field, we can draw on over twenty years of active research and development in the SOFC field. The cells, interconnects, manifolds along with fabrication and testing infrastructure used in this test were all initially developed in the '90s and refined in the early part of this decade as fuel cell technology. Cells are electrolyte supported scandia stabilized zirconia, with screen-printed electrodes of proprietary compositions. Interconnects (bipolar plates) are stamped sheet metal using commercial alloys and proprietary coating processes. The test stand including reactant/product recuperative heat exchangers and steam generator was a modified SOFC test stand.

Results

Installation of the half-ILS module was completed and the test unit heated. Once the air inlet temperature had reached the 825°C setpoint, and stack temperatures had stabilized, the initial load sweep was started. A series of eight load steps was taken from open circuit to a stack voltage of 78 V (average 1.3 V/cell). The stack open circuit voltage of 49.4 V at the beginning of the sweep compared very closely to the theoretical value of 49.6 V calculated based on selected reactant flow rates. Alternate load steps were held for 20-30 minutes to allow thermal equilibration of the stack. The intermediate voltage steps were held only long enough to obtain a stable current and dew point reading. Once the full load voltage of 1.3 V/cell had been reached the stack was left under steady load. Initial full load performance was slightly better than expected. The calculated hydrogen production rate was 1.22 normal m^3/hr. At the beginning of full load operation, the stack operating point was 78.06 V, 48.74 A, with 70.96°C dew point, down from the inlet dewpoint of 93.2°C. Input power was 3.8 kW, with the stack under steady operation at a steady-state temperature.

Outlet product stream compositions were analyzed by gas chromatograph (GC) after approximately 18 hours of operation at 1.3 V/cell. Nitrogen flow in the stack gave an internal reference. The hydrogen product gas stream composition by CG (dry basis) is 96.5% H_2 and 3.5% N_2. Calculated compositions based on measured reactant feed rates and 48.74 A operating current is 96.0% H_2 and 4% N_2. The average measured dew point in the first 18 hours under load was a 71.3°C, with an average current of 48.63 A, while the calculated dew point for that condition is 70.3°C. Inlet dew point calculated from the inlet flow rates should be 92.0°C. The measured value was quite close, averaging 92.9°C with stack at open circuit voltage (OCV). The good correlation of calculated and observed dew point and GC data does seem to confirm that the seals are effective.

In an effort to halt stack degradation resulting from thermal management issues associated with the steam generator, the stacks were operated on a mixture of CO_2 and steam for about half of the 2,000-hour period of operation. The stack was taken from OCV to 78 V, showing about 31 A, compared with 34-35 A on steam alone. However, this was still well below thermal neutral voltage for CO_2/H_2O. The stack voltage was set to 82 V, based on the calculated thermal neutral voltage of 1.37 V, for a 50:50 H_2O:CO_2 mixture with 50% electrochemical conversion. This brought the four stack temperature readings into a much tighter grouping. The higher operating voltage also pushed the current to over 40 A, for a combined H_2+CO production rate of over 1.0 Nm^3/hr. Operation was relatively steady in the initial 24-hour period after this change, when the

product gas was again analyzed by GC. The gas samples in Table 1 show very repeatable results.

TABLE 1. Product Gas Composition (dry basis) at Beginning of CO_2 Operating Segment

Sample	Current	Dewpoint	$\%H_2$	$\%N_2$	%CO	$\%CO_2$
INL.7	40.4 A	58.4°C	44.5	0.42	33.2	21.7
INL.8	40.4 A	58.4°C	44.5	0.50	33.1	21.7

The module was operated without change in configuration or flow rates for the following six weeks. In this six weeks span, over 3,500 lb of CO_2 was processed, with a 60% conversion of carbon dioxide to fuel gas. Over 100 gallons of synthetic diesel fuel could have been produced using this syngas stream.

Once steam generator modifications were complete the system was returned to steam electrolysis operation (CO_2-free) for the remaining operating period. This later period of steam electrolysis operation showed very steady performance. A complete summary of the data with photographs and graphs have been presented in the DOE milestone reports and conference papers listed below.

Conclusions and Future Directions

Several important results have been obtained in carrying out this test. First, the Half-ILS module has shown that short stack performance is attainable in tall stacks. This result undoubtedly derived benefit from the periodic paralleling of cell groups as implemented in this configuration. Secondly, most of the module performance issues of concern in the test were related to the test system peripheral to the stack itself. In particular, given the Arrhenius response of the stack, temperature uniformity is crucial. The system issues must be resolved before a clear picture of stack performance and stability can be obtained. However, given the results of this test, acceptable initial performance has been demonstrated, leaving stack lifetime or degradation rate as the topic demanding a larger share of future resources.

The large quantity of hydrogen and syngas produced by this relatively compact module demonstrate the great potential of high temperature electrolysis to become a key solution to our current energy problems.

FY 2007 Publications/Presentations

Journal Articles

1. O'Brien, J. E., Stoots, C. M., Herring, J. S., and Hartvigsen, J. J., "Performance of Planar High-Temperature Electrolysis Stacks for Hydrogen Production from Nuclear Energy," in press, *Nuclear Technology*, Vol. 158, pp. 118 - 131, May, 2007.

2. Elangovan, S. Hartvigsen, J.J., and Frost, L.J., "Intermediate Temperature Reversible Fuel Cells," in press, *International Journal of Applied Ceramic Technology*, Vol. 4 [2], pp 109–118, 2007.

Conference Papers

1. O'Brien, J.E., Stoots, C.M., Hawkes, G.L., Herring, J.S., and Hartvigsen, J., "High-Temperature Coelectrolysis of Steam and Carbon Dioxide for Direct Production of Syngas: Equilibrium Model and Single-Cell Tests," submitted, Fifth International Conference on Fuel Cell Science, Engineering & Technology, June 18–20, 2007, New York, USA.

2. Stoots, C.M., O'Brien, J.E., and Hartvigsen, J., "Syngas Production Via High-Temperature Coelectrolysis of Steam and Carbon Dioxide In A Solid-Oxide Stack," submitted, Fifth International Conference on Fuel Cell Science, Engineering & Technology, June 18–20, 2007, New York, USA.

3. O'Brien, J.E., Stoots, C., Herring, J.S., and Hartvigsen, J., "High-Temperature Co-electrolysis of Carbon Dioxide and Steam for the Production of Syngas: Equilibrium Model and Single-Cell Tests," ANS Embedded Topical: International Topical Meeting on the Safety and Technology of Nuclear Hydrogen Production, Control, and Management, June 24 - 28, 2007, Boston, Massachusetts, USA.

4. Stoots, C.M., O'Brien, J.E., and Hartvigsen, J., "Test Results of High Temperature Steam/CO2 Coelectrolysis in a 10-Cell Stack," ANS Embedded Topical: International Topical Meeting on the Safety and Technology of Nuclear Hydrogen Production, Control, and Management, June 24 – 28, 2007, Boston, Massachusetts, USA.

5. Hawkes, G.L., O'Brien, J.E., Stoots, C.M., Herring, S.J., and Hartvigsen, J., "3D CFD Model of High Temperature H2O/CO2 Co-Electrolysis," ANS Embedded Topical: International Topical Meeting on the Safety and Technology of Nuclear Hydrogen Production, Control, and Management, June 24 – 28, 2007, Boston, Massachusetts, USA.

6. Hartvigsen, J., Elangovan, S., Stoots, C.M., O'Brien, J.E., and Herring, J.S., "Pre-ILS Demonstration Of Planar Solid Oxide Fuel Cell Technology Readiness For Application In Nuclear Hydrogen Production," ANS Embedded Topical: International Topical Meeting on the Safety and Technology of Nuclear Hydrogen Production, Control, and Management, June 24 – 28, 2007, Boston, Massachusetts, USA.

7. Herring, J.S., O'Brien, J.E., Stoots, C.M., Hartvigsen, J., Petri, M.C., Carter, J.D., and Bischoff, B.L., "Overview of High-Temperature Electrolysis for Hydrogen Production," ANS Embedded Topical: International Topical Meeting on the Safety and Technology of Nuclear Hydrogen Production, Control, and Management, June 24 – 28, 2007, Boston, Massachusetts, USA.

8. Elangovan, S. Hartvigsen, J., Heck, B., Larsen, D. and Timper, M., "Cell and Stack Testing of High Temperature Electrolyzers," American Ceramics Society, 4th International Symposium On Solid Oxide Fuel Cells (SOFC): Materials, Science, And Technology, 31st International Conference and Exposition on Advanced Ceramics and Composites, Daytona Beach, FL, January 2007.

9. Stoots, C. M., O'Brien, J. E., Hawkes, G. L., Herring, J. S., and Hartvigsen, J. J., "High Temperature Co-Electrolysis of H2O and CO2 for Syngas Production," 2006 Fuel Cell Seminar, paper no. 418, Nov. 13 – 17, 2006, Honolulu, HI.

10. O'Brien, J. E., Stoots, C. M., Herring, J. S., Hawkes, G. L., and Hartvigsen, J. J., "Thermal and Electrochemical Performance of a High-Temperature Steam Electrolysis Stack," 2006 Fuel Cell Seminar, paper no. 417, Nov. 13 – 17, 2006, Honolulu, HI.

11. Hartvigsen, J. J., Elangovan, S., O'Brien J. E., Stoots, C. M., and Herring, J. S., "Solid Oxide Fuel Cell Technology Demonstrations in the Nuclear Hydrogen Initiative," 2006 Fuel Cell Seminar, paper no. 140, Nov. 13 – 17, 2006, Honolulu, HI.

12. Hartvigsen, J. "Exchange Rates in the Hydrogen Economy," Uinta Headwaters RC&D (USDA) Renewable Energy Conference, Nov. 10, 2006, Park City, UT.

13. Elangovan, S., Hartvigsen, J., Stoots, C,. O'Brien, J., Herrring, J.S., and Frost, L., "Metal Interconnects for High Temperature Steam Electrolysis," Materials Science & Technology 2006, October 15–18, 2006, Cincinnati, Ohio.

14. Hartvigsen, J. J., Elangovan, S., Stoots, C. M., O'Brien J. E., Herring, J. S., and Frost, L.J., "Results From Recent High Temperature Electrolysis Testing at Ceramatec, Inc.," Hi2H2 Workshop on High Temperature Water Electrolysis, Sep. 18–19, 2006, Roskilde, Denmark.

15. Stoots, C. M., O'Brien, J. E., Hawkes, G. L., Herring, J. S., and Hartvigsen, J. J., "High Temperature Steam and Carbon Dioxide Electrolysis Experiments at INL," Hi2H2 Workshop on High Temperature Water Electrolysis, Sep. 18–19, 2006, Roskilde, Denmark.

DOE Milestone Reports

1. Hartvigsen, J.J., and Herring, J.S., "Initial Dual-stack, Half-Module Test Results, in preparation for the Integrated Laboratory Scale Experiment, June – September 2006," September 20, 2006.

Special Recognitions & Awards/Patents Issued

1. Patent Applications: Several patent applications have been submitted or are in preparation. None have been issued. Details will be made public as patents are issued.