II.G.4 Laboratory-Scale High Temperature Electrolysis System

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Start Date: January 23, 2003
Projected End Date: Engineering Demo, 1 MW, 2018

Objectives

- By 2012, reduce the cost of central production of hydrogen from high temperature electrolysis to $3.10/gasoline gallon equivalent (gge) at plant gate, by 2017, reduce the cost of central production of hydrogen from high temperature electrolysis electrolysis to <$2.00/gge at plant gate.*
- 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.

** Since there are no objectives specific to the nuclear production of hydrogen, we have adopted those cited from wind water electrolysis.*

Technical Barriers

This project addresses the following technical barriers of the Multi-Year Program Plan p. 3.1-24 and the Nuclear Hydrogen Initiative:

- **G. Capital Cost** – Development of larger systems is also needed to take advantage of economies of scale.
- **H. System Efficiency** – New membrane, electrode and system designs are needed to improve system efficiency.
- **I. Grid Electricity Emissions** – Low-cost, carbon-free electricity generation is needed. Electrolysis systems that can produce both hydrogen and electricity need to be evaluated.

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: $3.10/kg – centralized production only, 2012.
- Life of cells: 20,000 hours of continuous operation.

Contribution to Achievement of DOE Systems Analysis Milestones

This project contributed/will contribute to achievement of the following DOE systems analysis milestones from the Systems Analysis section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- **Milestone 2:** Complete baseline economic, energy efficiency and environmental targets for fossil, nuclear and renewable hydrogen production and delivery technologies. (4Q, 2005)
- **Milestone 8:** Complete analysis and studies of resource/feedstock, production/delivery and existing infrastructure for technology readiness. (4Q, 2014)
- **Milestone 10:** Complete an analysis of the hydrogen infrastructure and technical target progress for technology readiness. (2Q, 2015)
- **Milestone 11:** Complete environmental analysis of the technology environmental impacts for the hydrogen scenarios and technology readiness. (2Q, 2015)

Accomplishments (Fiscal Year 2009)

- Demonstrated improved performance of oxygen and steam hydrogen electrodes through the deposition of electrocatalytic materials by atomic layer deposition or other infiltration processes. (Argonne National Laboratory) 9/30/2008
- Completed analysis of hot oxygen handling and cooling options based on collaboration with Atomic Energy Canada, Limited. 7/2/2008
- Tested the three-module (each module 4 x 60 cells) for the ILS experiment for 1,080 hours in Idaho Falls with a maximum output of 5.65 N m^3 H_2/hour (0.504 kg H_2/hour). 9/5/2008-10/20/2008
- Completed analysis report on corrosion testing of the second series of balance of plant materials on 9/30/2008.
- Completed report on experimental data from the operation of the High Temperature Electrolysis (HTE) ILS experiment on 2/17/2009.
- Completed report on critical causes of stack degradation in ILS cells on 5/27/2009.
- Demonstrated low-degradation performance of a 10-cell stack using improved oxygen electrodes, 5/27/2009-9/8/2009.

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Introduction

A research program 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 program includes both experimental and modeling activities. 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. In the last year we have begun testing various electrode-supported configurations as well.

During FY 2009, experimental results were obtained from three modules of four 60-cell stacks each, fabricated by Ceramatec, Inc. Additional experiments at ANL, MIT, and INL investigated the characteristics of cells used in previous tests and the performance of a variety of materials being considered for use in the balance of plant for HTE hydrogen production plants. We also organized a workshop on the long-term degradation of solid oxide cells used in high temperature electrolysis in conjunction with the Fuel Cell Seminar in Phoenix, October 27, 2008.

Approach

High-temperature electrolysis testing of single button cells and stacks has been in progress at the INL and the subcontractor Ceramatec Inc. (Salt Lake City, UT) since 2003. The scales of testing accomplished to date at the INL are pictured in Figure 1. The ILS facility operated at a maximum hydrogen production rate of 5,650 normal (273°K, 1 atm) L/hr (0.504 kg H_2/hr) with an input of 18 kWe. The ILS operated continuously for 1,080 hours from September 5 through October 20, 2008. Because of degradation of the oxygen electrodes, the hydrogen output decreased to 800 NL/hr in the course of the test.

After each of the tests, the stacks have been disassembled and the cells have been destructively examined to determine elemental migration during the test and to detect changes in the morphology of the electrolyte and electrodes. These examinations have also been conducted on cells in the test of the three-module ILS.

FIGURE 1. General View of the HTE ILS Experiment

Results

The full, three-module ILS experiment operated in Bay 9 of the Bonneville County Technology Center, Idaho Falls, from September 5 through October 20, 2008, a total of 1,080 hours [1]. A 420-hour test of the ILS with a single module was conducted in September and October 2007 [2]. The general layout of the experiment is shown in Figure 1, with the numbered components listed in Table 1.

TABLE 1. Identifiers for Figure 1

ID	Component
1	Hot zone enclosure lid
2	Power supply and instrument racks
3	Electrical distribution cabinet
4	Data acquisition and control monitors
5	Deionized water system
6	Steam generator
7	Steam and H_2 superheaters
8	Air compressor
9	Patch panel
10	Product finned cooler
11	Steam condenser
12	Mass flow controllers
13	H_2 vent
14	Air and O_2 vent

As can be seen in Figure 1, the ILS contains most of the components needed in a full-scale high temperature electrolysis plant, with exception of the nuclear reactor and facilities for using the hydrogen produced. The three modules, with the associated voltage taps and thermocouples are shown in Figure 2a, immediately before the closure of the hot zone lid.

Two capabilities were added to ILS between the single module and the three-module tests. In order to more closely simulate the operation of a commercial hydrogen plant, we added a system for recycle of 10-30% of the product hydrogen back into the input steam to maintain reducing conditions on the steam/hydrogen electrodes of the cells within the modules. Secondly, we installed heat recuperators under the modules to allow the use of steam at 350° to 450°C, rather than the 800°C required at the input to the modules. As shown in Figure 2b, the heat in the hydrogen and oxygen product streams at ~820°C is used to heat the input streams to ~790°C. The inherent ohmic heating in the cells provides the required 20°-40°C temperature difference between the countercurrent streams.

Heat up of the INL three-module ILS system began at approximately 5 PM on September 4, 2008. Peak hydrogen production as well as peak electrolysis power input was achieved at approximately 10 AM on September 5, 2008 (~17 hours elapsed test time). At this time, the electrolysis modules were consuming 18 kW of electrical power and producing over 5.7 Nm^3/hr H_2. Early test module area-specific resistance (ASR) values ranged from 1.25 Ω-cm^2 to just over 2 Ω-cm^2. Testing continued for 1,080 hours. Significant module performance degradation

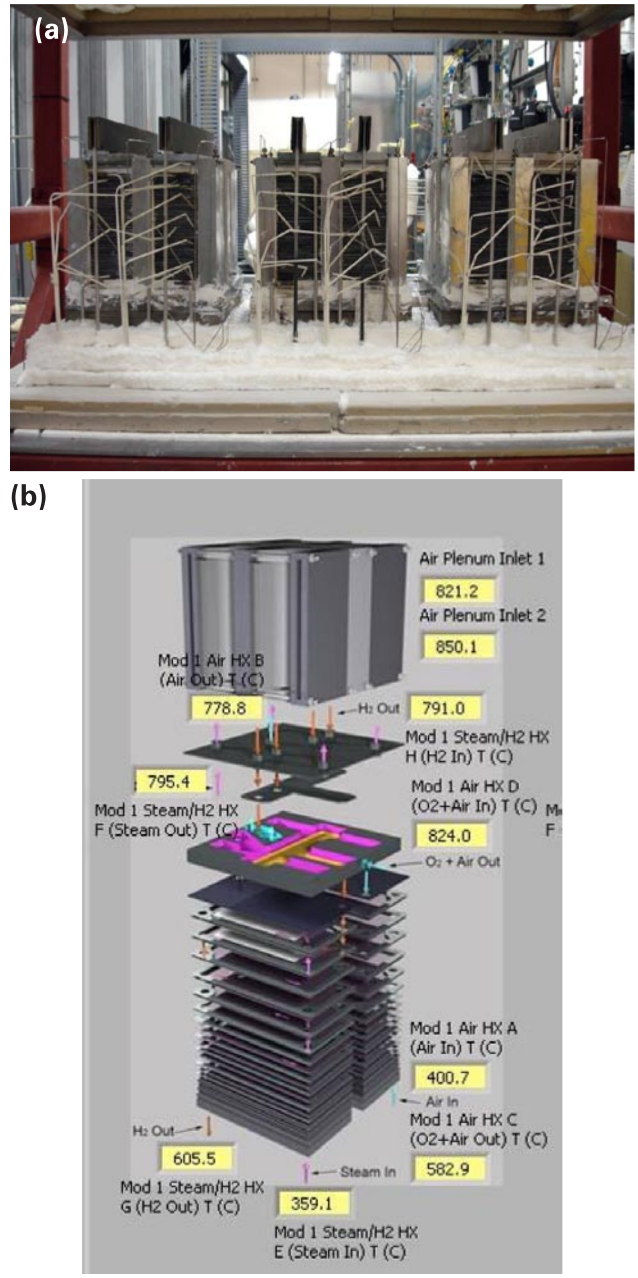

FIGURE 2. (a) Three modules each containing four stacks of 60 cells. The six recuperators are partially covered in insulation below the modules. (b) A screen print from our LabView® control software, showing the temperatures of the various streams entering and leaving the two recuperators under module 1.

was observed over the first 480 hours, after which no further degradation was noted for the remainder of the test for modules 1 and 2, while module 3 continued to degrade. The total H_2 production rate decreased from 5.7 Nm^3/hr to a steady-state value of 0.7 Nm^3/hr, primarily due to cell degradation. Most of the test was conducted at a constant electrolyzer power supply voltage of 76 V.

Once all test objectives had been successfully met, the test was terminated in a controlled fashion. Two relatively minor problems were encountered during testing. First, one N_2 mass flow controller failed and was replaced with a rotameter without any disruption of the experiment. Second, water condensation occurred in the H_2 recycle system. This was due to the elevated pressure required in the H_2 recycle loop. The condensate would sometimes collect in and interfere with the inlet H_2 mass flow controllers, leading to periodic interruptions in the hydrogen recycle flow. This may have caused accelerated degradation of the ILS modules. In particular, the flow interruptions could have caused electrode oxidation to occur in the modules. Once H_2 flow resumed, reduction would occur on the electrode surfaces. This cycling of oxidation and reduction may have contributed to the high rate of performance degradation observed. Around 480 hours elapsed test time, the condensate problem was finally fixed by installing a water trap just upstream of the H_2 mass flow controllers. Subsequent to this, the ASR values for modules 1 and 2 improved and finally leveled out with no further degradation. The reason for the continued performance degradation of module 3 after the condensation problem was solved while modules 1 and 2 improved is not understood.

The effectiveness of the heat recuperation system was estimated. A comparison was made between heater loads, steam generator loads, and hot zone heat losses between the earlier single module test (no heat recuperation) and the present three module test (with heat recuperation). The total per-module electric heater power was reduced by 50% in the case of heat recuperation.

Once the condensation problem was solved, the hydrogen recycle system worked flawlessly and made unnecessary the use of any external hydrogen source for maintaining reducing conditions in the cells.

The facility instrumentation and data acquisition/control system performed as designed except for condensation interfering with the inlet hydrogen mass flow controllers and failure of one nitrogen mass flow controller. Both problems were circumvented/solved without any disruption of the experiment.

Overall, the facility successfully demonstrated the largest scale high temperature solid oxide-based production of hydrogen to date.

Post-test examination of the representative cells from the three-module ILS test indicated that the oxygen electrode had delaminated from the electrolyte over part of the electrolyte area [3]. After a series of short-term tests of various diffusion-barrier layers in the cells, a long-duration test of a 10-cell stack was started May 27. As of this writing, that stack has operated ~1,000 hours and has shown only about 5% degradation. It is expected that this test will run until the end of FY 2009.

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