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TEST RESULTS FROM THE IDAHO NATIONAL LABORATORY 15KW HIGH TEMPERATURE ELECTROLYSIS TEST FACILITY

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ABSTRACT

A 15 kW high temperature electrolysis test facility has been developed at the Idaho National Laboratory under the United States Department of Energy Nuclear Hydrogen Initiative. This facility is intended to study the technology readiness of using high temperature solid oxide cells for large scale nuclear powered hydrogen production. It is designed to address larger-scale issues such as thermal management (feed-stock heating, high temperature gas handling, heat recuperation), multiple-stack hot zone design, multiple-stack electrical configurations, etc. Heat recuperation and hydrogen recycle are incorporated into the design. The facility was operated for 1080 hours and successfully demonstrated the largest scale high temperature solid-oxide-based production of hydrogen to date.

INTRODUCTION

The world's present transportation system has developed using light (i.e. low viscosity), sweet (i.e. low-sulfur) petroleum which can be easily refined into gasoline, diesel and jet fuels. Over the last century, the world has consumed the majority of those light sweet petroleum resources (conventional oil) and has had to turn more and more to heavier, higher-sulfur crude oil reservoirs (unconventional oil). The addition of hydrogen to the crude oil during the refining process is the primary method for breaking the large, viscous hydrocarbon molecules into smaller pieces and for the removal of sulfur. Historically, the necessary hydrogen has been produced primarily through a catalytic reaction between steam and natural gas, known as Steam Methane Reforming.

In 1980 we used about 0.7 kg of H₂ to upgrade and refine a barrel of crude, rising to 1.4 kg of H₂ per barrel in 2005. Thus the refining of the present US petroleum demand, 20 million barrels per day, requires about 10 million tons of H₂ annually, consumes 5% of our total annual natural gas, and releases 58 millions tons per year of CO₂. In the immediate future, more and more unconventional sources of liquid fuels, such as very heavy crude oils, oil sands, oil shale, and coal, will require increased amounts of hydrogen for their upgrading. For example, the Athabasca oil sands in Alberta require 3-4 kg of H₂ per barrel for upgrading and refining. Coal-to-liquids processes require 5-8 kg of H₂ to produce useable transportation fuels. The amounts of CO₂ released in the course of producing the required H₂ via either steam-methane reforming using natural gas or the water-gas shift reaction using coal are major obstacles to the implementation of coal-to-liquids processes. Because of the additional CO₂ releases in the production of hydrogen using natural gas or coal, the CO₂ emissions per vehicle mile using proposed coal-to-liquids technologies are double the releases using light sweet crude oil.

Regardless, conventional as well as unconventional world oil production will eventually peak and subsequently decline. Predictions of when this peak will occur are controversial and difficult to make due to geological complexities, measurement problems, pricing variations, demand elasticity, and political influences [1]. However, the impact of peak oil could be tremendous and unprecedented. In the present, demands upon

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existing reserves and production capacity are growing, primarily due to industrializing nations such as China and India [2]. This has resulted in instabilities in crude oil and gasoline prices, along with worries concerning how reliable crude oil supplies are against natural disasters, wars, and terrorist attacks. Finally, there are the climate change effects of oil usage. Most of the United State's energy-related CO₂ emissions come from oil. These issues have been driving the development of a non-carbon, renewable, H₂-based energy infrastructure [3].

The demand for H₂ has been increasing by 10% per year and there is a strong interest in developing hydrogen as a second energy carrier for the non-electrical market. The goals of a hydrogen-based energy economy are reduced oil consumption, foreign energy independence, and reduced greenhouse gas emissions. Since hydrogen is an energy carrier and not an energy source, attaining these goals is conditional upon development of suitable renewable energy sources and/or nuclear energy to power water-splitting technologies for carbon-free hydrogen production.

Water-splitting for hydrogen production can be accomplished via high-temperature electrolysis or thermochemical processes, using high-temperature nuclear process heat. In order to achieve competitive efficiencies, both processes require high-temperature operation. High-temperature electrolytic water-splitting supported by nuclear process heat and electricity has the potential to produce hydrogen with an overall system efficiency near those of the thermochemical processes [4], but without the corrosive conditions of thermochemical processes and without the fossil fuel consumption and greenhouse gas emissions associated with hydrocarbon processes.

The Idaho National Laboratory (INL), in conjunction with Ceramtec Inc. (Salt lake City, USA) has been researching the use of solid-oxide fuel cell technology to electrolyze steam for large-scale nuclear-powered hydrogen production. The scope of activities includes computational fluid dynamics modeling [5], process flow sheet analyses, and experimental testing [6, 7, 8, 9]. Experimental testing has followed a logical progression in scale. Button cell (~2 watt) and short stack (~500 watt) tests have primarily concentrated upon quantifying material and cell performance and have not addressed larger-scale issues such as thermal management (feed-stock heating, heat recuperation, and high-temperature gas handling), hydrogen recycle, multiple-stack hot-zone design, multiple-stack electrical configurations, and other "integral" issues. For example, in button cell and bench-scale stack testing, steam is introduced into the inlet gas stream by saturating a carrier gas via a heated humidifier. Furthermore, the cell or stack is located inside of a furnace and the inlet gases are heated to the stack inlet temperature by the same furnace. This approach for steam production and feed-stock heating is not realistic for larger scales of electrolysis.

An integrated laboratory scale (ILS) high-temperature electrolysis facility has been developed at the Idaho National

Laboratory under the DOE Nuclear Hydrogen Initiative. The ILS facility is designed to address larger-scale issues not addressed at the smaller bench scale. Initially, a single 5 kW solid oxide module was used for the first operational tests. This module was comprised of 4 stacks of 60 64 cm² cells each. No heat recuperation or hydrogen recycle was incorporated. Initial operation of this facility was in 2007 and resulted in over 400 hours of operation with an average hydrogen production rate of approximately 0.9 normal cubic meters per hour (Nm³/hr) [10].

Subsequently, the ILS facility has been expanded to a design-based nominal hydrogen production rate of 14.1 kW based on lower heating value (LHV, equal to 120 MJ/kg for hydrogen), or 4735 Normal (273°K, 1 atm) L/hr [10]. Expansions included inclusion of 3 modules rather than 1, incorporation of 3 gas heating and delivery loops versus 1, addition of hydrogen recycle, and addition of 3 pairs of recuperative heat exchangers (one pair for each module loop).

Solid-oxide electrolyzer cells are comprised of a variety of materials (ceramic, cermets, as well as metals) bonded together. These bonds include metal brazing and ceramic seals. Mismatches in thermal expansion coefficients between the various materials can cause stresses and result in weakening or cracking in the seals as the cells are either heated or cooled. Added to this is the high capital investment in solid-oxide cells for this test facility. To help assure highest performance of the cells, thermal cycling of the cells is to be avoided. Most solid-oxide cell applications are large-scale and stationary in nature, and are designed to avoid thermal cycling.

Testing of the INL ILS experimental facility with three modules commenced on September 4, 2008. Testing continued for 1080 hours. The test average H₂ production rate was approximately 1.2 Nm³/hr based upon current, with a peak measured value of over 5.7 Nm³/hr based upon current. Significant module performance degradation was observed over the first 480 hours. Some of this degradation was due to condensation in the hydrogen recycle loop affecting the performance of the hydrogen mass flow controllers and hence module inlet hydrogen mass flow rates. This problem was corrected at approximately 480 hours elapsed test time. Beyond 480 hours, modules 1 and 2 showed no further degradation while module 3 continued to show degradation in performance. Once all test objectives had been successfully met, the test was terminated in a controlled fashion.

OVERVIEW OF FACILITY

A brief introduction to the facility is presented here. A more comprehensive description may be found in [11]. The piping and instrumentation schematic for the ILS single-module experiment with no heat recuperation or hydrogen recycle is shown in Fig. 1. The electrolysis module requires a support system supplying electrical power for electrolysis, a feedstock gas mixture of hydrogen and steam, a sweep gas, and appropriate exhaust handling. In particular, this system must

HIGH TEMPERATURE ELETOLYSIS INTEGRATED LABORATORY SCALE P & ID

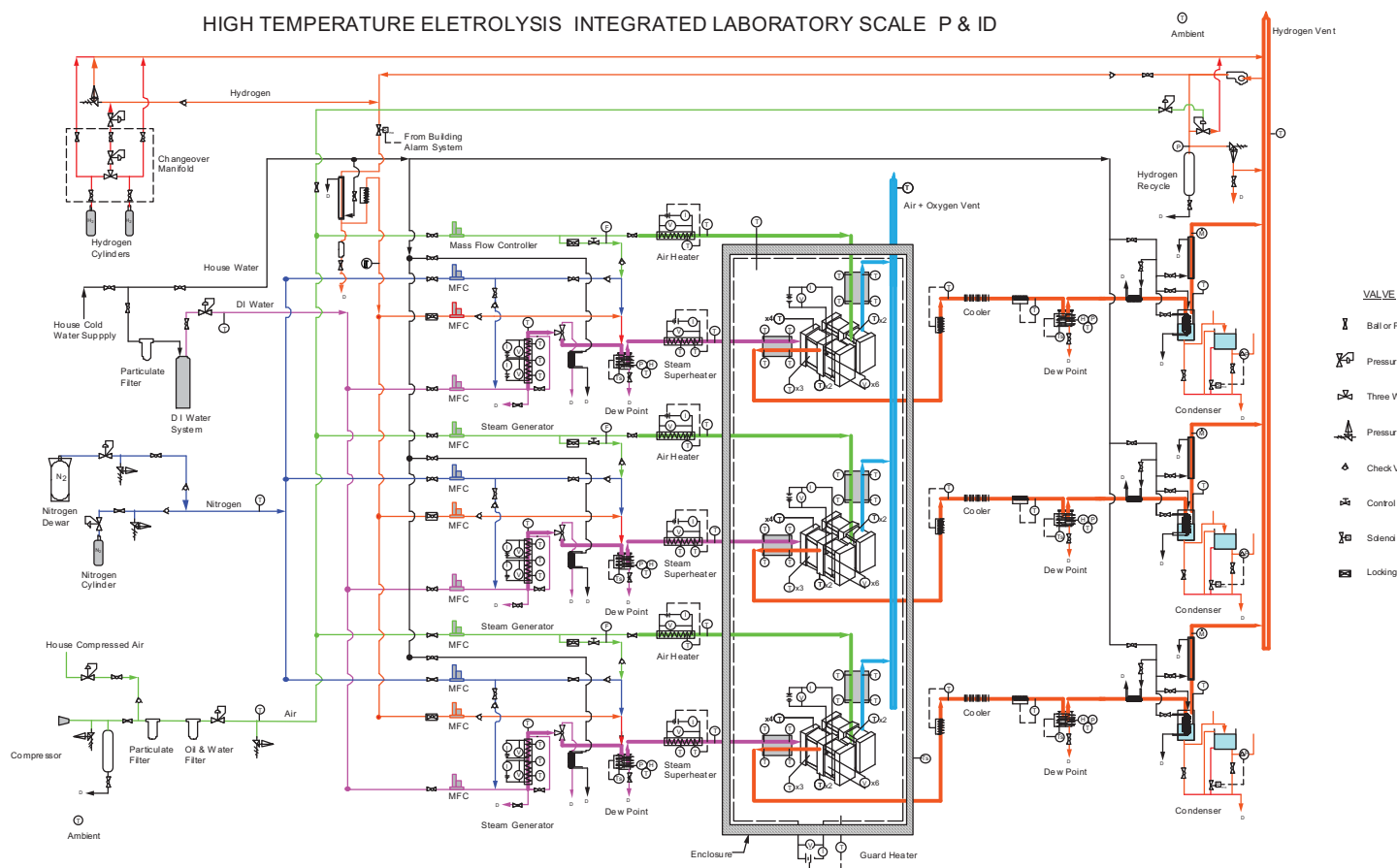


Figure 1. ILS piping and instrumentation schematic.

include means for controlled steam generation, mixing hydrogen with the steam, feedstock and product dewpoint measurements, heating the feedstock and sweep gas to the appropriate electrolysis temperature (via a superheater), cooling the electrolysis product stream, condensing any residual steam out of the product stream, and venting the hydrogen product. Since three electrolysis modules (1 module consists of 4 stacks of 60 cells each) were tested, the ILS support system actually consists of three parallel systems that supply feedstock, sweep gas streams, and electrical power basically independent of each other to each of the modules. All three modules were located within a single hot zone. Heat recuperation and hydrogen product recycle was also incorporated into the facility. To aid in interpretation of Fig. 1, the hydrogen / steam feedstock is represented by the color magenta, the product stream by orange, the inlet sweep gas by green, and the outlet sweep gas by blue.

Gas handling and basic components

Liquid water feedstock is fed at a controlled rate into the system by means of a Bronkhorst CORI-FLOW coriolis mass flow meter / controller. The water is then vaporized in an inline electrically-powered boiler. The boiler was fabricated by attaching a combination of twenty 200 and 300 watt clamp-on

electric heaters to the outside of a 1" diameter stainless steel tube. The heaters are covered with 2" of thermal insulation, then topped by an aluminum covering. The heaters are spaced such that a higher heat flux is obtained in the boiling region and lower heat flux in the single-phase regions. The heaters are all wired in parallel so that each operates at the same voltage. The tube interior is filled with a copper foam material which reduces flow perturbations and increases temperature uniformity in the boiling region. The saturated steam is immediately slightly superheated by a separately controlled electrically-powered heater. The outlet temperature is controlled by carefully adjusting the input power supplied by a DC power supply to obtain the desired superheat temperature. The steam mass flow rate is verified by monitoring the rate of electrical energy supplied to the boiler from the DC power supply.

The slightly superheated steam exiting the heater is mixed with hydrogen, which is required on the inlet side of the stack in order to maintain reducing conditions at the steam/hydrogen electrode. Nitrogen can also be introduced here and is sometimes used to increase the average molecular weight of the gas mixture and hence reduce diffusive losses (leaks) in the electrolysis modules. During startup, the inlet hydrogen is supplied from a compressed gas bottle. Once steady-state

electrolysis conditions are achieved, the inlet hydrogen is supplied via recycle of a fraction of the hydrogen product. In either case, the hydrogen flow rate is controlled by a Bronkhorst gas mass-flow controller and the data acquisition / control system. The recycle stream must be pressurized to overcome the significant pressure drop incurred by the mass-flow controller. Downstream of the mixing point, the temperature, pressure, and dewpoint of the steam/hydrogen gas mixture are measured. The absolute pressure is directly measured at the dewpoint measurement station in order to allow for accurate determination of the steam mole fraction. Precise measurement of the dewpoint and pressure allows for independent determination of the inlet gas composition.

A high-temperature electrically powered inline superheater then boosts the feedstock stream to approximately 350° - 450°C. Heat is supplied from six semi-cylindrical ceramic-fiber heaters with embedded coiled elements. Each heater section is capable of providing 1800 watts of power when operated at 240 volts, but they are operated at a much lower voltage for this application. Power is supplied to the heaters

from 3.3 kW DC power supplies. Heater power is feedback-controlled based on thermocouples located inside the ceramic fiber heaters. Two inch thick high-temperature thermal insulation is wrapped around the heaters and covered with an aluminum skin.

The primary material for process tubing is 316 stainless steel. An exception is the steam and air superheaters, where Inconel 600 is used to minimize high temperature corrosion.

The electrolysis modules and heat recuperators are mounted in the hot zone enclosure where they are maintained at the desired operating temperature using radiant heaters installed in the sides and top of the removable lid. As explained in references [12, 13], when the electrolysis process is operated below the thermal neutral voltage ($V_{in} = 1.287$ V/cell for 800°C operating temperature), heat must be added to overcome the endothermic reaction heat requirement. At thermal neutral conditions, the module operation is adiabatic and isothermal. If, however, the module is operated above the thermal neutral voltage, heat must be removed from the system.

The heat recuperators heat the inlet process gases to the



Figure 2. Right side view of INL ILS facility, with major components labeled.

Table 1. Identifiers for Figs. 3-5.

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

electrolyzer operating temperature of 800-850°C via heat transfer with the outlet process gases. In a large-scale high-temperature electrolysis plant, heat recuperation will be

absolutely critical in order to minimize the plant net heat requirement and maximize the overall hydrogen production efficiency. In the ILS, incorporation of heat recuperation is also beneficial from an operational standpoint because it reduces the required outlet temperature of the electrically heated steam and air superheaters and it reduces the thermal load on the hot zone metal base plate. An internally manifolded plate-fin design concept was selected for the heat recuperator application. This design provides excellent configuration flexibility in terms of selecting the number of flow elements per pass and the total number of passes in order to satisfy the heat transfer and pressure drop requirements. Theoretical counterflow heat exchanger performance can be approached with this design.

The gas mixture exiting the electrolyzer will be significantly enriched in hydrogen, typically to at least 50% hydrogen mole fraction, with the remainder being residual steam. The product stream is first cooled via a natural-convection air-cooled heat exchanger. The product stream temperature exiting this cooler is controlled such that no condensation can occur. Then the product gas mixture enters the outlet dewpoint measurement station. The measurement of both inlet and outlet dewpoint temperatures allows for direct determination of the steam consumption rate, and the corresponding hydrogen production rate. This rate can be compared to the electrochemical hydrogen production rate

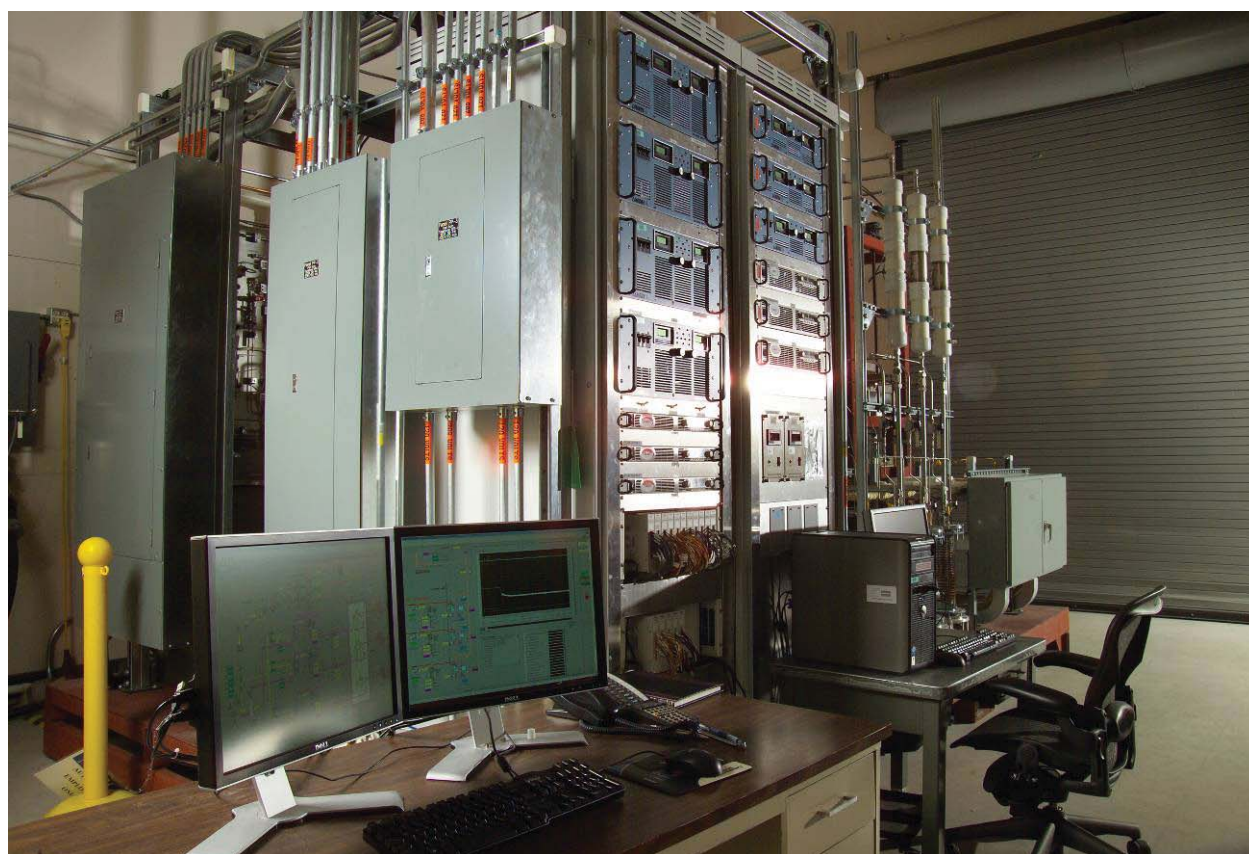


Figure 3. Left side view of INL ILS facility, with data acquisition computer monitor and power supply racks in foreground.

determined from the stack electrical current.

The outlet hydrogen/steam flow then enters a condenser where the majority of the residual steam is removed. The rate of water condensation is monitored via tank level, providing an additional independent measure of steam consumption. At this point, the product stream is ambient-temperature, saturated hydrogen gas, with about 3.7% residual water vapor. Nitrogen may also be present, as explained earlier. The flow rate of this product gas is measured with a low-pressure-drop mass flow transducer. Comparison of the condensate and hydrogen product mass flow rates with the electrolyzer inlet mass flow rates helps quantify any stack leakage that may occur. Most of the hydrogen product is then vented from the building, and the remainder pressurized and recycled to the inlet steam feed.

Air is used as a sweep gas to remove excess oxygen from the ILS system. Filtered compressed air flows through a mass-flow controller and into an electrically-powered heater to preheat the inlet air to the stack operating temperature. Downstream of the electrolyzer, the hot oxygen-enriched air stream is then vented from the building to the environment.

All of the system components and hardware were mounted on a skid that is 16 ft. long by 10 ft wide. Photographs of the skid with the components identified are presented in Fig. 2. The components are listed in Table 1 by identification number. A custom LabView (National Instruments) program was developed for ILS data acquisition and instrument control using SCXI data acquisition hardware. This is pictured in Fig. 3, along with the racks of power supplies.

Nitrogen gas can be injected into the water feed upstream of the boiler or directly into the gas stream upstream of the steam superheater. Air can be introduced into the steam-side feed stream downstream of the boiler. These capabilities are primarily used during heatup of the modules. From room temperature until approximately 400°C, air is flowed through both sides of the modules (steam side and oxygen evolution side). This is done to ensure complete combustion of any organic binders used during stack fabrication. From 400°C to approximately 600°C, a dry mixture of nitrogen and hydrogen is flowed through the side of the modules, and air is continued through the oxygen evolution side. Transitioning from air to the dry nitrogen hydrogen mixture requires purging the steam side of any air with a pure nitrogen flow.

Finally, from 600°C to 800°C, a mixture of nitrogen, hydrogen, and ~3% water is flowed through the steam side, with air through the oxygen evolution side. These latter flows help to ensure full reduction of the steam side electrode.

Data acquisition and control

A National Instruments SCXI – USB data acquisition and control system was used to collect data and provide control signals for operation of the ILS. 233 channels of data was collected and stored to disk at rates up to 1 Hz. The system included 28 independent PID feedback control loops. These feedback loops included the various heaters, steam generators, recycle pump, and condensate collection. When the ILS single

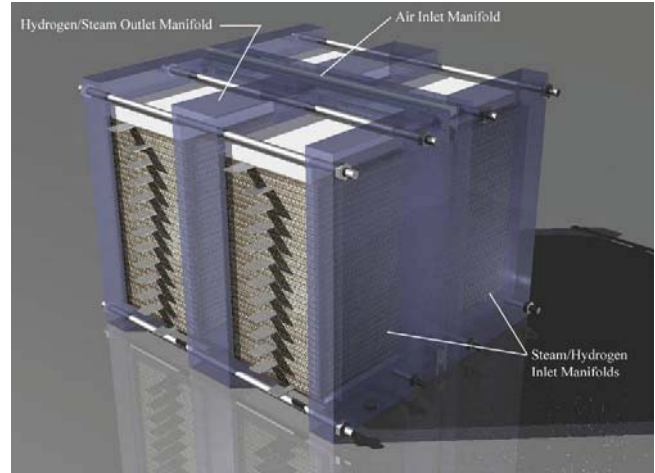


Figure 4. ILS 4-stack module.

module test was performed in the summer of 2007, only one SCXI – USB chassis was used for all signal measurement and control. Some problems arose in the first test concerning the internal stack thermocouples. These thermocouples are 0.020" OD, inconel-sheathed, ungrounded, Type K. They must be ungrounded since they are inserted into the middle of the electrolysis module cells and hence are exposed to the electrolysis voltage potential. Although the cell-to-cell potential is only 1 to 2 volts, the cell-to-ground potential may be as high as 80 volts. Long exposure to high temperatures (830°C) caused the thermocouple internal insulation to break down, losing electrical isolation between the sheath and the internal thermocouple wires, and causing a high bias voltage to be imposed on the thermocouple signal as well as thermocouple failure. This bias voltage exceeded the rating for the DAS and caused erratic behavior in the SCXI until the culprit thermocouple could be disconnected.

To minimize any bias voltage problems in the full ILS test, the measurement and control channels were split between two SCXI chassis. One SCXI chassis handled all measurements that were susceptible to high bias voltages. In other words, any measurements in direct contact with the electrolysis cells: stack operating voltage, stack intermediate voltages, and internal stack temperatures. All other measurements and all control signals were handled by a second SCXI chassis. In this manner, it is impossible for any bias voltage to affect measurements or control signals in the other chassis.

Control of the above multi-SCXI system was implemented using a personal computer and an INL-developed custom program using the National Instruments LabView software. In addition to the SCXI system, the INL-developed software also coordinated communication and control of the 12 serial interface based mass flow controllers as well. The software provided a graphical user interface for real-time operation and control of the ILS, provided on-screen real-time graphical data representation, and logged experimental test data to disk. Logged data was written in Excel spreadsheet format to simplify post-test data analysis.

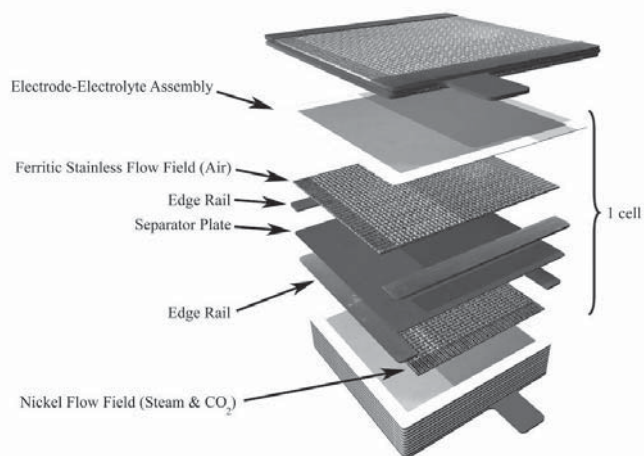


Figure 5. Diagram of solid-oxide stack components.

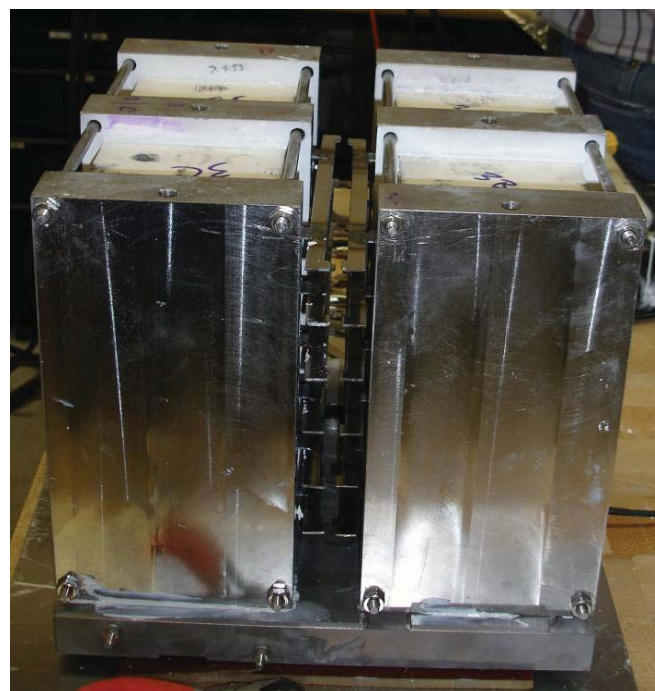


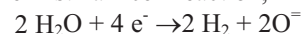
Figure 7. Two ILS half modules, showing electrical interconnections and air plenum space.

ELECTROLYSIS MODULES

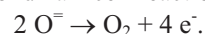
Stacks used for testing by the INL are fabricated by Ceramtec, Inc., of Salt Lake City, UT. They have an active area of 64 cm^2 per cell, providing a total active area of 3840 cm^2 in a sixty-cell stack. Four stacks comprise a module. Three modules were installed and tested, totaling 720 cells. The modules are designed to operate in cross flow, with the steam / hydrogen gas mixture entering the inlet manifolds on the right and left sides in Fig. 4, and exiting through the outlet manifold visible in Fig. 4. Airflow enters at the rear through an air inlet manifold (Fig. 4) and exits through the front and back open faces directly into the hot zone enclosure.

The electrolyzer is basically a solid oxide fuel cell run in reverse. In the electrolysis cell, steam and a small amount of hydrogen (to maintain reducing conditions at the nickel-

zirconia electrode) are introduced at one edge of the planar cell. Steam diffuses to the interface between the electrode and the electrolyte, where the first half-cell reaction,



takes place. The oxygen ions carry the electrons through the gas-tight solid-state electrolyte to the electrolyte / anode interface, where the second half-cell reaction occurs:



Construction Details

The internal components of the stack are shown in Fig. 5 and are comprised as follows. The interconnect plate is fabricated primarily from 441 stainless steel. It includes an impermeable separator plate with 441 stainless edge rails and two corrugated flow fields, one on the sweep-gas side and one on the steam / hydrogen side. The edge rails were glued and sealed to the separator plate using a high surface area YSZ paste. The height of the flow fields is 1.0 mm. Each flow field includes 32 perforated flow channels across its width to provide uniform gas-flow distribution. The steam / hydrogen flow fields are fabricated from nickel 201 foil. The air-sweep flow fields are made from Hast-X steel. The interconnect plates and flow fields also serve as electrical conductors and current distributors. To improve performance, the sweep-side separator plates and flow fields are surface-treated to form a rare-earth stable conductive oxide scale. A perovskite rare-earth coating is also applied to the separator plate oxide scale. This was one area where the 3 modules differed from each other. The stacks of module 3 were assembled with CuMn (oxide) ink rolled onto the cell side of the Hast-X (air side)



Figure 6. One-half of ILS module showing electrical interconnections.

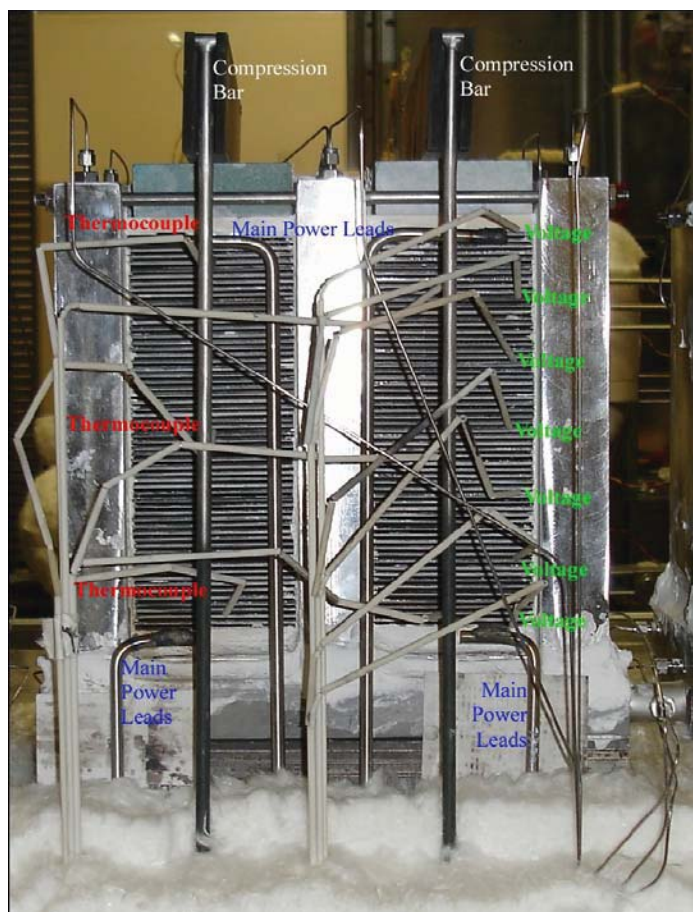


Figure 8. Final installation of one module with instrumentation and power attachments.



Figure 9. HTE ILS hot zone with three modules installed.

of stack assembly. So perhaps there were some seasonal (humidity) effects in tape casting and the sequence of ink batches. Ceramtec has maintained logs of the ink batches and where they went in the stacks. Also, the module #3 manifolds were very tight fitting (too much plasma spray alumina) and had to be ground a bit to fit. All of the inlet manifolds had to have the inlet port bored eccentric to fit on the base plates (base plate stubs were too close for the assembled stacks).

The electrolysis module is composed of four stacks, each consisting of sixty cells. Two stacks with individual inlet plenums are connected together with a common outlet plenum to form a half-module (Fig. 6). To preclude the loss of an entire stack if a single cell fails, the four stacks are electrically interconnected at every fifth cell. This is done by first interconnecting the pair of stacks in each half module, as

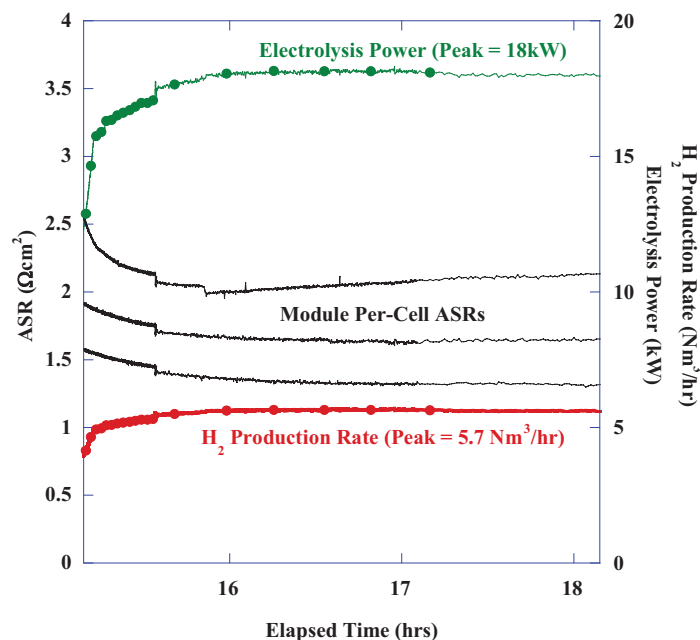


Figure 10. Peak electrolysis power and H₂ production rate.

flow field in place of LSCo ink that is normally used there. This was done to see if it was effective as a Cr transport barrier. The 441 and Hast-X parts were all treated with a rare earth scale stabilization layer underneath a LaCr perovskite conductive coating. On the steam / hydrogen side of the separator plate, a thin nickel metal coating is applied.

The electrolyte is scandia-stabilized zirconia, ~220 μm thick. The sweep-side electrode (anode in the electrolysis mode) is a strontium-doped manganite. The electrode is graded, with an inner layer of manganite/zirconia immediately adjacent to the electrolyte, a middle layer of manganite, and an outer bond layer of cobaltite. The steam / hydrogen electrode (cathode in the electrolysis mode) is also graded, with a nickel cermet layer immediately adjacent to the electrolyte and a pure nickel outer layer. Total average cell thickness (not including flow fields or interconnect) is approximately 300 μm.

Manifolds were Fecralloy (an alloy of approximately 20% Cr, 5% Al, >0.1% Y, 0.3% Si, 0.08% Mn, 0.03% Cu, 0.03% C, and the balance Fe). These were the first stack manifolds tested with bottom port inlets. Some unintentional differences were that the cells of Module #1 were mostly fabricated in 2007, and completed in early 2008, while module #2 was spring of 2008 and module #3 was summer of 2008, just ahead

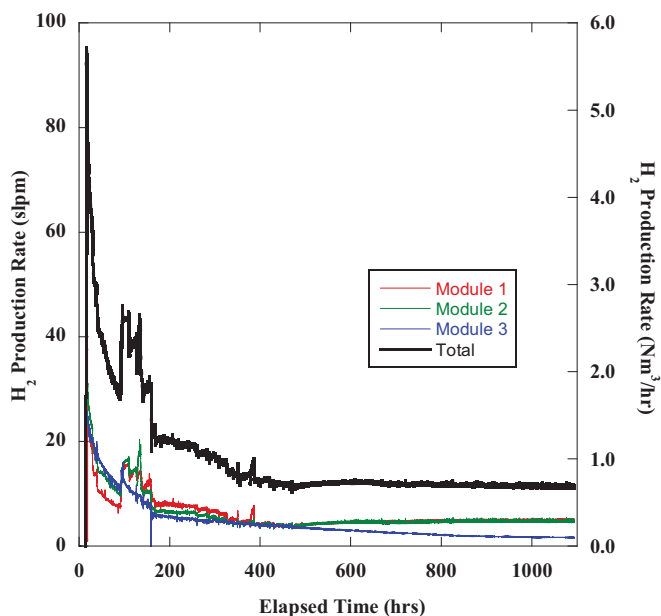


Figure 11. History of H₂ production rate.

shown in Fig. 6, and then interconnecting the two half modules when they are in final position (Fig. 7). When the two half-modules are placed back-to-back a common air inlet plenum for all four stacks is formed. Spring loaded bars are placed over the stacks to maintain a compressive load on the stacks during operation. Power leads to each stack, intermediate voltage taps and interior thermocouples were then attached, and subsequent sealing of gaps completed the installation (Fig. 8). All three of the ILS modules were incorporated into a single hot zone for the ILS, as displayed in Fig. 9.

ILS FACILITY TEST RESULTS

The ILS was operated for 1080 hours. Peak H₂ production as well as peak electrolysis power input was achieved at approximately 17 hours elapsed test time (Fig. 10). At this time, the electrolysis modules were consuming 18kW of electrical power and producing over 5.7 Nm³/hr H₂. Module ASRs were respectable and ranged from 1.25 Ωcm² to just over 2 Ωcm². A complete history of H₂ production rate for the test duration is displayed in Fig. 11. Total H₂ production rate decreased from 5.7 Nm³/hr to a steady state value of 0.7 Nm³/hr. The decrease was primarily due to cell degradation. Some cell degradation may be attributed to interruptions in inlet hydrogen mass flow rate due to condensate forming in the hydrogen recycle system and entering the hydrogen mass flow controllers. The increase in H₂ production rate at approximately 100 hours elapsed test time was due to increasing the hot zone interior temperature from 800 to 830 C.

The hydrogen product passes through a condenser before the hydrogen recycle system. Since the hydrogen recycle system operates at elevated pressure, additional water condenses out in the recycle system. When designing the system, INL engineers assumed that this additional water condensate would collect in the hydrogen recycle storage tank.

Table 2. Operating conditions for module voltage sweep.

Hot zone temperature	800 C
Inlet water mass flow rate	42 ml/min
Inlet H ₂ flow rate	10 NI/min
Inlet N ₂ flow rate	10 NI/min
Inlet Air flow rate	20 NI/min
Measured OCV	49.6 V
Measured inlet dew point	86.0 C
Outlet dew point at OCV	90.0 C
Current density (A/cm ²)	0 – 0.31

In spite of the tank, some droplets would still form and interfere with the inlet hydrogen mass flow controllers, causing flow disruptions. These interruptions could have caused the modules to undergo redox cycles – oxidation and reduction of the hydrogen-side electrode. For times earlier than 480 hours test time, operators would periodically remove the suspect mass flow controller and shake liquid water out. When reinstalled, the mass flow controller would operate properly again until the behavior would reappear. Operators also installed heat tracing on the tubing upstream of the controllers, but this did not improve performance. Finally, operators installed a liquid water trap and receiver vessel immediately upstream of the mass flow controllers. This modification solved the problem and only required periodic emptying of the receiver vessel. Performance of modules 1 and 2 improved, while the performance of module 3 continued to deteriorate.

The ILS modules performance was tested by sweeping the module power supply voltage over the range of 50 to 80 V (0.83 V/cell to 1.33 V/cell). This range corresponds to operation from the open-cell voltage to slightly above the thermal neutral voltage. Sweep results are shown in Fig. 12

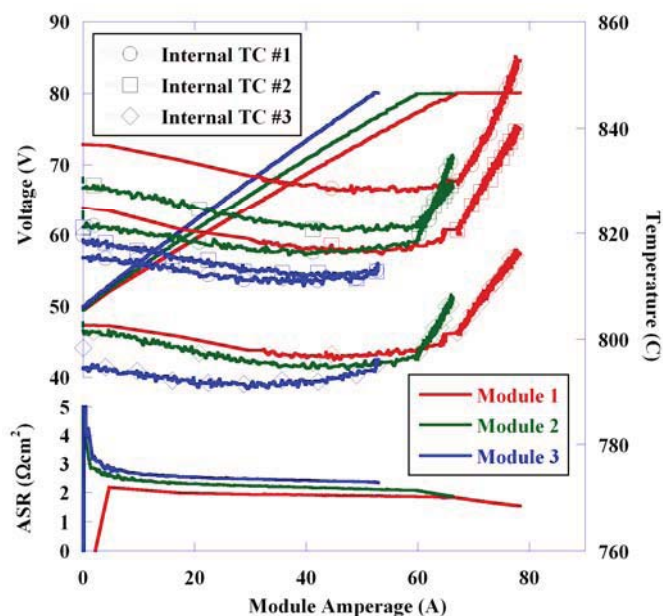


Figure 12. ILS 3 module voltage sweep / polarization curve data.

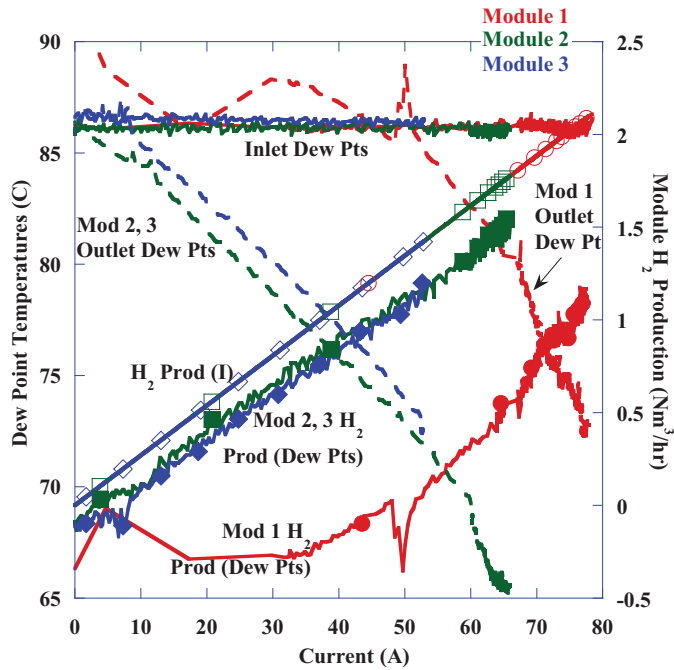


Figure 13. ILS module voltage sweep hydrogen production rates and dew points.

and the operating conditions for the ILS modules voltage sweeps are listed in Table 2. Module #1 was swept first and held at 80 V. Then, module #2 was swept and held at 80 V. Finally, module #3 was swept. Data corresponding to module #1 is red, data for module #2 is green, and data for module #3 is blue. The average ASR for the three ILS modules was measured to be just over $2 \Omega\text{cm}^2$, with module #1 exhibiting the lowest ASR and module #3 the highest ASR. This can also be seen in the slopes of the VI curves, with the curve for module #1 showing the lowest slope.

Stack internal temperatures, represented by lines with symbols in Fig. 12, initially decreased during the voltage sweep, due to the endothermic heat of reaction for water splitting. Once the operating voltage exceeded the thermal neutral voltage (77V for 60 cells), outlet gas temperatures quickly began to exceed inlet values. It is obvious that the voltage sweep rate was faster than the temperature response rate. It is usually most desirable to operate at the thermal neutral point where thermal stresses are minimized while the H_2 production rate is still reasonable. Operation at voltages in excess of the thermal neutral voltage might be desirable when recuperating heat from the products to preheat the feed stream.

Fig. 13 presents inlet and outlet dewpoint temperatures and hydrogen production rate (both based upon current as well as dew point difference) for each module for the sweeps described above. Solid lines without symbols represent inlet dew point temperatures. Dashed lines without symbols represent outlet dew point temperatures. Solid lines with open symbols show H_2 production rates based upon electrolysis current. Solid lines with filled symbols are H_2 production rates estimated from dew

Table 3. Long duration operating conditions for each ILS module.

Hot zone temperature	800-830 C
Inlet water mass flow rate	2.5-0.5 kg/hr
Inlet H_2 flow rate	10-6 NI/min
Inlet N_2 flow rate	10-5 NI/min
Inlet Air flow rate	20-30 NI/min
Measured inlet dew point	91 - 80 C
Module operating voltage	76 V
Current density	$0.3 - 0.03 \text{ A/cm}^2$

point temperature depression. Finally, the colors red, green, and blue symbolize modules 1, 2, and 3, respectively.

The inlet gas dew points remained essentially constant at 86°C throughout the duration of the sweep. In general, the outlet stream dew point temperatures decreased continuously through the sweep as the operating voltage and stack current increased. An exception to this is the outlet dew point temperature trace for module #1, which showed a complicated behavior probably due to condensation in the outlet dew point sensor fixture.

Agreement between the two means of estimating the H_2 production rate was reasonably good for modules 2 and 3. However, the erratic behavior of the outlet dew point sensor for module 1 affected the module 1 dew point-based H_2 production rate estimates. At the highest current levels, the total H_2 production rate for all 3 modules exceeded $5.3 \text{ Nm}^3/\text{hr}$ (88 slpm) during the sweep.

After establishing the ILS peak hydrogen production capacity and characterizing the performance of the modules, the ILS operating parameters were set to the long duration conditions listed in Table 3. ILS operations extended for 1080 hours. Inlet water mass flow rates, H_2 flow rates, N_2 flow rates, and air flow rates were adjusted at times during the test to maintain proper internal stack temperatures. The test had very few instrumentation problems. No high bias voltage problems were noted during this test. Furthermore, no internal stack thermocouples were lost (in contrast to the earlier single module test). Only two instrumentation problems arose. First, one nitrogen mass flow controller (Bronkhorst) failed (lost communications). A rotameter was installed to continue the test without interruption. The second problem involved condensation interfering with the inlet hydrogen mass flow controllers and was discussed earlier.

The effectiveness of the heat recuperation system was obvious from observing various inlet and outlet gas stream temperatures. 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.

SUMMARY

Full operation of the INL 3 module ILS system began in September 2008 and continued for 1080 hours. A peak H_2 production of $5.7 \text{ Nm}^3/\text{hr}$ H_2 was achieved, corresponding to a peak electrolysis power input of 18kW. Early test module ASRs ranged from $1.25 \text{ } \Omega\text{cm}^2$ to just over $2 \text{ } \Omega\text{cm}^2$ per cell. Significant module performance degradation 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. Some of this accelerated degradation may have been caused by recycle condensation interfering with the inlet H_2 mass flow controller. The total H_2 production rate decreased from $5.7 \text{ Nm}^3/\text{hr}$ to a steady state value of $0.7 \text{ Nm}^3/\text{hr}$, primarily due to cell degradation.

Evaluation of the heat recuperation performance indicated that the total per-module electric heater power load was reduced by 50% in the case of heat recuperation. The hydrogen recycle system was very effective 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 worked flawlessly 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. Future research will concentrate on reducing cell performance degradation during long term operation and further scale-up activities related to large-scale H_2 production.

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