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DESIGN OF AN INTEGRATED LABORATORY SCALE TEST FOR HYDROGEN PRODUCTION VIA HIGH TEMPERATURE ELECTROLYSIS

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The Idaho National Laboratory (INL) is researching the feasibility of high-temperature steam electrolysis for high-efficiency carbon-free hydrogen production using nuclear energy. Typical temperatures for high-temperature electrolysis (HTE) are between 800º-900ºC, consistent with anticipated coolant outlet temperatures of advanced high-temperature nuclear reactors. An Integrated Laboratory Scale (ILS) test is underway to study issues such as thermal management, multiple-stack electrical configuration, pre-heating of process gases, and heat recuperation that will be crucial in any large-scale implementation of HTE.

The current ILS design includes three electrolysis modules in a single hot zone. Of special design significance is preheating of the inlet streams by superheaters to 830°C before entering the hot zone.
The ILS system is assembled on a 10' x 16' skid that includes electronics, power supplies, air compressor, pumps, superheaters, , hot zone, condensers, and dew-point sensor vessels. The ILS support system consists of three independent, parallel supplies of electrical power, sweep gas streams, and feedstock gas mixtures of hydrogen and steam to the electrolysis modules. Each electrolysis module has its own support and instrumentation system, allowing for independent testing under different operating conditions. The hot zone is an insulated enclosure utilizing electrical heating panels to maintain operating conditions.

The target hydrogen production rate for the ILS is 5000 Nl/hr.

I. INTRODUCTION

Under the Nuclear Hydrogen Initiative (NHI), the United States Department of Energy (DOE) is currently researching the feasibility of two methods of hydrogen production based on nuclear energy: (1) thermochemical processes, primarily the Sulfur-Iodine (SI) process, and (2) high-temperature electrolysis (HTE). HTE typically operates at temperatures ranging from 800 to 900ºC consistent with the planned coolant outlet temperature range of advanced high-temperature nuclear reactors.

The long-term plan for the NHI includes demonstration of these technologies at increasingly larger scales to address overall system issues such as thermal management and heat recuperation. Since these two nuclear hydrogen research tracks are on parallel paths in the NHI program, both tracks are expected to advance to the next scale of research and deploy an Integrated Laboratory Scale (ILS) system in FY07. Target hydrogen production rates for the two ILS systems are 5000 Nl/hr for the HTE ILS and 200 Nl/hr for the SI ILS.

This paper describes the design of the HTE version of the ILS under development at the INL. In accordance with the Research and Development Plan for the NHI, the HTE research track is progressing from small-scale bench testing to large-scale demonstration. For HTE, the experimental research objectives have been organized as follows with increasing scale:

- Button cell fabrication/testing (~1 W) -- cell material development and performance
- Stack development/testing (200 W - 5 kW) -- stack design (electrode and electrolyte materials, interconnect and flow field materials and fabrication, inter-cell electrical contact, cell and manifold sealing issues, cell durability)
- Integral issues (15 kW) -- feed-stock heating, high-temperature gas handling, multiple-stack thermal management, heat recuperation
- Facility (200/500 kW and 5 MW) --production issues including energy management, utility requirements, high pressure operation, product purification.

HTE testing of button cells and stacks of up to 25 cells has been in progress at the INL and the subcontractor Ceramatec Inc. (Salt Lake City, UT) for more than 3 years. These tests primarily concentrated upon quantifying material and cell performance and have not addressed larger-scale issues. For example, in button cell and bench-scale stack testing, steam is introduced into the inlet gas stream using a humidifier approach in which the gas is bubbled through a heated water bath. 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

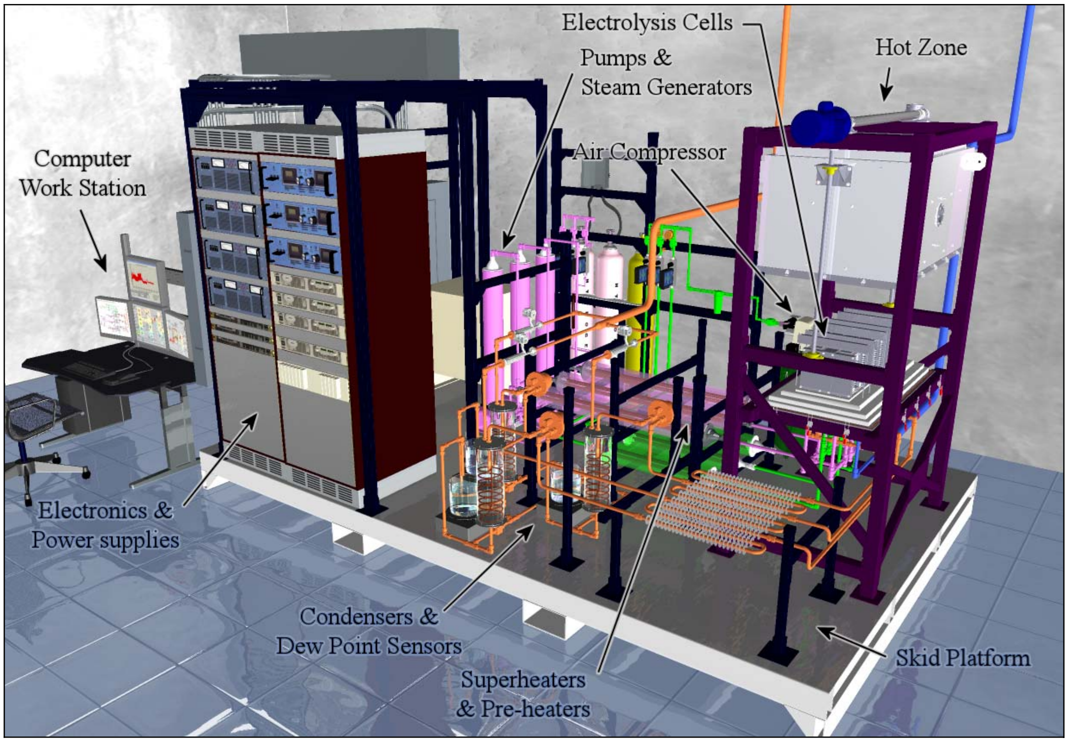

Figure 1. HTE ILS system overview.

steam production and feed-stock heating is not realistic for larger scales of electrolysis. The HTE ILS system has been envisioned to address these issues.

II. ILS OVERVIEW

The HTE ILS facility is designed to study large-scale hydrogen-production issues such as thermal management, hot-zone design, multiple-stack electrical configuration, pre-heating of process gases, and heat recuperation. In the ILS, the inlet steam/hydrogen flow will be preheated to the full 830°C electrolyzer operating temperature prior to entering the hot zone using superheaters. This scenario more closely simulates the inlet stream heating performed in a full-scale hydrogen production plant coupled to an advanced nuclear reactor. A full-scale plant would utilize both electrical power and high-temperature process heat from the reactor system.

Also of design note is the configuration of the electrolysis modules in the hot zone. Each module has three separate, parallel support systems that supply power, superheated steam, preheated air, and appropriate exhaust handling. By utilizing three separate gas streams, failure of an individual module will not affect the performance or testing of the other modules. Thus the ILS system design allows each module to be tested independently.

The HTE ILS system will be assembled on a large skid (10' x 16'). Primary skid-mounted components include electronics, power supplies, air compressor, pumps, superheaters, hot zone, condensers, and dew-point sensor vessels. Next to the skid will be a computer work station to electronically control the equipment. Fig. 1 shows the ILS system configuration with important components labeled. The piping and instrumentation diagram (P&ID) for the HTE ILS system, without heat recuperation or hydrogen recycle, is shown in Fig. 2. In both Fig. 1 and Fig. 2, the hydrogen/steam feedstock is represented by the color magenta, the product stream by orange, and the inlet sweep gas by green, and the outlet sweep gas by blue.

The ILS system is currently planned for an initial nominal hydrogen production rate of 14.1 kW based on the lower heating value (LHV) of 120 MJ/kg. This production rate corresponds to 4735 Normal (273°K, 1 atm) L/hr of hydrogen. Table 1 lists the nominal and extreme design cases for which the ILS system has been designed. The nominal design case assumes an average electrolysis cell ASR of 1.5 ohm cm^2 and operation at the thermal neutral voltage of 1.283 volts per cell. These operating conditions result in an electrolyzer required electrical power input of 14.54 kW and a hydrogen production rate of 4735 normal liters/hr. The extreme design case assumes an improvement in the per-cell ASR to 1.0 ohm cm^2. Operation at the thermal neutral voltage at this ASR results in a current density of 0.37 amps/cm^2, an electrolyzer electrical power requirement of 21.8 kW, and a hydrogen production rate of 7103 NL/hr. All ILS components were sized to accommodate this extreme design case.

In the initial HTE ILS deployment, heat recuperation will not be used. The system is designed to allow later

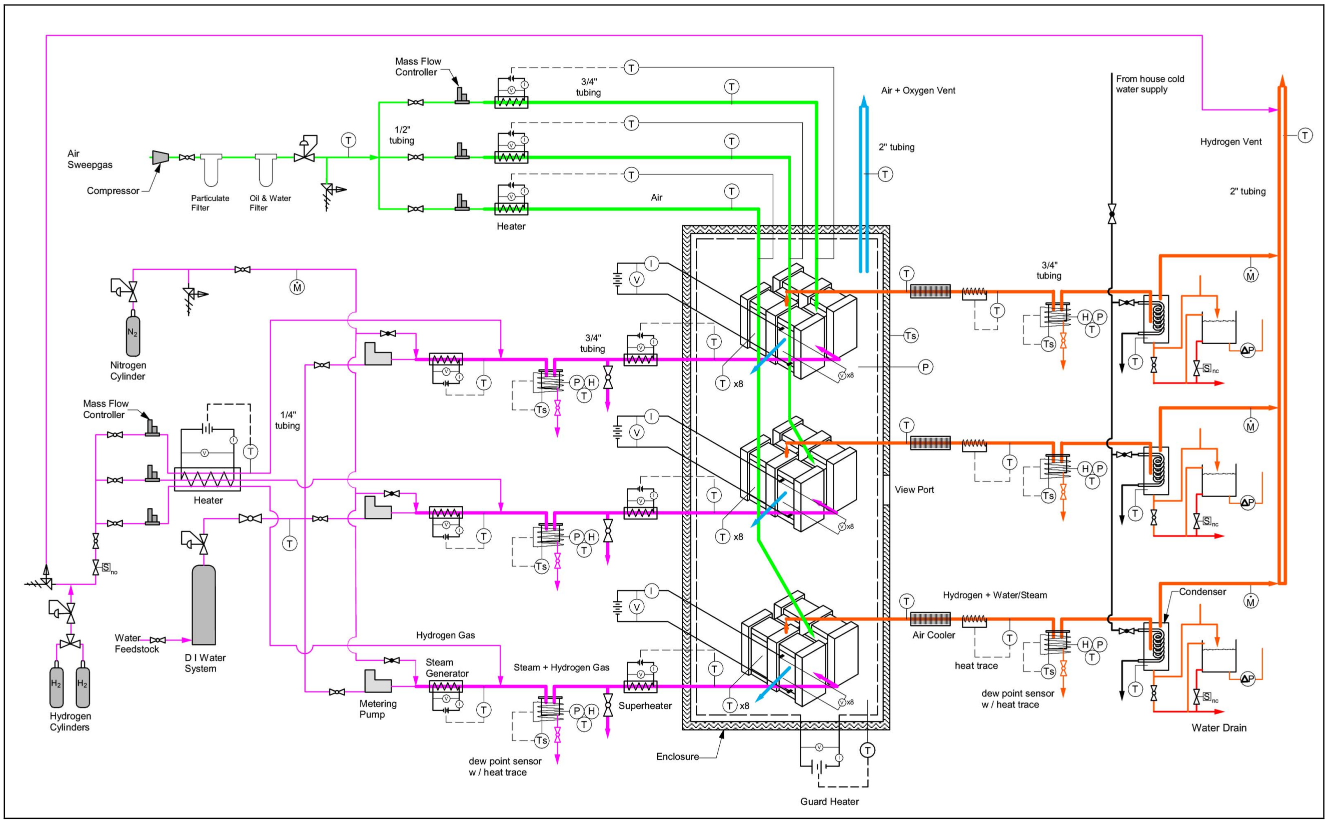

Figure 2.HTE ILS P&ID.

incorporation of recuperative heat exchangers. The use of recuperation minimizes the net heat addition required to heat the process gas streams up to the HTE operating temperature, improving the overall efficiency of the system

III. INLET DESIGN

To begin the process, deionized liquid water feedstock is fed at a controlled rate into the system by means of a positive-displacement metering pump. The water is then vaporized and slightly superheated in an inline electrically-powered steam generator, the design of which is discussed in greater detail below. Electrical power for the steam generator is supplied by a computer-controlled DC power supply that, based upon outlet steam temperature, is feedback-controlled through a computer data acquisition and control system (DACS). The steam mass flow rate is verified by monitoring the rate of electrical energy supplied to the steam generator from the DC power supply.

Table 1. Comparison of nominal and extreme design cases.

	Nominal Case	Extreme Design Case
ASR (ohm cm^2)	1.5	1.0
Current Density (A/cm^2)	0.25	0.37
Per-cell Voltage, V()	1.283	1.283
Electrolysis Power (kW)	14.54	21.8
Hydrogen Production Rate (NL/hr)	4735	7103

The slightly superheated steam exiting the steam generator is mixed with hydrogen, which is required on the inlet side of the electrolysis stack in order to maintain reducing conditions at the steam/hydrogen electrode (Nickel cermet). In the initial ILS configuration, the inlet hydrogen will be supplied from compressed gas bottles. The hydrogen flow rate is controlled by a mass-flow controller and the DACS. The inlet hydrogen must be preheated to the steam generator outlet temperature in order to prevent cooling of the steam and possible steam condensation. This is accomplished by temperature-based feedback control of the hydrogen supply line heat tracing and a DC power supply in conjunction with the DACS. Downstream of the mixing point, the temperature, pressure, and dew-point of the steam/hydrogen gas mixture is measured. Precise measurement of the dew-point and pressure allows for independent determination of the inlet gas composition.

Air will be used as a sweep gas for the ILS system. Compressed air flows through a mass-flow controller and

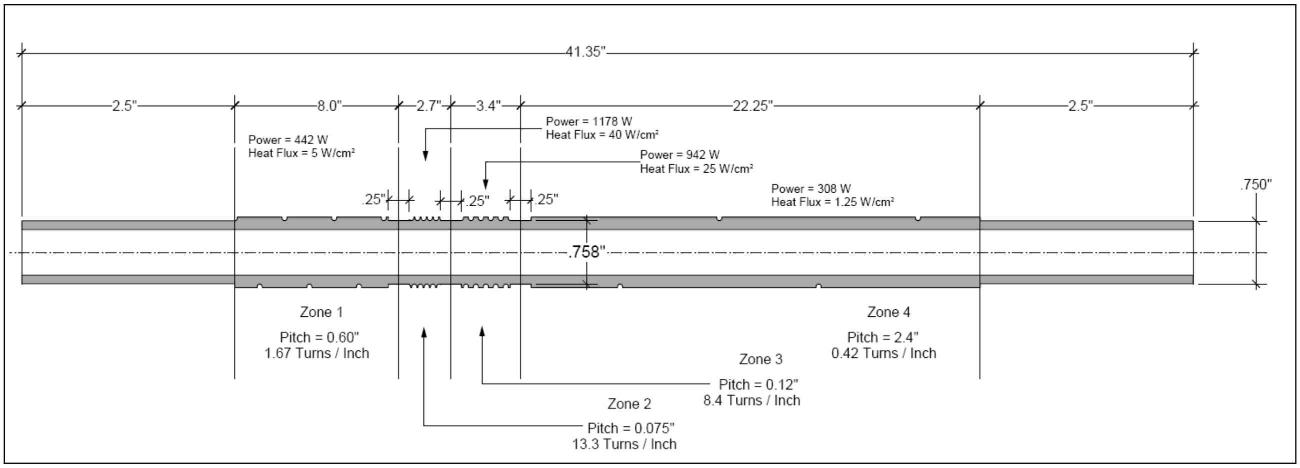

Figure 3. Steam generator details.

into an electrically-powered heater to preheat the inlet air to the stack operating temperature.

IV. ILS HEATING COMPONENT DESIGN

A search of steam generator/superheat equipment failed to find off-the-shelf equipment that would provide a steam outlet temperatures of 830°C, so custom designs for both the steam generator and superheater system were engineered at the INL to meet the specific requirements of the ILS.

IV.A. Steam Generator

A metering pump will supply the required liquid water flow rate to the inlet of the vertical steam-generator section. The steam generator is fabricated from a ½" Schedule-80 stainless steel pipe. Several heat transfer regimes exist over the length of the steam generator as shown in Fig. 3, which necessitated several different heat transfer rates to the fluid. These heat transfer rates were optimized for the ILS extreme case operating conditions to minimize the length of each segment and also to provide a safe operating temperature of the pipe wall.

Heat will be supplied to the fluid using 0.0226" diameter Kanthal A1 resistance wire laid in grooves machined in the outside of the pipe wall. This wire has a maximum continuous operating temperature rating of 1400°C. A thin layer of a ceramic insulator will be thermally sprayed onto the grooves prior to winding the wire to prevent electrical contact between the pipe and the resistance wire. Fig. 4 shows a photo of the steam generator pipe with the application of the insulator and resistance wire.

The power to the resistance wire will be supplied by a dedicated DC power supply controlled by the ILS DACS with a proportional-integral-derviative (PID) feedback control loop using the steam outlet temperature as the process variable. The outlet temperature will be controlled to approximately 50°C above the saturation temperature to prevent any condensation in the piping or at the dew-point sensor as a result of heat losses or the addition of the hydrogen gas which is injected downstream of the steam generator.

For the extreme design case the steam generator will require 2.82 kW of power. The design operating voltage is 250 volts, requiring a current of 11.3 amps for this power level. The selected power supply can operate from 0-300 volts and 0-16 amps.

The steam generator pipe will be enclosed in approximately 1.5" thick alumina-based insulation to minimize heat losses. The steam generator will be oriented vertically with flow from bottom to top.

IV.B. Superheater

After the steam/hydrogen gas mixture exits the dew-point measurement station, a high-temperature electrically powered inline superheater will boost the feedstock stream to the final electrolyzer operating temperature of

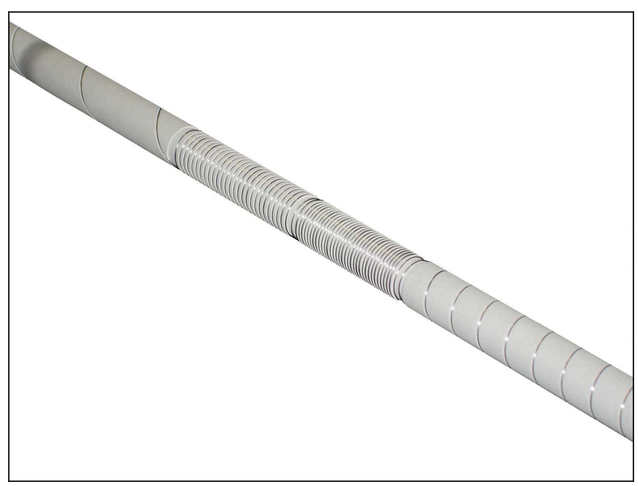

Figure 4. Photograph of steam generator with wound heater wire.

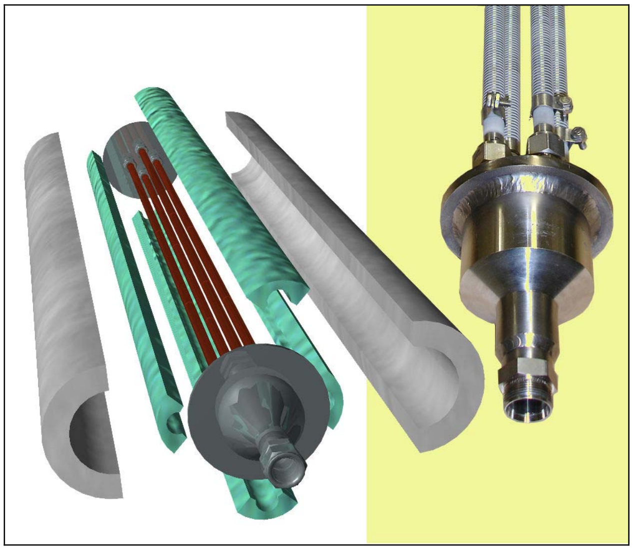

Figure 5. Superheater cut-away view.

800° - 830°C. The primary material of construction for the low-temperature tubing and components up to the superheater is 316 stainless steel.

The steam superheater design is very similar to that of the steam generator. However, because of the relatively low heat transfer coefficients to the single-phase steam and hydrogen gas mixture, four parallel ¼" schedule 80 pipes will be used instead of the single pipe used in the steam generator in order to increase the heat transfer surface area (see Fig. 5). Using four ¼" pipes instead of a single ½" pipe increases the flow cross-sectional area by about twenty percent. This does reduce the flow velocity by the same amount, but the inside heat transfer surface area of the pipe is increased by 120% allowing a significant reduction in the required length of the superheater. The four pipes will be surrounded with insulation as shown in the left of Fig. 5.

Each individual steam superheater pipe will have three heat zones with local wall heat flux determined by the local pitch of the resistance wire. This insures that the tube wall temperature will not exceed the design limit as the steam heats up along the pipe length. The orientation (horizontal or vertical) of the superheater pipe is arbitrary as there is no phase change along the length of the pipe. The resistance wire will be 0.032" diameter Kanthal A1 wire.

A single DC power supply will provide power to all four superheater pipes in parallel. As in the steam generator, the superheater power supply will be controlled by the ILS DACS with a PID feedback control loop using the steam temperature just before it enters the electrolysis stack as the process variable. By using this temperature for control, any heat losses in the piping between the superheater outlet and the electrolysis stack can be overcome by increasing the power to the superheater. For the extreme design case, the steam superheater will require 1.77 kW of power. The design operating voltage is 120 volts, requiring a current of 14.75 amps. The selected power supply can operate from 0-150 volts and 0-22 amps.

The superheater insulation consists of 1" thick zirconia-based insulation next to the pipes covered with an additional 1.5" of alumina-based insulation.

IV.C. Air Heater

Similar to the superheater, the air heater design separates the air flow into four parallel tubes. The only difference is the shorter length and number of heat zones for the individual pipes and the pitch of the resistance wire coil. In the extreme design case, up to 607 W of power could be required for the air heater. The design operating voltage is 75 volts requiring a current of 8.1 amps. The selected power supply can operate from 0-100 volts and 0-15 amps. Insulation for the air heater will consist of 1" thick zirconia-based insulation next to the pipes covered with an additional 1.5" of alumina-based insulation.

IV.D. Hot Zone

Downstream of the superheaters, the steam/hydrogen gas mixture will reach the three electrolysis modules incorporated into a single hot zone, as displayed in Fig. 6.

The ILS hot zone will include guard heaters designed

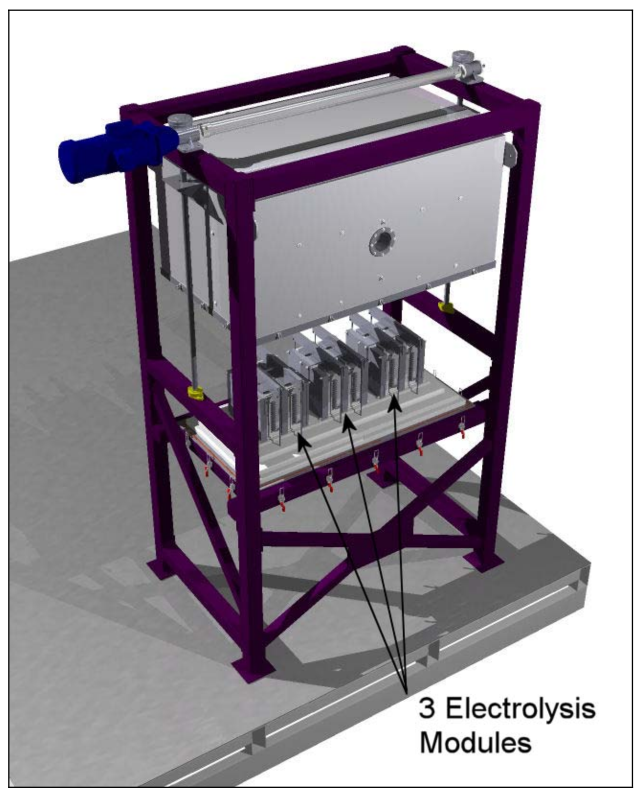

Figure 6. Hot zone.

to maintain the electrolysis stacks at the operating temperature during testing. In order to provide sizing information for the hot zone guard heaters, an estimate of the heat loss from the ILS hot zone enclosure was obtained using a one-dimensional steady-state model. This estimate accounts for conduction, convection, and radiation heat transfer. Hot zone outer dimensions, physical properties, and other important assumptions used in the calculation are listed in Table 2.

Based on these values, the estimated hot-zone heat loss at steady-state conditions is 3.55 kW. An additional heat load will occur when electrolysis is performed at an operating voltage lower than the thermal neutral voltage. For example, for the design case (ASR = 1.0 Ohm·cm^2, 50% steam utilization) operating at the thermal minimum voltage (~1.05 V), an additional steady-state heat load of 1.47 kW will be required under isothermal operating conditions to balance the endothermic heat requirement associated with the electrolysis process. Thus the estimated hot zone steady-state heater requirement at this operating condition would be 5 kW. To allow for a reasonable initial heatup time, and to provide some design margin, a hot-zone heater power requirement of 15 kW was specified.

V. ILS ELECTROLYSIS MODULE DESIGN

The ILS design includes three four-stack modules, with 60 electrolysis cells in each stack, for a total of 12 stacks and 720 cells in a single hot zone.

Table 2. HTE ILS nominal performance characteristics.

Independent Design and Operational Parameters	
active cell area	64 cm^2
cells per stack	60
number of stacks	12
stack operating temperature	830 °C
steam utilization	50%
stack operating voltage	77 V
per-cell ASR	1.5 Ohm cm^2
inlet steam mole fraction	0.9
inlet hydrogen mole fraction	0.1
Calculated Performance Values	
per-cell operating voltage	1.283 V
current density	0.25 A/cm^2
stack power	1232 W
total power (electric)	14.54 kW
inlet hydrogen flow rate	17.5 NLPM
inlet steam flow rate	158 NLPM
inlet liquid water flow rate	2.1 g/s
air flow rate	67.9 NLPM
hydrogen production rate	4735 NL/hr
heating value of produced H_2	14.1 kW (LHV)

This configuration utilizes solid oxide cell technology similar to the stacks previously tested at INL. The electrolysis module is configured so the steam/hydrogen gas mixture enters through the bottom of the steam inlet manifold (shown on the module's ends in Fig. 7) and exits through the bottom of the outlet manifold in the module's center. Airflow enters through the bottom of the air inlet manifold, flows through the stack in cross-flow with the steam/hydrogen gas mixture, and exits at the front directly into the hot zone. Dimensions of the ILS stacks, manifolds, and hot zone based on this preliminary design are provided in Table 3.

The three electrolysis modules will require separate and parallel support systems 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 include means for controlled steam generation, mixing hydrogen with the steam, inlet and product dew-point 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. Furthermore, each electrolysis module has its own independent instrumentation system. Because the feedstock and product gas streams for each electrolysis module are to be monitored and controlled separately; the inlet and outlet gas composition and flow rate will be known specifically for each module. This allows for independent testing of each of the three HTE ILS modules under different operating conditions (gas mixture, current density, etc.). In this way, a test matrix covering a range of operating conditions can be covered quickly. Additionally, if a module fails, that particular module can be shut down without affecting the performance or testing of the other modules.

Each module has multiple power lead attachment tabs (visible in Fig. 7) in addition to intermediate interconnect tabs. These intermediate tabs allow for electrical interconnection of adjacent stacks, thereby minimizing

Table 3. ILS stack and hot zone dimensional details.

Cells per stack	60
Stacks per module	4
Number of modules	3
Total number of stacks	12
Stack height	15 cm
Stack volume	1500 cm^3
1 Module volume	4260 cm^3
3 Module volume	0.051 m^3
Interior hot zone length	1.73 m
Interior hot zone height	0.75 m
Interior hot zone width	0.95 m
Hot zone volume	1.230 m^3

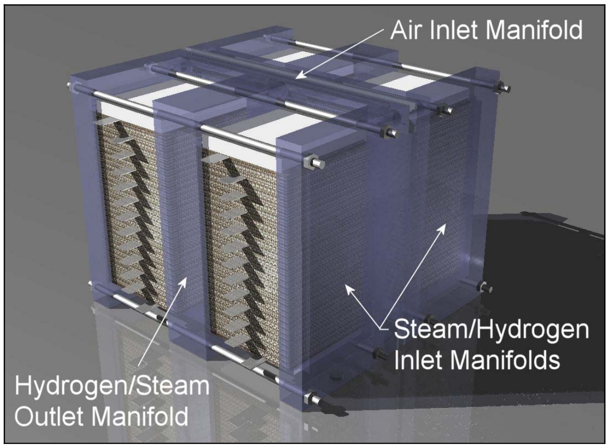

Figure 7. Electrolysis module with four stack of 60 cells.

the impact of a possible single-cell failure.

To measure fuel cell integrity, the area specific resistance (ASR) value is closely monitored. Testing of fuel cells shows that the integrity of the cell gradually decreases over time, hampering the cells' ability to produce hydrogen as efficiently. This degradation is evidenced by an increase of the calculated ASR values. As technology of cell fabrication improves, the cell's ability to efficiently produce hydrogen over long time periods should improve.

VI. EXHAUST GAS HANDLING DESIGN

There are two main exit streams from the fuel cell: a hydrogen/steam product stream and an air/oxygen sweep stream.

For each of the three electrolysis modules, there is a hydrogen/steam product stream emerging from the hot zone. These gas mixtures 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 finned tube, but not to the point that condensation can occur. Then the product gas mixture enters the outlet dew-point measurement station to directly determine the steam consumption rate and the corresponding hydrogen production rate. This rate can be compared to the electrochemical hydrogen production rate determined from the stack electrical current. The steam in each of the hydrogen/steam streams is removed by a condenser, creating three condensate water streams and three exhaust hydrogen streams. The condensate water flow rates from each stream are measured, and the water is then piped to a floor drain. The hydrogen stream flow rates are independently measured; then the three streams are combined into a single hydrogen exhaust stream. When hydrogen recycle is implemented into the HTE ILS, a line will be tapped at this point to provide hydrogen to be mixed with the inlet steam feedstock for the electrolysis process.

The air/oxygen sweep stream flows from the fuel cell stacks into the hot-zone enclosure where it is vented to the outside as a single stream.

VII. ELECTRICAL REQUIREMENTS

The electrical requirements for the ILS components run from less than 100 watts for the feedwater metering pumps to nearly 15 kW for the hot zone heaters. To meet these varied requirements a 480-volt 3-phase, a 208-volt 3-phase and single-phase, and a 120-volt single-phase circuits will be utilized. A single 200-amp 480-volt 3-phase tube and sleeve plug will supply all the power for the ILS skid. The four main power supplies, 10 kW or greater, will receive power from a 480-volt 3-phase power panel. A 75 kW transformer will convert the balance of the power to 208 Y/120 V to be distributed to the other components as required. The maximum power consumed by the ILS will approach 100 kW.

VIII. CONCLUSIONS

This report has presented the performance, space, and power requirements for the HTE ILS experiment to be deployed at the INL. The ILS will demonstrate larger scale hydrogen production while simultaneously addressing integral issues such as multiple-stack gas manifolding, stack electrical integration, and inlet / outlet gas handling and thermal management. The INL team has defined nominal and extreme design-basis operating conditions and a P&ID for a 15 kW HTE ILS system. This design includes specifications for the electrolysis stack modules as well as instrumentation and control requirements. Detailed process flow sheets have been developed for this design using the commercial system-analysis code HYSYS. These flow sheets include all of the components that would be present in the actual HTE ILS system such as pumps, compressors, heat exchangers, and the electrolyzer itself.

In this design, liquid water feedstock will be heated and electrolyzed using solid-oxide electrolysis cells at approximately 830°C to produce approximately 5000 NL/hr of hydrogen, corresponding to a hydrogen low-heating-value production rate of approximately 14.1 kW. To accomplish this, the initial HTE ILS configuration will have 12 planar HTE stacks of 60 cells per stack, with a per-cell active area of 64 cm^2. These stacks will be arranged in 3 modules of 4 stacks each and will be electrically interconnected to minimize the impact of any possible single-cell failure. All three modules will be enclosed within an insulated hot zone. Alternate cell and stack designs will be tested during the ILS extended operation period. The ILS will operate at approximately atmospheric pressure.

During extended operation, the ILS system will allow researchers to study options in heat recuperation to minimize the net heat addition required to heat the process gas streams up to the HTE operating temperature. Hydrogen recycle will also be implemented during ILS extended operation.

ACKNOWLEDGMENTS

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