Laboratory-Scale High Temperature Electrolysis System

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PD17

Idaho National Laboratory

High Temperature Electrolysis Overview

Technical Objectives

- Develop and demonstrate energy-efficient, high temperature solid oxide electrolysis cells (SOECs) and stacks for hydrogen production from steam.
- Demonstrate technology at progressively larger scales
- Perform flowsheet analyses of systems-level HTE processes to support planned scale-up to Integrated Laboratory-scale, Pilot-scale and Engineering Demonstration-scale experiments.
- Develop detailed CFD models of operating SOECs; validate with experiment data
- Investigate alternate cell materials (e.g. alternate electrode and/or interconnect materials) alternate cell configurations (e.g. porous-metal substrates, tubular cells, porous electrodes) and applications of inorganic membranes

Strategy

- Wherever possible, build on previous development of solid oxide fuel cells by DOE-EE, SECA and others.

High Temperature Electrolysis Overview

Timeline:

Project start date: Jan 2003, Button cell tests,
Project end date: Engineering Demo, 1 MW, 2015

Budget

Total project funding (all DOE-NE)

FY05: $1440 k

FY06: $4695 k

Barriers (next slide)

Partners

Ceramatec, Inc.
Argonne National Laboratory
Oak Ridge National Laboratory
University of Nevada, Las Vegas

Technical Barriers

Adapted from

3.1.4.2.2 Hydrogen Generation by {Water} Electrolysis [A-K]

G. Capital Cost - *R&D is needed to develop lower cost materials with improved manufacturing capability to lower capital costs while improving the efficiency and durability of the system. Development of larger systems is also needed to improve economies of scale.*

H. System Efficiency *– Development is needed for low-cost cell stack optimization considering efficiency, electrochemical compression and durability.*

I. Grid Electricity Emissions – *Low-cost, carbon-free electricity sources are needed.*

K. Electricity Costs – *High Temperature solid oxide electrolysis can use lower cost energy in the form of steam for water splitting to decrease electricity consumption. Technically viable systems for low-cost manufacturing need to be developed for this technology.*

3.1.4.2.3 Separations and Other Cross-Cutting Hydrogen Production [L-U]

L. Durability – *need to reduce amortized capital costs and allow more thermal cycles in lifetime*

N. Defects – *particularly in the oxygen handling service*

P. Operating Temperature – *take-off among reaction thermodynamics, materials limitations and use of product hydrogen and oxygen*

T. Oxygen Separation (and Handling) Technology – *Especially cooling of enriched oxygen/air mixtures from 850° C and transport to beneficial uses*

Approach (vs Objectives)

- Develop energy-efficient, high-temperature, solid-oxide electrolysis cells (SOECs) for hydrogen production from steam.
  - Optimize energy efficiency, cost and durability
    - optimize electrolyte materials (e.g., YSZ, ScSZ, sealants)
    - investigate alternate cell configurations (e.g., electrode-supported or tubular)
- Develop and test integrated SOEC stacks operating in the electrolysis mode with an aim toward scale-up to a 200 kW Pilot Plant and a 1 MW Engineering Demonstration Facility
  - Increase SOEC stack durability and sealing with regard to thermal cycles
  - Improve material durability in a hydrogen/oxygen/steam environment
  - Perform a progression of electrolysis stack testing activities at increasing scales and complexities
  - Develop computational fluid dynamics (CFD) capability for SOEC
  - Utilize advanced systems modeling codes (e.g. HYSYS, ASPEN)
  - Perform Cost and Safety Analyses

HTE FY-06 Task Area Overview

Workpackage	P.I.	FY-06 ($k)	Description/Goal
HTE Systems Definition			
ID16EL11	O'Brien	1445	Engineering analyses needed to define future high temperature electrolysis plants. Design of Integrated Laboratory Scale Experiment.
CH16EL11	Petri	750	Flowsheet and CFD Analyses. Materials development of higher conductivity electrodes.
HTE Experiments			
ID16EL21	Stoots	2250	HTE stack experiments. Equipment for ILS. Equipment and operation of materials testing loop.
OR16EL21	Bischoff	100	Analysis and experiments to determine the applicability of inorganic high-temperature membranes for steam/hydrogen separations.
CH16EL21	Petri	75	Modeling and x-ray and electrochemical characterization of patterned dense thin-film oxygen electrodes on electrolyte substrates produced by conventional methods.
ID06EL21	Hechanova	75	Investigate structure-property-performance relationships for oxygen and hydrogen electrodes and electrolytes, as well as the fabrication of thin-film electrolytes using atomic layer deposition (ALD) techniques.

High Temperature Electrolysis Overview

Key Milestones (FY-06)

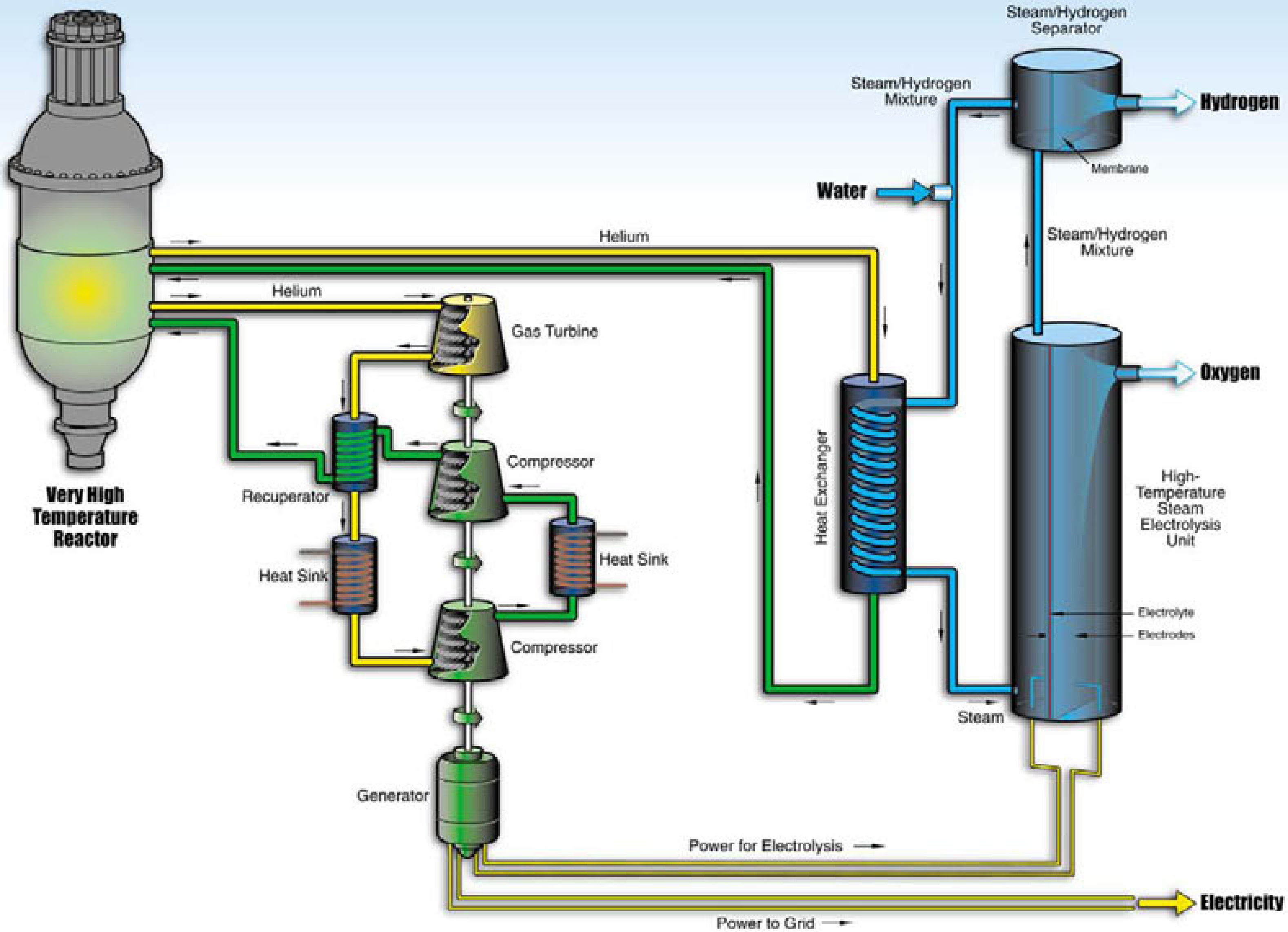
- Finalize Integrated Laboratory Scale experiment performance, space and power requirements. [INL 2/1/06] {completed 2/1/06}
- Complete document entitled "Integrated Laboratory-scale Stack Specification and Mechanical Design.“ [INL 9/1/06]
- Complete Final Report on use of HTE for Upgrading Athabasca Oilsands. [INL 9/15/06]
- Complete review article on Sealing Technologies Applicable to Solid Oxide Electrolysis Cells. [INL 11/30/05] {completed 11/30/05}
- Operate 20-25 cell stack at 100 Normal liters per hour for 1000 hours {elevated to Level 1 milestone} [INL 3/31/06] {completed 2/16/06}
- Begin testing of initial dual stack ILS module at Ceramatec Facility (2 x 60 cells) [INL 7/14/06]
- Initial operation of materials testing loop [INL 7/1/06]

High Temperature Electrolysis Overview

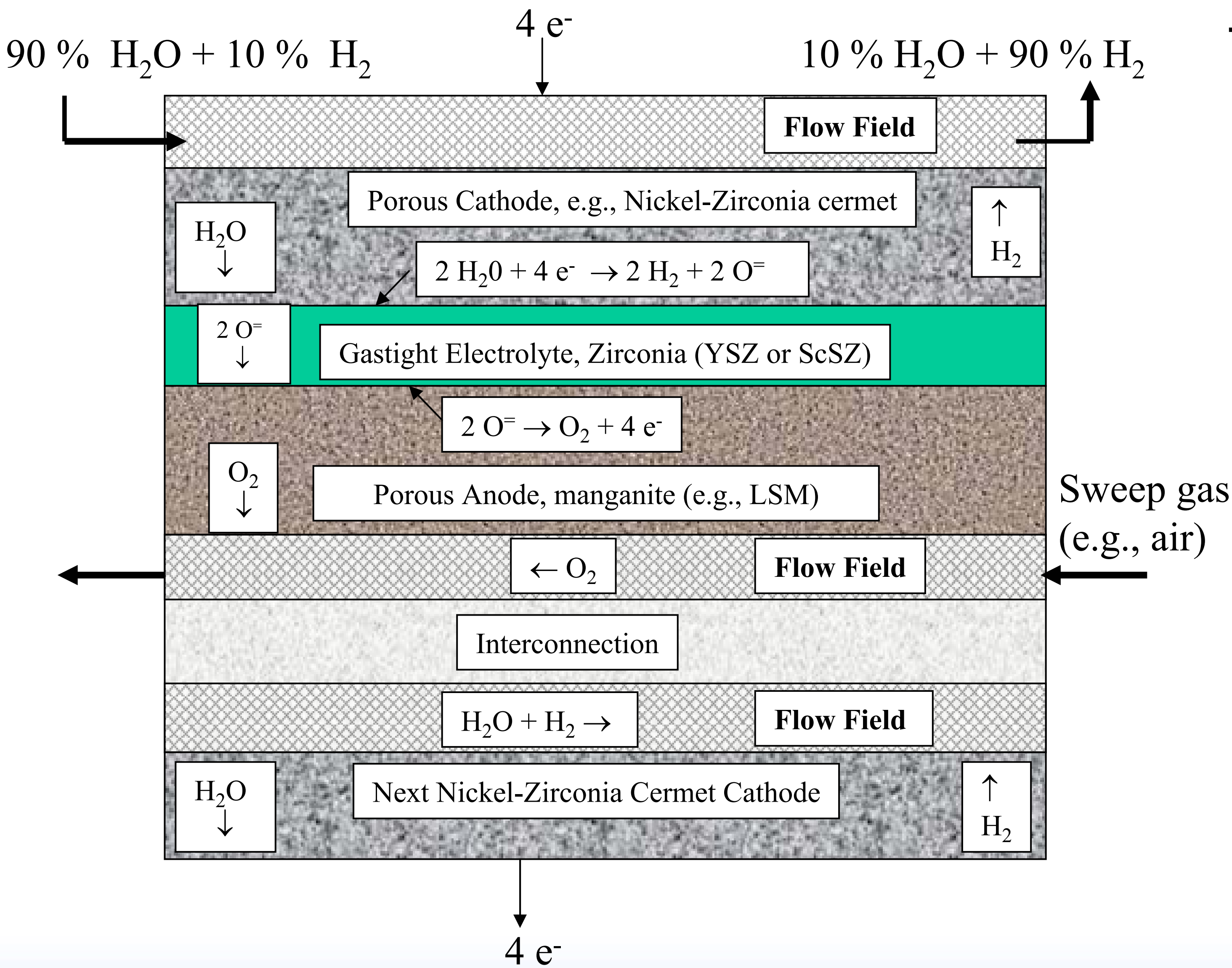
Key Milestones (FY-06)

- The ANL contribution to the joint INL/ANL report will include plant flowsheet analyses to thermally optimize the HTE-nuclear plant combination, particularly in the areas of heat recovery from the hydrogen and oxygen product streams. [ANL 9/15/06]
- The ANL contribution to the joint INL/ANL report will include Computational Fluid Dynamics analyses of HTE cells in support of the joint ANL/INL testing of these cells. These analyses and the ensuing report will summarize the investigation of the applicability of this configuration. [ANL 6/30/06]
- Complete testing of high temperature inorganic membranes for the separation of hydrogen and steam, based on flowsheet conditions and capabilities identified during FY-05. Specifically, complete a long-term test (approximately 1000 hours) at temperature suitable for high temperature electrolysis plant operation. [ORNL 9/15/06]

High Temperature Electrolysis Plant



Planar Solid-Oxide Electrolysis Stack



Typical Layer thicknesses

Electrolyte-supported	Cathode-supported
30 µm	1.500 mm
100 µm	10 µm
30 µm	50 µm
1 – 2.5 mm	

Oxygen electrode materials

- March '06 status:
  - Our best performing sample was PSC powder/PSC gel/CSO gel/YSZ electrolyte
    - *PSC = $Pr_{0.5}Sr_{0.5}CoO_{3\pm\delta}$, CSO=$Ce_{0.8}Sm_{0.2}O_{2\pm\delta}$*
- Current status:
  - We have reproduced the performance of the above sample with two PSC powder/CSO gel/ScSZ samples
  - Our current best sample is PSC powder/CSO gel/ScSZ
    - *Fired at 1100°C (as opposed to 1000°C) and with better electrode flatness*

Sample	ASR at 800°C	Voltage loss (800°C)	ASR at 900°C	Voltage loss (900°C)
PSC/PSC/CSO/YSZ	0.29 Ω-cm^2	58 mV	0.14 Ω-cm^2	28 mV
PSC/CSO/SSZ (1)	0.34 Ω-cm^2	68 mV	0.14 Ω-cm^2	28 mV
PSC/CSO/SSZ (2)	0.071 Ω-cm^2	14 mV	0.032 Ω-cm^2	6.4 mV
PSC/CSO/SSZ (3)	0.31 Ω-cm^2	62 mV	0.16 Ω-cm^2	32 mV

  - The electrode fabrication process is being improved by spraying CSO gel on heated substrates. This allows faster application rates of thicker ceria protection layers. We have also begun to spray powders to create flatter electrodes.

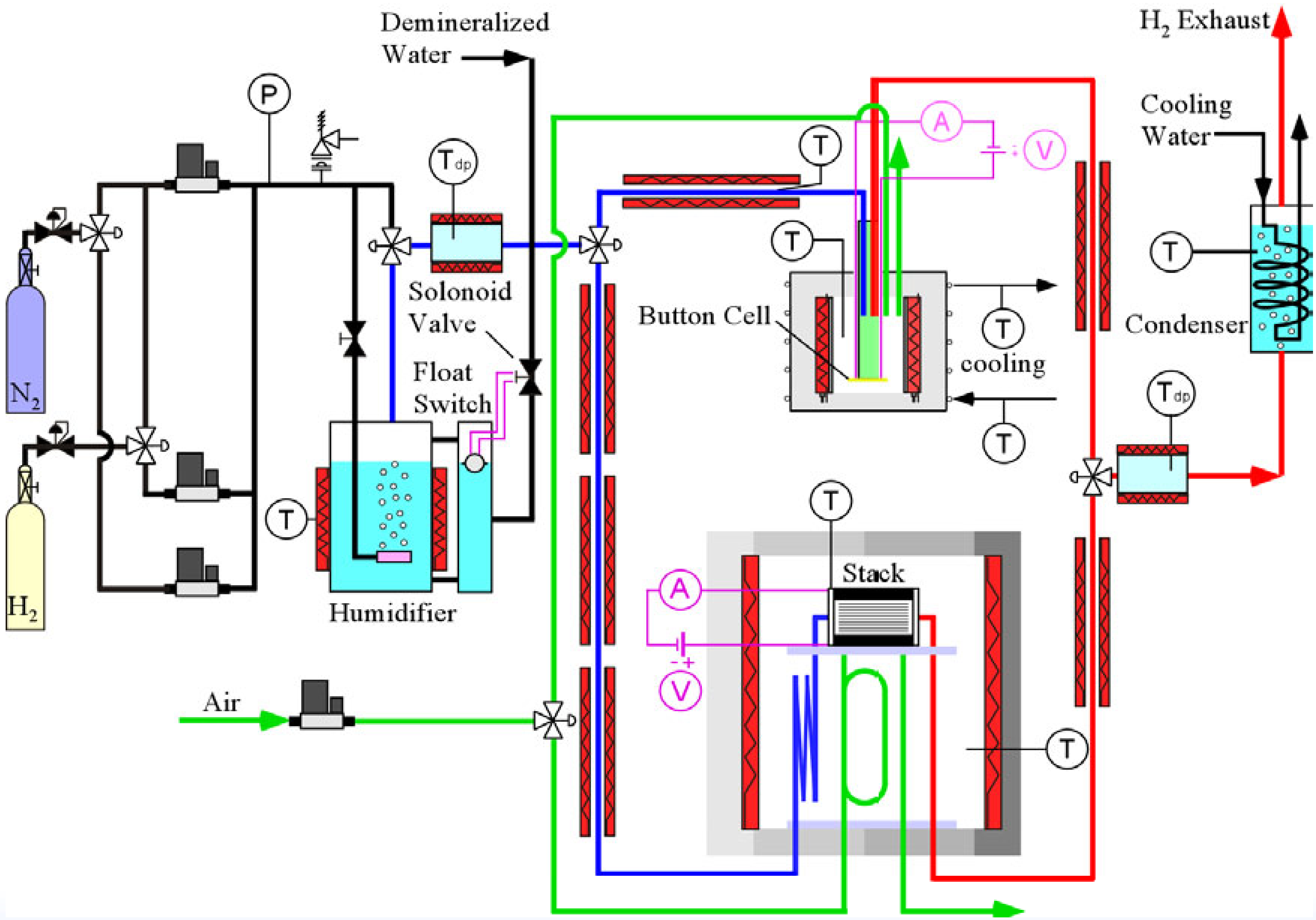
Hydrogen/Steam Electrode Development

- Measured Ni-8YSZ electrode ASR changes with steam/hydrogen ratio at 800°C.

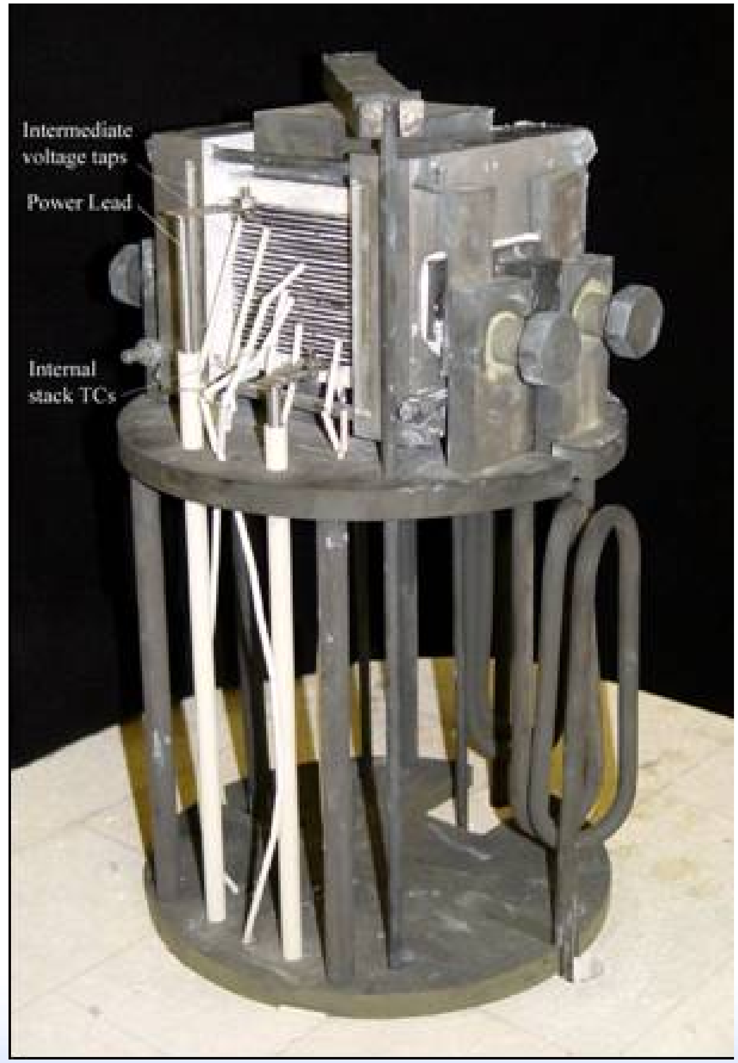
Sample	$H_2O:H_2 = 0.8$	$H_2O:H_2 = 9$	$H_2O:H_2 = 166$
Ni-8YSZ (5% CeO_2)	2.2 Ω-cm^2	1.7 Ω-cm^2	2.9 Ω-cm^2
Ni-8YSZ (1% Ru)	81 Ω-cm^2	77 Ω-cm^2	108 Ω-cm^2

- Alternate materials update—currently in the process of fabricating:
  - NbB_2/ScSZ (powder mixture) electrode
  - $TiNb_2O_7$/8YSZ (powder mixture) electrode
  - $La_{0.8}Sr_{0.2}Cr_{0.7}Ni_{0.3}O_3$ perovskite thin layer with current collector
- Additional funding needed to
  - Accelerate work on degradation-resistant hydrogen electrodes and improved oxygen electrodes.
  - Perform experimental studies to determine and mitigate the source of electrical resistance in stack configurations.

Schematic of Stack Testing Apparatus

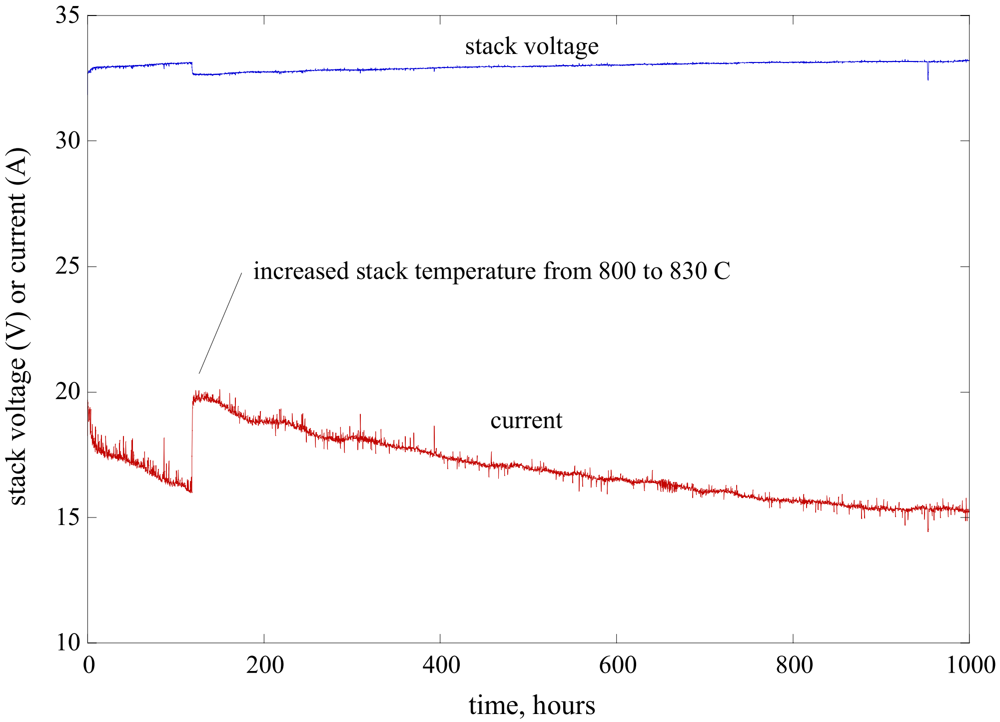


25-cell stack used in 1000-hour test Jan. 4 – Feb. 16, 2006

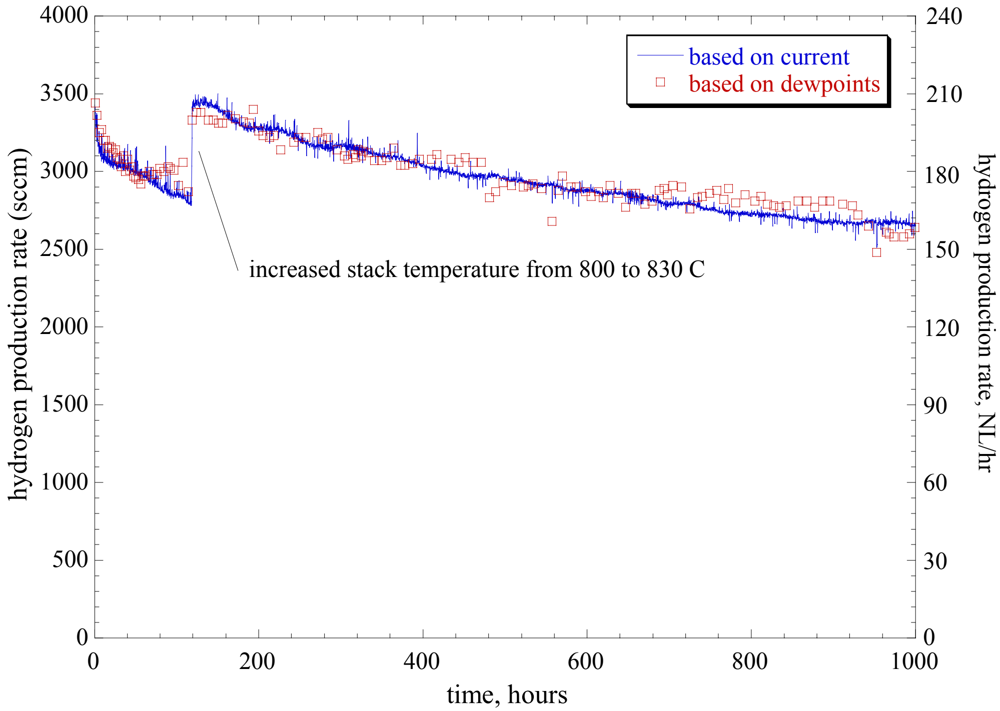


1000-hour electrolysis test, 25-cell stack

Stack voltage and current



Hydrogen production rate



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Design of Integrated Laboratory Scale Experiment

Test objectives

	Button	Stack		Facility	
	(1.5 W)	Bench (500 W)	Integrated Lab (15 kW)	Pilot (200 kW)	Engineering Demonstration (5 MW)
Electrode / Electrolyte Materials	x				
Electrode/Electrolyte performance	x	x			
Basic cell design	x	x			
Stack design		x			
Stack sealing		x	x		
Stack performance		x	x		
Manifolding		x	x	x	
Electrical configuration			x	x	
Instrumentation development	x	x	x	x	
Heating of feedstock			x	x	x
Product gas heat recuperation			x	x	x
Hydrogen recycle			x	x	x
High-temperature oxygen handling			x	x	x
Stack lifetime			x	x	x
Hydrogen purification				x	x
System startup and control			x	x	x
System maintenance				x	x
High-pressure operation				x	x
Hydrogen storage					x
Demonstration of large-scale hydrogen production					x

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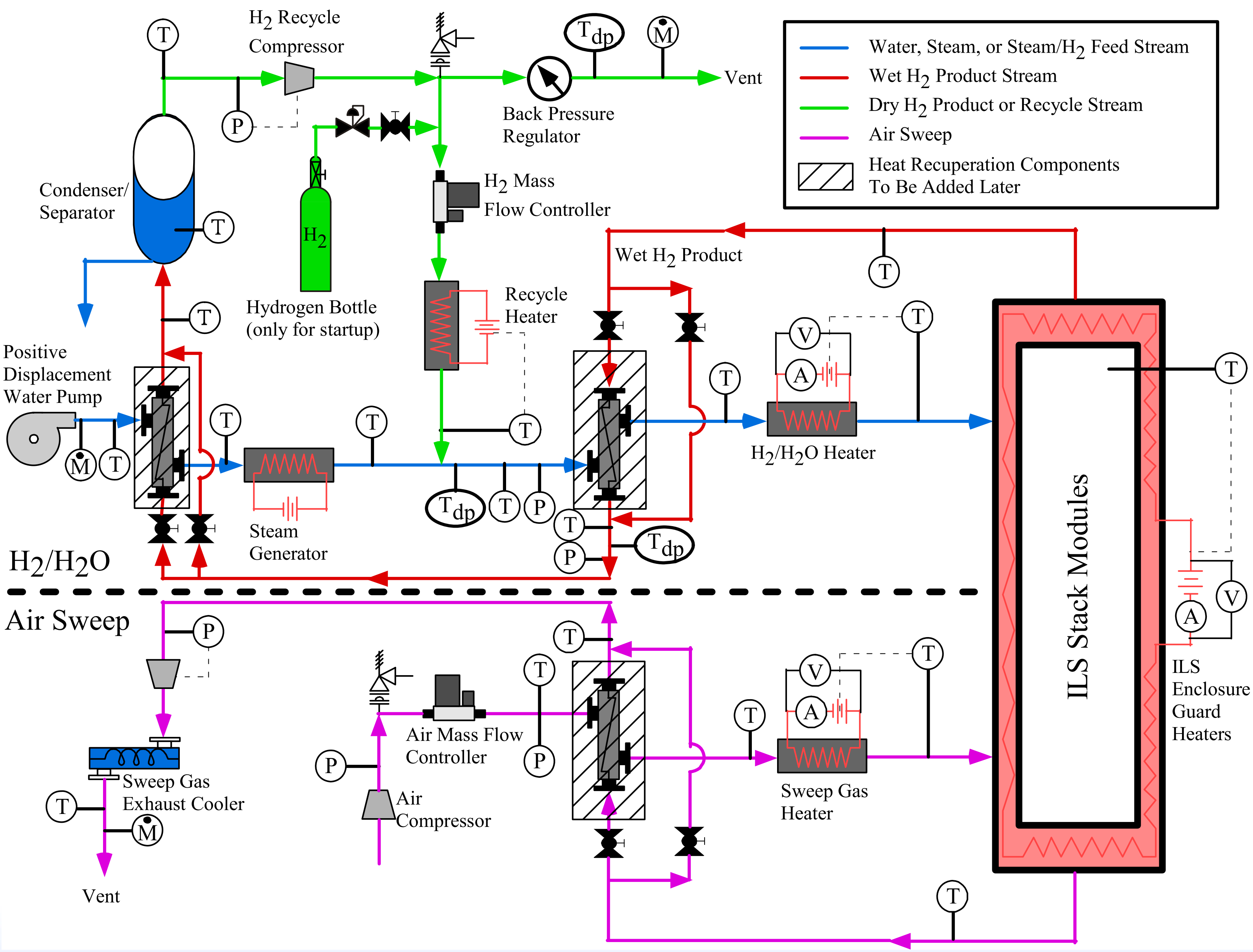
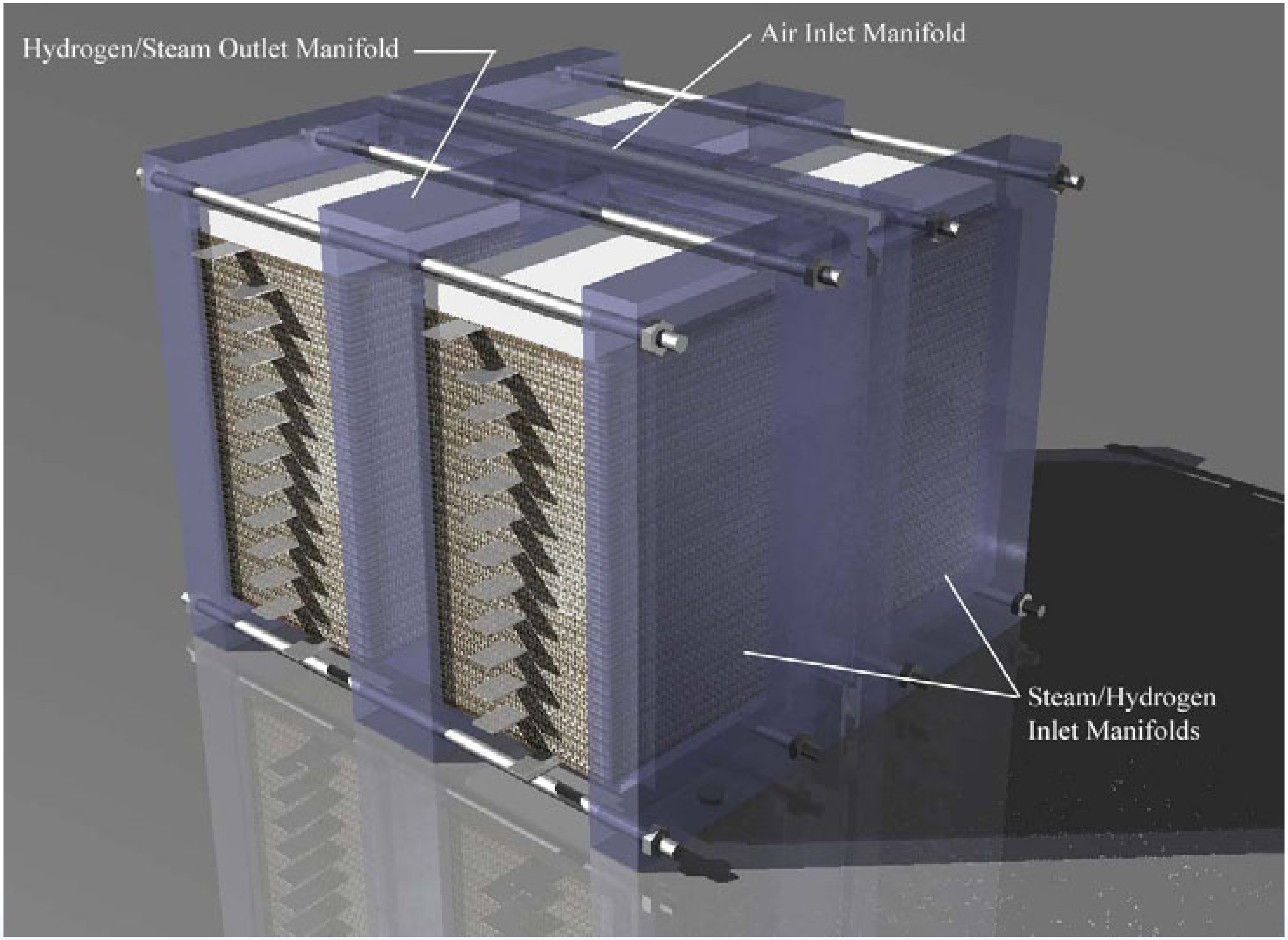
Advanced Materials & Electrochemical Technologies

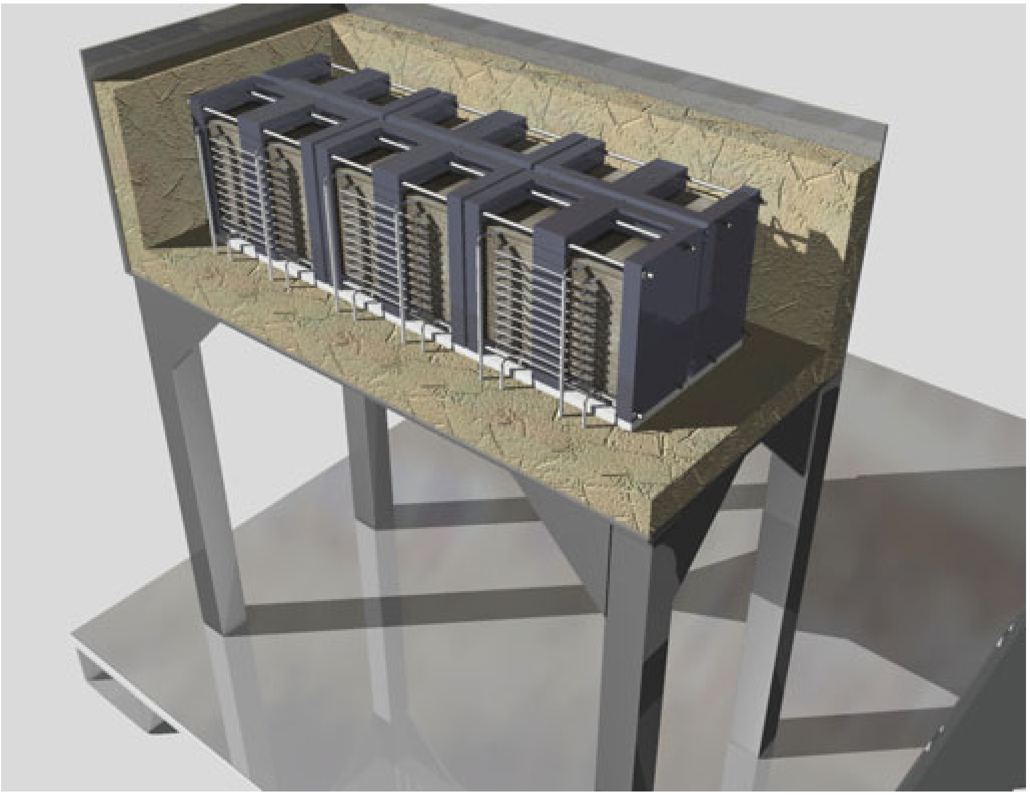
Schematic of Integrated Laboratory-scale system for HTE



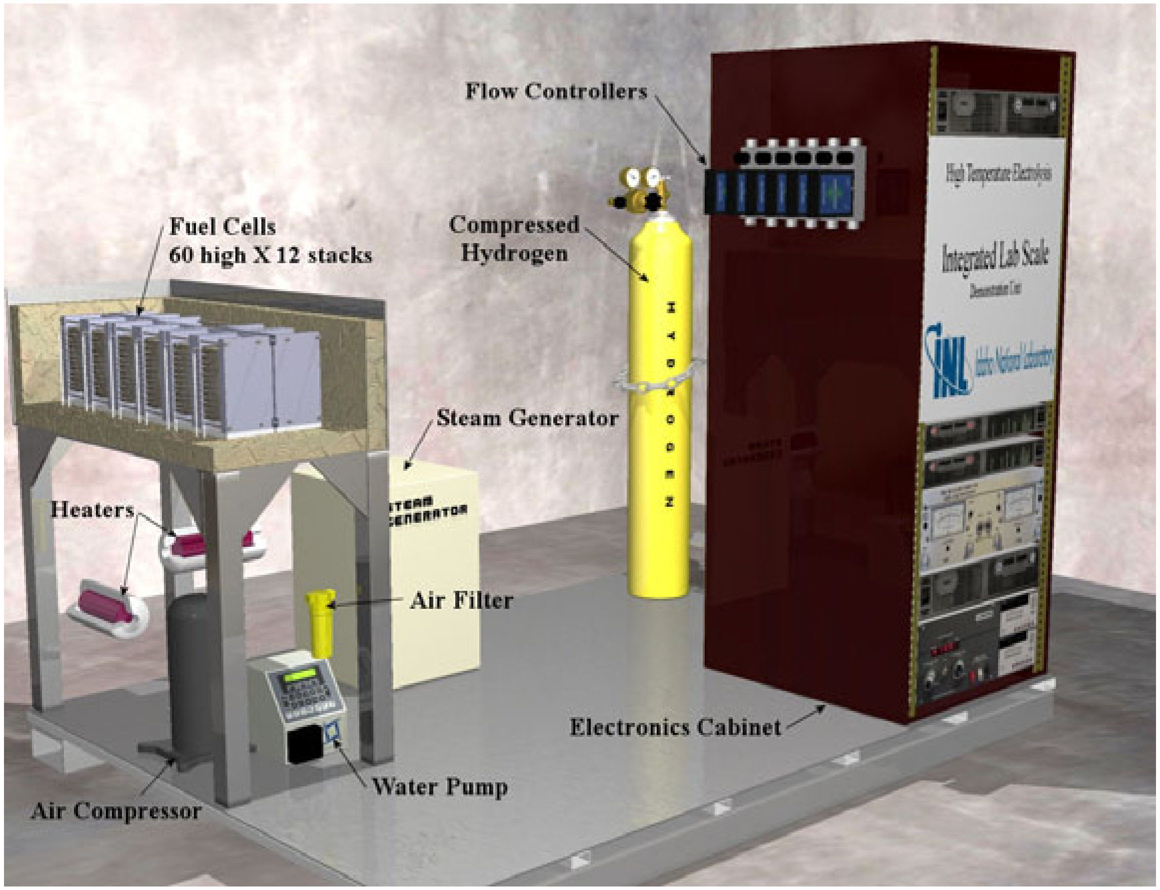
Integrated Laboratory-scale Experiment
Four-stack module



ILS Stack and Hot Zone dimensional details	
Cells/stack	60
Stack height	15 cm
Stack volume	1500 cm^3
Stack volume with manifolds	4260 cm^3
Number of stacks	12
Total volume of stacks with manifolds	0.051 m^3
Hot volume	0.100 m^3
Stacks per module	4
Number of modules	3
Hot volume height	0.28 m
Hot volume width	0.38 m
Hot volume length	0.94 m



ILS system mounted on skid

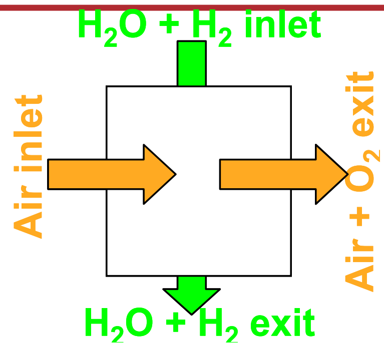


Independent Design and Operational Parameters	
active cell area	64 cm^2
cells per stack	60
number of stacks	12
stack operating temperature	830°C
steam utilization	50%
stack operating voltage	78 V
per-cell ASR	1.5 Ohm cm^2
inlet steam mole fraction	0.9
inlet hydrogen mole fraction	0.1

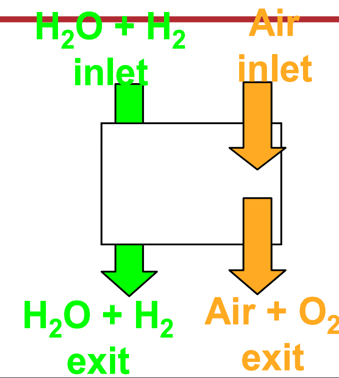
Calculated Performance Values	
Per-cell operating voltage	1.3 V
current density	0.271 A/cm^2
stack power	1352 W
total power	16.2 kW
inlet hydrogen flow rate	19.3 SLPM
inlet steam flow rate	174 SLPM
inlet liquid water flow rate	140 cc/min
air flow rate	103 SLPM
hydrogen production rate	5220 NL/hr
heating value of hydrogen produced	15.6 kW

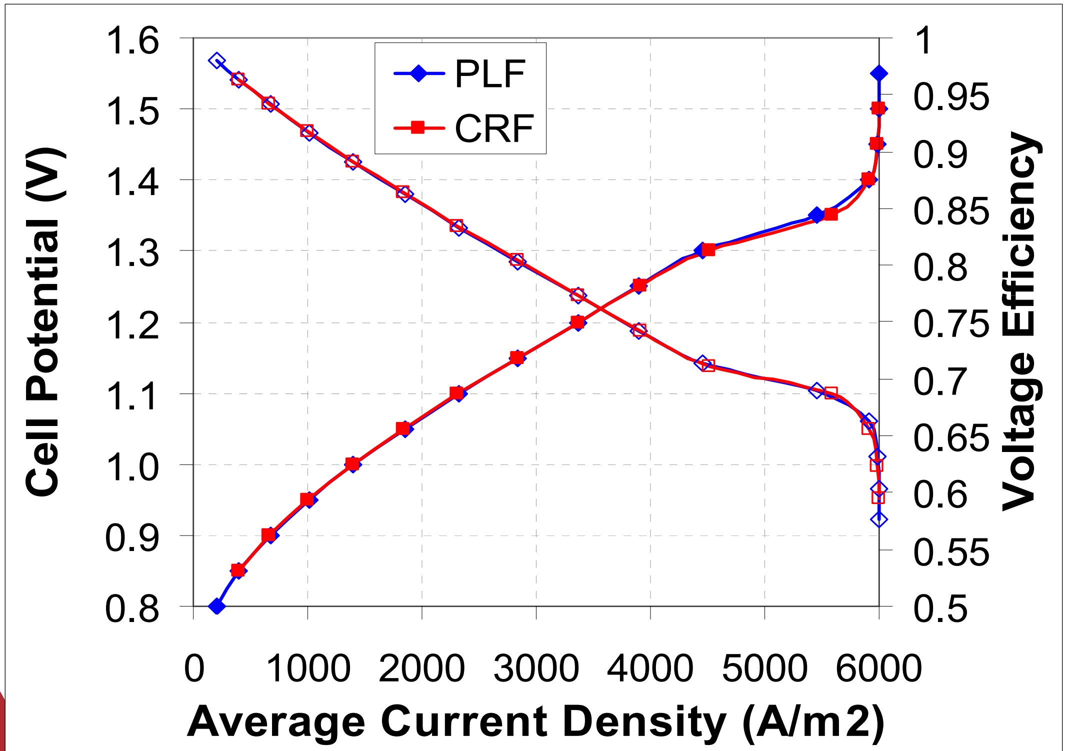
Effect of Flow Inlet Configurations: Polarization and Average Cell Temperature

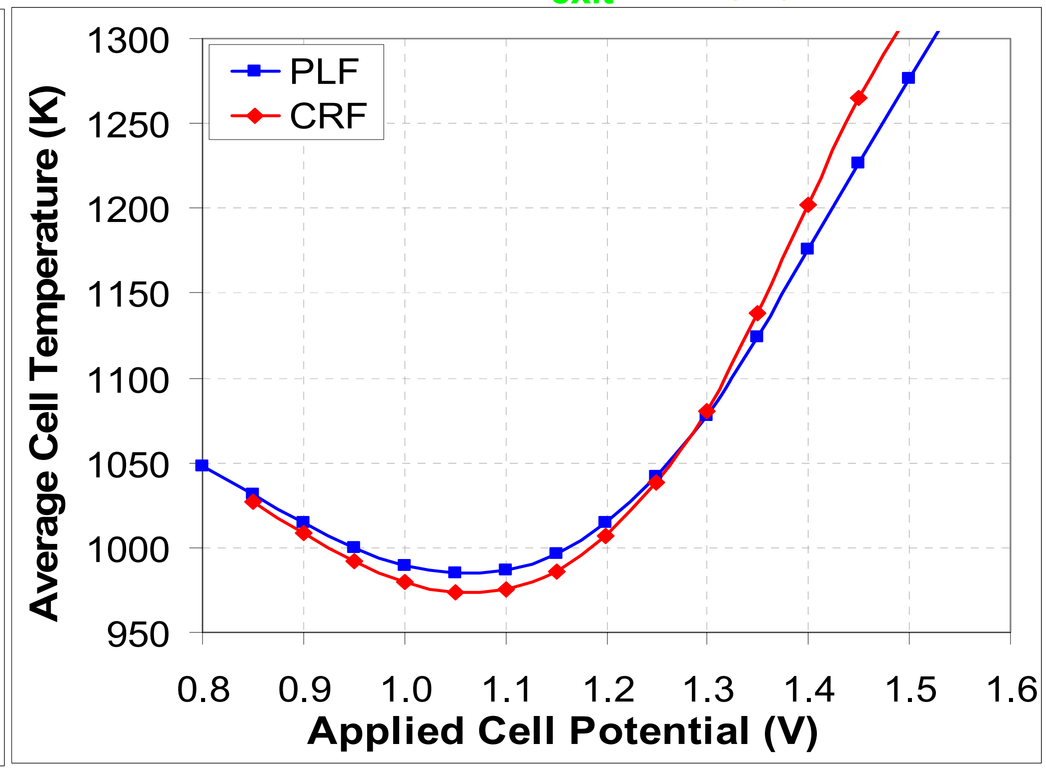
Cross Flow (CRF)



Parallel Flow (PLF)

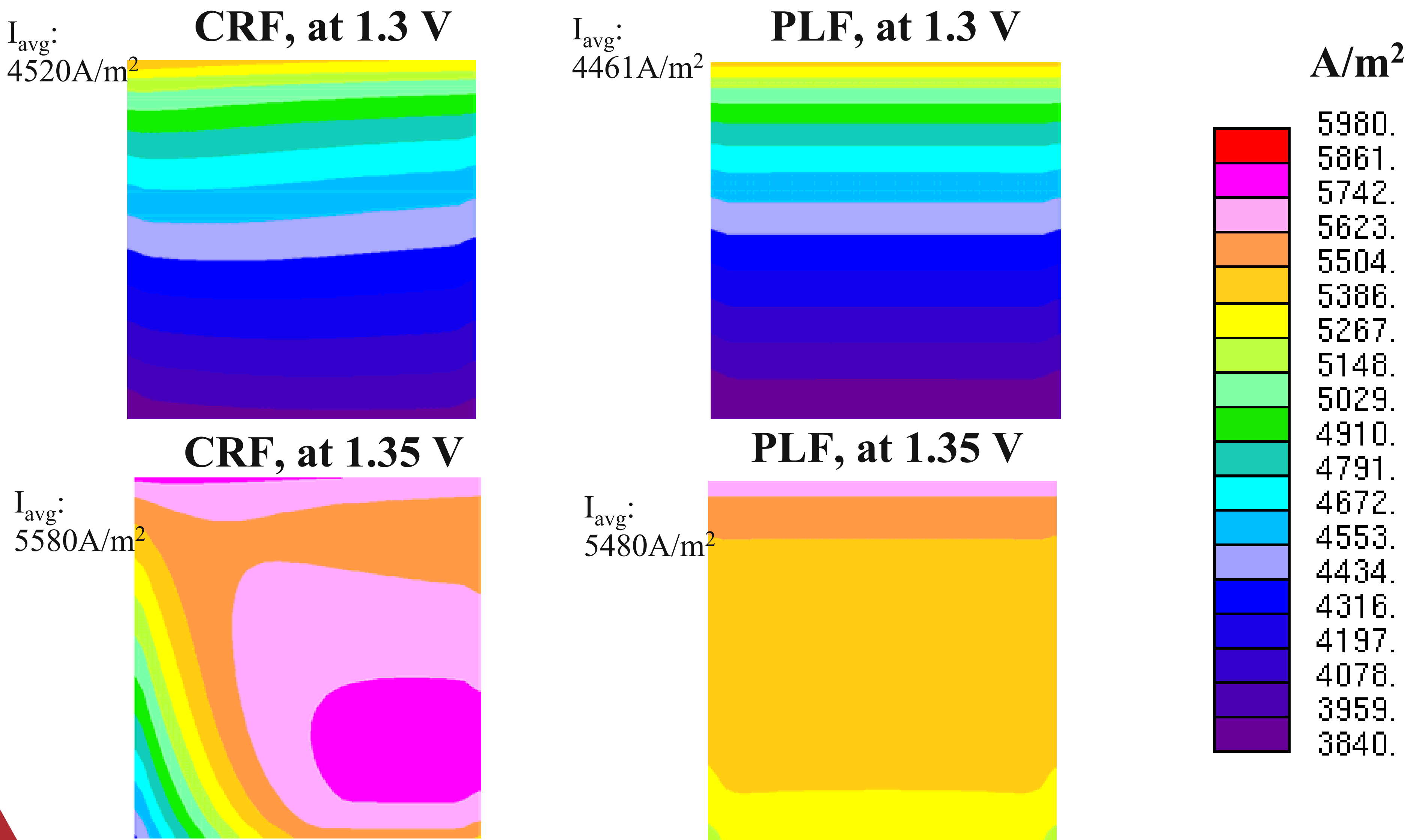






- Similar overall performance.
- Different temperature and current density profiles.

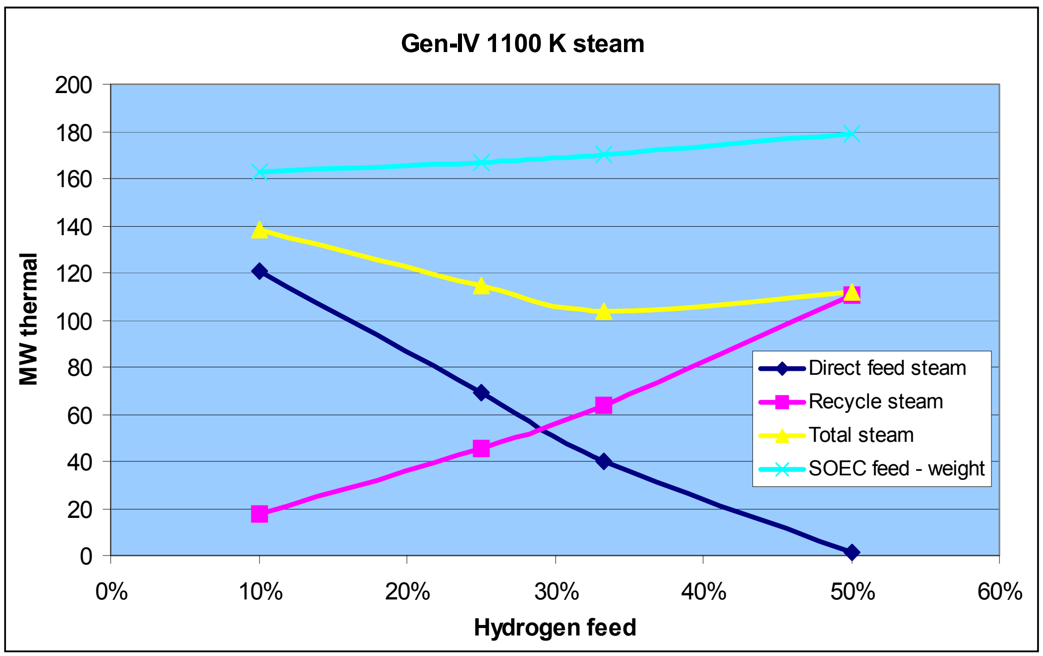
Simulated Current Density Profile for Cross and Parallel Flow Configurations

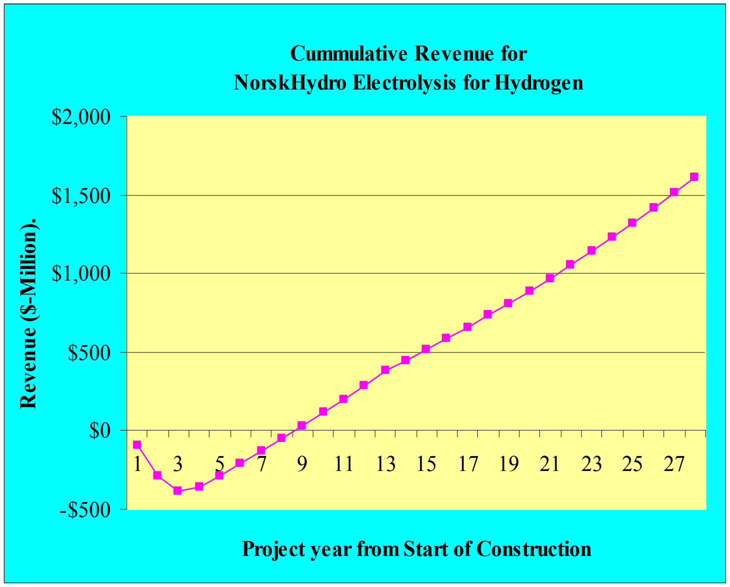


- More uniform utilization of steam with the parallel flow configuration at higher potentials.

Flowsheet Analysis of Solid Oxide Electrolyzer Cells and Electrolyser Plants

- Premium *(Electric)* Power demands are much lower for SOEC
- Premium heat demands now needed for SOEC
- Physical plant footprint will be larger for SOEC
- Gen-IV 1100K thermal power input can be minimized by thermal load management (50% conversion)
- Detailed economic assessment (beyond H2A)





Response to 2005 Reviewers' Comments:

Comment: Wouldn't a Mass Spec or GC measurement of hydrogen production be more direct than the dewpoint and electrical measurements?
Response: In the past year we have occasionally used a gas chromatograph (Agilent 3000A Micro GC), but found that the results were less consistent than the dewpoint and electrical measurements.

Conclusions

- **Experimental results from a 25-cell stack, 64 cm^2 active area, fabricated by Ceramatec:**
  - **Hydrogen production rates in excess of 160 NL/hr were maintained with a 25-cell solid-oxide electrolysis stack for 1000 hours over the time period from January 5 to February 17, 2005.**
  - **The stack endurance test was terminated due to completion of the milestone and not due to any problem with the stack itself.**
  - **Stack performance as measured by the per-cell ASR was good, beginning at 1.60 ohm-cm^2 and finishing at 2.15 ohm-cm^2.**
- **The Integrated Laboratory Experiment will produce ~30x more hydrogen, while incorporating, at reduced scale, all of the components of a commercial plant.**
- **The collaboration between ANL and INL and in the development of electrodes and electrolytes has been very fruitful.**
- **INL, FLUENT and ANL developed electrochemical CFD models for electrolysis that predicts product flow, current density, temperature and ASR distributions within the SOEC.**
- **Thermal and electrochemical performance of SOECs can be greatly improved through design.**

Publications

- Herring, J. S., O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "Progress in High-Temperature Electrolysis for Hydrogen Production using Planar SOFC Technology," accepted for publication in the *International Journal of Hydrogen Energy,* 2006.
- O'Brien, J. E., Stoots, C. M., Herring, J. S., and Hartvigsen, J. J.,"Hydrogen Production Performance of a 10-Cell Planar Solid-Oxide Electrolysis Stack," *Journal of Fuel Cell Science and Technology*, May, 2006.
- Stoots, C. M., O'Brien, J. E., McKellar, M. G., Hawkes, G. L., and Herring, J. S., "Engineering Process Model for High-Temperature Steam Electrolysis System Performance Evaluation," AIChE 2005 Annual Meeting, Cincinnati, Oct. 30 – Nov. 4, 2005.
- Herring, J. S., O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "High Temperature Electrolysis for Hydrogen Production Using Nuclear Energy" Paper #501, GLOBAL 2005, Paper #501, Tsukuba, Japan, Oct. 9 – 13, 2005.
- O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "Comparison of a One-Dimensional Model of a High-Temperature Solid-OxideElectrolysis Stack with CFD and Experimental Results," presented at the 2005 ASME International Mechanical Engineering Congress and Exposition, Nov. 5 – 11, Orlando.
- Hawkes, G. L., O'Brien, J. E., Stoots, C. M., Herring, J. S., Shahnam, M., "Thermal and Electrochemical Three Dimensional CFD Model of a Planar Solid Oxide Electrolysis Cell," Proceedings, 2005 ASME Heat Transfer Conference, July 17-22, 2005, San Francisco.
- O'Brien, J. E., Stoots, C. M., Herring, J. S., Lessing, P. A., Hartvigsen, J. J., and Elangovan, S., "Performance Measurements of Solid-Oxide Electrolysis Cells for Hydrogen Production from Nuclear Energy," *Journal of Fuel Cell Science and Technology*, Vol. 2, August 2005, pp. 156-163.
- Herring, J. S., O'Brien, J. E., Stoots, C. M., and Hawkes, G. L., "Progress in High-Temperature Electrolysis for Hydrogen Production using Planar SOFC Technology," 2005 AIChE Spring Annual Meeting, April 10 – 14, 2005, Atlanta, GA.
- O'Brien, J. E., Stoots, C. M., Herring, J. S., and Hartvigsen, J. J.,"Hydrogen Production Performance of a 10-Cell Planar Solid-Oxide Electrolysis Stack," Proceedings, ASME 3rd International Conference on Fuel Cell Science, Engineering, and Technology, May 23 – 25, 2005, Ypsilanti, MI.
- Hawkes, G. L., O'Brien, J. E., Stoots, C. M., Herring, J. S., Shahnam, M., "CFD Model of a Planar Solid Oxide Electrolysis Cell for Hydrogen Production from Nuclear Energy," to be presented at the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics NURETH-11, Popes Palace Conference Center, Avignon, France, October 2-6, 2005.
- O'Brien, J. E., Herring, J. S., Stoots, C. M., Lessing, P. A., "High-Temperature Electrolysis for Hydrogen Production From Nuclear Energy," to be presented at the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics NURETH-11, Popes Palace Conference Center, Avignon, France, October 2-6, 2005.