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## LIST OF ACRONYMS

BAS	Bioanalytical Systems
CCE	Catalyst Coated Electrode
CCM	Catalyst Coated Membrane
CV	Cyclic Voltammogram
DI-water	Deionized water
DM	Direct Methanol
DMFC	Direct Methanol Fuel Cell
DOE-NE	Department of Energy, Office of Nuclear Energy
EDAX	Energy Dispersive Spectroscopy
EIS	Electrochemical Impedance Spectroscopy
EW	Equivalent Weight
GES	Gener Electrochemical Systems
HyS	Hybrid Sulfur
ILS	Integrated Lab-Scale
LSV	Linear Sweep Voltammogram
MEA	Membrane Electrode Assembly
NHI	Nuclear Hydrogen Initiative
OCP	Open Circuit Potential
OPM	Oxford Performance Materials
PA	Phosphoric acid
PAFC	Phosphoric Acid Fuel Cell
PBI	Poly-Benzimidazole
PEM	Proton Exchange Membrane and Polymer Electrolyte Membrane
PEMFC	Polymer Electrolyte Membrane Fuel Cell
PFSA	Perfluorinated Sulfonic Acid
RT	Room Temperature, 25 °C
SCUREF	South Carolina Universities Research and Education Foundation
SDAPP	Sulfonated Diels-Alder Polyphenylenes
SDE	Sulfur Dioxide-depolarized Electrolyzer
SEM	Scanning Electron Microscopy
SHE	Standard Hydrogen Electrode
SNL	Sandia National Laboratory
SPEK	Sulfonated Poly-Etherketone
SPEKK	Sulfonated Poly-Etherketone-ketone
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

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## EXECUTIVE SUMMARY

The proof of concept of SO<sub>2</sub> electrolysis for the hybrid sulfur (HyS) process is a priority research target for FY 2008. Technical options must be better defined and the challenges better understood. The SO<sub>2</sub>-depolarized electrolyzer development is proceeding from single cell testing to larger multi-cell stack testing. Improvement of cell performance is being addressed through component development and testing. The overall HyS process development is being advanced through process flowsheet improvement, conceptual design studies for commercial equipment, and process design in partnership with industrial companies. Work during the first quarter of FY08 was conducted in the areas of Component Development, Single Cell Testing, Multi-Cell Stack Testing, and ongoing tasks related to Conceptual Design Study for Commercial Electrolyzer and HyS Flowsheet and Process Design in conjunction with Westinghouse Electric.

During the first quarter of FY 2008, SRNL continued its efforts with the Electrolyzer Component Development. SRNL tested platinum catalyzed carbon as an anode electrocatalyst. This approach was utilized to improve catalyst utilization and mass transport of sulfur dioxide to the active sites. Different treatments to the carbon surface were implemented in order to improve anchoring of the catalyst particles. Platinum alloys obtained from Columbian Chemicals Company were tested for sulfur dioxide kinetics. The alloys tested are platinum-cobalt, platinum-cobalt-nickel, platinum-cobalt-chromium and platinum-iridium-cobalt. So far testing has shown an improvement in the oxidation kinetics of 20 mV in comparison with pure platinum.

A key component of the SDE is the ion conductive membrane through which protons produced at the anode migrate to the cathode and react to produce hydrogen. One of the goals of SRNL's FY07 research program was to evaluate commercial and experimental membranes for the SDE. A milestone report, WSRC-STI-2007-00172 Rev 0, "Baseline Membrane Selection and Characterization for an SDE", was issued to the Department of Energy Office of Nuclear Energy during the third quarter of FY2007. In this report, SRNL provided a summary of the results from the characterization studies of the commercial and experimental membranes for the SDE. This work continued during the FY08 First Quarter. Several new experimental membranes were selected for characterization, including membranes from Case Western University, Clemson University, Sandia, Penn State, UNC and Giner. SRNL also prepared platinum treated membranes in an effort to reduce SO<sub>2</sub> transport. In addition, new membrane samples provided by Clemson University were tested. The samples are fluorocarbon based with different conducting groups. The membranes showed reduced crossover while maintaining ionic conductivity.

Work was initiated on a new component development task that that will greatly increase our ability to rapidly characterize various cell components under differing operating conditions without the need for testing in the larger electrolyzer facility. This is particularly important at this time since the single cell electrolyzer facility will not be available for MEA characterization testing for several months in order to permit testing of the multi-cell stack. A new bench-scale electrolyzer test station was designed; fabrication and assembly will be completed next month. It will be capable of testing approximately 1 cm<sup>2</sup> samples of MEA's

under ambient pressure conditions. This compares to the 60 cm<sup>2</sup> MEA required for the single cell test facility.

Single cell electrolyzer testing continued during the reporting period. Seven new MEA's were tested, with the primary objective of providing a better understanding of the design and operating conditions that effect the formation of a sulfur layer inside the cell MEA. Based on detailed analyses of past single cell electrolyzer testing, it was observed that the formation of a sulfur-containing layer between the membrane and the cathode electrode was influenced by type of catalyst support and the morphology of the platinum electrocatalyst. To further examine this effect, we initiated preparation of MEAs containing an unsupported platinum catalyst (i.e., Pt black). An MEA with the revised catalyst layers was fabricated and tested in the single cell electrolyzer. Analysis with a scanning electron microscopic examination revealed a sulfur layer similar to that previously observed. However, the examination also revealed higher than normal carbon residues on the cathode catalyst, which is likely the byproduct of glycerol used in the fabrication process. We are currently modifying the fabrication method to reduce the carbon content, since carbon is a potential contributor to the sulfur layer formation. Several new MEA's using Pt-black cathode catalysts were also procured from Giner Electrochemical and tested. Post-test examinations are in progress, and the results will be reported in the next quarterly report.

Work was initiated on the design of a mounting fixture to allow the installation of the larger multi-cell stack into the test facility. This stack has a total electrolyzer cell area that is 8 times larger than the current single cell electrolyzer. The installation will require changes to the plumbing of the system to include the inlets and outlets for the additional cells while maintaining the separation of current between the individual cells. The pressure protection requirements are being evaluated to determine if any changes are required due to the expected increase in hydrogen production and the addition of the flow loops for the additional cells. Physical modifications to the test facility and installation of the multi-cell stack will begin immediately following completion of single cell testing of MEA #25. A Level 1 milestone is scheduled for March 15, 2008 to complete multi-cell stack testing.

A subcontract change with Giner Electrochemical was initiated to allow design and testing of a Gas Diffusion Electrode. The initial part of the contract will include the design and fabrication of a test-stand. Work also continued on the process design and cost analysis of a commercial HyS electrolyzer. Improved HyS flowsheets were developed in conjunction with Westinghouse Electric and PBMR under the Technical Consulting Agreement. This work is planned to be completed next quarter.

## 1.0 INTRODUCTION

Hydrogen has been identified as a leading candidate to replace petroleum as part of the transition to a sustainable energy system, and major efforts are being conducted worldwide to develop the technologies and supporting activities required for this transition. In the United States, the federal research efforts are led by the U.S. Department of Energy (DOE). The U.S. DOE Hydrogen Program is an integrated inter-office program being conducted by the Office of Energy Efficiency and Renewable Energy, Office of Nuclear Energy (DOE-NE), Office of Fossil Energy and Office of Science. The primary objective of the DOE-NE Nuclear Hydrogen Initiative (NHI) is to develop the nuclear hydrogen production technologies necessary to produce hydrogen at a cost competitive with other alternative transportation fuels. The focus of the NHI is on thermochemical cycles and high temperature electrolysis.

The Savannah River National Laboratory (SRNL) has been tasked with the primary responsibility to perform research and development in order to characterize, evaluate and develop the Hybrid Sulfur (HyS) thermochemical process. The HyS Process uses a sulfur dioxide depolarized electrolyzer (SDE) to split water and produce hydrogen. During FY05 and FY06, SRNL designed and conducted proof-of-concept testing for a SDE using a low temperature, PEM fuel cell-type design concept. The advantages of this design concept include high electrochemical efficiency and small footprint, characteristics that are crucial for successful implementation on a commercial scale. During FY07, SRNL extended the range of testing of the SDE to higher temperature and pressure, conducted a 100-hour longevity test, and designed and built a larger, multi-cell stack electrolyzer.

The proof of concept of SO<sub>2</sub> electrolysis for the HyS Process is a priority research target for the FY 2008 NHI Program. Technical options must be better defined and the challenges better understood. The current status of electrolyzer performance must be established by operation at elevated temperature (>90C) and pressure (>10 atmospheres) and during a long duration run (>100 hours). SRNL is pursuing the liquid-phase sulfur dioxide decoupled electrolyzer (SDE) option, which is the main focus of the NHI work. The rate of development of HyS will depend on the identification of a promising membrane or an alternative means for controlling sulfur formation at the cathode of the cell. SRNL will work with Sandia National Laboratory (SNL), universities, and industry to address this issue. Electrolyzers of larger size will be required as the process development proceeds, and SRNL will test a multi-cell stack that was built in FY 2007. Work will be initiated for a Hybrid Sulfur Integrated Laboratory-Scale (ILS) Experiment that will combine a SRNL electrolyzer with the sulfuric acid decomposer developed by SNL for the S-I ILS. A review will be held at mid-year, and if progress warrants, work will progress to the ILS level including ILS system design, electrolyzer fabrication and infrastructure development. Benchmarks to be considered include electrochemical efficiency, membrane durability, and minimization of SO<sub>2</sub> crossover.

Work performed on NHI Work Package N-SR07TC0301 during the first quarter of FY08 is presented herein. Technical Progress is given in Section 2.0. The Project Management activities are reported in Section 3.0. Future Work is discussed in Section 4.0.

## 2.0 TECHNICAL PROGRESS

The SRNL Work Package for FY08 contains the following major tasks:

- Conceptual Design Study for Commercial Electrolyzer
- HyS Flowsheet and Process Design (Westinghouse TCA)
- Electrolyzer Component Development
- Development of Gas Diffusion Electrode
- Single Cell Performance Testing
- Multi-cell Stack Testing
- Upgrade Test Facility to 90-100 C and >10 atm
- Process Flowsheet Optimization
- ILS Planning and Design

The progress on each of these tasks during the First Quarter of FY08 is discussed in the following sections.

### 2.1 CONCEPTUAL DESIGN STUDY FOR COMMERCIAL ELECTROLYZER

The purpose of this task is to develop a conceptual design and cost estimate for a full-size commercial electrolysis system for the HyS Process. The following preliminary specification for a commercial electrolyzer module was developed:

Table 2.1  
Commercial Electrolyzer Specification and Design Requirements

Type	Proton Exchange Membrane
Design Concept	Bi-polar MEA Cell
Operating Temperature	100°C
Operating Pressure	20 atm
Current Density	500 mA per cm <sup>2</sup>
Avg Cell Voltage	600 mV
Active Area per Cell	1.0 m <sup>2</sup>
Cells per Module	200
DC Input per Module	600 kWe
H <sub>2</sub> Output per Module	37.6 kg per hour
Anolyte Inlet Acid Conc.	50wt% (excluding SO <sub>2</sub> )
Anolyte Inlet SO <sub>2</sub> Conc.	15 g per 100 gm acid
SO <sub>2</sub> Utilization	40%

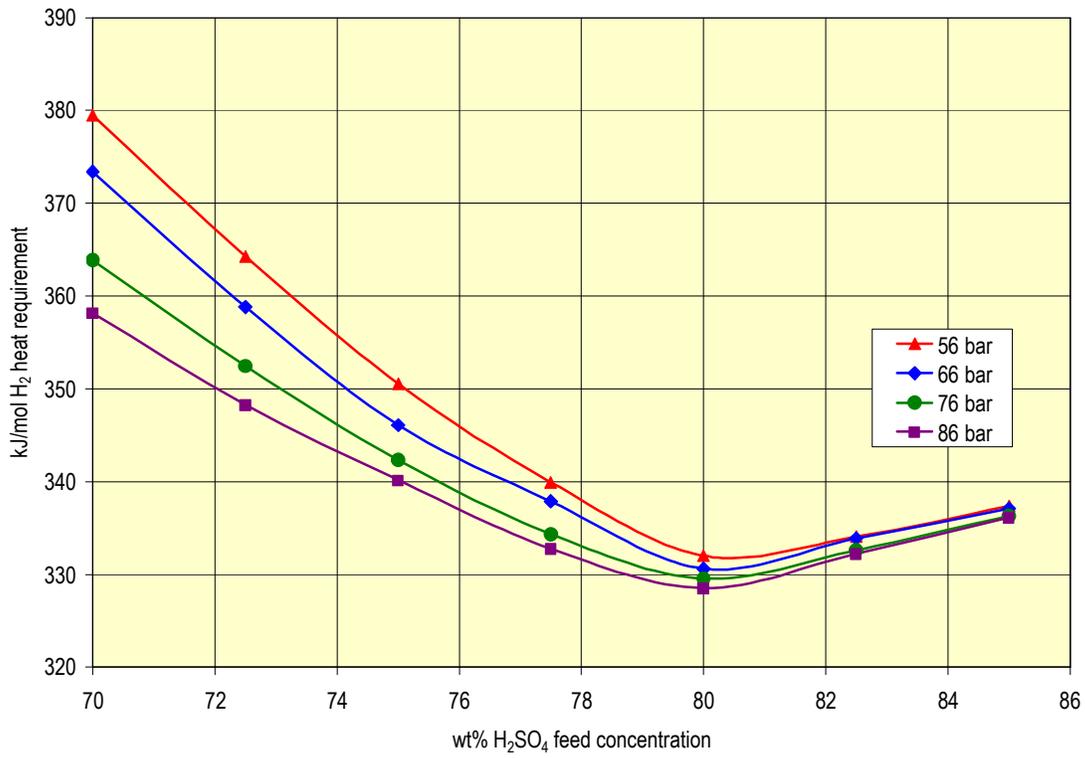
The development of the electrolyzer conceptual design will leverage existing design experience for similar electrochemical cells to as great an extent as possible. A subcontract has been let to Giner Electrochemical LLC for the purpose of reviewing the current state-of-the-art technologies for electrolyzers. This report will provide an industry perspective on current electrolyzer technologies and their potential application to the HyS Process. This report will describe industrial electrolysis systems at the individual cell level, the electrolyzer level, and the industrial plant level. Different cell types will be compared and contrasted with respect to the HyS application, and the pluses and minuses of the technologies will be discussed.

A discussion of industrial plants will be included focusing predominantly on the chlor-alkali and chlorate plants, but also mentioning, for contrast, aluminum electrowinning, high-speed electrogalvanizing, and electrodialysis. The fluid handling strategies to be considered in plant design will also be discussed. Electrical issues will include trade-offs of series and parallel stack connections, with an investigation of hybrid strategies. Differences in industrial electrolyzer equipment and operating costs, efficiencies, and maintenance issues will be discussed, as they relate to these electrical issues.

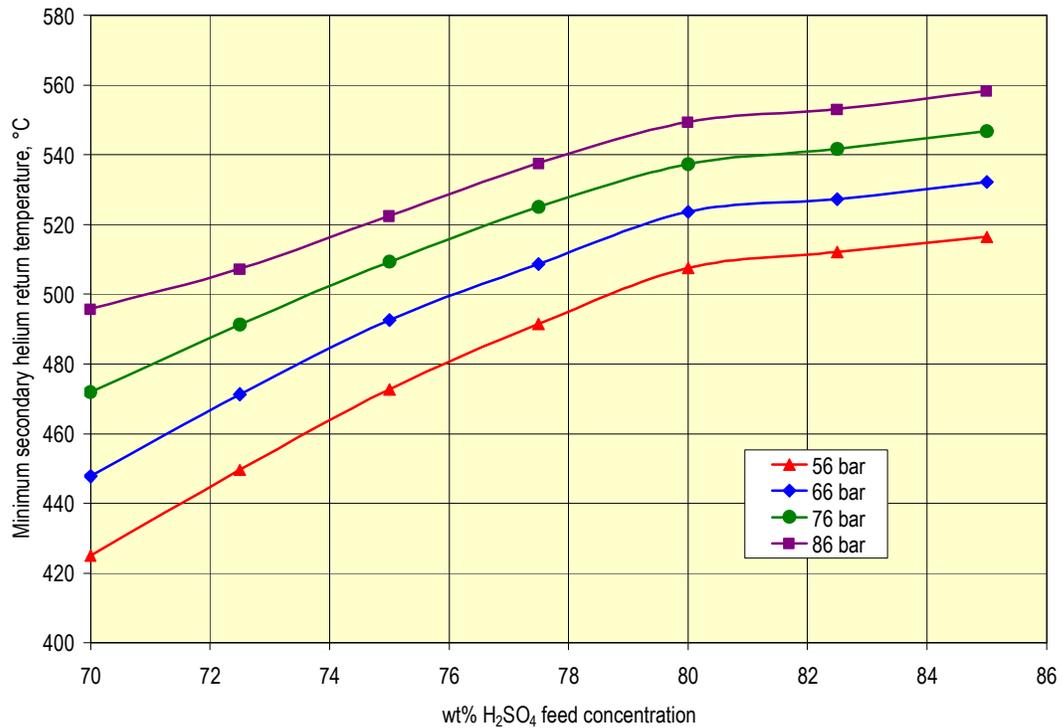
## **2.2 HYS FLOWSHEET AND PROCESS DESIGN**

During FY07 SRNL entered into a Technical Consulting Contract (TCA) with Westinghouse Electric Company. The purpose of this TCA is to permit SRNL to provide technical consulting services related to consolidating a reference design and technology program for the Hybrid Sulfur (HyS) Process Development for hydrogen production in conjunction with a High Temperature Gas-Cooled Reactor. Specifically, SRNL will incorporate Westinghouse's NGNP-based effort using the latest HyS flowsheet plus other HyS related advances from DOE's Nuclear Hydrogen Initiative (NHI) Program into a consolidated HyS design concept, a technology program plan and a cost estimate.

During the First Quarter of FY08, SRNL continued to update and optimize the HyS process flowsheet in conjunction with the design requirements provided by Westinghouse for the Pebble Bed Modular Reactor nuclear heat source. The overall heat integration system between the reactor and the hydrogen production process was optimized in order to maximize hydrogen production. The HyS flowsheet was modified to improve the efficiency of the acid concentration and H<sub>2</sub>SO<sub>4</sub> decomposition system. A revised flowsheet based on use of the Sandia National Laboratory's bayonet acid decomposer design was developed. Trade-off studies were conducted to determine the effect on energy requirements by varying the acid concentration in the bayonet feed stream. Figure 2-1 shows that the minimum energy requirement for the reactor is approximately 330 kJ/mol and occurs with an inlet acid concentration of 80wt%. Figure 2-2 shows the minimum secondary helium exit temperature from the reactor based on pinch analysis. Combined with knowledge of the secondary helium flowrate and temperature, which are functions of the nuclear reactor and secondary heat transfer system design, these figures can be used to calculate the maximum hydrogen output per reactor.



**Figure 2-1.** Helium heat target for Bayonet Decomposition Reactor



**Figure 2-2.** Secondary Helium Pinch Temperatures for Bayonet Decomposition Reactor

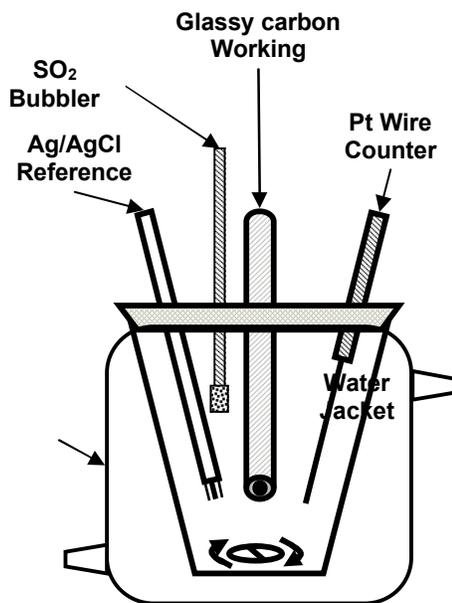
A complete mass and energy balance was completed for a PBMR reactor, secondary heat transport system and HyS Process. This is being reviewed by Westinghouse and will be revised based on their input. Work will continue on this task in the next quarter, including cost estimating. The results will be documented in a final topical report.

## 2.3 ELECTROLYZER COMPONENT DEVELOPMENT

The Electrolyzer Component Development task includes characterization, development and testing of each of the major components in the SDE, including the electrocatalysts, the proton exchange membrane, and the reactant diffusion layers. Based on results of component testing, components are selected for larger scale testing in the single cell electrolyzer. This task also includes the fabrication of Membrane Electrode Assemblies (MEA's) for the electrolyzer tests.

### 2.3.1 Catalyst Characterization

The short term stability of Pt alloy catalysts obtained from Columbian Chemicals Company was evaluated using the three electrode cells shown in 2-3. The cell consists of a glass vial with a PTFE cap and a water jacket. The three electrodes, which included a silver-silver chloride reference electrode, a platinum wire as the counter electrode, and a glassy carbon disk electrode, were inserted through the PTFE cap.

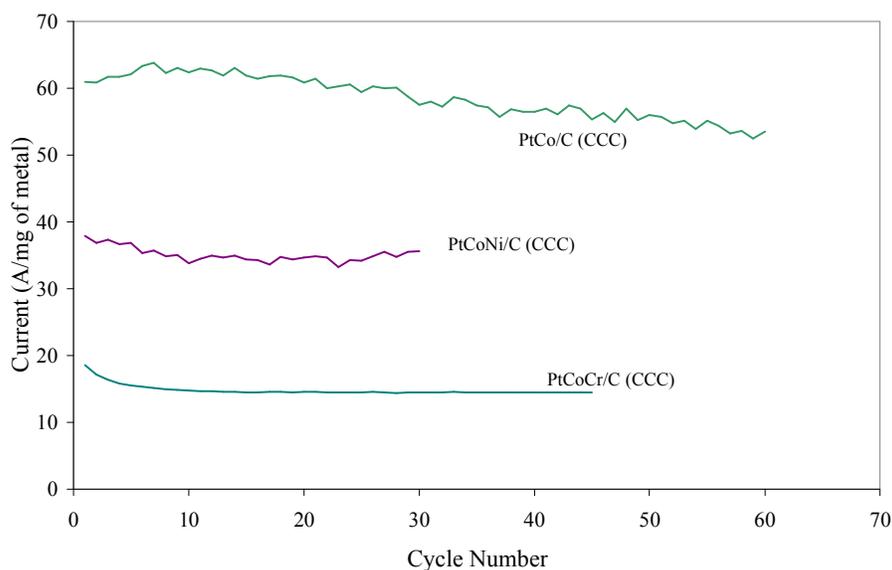


**Figure 2-3.** Simplified schematic of the catalyst characterization cell

During measurements the vial was filled with concentrated acid and purged of oxygen by flowing nitrogen. The catalyst's electrochemical characterization consisted of cyclic voltammograms (CVs) in the solution purged with nitrogen. The CVs were performed at a scan rate of 50 mV/sec and in a potential window between 1004 mV and -100 mV vs.

Ag/AgCl. The experiments were carried out at a temperature of 70 °C and 10.4 M sulfuric acid solution. The measurements were performed starting from the anodic potential and going in the cathodic direction.

Consecutive CVs were performed to study the stability of the catalyst and the different electrochemical reactions occurring at the surface of the electrode in the absence of SO<sub>2</sub>. During the CVs, the current was monitored as a function of a set potential which is varied at a constant rate. In general, the area under the hydrogen adsorption-desorption peak gives an idea of the electrochemically active surface area available for reaction. The difference in the hydrogen desorption peak height after consecutive cycling can be observed in Figure 2-4.



**Figure 2-4. Hydrogen desorption peak height after consecutive cycling for PtCo/C, PtCoNi/C and PtCoCr/C in 10.4 M H<sub>2</sub>SO<sub>4</sub> and 50 °C purged with N<sub>2</sub>.**

The highest peak is observed for Pt<sub>3</sub>Co/C catalyst followed by PtCoNi/C > PtCoCr/C. As can be observed the short term stability test show little or no degradation of the Pt alloy materials. An encouraging result as these catalysts shows enhanced kinetics for the oxidation of SO<sub>2</sub>. Further testing will be performed to select the most promising catalyst.

### 2.3.2 Membrane Electrode Assembly Preparation

The membrane electrode assemblies (MEAs) were prepared using two different procedures. In the case that platinized carbon is used as the catalyst material for the anode and cathode, the MEA was prepared by spraying the Pt catalyst ink directly on the membrane. The catalyst ink was prepared by ultrasonically blending a mixture of platinized carbon catalyst, with Nafion dispersion in the hydrogen form, water and methanol in an ultrasonic mixer for 1 hour. At the moment of spraying, the membrane is kept hot (80°C) to prevent the membrane from swelling and to increase the vaporization rate of the solvents. After the catalyst layers are sprayed on each side, the MEAs are pressed in a heated press for several minutes.

In the case platinized carbon is used as the anode catalyst and platinum black is used as the cathode material, a different approach was used to improve the dispersion of the Pt black in the catalyst ink (this is due to the high density of the Pt black). In this case the catalyst material is sprayed on to a PTFE decal that will latter be pressed on to the membrane. The catalyst inks were prepared by ultrasonically blending a mixture of platinum catalyst, Nafion dispersion in the tetrabutyl ammonium form, glycerol, water and methanol in an ultrasonic mixer for 3 hours. After spraying thin layers of the catalyst ink on the decal, the ink is allowed to dry in an oven. Once the spray process is completed, the catalyst layer on the decal is hot pressed on to the membrane for several minutes.

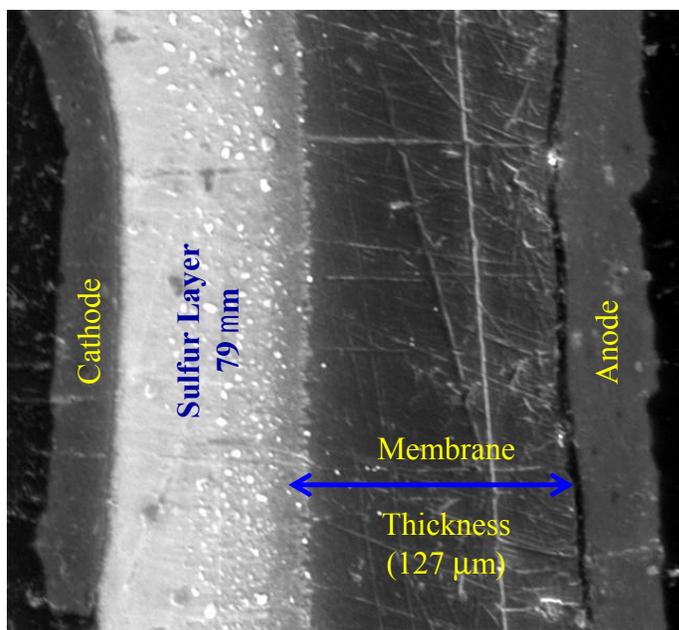
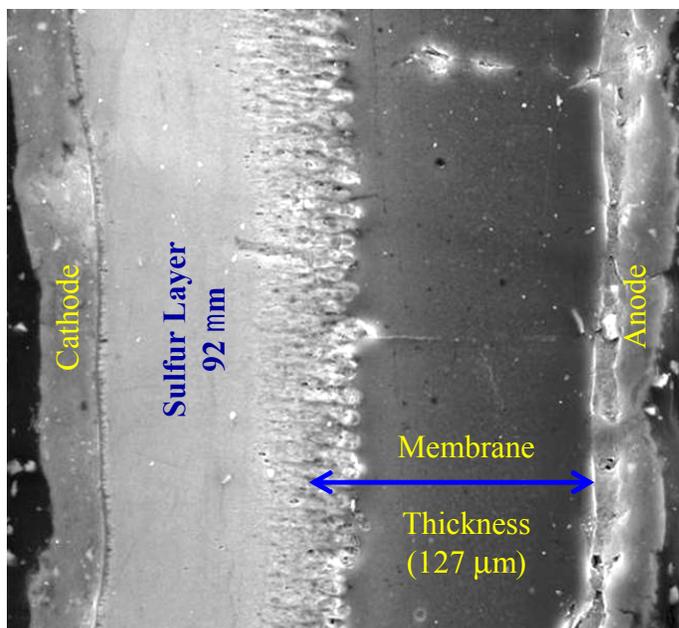
### **2.3.3 SEM Sample preparation**

Tested MEAs were post-test analyzed using scanning electrode microscope (SEM) with energy dispersive X-ray spectroscopy (EDX) to view the cross-section of the MEA and obtained an elemental analysis of the different layers. To prepare the sample for analysis a part of the MEA is dried and is embedded on epoxy. After the epoxy cures, the MEA sample is exposed by polishing to a mirror finish. Before the SEM is used, the sample is coated with a thin layer of gold to make the surface conductive.

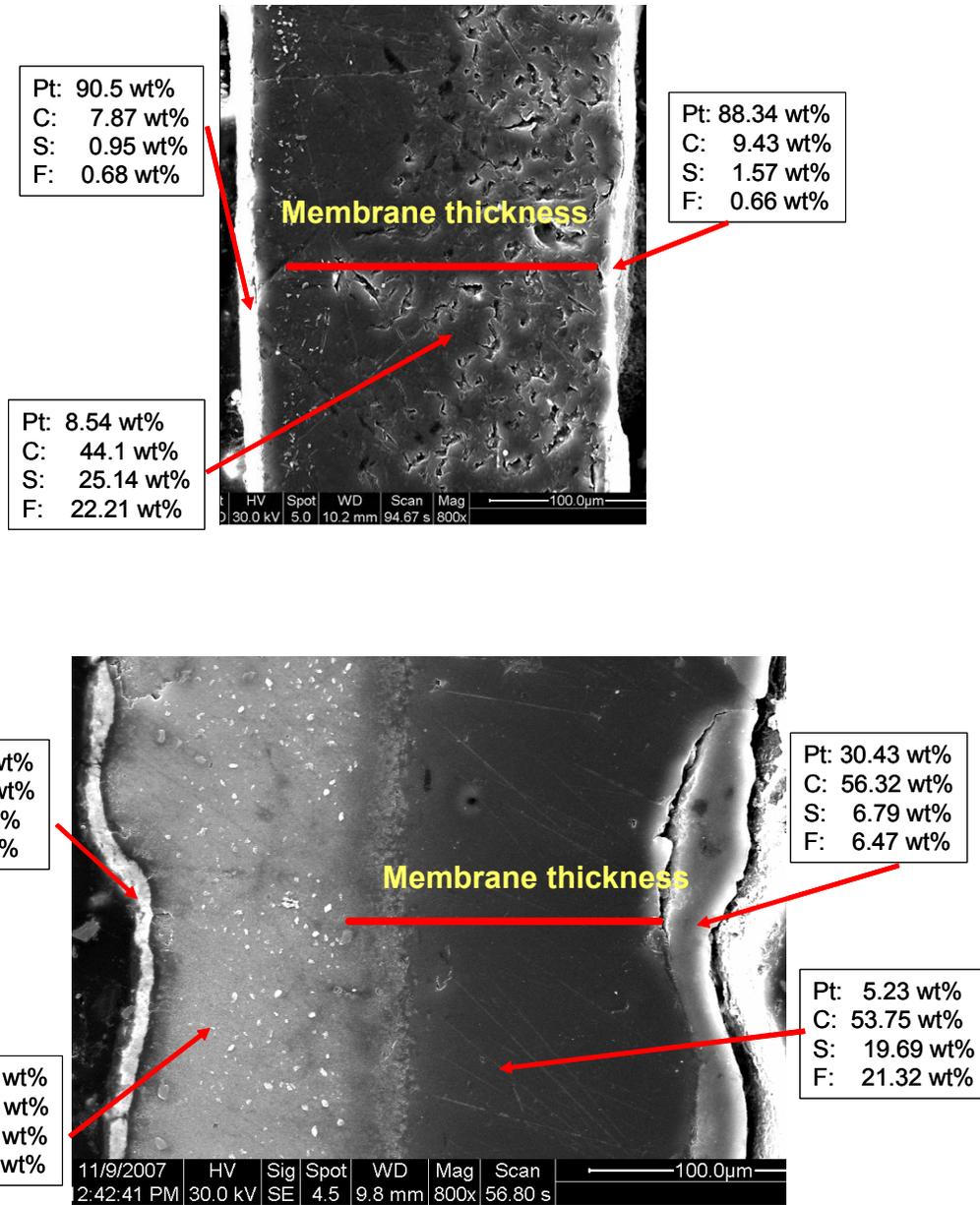
### **2.3.4 MEA Characterization: Post-Mortem Studies**

Four MEAs were analyzed after being tested in order to study the sulfur layer growth after several hours of operation. The first two MEAs were prepared almost identical by depositing the catalyst layer directly on Nafion® 115. Figure 2-5 shows the cross-section SEM micrograph of the MEAs after been tested at 80 °C and 4 atmospheres. From the post-test images of the MEAs, one can observe how the un-reacted sulfur dioxide that crosses from the anode is reduced to sulfur as soon as it encounters the cathode catalyst layer. Some of the reduced sulfur dioxide gets trapped in between the membrane and cathode catalyst layer where it accumulates and grows.

There is the possibility that the transported SO<sub>2</sub> is reduced by the carbon support at the cathode. This supposition comes from the fact that when the catalyst used is platinum black, the sulfur rich layer is not observed, however when the elemental analysis of the catalyst layer shows the presence of high carbon content, the sulfur rich layer is observed. Figure 2-6 (top image) shows the absence of the sulfur rich layer from an MEA prepared by Giner Electrochemical Systems using platinum black at both the cathode and the anode. From the elemental analysis we can observe that the carbon content is less than 10 wt%. Using this result to test our supposition, MEA 20 was prepared. On this MEA the cathode catalyst layer consisted of Pt black and the anode catalyst layer consisted of Pt/C. However, even thou Pt black was used at the cathode, the sulfur rich layer can be observed, as seen in Figure 2-6 (bottom image). From the elemental analysis one can observe that the carbon content is high compared to MEA 9, providing a possible explanation as to why we observe the sulfur deposition on MEA 20 but not MEA 9. The carbon in the catalyst layer is believed to be a residue from the MEA preparation due to the incomplete decomposition of glycerol in the catalyst ink. Further testing will be conducted with MEA's more closely following the MEA#9 approach (low carbon cathode) to better understand this phenomenon.



**Figure 2-5 Cross-section SEM micrograph of the MEAs after been tested for different times at 80 °C and 4 atmospheres: MEA 12- 105 hours (top) and MEA 13- 20 hours (bottom). Membrane for both MEAs is Nafion® 115 and Pt/C catalyst for anode and cathode.**



**Figure 2-6 Cross-section SEM micrograph of the MEAs after been tested at 80 °C and 4 atmospheres. MEA 9 (top) uses Nafion® 117 and Pt black on both sides. MEA 20 (bottom) uses Nafion® 115 and Pt black on the cathode and Pt/C catalyst for anode.**

## 2.4 DEVELOPMENT OF GAS DIFFUSION ELECTRODE

SRNL is working with Giner Electrochemical Systems LLC to develop an alternative electrolyzer design approach known as the gas-diffusion-electrode (GDE). The GDE could potentially provide a means to address the sulfur dioxide crossover issue by separating the SO<sub>2</sub> electrode from the PEM electrolyte. During this quarter, work was initiated on the building of a test stand for this task. The stand will provide the means to evaluate the electrochemical performance and SO<sub>2</sub> utilization of the cell under SO<sub>2</sub>-depolarized conditions. During the next quarter, the test stand construction will be completed, and Giner will implement a test program to evaluate the use of the GDE for the Hybrid Sulfur process. The system will then be compared and contrasted to the other electrolyzer technologies.

## 2.5 SINGLE CELL ELECTROLYZER TESTING

SRNL continued single cell electrolyzer testing during the First Quarter of FY08. Seven new MEA's were tested, with the primary objective of providing a better understanding of the design and operating conditions that effect the formation of a sulfur layer inside the cell MEA.

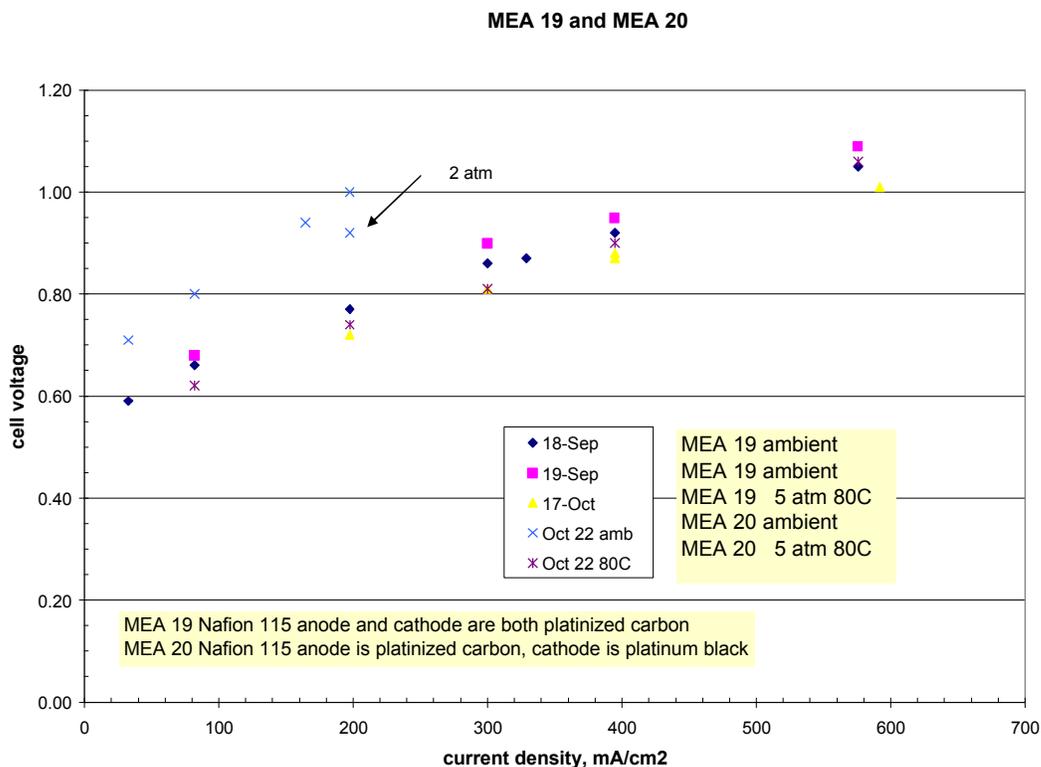
The test facility pumping problems experienced at the end of FY07 were addressed and satisfactorily solved. The pumping problems had limited testing time and had resulted in several pressure upsets, potentially damaging the MEA in the test electrolyzer. A new pump for feeding liquid SO<sub>2</sub> to the absorber was installed. Also, the gear pump used for anolyte feed supply was replaced with a similar pump that used alloy rather than Teflon gears. It was determined that 1-1/2 years of operation had worn the Teflon gears and significantly reduced the pump's performance. Operation of the alloy pump is much better and avoids many of the vapor-lock problems that were experienced with the Teflon pump.

The characteristics of the various MEA's tested this quarter are shown in Table 2-2. All tests used a carbon paper diffusion layer (7 mils thick) for the anode and a carbon cloth diffusion layer (12 mils thick) for the cathode.

**Table 2-2.** Characteristics of MEA's tested during FY08 First Quarter

MEA	Membrane	Thickness, mils	Anode Catalyst	Cathode Catalyst	Anode Pt, mg/cm <sup>2</sup>	Cathode Pt, mg/cm <sup>2</sup>	Active Area, cm <sup>2</sup>
19	Nafion 115	5	Pt-Carbon	Pt-Carbon	0.83	0.70	60.8
20	Nafion 115	5	Pt-Carbon	Pt-Black	0.782	2.67	60.8
21	Nafion 115	5	Pt-Carbon	Pt-Black	0.6	2.9	60
22	Pt-treated Nafion 117	7	Pt-Black	Pt-Black	1.0	1.0	61.2
23	Pt-treated Nafion 117	7	Pt-Carbon	Pt-Black	1.0	1.0	61.2
24	Pt-treated Nafion 117	7	Pt-Carbon	Pt-Black	1.0	1.0	50
25	Nafion 117	7	Pt-Black	Pt-Black	4.0	4.0	60

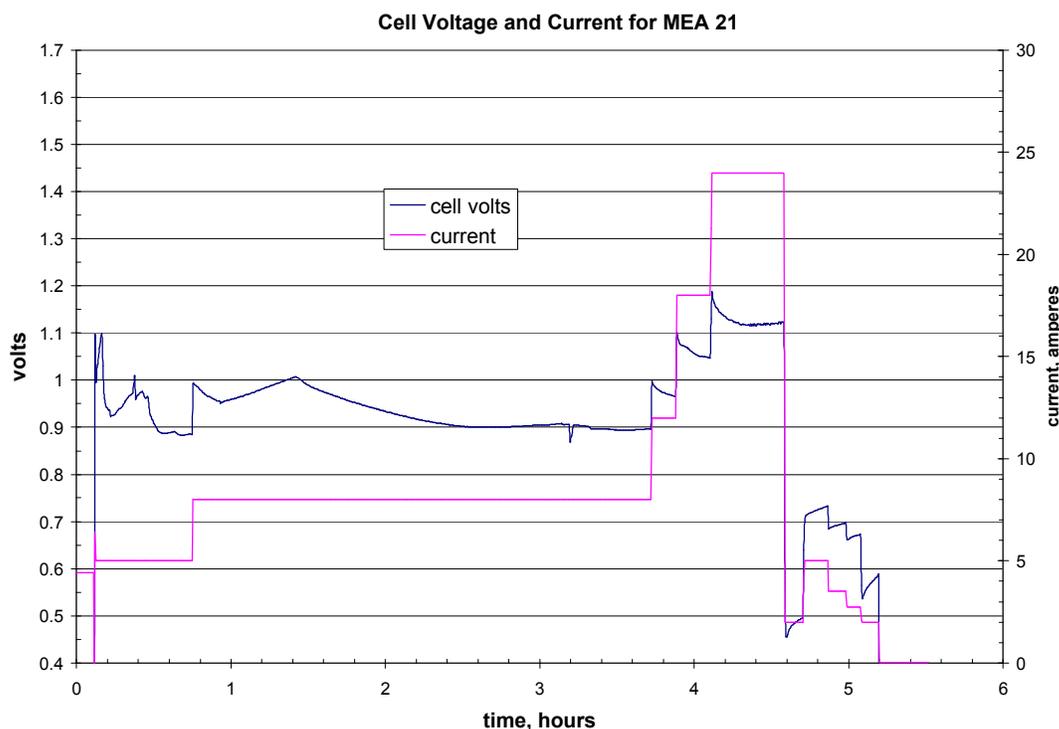
Polarization test results for MEA's #19 and #20 are shown in Figure 2-7. MEA #20 consisted of a Pt-carbon anode and a Pt-Black cathode, fabricated by SRNL. The performance of the two cells is similar, although slightly worse than some of the earlier MEA's tested. The major advantage of MEA #20 was felt to be the prevention of sulfur buildup due to the use of the Pt-black cathode. This was discussed in the previous section of the report.



**Figure 2-7.** Polarization data for MEA #19 and MEA #20.

MEA #21 was nominally identical to MEA #20, but the performance was considerably worse. Preparation of a Pt-Black cathode, as used in these two MEA's, is much more difficult and requires higher temperatures than preparation of Pt-Carbon electrodes. As a result, we continue to refine our fabrication methods to optimize the catalyze formation.

During MEA #21 testing, it was observed that the cell voltage responded much slower to changes in current than was the case with previous MEA's. This can be seen in Figure 2-8. For example, the transition at 4.2 hours required 12 minutes. With most previous MEA's, five minutes of transition was adequate. Also, there was a pronounced overshoot in voltage. These characteristics are likely the result of low porosity of the anode. Although MEA #21 had a Pt-carbon anode, it was subjected to different processing conditions during fabrication than previous Pt-carbon anodes due to the methods used for the Pt-black cathode preparation.

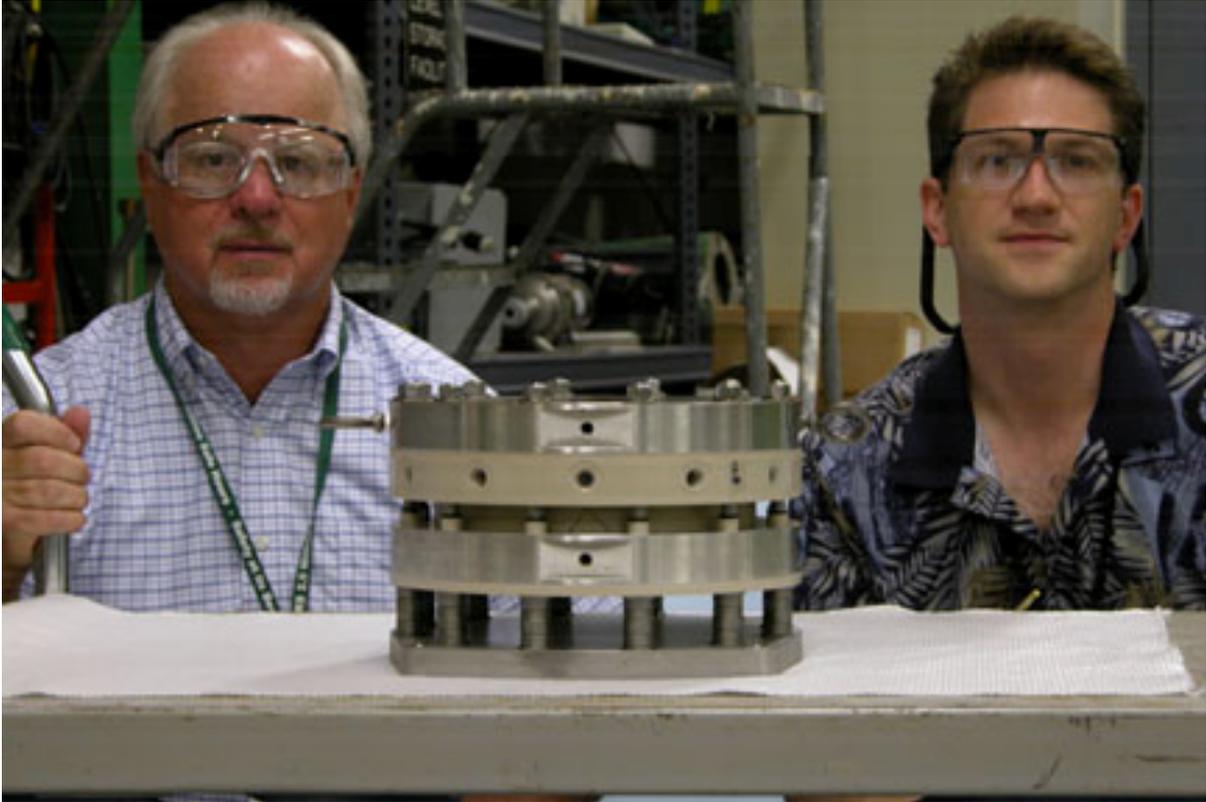


**Figure 2-8.** Transient response for MEA #21.

MEA's #22, #23, #24, and #25 were fabricated by Giner Electrochemical. They each used a Pt-black cathode (similar to MEA #9). MEA #22 and #25 also had a Pt-black anode, whereas #23 and #24 had Pt-carbon anodes. Pt-carbon anodes are expected to provide better voltage performance with the same catalyst loading than Pt-black anodes, due to a much higher catalyst surface area. MEA's #22, #23, and #24 used a Pt-treated membrane, whereas MEA #25 had an untreated Nafion 117 membrane. Testing of these latest MEA's was conducted near the end of the reporting period, and the results will be reported in the next Quarterly Report.

## 2.6 MULTI-CELL STACK TESTING

The multi-cell stack construction was completed at the end of FY07. A photograph of the finished electrolyzer is shown in Figure 2-9. It consists of three cells with an active area of 160 cm<sup>2</sup> each. The rated hydrogen output is 100 liters per hour. The MEA consists of Pt-Carbon anode, Nafion 117 electrolyte, and Pt-Carbon cathode. The construction is a modification of standard Giner water electrolyzer designs, although all wetted parts have been changed to carbon or polymer materials. The multi-cell stack will be installed in the SRNL electrolyzer test facility and characterization testing will be conducted next quarter.



**Figure 2-9.** Multi-cell Stack Electrolyzer with 100 lph rated hydrogen capacity

During this reporting period, work was initiated on designing the facility changes and mounting fixtures needed to install the larger multi-cell stack into the single cell electrolyzer test facility. The mounting fixture for the multicell stack has been designed and is currently under fabrication. The mounting fixture will be used to install the multicell stack in the test facility where the single cell electrolyzer is currently located. Due to the size differential of the 3 cell electrolyzer, several of the single cell system components will have to be adapted for the 3 cell stack. Current breaks for both the anolyte and catholyte will consist of long lengths of tubing to prevent short-circuiting across the cells. Each line will be a minimum of 8 feet in length. The length was determined by the resistance of the process fluid. The length of tubing calculated will provide enough electrical resistance to prevent a measurable amount of current flow and therefore effectively isolating one cell from another.

The 3 cell stack will be oriented vertically to allow for easy access to the plumbing connections. The existing support structure is currently being evaluated to assure that it will support the additional weight of the three cell stack. The mounting frame will attach to the existing structure of the single cell test facility with any reinforcements necessary.

A flow manifold has been designed and fabricated to allow for the distribution of flow to the three individual cells from a single pump. This distribution system had to be designed since no commercial parts are available in the required size that would withstand the operating conditions of the stack.

The pressure protection requirements for the electrolyzer system are being reviewed. Initially, the 3 cell stack will be operated at the same conditions as the single cell; a maximum pressure of 80 psi and a maximum temperature of 90 C. The active area will increase from 60 cm<sup>2</sup> for the current electrolyzer to 160 cm<sup>2</sup> per cell (a total of 480 cm<sup>2</sup>) for the multi-cell stack. This requires an 8-fold increase in current to operate the new stack with the same current density as the single cell electrolyzer. A new power supply has been installed. The increase in hydrogen production could potentially alter the relief settings required for the electrolyzer system. The addition of the 8' current breaks will also affect the setting of the pressure relief device. It is anticipated that a single pressure relief device will be used down stream of the current breaks. This will require the additional pressure drop of the tubing to be accounted for in the relief device calculations.

## **2.7 UPGRADE TEST FACILITY TO 90-100 C AND >10 ATM**

No work was scheduled during this reporting period.

## **2.8 PROCESS FLOWSHEET OPTIMIZATION**

No work was scheduled during this reporting period.

## **2.9 ILS PLANNING AND DESIGN**

No work was scheduled during this reporting period.

### **3.0 PROJECT MANAGEMENT**

SRNL has issued the project work packages for the FY 2008 work plan which were approved by the program management. A resource-loaded schedule has been prepared and is being used to manage the project.

As previously discussed with the DOE-NE Program Management Team, several of SRNL's tasks have been slightly delayed due to funding issues brought about by the continuing resolution. At the start of the FY2008, SRNL was granted incremental funds; however, funding was not posted in the financial plan until November 2008. SRNL has experienced delays in the project work which has resulted in schedule and deliverable adjustments. However, unless further significant delays in funding or funding reductions occur, SRNL expects to successfully complete all work tasks contained in the NHI Work Package.

SRNL personnel participated in the DOE Nuclear Hydrogen Initiative Semi-Annual Meeting which was held in Idaho Falls, Idaho, Oct. 23-25. At the request of the NHI Program Manager, SRNL also attended the Solar Thermochemical Hydrogen Program workshop in Boulder, CO on November 15, 2008.

SRNL presented papers on the Hybrid Sulfur project at the Global 2007 nuclear conference in Boise, ID in September and the AIChE Annual Conference in Salt Lake City in November, 2007.

### **4.0 FUTURE WORK**

Work will continue on refining and improving the SO<sub>2</sub>-depolarized electrolyzer design. The Component Development task will be expanded to include construction and operation of a small-scale electrochemical cell. This will permit more rapid characterization of cell components under actual electrochemical operating conditions.

Testing of single cell electrolyzer will be temporarily halted after completion of MEA #25 testing. This is necessary in order to modify the test facility for larger scale testing of the multi-cell stack. The new stack will be installed in the facility and tested over a range of operating conditions. A Level 1 Milestone for the Hybrid Sulfur development program is the completion of Multi-cell Stack Testing, which is scheduled for March 15, 2008. Work toward this milestone is on schedule.

Work on the Commercial Electrolyzer conceptual design task and the Westinghouse TCA flowsheet task will be completed next quarter. Work will be initiated on the ILS planning task and the upgrade of the test facility for higher temperature and pressure operation.