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Abstract

The following report documents the technical approach and conclusions made by Acumentrics Corporation during latest budget period toward the development of a low cost 10kW tubular SOFC power system. The present program, guided under direction from the National Energy Technology Laboratory of the US DOE, is a nine-year cost shared Cooperative Agreement totaling close to \$74M funded both by the US DOE as well as Acumentrics Corporation and its partners. The latest budget period ran from January of 2005 through June 2005. Work focused on cell technology enhancements as well as BOP and power electronics improvements and overall system design. Significant progress was made in increasing cell power enhancements as well as decreasing material cost in a drive to meet the SECA cost targets. The following report documents these accomplishments in detail as well as the layout plans for further progress in next budget period.

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INTRODUCTION

Acumentrics Corporation has focused during the latest six-month budget period of the SECA program on the design and manufacture of micro-tubular SOFC power systems approaching twice the power density now achieved from state of the art anode supported tubular designs. By developing a common stack design with high power density cells capable of meeting the SECA cost targets, a number of markets will be opened to this technology. Present markets being focused upon include telecommunication, remote residential, and military markets. Operation on fuels including natural gas and propane will be developed for the telecommunication and remote residential markets. These fuels will be developed and demonstrated during Phase I of the program. Operation on liquid fuels, including diesel and JP-8, will be developed for the military markets. These fuels will be developed and demonstrated during Phases II and III of the program.

The overall goals of Phase I of the program, which represents three years of development, include:

1. Design of a common low cost generator to meet all chosen markets.
2. Development of an anode supported micro-tubular cell capable of twice the power density presently achieved.
3. Prototype testing of a natural gas or propane fueled unit meeting and exceeding SECA goals.

The research and development to achieve the above goals can be listed in five major sub-tasks:

1. **System development and integration** – In this task work is focused on the functionality and cost reduction of major BOP components. Thermalhydraulic components are being developed and tested as well as the necessary control strategies. Power electronics and control hardware is being refined and cost reduced to meet the goals of the program. Work is also concentrating on thermal recovery and burner designs for stationary and mobile applications.
2. **Cell Technology Development** – In this task work is focusing on improvements in cell power density through material changes and refinements. Composition and morphology of the anode, electrolyte, and cathode are all being addressed to increase cell power.
3. **Stack Technology Development** – In this task work is focused on generator design and assembly to reduce cost and improve reliability. Connections to the anode and cathode for current collection are being optimized. Casting of insulation to net shape or near net shape is also a focal point for cost reduction.

EXECUTIVE SUMMARY

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4. **Fabrication and Processing Technology Development** – Work is concentrating around cell manufacturing processes and the interaction between processes. Assurance of the reliability of each major process without the compromise of cell power is critical in this task.

During this past budget period, a number of accomplishments were made. They include:

- **Isostatic Anode Tube Manufacture:** tubes were demonstrated to be manufactured through isostatic pressing rather than the existing extrusion process. This allows for a number of potential improvements such as inclusion of a closed end, grading of the anode material, and lower cost manufacturing through a reduction in steps. Further work to evolve this technology will be performed in the next period.
- **Completion of Phase One Independent Cost Validation:** During the budget period, a team of auditors independent of DOE assessed the viability of the Acumentrics technology achieving the cost goals which exist for phase I. Namely, can the Acumentrics unit achieve less than \$800/kW cost at 50,000 units per year production. After months of review and data collection, that team concluded that the Acumentrics design should be in the \$560-\$760/kW range depending upon the operating point.
- **Testing of a Ceramic Recuperator:** The Acumentrics team also performed validation testing on two different types of ceramic recuperators during the budget period. By the end of the period, the ceramic recuperator was found to be thermally stable over long periods of operation and through many thermal cycles. The challenge remains to increase the effectiveness from the 70% level up to the 85% level found in metallic designs.
- **Complete Cell Computer Model:** During the past period, an electrochemical model of the existing tubular cell was completed. This model was then accurately validated against empirical cell test data. The model is now being used to predict impacts of diameter changes and multiple current collection points.
- **Improved Cell Power through Chromite Interconnections:** During the last period, the processing issues with regard to deposition of a conductive ceramic layer for dual atmosphere used were resolved. A number of electrical tests were then performed which demonstrated the expected power gain over a tube with a single end point take-off. During the next reporting period, further data will be compiled.
- **Stable Silver/Chromite Connections:** In order to utilize a chromite connection in dual atmosphere, a low resistance joint must be made which is stable in air. During the past reporting period, a connection was tested for over 1200 hours with low resistance. This test will continue into the next reporting period.
- **Delivery of a 5kW Demonstration to NETL:** During the past reporting period, a 5kW SOFC CHP demonstration unit was delivered to NETL. This was the first unit delivered by any SECA team and will be utilized to shakedown the Phase I test rig at DOE.

- **Demonstrated Microwave Drying:** During the latest reporting period, the ability to microwave dry and debind the tube was demonstrated. This has the capability to reduce the overall processing time from greater than 48 hours to well under 6 hours.

Task 1.0 - System Development and Integration

Subtask 1.1 – Prototype Stationary System Design

Generator performance calculations were made using the latest cell performance data. A 10 kW, 432 cell, 72 manifold geometry was evaluated. State points for both maximum power and design efficiency operating conditions were determined and are summarized below.

Figure 1: Design efficiency – 35% Net DC

Design Power - 10 kW

Cell Voltage
No of Cells
Fuel Utilization
K Factor
Heat Loss (w/m^2)
Power
BOP Power

Current
Gen Eff AC
O/C
Air Stoichs
Heat Exchanger Eff

DC Power
Gen Eff DC
Net DC Eff
Heat Loss

Slope
En
Cell Temp

Cell Area (cm^2)
Gen Eo
Gen V

Anode Fuel (lpm)
Cathode Air (lpm)
Cooling Air (lpm)

Figure 2: Design Power – 10kW

Additional safety features have been brought into operation in the gas generator test area. These include a pressure sensor to detect low-flow in the exhaust hoods Exhibit 1, a combustible gas detector, a control relay, and a natural gas shut-off valve Exhibit 2. Low exhaust flow or gas leaks will operate the gas shut-off valve. The control relay also signals to selected test stands the in the event of low exhaust flow to initiate shutdown of those tests running on bottled gas such as propane.

Exhibit 1: Pressure sensor to detect low-flow in the exhaust hoods

Exhibit 2: Combustible gas detector, a control relay, and a natural gas shut-off valve

Generator performance calculations were made for a generator utilizing 432 – 22 mm diameter, 28 cm long cells. A range of cell power densities was evaluated as shown in Figure 3. The power densities represent a single ended current collection cell, a double-ended current collection cell and an enhanced performance double-ended current collection cell.

Figure 3: Cell Power Densities

Subtask 1.3 - Control Strategy Development

Work is proceeding to standardize software across the various bundle, stack, generator, and simulator test platforms. The new state sequencers, control algorithms, and communications modules are being upgraded to those used in the current 5kW generator product.

Non-linearity in the A/D conversion process has been identified and improved for cell voltage and thermocouple measurements. Precise voltage measurement is necessary when using cell voltage as an indicator of fuel utilization. Terminating long analog printed circuit traces to the A/D on-chip made the improvement multiplexor with 100-Ohm resistors. The on-chip multiplexor injects charge into these lines when it switches.

The HMI interface has been improved and simplified reducing need to update HMI configurations.

A new circuit board has been designed and laid out to allow for direct reading of all the cell voltages in a generator. The design uses opto-FET ICs to multiplex directly into the present controller voltage sense inputs. Up to 90 differential voltages may be instrumented when 3 circuit boards are utilized. The layout is shown in Figure 4.

Figure 4: Circuit Board Design

The first of the distributed control printed circuit designs has been received and is in debug and is shown in Exhibit 3. This design accepts 16 thermocouple inputs and communicates with other control elements via the CAN bus.

Figure 3: Distributed control printed circuit design

New software has been implemented for the HMI that allows for remote control and monitoring over a network or the Internet using a second computer. Both local and remote computers may interact with the fuel cell simultaneously.

A low battery voltage disconnect feature has been added to the fuel cell operating firmware. Previously this feature was present in the bundle and stack testers only. A voltage input trip coil is used to trip the 200ADC battery circuit breaker if the 48V battery voltage falls below a safe level.

An additional thermocouple has been added to compensate for battery changes over temperature when in the grid-connect mode. In this mode, the inverters load the battery until a temperature compensated float voltage is reached. This must be coordinated with the fuel cell interface converter (FCIC) that controls battery float voltage in the stand-alone mode.

A feature to automatically reconnect inverters in the stand-alone mode following a load fault has been implemented. The current algorithm calls for a reconnect attempt every 2 minutes.

New hardware and firmware has been developed that monitors and reacts to fuse faults in the BOP power distribution circuit. Before, only indirect effects could be judged to determine a blown fuse. Now, each fuse is directly monitored. These additional faults have also been added to the HMI.

The algorithm that controls the electronics area cooling fans has been improved. To prevent condensation during startup the fans will not draw in outside air until the inside of the cabinet has warmed to 50C. Also, the hysteresis in the on/off control has been increased to reduce the number of fan cycles.

Currently the control system is able to communicate over the Internet via an Ethernet network connection. This feature can be used to remotely monitor and control the fuel cell power plant. For remote locations without network access, a satellite Internet capability is being designed in. A DirectTV dish antenna, RF transceiver, and modem will be used to interface with the satellite. To handle the wide temperature range expected at remote locations, an imbedded PC with FLASH memory drive is being used. The PC will obtain power from the 24VDC BOP supply and interface directly with the fuel cell controller and the satellite modem. It will run a subset of Microsoft WindowsXP. The PC has been obtained and is now being configured.

The Acumentrics designed fuel cell control printed circuit assembly (P/N 51-0133) is being upgraded for higher performance and lower cost. A summary of the new features and comparison to the present controller is shown in Table 1.

<i>Controller Variations</i>		
Function	Present F2407	New F2810
ANALOG IN	4 x 0-5V 4 x 0-20mA 1 x 100mV, 1 x 300mV	4 x 0-5V 4 x 0-20mA 2 x 100mV 2 x BOUT readback (100mV)
ANALOG OUT	6 x 0-10V, 4 x 24V PWM	6 x 0-10V or 6 x 24V PWM 2 x 0-24V, 1A (Maxitrol)
BINARY IN	10 x 24V 6 x 5V (no protect)	5 x 24V, 10 x 24V blown fuse 8 x 5V (no protect)
BINARY OUT	6 x 24V 2 x relay form c	16 x 24V (2 w/ current readback) 4 x relay form c
THERMOCOUPLES	16	32
VSENSE	16	64
COMM	1 x iso RS232 1 x CAN	1 x RS232 (to inverter) 1 x RS232 or DIGI 1 x CAN
POWER CIRCUITS		1 x 12V isolated for TSI 10 x 24V fuse

Table 1: New Controller Requirements

The new controller will have additional channels of thermocouple and cell voltage meter inputs to support larger stacks. These circuits are being redesigned to make them smaller, lower cost, and higher performance. It is expected that each thermocouple channel will drop in cost from around \$2 per channel (FCTC design) to around \$0.34 per channel. The cell voltage sense channels are expected to come in at \$0.50 per node. A Digi brand Ethernet adapter will be integrated onto the controller replacing the current Lantronix chassis mount adapter.

In addition to new controller design work, the existing controller design has been modified to be less expensive to manufacture and more convenient to work with. The circuit board layout has been revised to eliminate all rework. The thermistor that

measures PCB temperature has been relocated at the thermocouple connector to improve cold junction compensation. The linearity of the cell voltage input channels has been improved by changing the source impedance of the analog-to-digital converter (ADC) buffer circuits. The emulator connector has been relocated to allow access by the emulator pod from the side of the controller instead of from the top only. The CAN bus terminator has been made configurable to improve signal integrity when more than 2 CAN bus devices are present.

Blower speed measurements can now be completed by using a new software based tachometer algorithm. This software approach frees up DSP hardware counters for other uses (such as pulse-width-modulation) and expands the number of tachometer channels to include all the binary inputs.

Subtask 1.4 - Heat Recovery

Testing of a second ceramic recuperator was conducted during the budget period. Leakage measurements were made on the air to exhaust side on Casting #2. Figure 5 shows the leakage data with and without the plenum covers installed. The “with ends blocked” data is the leakage of the ceramic core itself while the “with plenums” data is with the plenum covers installed and the exhaust outlet chute taped shut. As shown in the graph, there was considerable less leakage for Casting #2 compared to Casting #1. It is important to note however that the Casting #1 data is after the unit was tested several times and therefore had been up to operating temperature with several thermal cycles.

Figure 5: Leakage data with and without the plenum covers installed

Casting #2 recuperator was then installed in the heat exchanger test stand and two long duration tests were conducted. Figures 6 and 7 summarize the results of the first test. The first graph gives the temperatures for the entire 24 hour test period and the second

graph gives the temperatures and effectiveness during the period of steady state operation and changes in test conditions as noted. At the design flow rate of approximately 1000 lpm the unit achieved an effectiveness of 68%. At higher flow rates the effectiveness dropped slightly. The effectiveness also dropped slightly when a portion of the exhaust passages was blocked. Placing an insulation board on top of the unit next to the air inlet plenum caused this occurrence.

Figure 6: Temperature Data for Recuperator #2

Figure 7: Temperatures & effectiveness during steady state operation

In the second test, the system was operated over a 50-hour period with similar results. The effectiveness was increased to 72% with the placement of perforated tiles on the top of the unit. It is believed that these tiles improve the performance by reducing the radiation loss from below the recuperator. Figure 8 shows the test conditions and calculated effectiveness.

Figure 8: Test conditions and calculated effectiveness (1)

Figure 9: Test conditions and calculated effectiveness (2)

After the second test, the unit was removed and leakage tests were conducted. Figure 10 shows the leakage data for the core before and after testing. Also shown on the graph is the core leakage data for Casting #1 after it had been tested. As seen in the graph, the leakage of Casting #2 after testing was over 300% greater than before testing and is greater than Casting #1. Hairline cracks were detected on the exhaust inlet side of the core, which may explain the increased leakage. Photos of the cracks are shown in Exhibit 4; they were located on only one side of the core, roughly at the centerline of the unit.

Figure 10: Leakage data for the core before and after testing

Exhibit 4: Hairline cracks were detected on the exhaust inlet side of the core

Blasch has cast a third casting. This casting was made using Blasch's BP 67/8-SC an oxide bonded, silicon carbide. The material is nominally 67.5% SiC, 29.5 Alumina and 3.5 Silica and has excellent shock resistance and approximately double the thermal conductivity of the alumina. A photograph of the unit is shown in Exhibit 5 below. A leakage test was performed on casting #3 and as shown in Figure 11, the leakage for this unit was slightly greater than the as-received casting #2 and significantly better then casting 1 & 2 after thermal cycling. The plenums were installed on the casting and the unit placed in the recuperator test stand. The plenum retaining frames were machined to a slightly larger opening to fit over the flanges of the SiC casting which had more surface imperfections than the alumina castings.

Exhibit 5: Casting #3 received from Blasch Precision Ceramics

Figure 11: Results of leakage test were performed on casting #3

Two performance tests were made with the unit and the results are presented in the graphs in Figure 12 and Figure 13. In the first test the unit was operated at the design flow rate. An effectiveness of 0.65 was achieved with the exhaust face open to atmosphere and an effectiveness of 0.68 was measured with perforated tiles placed on top of the unit. The effectiveness gain is due primarily to an increase in temperature uniformity of the preheated air leaving the recuperator. This test was terminated prematurely due to a power failure and during shutdown the recuperator air and exhaust flows were stopped abruptly. In a normal shutdown the cathode fan is left on resulting in continued heat transfer by the recuperator as the entire system cools. There were no obvious signs of problems resulting from the “stagnant” shutdown.

Figure 12: First performance test

A second test was performed to confirm the earlier results and to obtain performance data above and below the design flow rates. Figure 13 shows the test conditions. The test was started with the tiles at the exit of the recuperator and removed once a baseline reading was obtained. Once again an effectiveness of 0.68 was measured indicating there was no change in the recuperator after the initial thermal cycle. The tiles were removed and the system operated at airflow rates of 830, 1040 and 1450 lpm. During the operation at 1450 lpm, the inlet temperature to the recuperator was increased by 30°C. As shown in the graph the recuperator performance is relatively insensitive to changes in load for the range of flows investigated.

Figure 13: Second performance test

Thermal cycle testing of casting #3 enabled an evaluation of the ability of the SiC material to handle startup, shutdown and steady state temperature gradients. The test procedure began with a morning start of the recuperator test stand and ran in a steady state mode for several hours and shutdown at the end of the day for cooling overnight. Figure 14 shows the life graph for the testing.

Figure 14: Life Graph

A between test check of the inlet pressure to the recuperator at two flow rates verified that there was no cracking of the ceramic monolith. There was no significant change in the inlet pressure during the 14 thermal cycles. After the 14th thermal cycle the recuperator was removed from the test stand and inspected. Again, there were no signs of cracking.

Figure 15 compares the core leakage rate before and after the testing. The graph shows the leakage rate of casting #2 after three thermal cycles and evidence that there was no change in the leakage rate as a result of the thermal cycles. Also of importance, is the leakage rate with the covers installed being equal to that of the core itself indicating that the covers remained sealed to the monolith even after 14 thermal cycles. Inspection of the cover gaskets revealed that they were compressed tightly between the covers and casting as evidenced by a compression outline on the gaskets.

Figure 15: Core Leakage Rates

Exhibits 6 and 7 show the top and bottom surfaces of the casting after the thermal cycle testing; there are no signs of cracks as had developed on the alumina castings, (casting #1 and #2).

Exhibit 6: Exhaust Outlet Surface

Exhibit 7: Exhaust Inlet Surface

Metallic, Counter-flow Recuperator

A metallic, counter-flow, recuperator using the Toyo Flexible Fin Core (FFTex) finned rectangular tubing geometry was also tested. The test stand was modified to direct the exhaust flow towards the downcomer end of the stack chamber and into the inlet of the recuperator. The recuperator was modified so that exhaust gas entered only one side of

the unit and likewise the exhaust outlet was modified so that the exhaust gas exited the unit from only one side. A sketch of the test setup is shown below Figure 16 along with a photo of the assembly in Exhibit 8. The top insulation, consisting of 2.5 cm ceramic fiber blanket and 7.6 cm of mineral wool is removed in the photo. The overall dimensions of the unit are 16" wide, 25" long and 4" high which is considerably smaller than the metallic and ceramic 3 pass, cross flow units.

Figure 16: Sketch of the test setup

Exhibit 8: Photo of the assembly

The sketch in Figure 16 shows the thermocouple locations. Except for the thermocouples at the exhaust gas inlet to the recuperator, the thermocouple location remained the same. Four thermocouples measured the gas temperature exiting the lower plenum and two were located above the radiation shield. There was a single thermocouple approximately on centerline at the exhaust inlet to the recuperator.

In the first test, 3/22/05, the exhaust from the recuperator was open to atmosphere. Due to the relatively high-pressure drop across the exhaust side of the recuperator there was a loss of exhaust gas from the stack chamber resulting in low recuperator effectiveness. Although care was taken to seal all insulation joints on the stack chamber, the system was not designed to operate at elevated pressures, and therefore, there was a fair amount of gas leaking out of the insulation package. The pressure data suggests that 30% of the exhaust gas may have been leaking from the system.

A second test, 3/28/05, was performed, this time with a modification to the test setup to reduce the backpressure at the exhaust inlet side of the recuperator. An exhaust tube attached to the recuperator exhaust outlet was directly connected to the test facility exhaust system, which operates at approximately $-0.5''$ w.c. With the reduce stack chamber pressure there is a reduction in the exhaust short-circuiting the recuperator and an improvement in the recuperator performance. The Figure 17 shows the temperatures during the course of the 3/28/05 test.

Figure 17: Temperatures during the course of the 3/28/05 test.

Subtask 1.6-Gas Utilities

Valve Development

Through extended valve testing, a number of issues have arisen related to fuel flow stability. Stiction has been observed with the Burkert 6022, and more recently, 6023 proportional valves. Over the three years these valves have been in service, a number of issues have arisen. In particular, valve stiction, occurs when the valve is operating at set flow conditions for extended periods. This stiction results in erratic jumps in fuel flow rate, which temporarily destabilizes system operation. The exact period required before the valves begin to exhibit these problems is difficult to determine, an approximation would be within 500-1000 hours depending on the testing schedule/operating modes.

An illustration of the valve assembly is given in Figure 1.6.5. A new, used and the failing valve were disassembled. The used and failing valve plungers, when compared to a new plunger were worn (Figure 1.6.6). Wear grooves had formed circumferentially on these plungers, with the failing valve appearing to exhibit the most wear. Failing valve pertains to valve exhibiting frequent stiction characteristics. In contrast to the 6022, the 6023 valves contain two slip rings (Figure 1.6.7). The design of the 6023 does not permit the same wear as that observed with the 6022. However, problems have been experienced with the 6023, which were fixed temporarily (~4 days) by applying a dry lubrication (Accupro /Teflon® Dry Lube) to the plunger and inside the armature guide tube.

Burkert's response was that valve stiction was common with the 6022 and 6033 under the operating conditions described above. Two alternatives were offered, the 2833 (frictionless design) and the 6024. Exact specifications, pricing and availability are being reviewed.

Figure 18: 6022 Valve Schematic

Exhibit 9: Three valve plungers compared

Exhibit 10: Disassembled 6023 valve

Testing to evaluate the M420 Modulator Valve for fuel control in generators and test equipment continued. A bench test was set up to gauge M420 valve performance and various controller options. A blower (25-0028) was used to simulate fuel pressure. The test set-up is shown below in figure 1.6.8.

Figure 19: Bench test to gauge M420 valve performance and various controller options

The Bench top testing was undertaken to compare the valve performance when directly powered by a bench top variable power supply and indirectly powered by sending a 4-20mA signal to a Maxitrol A200 signal conditioner (Exhibit 11 [a]) or 0-10V signal to the low cost valve controller (Exhibit 11 [b]).

Exhibit 11: Maxitrol A200 signal conditioner and 0-10V signal to the low cost valve controller

Figure 20 illustrates the valve performance with each control method.

Figure 20: Valve performance with each control method.

System testing was next undertaken on Stack Tester II, using the low cost valve controller to control the M420. Pressure gauges were installed after the M420, and at the outlet of fuel air mix line. The fuel passes through the sulfur remover before mixing with the air (not hot during test). A manual valve was installed on the outlet to test the M420 under conditions of increasing backpressure, essentially simulating a stack.

1. The M420 will pass a maximum of 47 Lpm of natural gas, when the anode air system is off. Maximum flow was limited by the valve controller setting of ~ 20V in Figure 21.

M420 Fuel Flow Rate Versus Pressure at Outlet (P2)

Figure 21: M420 fuel flow rate vs. pressure at outlet

2. The minimum flow setting was determined to be less than 1 Lpm. Problems with system noise made it difficult to determine accurately the lower flow limits of the valve. Initial impressions are the valve will reliably control down to 0.5 Lpm and achieve a shut off < 0.25 Lpm.
3. The air:fuel was set to 3:1, the maximum fuel flow was 30 Lpm, and 90 Lpm air at 7" w.c. (in Figure 22). The fuel is limited by the outlet pressure of 7" w.c., the maximum setting for the M420. The fuel was constrained by the 1:3 ratio (blower set point).

Figure 22: M420 fuel flow rate vs. pressure at outlet

4. The valve was subjected to increasing backpressure applied using the manual valve at the outlet (P3). The blower maintained a consistent airflow of 75 Lpm. The backpressure at the valve outlet (P2) increased to 8.2 Lpm.

Figure 23: M420 fuel flow rate vs. pressure at outlet

Increasing the spring constant of the plunger spring can increase the outlet pressure of the valve. This action may affect shut off, and will need further testing. Alternatively, the M420H is capable of a total modulation span of 10\"w.c. However, “the sensitivity of outlet pressure change relative to voltage change could cause hunting to occur”.

The following conclusions and recommendations were made:

- Outlet pressure from the M420 is regulated to a maximum of 7" w.c., reducing the maximum fuel flow rate.
- The valve response is consistent when applying 0-20 VDC directly using a bench top power supply, or using a signal conditioner/ custom valve controller (Figure 2)
- Maximum power consumption recorded was 7.2 W
- The in-house low cost valve controller operated the valve satisfactorily, but will require modifications before practical application
- The A200 signal conditioner regulates the valve voltage to ~ 18 V
- Minimum achievable flow rate was less than 1 Lpm (limited by test setup)
- Data collection was limited by flow meter used on the bench test (1000 Lpm) and system noise (electrical) when testing in Stack tester II (0.5-1 Lpm).
- Order M420H (higher outlet pressure setting) for testing.
- Begin testing of the M420 & 6022/3 at a set flow condition.
- Determine requirements for integration of M420 (or similar valve) into GUM.
- Continue development of low cost valve controller.

TSI Flow Meter Study (I)

The integration of the TSI flow meters into test stands and generators have consistently suffered from two problems, power issues with flow meter fuses being blown and reliability of the flow meter accuracy/calibration. The latter has only become more obvious over the past year, with flow meters on various test stands being calibrated using a gas totalizer, displaying the gradual decay of signal feedback with flow rate. Work was undertaken to determine the effect that a clogged screen would have on the calibration of a TSI flow meter, and to determine if the current filters are adequate.

The TSI flow meter contains an exposed thermal flow sensor. The flow meter is designed to strain air uniformly over the sensing element. Mesh screens are located at the flow meter inlet and outlet to further help strain the flow and protect the sensor. These screens were filled with particulates, which reduce the flow meter accuracy (calibration). Cell Engineering personnel have been cleaning these screens over the past year to remedy the problem of reduced flow readings. However, checks on the calibration following a service have not been undertaken.

Several flow meters were calibrated and it was determined that depending on length of operation and meter location, a 50% error in measured flow was possible. Based on the calibration results, it was determined that the filters currently used do not filter the air sufficiently to prevent the flow sensor from falling out of calibration. The filters currently used are rated for 5-10 micron filtration whereas the manufacturer recommends a HEPA (High Efficiency Particulate Arrestance) grade filter (>99.7% of all particulates >0.3 micron). Improper filtration results in the inlet screen becoming blocked, changing the flow of the air, and the flow sensor element itself becomes covered in foreign material, which reduces sensitivity. Cleaning of the flow meter screen only improves

calibration marginally. A HEPA grade filter has been identified which should eliminate this contamination issue. Test stands will be outfitted with these filters and the flow meters checked periodically to verify that calibration is maintained.

Several methods of cleaning the flow meter sensor element were evaluate to determine if the flow meters could be returned to their original calibration. Cleaning the screen only improved the calibration a little (81% to 85% of actual). Washing the flow sensor with soapy water (Figure 24) or using an electrical cleaner almost restored the original calibration (Figure 25). The effect of cleaning the screen on FM-C, or using compressed air to clean the flow-sensing element was negligible.

Figure 24: Illustrating effect of cleaning screen, and washing flow sensing element with water

Figure 25: Illustrating effect of cleaning flow sensing element with compressed air and using ASF mist (Flow meter C, after 13 days operation)

Voltage Sense

There have been a number of approaches to connecting the high temperature voltage sense wires to the manifold voltage sense taps. Early concepts used high temperature (900°F) butt connectors. These were attached to the voltage tap by crimping, which made it difficult when the stacks were removed for repair. The next approach used a range of socket (molex) connectors that offered the flexibility of easy removal. However, these connectors are typically lower temperature, have thin wall construction and consequently deteriorate quickly.

Two solutions are being investigated:

Use a high temperature spade and socket connector

Modify the simple butt connector by crimping to achieve a firm fit on the voltage tap

Using a 9/64 lathe collet and a spacing pin, the butt connector was inserted 1/3rd into the collet and pressed. The exact number of presses, time pressed and orientation of the butt connector seam within the collet varied the fit. The exact settings for the lathe collet will be determined before modifying a run of butt connectors.

Notes:

Butt connector ID ~ 0.07"
(Measured values ranged 0.065-0.07)

Butt connector OD ~ 0.14"

Voltage Tap OD 0.0625"

Exhibit 12: Connector Descriptions

Conclusions

- The spade and socket connectors are supplied in 16-14 AWG only, therefore do not crimp as effectively as 22-18 AWG.
- The spade and socket connector also takes up more space and requires a larger insulating sleeve than the butt connector.
- The straight butt connector is troublesome to modify but offers a streamlined option.

For the reasons above the modified butt connectors will be used in future FC Module testing.

Subtask 1.7 - DC/AC Inversion

Battery Ripple

With improvements in fuel cell generator life and performance, the stress placed on the lead-acid batteries in the system has increased. The primary stress comes from 120Hz ripple current created by the inverters when the batteries are at float. This occurs when the fuel cell is supplying the user load and has fully charged the battery bank. The 120Hz ripple current causes resistive heating in the batteries and, if large enough, can shorten the life of the batteries by creating charge/discharge cycles. Typically, GEL cell battery manufacturers will recommend a ripple voltage less than 4% of the DC float voltage.

An experiment to observe the effects of ripple current is shown in Figure 26. An adjustable DC power supply is used to keep a parallel arrangement of two 12V batteries at float (fully charged). A current transformer is used to inject AC current at 60Hz or

TO 0-120V 10A VARIAC

1:20 CURRENT XFRMR

50A

1 OHM 200W

LAB DC SUPPLY
0-20V, 0-3A

12V 70AH GEL

12V 70AH GEL

SWITCH FOR CHARGE VOLUME TEST

Figure 26: Experiment to observe the effects of ripple current

The battery ripple current experiment was conducted with 71 A rms at 60Hz applied to the generator lead-acid gel batteries at float (13.9V at 25C per 12V nominal battery). Results of the experiment on battery impedance and charge volume are shown in Figure 28 and 29. After 14 days no degradation has been observed.

Figure 28

Figure 29

Inverter Control Design

A damping resistor / capacitor was added to the inverter filter to increase system damping at the filter frequency. The derived model is shown in Figure 30, with a mathematical representation given by equation 1. Note that the model includes feedback filters for both the inductor current (V_{shunt}) and the filter output voltage (V_{out_fb}).

Figure 1.7.C: Inverter Model

Equation 1

$$\begin{pmatrix} \frac{d}{dt} I_{Lf} \\ \frac{d}{dt} V_{cf1} \\ \frac{d}{dt} V_{cf2} \\ \frac{d}{dt} x_{filt1} \\ \frac{d}{dt} x_{filt2} \end{pmatrix} = \begin{bmatrix} -\frac{(R_{cf1} + R_{cf2}) \cdot R_f + R_{cf1} \cdot R_{cf2}}{(R_{cf1} + R_{cf2}) \cdot L_f} & -\frac{R_{cf2}}{(R_{cf1} + R_{cf2}) \cdot L_f} & -\frac{R_{cf1}}{(R_{cf1} + R_{cf2}) \cdot L_f} & 0 & 0 \\ \frac{R_{cf2}}{C_{f1} \cdot (R_{cf1} + R_{cf2})} & -\frac{1}{C_{f1} \cdot (R_{cf1} + R_{cf2})} & \frac{1}{C_{f1} \cdot (R_{cf1} + R_{cf2})} & 0 & 0 \\ \frac{R_{cf1}}{C_{f2} \cdot (R_{cf1} + R_{cf2})} & \frac{1}{C_{f2} \cdot (R_{cf1} + R_{cf2})} & -\frac{1}{C_{f2} \cdot (R_{cf1} + R_{cf2})} & 0 & 0 \\ K_{shunt} \cdot \omega_{c_shunt} & 0 & 0 & -\omega_{c_shunt} & 0 \\ \frac{K_{volt} \cdot R_{cf1} \cdot R_{cf2}}{R_{cf1} + R_{cf2}} & \frac{K_{volt} \cdot R_{cf2}}{R_{cf1} + R_{cf2}} & \frac{K_{volt} \cdot R_{cf1}}{R_{cf1} + R_{cf2}} & 0 & -\omega_{c_volt} \end{bmatrix} \cdot \begin{pmatrix} I_{Lf} \\ V_{cf1} \\ V_{cf2} \\ x_{filt1} \\ x_{filt2} \end{pmatrix} + \begin{bmatrix} \frac{V_{bus}}{L_f} & \frac{R_{cf1} \cdot R_{cf2}}{(R_{cf1} + R_{cf2}) \cdot L_f} \\ 0 & \frac{R_{cf2}}{C_{f1} \cdot (R_{cf1} + R_{cf2})} \\ 0 & \frac{R_{cf1}}{C_{f2} \cdot (R_{cf1} + R_{cf2})} \\ 0 & 0 \\ 0 & \frac{K_{volt} \cdot R_{cf1} \cdot R_{cf2}}{R_{cf1} + R_{cf2}} \end{bmatrix} \cdot \begin{pmatrix} V_{cont} \\ I_{Load} \end{pmatrix}$$

Control design using Equation 1 is ongoing. The first step in the design will be to close the inductor current feedback loop. The transfer functions from the control input to the model outputs are shown in Figure 31. The current loop will be closed around the shunt amplifier voltage (V_{shunt} in Equation 1). The impact of various resistive, inductive, and capacitive loads on this transfer function is shown in Figure 32 (compare to the V_c to V_{shunt} transfer function in Figure 33).

Figure 31: Control Input to Model Output Transfer Functions (No Load)

Figure 32: Control Input to V_{shunt} Output with Various Loads

The challenge, given the various plants shown in Figure 1.7.E, is to pick a stable compensator with adequate tracking at 60 Hz. The loop topology is shown in Figure 1.7.F. A PI compensator is implemented, and a combination of simulations and tuning using the inverter simulator hardware lead to the following gains:

$$H_{PI}(s) = K_P + \frac{K_I}{s}$$

$K_P = 0.5 \qquad K_I = 1000$

Figure 33: Voltage / Current Loop Control Topology

Figure 34: Closed Loop Current Response with Various Loads

Exhibit 13: High Current PCB Connection Test Setup

Current	Duration	Temperature 5/8" Lug	Temperature 1/2" Lug
130A	32 min	61.2 deg C	60.2 deg C
140A	33 min	68.0 deg C	67.0 deg C
140A	55 min	69.5 deg C	70.6 deg C

Table 2: High Current PCB Connection Test Results

Lithium Ion battery technology was evaluated as an alternative to the lead-acid batteries presently used in the fuel cell power plant. A vendor was contacted and a meeting was held at Acumentrics to discuss the application. In general, the smaller size, lighter weight, and longer life of Lithium Ion technology does not make up for its high cost in a stationary fuel cell application. Also, it is unclear how well the Lithium Ion battery will perform under 120Hz ripple current conditions at float.

Task 2.0 - Cell Technology Development

2.1 - Anode Tube Composition Optimization

Isostatic Pressing of Tubes

A collaboration with Praxair has been set up to produce a number of closed-one-end (COE) tubes by isostatic pressing (IP). These tubes will then be fabricated into cells at Acumentrics and evaluated for performance. A number of advantages will be realized by moving to a COE tube, these include;

- Decreased number of seals per active area of ceramic. This is expected to increase reliability of the cell.
- Decreased number of overall seals therefore reducing the total cost of the system.
- Efficiencies in time will be realized at the front end of the tube production process.
- Efficiencies in time and yield improvements will be realized during vacuum deposition of the electrolyte.

Experimental work focused upon pressing tubes at Praxair for evaluation at Acumentrics. The areas of investigation included:

1. TGA/DTA
2. Bisque firing of the green tube
3. Electrolyte coating the bisque fired tube

1. TGA/DTA

TGA/DTA analysis was conducted upon the post pressed powder to determine the temperature at which binder burnout occurs and therefore allow a bisque firing schedule for the pressed tubes to be elucidated.

The following two graphs below Figure 38 shows TGA and DTA data for the post press powder. The TGA shows that binder burnout occurs most aggressively around the 205°C region. This is confirmed by the DTA, which shows an exothermic reaction, taking place in the same temperature region with a peak at 208°C. Burnout is largely complete by 350°C and the total additives in the system account for just under 6% by weight of the total material.

Figure 38

2. Bisque firing of the green IP tube

Exhibit 14 shows a green tube as received from Praxair.

Exhibit 14: COE IP Green Tube Pressed by Praxair

In an initial experiment, the flange on the end of one green tube was cut off and used as a test piece prior to bisque firing an actual tube sample. This was to determine shrinkage and whether the debinding stage of the bisque fire schedule was gentle enough to prevent defects in the fired piece. Exhibit 15 shows the cut off flange in the green state.

Exhibit 15: Flange from the IP Green Tube

The flange was then bisque fired to 1137°C for 4 hours on a bed of zirconia powder supported by a SiC batt. The flange survived the bisque firing intact suggesting that the burnout schedule used should be acceptable for an actual IP tube sample. Shrinkage data was then collected for the bisque fired flange.

The IP flange was then sintered to 1510°C for 4 hours to determine the amount of shrinkage that occurs during sintering. Post sintering, the flange appeared to be intact as shown in Exhibit 16.

Exhibit 16: Sintered IP Flange

The average bisque to sintered shrinkage calculated is 19.4%. This compares favorably with Acumentrics current standard production shrinkage value and should, as a result, lead to an intact leak tight electrolyte layer after sintering. The combined shrinkage of the bisque and sinter firing steps for the ISP tube was approximately 26%.

An actual IP tube was then bisque fired using the same schedule that was used for the flange. Examination of the tube after firing revealed it to be cracked and slumped. The cracking of the tube appeared to be because the wall thickness was too thin to support itself, particularly at the open end once the binder had been burned off. Exhibit 17 shows the tube after the first bisque-firing attempt.

Exhibit 17: IP Bisque Fired Tube with Flange Removed

Another attempt was then made at bisque firing an IP tube, this time however the majority of the flange was left intact to give the tube more support at the open end once the binder had burned off. One side of the flange was ground down with a drum sander so as not to impede shrinkage of the tube during firing, the ground side was obviously placed sand side down. After bisque firing, the IP tube was still intact with no detectable cracks. The tube had however sagged slightly just as the first IP tube did due to the thin wall thickness. Exhibit 18 shows the ground end of the flange after bisque firing.

Exhibit 18: Ground Flange of the IP Tube that Survived the Second Bisque Firing

3. Electrolyte coating the bisque fired tube

The intact IP tube was then successfully vacuum dip coated with YSZ electrolyte slurry, in Exhibit 19.

Exhibit 19: IP Tube Dip Coated with YSZ Electrolyte

Further tubes with a thicker wall are currently being pressed in an attempt to overcome the slumping issue during the bisque fire. Tubes will also be electrolyte coated and fired to determine the processing issues associated with both steps.

Vertical firing kiln furniture design

All horizontal firing trials conducted to date have resulted in slumping of the IP tube whether during the bisque or electrolyte firing stages. A limited amount of vertical firing data has also been collected which indicates that the IP tubes remain straight and round when fired in this plane.

Developmental kiln furniture has therefore been designed to allow vertical firing of the tubes in Acumentrics current furnaces. 3D representations of the proposed furniture are shown in the following diagrams.

Exhibit 20: Silicon Carbide Beam

Exhibit 20 shows a silicon carbide (SiC) beam that has cutouts removed from it whilst still green. These cutouts are for tube positioning and as such, 8 tubes may be fired on each SiC beam. Each COE tube to be fired has an alumina collar, which is slightly larger than the tube diameter, slipped over its closed end and fed along the tube length until it engages the tube flange. The tube with the alumina collar is then electrolyte infiltrated and subsequently loaded onto the kiln furniture. The alumina collar is the only material in contact with the SiC beam

Exhibit 21: COE Tubes on Furniture

The top and underside 3D image views show eight COE tubes loaded onto the furniture along with the alumina retaining collars. Also shown are the support posts that are positioned either end of cutout beam, these are also fabricated from SiC.

This actual furniture design is in the process of being fabricated. Elements of the design such as the alumina washers have already been evaluated and work effectively. A further advantage of firing vertically is that the electrolyte on the tubes is not in contact with the kiln furniture and as such contamination is minimized.

Subtask 2.2 - Electrolyte Composition

Electrolyte Thickness Evaluation

The reason for Acumentrics remaining cell degradation is still not fully understood. In an effort to determine what role the electrolyte might play, a batch of tubes has been split into three equal sets before coating with electrolyte. Each set was then dipped with YSZ for a different time interval, 8, 15 or 24 seconds, resulting in three sets of tubes with different electrolyte thickness.

Cells from each different set of dipping conditions were then selected and submitted to cell testing for performance analysis. Cells were also selected for their firing position in the furnace for the three different firing steps in an effort to elucidate any effects of firing position on performance. The cells are presently in the cell-testing queue.

Subtask 2.3 - Cathode Composition

A vendor provided Acumentrics with a sample of LSM containing higher strontium levels than currently used for standard production cells. The lot number for the powder is T2065. The chemical formula for the sample is $\text{La}_{0.8}\text{Sr}_{0.2}(\text{MnO}_3)$.

A comparison of the performance provided by this experimental LSM with respect to the current production material has been detailed. In addition to a potential performance enhancement, this material is more than 5 times cheaper than the current LSM purchased.

C1 and C2 slurries were first formulated from the (80/20) LSM cathode material and YSZ (-1 μ). The formulations for the C1 and C2 slurries are shown in Table 3.

Table 3: Experimental LSM Slurry Formulation

Cathode Slurry	Material Percentages by Weight			
	LSM	YSZ	Terpineol	Iso-Propanol
C1 Slurry	16%	16%	13%	58%
C2 Slurry	40%	NA	13%	47%

The YSZ fraction for the C1 slurry was milled in iso-propanol for 24 hours before the terpineol and LSM powder were added. The final formulation was then milled for an additional 15 minutes. The C2 slurry was milled for 15 minutes with the addition of all the materials. The slurries were then tested for particle size distribution and viscosity before applying them to cells. The material testing data collected, for both cathode slurries, is compared to standard production cathode slurries, in Table 2.3.2.

Table 4: Comparison of the Slurry Properties for the Vendor and the Production Cathode

Slurry ID	D₅₀ for YSZ (μm)	Final D₁₀ (μm)	Final D₅₀ (μm)	Final D₉₀ (μm)	Viscosity @ 160 rpm (cP)
C1	0.432	0.352	0.635	1.332	9.31
Production C1	0.596	0.322	0.596	1.756	8.30
C2	NA	1.038	1.610	2.486	9.85
Production C2	NA	0.897	1.838	4.015	8.66

Tubes were then coated with the experimental slurries and sintered at two different high temperatures for hours. The two different temperatures were:

- 1175°C for 2 hours
- 1250°C for 2 hours

Outcome

After sintering, the cathode resistances for both firing regimes were measured along the length of the tube. As expected, the tubes that were sintered to 1250°C had a lower resistance than the tubes sintered to 1175°C. It was also noted that the AE cathode fired to 1175°C was under sintered, evident by the fragility of the resulting coating. Table 5

shows the cathode resistance compared to the firing temperature and the sintered weight of the cathode applied to the tube.

Table 5: Summary of the Cathode Resistance and Weight Compared to the Sintering Temperature

Tube Number	Firing Temperature	Resistance (ohm)	Cathode Weight (g)
B180351	1250°C 2hr	706	0.9860
B180374	1250°C 2hr	525	0.9731
B180358	1250°C 2hr	627	0.9541
B180346	1175°C 2hr	806	1.0376
B180360	1175°C 2hr	734	1.0188
B180354	1175°C 2hr	1050	0.8955

Acumentrics standard cathode coating has a cathode end-to-end resistance 2-3 times higher than the values recorded in the table above even though the weight of applied cathode is typically 300-500 mg greater with the standard formulation.

Two of the 1250°C fired cells, B180374 and B180358 were then cell tested at standard conditions of 150mA/cm², 75% fuel utilization at 800°C. The cells were on test for 498 hours before being removed from the cell test stand to make way for other tests. Both cells displayed peak voltage values comparable to the highest performing cells fabricated from Acumentrics current production LSM material. Table 6 displays a summary of the cell test results produced.

Table 6: Summary of the Cell Test Results for the Initial Cell Test of the Cathode

<u>Tube Num.</u>	<u>Peak Voltage (mV)</u>	<u>End Voltage (mV)</u>	<u>IR (mΩ)</u>	<u>OCP (V)</u>	<u>Time on Test (hr)</u>	<u>Thermal Cycles</u>	<u>Fuel Utiliz.</u>	<u>Degradation per 1000 hrs</u>
B180374	677	667	1.049	1.094	493	2	75%	-7.2%
B180358	682	669	1.014	1.091	493	2	75%	-9.8%

Figure 39 shows the potential output versus the time the cells were initially on test.

Figure 39: Potential output versus the time the cells were initially on test.

This initial cell test showed that these cells had a significant increase in potential (48mV and 53mV) when compared with the cell that was sintered to 1175°C. This increase could be attributed to the simple fact that the cathode was sintered to a higher temperature allowing for a lower cathode resistance and thus better connectivity among the cathode particles. In agreement, the IR values observed for both cells fired to 1250°C are lower than the 1175°C cell.

The IR versus time in Figure 40 shows that the IR is decreasing during the first 380 hours of testing.

Figure 40: The IR is decreasing during the first 380 hours of testing.

The 1250°C cells were then put back on test in a different cell test stand (CTS 10) from the initial one utilized. Table 7 displays a summary of the cell test results produced for the second time the cells were placed on test.

Table 7: Summary of the Cell Test Results for the Continued Cell Test

<u>Tube Num.</u>	<u>Peak Voltage (mV)</u>	<u>End Voltage (mV)</u>	<u>IR (mΩ)</u>	<u>OCP (V)</u>	<u>Time on Test (hr)</u>	<u>Thermal Cycles</u>	<u>Fuel Utiliz.</u>	<u>Degradation per 1000 hrs</u>
B180374	631	487	1.114	0.973	1494	6	75%	-22.82%
B180358	633	525	1.199	1.092	1494	6	75%	-17.06%

Figure 41 shows the potential output versus time on test for the second cell test of the original AE cathode cells tested in Figure 39.

Figure 41: Potential output versus time on test for the second cell test of the original cells tested in Figure 39.

The results laid out in Table 7 above are interesting in that when the cells were put back on test, both had experienced a 34mV drop from the end of one test in CTS12 to the beginning of the second test in CTS 10. Both cells also experienced a nominal drop of 26mV after a 15°C adjustment down in temperature of the cell test stand, 17.3 hrs into the second test run. These two factors alone explain at least some of the increase in degradation numbers observed between the first and second tests.

Figure 42 shows that IR increases with time for both cells under test. The IR for cell B180358 continued to increase during the duration of the cell test while the IR for cell B180374 did not change significantly during the last 400 hours on test. This increase in IR, for both cells contributed to the continuous decline in the cells' potential during the lifetime of the test.

In Summary

The new cathode material must be sintered to at least 1250°C for 2 hours to achieve good connectivity among the cathode particles and therefore a lower resistance. Cathodes fabricated from the same material and fired at 1175°C showed poor mechanical stability and a nominal 50mV reduction in peak potential when tested under standard operating conditions.

Acumentrics standard cathode coating has a cathode end-to-end resistance 2-3 times higher than the values recorded for the new LSM even though the weight of applied cathode is typically 300-500 mg greater with the standard formulation.

Both tested cells displayed peak voltage values comparable to the highest performing cells fabricated from Acumentrics current production LSM material.

The observed cell potential and hence degradation values are much worse for both cells when transferred to a different cell test stand after 493 hours of operation.

In addition, the 34mV drop in cell potential from the end of the first test to the start of the second is difficult to reconcile as simply due to the thermal cycle between the two tests.

This is stated since during the second cell test, the cells were thermal cycled 6 times and their potential came back up to or within a couple of mV of the potential observed pre

cool down. This rebounding trend does not support the 34mV decrease in the potential observed for both cells between the two cell tests.

Before a confident determination of the material's merit, with regard to the standard LSM, can be concluded further analysis and cell testing will be conducted.

Cathode and Anode GDC Impregnations

Polarization losses in the cathode should be reduced by the addition of the mixed conductor Gd-doped ceria (GDC) due to its facility for oxygen reduction. Incorporation of GDC into the C1 layer (i.e. the cathode) can be achieved by a number of means. LSM/GDC composites and GDC doped LSM/YSZ cathodes have both been reported in the literature. Aqueous solutions will infiltrate the porous cathode and so a first attempt at boosting cell performance will be made using by impregnation with aqueous Gd and Ce salts. The appeal of this method is that it is simple and can be used on cells retroactively.

A number of methods to impregnate existing cells were tried. The dipping method was used to impregnate the cathode of one cell, and the anode and cathode of two cells; the painting method was used to impregnate the cathode of a fourth cell; a fifth cell was a control. Two cells were impregnated using the painting method 120 hours into the experiment

Cell No	Trial	Metal load/g	Coating Method
<i>Initial</i>			
C120033	1	0.29	painted
C120031	1	0.10	dipped
C120034	1	0.77	dipped
C120040	1	0.71	dipped
C120037	1	0	
After 120 hrs			
C120031	2	0.68	painted
C120037	2	0.43	painted

Table 8: GDC Impregnation Trials

Time/hrs	18.9	27.3	119	138.6	145.5	164.4
Temp./°C	800	845	700	800	700	845
Voltage/V						
C120033	0.648	0.654	0.485	0.600	0.442	0.623
C120031	0.629	0.642	0.465	0.638	0.579	0.636
C120034	0.659	0.671	0.517	0.633	0.496	0.653
C120040	0.640	0.647	0.499	0.586	0.454	0.608
C120037	0.596	0.615	0.394	0.622	0.527	0.637

Table 9: Cell Performance (shaded cells are control voltages)

All impregnated cells generally operated better than the control with an apparent 30-60mV gain at 800 and 845 °C

However, the results are not definitive on account of the anomalously high area specific resistance of the control (Figure 43). It is possible for the impregnation to play some role in improving C1 conductivity, however in the worst case, *i.e.* that the resistance is unaffected by the impregnations, the results should be corrected. In this case there does not appear to be an unequivocal improvement in the cell potential at 800 or 845 °C. This is also no significant difference in the apparent exchange current densities at these temperatures (Figure 44).

Figure 43: Area Specific Resistance

Figure 44: Exchange current density

However at 700°C, there is a clear improvement in the impregnated cells over the control of ~100mV. Correcting for resistance would lead to at least a 40mV increase in performance over the control, for 3 of the impregnated cells, but no improvement for C120031. This is also indicated in Figure 44 where the exchange current densities are relatively high for only three of the impregnated cells.

LSM/GDC Cathodes

Eight cells were prepared with GDC/LSM cathodes. Unfortunately the GDC (nano-particle, NexTech) particle size distribution was broad with $D_{50} \sim 3.0\mu\text{m}$ and $D_{90} \sim 10\mu\text{m}$ even after milling. The nano-particle features also engendered a high viscosity in the resulting 34 solids wt% C1 slurry (53 wt% $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_x$, 47% wt LSM) making it difficult to coat cathodes. It was necessary to reduce the solids loading to 21.6 wt% to produce a slurry of the correct consistency. All eight cells were further coated with standard LSM C2 slurry.

The literature reports that a highly resistive GDC/YSZ solid solution starts to form in the vicinity of 1200-1300°C, and thus four cells were fired at 1250°C for 2 hours and the remainder fired at 1200°C. In both cases it was found that the cathodes did not bond well to the electrolyte, and could be rubbed off from the electrolyte. Approximately 5-10% of the cathode area immediately fell from the tube on removing from the furnace.

The subsequent reduction did not seem to further damage the cathode, and ultimately four of the eight cells (2x1250°C, 2x1200°C) were wound with silver, brazed and submitted to cell testing.

LSF Cathodes

Eight cells were fabricated with LSF cathodes using the standard dip coating process. The solids slurry was made using powder from combustion spray pyrolysis. Four of the cells were dipped twice in order to increase the cathode thickness. Four cells (2x 1 coat, 2x 2 coats) were fired at 1100°C for 2 hours, and the remainder at 1250°C . In both cases it was noted that the cathode coating bonded well to the electrolyte.

Other than the cathode, the cells were manufactured as standard cells, and 5 cells were ultimately tested (Table 10).

Cell No	Weight LSF/g	Firing Temp/°C
C180284	0.45	1100
C180252	0.77	1250
C180240	0.72	1250
C180249	0.80	1100
C180294	0.82	1100

Table 10: LSF Cell Trials

		Time/hrs	90	95	113	233	235	236
		Temp./°C	800	700	850	850	800	700
Cell No.	J/mA/cm ²	%FU						
C180284	150	75	0.653	0.405	0.685	0.679	0.651	0.376
C180252	80	40	0.169		0.363		0.324	
C180240	80	40	0.313		0.441	0.342	0.227	
C180249	150	75	0.669	0.423	0.699	0.674	0.629	0.327
C180294	150	75	0.651	0.365	0.689	0.676	0.633	0.319

Table 11: Cell potential in V, near peak potential (~100hrs) and at the end of the cell test (blank cells indicate voltages sank below V_{min} of 0.1V)

The cell performance of the two cells fired at 1250°C was extremely poor, in Table 11, and the low apparent exchange current densities (~4 mA/cm²) suggests that this could be a result of low surface area due to over-sintering.

Subtask 2.4 - Cell Testing

VJ Overview

A new database was created for the statistical analysis of low end VJ data.

Figure 45: Apparent current exchange densities plotted against week of fabrication (open symbols are Ni foam variants, closed symbols are standards)

Figure 2.4.1 displays some of the results of low-end VJ analyses for cell tests conducted in 2004. Clearly temperature is seen to have a big effect on the apparent exchange current density, but due to the sparse data set it is difficult to accurately elucidate any trend for the foam cells. J_0 appears to be reasonably consistent and reproducible but modeling work will be needed to elucidate the sensitivity of the apparent exchange current density to changes in the anode and cathode exchange densities.

Cell Modeling

Using the cell model developed in the past budget period, VJ curves were prepared for the following current collection scenarios:

- 1) Double braze injector cells
- 2) Ni foam current collector enhancements
- 3) Large diameter cells 22mm OD
- 4) Thicker walled anodes (1.88 mm wall)
- 5) Multiple takeoffs

No further parameter fitting was performed - materials parameters used previously for modeling of the standard single braze cell (Table 12) were held over so that a comparison of the potential gains for each of the current collection cases could be immediately explored. The anode exchange current density in all cases was fixed high at 2 A/cm² and the cathode exchange density was preserved at 0.1A/cm², as found from the fit to experimental data. For both anode and cathode, the exchange current density is assumed to be independent of reactant concentrations

Symbol	Description	Value	Unit
σ_E	Electrical conductivity of electrolyte	0.024	S cm ⁻¹
σ_A	Electrical conductivity of anode cermet	3000	S cm ⁻¹
σ_C	Electrical conductivity of LSM	60	S cm ⁻¹
σ_{Ag}	Electrical conductivity of silver	142857	S cm ⁻¹
σ_{Ni90}	Electrical conductivity of Ni foam 90ppi	312	S cm ⁻¹
σ_{Ni60}	Electrical conductivity of Ni foam 60ppi	883	S cm ⁻¹
t_{Ni90}	Ni foam 90ppi thickness		S cm
t_{Ni60}	Ni foam 60ppi thickness		S cm
t_E	Electrolyte thickness	2.5×10^{-3}	cm
t_A	Anode support tube thickness	0.15	cm
t_C	Cathode LSM current collector thickness	3.5×10^{-3}	cm
A_{Ag}	Cross sectional area of silver bus-bar/collector		cm ²
J_{OC}	Cathode exchange current density	0.1	A cm ⁻²
J_{OA}	Anode exchange current density	2.0	A cm ⁻²
D_{H_2}	Binary diffusivity of H ₂ -H ₂ O	8.6	cm ² s ⁻¹
D_E	Effective H ₂ diffusivity in anode cermet	0.86	cm ² s ⁻¹

Table 12: Model parameters (800°C)

Figure 46: Model VJ (75% FU) predictions of various cell modifications

Figure 46 shows the model VJ for the double brazed cells with injector tubes (*i.e.* gas inlet at opposite end to the current collection), for the next generation 22mm cell, and for Single braze (SB) variants each with improved anode side collection. Note that, in qualitative agreement with experiments (not shown), the VJ predictions are very similar, with anode current collection improvements only showing nominal performance gains.

Figure 47: Thick wall VJ (75% FU) predictions and experiment

Figure 47 compares experimental data for the single braze standard cells and the thick walled anode cells, to the model prediction. Immediately noticeable is that experimentally, there is negligible improvement of the cell performance, whereas the model predicts a potential gain of ~25mV. The model provides further justification for re-testing new thick walled cells, but with closer attention to be paid to the anode, in particular, the conductivity of the anode tube should be quantified prior to the experiment.

Figures 49 compares predictions for three different methods of current collection on double brazed cells with injectors (Figure 48).

Figure 48: Current collection alternatives, (a) 1 takeoff, (b) 2 takeoffs, (c) 3 takeoffs

Figure 49: Predicted VJ curves (75% FU) for multiple take-offs

Significant gains in performance are obtained, presumably because of a reduction in collector resistances, but also perhaps due to lower activation polarization losses.

LaCrO₃ Testing Results

A normalized graph of performance of all interconnection cells tested to date was compiled. From the results, it was clear the cell performance is significantly higher than that of our current standard (0.650V), and that these cells, with one interconnection, could achieve potentials on average about 0.710V. The second aspect that became clear was the detrimental effect of thermal cycles on the cells. The two initially tested cells (one of which was thermal cycled early, and one of which was thermal cycled later in life) showed excellent performance. However, the cell that was tested for over 1000hrs proved that even one thermal cycle can have a significant effect on performance (loss of 0.030V).

Figure 50: LaCrO₃-IC All Cells Plotted at Standardized Conditions of 150 mA/cm²

The performance enhancement observed was due to the lowering of the effect cell resistance, borne out by the observed ohmic drops measured throughout the cell lifetime. See Figure 51.

Figure 51: Ohmic Drop Measurements

Clearly, the larger effect is the elimination of the loss due to thermal cycles. To that end, cells are being tested with different current-pickup configurations, and different bonding layers for the chromite interconnection.

Re-Braze Experiment

In an effort to further ascertain the contribution of the copper braze degradation, the inlet braze cap of five D15/L330-BB-ST cells tested in excess of 6000 hours, were cut off, the cell inlets re-brazed, and the cells re-submitted for testing. At the end of the initial long-term cell test, the degradation rates had spanned from -1.8% /khr to -3.7% /khr with an average of -3% /khr. Moreover, by the test end the cells had been struggling to maintain a stable performance for times greater than a few hours.

After re-brazing, stable performances were obtained and re-calculated degradation rates spanned from 0.0% /khr to -0.9% /khr, with an average of -0.6% /khr. Note that this latter degradation calculation accounts for the section of anode that was necessarily cut from the original cells in order to re-braze them. At least 70% of the degradation appears attributable to degradation of the braze, although there was conflicting evidence as to what was the specific mechanism for the loss. The degradation could not be quantitatively attributed in its entirety to contact issues or cell leaks. Other mechanisms for degradation must still be explored.

Based on the results of cells with nickel inserts, a parallel contactor from the nickel cermet to the nickel braze cup might be a worthwhile contingency effort.

2.5cm Cell Testing

A large 3-IC cell was placed on test. At 50%FU, 850°C, 300mA/cm², 42W was achieved. This is not a peak power. It has also been determined that the cell has a substantial leak that is affecting performance. This is evidenced by the slow climb back to open circuit and the low OCP for some IC positions. This has been traced to a spraying alignment issue where the fixturing for this first cell was not properly aligned with the IC area, resulting in insufficient overlap of the spray.

However, the test has been highly successful, and suggests Acumentrics is very close to achieving a 60W cell.

Task 3.0 – Stack Technology Development

Subtask 3.1 – Cathode Current Collector Improvements

Cathode current collection losses were explored in detail using analytical and finite-element techniques. The impetus for the investigation was to predict the order of magnitude of improvement in cell voltage that could be obtained by substituting LSM for LSCF (La_{0.6}Sr_{0.4}Co_{0.8}Fe_{0.2}O_{3-d}). The two modeling techniques were consistent and predicted the losses for the standard cells' cathode operating at 150mA cm⁻² to be only on the order of 5mV. Moreover the presence of a 3 micron silver layer on top of the LSM was predicted to be sufficient to reduce the losses to essentially zero even at 1A.cm⁻². Thus if LSCF is pursued it should be from the point of view of reducing activation polarization. A further result of the analysis was the prediction that the amount of silver wire used for collecting the current can be probably halved without seeing an appreciable increase in resistive losses in the collector.

Subtask 3.2 – Anode/Cathode Current Collection

A silver/chromite connection experiment shown below in Figure 22 continued on test for over 1200 hours. After 1272 hours and 53 thermal cycles, the resistance has remained stable at approximately 2.8 Ω at 838°C.

Exhibit 22: Silver/chromite connection experiment

Figure 52: Silver/chromite connection experiment

One of the limiters in terms of cell performance is the conductivity of the anode bus. This issue is presently being addressed in two ways.

1. Higher Ni content and differing particle morphology cermets are presently being evaluated.
2. Work has been initiated to apply lanthanum chromite bands along the length of the cell. This will allow the fabrication of longer length cells with a number of current take off positions or chromite bands along the length.

Initial investigations into plasma spraying a chromite band took place 08- 24- 04. Evidence from these previous plasma spray trials suggested that the applied chromite delaminates from the electrolyte during thermal treatment. Subsequent microscopy of the as sprayed tube x-sections have however shown that is not the case.

Exhibit 23 shows a section through the anode, electrolyte and chromite layers after plasma spraying. This sample has undergone no post spray thermal treatment.

The first point to note is the interfacial gap between the electrolyte and chromite coatings after plasma spraying, suggesting that the chromite layer does not effectively wet the electrolyte layer during spraying. This suggests that the de-lamination observed in the previous study during thermal treatment of the sample was not in fact de-lamination since it appears that there was never any real bond between the two layers in the first place.

Exhibit 23: Anode, electrolyte and chromite layers after plasma spraying (1)

The second point to note is the splat structure of the plasma sprayed chromite layers. The image below Exhibit 24 shows the same sample after heat treatment at 1400°C for two hours. The splat structure of the plasma sprayed chromite that was evident pre heat treatment has disappeared completely. The interfacial gap between the chromite and the electrolyte is again clearly visible. There does however appear to be good contact between the chromite and the anode even after thermal cycling of the cell. It is this intimate contact with the anode that makes it visually appear to look like the chromite is also bonded to the electrolyte itself after spraying.

Exhibit 24: Anode, electrolyte and chromite layers after plasma spraying (2)

From the images in Exhibits 23 and 24 it has therefore been concluded that the chromite never really wets the electrolyte surface during plasma spraying and therefore a bond is not formed between the two layers. The smooth, dense nature of the fully sintered electrolyte does not allow the chromite to mechanically key into the surface of the material.

Based upon these early results a number of different parameters have been investigated to try to resolve the chromite adhesion issue. The variables investigated include:

1. Bond coat application before spraying the chromite.
2. Roughened tube surface either side of the chromite band with YSZ powder.
3. Partial sintering of the electrolyte, followed by plasma spraying of the chromite, then fully sintering the electrolyte and chromite in an additional firing step.

Sample Preparation

1. Bond coat application before spraying the chromite.

This investigation involved the application of a bond coat through plasma spraying prior to application of the chromite layer. The bond coat was applied to conventionally processed tubes. The tubes were prepared in the standard way prior to plasma spraying, in Exhibit 25, of tubes after electrolyte firing.

Exhibit 25: Tubes after electrolyte firing

2. Roughening the surface either side of the chromite band with YSZ powder

It was considered that roughening the surface of the electrolyte in the chromite overlap region would increase the mechanical keying or interlocking taking place between the chromite and electrolyte.

To achieve this roughened surface region, the bisque fired tube was first vacuum infiltrated with electrolyte. Whilst the tube was still moist with solvent, YSZ powder was

applied liberally over the circumference of the tube in the overlap region. Excess YSZ was then blown from the tube using compressed cylinder air and the coated tubes electrolyte sintered.

Exhibit 26: Roughened surface tubes at post electrolyte sintering

The uniformity of the roughened overlap region is not good but it was considered that the above samples were adequate to determine if the roughened surface path of investigation had merit.

3. Partial pre-sintering of the electrolyte.

By only partially sintering the electrolyte coated tube before plasma spraying with chromite, it was considered that the reduced electrolyte density would provide a more favorable surface for the chromite to adhere to.

Tubes were prepared for spraying with chromite in the usual way apart from the fact that the electrolyte sinter temperature was reduced from 1500°C to 1300°C. An image of the tubes after electrolyte firing is provided in Exhibit 27.

Exhibit 27: Tubes after electrolyte firing

Plasma Spraying

All three sets of tubes were then plasma sprayed with chromite using the same set up and fixturing. The gun power and chromite feed rate were set to the same values for each trial.

No problems were experienced during spraying of the bond coat or roughened surface samples in that, to the naked eye, all applied chromite coatings appeared intact after spraying and all tubes survived the spraying process. All the reduced firing temperature samples however broke either as soon as the tube began spinning in the chuck or after the second pass with the plasma spray gun. No further trials using reduced sintering temperature tubes will be conducted.

Post Spray Testing

B490172 was the first tube to be evaluated after plasma-spraying trials were completed. The conductivity at 800°C of the chromite band was initially investigated using a 4-point probe methodology. As with the previous plasma spray trial, the conductivity along the length of the chromite was found to be in the same region. Upon cooling this sample, no de-lamination of the chromite was observed. The tube was then reduced in the standard reduction furnace under standard conditions. Exhibit 28 shows the tube after reduction, which is clearly not fully reduced. The reduction front can be seen $\frac{3}{4}$ of the way along the length of the tube, suggesting that the chromite was porous to some extent.

Exhibit 28: Tube after Reduction

The leak rate of the partially reduced tube was then measured using Acumentrics semi-automatic QC checker and was found to be high, again suggesting that the chromite region was too porous to enable complete reduction. Sections of this tube have been sent for microanalysis and chromite layer thickness determination.

An effort was then made to try to densify the as sprayed chromite layer of the remaining bond coat tubes. As such, tube B490174 went through an extra sintering step before application of cathode to the cell. After thermal treatment at this temperature, no delamination of the chromite was detected. Cathode was then applied to the cell by masking off the chromite band with scotch tape prior to dipping. The cathode was then fired and the cell successfully reduced utilizing the standard reduction furnace and conditions. The measured leak rate for this tube in the reduced form was much improved over the previous tube B490172 and was similar to that of production cells. The extra sintering step results in a chromite band dense enough to allow full reduction of the tube.

All the samples that had a bond coat layer as an intermediate survived all thermal cycling procedures that they experienced without de-lamination whether grit blasting took place prior to spraying or not.

The series of tube samples with the roughened surface either side of the chromite/electrolyte overlap region did not show any visual signs of chromite de-lamination post spraying. Upon thermal treatment however, all the roughened surface samples without an applied bond coat showed de-lamination of the chromite layer, although much reduced with respect to the non roughened samples prepared in the previous trial. See the image in Exhibit 29.

Exhibit 29: After thermal treatment

Interestingly, all the roughened surface tube samples showed chromite de-lamination from one side of the overlap area only, the other side appearing to remain intact after thermal treatment. Grit blasting of the sample again appeared to make no difference to the de-lamination behavior of the chromite upon thermal treatment.

In addition to having a roughened surface, tube B490169 also had 1 bond coat applied prior to plasma spraying of the chromite. Upon thermal treatment of the tube, the applied chromite layer remained intact upon the tube. The tube could not be fabricated into a cell however since as was the case with B490171 described previously, during the densification step the electrolyte became compromised by contaminant from the furnace lining. Both tubes were fired at the same time in the same furnace.

A new cell design is being developed that utilizes a doped lanthanum chromite band wrapped around the center of the anode support (AS) tube. This material has the unique properties of electronic conduction in both oxidizing and reducing atmospheres. Thus it is highly suitable for use as an interconnect material for solid oxide fuel cells. To build on the successful single cell tests that have been conducted, a 30-cell bundle test will be performed to further evaluate cell performance, evaluate cell-to-cell connection issues and obtain stack configuration performance. Before this can be accomplished though, a method of current collection between cells must be developed. The system must be capable of forming series connections between the cells, that is, passing current from the anode (chromite band) to the cathodes on the next cell in the row. The material of choice for this system is silver due to its oxidation resistance at the operating temperature of 800°C.

A number of different current collection configurations are presently being developed. The first, which builds on the results of the furnace tests described below, consisted of a stamped and formed silver clip, which ‘snaps’ over the chromite band. Since silver is a very soft material at 800°C, another clip is ‘snapped’ over the silver clip to maintain some degree of compression onto the chromite band.

Microscopy

A number of samples were investigated further through micro analysis to determine coating thickness and integrity.

Tube B120011 did not have an applied electrolyte coating before plasma spraying with bond coat and chromite. After plasma spraying, the tube was heated to check for the occurrence of chromite de-lamination upon thermal treatment and to densify the chromite layer. Upon inspection of the tube after thermal treatment, no de-lamination of the plasma sprayed band was apparent. The cell was then sectioned for micro and is shown in Exhibit 30.

Tube B120011

Exhibit 30: Cell sectioned for micro images

The images above indicate that the coating thickness for 10 passes of the chromite topcoat is in the region of x microns thick. The bond coat is not readily visible on this sample but may be the region between the anode and chromite layers, which contains increased porosity. The density of the applied chromite layer appears good with some closed porosity

Tube B490172 had an applied electrolyte layer. Post spraying of the chromite, the tube was thermally treated, after which no observable de-lamination of the chromite had taken place. The tube was then reduced in Acumentrics standard reduction furnace using standard conditions, after which it was sectioned for micro. During reduction the tube only reduced $\frac{3}{4}$ of the way along its length and thus it is considered that the chromite must have been porous, allowing hydrogen to pass through it.

Tube B490172

Exhibit 31: The bond coat layer to be clearly visible between the electrolyte and chromite layers

The images in Exhibit 31 show the bond coat layer to be clearly visible between the electrolyte and chromite layers. Further, the bond coat layer appears to contain numerous very large pores, the origin of which is unknown at this stage. To maximize coating integrity and also electrical conductivity through the chromite, these pores must be eliminated in future samples.

At this stage it is not known when the pores form, whether it be during plasma spraying, thermal treatment or reduction. The pores do however appear somewhat rounded in nature and very large, suggesting that degassing of some phase may be responsible but it is not known when or how this could occur since any potential degassing would be expected to occur preferentially in the plasma spray stage.

The thickness of the bond coat appears to be in the region of x microns while the chromite layer is in the region of y microns for tube B490172.

The images above also show some evidence of what appears to be the electrolyte fracturing, possibly due to the TEC mismatch of the bond coat and YSZ layers, although unexpected since the bond coat layer is very thin. This is however speculation at this stage and is based upon the microscopy images from one sample and as such additional data must be collected to verify this.

Summary Cell Test Results

Two chromite plasma sprayed tubes were then submitted to cell testing for performance analysis.

Both cells were processed in the same way. After plasma spraying was complete, the cells were thermally treated, after which they were reduced in Acumentrics standard reduction furnace and then leak checked before being evaluated for performance.

The interconnect current collection system was different for each cell put on cell test. For the initial cell, B490174, silver paint was first applied to the chromite layer, followed by a silver braid wrap. Cell B490167 however first had silver paint applied followed by a silver clip, on top of which was placed spring clip to compress the silver clip onto the applied silver paint.

Cell B490174 was the first chromite interconnect cell to be assessed for performance under Acumentrics standard testing conditions of $150\text{mA}/\text{cm}^2$, 75%FU and 800°C . Figure 53 is its life curve to date which shows low degradation over the first 1319 hours, excluding the first 24 hours under test. The observed cell potential was also encouraging in that it was 60-70mV higher than would be expected for a standard production cell run under the same standard operating conditions.

Figure 53: Life curve showing low degradation over the first 1319 hours of 1.3%, excluding the first 24 hours under test

The IR was also measured for cell B490174 over its lifetime and moved within a narrow range from 0.62 at the start of the test to 0.68 ohm.cm^2 when the latest available reading was taken at 918 hours. It is apparent that the reduction in IR with respect to standard cells accounts for the improved performance shown by the interconnect cells. This is obviously expected since the anode bus length is essentially cut in half through the introduction of the chromite band.

Cell B490167 was the second chromite interconnect cell to be assessed for performance under Acumentrics standard testing conditions of 150mA/cm^2 , 75%FU and 800°C . Figure 54 is the life curve recorded to date, which shows that the cell has already undergone seven thermal cycles. Each thermal cycle resulted in a voltage drop, which on some occasions was recovered with time but on others the potential remained below that observed before the cycle. This behavior was particularly pronounced early on in the cell lifecycle. The observed cell potential was again encouraging in that it was 60-70mV higher than would be expected for a standard production cell. This cell could not however be run under standard operating conditions in that a fuel utilization of 55% rather than the standard 75% had to be used in order to allow acceptable operation of the cell. The IR for the cell was measured to be 0.7 ohm.cm^{-1} at the outset of test and climbed gradually to 0.8 ohm.cm^{-1} by 624 hours, the latest reading taken so far. At hour 618 the current density was increased to 250 mAcm^{-2} to allow further analysis of the cell to take place.

Both cells are currently undergoing further cell testing and performance evaluation.

Figure 54: Life curve showing that the cell has already undergone seven thermal cycles

Current Collection

Current collections methods capable of working in conjunction with the lanthanum chromite banded cell geometry were investigated.

A number of ‘dummy’ cells were fabricated and screwed into two manifolds to simulate two layers in a stack. This is the minimum number of layers necessary to form a series connection. Two different current collection methods were developed and the corresponding silver/spring clips were fabricated. These clips were designed to work with the baseline manifold geometry wherein the manifold layers are stacked together after cells are installed along with the appropriate current collector.

It was found that a large amount of patience was necessary to align the individual silver tabs and the spring clips in order to ‘snap’ the next layer in place. This method, while successful, would certainly test the fortitude of the assembler.

Subtask 3.3 – Generator Design

Power Lead Connection

Testing of the power lead connections continued throughout the reporting period with periodic measurements taken. Figure 55 shows the connection resistance with time. After 2088 hours and 22 thermal cycles, the resistances are stable.

Figure 55: Connection resistance with time. After 2088 hours and 22 thermal cycles, the resistances are stable.

Thermoelectric Effects

Absolute Thermo power data for a number of metals was obtained from the literature and used to calculate the thermoelectric effect for the Ag-Ni couple. For a conducting junction at 800°C the potential difference between Ni and Ag is estimated at 21.1mV, which compares well with an experimental estimate by the Generator Design team of 20mV.

Off gas Recirculator

Final piping and insulation work was completed on the off gas recirculator test loop. A schematic of the configuration is shown in Figure 56. Shakedown testing of the system was conducted and an initial 30-hour test performed to gather baseline performance data. Figure 57 shows the key process temperature. The stack tester furnace chamber was held at 800 C and the simulated off gas flow rate was held at 11 kg/hr. During a portion of the test, 20 slpm of purge gas (3%H₂/N₂ Balance) was added through the fuel inlet port. The recirculator pressure boost was 10" w.c.

Figure 56: Final piping and insulation work completed on the off-gas recirculator test loop.

Figure 57: Key process temperature

Baffle Design

During testing of the fuel cell module with the metallic, between stack, cathode air baffles the cathode air baffles on adjacent stacks shifted and created a stack to stack short. It was proposed to fabricate these baffles using a mica board of 1/8" thickness to replace the metallic fins, thus eliminating a possible short circuit. A baffle was fabricated and welded (with bracketing) to the standard tube support. This assembly was placed into a furnace and thermally cycled to 850°C every 24 hours for a total of four cycles. After that, the assembly was soaked at 800°C for 100 hours. After testing, the mica was still rigid and securely held to the tube support. This design solves the problem of short-circuiting. More thermal cycling is currently being performed to provide long-term test results. Given the rigidity and integrity of the mica after thermal testing, a design will be produced which eliminates the metal support tube and backing bar. It is anticipated that the 1/8" mica is sufficiently strong to be simply supported at the manifold at one end and sandwiched between the end of cell insulation material at the other end.

Stack Tests

Additional stack tests were conducted to further characterize the performance of the cells and integrated manifolds. Particular attention was paid to acquiring performance data at higher fuel utilizations. Figure 58 shows the fuel utilization for three stacks operating at 75% fuel utilization. These stacks consisted 21 manifolds, 6 cells per manifold and the cells had single ended current take off with ¼” npt braze caps and 3/16”.

Figure 58: Fuel utilization for three stacks operating at 75% fuel utilization

Generator Test

A propane FC module was assembled to further evaluate stack performance on LPG and to evaluate the full-scale LPG pre-reformer and associated piping. The testing will also enabled the LPG generator controls software to be fully debugged and tuned. A CAD model of the pre-reformer and associated piping was developed and the unit was built. The pre-reformer is located within the FC module insulation package adjacent to the recuperator, above the stacks. A “leaf spring” mount, similar to that used for the stacks, provides horizontal stability but the reactor and piping are free to expand vertically.

Generator Manufacture

The manufacture and assembly of a 5 kW generator for shakedown testing of the NETL Fuel Cell Test Facility was carried out. Four stacks, 126 cells per stack, were fabricated and tested along with the gas utility module, control hardware and power electronics. The unit is capable of grid and stand-alone and has undergone integrated factory testing.

Stack Tests

The four stacks for the NETL generator were tested in Stack Tester #1. Each stack was tested for approximately 20 hours at 60 to 90 amps and 60 to 75% fuel utilization. The graph labeled Figure 59 gives the voltage/current points for each stack.

Figure 59:
Voltage/Current Points in Stack Tests

A pressure and flow check was performed on each stack after the stack testing and based on the QC data and stack test data, the stacks were deemed ready for placement into the fuel cell module assembly.

Bundle Test

A 5 manifold, 30 cell assembly was fabricated to determine the performance of double ended current collection cells in a stack configuration. The manifolds had injector tubes with orifices. The manifolds were the blank Ni 200 MIM version (machined in-house). A special endcap was fabricated with current collection features. The endcap was fabricated from braze caps by closing the through-hole with a nickel plug and welding in place. A new hole was drilled and reamed through this nickel plug to accommodate a press-fit silver pin. The silver pin had a hollow section in the outer end for the braid, which was then crimped into place. Exhibit 32 shows the overall bundle ready for testing.

Exhibit 32:
Double End Current Collection Bundle Test

The test failed due to a malfunction in the control system. The unit will be reassembled for a second test.

Chromite Cell Generator Design

A conceptual design of a 180 cell, chromite interconnect, stack configuration was prepared. Each manifold consists of two rows of 10 cells (2 x 10), connected in series along the ten. Since the manifolds hold only 2 rows of cells, current collection assembly

between cells is easily accomplished prior to the assembly of the stack, and manifold-to-manifold connections can be made on the outboard sides of the assembly.

The cell-to-manifold connection for this design must be electrically insulating. This can be performed in two ways. Either the cap, which holds the cell, is electrically insulating or the mechanical connection of the cap to the manifold is electrically insulating. At this stage the design is configured with a metallic braze cap (with parallel thread) and an electrically insulating connection to the manifold. Gaskets, washers and ceramic sleeves are used to isolate the cap from the manifold.

Subtask 3.4 – Manifold and Cap Development

MIM Manifolds and Caps

A first run of MIM threaded braze caps (~50) were received from the manufacturer, Flomet. The brazing fixtures have been modified as necessary to permit brazing of these caps to cells and brazing trials will be conducted next month.

Difficulties experienced with injection molding of the manifolds has been identified to be, in part, a problem in the barrel system of the press used for injection molding of these parts. Additional manifolds will be shot, now that the barrel/injector issues have been resolved.

Deep Drawn End Caps

The deep drawn end caps that were on test for 500 hours as part of a 5 manifold subassembly test were inspected. The end caps were well attached to the cells and in good condition.

Deep Drawn Threaded Braze Cap

A two piece deep drawn braze cap with rolled threads was designed and a quote for this part received from Truex Inc. Based on the preliminary quote it is projected that the cost of a cap using this construction technique could be below \$0.30.

Manifold & Cap Configuration with Chromite Interconnect

The cyclic furnace testing of the magnesia based ceramic cap assemblies continued. After 1080 hours and 48 thermal cycles, the leakage was $<0.84 \text{ mL min}^{-1}$ @ 0.5" w.c. for both samples and approximately 1.0 mL min^{-1} @ 1" w.c. This is considered very low leakage and thus this geometry/materials setup is successful. The figure 3.4.1 below shows the reduction in leakage with increasing number of thermal cycles and time at 838°C.

Figure 60 shows the reduction in leakage with increasing number of thermal cycles and time at 838°C.

Manifold Coatings

Additional hours and thermal cycles were accumulated on the coated 304 and 1019 samples. The type 304 aluminized samples were still stable and the 1010 samples continued to oxidize.

The non-coated 304 and 309 control samples also remained on test. After 1300 hours and 15 thermal cycles, the weight of the 304 sample has decreased and the 309 sample remains stable.

The AL 347HP and AL 20-25+Nb samples also remained on test. After 2064 hours and 86 cycles the AL 347HP had lost 0.0116 g (0.75% of total weight) and the AL 20-25+Nb had gained 0.0024 g (0.11% of total weight).

Injector Design

Due to the inconsistency of the pressure drop through the 1/8" diameter injectors and the need to reduce the overall injector pressure drop, it was decided to use 3/16" injectors with an orifice to restrict the flow. A number of experiments were performed to determine the easiest and most consistent method of manufacturing injectors.

Firstly, lengths of 3/16" Inconel[®] 600 tube were cut and de-burred. Orifices were machined with a drilled and reamed hole size of 0.035", and length of 0.15". After pressure testing 12 of these injectors, a pressure deviation of 2.4" w.c. was observed. A tolerance of ± 0.2 " w.c. was the criteria. It was assumed that the orifices were leaking around the outer edge and this was stopped with RTV. Another pressure test confirmed that there was indeed leakage around the outside but this did not account for all of the pressure difference.

Some existing deep drawn parts (with orifices) were tested at a similar gas flow and it was found that the parts all had a consistent pressure drop. This suggested that if an orifice could be deep drawn or stamped, then this method would be sufficient for manufacturing large quantities of parts.

Another set of injectors and orifices were fabricated to try for repeatable results. More care was taken during the machining process and a larger orifice interference fit was used. The results indicated that a tolerance of ± 0.2 " w.c. was unattainable.

A new method was initiated where an orifice (0.0385") was drilled into the side of the tube (1/2" from the end), with the end of the tube being flattened and welded tight. A number of complete injectors were fabricated and tested in this manner. The results indicated a pressure drop of 6.4" w.c. ± 0.4 " w.c. @ 1647 mL min⁻¹ air-flow for a sample size of 9.

After building one stack and realizing that the orifice could be blocked if it touched the inside of the cell, a modification to the design was implemented. The flattened end of the injector tube was bent approximately 45° in the same direction as the orifice to form a flap that would not allow the orifice to hit the inside of the cell tube wall. It was determined that a gap of 0.050" was enough to eliminate any orifice flow restriction.

Thus, a method of manufacturing injectors with a tolerance of ± 0.4 " w.c. was conceived. To get this tolerance down to ± 0.2 " w.c, each injector was placed into the pressure testing apparatus and the orifice was modified with a hand tool until it was within specification. Obviously, this method is time consuming, but the outcome was within the tightest tolerance.

In the near future, a new method must be developed to allow injectors/orifices to be fabricated automatically without the customization that is necessary with the current method.

A stamped orifice was designed, which would allow an orifice to be press-fitted into the end of an injector tube, thus simplifying assembly. A quote was obtained which showed the cost to be approximately \$0.05 each (for a quantity of 500,000 pieces). Figure 61 shows the general design for a stamped orifice.

Stamped orifice design.

Figure 61: General design for a stamped orifice

A stack with the new, radial hole, injector geometry was built and tested. Figure 62 shows the average injector pressure drop for the radial hole injector along with the pressure drop across the 1/8" OD – 0.035" wall injector. Also shown on the graph is the pressure drop of an injector with a 0.041" machined orifice. It is important to note that with this low injector pressure, flow distribution to the cells throughout the stack height was uniform as indicated by manifold voltages.

Figure 62: The average injector pressure drop for the radial hole injector along with the pressure drop across the 1/8" OD – 0.035" wall injector

MIM Manifolds and Caps

Brazing trials were conducted on the MIM braze caps. Successful results were obtained.

Twenty manifolds have been processed through sintering and have been sent out by Flomet for final machining. Two blanks have been inspected by Acumentrics and should be acceptable. Their final machining of these manifolds will be done by the Acumentrics in-house shop for comparison to the outside vendor.

Manifold & Cap Configuration with Chromite Interconnect

The cyclic furnace testing of the magnesia based ceramic cap assemblies continued. After 1488 hours and 65 thermal cycles, the leakage was $<0.84 \text{ mL min}^{-1}$ @ 0.5" w.c. for both samples and approximately 1.8 mL min^{-1} @ 1" w.c. This is considered very low leakage and thus this geometry/materials setup is successful. At approximately 900 hours, the gas composition was changed from argon/3% hydrogen to nitrogen/3% hydrogen. The leakage at 1" w.c. immediately dropped to 1 mL min^{-1} . At 1344 hours, the gas was changed back to the original composition and the leakage went back up to where it was previously. Figure 63 shows the leakage with increasing number of thermal cycles and time at 838°C.

Figure 63: Sample leakage after thermal cycling to 838°C every 24 hours. Leakage data was taken at room temperature.

Work has commenced on incorporating the chromite interconnect cells into a stack design. Since the manifold no longer needs to carry current, a fuel delivery/off gas collection arrangement using a common manifold assembly is possible. It has been proposed to connect 2 cells in parallel within a stack. For ease of assembly, it is most likely necessary to develop a manifold that consists of two rows of cells that would allow the current collection connections to be constructed. Figure 64 shows the latest iteration of a stamped stack design, which utilizes 2-cell manifolds. This particular design is a complete 5 kW unit, using 120 x 50 W cells. Request for quotations were sent to three stamping companies to determine if this is an economically viable option for stack manufacturing.

Figure 64: 5 kW unit assembled from stamped manifolds.

When using a common manifold stack design, the injectors must be electrically isolated from the cells. A number of different ways to isolate the injectors were realized. An alternative configuration to forming a bulge would be to stamp enough of a crease into the tube to form a barrier for the spacer. This would be a more economical method of fabrication.

Figure 65

Manifold Coatings

The final aluminized Type 304 (VaporKote®) sample is on test (total of 2028 hours and 24 thermal cycles). The Type 304 sample gained a total of 0.0410 g (0.12% of total weight). These results agree with the initial aluminized samples in that aluminized Type 304 can withstand the rigors of the stack operating conditions (oxidizing atmosphere). The experiment has been terminated.

Figure 66: Graph of weight of pack aluminized samples after 1308 hours at 816°C with thermal cycling to room temperature every 24 hours

A set of control samples is also being tested. These consist of coupons of Type 304 and Type 309. After 1600 hours and 16 thermal cycles, the weight of the Type 304 sample has decreased and the Type 309 has stabilized, under the oxidizing conditions. Figure 67 shows the initial high rate of weight gain (oxide layer formation) and the slow weight decrease of the Type 304 sample.

Figure 67: Weight change of the Type 304 and Type 309 over time at 838°C, with thermal cycling every 24 hours.

After 2028 hours, the aluminized Type 304 sample was still stable with respect to oxidation resistance at 815°C. These results show that the process of pack aluminizing forms a coating onto Type 304 that can withstand stack operating conditions.

The Type 309 (non-aluminized) sample also seemed to be stable. This is evidence to suggest that a manifold fabricated from this material will withstand the stack operating conditions. However, the actual atmosphere within the stack is different (fuel-rich and also high steam concentration), thus long term stack testing is required for a definitive answer.

After 2472 hours and 103 thermal cycles the AL 347HP had lost 0.0136 g (0.88% of total weight) and the AL 20-25+Nb had gained 0.0015 g (0.07% of total weight). At this stage of the experiment, it seems that the AL 347HP is slowly losing weight and the AL 20-25+Nb is relatively stable. This experiment will continue for at least 2500 hours. Experiments using the fuel and exhaust gases from the stack must be implemented to ensure longevity of the materials.

Figure 68: Weight of Allegheny Ludlum alloys with thermal cycling every 24 hours to 838°C in air

MIM Manifolds and Caps

New braze cap furnace furniture was used to reduce the amount of distortion of the braze cap part during sintering. The furniture allows the part to be fired with the threads up and will reduce the amount of post sintering treatment required to bring the parts within specification.

The final machining of three manifolds was carried out by Acumentrics' in-house shop confirming that the manifolds can be drilled, tapped and machined to final configuration without problems. These manifolds have been installed in a stack subassembly to evaluate their performance under generator conditions.

Post-sinter machining of manifolds by an outside machine shop has proceeded but at a slower pace than earlier planned. Additional tooling and a thread tapping mill was ordered.

108 manifolds were processed through sintering and are available for post-sinter processing. Evaluation of outside shop first article samples will be made prior to further work on the 108 manifolds.

Manifold & Cap Configuration with Chromite Interconnect

The cyclic furnace testing of the magnesia based ceramic cap assemblies continued. After 1872 hours and 81 thermal cycles, the leakage was $<0.84 \text{ mL min}^{-1}$ @ 0.5" w.c. for both samples and approximately 2.0 mL min^{-1} @ 1" w.c. This is considered very low leakage and thus this geometry/materials setup is successful. Figure 69 shows the leakage with increasing number of thermal cycles and time at 838°C.

Figure 69: Sample leakage after thermal cycling to 838°C every 24 hours. Leakage data was taken at room temperature.

Discussions were held with a stamping manufacturer to improve the manufacturability of the stamped manifold design presented in the previous monthly. A number of changes were made to the designs and a quote will be furnished within the next two weeks. Calculations were made to determine the bolt hole spacing and flange dimensions for proper manifold sealing using mica gaskets.

MIM Manifolds and Caps

MIM manifolds accumulated 160 hours of operation in a 5 manifold bundle test and inspection of the manifolds did not show any deleterious effects from being at temperature in the fuel cell environments. There was also no performance degradation of the bundle attributed to the manifolds. A photo of one of the manifolds is shown below in Exhibit 33.

Exhibit 33: MIM manifold

Flomet has received the post-sinter machined manifolds from their outside machine shop and the manifolds are under going QC. They will be sent to Acumentrics for inspection and evaluation.

92 MIM manifold blanks were received from Flomet and are being readied for machining by Acumentrics' internal machine shop. Approximately 10 percent of the manifolds have some "material separation" at the fillets. These "material separations" occur in regions of high injection shear and result from a separation of the metal particles from the binder during injection. The separations that appear as cracks are being evaluated further. Tooling modifications to increase the fillet radius will be made to reduce the high shear zones.

Manifold & Cap Configuration with Chromite Interconnect

The cyclic furnace testing of the magnesia based ceramic cap assemblies continued. After 2640 hours and 111 thermal cycles, the leakage was $<0.84 \text{ mL min}^{-1}$ @ 0.5" w.c. for both samples and approximately $2\text{-}3 \text{ mL min}^{-1}$ @ 1" w.c. This is considered very low leakage and thus this geometry/materials setup is successful. Figure 70 shows the leakage with increasing number of thermal cycles and time at 838°C.

Figure 70: Sample leakage after thermal cycling to 838°C every 24 hours. Leakage data was taken at room temperature.

End Cap Design (Current Collection)

As a fall back position from the chromite cell being developed, the incorporation of current collection at both ends of the baseline cells is being pursued. Cell testing has clearly shown that a significant improvement in cell power can be realized by tapping both ends of the cells. Since the endcaps currently utilized are deep drawn, thin wall caps, a modification to these caps or a new cap design is required to allow end of cell current collection. A number of designs have been detailed.

For immediate testing of the double ended current collection configuration, modifications have been made to MIM threaded braze caps to allow their use as an end cap. As shown in the Figure 71, the NPT thread portion of the cap is modified to receive an internal insert that is drilled and reamed to accept a silver pin. Thirty of these modified caps have been made and will be utilized in a 30 cell, 5 manifold bundle test.

Figures 71 (a) & (b): Solid models of modified braze caps to accept a silver pin for current collection.

A machined design consisting of two parts that form the annulus of the brazing cavity could also be utilized. In the center insert, a hole was drilled and reamed to accept a silver pin originating from the silver braid of the series connected cell. The silver pin would have a hollow section in which the silver braid could be inserted and crimped tight. Figure 72 illustrates this concept.

Figure 72: Solid model of the screw-machined version of a current collecting end cap

The long-term solution is to manufacture a new deep drawn part that has an extra feature to allow the silver pin to press into the end, thus providing a cheaply manufactured part that is capable of current collection. Figure 73 illustrates a possible configuration with silver pin inserted.

Figure 73 Deep drawn end cap with extra feature to allow a silver pin to be pressed into it

Electrically Insulating Cell to Manifold Connection

Along with the development of a non-conducting braze cap configuration, alternative methods of attaching and electrically isolating the cell from the manifold are being pursued. Cell attachment configurations, which allow the metallic braze cap to be isolated from the manifold using insulating washers and sleeves have been detailed. Additional versions of a cemented ceramic cap with a metallic attachment feature have also been detailed.

One of the major problems to overcome with the use of an interconnection is the elimination of any current path at the end of the cell. Currently, a highly conductive braze joint is used that allows current conduction as well as forms a tight gas seal. For generator designs with the interconnection connection, a non-conductive seal is desired at the gas manifold junction. To that end, a number of experiments have been tried.

A number of setups have been attempted, including the use of alumina powder, insulation, and zirconia powders as fillers with Cu washer sealing; the double-dipping of the ends into ZrO_2 slurry coating, and then brazing, and the use of alumina/phosphate cements as an end seal with Cu sealing along the EL on the side of the cell. While the cement and alumina powder have shown some promise, no technique has, to date, been completely or consistently successful. Some of this inconsistency has been due to application technique

(i.e. leaks have been observed along sections where excess cement was accidentally applied to the cup or cell surface, not allowing a tight gas seal between the cup, Cu, and EL.). Technique is currently being corrected to maintain a consistent application of materials for the next round of testing.

MIM Manifolds and Caps

92 MIM manifold blanks were received from the manufacturer along with 10 machined manifolds. A visual inspection was performed on the manifolds and a list of recommendations was generated, for implementation for future manifold orders. Following are items, which will be addressed in future machined manifolds:

1. There was a lip on the back edge of the current collection groove. This must be removed by extending the width of the current collection groove by 0.005”.
2. There was a burr on the back of the NPT thread. This must be removed.
3. The NPT thread was not deep enough. Cut to the depth indicated by the 1/4” NPT gauge (ie. a typical 1/4” NPT thread depth).

In general, the manifolds did not meet the flatness requirements originally set for the part but it was found that the flatness could be improved by compressing each manifold in a hydraulic press. To determine if adequate sealing of the fuel and offgas chambers could be obtained, six MIM manifolds were prepared with current collection grooves for stacking into a 6-manifold ‘mini-stack’. Compression plates and rods were fabricated in order to complete the stack. A number of ‘blank’ brazing caps were fabricated to block off the 1/4” NPT cell holes in the front face of the manifolds. Silver tabs were fabricated from silver sheet.

The individual manifolds were leak checked to ensure that there was no leakage either through the braze caps of the 1/4” NPT threads. The stack was assembled with the same methods used to build a full size unit. No ceramic paint was used to seal the ends of the silver tab to the manifold.

The stack was torqued to 300 lb-in before performing a leak check. The unit was then placed into a furnace and thermally cycled to 800°C with a dwell of 6 hours and a ramp rate of 10°C min⁻¹.

Exhibit 34: Stack of MIM manifolds

Individual manifolds had a leakage of $<0.84 \text{ mL min}^{-1}$ @ 10" w.c. This was considered to be below the measuring limit of the rotameter (Omega 25.3 mL min^{-1}).

The stack of manifolds before thermal cycling had a leakage of $<0.84 \text{ mL min}^{-1}$ @ 10" w.c. After the thermal cycling and retorquing to 300 lb-in, the leakage was:

Pressure	2"	3"	4"	5"	6"	"w.c.
Leakage	9.38	12.4	15.8	20.8	23.6	mL min^{-1}
Leakage	0.26	0.35	0.44	0.58	0.65	mL min^{-1} cell^{-1}

Table13: Leakage Data

This is much tighter than a typical full size stack, which would leak approximately $10 \text{ mL min}^{-1} \text{ cell}^{-1}$.

The current collection grooves varied in depth from 0.017" to 0.024". Gasket thickness after thermal cycling ranged from 0.047" to 0.055"

The manifolds were also measured for 'flatness' by measuring the amount of movement front-to-back and left-to-right (i.e. the amount of rock). Two of the manifolds had displacements of 0.006". After thermal cycling, the displacement had been reduced to 0.001" and 0.003". This suggests that the combined heat and compression actually flattens the manifolds.

One of the blank braze caps that was screwed into the manifold caused the front wall to bulge (increased height). Thus the manifold was not flat after this operation. This did not affect the sealing ability of the mica gaskets. The difference in height caused by the bulge was 0.008”.

Manifold & Cap Configuration with Chromite Interconnect

The cyclic furnace testing of the magnesia based ceramic cap assemblies continued. After 2976 hours and 125 thermal cycles, the leakage was $<0.84 \text{ mL min}^{-1}$ @ 0.5”w.c. and approximately 2-3 mL min^{-1} @ 1”w.c., for both samples. This is considered a low leakage and thus this geometry/materials setup is successful. At approximately 900 hours, the gas composition was changed from argon/3% hydrogen to nitrogen/3% hydrogen. The leakage at 1”w.c. immediately dropped to 1 mL min^{-1} . At 1344 hours, the gas was changed back to the original composition and the leakage went back up to where it was previously. Figure 21 shows the leakage with increasing number of thermal cycles and time at 838°C.

Figure 74: Sample leakage after thermal cycling to 838°C

Currently experiments are being conducted to develop and isolate braze cap for use with an interconnection on the Acumentrics fuel cell. Such cup will allow for the development of a simple manifold system, or the use of different geometries within the hot box. These experiments have been isolated on the testing of pastes and powders for isolation of the seal, as well as cup modification and the brazing of oxidized, rather than reduced cells. Results of these experiments so far indicate that a sealed, electrically

isolated braze joint may consist of a Zr paste, Cu washer for sealing, and a modified cup that does not have an inner wall. Results will be compiled and detailed in the next period.

MIM Manifolds and Caps

As reported previously, four complete stacks using MIM manifolds were built and tested. Manifolds were final-machined in Acumentrics' in-house shop involving threading of the cell hole and machining of the current collection groove. Exhibits 35 and 36 show the manifolds before and after final machining. An important indicator of the overall acceptability of the MIM manifolds is the leakage data of the stacks. Figures 75 and 76 present the leakage data for stacks assembled with plate stock/machined manifolds and MIM manifolds before and after performance testing. Although the average leakage for the MIM manifolds is higher than the machined manifolds, these leakage levels have been found to be acceptable in the past. It is believed that the extra leakage is a combination of the thinner wall and therefore reduced gasket seating surface of the MIM manifolds and the uneven current collection surface.

Exhibits 35 and 36: MIM Manifolds Before and After Machining

Figure 75: Manifold Leakage Data Prior to Performance Testing

Figure 76: Manifold Leakage Data After Performance Testing

Manifold & Cap Configuration with Chromite Interconnect

To take full advantage of the current collection geometry afforded by chromite-banded cells require an electrically insulated cell-to-manifold connection. With an electrically isolated cell-to-manifold connection, the fuel plenum can potentially be a single chamber, stamped housing, greatly reducing the cost of the fuel manifold. One option is to fabricate an electrically insulating cell-to-braze cap connection. The braze cap can then be threaded or press fit into the fuel plenum without any additional isolation features. Three

cells were cemented into standard 1/4" NPT and 1/16" NPT braze caps, leak checked, thermally cycled and cell tested. The first was a standard 1/4" NPT cap in the reduced condition. The second was a 1/16" NPT cap with slots cut in the outside for anchoring the cement and the third was an oxidized 1/4" NPT braze cap with anchoring slots. The deep drawn endcaps were also cemented into place.

The other end of the cells were also soaked in water before inserting into the 1/2 filled deep drawn braze caps.

Exhibit 37: Three cells after cementing caps in place

Exhibit 38: Details of the cemented caps for Cell #1 (C150418)

Then, enlarged for more detail

Exhibit 39: Details of the cemented caps for Cell #2 (C150400)
Then enlarged for more detail.

Exhibit 40: Details of the cemented caps for Cell #3 (C150383)
Then enlarged for more detail.

Once the cells had been cured, the current collection was assembled. Three different methods were used. Cell #1 used the twisted and flattened braid pressed onto the chromite band with a spring clip. Cell #2 used a silver tab and spring clip pressed onto the chromite band. Cell #3 used a silver butt splice connector crimping the braid from the chromite band and the braid from the current collection together. All cells had a silver wire winding over the chromite band.

Exhibit 41: Current collection methods used to connect the cells to the test fixture

After the cementing and curing at 70°C overnight, the cells were leak tested, both with the cell leak tester (vacuum) and the flow tester (2" w.c. pressure).

When compared to the standard metallic braze joint, the leakage of the cemented cells was very high. A typical leak rate value for a standard braze joint is less than 50 torr min⁻¹. Table 15 describes the leak rates obtained from the cemented cells. Also included are the flow test numbers.

Cell Number	Leak Rate	Flow Rate	Pressure
C150418	1600 torr min ⁻¹	<0.8 mL min ⁻¹	0.5" w.c.
C150418	1601 torr min ⁻¹	0.8 mL min ⁻¹	1.0" w.c.
C150418	1579 torr min ⁻¹	3.0 mL min ⁻¹	1.5" w.c.
C150418		5.7 mL min ⁻¹	2.0" w.c.
C150400	1441 torr min ⁻¹	<0.8 mL min ⁻¹	0.5" w.c.
C150400	1439 torr min ⁻¹	9.1 mL min ⁻¹	1.0" w.c.
C150400		13.8 mL min ⁻¹	1.5" w.c.
C150400		16.0 mL min ⁻¹	2.0" w.c.
C150383	697 torr min ⁻¹	<0.8 mL min ⁻¹	0.5" w.c.
C150383	681 torr min ⁻¹	<0.8 mL min ⁻¹	1.0" w.c.
C150383		<0.8 mL min ⁻¹	1.5" w.c.
C150383		<0.8 mL min ⁻¹	2.0" w.c.

Table 14: Leak Testing of the Cemented Cells

At the off gas side of the manifold, the typical operating pressure was approximately 0.7" w.c. with a maximum of approximately 1.5" w.c. at maximum flow. This suggested that Cells C150418 & C150383 were leak tight enough for use in a stack. Cell C150400 showed a high leakage of 9.1 mL min⁻¹ at 1" w.c. It is unknown what the maximum limit to the off gas leakage is at this stage.

These cells have been submitted for cell testing to evaluate the connection method.

The cyclic furnace testing of the magnesia based ceramic cap assemblies continued. After 3504 hours and 147 thermal cycles, the leakage was $<0.84 \text{ mL min}^{-1}$ @ 0.5" w.c. and 1.7 mL min^{-1} and 3.0 mL min^{-1} @ 1" w.c., for both samples. This is considered a low leakage and thus this geometry/materials setup is successful. At 1344 hours, the gas was changed back to the original composition and the leakage went back up to where it was previously. Figure 77 shows the leakage with increasing number of thermal cycles and time at 838°C

Figure 77: Sample leakage after thermal cycling to 838°C every 24 hours.
Leakage data taken at room temperature

Task 4.0 – Fabrication and Processing Technology Development

Subtask 4.1 – Tube Drying

Microwave energy has been evaluated for the post extrusion drying process in addition to de-binding in the same unit. Initial trials have been undertaken where Acumentrics extruded fresh tubes, placed them in standard drying holders and then wrapped the entire

holder with plastic wrap to minimize evaporation. The tubes were then transported to a vendor for microwave drying and de-binding trials.

At the vendor, sections of the wet tubes were taken out of the drying holders and placed in the same alumina kiln furniture used for de-binding trials. The tubes were first brought up to 85°C and were then held for two hours to allow drying to take place. The drying schedule was programmed to go into a heating rate profile similar to the one previously used to successfully de-bind green tubes before bisque firing. See Figure 78.

Figure 78: Microwave tube drying and de-binding attempt.

The profile above shows that during the first hold at 150°C of the de-binding phase, the tube surface temperature significantly increased due to the exothermic reaction of the binder burning off. This type of temperature spike inherently leads to tube cracking and as such, this test was ended prematurely.

Further combined microwave drying/de-binding trials will be completed when a more powerful microwave unit becomes available. This unit will also have air blowers to aid in the removal of degradation products during binder removal, which will reduce the propensity for cracking during binder removal.

Subtask 4.2 – High Volume Tube Firing

A number of conclusions were drawn concerning the microwave de-binding of Acumentrics green tubes, some of which include:

- Microwave de-binding cuts the processing time from 13 hours to 3.75 hours, a time saving of 9.25 hours.
- The total time to microwave de-bind and belt bisque fire tubes is approximately 10 hours compared to the current 44 hours required when using Acumentrics standard production furnace. This time can be reduced even more if a microwave unit is placed in line with the belt furnace, thus allowing for the bisque firing to commence right after the tube has been de-binded.
- Alumina furniture is suitable for use in both the microwave de-binding step and the belt bisque firing stage.

From this work, it has been possible to fully fabricate one cell, which went through the microwave de-binding step during the bisque firing process, thus reducing the bisque firing time from 44hrs to approximately 10 hours. This cell has since been tested for performance.

Table 15 below shows a summary of the cell test data collected after approximately 578 hours on test.

<u>Tube Num.</u>	<u>Peak Voltage (mV)</u>	<u>End Voltage (mV)</u>	<u>IR (mΩ)</u>	<u>OCP (V)</u>	<u>Time on Test (hr)</u>	<u>Thermal Cycles</u>	<u>Fuel Utiliz.</u>	<u>Degradation per 1000 hrs</u>
B390277	666	574	1.369	1.063	578	1	75%	-24%

Table 15: Summary of the cell test data collected after approximately 578 hours on test

The cell performed on a par with conventionally processed cells in terms of potential, showing a peak of 666mV. At 24%, the degradation observed for the cell is however much greater than that observed for cells processed in a standard way. An explanation for this enhanced degradation at this stage is unknown. Further testing of other microwave de-binded cells must therefore take place to determine if the enhanced degradation is due to microwave de-binding of the tube or something else.

Figure 79: Cell voltage versus the time on test for cell B390277

The IR for the microwave de-bound cell was also measured during cell testing. The IR values are plotted in Figure 80 and are increasing with time, as would be expected for a degrading cell.

Figure 80: The IR values are plotted

Task 5.0 – Fuel Processing Technology Development

Subtask 5.1 – Reforming Technology-Light Hydrocarbons

Propane CPOX Reactor

A larger diameter propane CPOX reactor was designed and built to permit testing at higher propane flows and to evaluate scale up issues. The reactor has an outer diameter of x inches and an overall height of y inches. Exhibit 42 shows the reactor geometry. The test stand was also modified to reduce the backpressure in the reactor outlet piping and to add additional cooling air to the furnace chamber where the reactor products are oxidized.

Exhibit 42: The reactor geometry

The Propane CPOX test ran for 618 hours without any outward signs of deactivation. The O/C ration increased almost linearly over the test from ~1.1 to 1.25.

Approximately 15 LPM of LPG is required to provide fuel for a 5kW system via the CPOX route (O/C=1.2). The maximum flow of LPG in the test was 12.3 LPM. The LPG flow rate was limited by the air the blower and system backpressure ($\sim 10''$ W.C.).

Figure 81: Testing of a 5kW LPG CPOX reactor

The pressured drop across the reactor itself is shown in **Figure 82**. The pressure does not correlate exactly with the gas flow rates due to a drift in the calibration of the airflow sensor. This can also be seen in **Figure 83** where both the temperature of the inlet and the O/C ratio are plotted against time. As expected, temperature and O/C ratio are well correlated; above the stoichiometric ratio for syngas extra oxygen results in exothermic total oxidation. Increases in the O/C have a double positive effect on the pressure drop on account of the increased airflow and increased temperature. Propane conversion was 100% throughout most of the test, and hydrogen and carbon monoxide selectivities, as in previous tests, were between 80 and 90%. In the case of high flow rates (12.4 LPM) and very low O/C (1.02), a small amount of propane (0.1 vol%) appeared in the outlet GC trace ($>98\%$ conversion) probably because of the lower reactor temperature.

Two more important points should be drawn from the figures: (1) there is no evidence of reactor plugging even at the low O/C ratio (~ 1) in the last 24 hours of the test (2) bed temperatures are very high ($\sim 1050^\circ\text{C}$) at O/C=1.2

Figure 82: Pressure drop across the CPOX reactor (as measured at sample points)

Figure 83: Variations in temperature and O/C ratio over the test time

Since catalyst (and reactor materials) lifetime is significantly reduced at high temperatures, it is preferable that the reactor operates at as low O/C as possible. The final

24 hours of the test was conducted at an O/C only slightly greater than 1.0 resulting in a low reactor temperature of $\sim 850^{\circ}\text{C}$. Despite the higher propensity for carbon formation no outward sign of activity degradation was observed. After the reactor was cooled on 3% H_2/N_2 the reactor contents were inspected (Exhibit 43) and no significant evidence of carbon deposition was found on the catalyst.

Exhibit 43: (a) bottom of the igniter, (b) top of catalyst bed, (c) catalyst sample

A propane reactor was then installed on a stack tester to enable integrated operation of a stack on reformed LPG fuel. The reactor diameter was scaled down but contained the same catalyst support, catalyst depth, burner geometry, and thermocouple arrangement as a full-scale unit. The reactor contained catalyst with alumina on top. The reactor was connected to the inlet flange of the stacks tested via a $\frac{1}{2}$ " interconnect tube. The stacks each contained a standard POX reactor, as utilized on natural gas generators. The table below gives the inlet and outlet reactor gas compositions for two sets of gas samples. As with the earlier reactor testing, complete conversion of the propane was achieved in the reactor. Table 16 shows the temperature trace of the reactor for a typical stack test followed by a graph (Figure 84) of the initial light off period for the reactor. A hot surface igniter is used to light-off a fuel lean air/fuel mixture to bring the catalyst above the ignition temperature. Once the correct catalyst temperature is achieved the air and fuel flows are restarted at the correct O/C ratio.

	Reactor Inlet 12:24	Reactor Outlet 12:24	Reactor Inlet 15:05	Reactor Outlet 15:05	Stack Offgas 16:00
Anode Air (lpm)	16.5		16.5		
Anode Fuel (lpm)	1.96		1.93		
O/C GC	1.200		1.249		
Concentration %					
CH ₄	0.00%	0.84%	0.00%	1.68%	0.00%
H ₂	0.00%	24.04%	0.00%	24.31%	3.32%
H ₂ O	0.00%	1.96%	0.00%	1.73%	16.33%
CO	0.00%	17.82%	0.00%	17.22%	0.15%
CO ₂	0.03%	2.79%	0.07%	3.16%	20.16%
O ₂	19.27%	1.30%	19.29%	0.92%	1.25%
N ₂	70.00%	51.25%	70.37%	50.98%	58.79%
C	0.00%	0.00%	0.00%	0.00%	0.00%
C ₃ H ₈	10.70%	0.00%	10.27%	0.00%	0.00%
C ₂ H ₆	0.00%	0.00%	0.00%	0.00%	0.00%

Table 16: Temperature trace of the reactor for a typical stack test

Figure 84: The initial light off period for the reactor

Subtask 5.2 – Light fuel desulphurization

Design of an experimental desulfurizer based on the BASF adsorbents is shown in Figure 85

Figure 85: Catalyst packing

The current scheme relies on three adsorbents.

- 1) Activated carbon (for removal of thiophenes and mercaptans)
- 2) Mixed metal oxide for COS and mercaptan removal
- 3) Mixed metal oxide for polishing

Englehard and US Filter were approached for details on their mixed metal adsorbent and activated carbon, respectively. Based on information from US Filter, the activated carbon they initially supplied for use in generators is not optimal for the application. Details on an appropriate carbon should be sent to Acumentrics shortly.

Englehard had a mixed metal oxide based on Ni for low temperature adsorption of sulfur (CNG-1), as well as a selective oxidation catalyst/adsorbent, which operates at 200°C. Technical details should be forthcoming.

US Filter has now confirmed that as far as US Filter products are concerned, the coconut shell based carbon VC48C, is the best product for removing sulphur from LPG, despite the availability of caustic impregnated carbons. The caustic impregnated carbons convert mercaptans to their disulfides provided oxygen is present in the gas stream. The disulfides are more strongly adsorbed by the activated carbon, making them easier to remove. The caustic impregnated carbons however do not change the thiophane (suspected to be the major contaminant in LPG) and dimethyl sulfide and must adsorb them if they are to be controlled. In that the impregnated carbon has a significant portion of its pore space occupied by the impregnant; it does a very poor job of adsorbing the thiophane and DMS. The higher surface area coconut shell based carbons do a better job of adsorbing these contaminants. However quantitative estimates of the capacity cannot be made for the LPG application on account of the competitive adsorption of propane and higher components for the active sites for which they have no experimental data. The observation of the warming of the activated carbon bed when LPG is introduced is further evidence for hydrocarbon adsorption.

List of Acronyms and Abbreviations

AC	Alternating Current
ANSI	American National Standards Institute
ASCII	American Standard Code for Information Interchange
BOM	Bill of Material
BOP	Balance Of Plant
CAD	Computer Aided Design
CAN	Controller Area Network
CM	Common Mode
CPOX	Catalytic Partial Oxidation
DC	Direct Current
DSP	Digital Signal Processor
DTA	Differential Thermal Analysis
EEPROM	Electrically Erasable Programmable Read-Only Memory
EMC	ElectroMagnetic Compatibility
ESTOP	Emergency Stop
FCIC	Fuel Cell Interface Converter
FCS	Fuel Cell Stack
FCUPS	Fuel Cell Uninterruptible Power Supply interface
FET	Field Effect Transistor
FU	Fuel Utilization
GUM	Gas Utility Module
HMI	Human-Machine Interface
IC	Investment Casting
I/O	Input / Output
IR	InfraRed
LED	Light Emitting Diode
lpm	Liters per minute
MIM	Metal Injection Molding
mlpm	Milliliters per minute
MOR	Modulus of Rupture
MOSFET	Metal Oxide Semiconductor Field Effect Transistor
NPT	National Pipe Thread
NYSERDA	New York State Energy Research and Development Authority
PC	Personal Computer
PCB	Printed Circuit Board
P&ID	Piping and Instrumentation Diagram
PID	Proportional Integral Differential
POX	Partial Oxidation
PLC	Programmable Logic Controller
PWM	Pulse Width Modulation
ROM	Read Only Memory
RPM	Revolutions Per Minute
RPTS	Rapid Prototype Test Station
SEM	Scanning Electron Microscope

SiC	Silicon Carbide
SOFC	Solid Oxide Fuel Cell
SPI	Serial Peripheral Interface
TEC	Thermal Expansion Coefficient
TGA	Thermal Gravimetric Analysis
UPS	Uninterruptible Power Supply
TI	Texas Instruments
w.c.	Inches water column
YSZ	Yttria Stabilized Zirconia