

10 kW SOFC Power System Commercialization  
SECA Semi-Annual Report 41244R08

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# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

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# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## Abstract / Summary

Cummins Power Generation (CPG) as the prime contractor and SOFCo-EFS Holdings LLC (SOFCo), as their subcontractor, teamed under the Solid-state Energy Conversion Alliance (SECA) program to develop 3-10kW solid oxide fuel cell systems for use in recreational vehicles, commercial work trucks and stand-by telecommunications applications. The program goal is demonstration of power systems that meet commercial performance requirements and can be produced in volume at a cost of \$400/kW. This report summarizes the team's activities during the seventh six-month period (July-December 2005) of the four-year Phase I effort.

### **Summary of progress and future direction**

While there has been significant progress in the development of the SOFC subsystems that can support meeting the program Phase 1 goals, the SOFCo ceramic stack technology has progressed significantly slower than plan and CPG consider it unlikely that the systemic problems encountered will be overcome in the near term. SOFCo has struggled with a series of problems associated with inconsistent manufacturing, inadequate cell performance, and the achievement of consistent, durable, low resistance inter-cell connections with reduced or no precious materials. A myriad of factors have contributed to these problems, but the fact remains that progress has not kept pace with the SECA program. A contributing factor in SOFCo's technical difficulties is attributed to their significantly below plan industry cost share spending over the last four years. This has resulted in a much smaller SOFC stack development program, has contributed to SOFCo not being able to aggressively resolve core issues, and clouds their ability to continue into a commercialization phase.

In view of this situation, CPG has conducted an independent assessment of the state-of-the-art in planar SOFC's stacks and have concluded that alternative technology exists offering the specific performance, durability, and low cost needed to meet the SECA objectives. We have further concluded that there is insufficient evidence to reliably predict that SOFCo will be able to achieve the SECA performance and cost goals on a schedule consistent with SECA or CPG commercialization goals. CPG believes SOFCo have made a good faith effort consistent with the available resources, but have repeatedly fallen short of achieving the programs scheduled targets.

CPG has therefore initiated a process of application for extension of Phase 1 of our SECA program with the intent of transitioning to an alternative stack supplier with more mature SOFC technology, and demonstrating a system meeting the SECA Phase 1 goals by the end of calendar 2006. We have identified an alternative supplier and will be reporting the progress on transition and program planning in monthly technical reports, reviews, and in the next semi-annual report.

### **Performance against SECA Objectives:**

The following table summarizes performance achieved against objectives. A major shortfall is the achievement of target cost value for stacks, and the associated cost impact on the SOFC module. The stack cost miss is driven by a combination of higher-than-target ASR (thus requiring additional stack material to meet power requirements) and failure to successfully replace precious metals at the cathode to interconnect junction.

SOFCo made significant progress against degradation targets during the reporting period, but repeatable degradation progress is cross-linked to replacement of precious metals at the cathode to interconnect junction, which remains to be accomplished. A further concern is stability of the interconnection junctions under transients, which remains to be explored

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

following the material set replacements for precious metals. This is considered a high risk area due to the ridged nature of the ceramic/ via assembly.

### Total System Cost and Performance Status

Requirement	Criteria	C2 Prototype (Ph 1 Deliverable)	Comments
<b>Cost</b>	\$800/kW (mfg cost)	<b>\$2,133/kW (Best Projection is \$1683/kW)</b>	See Cost Status table for details
<b>Efficiency</b>	25% (mobile)	<b>&gt;25%</b>	Aspen system analysis
<b>Power Degradation</b>	<2% per 500 hrs at constant voltage	<b>A 67-cell stack was tested for 400 hours, and showed increasing power over the test period. Stack behavior was similar to recent short stacks.</b>	Two short stacks operated on reformed natural gas during the reporting period showed degradation rates ~2% / 500 hours. In addition, a 68-cell stack showed performance similar to recent short stacks; output increased over the 350 hour test period.
<b>Availability</b>	80% for 1,500 hr test	<b>On target to achieve</b>	Multiple 1,000 hr stack tests performed

### Component Systems Cost Status

System Item	Criteria	C2 Prototype (Ph 1 Deliverable)	Comments
<b>Stacks</b>	\$340/kW	<b>\$1,750/kW Best projection for deliverable SOFC module is \$1,300/kW</b>	Projected stack cost \$450/kW based on reduction of ASR from 0.6 to 0.5 $\Omega$ -cm <sup>2</sup> , and elimination of the high cost air-side bump material. Design changes to increase active area required to further reduce cost.
<b>Fuel Processor</b>	\$60/kW	<b>\$27/kW</b>	Successful development of a low-cost, waterless CPOX reformer.
<b>SOFC Module</b>	\$185/kW	<b>\$405/kW</b>	The C2 as-built cost is above the target due to the 4-stack implementation. The advanced power module design meets the Phase I target.
<b>Balance of Plant</b>	\$48/kW	<b>\$64/kW (same ↔)</b>	Air blower system, fuel supply system, plumbing, base, housing
<b>Controls &amp; Power Electronics</b>	\$146/kW	<b>\$107/kW (same ↔)</b>	Including control for fuel cell system, DC power conditioning, and DC/AC Inverter.
<b>Total</b>	\$779/kW	<b>\$2,133/kW (Best Projection is \$1683/kW)</b>	Costs are nominal and best projected values, not Crystal Ball distributions.

### Cell, stack, reformer, system, manufacturing:

#### Cell Evaluation

During 2005, SOFCo experienced a number of cell quality related issues which adversely impacted stack development efforts. In particular, electrolyte strength fell below required minimum values as a result of an apparent problem with the tape casting process. SOFCo worked closely with its cell supplier to overcome the electrolyte strength issue, and shipment of acceptable cells resumed in July. Two key outcomes of single cell tests performed in the course of the resolution process were the development of heat-up/pre-conditioning processes for the cells that were compatible with the stack assembly procedure and, more importantly, led

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

to improved cell performance. As a result, SOFCo repeatedly demonstrated cell ASR values of  $0.6 \Omega\text{-cm}^2$  at  $825^\circ\text{C}$  for ScSZ electrolyte-supported cells obtained from the external supplier.

### **Stack Development and Performance Assessment**

During the Phase I effort, SOFCo performed more than 200 stack tests, with most of these using 2-5 cell short stacks. Most of the early tests were performed using 3YSZ electrolyte-supported cells, and were aimed at establishing repeatable stack assembly procedures and validating the SOFCo "all-ceramic" stack design using the multi-layer ceramic interconnects. The work demonstrated effective cell-to-interconnect and stack-to-manifold sealing, and showed that high fuel utilization ( $>80\%$ ) could be achieved. In 2004, SOFCo began using ScSZ electrolyte-supported cells for stacks, and shifted emphasis toward achieving the stack performance (power density and degradation rate) required for the C2 system demonstration.

In 2004, SOFCo demonstrated that the required initial stack power density could be achieved, but degradation rates were too high. An internal "Tiger Team" performed an extensive study and developed a fundamental understanding of the primary sources for stack degradation; key mechanisms were identified and corrective actions were defined. As a result of this effort, short stack degradation was reduced from 10-20% per 500 hours to less than 4% per 500 hours (from peak power). More important, the contributions to stack degradation associated with the interconnect and the various electrical contacts within stacks were substantially eliminated. With subsequent refinements, particularly with the addition of compliant connections, a stack ASR of  $0.6 \Omega\text{-cm}^2$  was demonstrated using externally supplied ScSZ cells. Stack performance now appears to be largely driven by the cell behavior, for which degradation rates are on the order of 2% per 500 hours. At the end of this reporting period, SOFCo conducted two short stack tests using reformed natural gas as the fuel. Both of these stacks were operated for  $>1000$  hours and demonstrated power degradation rates of 2-3% per 500 hours, approaching the Phase I SECA target.

PCU development has proceeded to the point where SOFCo is ready to construct the C2 stacks and install them into the SOFC module. The flexible current lead and the compliant fuel-side connection have been successfully demonstrated. The ability to pre-condition stacks and then transfer them to the test stand has also been demonstrated. At the end of the Phase I period, SOFCo assembled and tested a final 67-cell PCU using reformed natural gas as the fuel. While this stack operated for only about 400 hours, this was the first tall stack that behaved similar to short stacks and single cells (i.e. initial power increased at constant voltage over a significant amount of time). Given that stack power was still increasing at the end of the test, these results suggest that tall stack degradation may be similar to that observed for short stacks (2-3% per 500 hours). Stack performance was sufficient to meet C2 operating conditions and to provide the required power output. Although all of the components required for C2 stack assembly (cells, interconnects, etc.) were in hand, a decision was made to not proceed with stack assembly and the C2 test.

### **CPOX Reformer**

SOFCo selection and development of CPOX reformer technology for the SECA Phase I program built upon prior fuel processing skills for SOFC and PEM applications that included extensive evaluations of steam reforming, autothermal reforming (ATR) and catalytic partial oxidation (CPOX) reforming approaches. With that prior understanding, SOFCo proposed a CPOX approach based on its best fit to operational requirements needed for the mobile APU application targeted by CPG and SOFCo for the SECA program.

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

Initial catalyst screening and operational tests were performed in SOFCo's micro-reactor at 0.3 kWe scale. Testing showed the viability of waterless CPOX performance for liquid propane (LP) and natural gas (NG) fuels. Scale-up and demonstration was next performed at 1kWe for both fuels. Full-size SECA Phase I developments were completed at 5 kWe scale.

The "waterless" 5-kWe CPOX fuel reformer for use in the Phase I demonstration test at CPG was extensively characterized with Alliance, OH, pipeline natural gas (NG). Reformer operation and material tests showed carbon-free performance during startup, normal operation and shutdown for operation in excess of 2000 hours. A 20:1 load turndown was demonstrated for the C2 reformer (exceeding the original 5:1 target design), thereby increasing the operational flexibility of SOFCo's fuel cell module. Effects of air flow and feed preheat temperature were examined to establish control boundaries.

The C2 reformer with instrumentation was fabricated and closely integrated into the C2 power module. The SOFCo design incorporated a removable catalyst housing for maintenance and replacement. Final installation and shipment to CPG for testing was completed in December 2005. Reformer light-off in the C2 system at CPG was first performed in August 2005.

### **SOFC Power Module (Includes: stack, manifolding, reformer, HX and insulated enclosure)**

Fabrication of the C2 power module was completed in 2004 and delivered to CPG in December. Initial hot functional tests in January 2005 with installed fuel cell stack simulators and manual control proved the operation of the power module's startup burner, heat exchanger and insulation. At the completion of this checkout, CPG continued fabrication and installation of BOP hardware, electricals and controls software. Hot functional testing resumed in early August that allowed CPG checkout of BOP component operation, controls and software during hot operation. Tests with stack simulators were completed in October with preparations underway to install the four, 70-cell stacks for C2 fuel cell Phase I test performance runs.

[REDACTED]

### **Performance Improvements and Cost Reduction**

#### **Advanced Cell Development**

ScSZ electrolyte-supported cells being developed by SOFCo are showing 825°C ASR values of  $<0.45 \Omega \cdot \text{cm}^2$ ; short-term performance degradation rates are modest, and are within expected values based on the known electrolyte aging phenomenon. Based on the promising results for SOFCo cells, short stack tests were performed prior to the end of Phase I. One stack that used the low-cost air-side bond layer in combination with an internal ScSZ cell had an initial ASR of  $0.61 \Omega \cdot \text{cm}^2$  and showed relatively stable performance. The stack ASR, however, did not meet Phase I target of less than  $0.5 \Omega \cdot \text{cm}^2$ .

#### **Low-Cost Interconnect and Stack Materials**

SOFCo made significant progress in the development of low-cost cell-to-interconnect connection materials. Early in the Phase I period, a Ni-cermet contact ink was implemented to connect the anode to the fuel-side of the interconnect. [REDACTED] The development of a low-cost air-side cell-to-interconnect connection material achieved mixed results, however. Low-cost air-side contact materials have been developed with sufficiently high conductivity and chemical compatibility. The low-cost bond layer was successfully demonstrated in combination with the new cathode on internally developed cells at three levels: single cell tests, repeat unit tests and a short stack test. However, attachment of the ScSZ cell to the 3YSZ interconnect with low-cost bump materials has not been successfully accomplished. Acceptable stack testing results were accomplished by continuing to use the Pt-based bumps to connect the low-

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

cost bond-layer and interconnect. As of the end of Phase I, SOFCo believes that the low-cost bond layer material is ready to implement into future stacks. Substitution of this material for the current Pt-based bond layer would serve to reduce the cost of the air-side connection by about 80%.

The baseline interconnects used throughout the Phase 1 program to demonstrate SOFCo's stack technology have used platinum-YSZ cermet vias. The initial selection of platinum cermet vias was based on the stability of platinum in both reducing and oxidizing environments. Although costly, the use of the platinum cermet allowed for an accelerated development of the manufacturing process for SOFCo's novel multi-layer ceramic interconnect and the successful demonstration of a promising stack technology. A successful SECA Phase 1 program required elimination of precious metals to meet the cost targets. Significant progress was made by SOFCo in the development of low-cost via materials, particularly during the last two years (2004-2005). Modified fuel-side and separator-layer via compositions developed during the final year of Phase I resulted in significantly lower ASR contributions and improved stability for the AVM interconnects. In particular, stack #227 was a significant outcome for SOFCo, in that three out of four AVM interconnects in this stack met the June milestone ( $ASR < 0.2 \Omega \cdot \text{cm}^2$ ). Moreover, two of the interconnects met the final ASR target ( $\leq 0.1 \Omega \cdot \text{cm}^2$ ) and showed good stability. SOFCo subsequently assembled and tested two additional short stacks that yielded positive performance data. In particular, Stack #243 was operated for 1000 hours and showed stable resistance values; the average ASR for the AVM interconnects at the end of the test was  $0.07 \Omega \cdot \text{cm}^2$ . Based on these results, SOFCo met the Phase I milestone for the low-cost via materials and is prepared to implement these materials into future interconnect production development efforts.

Initial fabrication of interconnects using a less expensive 3YSZ powder did not indicate any manufacturing issues that need to be addressed. A small batch of 15-cm interconnects was successfully fabricated using powder produced by the chosen supplier on their R&D line. Subsequently, SOFCo received 3YSZ powder from the supplier from their production line and fabricated a number of 10-cm interconnects that met quality requirements. Successful fabrication of 10-cm interconnects served to confirm that the low-cost 3YSZ powder is suitable for use in the production of SOFCo's multi-layer interconnects. As a result, a decision was made to perform future interconnect development efforts using the new low-cost 3YSZ.

### **Balance of Plant (BOP)**

During this period the low cost cold Balance of Plant (BOP) system was tested and developed with the C2 complete system. Significant testing has been accomplished with the complete BOP and C2 Hot box system. At this time all BOP hardware is functioning as required for system operation. Test results uncovered significant deterioration of the recuperator heat exchanger as the result of a fast start up transient, and the requirement for its replacement put a hold on further testing.

### **Controls and Power Electronics**

The control loops necessary for the phase 1 test were completed this reporting period. The stack inlet, outlet, and burner temperature control loops have been verified. All flow control and measurement devices have been implemented.

### **Future Plans.**

#### **Cell, stack, reformer, system, manufacturing**

Development of the stack and manifold assembly is being transitioned to an alternative supplier who has already demonstrated the ability of their stack to meet or exceed the SECA Phase 1

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

requirements, with reasonable potential to achieve SECA Phase 3 performance and cost goals. For completion of the Phase 1 demonstration, CPG is relying on approval of the requested extension and transition to the new supplier's stack and natural gas steam reformer. In parallel, CPG is actively working to identify and assemble a team to transition the fuel reforming effort to focus on technology development for diesel reforming for the Phase 2 effort.

### **Balance of Plant**

A completely new air management system is being designed and developed to provide a low cost system for the new Phase 1 extension plan. This new air system will incorporate the DC/DC boost cooling feature and other features consistent with the longer term design approach.

### **Controls and Power Electronics**

New CAN interface modules are being designed to provide control interface for the new air management system. The DC/DC boost converter is being redesigned for integration with the new stack.

## **Issues and Challenges**

### **Cell, stack, reformer, system, manufacturing**

Due to the time constraint of completing the Phase 1 system test by the end of calendar 2006, significant reliance will be placed on use of supplier provided thermally integrated hot section sub-systems, consistent with the hot-box / power module concept used with the SOFCo based system. Key areas of technological development in the cell and stack leading to transition to the Phase 2 system have been identified for focus during 2006. These areas will be addressed in more detail in the next report.

### **Balance of Plant**

Timing for the new air management system is short. The new system design is constrained by the requirement for compatibility with pre-existing subsystems. Although these constraints place some limitations on cost reductions and performance, the team believes BOP will be consistent with achievement of the Phase 1 SECA targets, and further refinement for Phase 2 will be possible.

### **Controls and Power Electronics**

New CAN interface modules need to function as both interpretation and control modules to the existing supplier control system. The DC/DC boost converter will be revised to meet the new power input and cooling flow requirements.



10 kW SOFC Power System Commercialization  
SECA Semi-Annual Report 41244R08

Table of Contents

<b><u>Section</u></b>	<b><u>Page</u></b>
Cover Sheet	1
Disclaimer	2
Abstract	3
Table of Contents	9
List of Graphical Materials	10
Executive Summary	13
Body	15
References	97
Bibliography	97
List of Acronyms	98

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## List of Graphical Materials

<u>List of Tables</u>	<u>Page</u>
Table 1 – DOE Requirements Status.....	4
Table 2 – CPOX Performance Comparison.....	50
Table 3 – 1 kWe CPOX Performance Comparison.....	52
Table 4 – Comparison of Reformer Scale-up Performance.....	56
Table 5 – Material Performance During Reformate Exposure.....	59
Table 6 – Summary of Test Data Sets and Measured Recuperator Heat Exchanger Inlet Temperature and Fuel Flow Rate.....	66
Table 7 – Summary of Parallel Path Improvements.....	71

<u>List of Figures</u>	<u>Page</u>
Figure 1 – Cell performance history.....	16
Figure 2 – Performance of external ScSZ electrolyte-supported cells subjected to two different anode reduction procedures.....	18
Figure 3 – Performance results at 825°C for vendor supplied ScSZ cells (68.1 cm <sup>2</sup> active area) using the modified anode reduction procedure.....	18
Figure 4 – Performance results at 825°C for external ScSZ cells (68.1 cm <sup>2</sup> active area) with the pre-conditioning procedure.....	19
Figure 5 – Strength of ScSZ electrolytes.....	20
Figure 6 – Long-term performance results for test SC362.....	21
Figure 7 – Progress in short stack performance.....	22
Figure 8 – Typical 5-cell short stack.....	23
Figure 9 – Manifold arrangements for short stacks.....	23
Figure 10 – Fuel utilization vs. current density for a short stack.....	24
Figure 11 – Stack 154 performance with reformate.....	25
Figure 12 – Initial short stack testing with ScSZ cells.....	26
Figure 13 – Performance of short stacks #203 and #213.....	27
Figure 14 – Performance of short stacks #223 and #224.....	28
Figure 15 – Long-term performance for stack #232.....	29
Figure 16 – Performance for stack #241 operated on desulfurized NG.....	30
Figure 17 – 20-cell stack in test stand.....	31
Figure 18 – 47-cell stack in test stand.....	31
Figure 19 – HST Facility.....	32
Figure 20 – FEA stress analysis results.....	32
Figure 21 – 48-cell stack #168 in the HST facility.....	33
Figure 22 – FEA stress analyses for stack #168.....	33
Figure 23 – Two 48-cell stacks installed in the C1 module.....	34
Figure 24 – 48-cell stack #189.....	35
Figure 25 – Steady-state operation of stack #189.....	36
Figure 26 – Cyclic testing of stack #189.....	36
Figure 27 – Rapid startup of stack #189.....	36
Figure 28 – 70-cell stack #214.....	37
Figure 29 – Test results from 17-cell stack #226A.....	40
Figure 30 – Test results from 50-cell stack #233.....	41

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## List of Figures (Cont'd.)

Figure 31 – PCU stack #244 .....	42
Figure 32 – Initial test results for Stack #244 .....	43
Figure 33 – Additional test results for stack #244 .....	44
Figure 34 – Monolithic stack .....	44
Figure 35 – Monolith catalyst (left) and quartz reactor top view (right) .....	47
Figure 36 – CPOX startup (LPG and NG light-off) .....	47
Figure 37 – Carbon formation effect on CPOX performance .....	48
Figure 38 – CPOX performance with LPG .....	48
Figure 39 – Effect of feed preheat temperature .....	48
Figure 40 – Effect of O <sub>2</sub> /C feed ratio .....	49
Figure 41 – Reformer performance with turndown .....	49
Figure 42 – Fuel processor test unit .....	50
Figure 43 – 1.5 kWe CPOX reformer .....	51
Figure 44 – Hot-gas filter .....	51
Figure 45 – NG CPOX performance [Catalyst B] .....	52
Figure 46(a) – Fuel conversion vs. load .....	53
Figure 46(b) – Reformer efficiency vs. load .....	53
Figure 47(a) – C <sub>2</sub> + slip for CPOX catalysts .....	53
Figure 47(b) – Reformer operating envelope .....	53
Figure 48 – C2 (5kWe) reformer test hardware .....	55
Figure 49 – Coupon test hardware .....	55
Figure 50(a) – Fuel conversion with load .....	57
Figure 50(b) – Reformer outlet temperature with load .....	57
Figure 51(a) – Air level effect on performance .....	58
Figure 51(b) – Preheat effect on performance .....	58
Figure 52(a) – 5 kWe C2 reformer durability .....	58
Figure 52(b) – 1 kWe C1 reformer durability .....	58
Figure 53 – C1 stack and cage assembly .....	61
Figure 54 – Stacks (2) installed and insulated in C1 hot box .....	61
Figure 55 – C1 power module .....	61
Figure 56 – Example C1 performance during operation and controls checkout .....	62
Figure 57 – Power module and BOP configuration at CPG for first power module heat up .....	63
Figure 58 – Test 1643 showing temperatures and startup burner fuel flow .....	63
Figure 59 – Test 1645 showing temperatures and fuel flows (startup burner and makeup) .....	64
Figure 60 – Test 1656 (August 12) showing temperatures and startup burner + makeup fuel flows .....	67
Figure 61 – C2 power module inspection pictures at CPG on October 10 .....	68
Figure 62 – Combustor and cordierite plate showing the heat-affected zone corresponding to the combustion of the makeup fuel jet .....	68
Figure 63 – Heat exchanger showing discoloration from heat affect .....	69
Figure 64 – Parallel path cost reduction roadmap .....	72
Figure 65 – Typical performance of SOFCo developed ScSZ electrolyte-supported cells at 825°C tested under humidified 50:50 H <sub>2</sub> N <sub>2</sub> fuel .....	73
Figure 66 – Degradation of SOFCo cells with and without manifold protection schemes .....	74
Figure 67 – Degradation results for SOFCo cells subjected to more extreme conditions expected within stacks .....	74

10 kW SOFC Power System Commercialization  
SECA Semi-Annual Report 41244R08

**List of Figures (Cont'd.)**

Figure 68 – Observed migration of strontium into the ScSZ electrolyte.....	75
Figure 69 – Performance of cells using a [ ]cathode with the addition of the SOFCo low-cost bond layer material .....	75
Figure 70 - 825°C performance of SOFCo cells using new MEIC cathodes (with low-cost bond-layer also present) .....	76
Figure 71 – Single cell degradation data (825°C) for SOFCo cells using an initial MEIC cathode composition .....	77
Figure 72 – [ ] .....	78
Figure 73 – Low-cost cathode bond layer resistance test.....	78
Figure 74 – Cathode bond layer in single cell tests .....	79
Figure 75 – Performance of low-cost cathode bond layer with internal cell .....	79
Figure 76 – Low-cost cathode-side connection test.....	80
Figure 77 – Performance of stacks with low-cost air-side connections .....	81
Figure 78 – Schematic of multi-layer interconnect section showing low-cost vias and SEM micrograph showing a cross-section through a column of vias.....	82
Figure 79 – Schematic showing arrangement for Stack #165 .....	83
Figure 80 – Resistance contributions for AVM interconnects during initial testing period for Stack #165.....	84
Figure 81 – SEM micrograph showing the interface between the separate and fuel-side vias .....	84
Figure 82 – Short stack #227 containing four AVM interconnects .....	86
Figure 83 – Short stack #234 containing four AVM interconnects .....	86
Figure 84 – Performance data for stack #243.....	87
Figure 85 – C2_SIM Development Testing at CPG .....	90
Figure 86 – C2 Recuperator Over Temperature Damage .....	91
Figure 87 – CPOX Light-Off.....	93
Figure 88 – C2 System Temperatures.....	93
Figure 89 – Control System Hardware Architecture .....	95
Figure 90 – Stack Inlet Temperature Control Loops .....	96
Figure 91 – Simulink Model of Recuperator Temperature Control.....	97
Figure 92 –Fuel Cell Boost Efficiency .....	99

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## Executive Summary

Although significant progress was made during the reporting period, performance falls short of SECA objectives in two key areas: cost and degradation. The major factor in the cost miss is stack performance. The major factor in degradation is stability of the cathode to interconnect junction. Highlights are provided for activities supporting the Phase I demonstration and development efforts directed at performance improvements and cost reduction.

**The status of the program related to the four key performance requirements is summarized below:**

Measure	Metric	Projected Deliverable Status
System Cost	\$800/kW (mfg cost)	\$2,133/kW (Best Projection is \$1683/kW)
Efficiency	25% (mobile)	>25%
Power Degradation	<2% per 500 hrs at constant voltage	2-3% / 500 hrs based on a 67-cell stack tested for 400 hrs that exhibited degradation behavior similar to recent short stacks.
Availability	80% for 1,500 hr test	Achievable based on multiple stack tests


## C2 Prototype Development and Demonstration

**Cell Evaluation.** Single cell tests were performed resulting in the development of heat-up / pre-conditioning processes that were compatible with the stack assembly procedure, and led to improved cell performance. SOFCo repeatedly demonstrated cell ASR values of  $0.6 \Omega\text{-cm}^2$  at  $825^\circ\text{C}$  for ScSZ electrolyte-supported cells vs. target of  $<0.5 \Omega\text{-cm}^2$  required for Phase 1.

**Stack Development and Performance Assessment.** Short stack degradation has been reduced from 10-20% per 500 hours to less than 4% per 500 hours and the contributions to stack degradation associated with the interconnect and the various electrical contacts within stacks were substantially eliminated. SOFCo conducted two final short stack tests using reformed natural gas fuel. Both stacks were operated for >1000 hours and demonstrated power degradation rates of 2-3% per 500 hours.

PCU development proceeded to the point SOFCo is ready to construct the C2 stacks and install them into the SOFC module. Flexible current lead and the compliant fuel-side connection and the ability to pre-condition stacks and transfer them to the test stand have been demonstrated. SOFCo assembled and tested a 67-cell PCU for 400 hours using reformed natural gas fuel. This was the first full scale stack that behaved similarly to short stacks and single cells (i.e. initial power increased at constant voltage over a significant amount of time). These results suggest that tall stack degradation may be similar to that observed for short stacks (2-3% per 500 hours). Stack performance was sufficient to provide the required minimum power. Components (cells, interconnects, etc.) were procured for C2 stack assembly.

**CPOX Reformer.** The C2 reformer was fabricated and closely integrated into the C2 power module. The design incorporated a removable catalyst housing for maintenance and replacement. Reformer light-off in the C2 system at CPG was first performed in August 2005.

**SOFC Power Module.** Hot functional testing resumed in early August that allowed CPG checkout of BOP component operation, controls and software during hot operation. Tests with stack simulators were completed in October. 

## Performance Improvements and Cost Reduction

**Advanced Cell Development.** ScSZ electrolyte-supported cells being developed by SOFCo are showing  $825^\circ\text{C}$  ASR values of  $<0.45 \Omega\text{-cm}^2$ ; short-term performance degradation rates are modest, and are within expected values based on the known electrolyte aging phenomenon. One stack that used the low-cost air-side bond layer in combination with an internal ScSZ cell

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

had an initial ASR of  $0.61 \Omega\text{-cm}^2$  and showed relatively stable performance. The stack ASR, however, did not meet Phase I target of less than  $0.5 \Omega\text{-cm}^2$ .

**Low-Cost Interconnect and Stack Materials.** [REDACTED] Development of a low-cost air-side cell-to-interconnect connection material achieved mixed results. Low-cost air-side contact materials have been developed with sufficiently high conductivity and chemical compatibility. The low-cost bond layer was successfully demonstrated in combination with the new cathode at three levels: single cell, repeat unit, and short stack. Attachment of the ScSZ cell to the 3YSZ interconnect with low-cost bump materials has not been successful. As of the end of Phase I, SOFCo believes that the low-cost bond layer material is ready to implement.

The baseline interconnects used throughout the Phase 1 program incorporate platinum-YSZ cermet vias. The ASR target for Alternative Via Material (AVM) is  $\leq 0.1 \Omega\text{-cm}^2$ . SOFCo assembled and tested Stack #243 for 1000 hours with stable resistance values. The average [REDACTED]

### Balance of Plant

A low cost BOP system was adapted to match evolving stack and hot box requirements, tested, and integrated with the C2 (simulator) system. Significant testing was accomplished with the complete BOP and C2 Hot box system, and all BOP hardware is functioning as required for system operation. It is currently able to provide the required performance from start-up, Normal Operating Condition (NOC), peak power, and shutdown for the SECA Phase 1 test. The system demonstrated stable operation for hundreds of hours at the NOC operating point. System modeling, vibration isolation, and enclosure design continued during this reporting period. The test cell instrumentation, programming, and calibration are complete for the phase 1 test.

However, during development testing a failure in the heat exchanger system has been detected in the data. Diagnostic testing indicated that a visual inspection of the recuperator was required. The inspection highlighted an over temperature failure in the recuperator exhaust inlet. This condition put a hold on further testing.

### Controls and Power Electronics Systems

The control loops necessary for the Phase 1 test were completed this reporting period. The stack inlet, outlet, and burner temperature control loops have been verified. All flow control and measurement devices have been implemented.

The fuel cell boost has been repackaged to allow system integration and production. In addition a new CAN interface board has been designed and built to allow the boost to be controlled via the CAN communication serial bus. Thermal verification testing was completed and the fuel cell boost was ready for phase 1 fuel cell testing to commence.

Delays in commercial development of the power output stage prompted design and development of a test system backup. A functional interim power output stage has been constructed to allow the fuel cell to startup from an AC source and invert the fuel cell's DC voltage to AC for the purposes of electrical loading. This interim system remains available as the backup plan pending availability of the commercial power output stage.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## ARTICLE 1. I. Prototype System Development and Demonstration

### ARTICLE 2. Subtask 2 Cell Evaluation

**OBJECTIVE:** Develop and/or select a cell that will facilitate development of a stack for the C2 Phase I test having an ASR of about  $0.75 \Omega \cdot \text{cm}^2$ , which implies a cell performance target of  $0.65\text{-}0.7 \Omega \cdot \text{s-cm}^2$  at  $825^\circ\text{C}$ .

**APPROACH:** SOFCo began the Phase I effort focusing on co-fired cells – that is cells in which the anode, electrolyte and cathode were fired in a single operation. Toward the end of the first year of the program (2003), it was determined that co-fired cells suffered from mechanical integrity issues. As a result, efforts were shifted toward a dual-path approach. One path involved evaluation of cells from commercial suppliers and adapting available cells to the SOFCo design. The second path involved continued cell development within SOFCo, but focusing primarily on cells using an advanced electrolyte and post-fired electrodes.

SOFCo initially selected YSZ electrolyte-supported cells for stack development efforts. Subsequently, ScSZ electrolyte-supported cells with an ASR of  $\sim 0.7 \Omega \cdot \text{cm}^2$  at  $825^\circ\text{C}$  were selected in 2004 for further stack development and the C2 prototype system. Because of vendor manufacturing problems and supply disruptions in 2004, additional qualification testing was performed in early 2005. A down selection was made in March 2005 to a vendor-supplied ScSZ cell with co-fired electrodes and a standard fuel-side nickel conducting layer was specified. Subsequently, SOFCo experienced quality problems with the cells. In particular, electrolyte strength fell below required minimum values as a result of apparent problems with the tape casting process. SOFCo worked closely with the cell supplier to address the electrolyte strength issue; significant testing of green tapes, fired electrolytes and cells was performed. Findings were communicated to the external cell supplier and corrective actions implemented to improve cell quality. By the final reporting period (Q3 2005), the cell vendor had resolved all quality problems, and as a result was able to deliver the required cells for the C2 demonstration.

**EXPERIMENTS:** During the early part of the SECA program, SOFCo used button cell testing as the primary tool for screening cells. However, testing at this scale was determined to be inadequate for projecting cell performance at the stack level. As a result, SOFCo developed a single-cell test stand that was capable of testing 10 cm (4" round or square) cells. Button cell testing was continued to support advanced cell development, but was no longer used after 2003 to evaluate performance/quality of cells intended for use in stack development efforts.

During the final two years of Phase I, SOFCo's approach continued to use the single-cell test stands to screen the performance of full-size cells prior to introducing such cells into short stack tests. The test stands were able to achieve representative fuel utilization as exhibited during short stack testing. Initial ASR calculations were performed using the slope of the VI curve between approximately 0.6 and 0.8 volts to compare cell performance and to determine whether performance was close to that required for C2. A more accurate ASR calculation was also performed based on SOFCo's electrochemical model of the single-cell test stand combined with the VI scan obtained for the cell, and the noted fuel utilization levels. Modeling of the single-cell test stand permitted prediction of the power level and operating voltage that a cell needs to exhibit at a constant voltage test condition (tested under a set fuel utilization and an initial current density), for a cell to meet the performance requirements of C2 ( $\text{ASR} < 0.7 \Omega \cdot \text{cm}^2$ ). This provided a quick assessment of whether cells met the target performance requirements without having to run the ASR model calculations of VI scans throughout the endurance testing of single-cells. In addition to the cell performance testing, mechanical testing of electrolytes and

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

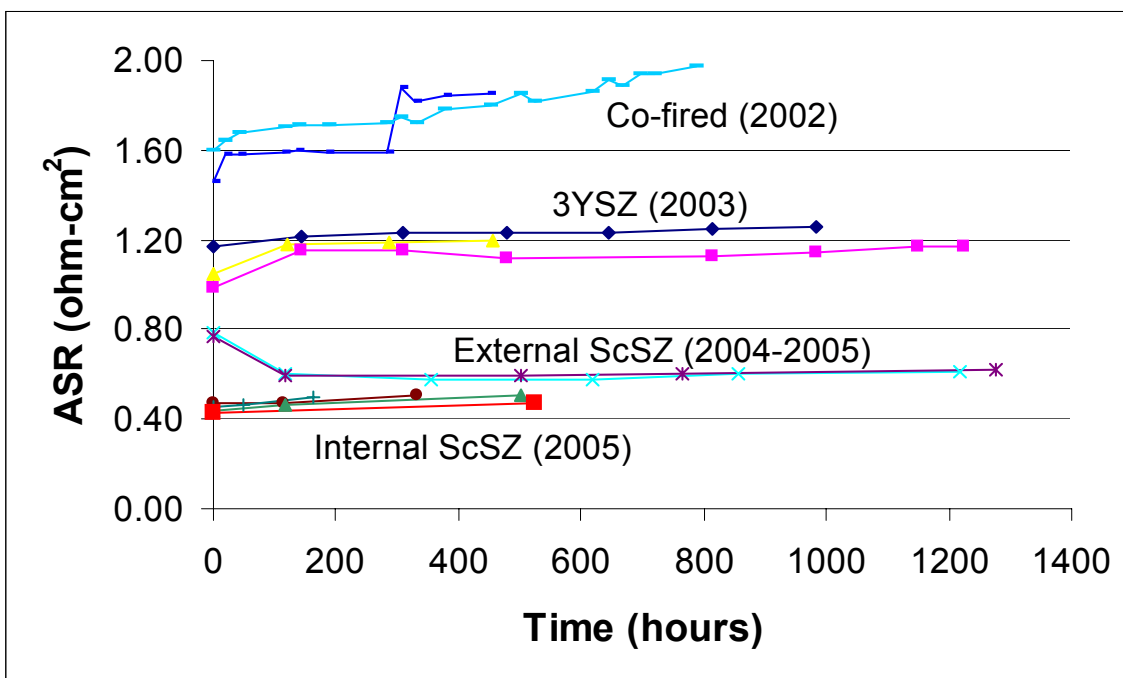
cells was performed. The standard conditions for the single-cell tests for external ScSZ cells were:

- 825°C test temperature
- 3% humidified hydrogen/nitrogen fuel gas
- constant voltage that achieves an initial 350 mA/cm<sup>2</sup> current density
- 37% fuel utilization.

SOFCo also explored ways to improve cell performance and investigated an anode reduction and a cell pre-conditioning process. The testing conditions for the pre-conditioned cells were the same as the standard ones, except for the heat up procedure. After the anode reduction step, the standard process was to test the cells at 825°C. [REDACTED]

### RESULTS AND DISCUSSION:

During Phase I, SOFCo made significant progress in the development of high-performance electrolyte-supported cells. This progress is illustrated in **Figure 1**, which provides representative performance results for cells tested during the 4-year period. SOFCo began the program focusing on cells in which the electrolyte, anode and cathode were co-fired in a single firing operating. These cells showed relatively poor performance, both in terms of power output



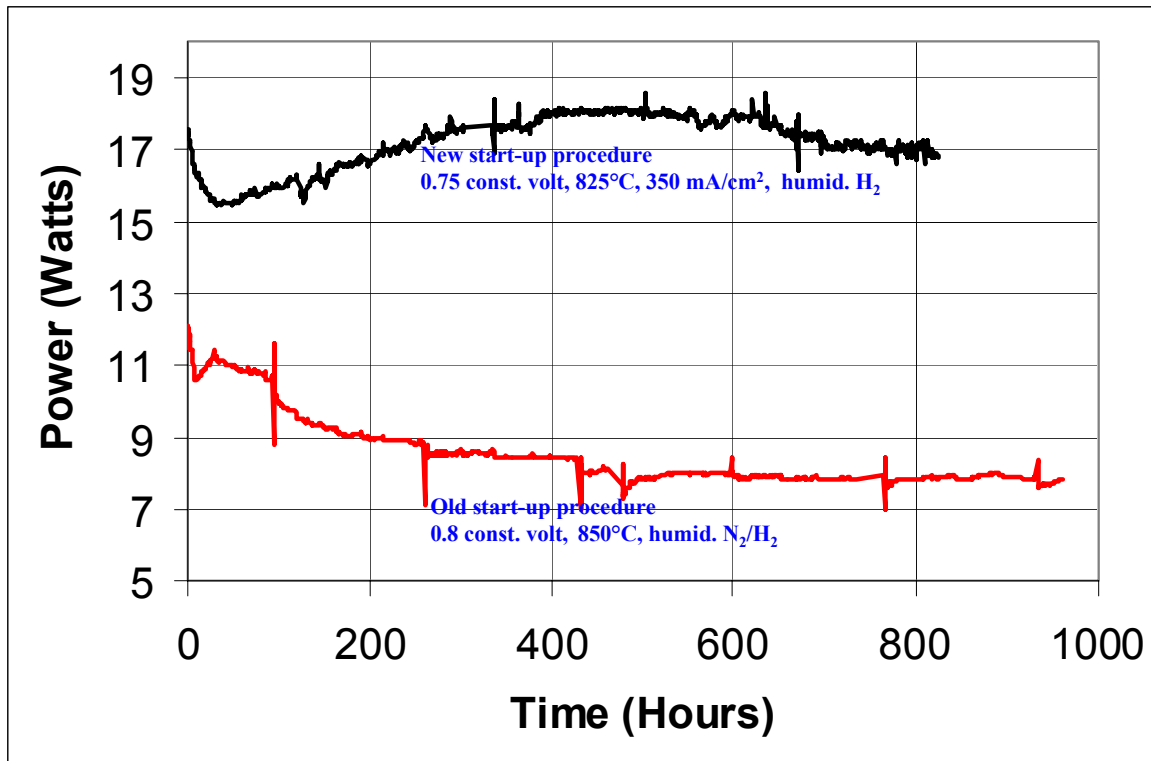
**Figure 1. Cell performance history**

(area specific resistance – ASR) and degradation behavior, and also suffered from a mechanical integrity issue. As a result, SOFCo terminated internal cell development efforts, and shifted emphasis toward cells supplied by external sources. A number of anode-supported cells and electrolyte supported cells were evaluated. After determining that anode-supported cells



## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

were not suitable for use with SOFCo's multi-layer ceramic interconnect, due to a mismatch in the coefficient of thermal expansion (CTE), a decision was made in 2003 to focus on commercially available electrolyte-supported cells using a 3YSZ electrolyte. While these cells were ideal as a baseline cell to support stack development efforts, they did not have the performance required to meet Phase I targets for the C2 prototype demonstration. As a result, SOFCo initiated a dual-path effort to develop an advanced electrolyte-supported cell using a ScSZ electrolyte material. In one path, discussed further below, SOFCo worked closely with an external supplier to substitute the new electrolyte material into its production operations using the standard anode and cathode materials. The result was a cell having an ASR in the range of 0.6-0.7  $\Omega$ -cm<sup>2</sup>. As a second path, SOFCo initiated an internal effort to develop a higher performance cell (ASR <0.45  $\Omega$ -cm<sup>2</sup>) using the same ScSZ electrolyte material, but focusing on improved electrodes. Development of the ScSZ cell provided by the external supplier is discussed further below; internal development of the higher performance ScSZ cell is reviewed in a later section.

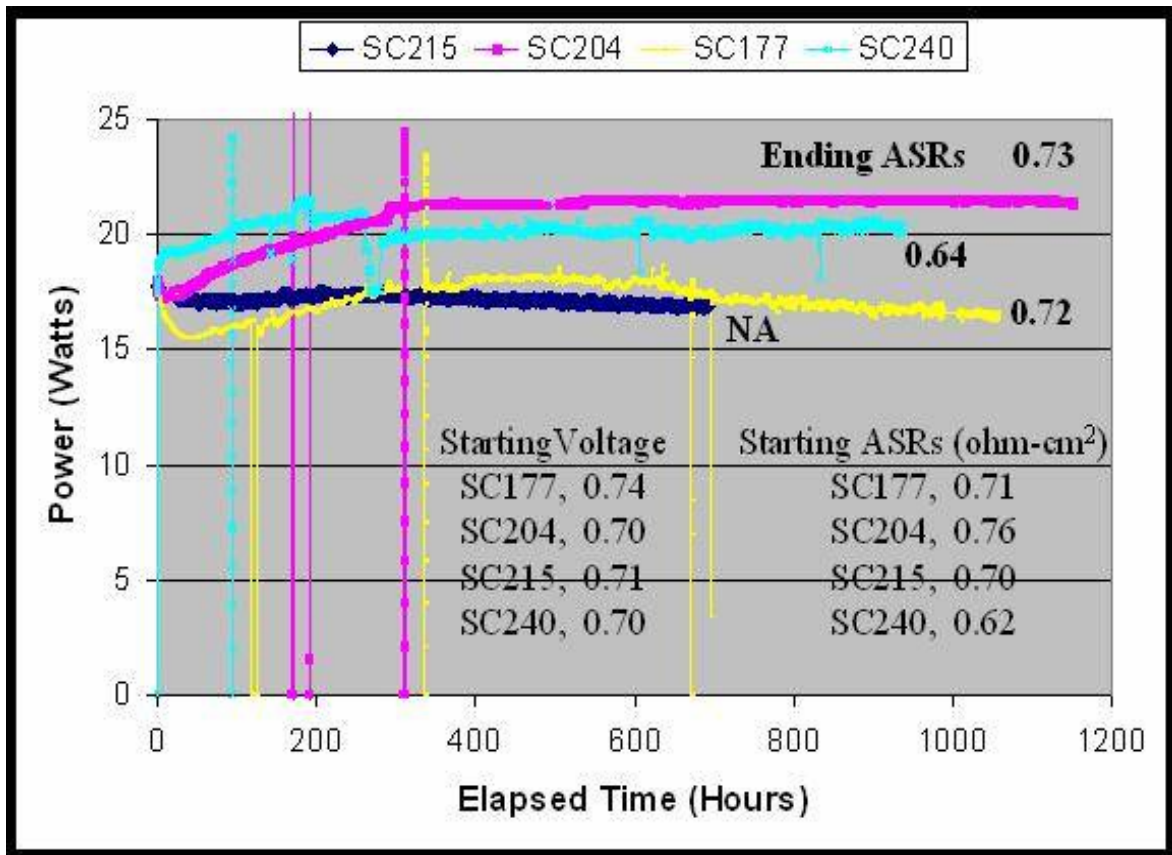


**Figure 2. Performance of external ScSZ electrolyte-supported cells subjected to two different anode reduction procedures**

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

In 2003, SOFCo initiated development of the ScSZ electrolyte-supported cell, and in Q3 2003 a ScSZ cell from an external supplier was selected for optimization and implementation into stacks targeted for the C2 demonstration. Early tests for such cells demonstrated initial ASR values in the range of 0.7-0.75  $\Omega\text{-cm}^2$ , but degradation rates were too high. Extensive testing of these cells, along with post-test examinations, revealed that the poor performance was related to a poor anode microstructure (and related high resistance). By changing the procedure for heating up these cells, and including the process for reducing the NiO in the anodes, a much more stable microstructure was obtained. The result, as shown in **Figure 2**, was a significant improvement in starting cell ASR (higher power output) and stability.

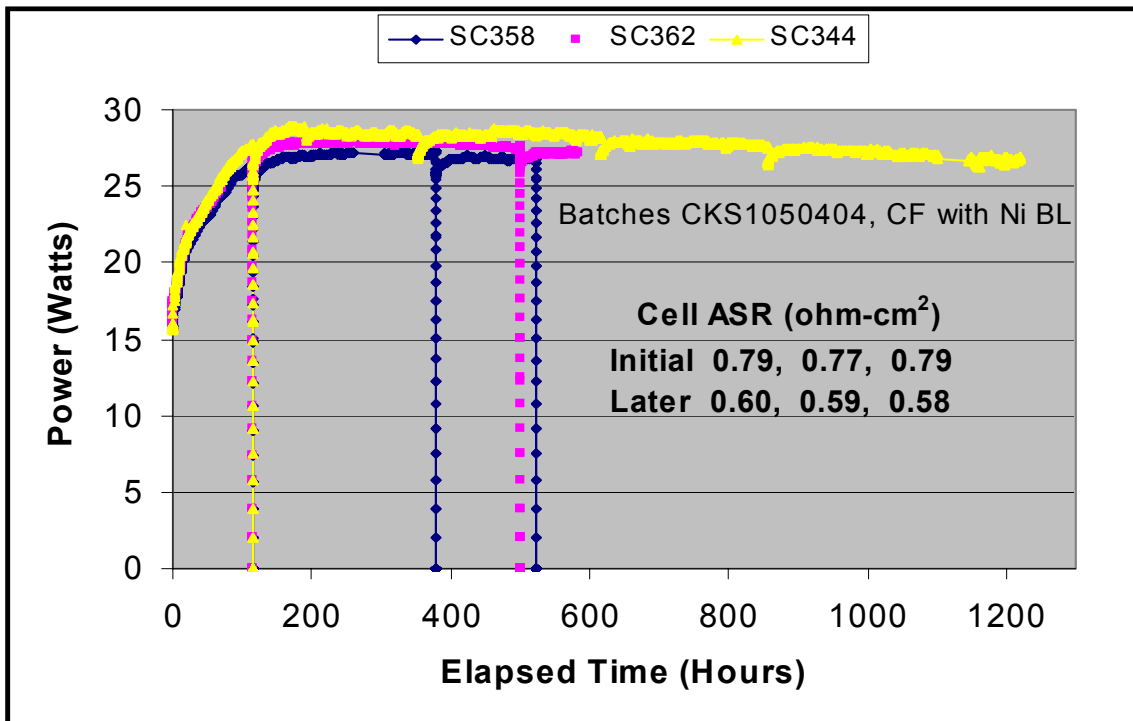
Subsequent testing of cells in 2004 (see **Figure 3**) showed cell performance in the required range (ASR  $\sim 0.7 \Omega\text{-cm}^2$ ), but performance was somewhat variable. Further development showed that cell performance was impacted by the heat up procedure. Upon adding a pre-conditioning step, cell ASR values of 0.6  $\Omega\text{-cm}^2$  were achieved, as shown in **Figure 4**.



**Figure 3. Performance results at 825°C for vendor supplied ScSZ cells (68.1 cm<sup>2</sup> active area) using the modified anode reduction procedure**

While significant progress was made in the area of improved electrochemical performance, a major quality problem arose with the externally supplied ScSZ cells in early 2005. Extensive evaluation of the cells revealed that the electrolytes contained defects which adversely affected cell strength. In some cases, cell strength fell. As a result, many cells failed in both single-cell and stack testing. This issue resulted in a temporary interruption in the supply of cells to SOFCo in May 2005, while an intense collaborative effort

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 4. Performance results at 825°C for external ScSZ cells (68.1 cm<sup>2</sup> active area) with the pre-conditioning procedure**

was conducted to solve the electrolyte strength problem. The strength issue was partially solved by the cell supplier by implementing a modified slip formulation and cell delivery resumed, with 98 ScSZ cells delivered to SOFCo the end of June. These cells were made from electrolytes (from tapes cast in May) having reasonably good strength. [REDACTED] However, another electrolyte sample from a tape cast in June showed low strength (<50% of the target value), indicating a process variability issue. Subsequently, SOFCo performed characterization of the green tapes to help understand the problem, along with extensive mechanical testing of fired electrolytes. SEM examinations were also performed for selected specimens. A number of defect sources were identified, and the results were communicated to the cell supplier. Implementation of corrective actions resulted in significant improvement of electrolyte strength and cell quality. In July the supplier completed a 9 day tape casting campaign that yielded sufficient green tape required to supply ~ 500 cells for stack development and the four 70-cell stacks for the C2 unit. Strength measurements were made for electrolytes produced from each batch of tape (see **Figure 5**). The measured electrolyte strength values were all greater than [REDACTED] of the target value. Shipment of ScSZ electrolyte-supported cells resumed in early August; the supplier completed shipment of all cells for the C2 demonstration in September.

Electrochemical testing was continued for the ScSZ cells through the end of the Phase I period to validate cell quality (for cells targeted for the C2 stacks) and to obtain long-term performance data. **Figure 6** shows the performance of SC362 over a testing period approaching 3000 hours. Degradation rates observed for this cell (from peak power) were in the range of 3.5-4.0% per 500 hours during the initial testing period, and slowed down to less than 2% per 500 hours toward the end of the test. This degradation rate was slightly higher than that observed for the previously longest running cell (SC344) shown in **Figure 4**. The degradation rate for this cell was measured at 2.9% per 500 hours from 185 to 1219 hours.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

While the power output (or ASR) from the external ScSZ cells appear to meet the Phase I targets for the C2 demonstration, the degradation rates are too high. To better understand the sources of degradation, SOFCo performance substantial testing and post-test examinations. Through this effort, a number of potential sources for excessive degradation were identified. The primary contributors were aging of the ScSZ electrolyte and contaminants from the gas supply tubes and manifolds used in the single cell test stands.

Extensive testing has shown that cell mechanical properties do not degrade over time at operating temperature. However, tests at PNNL have confirmed that electrical conductivity of the electrolyte does decrease by about 25% over the initial 1000 hours at typical stack operating temperatures. The resistance change appears to level out beyond 2000 hours. It is estimated that the electrolyte contribution to the cell ASR is about  $0.15 \Omega\text{-cm}^2$ , while the remaining  $0.55 \Omega\text{-cm}^2$  contribution is attributed to the electrodes. As a result, a 25% increase in electrolyte resistance is expected to cause the cell ASR to increase by about 2% per 500 hours during the initial 1000 hours of operation. Degradation rates are expected to be lower beyond 1000 hours.

Another key source of degradation was contamination of the electrodes of the Incoloy 800 manifolds that are used in the SOFCo single cell test stands. Of greatest significance, SEM/EDS analyses have often shown chromium contamination in the cathodes. In cells tested for long time periods and/or at high temperatures, the level of chromium contamination has been fairly high. Even with chromium present in the single cell tests, relatively low degradation rates ( $\sim 3\%$  per 500 hours) have been measured. This might be due to the fact that these cells utilize a LSM-based cathode that is known to be less susceptible to chromium poisoning than higher performance mixed-conducting ferrite-based cathodes. Also, examinations often revealed the presence of Si and P on the anode surface. Since the level of contamination was generally low, the impact of these species on anode performance is unknown. Toward the end of the Phase I period, SOFCo performed a series of tests where the test stands were modified to prevent contamination of the electrodes. While degradation rates appeared to be reduced, sufficient testing was not performed to quantify the reduction in degradation.

**CONCLUSION:** ScSZ electrolyte-supported cells with an ASR of  $\sim 0.7 \Omega\text{-cm}^2$  at  $825^\circ\text{C}$  were selected for the C2 stacks. Substantial qualification testing was performed and a final down

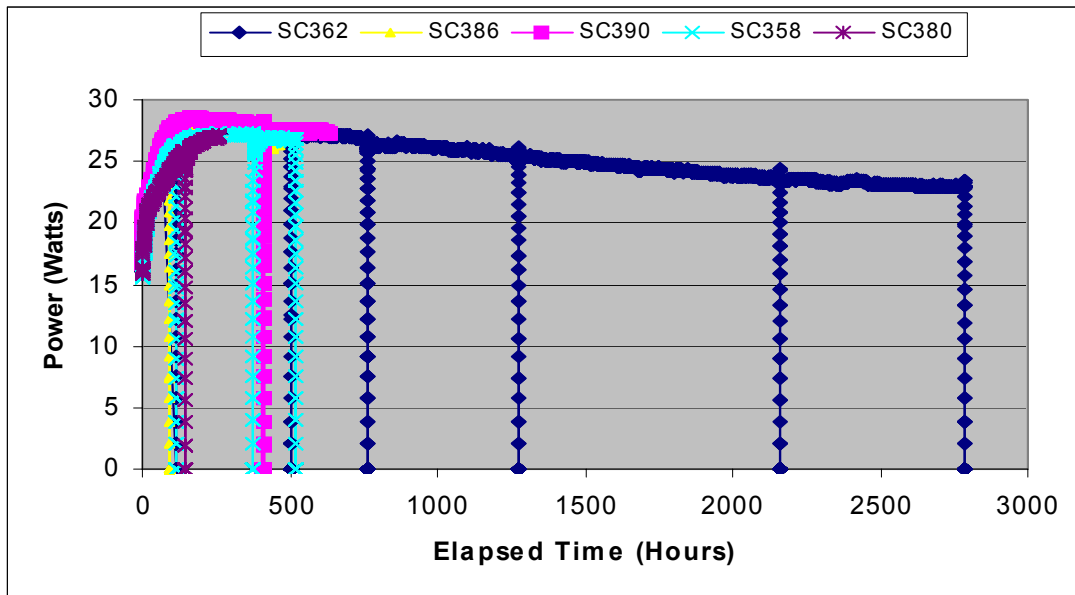


Figure 6 Long-term performance results for test SC362

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

selection was made in March 2005 to define the specifications for the cell to be used for PCU development and the C2 demonstration. Single cell tests confirmed that this type of cell had the expected electrochemical performance, with ASR of close to  $\sim 0.7 \Omega\text{-cm}^2$  and a degradation rate  $< 3\%$  per 500 hours. An electrolyte strength issue, as a result of poor tape quality, caused a temporary interruption in the supply of cells to SOFCo in May. An intense collaborative effort between SOFCo and the cell supplier was conducted to solve the electrolyte strength problem. Cell quality was significantly improved and the shipment of cells from the external supplier was resumed. It was concluded that these ScSZ electrolyte-supported cells have adequate electrolyte strength and sufficient performance for stack development and the C2 stacks. A pre-conditioning procedure has been developed that has a positive impact on cell performance. As a result, cell ASR values of  $0.6 \Omega\text{-cm}^2$  (at  $825^\circ\text{C}$ ) and more consistent cell performance have been achieved. This pre-conditioning procedure has been incorporated into both short stack and C2 stack tests.

## Subtask 3 Stack Development

### STACK PERFORMANCE AND DEGRADATION

**OBJECTIVE:** Establish a fundamental understanding of the primary contributions to stack performance degradation and develop solutions to drive degradation toward the Phase I target of  $< 2\%$  per 500 hours. Demonstrate low degradation in short stacks and implement appropriate improvements in materials and assembly techniques into tall stacks (power cell units – PCUs), based on risk factors and available time.

**APPROACH AND EXPERIMENTS:** During the first two years of the SECA program, stack testing at SOFCo was limited due to poor mechanical integrity of the co-fired cells (primarily in the first year – 2002) and shortage of multi-layer interconnects (primarily in 2003). During this period, significant effort was directed at achieving a reliable stack design, including cell-to-interconnect and stack-to-manifold seals. Testing was generally performed with 2-5 cells

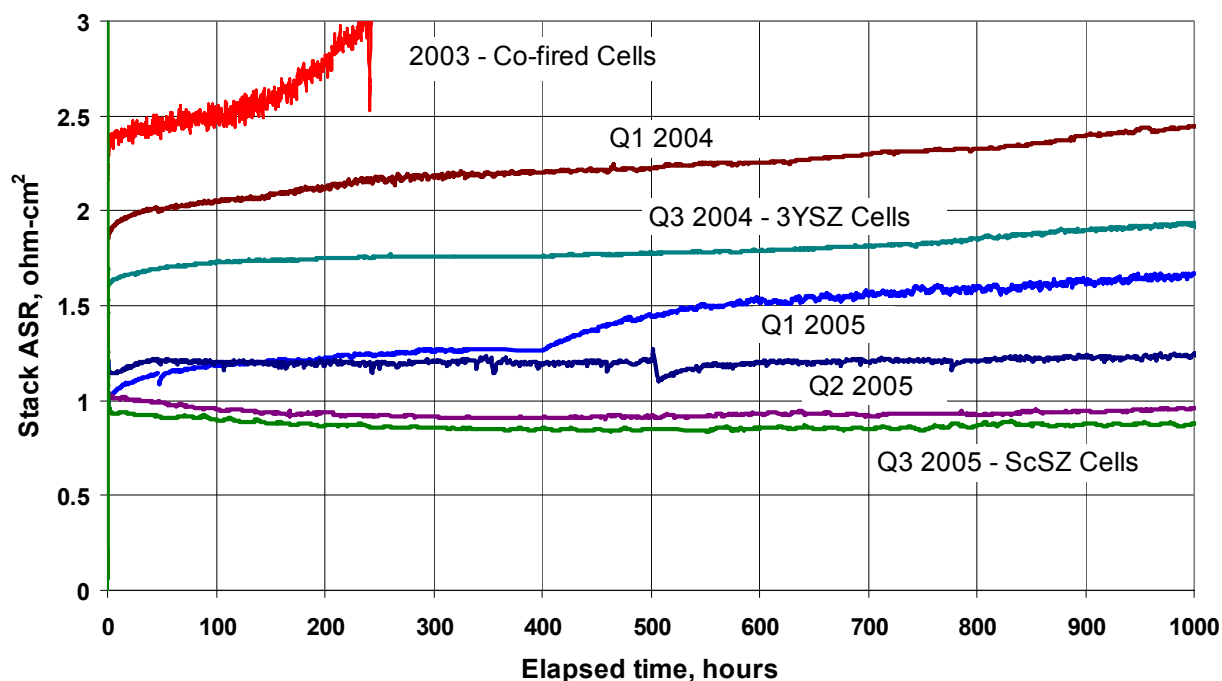


Figure 7. Progress in short stack performance

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

stacks. In addition, SOFCo developed a novel approach for inserting instrumentation into short stacks, thereby allowing direct measurement of the separate contributions to overall stack performance. This tool was used to isolate and identify the key contributions to stack resistance and performance degradation.

Through numerous short stack tests, first using 3YSZ electrolyte-supported cells and later using ScSZ electrolyte-supported cells, SOFCo observed stack performance behavior that could not be explained based on single cell performance. In particular, degradation rates were much higher than expected. To address this issue, SOFCo established an internal "Tiger Team" to identify and understand the likely sources for degradation in stacks. Significant testing was performed at multiple levels – materials characterization, single cells, coupons, and short stacks. Analytical models and post-test examinations have been used extensively to support this work.

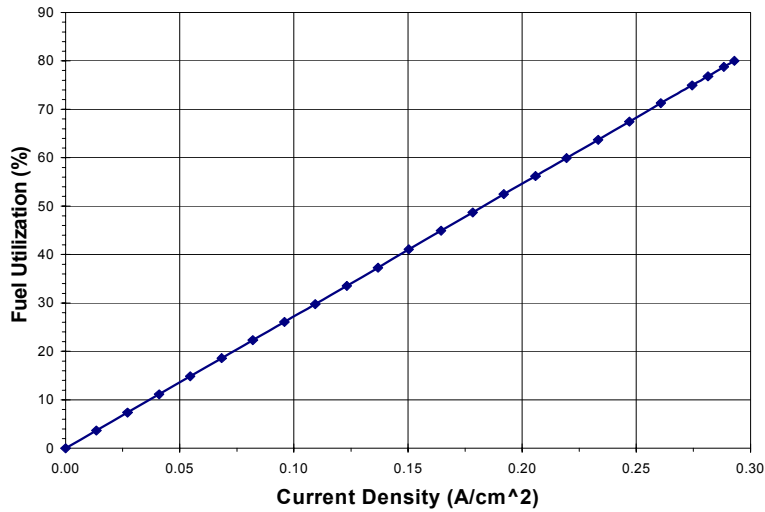


**Figure 8. Typical 5-cell short stack**

**RESULTS AND DISCUSSION:** During the SECA program, SOFCo performed more than 200 stack tests, with most of these being 2-5 cell short stack tests. As shown in **Figure 7**, early stack tests were generally performed using co-fired cells, and often resulted in high ASR values (typically starting at  $>2 \Omega \cdot \text{cm}^2$ ) and excessive degradation rates ( $>20\%$  per 500 hours). In early 2004, SOFCo began using 3YSZ electrolyte-supported cells obtained from an external supplier. These cells had a high mechanical integrity and stable performance; cell ASR was typically  $1.2 \Omega \cdot \text{cm}^2$  at  $825^\circ\text{C}$ . Using these cells, SOFCo was able to focus efforts on validating the stack design using the multi-layer ceramic interconnects, improving stack seals, and establishing methods to achieve stable cell-to-interconnect electrical contacts. During 2004, substantial progress was made in improving stack performance, as degradation rates were reduced to about 10% per 500 hours. However, ASR was about 3X higher than the target value for Phase I and degradation rates were still excessive. Toward the end of 2004, SOFCo began using higher performance ScSZ electrolyte supported cells for stack tests and established an intense internal effort ("Tiger Team") directed at resolving performance issues. Through 2005, significant progress was made in reducing stack ASR toward  $0.8 \Omega \cdot \text{cm}^2$  (using cells having an ASR  $\sim 0.7 \Omega \cdot \text{cm}^2$ ), and degradation rates were reduced to about 2% per 500 hours.

As mentioned above, most of the tests used 2-5 cell short stacks, such as that illustrated in **Figure 8**. These stacks generally contained instrumentation allowing for separate measurement of the individual contributions to stack resistance. The seal between the cells and interconnects was accomplished using a specially formulated ceramic paste. The manifold arrangement for the SOFCo co-flow stacks is illustrated in **Figure 9**. The stacks used an integral manifold within the interconnects to distribute fuel entering the inlet slots over the entire active surface of the cells. Sealing between the stack and the manifold was generally accomplished using a ceramic fiber gasket.

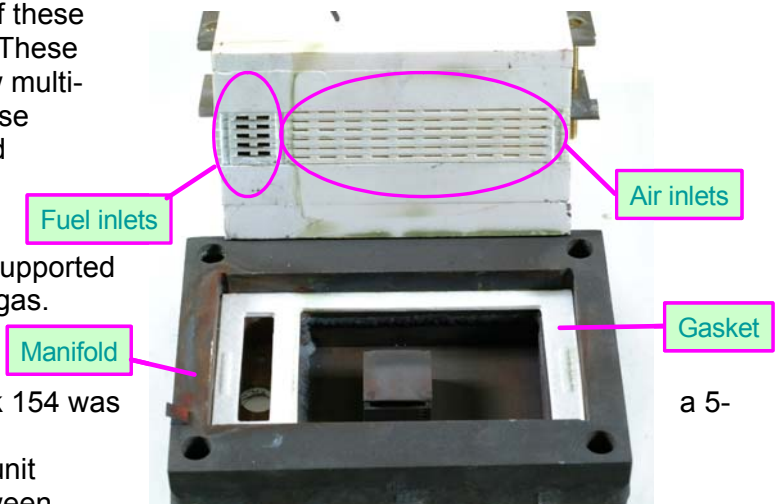
# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 10. Fuel utilization vs. current density for a short stack**

Upon switching from co-fired cells to the commercial 3YSZ electrolyte-supported cells, and obtaining relatively stable stack performance, a number of experiments were performed to validate the co-flow stack design and to assess the quality of the seals. As an example, flow measurements were made for the fuel flow channels to assess the uniformity of fuel flow through interconnects. It was determined that the SOFCo interconnect design achieved the designed flow distribution with a variation of +/- 5% across the flow channels. Moreover, electrochemical modeling confirmed that a 5% variation in fuel flow would support fuel utilization levels in excess of 85%. Separate experiments were performed to assess seal quality. In one such experiment, current density was increased at a constant H<sub>2</sub>/N<sub>2</sub> fuel flow rate for a short stack. As shown in **Figure 10**, a fuel utilization of 80% was achieved and a linear relationship between fuel utilization and current density was observed. This data, along with measured temperatures, was analyzed to confirm that less than 5% of the fuel was being lost due to seals and other factors. Separate experiments showed that fuel utilization levels in excess of 85% could be achieved. Moreover, some of these experiments included thermal cycling. These tests confirmed that the SOFCo co-flow multi-layer interconnect and stacks using these interconnects could achieve the desired performance levels.

Many of the early short stack tests performed using the 3YSZ electrolyte-supported cells used humidified H<sub>2</sub>/N<sub>2</sub> as the fuel gas. In early 2004, the first stack test using reformed natural gas (from a CPOX reformer) was performed. Stack 154 was cell stack that used commercial 3YSZ electrolyte-supported cells and repeat unit instrumentation (i.e., a voltage tap between each interconnect-cell repeat unit). The results for this stack are shown in

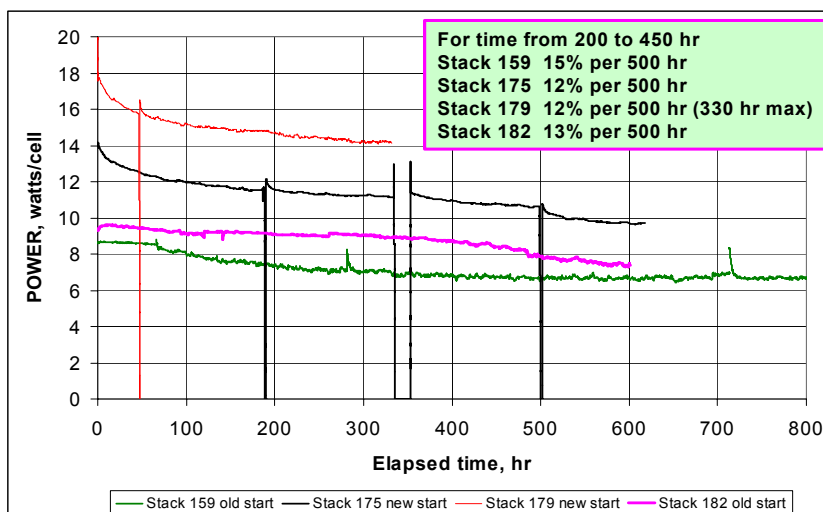


**Figure 9. Manifold arrangement for short stacks**

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

**Figure 11.** The stack was operated at a fuel utilization of 60% for about 1800 hours. The inlet fuel gas composition was: 47.5% nitrogen, 29.9% hydrogen, 15.8% carbon monoxide, 4.1% methane, 2.2% carbon dioxide and 0.5% argon. The fuel utilization was based on hydrogen, carbon monoxide and methane. The stack ran initially at 0.6 volts/cell and 17.3 amps, with an ASR of  $1.47 \Omega\text{-cm}^2$ . The average constant-voltage power degradation over the full 1800 hours of the degradation test was 3.6% per 500 hours. The average constant-voltage power degradation over the last 1500 hours was 2.6% per 500 hours.

While demonstrating reasonable stack performance using 3YSZ cells was encouraging, the power output per repeat unit was about a factor of three too low. A stack ASR of about  $0.75 \Omega\text{-cm}^2$  was needed for the C2 stacks and an ASR of  $0.5 \Omega\text{-cm}^2$  was needed to achieve the Phase I stack cost target. As discussed previously, a decision was made to switch to ScSZ electrolyte-supported cells to improve stack performance. It was expected that stack ASR of about  $0.75 \Omega\text{-cm}^2$  could be achieved using ScSZ cells obtained from a commercial supplier, and that an ASR approaching the  $0.5 \Omega\text{-cm}^2$  target might be possible by improving cell performance. The transition to ScSZ cells was initiated in late 2003 and the first stack tests were performed in 2004.



**Figure 11. Stack 154 performance with reformat**

Initial short stack tests using ScSZ electrolyte-supported cells showed improved starting ASR values (on the order of  $1.0\text{-}1.2 \Omega\text{-cm}^2$ ), but high degradation rates that often exceeded 15% per 500 hours. Changes were made to the air- and fuel-side contact inks used to electrically connect the interconnects to the cells and also to the procedure for the initial heat up of the stacks. These changes resulted in improved starting performance, as shown in **Figure 12**, but degradation rates were not significantly reduced. In particular, Stack 179 was the first stack to demonstrate the ASR targeted for the C2 demonstration. The initial repeat unit ASR was  $0.75 \Omega\text{-cm}^2$  at  $825^\circ\text{C}$ , with a current density of  $0.35 \text{ amps/cm}^2$  and a fuel utilization of 32% (using humidified  $\text{H}_2/\text{N}_2$ ). After about 330 hours, the test facility tripped and caused a problem with the subsequent data. The test was continued, however, and the degradation rate slowed to about 7% per 500 hours during the final period of operation.

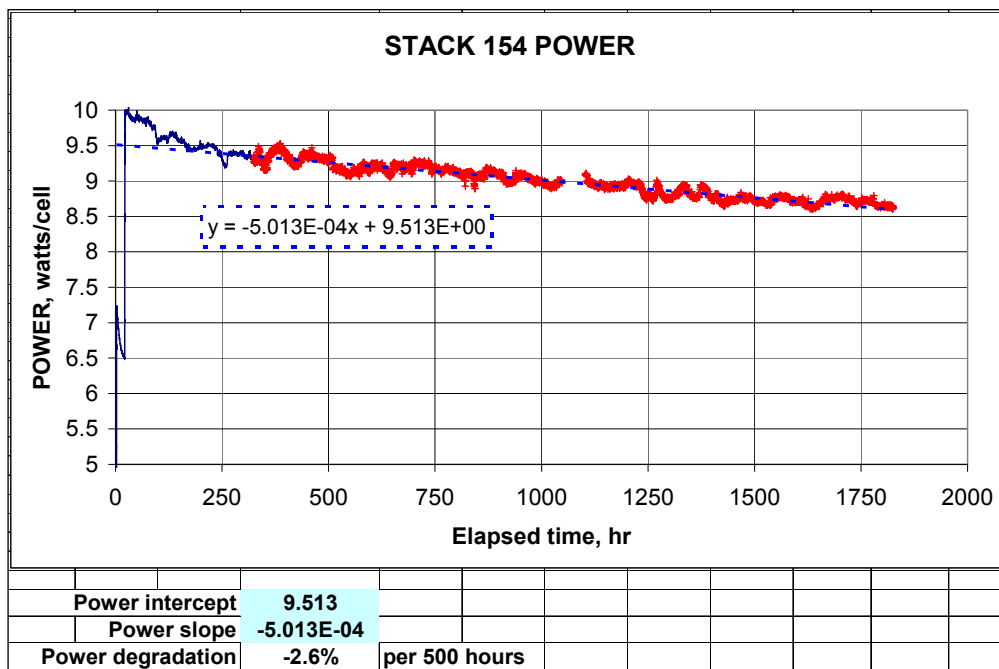
During the later half of 2004, stack tests consistently showed that degradation rates for short stacks using 3YSZ cells was on the order of 2-3% per 500 hours, while rates for shorts stacks using ScSZ cells were on the order of 7-15% per 500 hours. These stacks used similar



# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

materials and assembly methods during stack construction. Given that the cells showed relatively low degradation rates compared to those observed for the stacks, it was concluded that some fundamental issues existed for stacks utilizing the ScSZ cells.

In late 2004, an internal “Tiger Team” was organized to attack the stack degradation issue with much greater intensity and focus than previously directed at this area. The team reviewed cell and stack performance data, along with available information from post-test examinations. The team organized itself into small sub-groups directed at air-side contacts, fuel-side contacts, cell performance and interconnect resistance. A range of activities were conducted to identify and understand likely sources of degradation, if any, for each area using a number of approaches: analysis of cell and stack performance data, modeling, materials characterization, coupon tests, and post-test SEM examinations.



**Figure 12. Initial short stack testing with ScSZ cells**

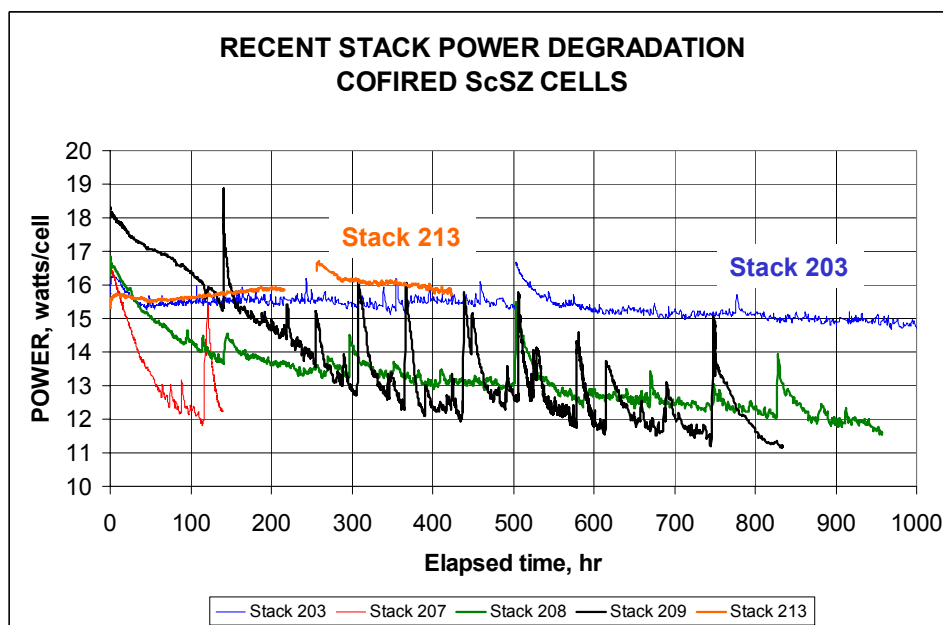
During Q1 2005, the SOFCo “Tiger Team” made significant progress in developing an understanding of stack performance degradation. Efforts to identify key sources for degradation and to define corrective actions were completed. As summarized below, the air-side and fuel-side electrical contacts between the cells and interconnects were found to be the primary contributors to excessive degradation rates.

- **Air-side Contacts** – The SOFCo investigation showed that failed connections often occurred at the interface between the cathode bond layer (CBL) and the cathode in the regions adjacent to the air-side bumps. The generally observed failures appeared to be the result of weak bonding between the CBL and the cathode and excessive shrinkage of the air-side bump material at high temperatures (i.e., >10% shrinkage during heat up and operation). A two-part corrective action was defined: a) pre-firing the CBL after screen printing onto the cathode to improve the bonding; and b) reformulation of the bump ink to reduce sintering shrinkage.

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Fuel-side Contacts - The diffusion of Ni from the bump material into the Pt-YSZ pads on the interconnects appeared to be one source for degradation. In addition, the relatively high CTE of the bump material (due to a high Ni content) relative to the interconnect may adversely impact integrity of the contacts. The investigation led to a two-part corrective action for the fuel-side contacts: a) adding Ni to the Pt-YSZ pads to reduce the driving force for diffusion of Ni out of the bumps; and b) reducing the Ni content of the bump material (to reduce CTE), and possibly using different Ni and YSZ powders for the ink formulation.

During the investigation, a series of short stack tests were performed, first to confirm certain hypotheses regarding likely degradation mechanisms and then later to evaluate corrective actions. Short stacks #203 and #213 provided valuable support for the hypothesis that a [ ] fuel-side connection is important in improving the integrity of electrical contacts and maintaining stable stack performance. These two stacks employed standard construction methods and materials, but used ScSZ electrolyte-supported cells in which a Ni layer on the anode had been added by the external supplier. [ ] Stack #203 was operated for over 1000 hours and exhibited a relatively low degradation rate of 3% per 500 hours. As shown in **Figure 13**, this was a significant improvement over the 10-20% per 500 hour degradation rate observed for prior short stacks [ ]. Short stack test #213 was conducted to validate this result. This stack showed almost no degradation over the first 250 hours of testing. However, this stack experienced an operational upset which appeared to affect performance. The stack was operated for >1000 hours, and the degradation rate during the final 800 hour period was >5% per 500 hours.



**Figure 13. Performance of short stacks #203 and #213**

The next advancement in stack performance was demonstrated with short stacks #223 and #224 (see **Figure 14**). The key results for these two stacks are summarized, as follows:

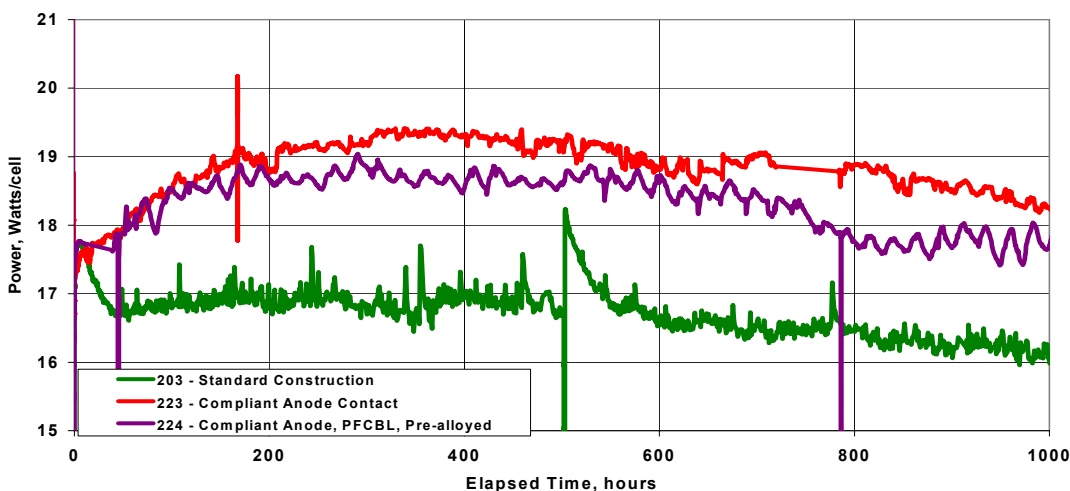
- Stack 223 – This stack was constructed using three ScSZ electrolyte-supported cells and the standard assembly method/materials. This stack included a [ ]

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

connection on the fuel-side of each repeat unit; the [ ] fuel-side connection is described later in this report. The initial ASR for the stack was  $0.81 \Omega\text{-cm}^2$ , but decreased over the first 380 hours of operation to a minimum of  $0.70 \Omega\text{-cm}^2$ . Over the next 700 hour period, the stack power degraded at a rate of 4% per 500 hours, and at 1050 hours of operation the ASR was  $0.75 \Omega\text{-cm}^2$ . The stack was then subjected to a period of high temperature operation ( $925^\circ\text{C}$ ) and a thermal cycle; higher than expected degradation was observed.

- Stack 224 – This stack was also constructed using two ScSZ cells, but used the two corrective actions selected by the team: a) cathode bond layer pre-fired at  $1000^\circ\text{C}$ ; and b) addition of Ni to the Pt-YSZ pads on the fuel-side of the interconnects. This stack also included the [ ] connection on the fuel-side of each repeat unit. After undergoing a pre-treatment cycle, the initial ASR was  $0.79 \Omega\text{-cm}^2$ , but decreased to about  $0.71 \Omega\text{-cm}^2$  during the initial 250 hours of operation. Over the next 750 hour period, the stack power degraded at a rate of 4-5% per 500 hours (ASR =  $0.74 \Omega\text{-cm}^2$  at 1000 hours). The stack was then subjected to a number of different operating conditions. A post-test examination was performed after terminating the test.

While special tests suggest that the two selected corrective actions should provide for more stable electrical connections, improved stack performance was not observed (comparing Stacks 223 and 224). It is likely that the [ ] connection substantially improved the robustness of the electrical contacts within the stack, but some other degradation mechanism was still active. At this point, a decision was made to perform a long-term test to determine whether the two corrective actions had a measurable impact on stack performance.



**Figure 14. Performance of short stacks #223 and #224**

It should be noted that detailed performance analyses for both stacks suggested that the electrical connection between the outer interconnects and the metal current collectors was degrading more rapidly than desired. A separate effects test confirmed this observation, and suggested that some portion of the 4-5% per 500 hour degradation was related to the current collector attachment. As a result, evaluation of an alternative current collector attachment method was performed, leading to the development of a [ ] connector (placed between the outer interconnects and the metal current collectors). Stack #232 was constructed to validate the benefits of [ ] fuel-side connections and [ ] current collector

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

connections. This stack also included the pre-alloyed pads on the fuel-side of each interconnect. Initial stack ASR at 825°C was 0.69  $\Omega$ -cm<sup>2</sup>; performance improved to an ASR of 0.59  $\Omega$ -cm<sup>2</sup> after about 450 hours (power density >280 mW/cm<sup>2</sup> at 0.7 volts), as shown in **Figure 15**. Thereafter, stack performance began to degrade at a rate of slightly less than 4% per 500 hours (from peak power). At 2732 hours, the stack ASR was 0.77  $\Omega$ -cm<sup>2</sup> at 60% fuel utilization. The stack temperature was then increased to 875C and held for about 200 hours; during this period, stack degradation was about 3% per 500 hours. It should be noted that testing of single cells also shows similar trends, with degradation rates on the order of 3% per 500 hours. Given these results, it now appears that much of the observed stack degradation is related to the cell, with little or no contribution from the interconnect and electrical connections. Moreover, it is believed that aging of the ScSZ electrolyte may account for more than one-half of the degradation during the first 2000 hours of operation. The stack test was terminated in Q4

**Average RU Power Degradation - Stack #232**



**Figure 15. Long-term performance for stack #232**

2005 at the end of the Phase I period, followed by post-test examinations.

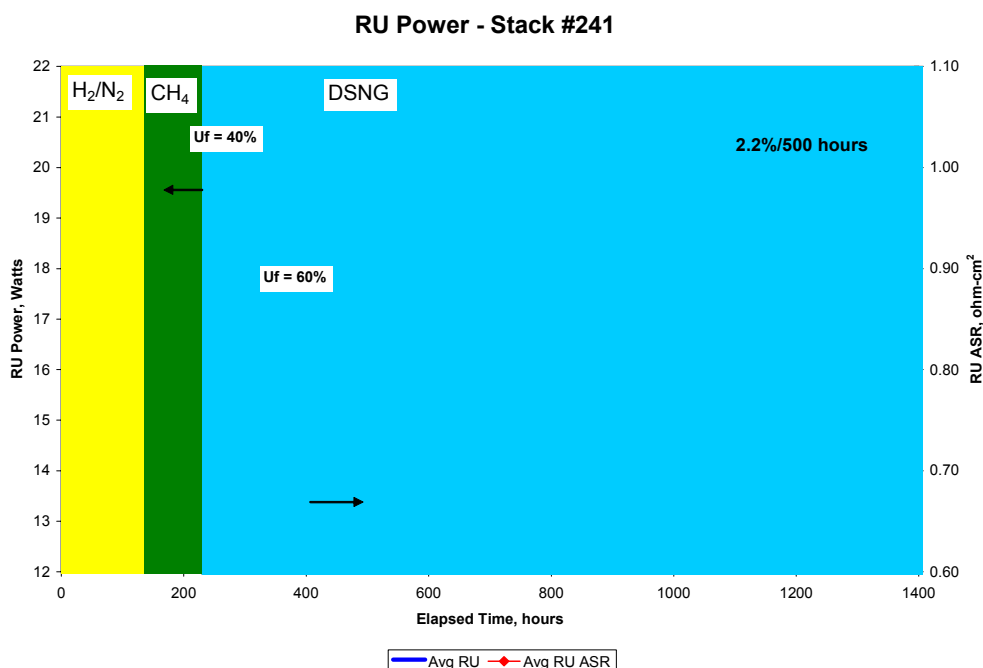
Toward the end of the Phase I period, two final short stacks were tested to evaluate stack performance with reformed natural gas:

- **Stack 240** – This short stack was constructed using ScSZ electrolyte-supported cells and the standard construction method, except that the [ ] fuel-side connection was not used. Testing was initiated using the H<sub>2</sub>/N<sub>2</sub> mixture, and was then switched to reformed natural gas (without any clean-up). The stack was operated under steady-state conditions for more than 1400 hours with a relatively low degradation rate of about 1.5% per 500 hours. The stack ASR was on the order of 0.95  $\Omega$ -cm<sup>2</sup> at 825°C and a fuel utilization of 46%. This stack showed relatively stable performance with reformed natural gas, in contrast with other recent stacks using the [ ] fuel-side connection where ASR was much lower, but performance was degraded when reformed natural gas was used (without clean-up).

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Stack 241** – This short stack was constructed using ScSZ electrolyte-supported cells and the standard construction method, including the [ ] fuel-side connection. This stack was operated on H<sub>2</sub>/N<sub>2</sub>, reformed bottled methane, and reformed desulfurized pipeline natural gas. That is, a clean-up process was used for the natural gas prior to reforming. This stack was operated for over 1400 hours at 825°C and a fuel utilization of 60%. As shown in **Figure 16**, power increased over the initial 700 hour testing period (power density ~ 270 mW/cm<sup>2</sup>), and then decreased at a rate of about 2.2% per 500 hours (from peak power). The stack ASR at the end of the test was about 0.68 Ω-cm<sup>2</sup>.

The low degradation rates observed for both of these tests represented major accomplishments for the SOFCo effort. In particular, Stack 241 demonstrated that both high power density and low degradation rates could be achieved when operating on natural gas. Such degradation rates would be expected for the ScSZ cells, given the documented aging of the electrolyte during the initial 1000-2000 hours of operation.



**Figure 16. Performance for stack #241 operated on desulfurized NG**

**CONCLUSION:** During Phase I of the SECA program, SOFCo made significant progress in improving stack power density and reducing degradation. By implementing ScSZ electrolyte cells, stack ASR was reduced from about 1.4 Ω-cm<sup>2</sup> to less than 0.7 Ω-cm<sup>2</sup> at 825°C and high fuel utilization. An internal “Tiger Team” performed an extensive study and developed a fundamental understanding of the primary sources for excessive stack degradation; key mechanisms were identified and corrective actions were defined. As a result of this effort, stack degradation was reduced from 10-20% per 500 hours to about 2% per 500 hours, and thereby meeting the Phase I objective. Stack performance now appears to be largely driven by the cell behavior.

### PCU (TALL STACK) DEVELOPMENT

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

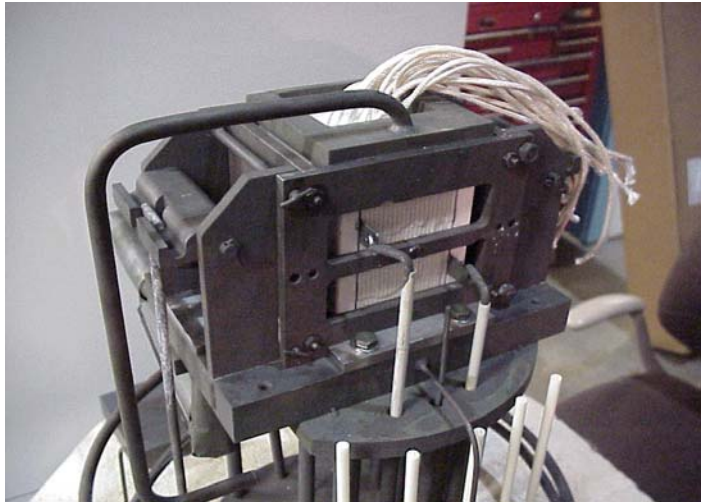
**OBJECTIVE:** Develop 10-cm full-size stacks (70 cells) that meet Phase I performance targets and supply four stacks for use in the C2 demonstration test.

**EXPERIMENTS AND RESULTS:** In the fall of 2003, SOFCo initiated concerted power cell unit (PCU) development activities. This work was initially aimed at developing a 48-cell stack design using 3YSZ cells for use in the C1 system and subsequently a 70-cell stack design using ScSZ cells for use in the C2 Phase 1 Demonstration System. The SOFCo “tall” stack design is a horizontal, co-flow configuration that simplifies manifolding of the inlet and outlet gas streams and also results in more uniform loading of each cell and interconnect than a vertical design. The PCU is contained within a cage-like structure to ease handling and to provide for axial load application. The initial PCU development activities used a 20-cell stack configuration that was tested in a modified version of the test stands normally used for short stack tests (generally 10 cells or less). Subsequently, 47-cell and 48-cell stacks were tested in a larger Horizontal Stack Test (HST) Facility prior to construction of the two stacks required for the C1 system test. All of these early stacks used nominally 140  $\mu\text{m}$  thick 3YSZ electrolyte-supported cells. After successful completion of the C1 system test at CPG, PCU development continued with scale-up to 70-cell stacks using the new nominal 110  $\mu\text{m}$  ScSZ electrolyte-supported cells. The following summarizes the significant PCU development stack tests and related activities from the fall of 2003 through the end of calendar year 2005.

## 20-Cell Stacks

A 20-cell mockup (without operating cells) and three operating 20-cell stacks were built and tested during development of the initial PCU concept from November 2003 through February 2004. The 20-cell stacks used 3YSZ cells. A 20-cell stack installed in its test stand is shown in **Figure 17**. These tests provided key information that allowed refinement of the PCU design concept prior to construction of the 48-cell development stacks and, subsequently, the two C1 system stacks. The 20-cell stack tests were key to refining the tall stack PCU design in the following areas:

- Assembly process
- Cage design
- Cell flatness requirements
- Stack sealing requirements
- Stack end load requirements
- Manifold gasket materials and application
- Stack insulation requirements
- Inlet-to-outlet thermal gradients
- High stress locations in interconnects and methods for mitigating



**Figure 17. 20-cell stack in test stand**

At the completion of the 20-cell test sequence, the PCU design had been highly refined and was ready for scale-up to a nominal 48-cell stack.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## C1 System Stack Development

The C1 system test was an intermediate test of a nominal 1kW fuel cell system aimed at providing CPG with stack operating experience and to assist in the design of a fuel cell automated control system. The C1 system required two 48-cell stacks using 3YSZ cells to achieve the nominal power output. Prior to building the actual C1 stacks, two stack tests were run to verify the scale-up from 20 cells to 48 cells. The following outlines the two C1 development stacks and general results from the C1 system test relating to stack design and general performance issues.



**Figure 18. 47-cell stack in test stand**



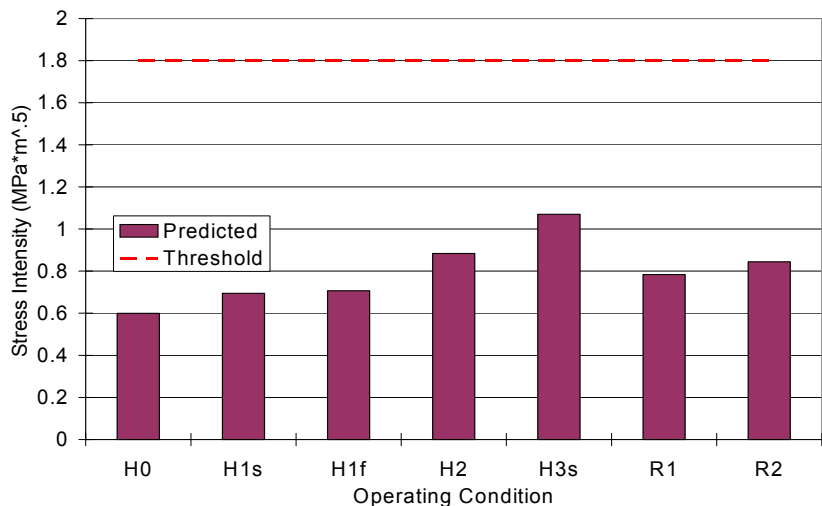
**Figure 19. HST Facility**

### 47-Cell Stack #162

The first C1 development stack had 47 cells (3YSZ) and was built in March 2004. This stack incorporated improved stack insulation and was the first PCU stack to be run in the Horizontal Stack Test (HST) Facility using reformed (CPOX) natural gas. The stack is shown in **Figure 18**, after installation in the HST. The HST Facility is shown in **Figure 19**. This stack was tested initially using hydrogen fuel and then switched over to reformed natural gas. The stack produced between 400 and 500 watts of power at fuel utilizations up to 70%. This stack showed that the PCU design and the 3YSZ cell could meet the C1

system performance requirements. Detailed thermal and FEA stress analyses (summarized in

**Figure 20**) were performed at key operating points to predict the stress state within the stack. This analysis showed the operating stress intensity levels for the each of the operating conditions were well below the allowable levels (red dashed line). This provided feedback on stack operating conditions relative to stress levels and began to define the boundaries for safe operating modes and transitions between modes for the PCU design.



**Figure 20. FEA stress analysis results**

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

At approximately 300 hours into operation, a malfunctioning load controller caused a high current surge through the stack and appeared to cause damage in one group of cells. Two days later, a reformer trip caused some additional upset and negatively affected the damaged group of cells. The stack was run for several more days until shutdown after 434 hours of operation. Post-test examination confirmed that the higher stressed outlet edge of the interconnects were undamaged, as predicted by the thermal and FEA analysis. Generally the inlet edge of the stack is expected to have lower operating stresses, however, three interconnects had inlet edge cracks. Two of the cracks were at thermocouple locations. These latter two cracks are believed to be related to the over-current condition and likely high current flow through the thermocouple sheaths, which were in electrical contact with the facility. One cell, adjacent to one of the cracked interconnects, was also damaged. This stack provided key operating experience with a tall stack and confirmed that the basic PCU design was viable.

### **48-Cell Stack #168**

A 48-cell stack (3YSZ cells) was built in May 2004 and incorporated a new C1-type outlet manifold and new outlet edge insulation material to better streamline the stack outlet region. These features are shown in the **Figure 21**. This stack operated on hydrogen for 250 hours and on reformed natural gas for 500 hours. The stack was operated at a maximum power of 480 watts and achieved fuel utilization as high as 82%. This stack was used to demonstrate power ramps from zero to full load at three different air flows to help finalize C1 operating conditions and transitions from full power to hot standby and back. Toward the end of the test, this stack



was

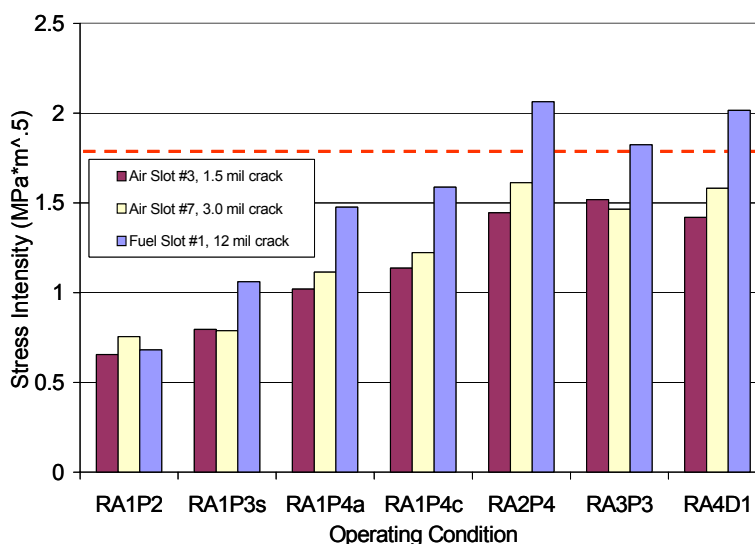
**Figure 21. 48-cell stack #168 in the HST Facility**

deliberately operated at conditions which exceeded the recommended stress levels to further define and confirm those limits and the effect on stack operation. The stack operated well at all conditions, even those over-stress conditions. Although somewhat higher initially, the degradation at steady state over the last 164 hours of operation was 4% per 500 hours. This stack was shutdown for inspection just prior to the start of construction of the two C1 stacks.



## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

Post-test inspections showed that a high percentage of the interconnects had fine, very tight cracks at the outlet edge, along with 3 cracked cells. None of these cracks appeared to have resulted in air-to-fuel burning. As indicated in **Figure 22**, the stress analysis had predicted that the allowable stress intensity (dashed red line) was exceeded during three sets of operating conditions. This test confirmed that the stress analysis predictions correlated well with actual observations and that the over-stress condition could cause cracking. This test provided all of the required information for moving forward with the construction of the two C1 system stacks and defined safe operating modes that met C1 test requirements.



**Figure 22. FEA stress analyses for stack #168**

### **C1 System Stacks #177 and #178**

Two 48-cell PCU stacks were constructed in June 2004 for the C1 demonstration. These stacks used 3YSZ cells, like the preceding PCUs. After assembly, the stacks were packaged and shipped to CPG, where they were installed in the C1 system by SOFCo personnel. **Figure 23** shows the two stacks installed in the C1 unit. The C1 system was started up on July 4, 2004. The stacks were pre-conditioned (solvent burn-off and anode reduction) in place. Shortly before switching over to the reformer, the system controller software errantly started the reformer air flow. This exposed the stack anodes to 700°C air for approximately 12 minutes, which would have re-oxidized the anodes.



**Figure 23. Two 48-cell stacks installed in the C1 module**

Although SOFCo had run tests showing that short stacks could survive this re-oxidization event, it is problematic for any stack to do so. Shortly after this event, a group of cells in one of the two stacks started showing abnormally high ASR. Subsequently, low fuel utilization conditions were identified that appeared to minimize the effect of the damage and the stacks continued to run. The control system development proceeded as planned and all objectives for the test were achieved, albeit at lower power levels than planned. These stacks and the C1 system provided

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

CPG with early stack operating experience and provided key information required for developing automated control algorithms.

## **C2 System Stack Development**

Following the completion of the C1 testing, PCU development concentrated on the stack developments that would be required for the C2 system demonstration. The C2 system would require four 70-Cell stacks and would use 110  $\mu\text{m}$  thick ScSZ electrolyte-supported cells to achieve lower ASR. Early shipments of the ScSZ cells proved to be too weak for use in tall stacks, where the stress levels are higher than in short stacks. While the vendor continued to refine the manufacturing process for the ScSZ cells to reduce defects and increase their strength, the initial ScSZ cell deliveries were used in early single cell and short stack (typically <5 cells) tests to obtain ASR and degradation information. To continue with the PCU development while awaiting higher strength ScSZ cells, the first C2 PCU stack used 3YSZ cells, like the previous C1 development stacks. Subsequent C2 PCU development stacks did use ScSZ cells, although early stacks were still using cells that had less than the specified strength. The cell strength gradually increased with time and the last two stacks in the series had full strength cells. The following summarizes general results from the C2 PCU development stacks, tested from September 2004 through December 2005.

### **48-Cell Stack #189**

The first PCU test in the sequence leading up to the C2 system stacks was another 48-cell stack. Because of limited availability of the ScSZ cells with acceptable strength, this stack retained the 3YSZ cells used in previous PCUs. The primary purpose of this stack was to start exploring the operating envelope for the C2 system.

This included plans for:

- Transient tests to determine the optimum methods for moving from full power to hot standby and return;
- Cycling to room temperature and restart, including a rapid startup; and
- Fuel utilization cycling tests.

48-cell Stack #189 was assembled and installed in the HST Facility in September 2004 with testing continuing through December 2004. The assembled stack is shown in **Figure 24**.



**Figure 24. 48-cell stack #189**

This stack was pre-conditioned in the HST using a more optimum startup and anode reduction procedure. Upon initial operation, the stack ASR was consistent with previous stacks using 3YSZ cells, approximately  $1.35 \Omega\text{-cm}^2$  (825°C). Data taken on September 18 was analyzed in detail to represent the initial stack performance as follows:

- 0.141 watts/cm<sup>2</sup> (491 watts for 48 cells with 72.7 cm<sup>2</sup> active area) at 0.676 volts/cell and 15.1 amps;
- Fuel utilization was 71%;
- Stack average temperature was 823°C (751°C inlet and 895°C outlet);
- Reformed natural gas composition was: 45.3% N<sub>2</sub>, 31.6% H<sub>2</sub>, 17.5% CO, 3.1% H<sub>2</sub>O, 1.6% CO<sub>2</sub>, and 0.9% CH<sub>4</sub>; and

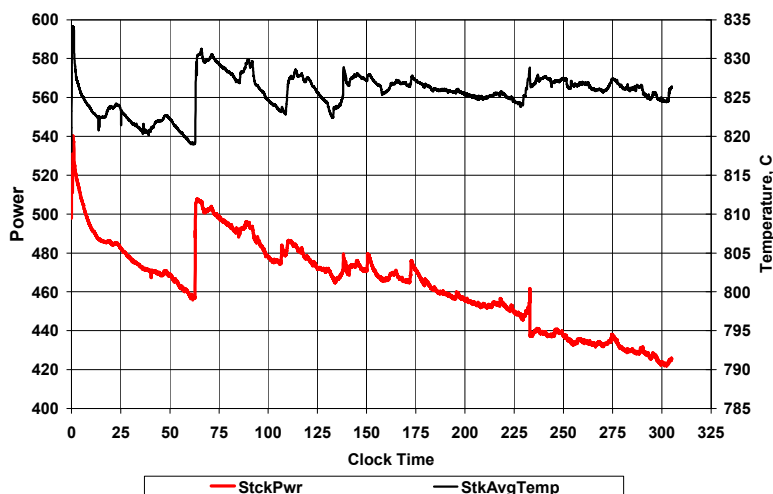
## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Performance was about 10% better than the previous full-size stack (#168) tested in the HST Facility and the improvement was attributed primarily to the new startup and anode-reduction procedure.

Shortly after startup of this stack, it was discovered during short stack testing that the ink used to create the interconnect contact pads was sub-standard, with higher resistance than normal. This was traced to a minor change in a preparation procedure that significantly affected the ink characteristics. This problem is believed to be responsible for higher than normal degradation that this stack experienced as testing progressed.

The first 288-hour test period covered steady-state operation. Then a 552-hour period of cyclic testing was performed. The first several power cycles were between powered operation and a hot idle (standby) condition (i.e. approximately 400 watts to 100 watts). This was followed by two cycles to room temperature at normal cooling and heating rates. The third cycle to room temperature and back included a faster heat up rate of 3°C/min. This is three times faster than the previous startup cycles and reduced startup time from 12 hours to 4 hours. This completed the 552-hour period of cyclic testing and brought the total test time to 840 hours. None of the cycles caused detectable changes in performance (power before and after the cycle was about the same).

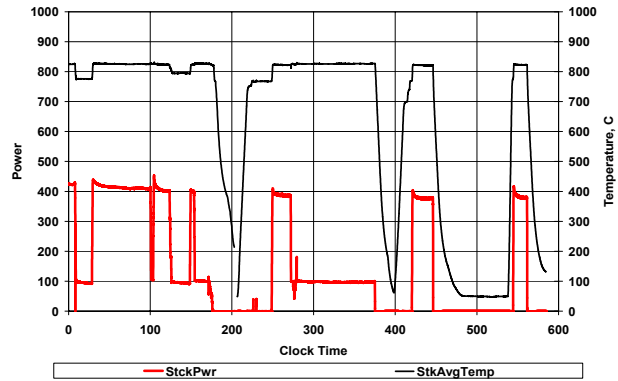
At this point, degradation calculations were made over the steady-state, cyclic and total test periods. The average degradation rate over the 288-hour steady-state period was 40% per 500 hours. This high degradation rate is believed to be related to the sub-standard ink used to print conductive pads on the interconnect, as described above. **Figure 25** shows the power and average stack temperature during this 288 hour period of operation from 17 to 305 hours.



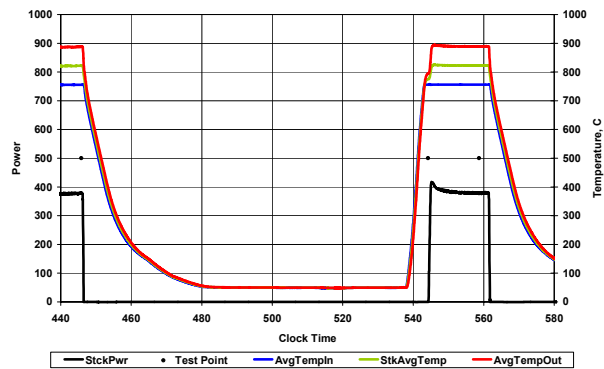
**Figure 25. Steady-state operation of stack #189**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

The average power degradation rate over the 552-hour cycle testing period was 8% per 500 hours. **Figure 26** shows the power and average stack temperature during this period. The 552-hour period is from 3 to 555 hours on the plot. The average overall rate for the entire 840-hour period was 19% per 500 hours. Note that the combination of power transition cycles and temperature cycles to room temperature and back had very little effect on stack performance, including one unplanned system trip. During one of the room temperature shutdowns, the HST Facility control system was modified to allow more automated control of the stack transients. **Figure 27** shows the rapid startup of the stack from room temperature to full power conditions (825°C average stack temperature) taking only four hours. There was no ill-effect from this rapid startup, with performance returning to pre-shutdown levels. One of the key results from this stack test was that various operating schemes for moving from full power to hot standby and back were thoroughly investigated and optimized to best meet C2 requirements. The stack stability during all of the transient testing, including five room temperature cycles, clearly demonstrated that stack performance was not significantly affected by these transients, thereby providing confidence in the basic PCU design and stack construction techniques. This stack was shutdown after over 1000 hours of operation, with about 70% of the time spent doing transient testing and the remainder at steady-state conditions.



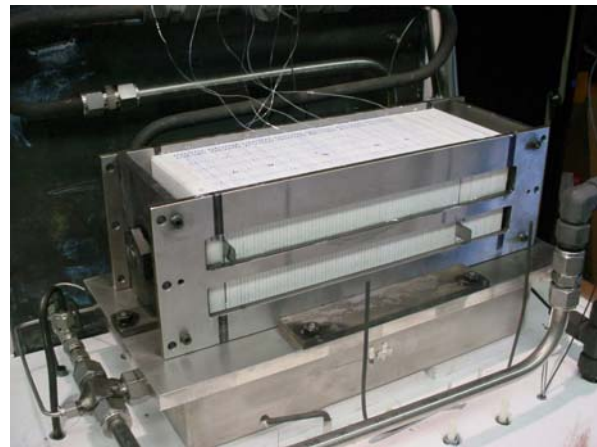
**Figure 26. Cyclic testing of stack #189**



**Figure 27. Rapid startup of stack #189**

## **70-Cell PCU Stack #214**

By March 2005, a sufficient quantity of ScSZ cells had been received to proceed with construction of a 70-cell stack using this new cell design (see **Figure 28**). There were basic two anode-side variations of the ScSZ cell with one using a vendor applied nickel conducting layer and the other without the conducting layer. There were also two methods for cutting the green electrolytes to size, and these were a conventional hot knife cutting operation and a laser cutting operation. Stack #214 incorporated cells with each of these variations.



**Figure 28. 70-cell stack #214**

The primary purpose for this stack was to:

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Gain experience at building and operating a full-size 70-cell C2 stack;
- Determine relative performance of ScSZ cells with and without the vendor-applied nickel layer on the anode; and
- Determine the acceptability of cells that were sized at the vendor using the laser cutting process.

The stack was installed in the HST Facility and pre-conditioned during startup. Initial operation used a H<sub>2</sub>/N<sub>2</sub> fuel mix. Almost immediately after startup, a non-uniform outlet temperature was observed in the section of the stack containing the laser cut cells (approximately 20 cells at one end of the stack). A high outlet temperature is usually an indication of a damaged section of the stack where fuel/air burning is taking place. This condition required the stack be operated at less than full power to prevent excess heat build up in the malfunctioning section. Under those limitations, a maximum power level of 850 watts at 70% fuel utilization was achieved. The stack was switched over to reformat fuel. With further degradation of the damaged section the maximum power on reformat was 700 watts and 50% fuel utilization. The section of the stack [REDACTED] was showing about double the degradation rate of a 10-cell section [REDACTED] that was located furthest from the damaged section (i.e., in the middle of the stack). An analysis of the data from this 10-cell segment of cells with the nickel layer indicated about 4% per 500 hours degradation.

Due to continued degradation of the stack and the desire not to cause any additional damage that would compromise post-test inspection, the stack was shut down. Post-test inspection of 25 cells from the damaged end of the stack (the rest of the stack was not disassembled) showed that 100% of the 20 laser cut cells were broken. A number of adjacent cells and interconnects were also broken from what was suspected to be a cascade failure from the damaged portion of the stack. There were two key conclusions from the short operation and post-inspection of this stack:

- The cell vendor was informed not to produce any additional cells using the laser cutting procedure. Evidently, this process results in some very undesirable stress state that is not apparent through inspection. Subsequent strength tests of these particular cells showed them to be very weak along the cut edges.
- The use of the [REDACTED] reduced power degradation. The vendor was told to provide all future cells with [REDACTED].

On the positive side, the assembly of the stack went very smoothly, showing that the scale-up from 48-cell stacks to 70-cell stacks did not introduce any stack build issues. Also, the best group of 10 cells was showing only 4% per 500 hours degradation early in the stack operation, where degradation is usually observed to be higher.

### **60-Cell PCU Stack #214A**

In April 2005, the 45 intact cells from Stack #214 were used in building a new 60-cell stack. Fifteen new cells and interconnects were assembled into a stack and the 45-cell segment from Stack #214 added to that new stack, thereby creating a 60-cell stack identified as #214A. The purpose of this stack was to:

- Gain additional operational time on a large stack by utilizing the good portion of Stack #214;
- Gain experience at rebuilding a portion of a stack;

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Process a stack using a separate Pre-conditioning Facility (PCF) for solvent burnout and anode reduction. This is the process planned for the C2 stacks. All prior PCU stacks had been pre-conditioned in the HST Facility; and
- Checkout performance and durability of some recently received ScSZ cells with defect indications.

It was noted immediately after construction that the new and old sections of the stack did not mate up as well as expected, primarily due to a slight skew in the original 45-cell section. The stack was installed in PCF and underwent the standard pre-conditioning conditions. The PCF can obtain very low power data from a stack as an initial check on performance and the stack seemed to be operating normally. After pre-conditioning, the stack was moved to the HST Facility where it was started up using a C2 startup schedule without any need for holds or anode reduction schedules. Shortly after startup, there were indications of stack leakage and the stack had to be shutdown almost immediately for inspection. The inspection confirmed that there was a major leak at the interface between the new and old sections of the stack. The specific procedure used for joining the two sections was identified as the primary cause of the problem and a new procedure was developed for use in future stacks. The experience gained with this stack and the development of a repair procedure was found to be invaluable on later PCU stacks where some required repairs such as interconnect/cell removal or replacement. Each of those repairs was successful and saved both time and dollars in the PCU development effort. The PCF also worked as planned and thereby established a viable pre-conditioning process for the C2 stacks, avoiding any need for special startup procedures or special gas compositions in the C2 system.

### **40-Cell PCU Stack #226**

The ScSZ cell supply continued to be low during the late spring of 2005, while the vendor tried to modify their process to reduce electrolyte defects and increase cell strength. Those cells that did arrive were showing numerous minor surface defects and had significantly lower strength and higher scatter in strength than desired. Efforts were made to thoroughly inspect and down-select only the best of the cells, but selection based on visual inspection could not rule out potential defects hidden under electrodes or defects too small to be seen, but significant to overall strength. It was decided to determine if the defects being seen were indeed unacceptable by subjecting them to actual stack testing conditions in a tall stack, where the stress state is higher than in a short stack (due to temperature gradients). Also, a [REDACTED] layer had been developed for use on the anode-side of the cells and this was showing great promise in short stack testing. The decision was made to proceed with a 40-cell C2 PCU stack test whose purpose would be to:

- Continue PCU development in terms of stack performance and degradation measurements;
- Determine the acceptability of the cells with defect indications; and
- Test the new [REDACTED] construction in a tall stack by assembling 20 cells with the standard construction and 20 cells using the new [REDACTED] construction. All of the cells were from the same Lot of cells that had numerous surface defect indications.

This stack was started up in May 2005 directly in the HST Facility to save the time required to remove a stack from the PCF and re-install it in the HST. Shortly after reaching operating conditions, there were indications of damage in the section of the stack without the [REDACTED] layers. The stack was immediately shutdown for inspection. The section of the stack without the [REDACTED] anode layer was removed for disassembly. In addition, three of the adjacent cells and interconnects from the [REDACTED] section of the stack were removed to determine if there

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

was any damage in that section, leaving a 17-cell section [ ] intact. Eighteen of 20 cells/interconnects in the [ ] section were damaged, likely from a cascade failure caused by one or more defective cells. The inspection of the three [ ] interconnects removed showed no damage and very good adhesion of the [ ] layer. [ ]. This was an important finding and needed to be confirmed. It was decided to re-install the intact 17-cell section with the [ ] layers for further testing.

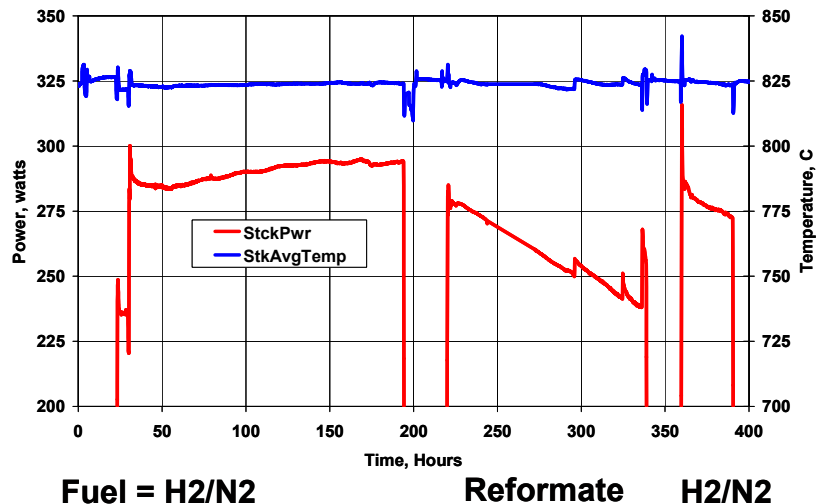
## 17-Cell PCU Stack #226A

A new current collector was installed on the end of the stack that had the 23 cells and interconnects removed and the stack was re-installed in the HST. The purpose of this stack test was to:

- Continue PCU development;
- Further investigate the potential stress reduction and performance benefit of the [ ]; and
- Collect additional operational data on ScSZ cells and performance with reformat.

This stack was installed in the HST Facility and started up and run for the first 200 hours on an H<sub>2</sub>/N<sub>2</sub> mix. During this time, the stack power actually continued to increase (see **Figure 29**), very similar to what was seen in single cell and short stack tests using the [ ] anode layer. The maximum power on hydrogen was 294 watts or 17.3 watts / cell (or 240 mW/cm<sup>2</sup>) and the stack ASR was 0.82 Ω · cm<sup>2</sup>. At about the 200 hour point, the stack was switched over to natural gas reformat. During the switchover, there was a perturbation which resulted in a brief exposure of the fuel side to O<sub>2</sub>/N<sub>2</sub> and a temperature gradient reversal in the stack (higher temperature at inlet than at outlet). Shortly thereafter, the stack began degrading in power. At the time of the incident, it appeared that the perturbation had caused damage and the subsequent degradation of the stack. However, it was later found in testing with other tall and short stacks that the reformat fuel was a possible source for the degradation seen in this stack and not any damage to cells/interconnects from the switchover perturbation. The stack continued to operate for a total of 492 hours, with a brief return to hydrogen fuel at the end of the test. The stack was shutdown and inspected.

The inspection showed two interconnects with relatively small cracks on the inlet edge and two cells, not adjacent to those interconnects, with small cracks near the inlet edge. Inlet edge cracks are not typical since the higher stresses are generally expected near the outlet edge where temperature gradients are higher. This damage was attributed to the reversal in temperature (higher at inlet) during the switchover to reformat and/or the exposure of the



**Figure 29. Test results from 17-cell stack #226A**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

fuel-side to O<sub>2</sub>. In general, the relatively minimal damage sustained seemed to confirm that the [REDACTED] layer does provide [REDACTED]. (In hindsight, the minimal damage also confirmed that there was another explanation for the high degradation seen when running on reformat.) The damage was in marked contrast to the nearly 100% cell failure that was in the [REDACTED] cell/interconnect pairs (same cell Lot) in Stack #226. Based on the success of this test and short stack test results, it was decided to use the [REDACTED] construction for future large stacks and for the C2 stacks. This decision was based on the benefits of improved starting ASR, subsequent decreasing ASR over the first couple hundred hours of operation, and [REDACTED]

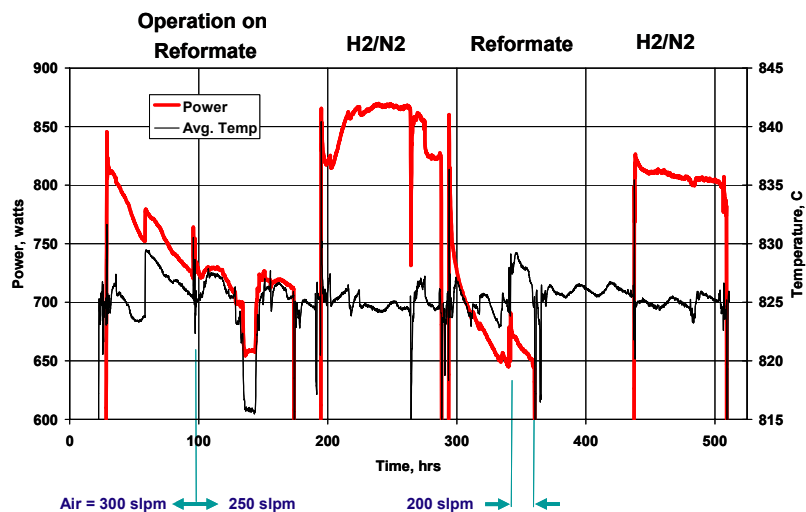
## **50-Cell PCU Stack #233**

The next C2 PCU stack was constructed in early July 2005. As stated previously, the strength of the cells available at that time was not as good as desired, being similar in strength and visible defects to those cells used in Stack #226. However, there were operating issues that had to be resolved and Stack #233 was constructed using the best available cells and the [REDACTED] fuel-side connection. Stack #233 was built to investigate several new issues:

- The potential performance issues related to running on natural gas reformat;
- Confirmation that a [REDACTED] stack benefits the overall stack ASR, improves degradation, and protects weak cells;
- Gain additional experience with running the Pre-conditioning Facility;
- Implementation and confirmation of performance improvements seen with a new startup procedure developed in single cell and short stack testing; and
- Testing of all of the features contemplated for the C2 stacks, including:
  - New cage material
  - New end loader
  - [REDACTED]
  - New [REDACTED] current collector connections.

Stack #233 was initially installed in the Pre-conditioning Facility where it underwent solvent burnout, anode reduction, short-term high-temperature conditioning, and then short-term operation at normal operating temperature. Limited low power operation confirmed the stack was performing well. Leak checks confirmed a good stack build.

The stack was shutdown and moved to the HST facility where it was started up directly on reformat. Initial leak checks confirmed the stack still had good sealing after the pre-conditioning cycle, removal and the re-installation procedure. The initial power (not necessarily maximum power) was 816 watts or 16.3 watts/cell. The stack immediately started showing degradation upon reaching operating



**Figure 30. Test results from 50-cell stack #233**



## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

temperature. Although all of the data was not in from other tests at this point, it was later shown that this high degradation was due to operating on reformed natural gas rather than the much lower degradation rate seen when operating on H<sub>2</sub>/N<sub>2</sub>. The degradation continued until the 175 hour point at which time the stack was taken to open circuit voltage (OCV) and then switched over to hydrogen fuel. The data shown in **Figure 30** shows that the stack power immediately increased at this point and continued to show steady improvement. The maximum power achieved on hydrogen was 866 watts or 17.3 watts/cell. This is the same watts per cell as was achieved with the 17 cells of Stack #226A. Stack #233 was alternately operated on reformat and hydrogen one more time each and showed improved power output on hydrogen and nearly stable operation, while for each cycle on reformat the stack exhibited much higher degradation.

During the filling of a bubbler (used to humidify the hydrogen fuel), a significant amount of excess moisture was introduced into the stack due to a malfunctioning level gage. Immediately, there was a small rise in the outlet temperature at one end of the stack. Although not deemed significant at the time, during a later change in stack operating conditions, the outlet temperature in this section of the stack rose rapidly, indicating stack damage at that end. The stack was shutdown in August at about the 500 hour point in operation for inspection of the damaged end. Thirty-five of the 50 cells were kept intact for possible restart as Stack #233A. An examination of the damaged end of the stack confirmed that 13 of the 15 interconnects and 10 of the 15 cells had cracks on the inlet edge. Again, the inlet edge is generally under relatively low stress during normal operation, so the damage was almost assuredly caused by the bubbler overfill accident which would have been seen primarily at the inlet. There was some apparent damage to the outlet edge of a few interconnects in the 35 cell section, likely from the same incident, but that part of the stack generally looked to be in good condition and it was decided to re-install that section as Stack #233A.

### **35-Cell Stack #233A**

Two new [REDACTED] current collectors were installed on the remaining 35 cells and this stack was installed directly into the HST, since there was no need for pre-conditioning. The primary purpose of this stack test was to investigate the degradation issue related to running on reformed natural gas. As such, at various times this stack was run on H<sub>2</sub>/N<sub>2</sub>, reformed bottled methane (<1 ppm Sulfur), reformed natural gas, and reformed natural gas with [REDACTED]. This test was run in conjunction with single cell tests and short stack tests that were also running on reformat and various other gas combinations. The net result of this concerted testing program to identify reformat operating issues, was that two stack fuel supplies were found that provided acceptable stack degradation performance during normal C2 long-term steady-state operating conditions. These were:

- Reformed bottled methane (<1ppm Sulfur); and
- Reformed natural gas using the [REDACTED].

Stack #233A continued to operate and provided valuable performance data related to reformat operation. At shutdown, the total operating time for the components in Stack #233 and #233A was 2300 hours. Operation included 11 power cycles, two room temperature cycles, a rebuild of Stack #233 into Stack #233A, and the wide variety of fuel types described above. Upon post-test inspection, two cells appeared to have been cracked sometime during operation and a number of the interconnects had unusual outlet edge cracks, most likely related to the bubbler overfill that damaged the opposite end of the stack at the inlet. None of these cracks appeared to effect the stack operation at the end of the test, since there were no obvious indications of stack damage in terms of operating issues or abnormal temperature measurements. Overall, this stack:

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Successfully demonstrated the pre-conditioning process and stack transfer to the HST (or other) test system;
- Had a starting ASR of  $0.9 \Omega\text{-cm}^2$ ;
- Exhibited an overall degradation of about 7%/500 hours, including:
  - effects of the initial bubbler damage
  - effects of the rebuild
  - 11 power cycles on various gas compositions, some of which caused severe degradation
  - power recovery when switching to more benign gas compositions
- Solved the natural gas reformat degradation problem by using [REDACTED].

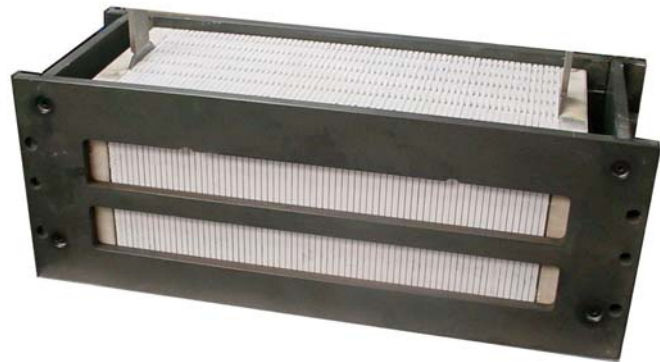
### **70-Cell PCU Stack #244**

In mid-November 2005, PCU Stack #244 with 70 cells was constructed using the best of the available cells, interconnects, and construction approaches/materials developed during the two-year PCU development program (See **Figure 31**).

The plan was to run this stack on reformed natural gas [REDACTED] developed for Stack #233A. Stack #244 was installed directly in the HST for pre-conditioning in that facility to minimize turnaround time. During the pre-conditioning cycle there was an indication of what later turned out to be a leak in the inlet manifold gasket. This

was a very unusual condition, since no previous PCU stack had experience any inlet or outlet gasket problems. The PCU horizontal design is relatively simple to seal. Through slight modification of the pre-conditioning procedure, the leakage problem was minimized and the pre-conditioning cycle was successfully completed. The stack was shutdown and removed from the facility to confirm the source of the leakage and to inspect the stack for any damage. The inspection showed that the cage had apparently moved relative to the stack and had likely lifted the stack load off of the gasket, creating the leak. A number of slight modifications were made to the cage to prevent a further occurrence. The stack inspection showed no damage to the stack. However, during handling in preparation for re-installation, cell #68 (which was protruding slightly out of the side of the stack) was damaged. Normally, a broken cell would be replaced, but since the cell was near the end of the stack, the last 3 cells and interconnects were just removed and a new current collector was installed. Stack #244 now became a 67-cell Stack and because there were no new cells installed, the pre-conditioning cycle did not have to be repeated. Stack #244 was re-installed in the HST and started up in early December 2005.

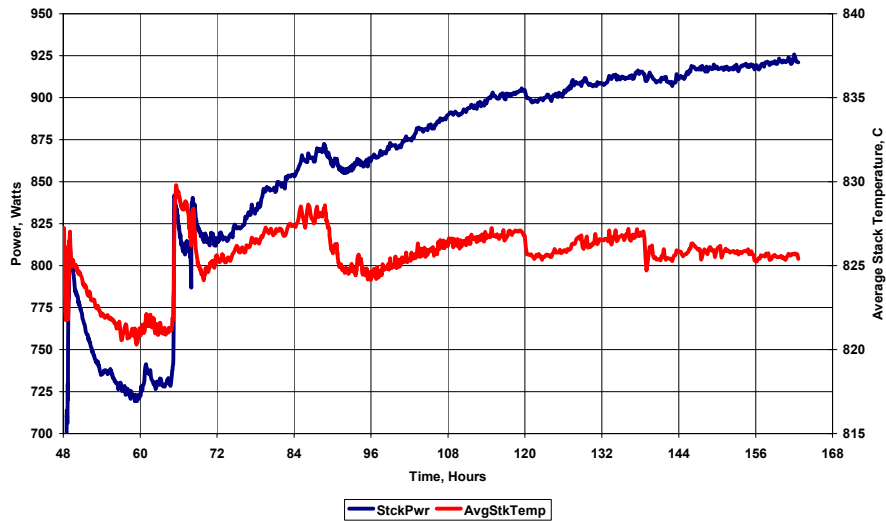
Stack #244 was initially set to 800 watts power output and put on automatic control over the first weekend. **Figure 32** shows the initial performance of the stack. The stack power dropped with temperature over the weekend, but quickly recovered when the temperature was increased at about the 65-hour point. The initial power sensitivity to temperature was unusual but as the power increased, this sensitivity seemed to decrease. At the 160-hour point the stack power was 925 watts and increasing. The stack ASR at this point was 0.98 and improving. The stack power was then increased to 1000 watts and ran steady for approximately 2 days (15.2 W/cell,



**Figure 31. PCU stack #244**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

22.7 amps, 0.67 V/cell). This represents 4.2 kW in the 4 stack C2 system. Thermal stress analysis indicated that higher power levels could be achieved without exceeding the maximum stress. It was decided not to push the stack to higher stress levels at this time.

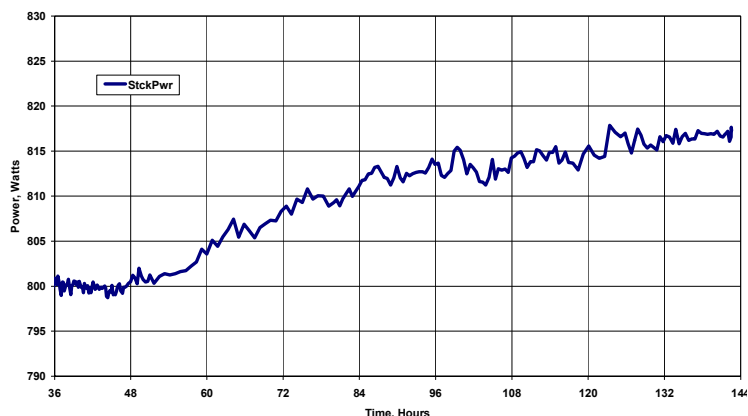


**Figure 32. Initial Test Results for Stack #244**

Stack 244 power was lowered to 800 watts (12.2 w/cell, 15.7 amps, 0.78 V/cell). This corresponded to 3.4 kW in C2 which represented its long-term operating condition. The stack ran for 144 hours at this condition with a 7% per 500 hour increase in power over time and an ASR of 0.93, as shown in **Figure 33**.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

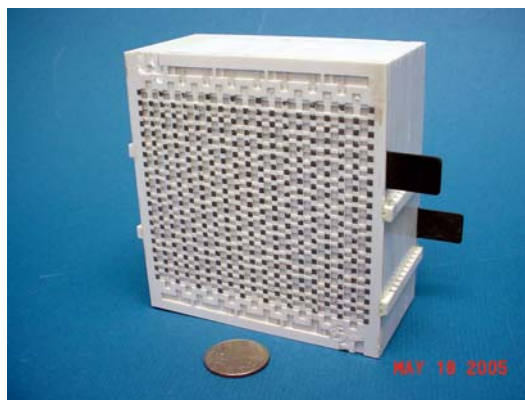
Overall stack #244 ran for over 400 hours on reformat from [REDACTED] natural gas. For the first time, the tall stack behaved similarly to that observed in short stack and single cell tests (i.e. initial power increase at constant voltage over a significant amount of time). Although in the time frame tested stack power was still increasing. These results suggest that the degradation behavior of this tall stack may also have been similar to that observed in recent short stack and single cell tests. Moreover, stack performance was sufficient to meet C2 long-term operating conditions at 3.2 kW. The highest power achieved in stack #244 represented 4.2 kW in C2, with a higher power output possible.



**Figure 33. Additional test results for stack #244**

## Monolithic Stack Development

The use of a metal cage for containment of the all-ceramic stack is a short-term solution to ease handling and to allow the application of an end load to a horizontal stack. The use of ceramic interconnects in the SOFCo stack design creates an opportunity for an all-ceramic stack that is termed the “monolithic stack” design. An early example of such a stack is shown in **Figure 34**. This early version used simple ceramic side rails made from scrap 3YSZ material generated during the interconnect manufacturing process, so the CTE match is identical to the majority of the stack material. These side rails were attached using a glass composition. Various glass compositions with differing glass crystallization temperatures were developed to tailor the glass cure point to various stack pre-conditioning temperatures. Additional work on the design of the side rails is required to develop more of a structural element that fully captures the stack and allows for ease of assembly. The monolithic design requires a new current collector concept with integral connection of the current collector to the end interconnects to assure good electrical contact throughout the operating temperature range of the stack. Such a development is underway with NASA personnel and others. The monolithic approach would significantly improve stack ruggedness as well as eliminate the metal cage and the need for a stack end loader. The former would improve the potential for application of the SOFCo design to mobile applications where vibration issues are prevalent, while the latter would reduce cost and weight and significantly simplify the overall PCU design. Development of a monolithic stack design was showing great promise, but other priorities related to stack performance issues and reformat operation delayed the monolithic stack development effort. Although the basic concept has been demonstrated, realistically the monolithic design is essentially a stack packaging issue and will become more relevant as the stack design and pre-conditioning requirements are finalized based on commercial product requirements.



**Figure 34. Monolithic stack**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

**CONCLUSION:** The major thrust of the PCU Development effort was started in the fall of 2003 with a 20-cell stack and was scaled up over the next two years to 48-cell stacks and eventually 70-cell stacks. In total, counting sub-sections of stacks tested, 15 “tall” PCU stacks were tested, including 13 stacks tested at SOFCo facilities and two stacks tested in the C1 system at CPG. Many short stack (10 cells or less) and single cell tests were performed in support of the PCU development effort and were invaluable in defining and identifying various operating and construction issues. As described above, a number of the tall stack operating issues had to do more with system operational issues and system component failures than with the fuel cell technology itself. System operational issues are to be expected with a new technology and the solutions to these types of problems are much more tractable. Development of automated control schemes to prevent out-of-spec conditions will be critical to all fuel cell systems and should go a long way to assuring that stacks are protected against these types of events.

In terms of stack issues, cell strength was a major problem that has now been largely overcome through the institution of better cell manufacturing processes and the use of the [REDACTED] fuel-side connection. Over the two years of concerted development effort, there was significant progress in understanding the required strength and other characteristics of both cells and interconnects and the best methods for operating the fuel cell system to maintain stack integrity. Significant progress has been made in building, pre-conditioning and operating large stacks like those designed for the C2 system. Fuel supply operating issues with reformed natural gas did not appear until more recent stacks when the higher power cells (i.e., higher current density) made with ScSZ electrolyte were fully integrated into the stacks. [REDACTED]. Longer term developments of more tolerant anodes are underway at a number of organizations, since this problem is generic to the fuel cells using the conventional Ni-cermet anodes.

Overall, the SOFCo PCU development has reached a point where it is ready to begin design refinement for specific commercial applications. Tall stack operation now largely duplicates the performance seen in short stacks and single cell tests and this indicates that stack construction issues have essentially been solved, although further improvement is always possible. Unfortunately, continued testing of Stack #244 was not possible due to reaching the end of Phase I. As a result, the long-term performance trends, including degradation behavior, were not established. However, given the performance of short stack #241, it is possible that degradation in the 2-3% per 500 hours may have been achieved.

## **Subtask 4 Fuel Processor Development**

**OBJECTIVE:** Develop a cost effective CPOX reformer with no external water supply requirements.

**APPROACH:** A Catalytic Partial Oxidation (CPOX) reforming approach was selected by the SOFCo/CPG team for its compactness, low-cost design and operational simplicity. During 2002 and 2003, SOFCo completed bench-scale and scaled-up CPOX performance evaluations for natural gas (C<sub>1</sub>-C<sub>2</sub>) and LP (C<sub>3</sub>-C<sub>4</sub>) fuels. The micro-reactor scale (0.3 kWe) and 1 kWe-scale data showed a good performance match. In the first half of 2004, several sub-scale tests were conducted to investigate CPOX durability, long-term operability of short SOFC stacks on reformed natural gas and “off-normal” operation to support C1 and C2 system startup/shutdown. A 5 kWe CPOX reformer was built and extensively tested with multiple SOFC stacks in preparation for the Phase I test at CPG. The 5 kWe reformer was installed in the C2 power module and delivered to CPG in December 2004.

**EXPERIMENTS:** The following text presents a summary of SOFCo’s CPOX reformer development at 0.3, 1 and 5 kWe scale performed during the SECA Phase I program.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## **Small-Scale (0.3 kWe) Fuel Processor**

Catalyst deactivation and fouling due to carbon deposition and thermal sintering are two significant risk factors affecting the long-term operability of a CPOX reformer given the high reaction temperatures and operation at substoichiometric oxygen levels. Both of these factors are accentuated in the waterless mode of operation. One goal of the Phase I fuel processor development program was to identify catalyst systems for the CPOX reforming of NG and LPG. Another goal was to carry out longer-term catalyst testing to characterize deactivation and carbon formation. The lighter hydrocarbon ( $C_1$ - $C_4$ ) fuels with their relatively high hydrogen content were expected to be more amenable to this approach. Abundant literature exists on the CPOX of  $C_1$ - $C_2$  type fuels (methane, NG, ethane), but examples for propane and butane are scarce. The CPOX of heavier liquid distillate fuels is expected to be more challenging, but technically feasible, based on SOFCo's extensive experience in gas- and liquid-fuel reforming, and remains a longer term goal.

Bench-scale catalyst testing carried out in Phase I of the SOFCo/CPG SECA program is described. The results support CPOX as an attractive alternative to SR and ATR processes.

## **Experimental**

All experiments were performed using a modified Zeton-Altamira (AMI-200) reactor system allowing the production of up to a 0.3 kWe electric equivalent of reformat gas. The system had thermocouples for the inlet and exit gas temperature measurements and a pressure gauge to measure the inlet bed pressure. After passing the reformat through a condensate trap, the product gas rate was measured with a calibrated wet test meter and analyzed online using a Varian Four-Channel Quad Micro GC. Monolith catalyst samples were loaded into a quartz U-shaped reactor with a 0.4 cm (0.17 in) ID preheater arm and 1.7 cm (0.67 in) ID reactor arm. **Figure 35** shows a monolith catalyst and a top view of the reactor arm. The monolith was wrapped in a ceramic paper to provide a seal between the monolith and reactor wall.

The LPG, NG, and air were purchased from industrial gas vendors. The LPG conformed to the ASTM-D1835 fuel specification and the NG contained ~96 mole % methane. The process air was commercial grade and contained less than 10 ppmv moisture.

Monolith catalyst samples (1.3 D x 2.5 cm L (0.5 x 1 in), 400-600 CPSI) were obtained from several catalyst suppliers. The CPOX testing was carried out at atmospheric pressure, steam-to-carbon (S/C) feed ratios of 0.0-0.5, molar oxygen-to-carbon ( $O_2/C$ ) feed ratios of 0.4-0.7, gas contact times of 15-100 msec, and feed preheat temperatures of 50-350°C. Carbon deposition was assessed by comparing the pre- and post-test monolith weights and by visual inspection of the used catalyst samples.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

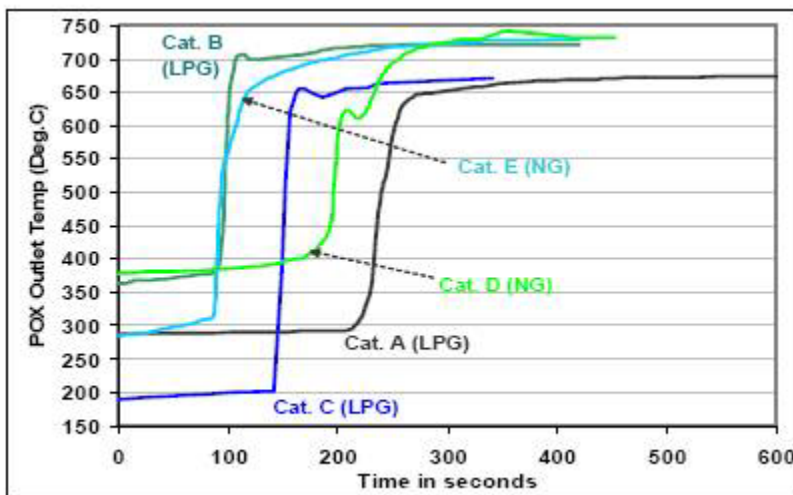
## Results

Several monolith catalysts were evaluated (30-50 hr) for the CPOX of LPG under the same test conditions. LPG was used for the initial screening because of its higher olefin and heavy hydrocarbon ( $C_3+$ ) content relative to NG. The testing provided a catalyst ranking based on hydrocarbon conversion [molar  $(CO+CO_2)_{out}$  / molar  $(fuel-carbon)_{in}$ ], reforming efficiency [ $LHV(H_2 + CO)_{out}$  /  $LHV(fuel)_{in}$ ], light-off temperature, and tendency for carbon formation. The light-off temperature depended highly on the catalyst formulation, the hydrocarbon type, and the



**Figure 35. Monolith catalyst (left) and quartz reactor top view (right)**

oxygen content of the feed (**Figure 36**). Light-off temperatures ranged from 200°-400°C and were characterized by a rapid increase in the reformer temperature. After light-off, the reformer was fully operational, generating reformat for immediate use. The curves in **Figure 36** demonstrate the rapid startup characteristics of the CPOX reformer. The response to load variations was also almost instantaneous.



**Figure 36. CPOX startup (LPG and NG light-off)**

**Figure 37** depicts the measured reformer performance degradation due to carbon formation in the bed. The data were collected for LPG at short contact times. The reformer inlet pressure served as a good indicator of bed plugging due to carbon accumulation. Among several tested catalysts, Specimens 2 and 3 (**Figure 37**) showed no inlet pressure rise, confirming non-carbon conditions over the 30-50 hr test. A flat reformer efficiency trend for Specimens 2 and 3 also supported this finding. However, the detrimental effects of carbon formation and the resulting flow blockage on the reformer performance were clearly evident for Specimen 1. Within 20 hr of operation, the CPOX reformer efficiency dropped almost 30% (from the low 70s to mid 40s), which coincided with a threefold increase in the inlet pressure (from 3 to 9 psig). A post-test inspection of Specimen 1 revealed more than a 50 % blocked

# 10 kW SOFC Power System Commercialization

## SECA Serr

flow area with hard carbon deposits. These tests proved very instrumental in achieving the first objective of identifying both CPOX catalysts and non-carbon process conditions for LPG and NG fuels.

Two catalyst compositions were selected for longer-term testing and parametric studies. **Figure 38** shows a 550 hr aging study carried out with LPG. High hydrocarbon conversions (>88%) were maintained with reformer efficiencies of 70-75%. The hydrocarbons in the product gas included methane, ethylene, ethane, propylene, and propane. A typical dry reformate gas contained 45-48% H<sub>2</sub>+CO, 1-3% CO<sub>2</sub>, 0.2-0.7% methane, <2% C<sub>2</sub>+ hydrocarbons, and balance nitrogen. After more than 500 hr of operation, there was no evidence of carbon formation on the catalyst surface. The results were very encouraging and suggest the CPOX reforming is a viable option for generating fuel cell reformate from LPG.

The effect of feed preheat temperature on the reformer performance is shown in **Figure 39**. As the inlet temperature was increased from ambient to 350°C, the hydrocarbon conversion and reformer efficiencies improved. The maximum reformer efficiency was ~80% at 350°C. It is anticipated that the fuel cell hot exhaust will be a major heat source for feed preheating in a thermally integrated system.

These tests also identified a feed preheat limit of 250°C at high O<sub>2</sub>/C levels (**Figure 39**). Above this limit, the reformer efficiency was adversely affected, due to fuel/air pre-ignition near the monolith inlet. At low O<sub>2</sub>/C ratios, the feed preheat temperature limit was extended beyond 350°C.

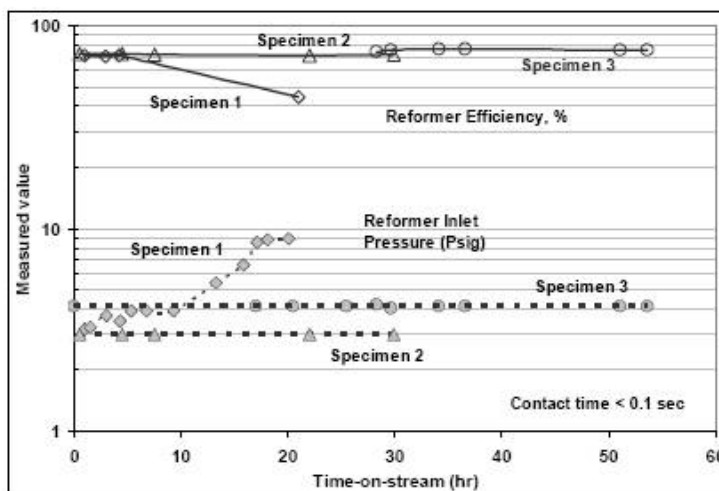


Figure 37. Carbon formation effect on CPOX performance

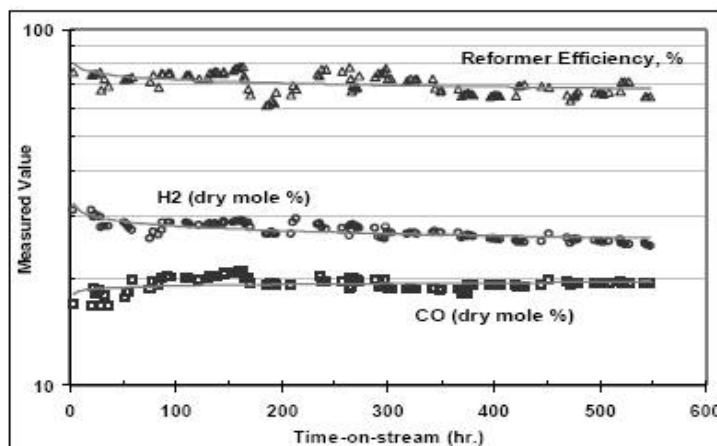


Figure 38. CPOX performance with LPG

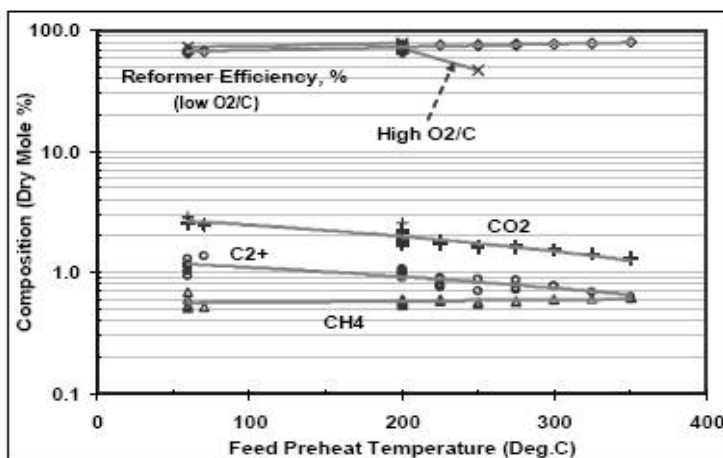


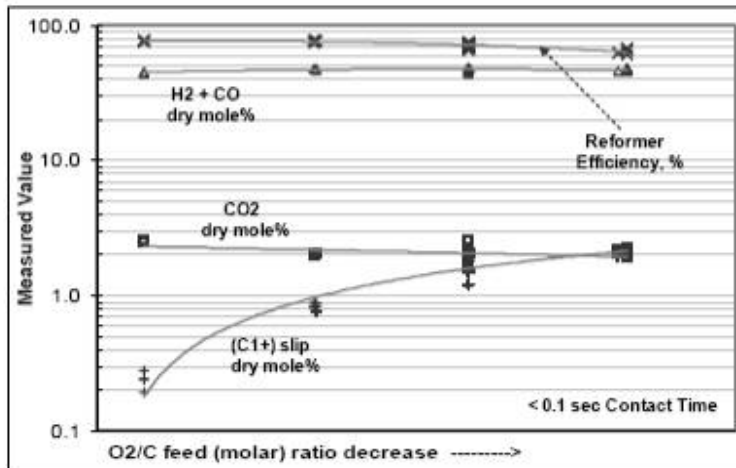
Figure 39. Effect of feed preheat temperature



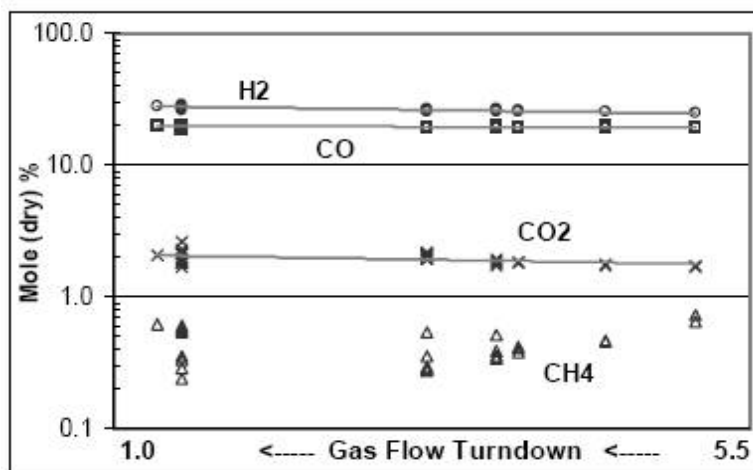
## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

Lowering the O<sub>2</sub>/C feed ratio resulted in a significant drop in the hydrocarbon conversion (**Figure 40**). The unconverted hydrocarbons content (C<sub>1</sub>+ slip) of the reformat increased from 0.2 % to 2.0%. Correspondingly, the reformer efficiency decreased from 75% to 60%.

**Figure 41** shows that the reformat gas composition was only slightly affected by a 5.5:1 turndown. The methane slip gradually increased with throughput but remained well below 1.0% over the flow range. The CPOX of NG was also evaluated under similar process conditions. Higher light-off temperatures were required for NG. The hydrocarbon conversions were above 90%, with reformer efficiencies approaching 80%. There was also an expected tendency for lower carbon formation with NG. High hydrocarbon conversions and H<sub>2</sub>+CO yields were maintained. A typical dry reformat gas contained 44-48% H<sub>2</sub>+CO, 3-4% CO<sub>2</sub>, 0.9-1.7% methane, <100 ppmv C<sub>2</sub>+ hydrocarbons, and balance nitrogen. No carbon formation was observed over the range of test conditions for 85 hr of operation.



**Figure 40. Effect of O<sub>2</sub>/C feed ratio**



**Figure 41. Reformer performance with turndown**

Table 2 compares averaged CPOX reformer data for NG and LPG. Higher hydrocarbon conversions and reformer efficiencies were obtained with NG. Both fuels gave CO+H<sub>2</sub> yields of 45-46%, but the NG had a 50% higher H<sub>2</sub>/CO product ratio, consistent with the higher hydrogen content of NG. The NG reformat contained 1-2% unconverted methane and traces of heavier hydrocarbons. The LPG product gas contained near-equilibrium methane levels and propane and propylene were the main C<sub>2</sub>+ hydrocarbons.

### Summary

The small-scale 0.3 kWe development achieved the overall objective of developing a CPOX reformer for integration with a 10 kWe SOFC system. The reformer was successfully operated over 500 hours and demonstrated high reforming activity for both NG and LPG feeds.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

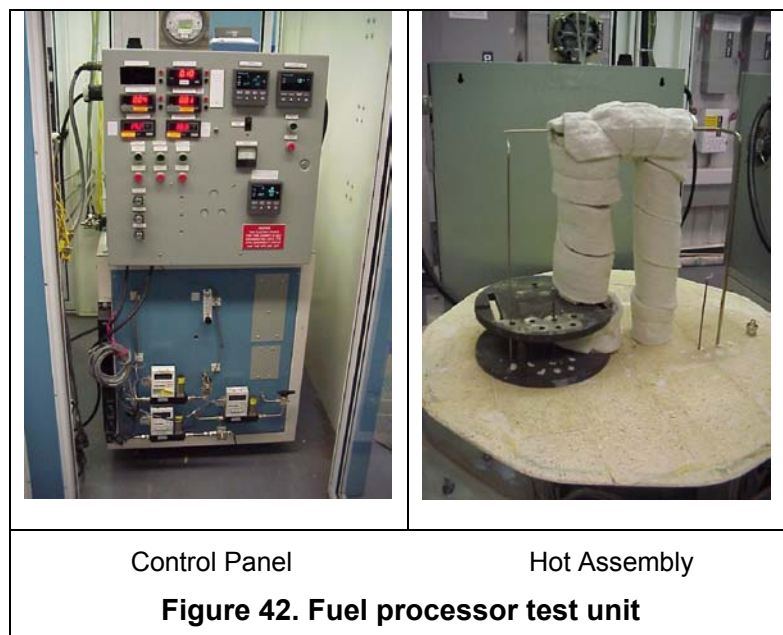
**Table 2. CPOX Performance Comparison**

	Methane: NG	Propane: LP
C in feed (wt %)	72.0	81.9
H in feed (wt %)	24.2	18.1
Sulfur in feed (ppmw)	20	185
H/C Conversion, $\chi$ %	92.9	88.5
CPOX Efficiency, $\eta$ %	76.9	72.5
Reformate (dry) Mole %		
H <sub>2</sub>	31.0	26.8
CO	14.6	18.6
CO <sub>2</sub>	3.3	2.6
CH <sub>4</sub>	1.3	0.5
C <sub>2</sub> +	0.01	1.4
N <sub>2</sub>	49.8	50.1
Product H <sub>2</sub> + CO	45.6	45.4
Product H <sub>2</sub> /CO ratio	2.13	1.44

## 1 kWe-Scale Fuel Processor

As a result of the positive CPOX performance at up to 0.3 kWe, reformer scale-up to 1 kWe was initiated with objectives to:

- Demonstrate 1 kWe reformer component testing for C1 installation;
- Demonstrate a fuel-flexible reformer design for NG and LPG; and
- Demonstrate short SOFC stack operability with the CPOX reformate.



A scaled-up (1.5 kWe) fuel reformer test unit was engineered to demonstrate its functionality at the operating conditions required during fuel cell power system operation. The small-scale reformer test data provided all the necessary design data for the scale-up. The test apparatus is shown in **Figure 42**.

The portable test skid was equipped with test components, test instrumentation, and monitored and controlled using an on-line Agilent Data Logger System. The unit was fitted with gas feed meters, reformer feed preheater, feed mixer, reformer, particulate filter, product gas

condensate trap and a gas analyzer. The test unit was also connected to an existing flare system for safe disposal of the product gas.

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 43. 1.5 kW CPOX reformer**



**Figure 44. Hot-gas filter**

The CPOX reformer was designed by SOFCo and fabricated in-house for 1.5 kW<sub>e</sub> demonstration. The light-weight CPOX reformer is shown in **Figure 43**. Its compact size displaced less than 100cc of volume. The reformer catalyst formulations are proprietary. The reformer design/scale-up strategy was developed to yield low pressure drops (<0.5 psi) and representative reaction times over the load range. The 1.5 kW<sub>e</sub> demonstration also included a downstream hot-gas filter (**Figure 44**) to trap particles and/or catalyst fines present in the gas stream.

Two types of fuels were tested: commercial-grade propane (LP) and natural gas (NG). Dry plant air was used for the CPOX reformer operation. The reformer testing was carried out at atmospheric pressure, oxygen-to-carbon (O<sub>2</sub>/C) feed ratios of 0.45-0.7, gas contact times of less than 1 second, and feed preheat temperatures of 100-300°C. The dried product gas flow rate was measured using a mass flow meter and analyzed for H<sub>2</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and unconverted hydrocarbon species (C<sub>2</sub>-C<sub>5</sub>'s) using a Varian 4-channel microGC. For all test data, the C and N-elemental balances were satisfied within 95-105%.

### Results and Discussion

In the first half of 2003, a CPOX catalyst (labeled A) was evaluated for LP. Performance was good. At DOE-NETL's direction, SOFCo/CPG were advised that the Phase 1 test could not use LP but could use NG. As a result, catalyst "A" testing was extended to NG in the first half of 2003 and additionally, an alternate catalyst (labeled B) was evaluated with NG.

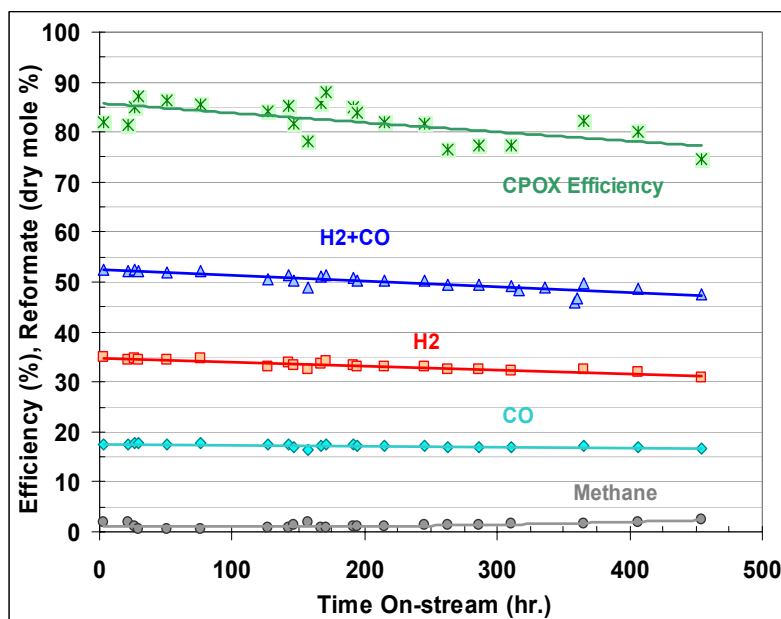
Table 3 summarizes the measured performances of Catalyst "A" with NG and LP fuels and that of Catalyst "B" with NG. As of December 2003, the C1-scale reformer logged 900 hr and 775 hr of cumulative run-time with LP and NG, respectively. The CPOX reformer for the C1 demonstration contained Catalyst "A".

The impact of fuel switching from LP to NG on reformer design and operation was minimal. The light-off temperature for NG was higher than that for LP. A constant feed preheat (300°C) was required for proper functioning of the NG reformer. In comparison, the LP reformer required a feed preheat (200°C) only during startup. The NG reformer was demonstrated for a 4:1 turndown versus 5:1 for LP. The fuel conversion and reformer efficiency were higher for NG than for LP. **Figure 45** shows the long term (460 hr) performance of the NG reformer using Catalyst "B".

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

**Table 3. 1 kWe CPOX Performance Comparison**

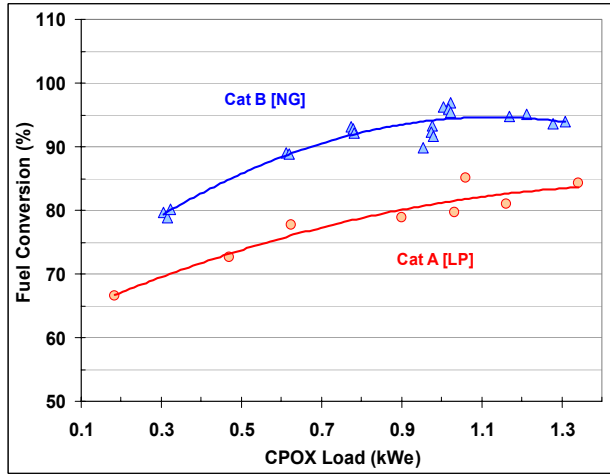
C1-Sale Reformer	Propane (LP)	Natural Gas (NG)	
Catalyst Designation	A	A	B
Time On-stream	900h	240h	535h
Feed Preheat	200 °C	300 °C	
<b>Operation</b>			
Turndown [% Load]	100 to 20	100 to 50	100 to 25
Air/Fuel Ratio, [FE]	2.7 – 3.3	3.0 – 3.7	3.0 – 3.3
<b>Performance</b>			
Fuel Conversion (%)	75 - 85	80 - 88	90 - 98
CPOX Efficiency (%)	65 - 72	66 - 73	75 - 85
H <sub>2</sub> +CO [dry mole %]	40 - 45	45 - 48	47 - 50
H <sub>2</sub> /CO Ratio	1.2	1.8	2.0
Methane Slip [dry mole%]	0.5 - 2.0	2.0 - 4.0	0.4 - 2.4
C <sub>2</sub> + Slip [dry mole%]	0 - 2.0	0 - 0.13	0 - 0.04



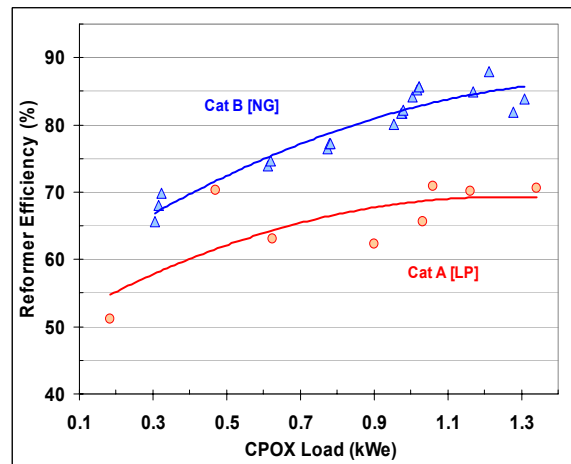
**Figure 45. NG CPOX performance [Catalyst B]**

The effect of throughput (or load) on reformer performance is shown in Figures 46(a) and (b) for LP and NG fuels. The fuel conversion and the corresponding reformer efficiency showed decreasing trends as the reformer load decreased from its full-scale value. This may be attributed to heat effects in the reactor affecting the relative amounts of total and partial oxidation over the load range. While the LP reformer was 70% efficient at 1.3 kWe load, the NG reformer efficiency peaked at 85%.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

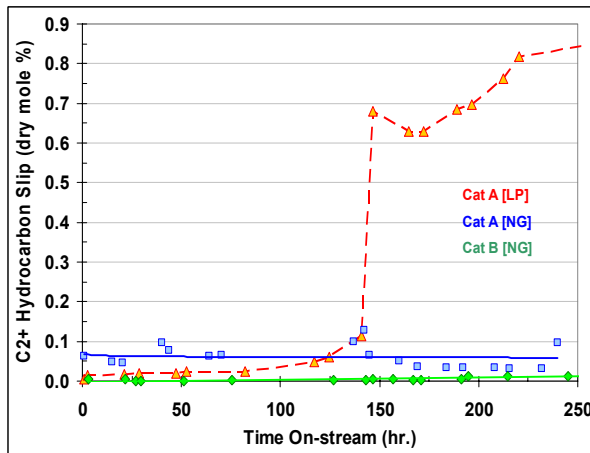


**Figure 46(a). Fuel Conversion vs. Load**

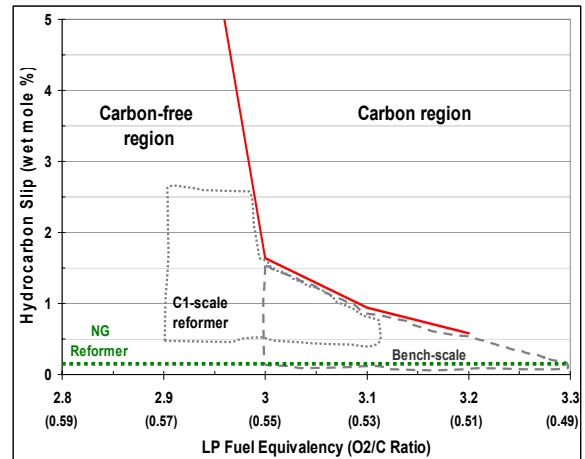


**Figure 46(b). Reformer Efficiency vs. Load**

As compared in **Figure 47(a)**, the hydrocarbon slip for NG was significantly lower than that measured for LP. The NG reformat therefore offers very little or no carbon deposition potential. The experimental data collected for NG supported the equilibrium carbon region predictions shown in **Figure 47(b)**. Additionally, oxygen-to-carbon [O/C] feed ratio, gas mixture temperature and component materials were identified to be important factors that controlled carbon formation.



**Figure 47(a). C<sub>2</sub>+ Slip for CPOX catalysts**



**Figure 47(b). Reformer Operating Envelope**

Short stack testing was started to evaluate SOFC operability with a NG reformat containing H<sub>2</sub>, CO, CO<sub>2</sub> and methane. By the end of December 2003, the SOFC stack logged 72 hrs on reformat slip-stream (30 mole% H<sub>2</sub>, 16% CO and 3-5% methane) to the stack. Gas samples collected before and after the stack indicate that ~90-95% of the methane slip was internally reformed in the stack. The SOFC cell was able to utilize a feed stream low in H<sub>2</sub>/CO ratio.

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

Constant system and stack inlet pressures suggest no blockages in the flow fields due to carbon deposition. Further, the SOFCo fuel cell stack (using 3YSZ electrolyte-supported cells) demonstrated sulfur tolerance at the conditions tested. The SOFC feed gas contained low levels of H<sub>2</sub>S (of the order of 0.5-2 ppmv) since the NG fuel was not desulfurized at the reformer inlet.

### Summary

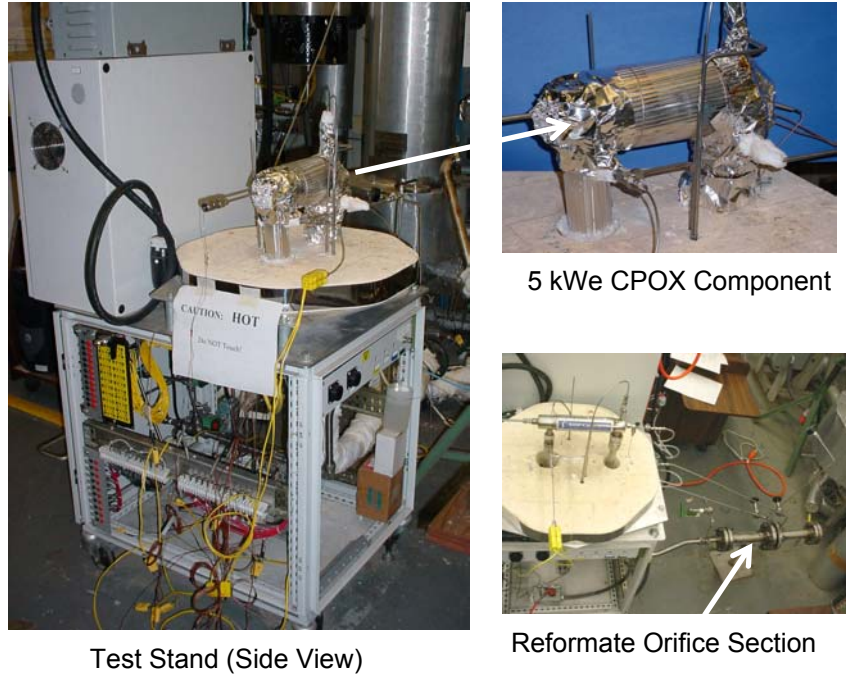
- Design, performance and parametric testing of the C1-scale reformer were completed for NG and LP fuels.
- The 1 kWe CPOX reformer was supplied for C1 installation/testing at CPG.
- Operational carbon formation boundaries were established through experiments and modeling.
- Stack testing with the “waterless” CPOX reformat provided an early indication that the SOFC was able to utilize a low H<sub>2</sub>/CO reformat feed with methane and H<sub>2</sub>S.
- Experiments confirmed that carbon formation is not a concern for the NG fuel.
- Carbon deposition may be an issue for LP, and SOFCo will perform further operational characterization of LP over the operating temperature range prior to scale-up. With the decision to use NG for C2 Phase 1 testing, there is no priority at this time to further characterize LP fuel processor operation.

### 5 kWe-Scale Fuel Processor

#### C2 Reformer Prototype Demonstration

**Figure 48** shows different sections of the 5 kWe C2 CPOX reformer demonstration facility, which was designed and constructed in early 2004 and operated into 2005. The 5 kWe CPOX component design (**Figure 48**) was based on the sub-scale performance data and CFD modeling of the internal components. The monolithic CPOX reformer was designed and fabricated in-house for 5 kWe demonstration. The unit was operated on untreated pipeline NG containing less than 20 ppmw sulfur species. Dry plant air was used as the oxidant for achieving the desired CPOX stoichiometry. The instrumentation included thermocouples, static pressure gauges, differential pressure transducers, and feed mass flow controllers. The hot reformat stream was passed through an orifice section (**Figure 48**) for product gas flow measurement. The reformer testing was carried out at atmospheric pressure, no water addition, molar oxygen-to-carbon (O<sub>2</sub>/C) feed ratios of 0.5-0.7, <1 sec gas contact times, and feed preheat temperatures of 20-350°C. Dry reformat samples were analyzed for H<sub>2</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and unconverted hydrocarbon species (C<sub>2</sub>-C<sub>5</sub>'s) using a Varian 4-channel, micro gas chromatograph. The differential pressures across the feed preheater and reformer were monitored throughout the tests. For all test data, the C-, O- and N-elemental balances were satisfied within 95-105%.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 48. C2 (5 kW) reformer test hardware**

## Material Compatibility Tests for Carbon Deposition

The C2 reformer test hardware was modified to expose material coupons to reformed natural gas. Two 3.8 cm (1.5 in) diameter x 20 cm (8 in) long coupon test sections (**Figure 49**) were installed in a series configuration at the reformer outlet with individual temperature controls in the 200-800°C range to bracket the thermodynamic carbon formation boundary.

Materials, typically used in SOFC stack and balance-of-plant (BOP) equipment, were considered for these tests. These included anode- and cathode-side materials, cell via materials, stack interconnect samples, coated metal alloys, nickel screens, wires, meshes and felts.

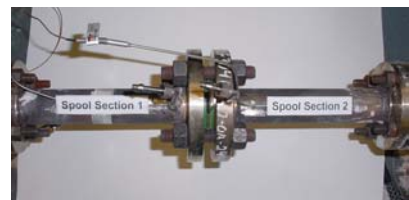
Several material compatibility tests were conducted to investigate carbon deposition at design and off-design reformer operating conditions. Under the design conditions, a steady stream of reformate with about 29 mole% H<sub>2</sub>, 16% CO, 1.75% CO<sub>2</sub>, 2.5% CH<sub>4</sub>, <100 ppmv C<sub>2</sub><sup>+</sup>, 0.5% Argon, 6% H<sub>2</sub>O and N<sub>2</sub> (balance) flowed over the specimens. Each test ran for 100 hours over a temperature range of 250 to 650°C.



(Coupon Tray Section)



(Tray Section installation)



(Coupon Test Section)

**Figure 49. Coupon test hardware**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## Results and Discussion

### **C2 Reformer Demonstration**

Table 4 summarizes the measured CPOX performance at C1 (1 kWe) and C2 (5 kWe) scales. The performance data for bench-scale (0.3 kWe) and scaled-up reformers show a good match over the 1:30 scale-up. A flow turndown of 20:1 was successfully demonstrated for the C2 prototype. The fuel conversion, efficiency and H<sub>2</sub>+CO product were improved with the reformer scale-up. This may be attributed to a smaller reformer heat loss portion out of the total heat release with scale-up.

**Table 4. Comparison of Reformer Scale-up Performance**

	<b>Bench-Scale</b>	<b>C1-Scale</b>	<b>C2-Scale</b>
Scale-up Factor (NG)	1	6	30
Design Load (kWe)	< 0.3	1.0	5.0
Monolith Volume (cm <sup>3</sup> )	10.1	47.8	121.8
OD Surface/Volume (cm <sup>-1</sup> )	3.15	2.0	1.05
Pressure Drop, psi	0.03 (est.)	0.3	0.3
Load Range (kWe)	-	0.3 – 1.3	0.3 – 6.7
Turndown (% of design load)	-	100 to 30	100 to 6
(% of maximum load)	-	100 to 23	100 to 4.5
Fuel Conversion, %	83 – 88	75 – 85	75 – 98
Reformer Efficiency, %	68 – 72	65 – 72	60 – 88
Reformate (dry mole %)			
H <sub>2</sub>	25 - 28	22 - 26	28 - 32
CO	18	18 - 19	14 - 18
CO <sub>2</sub>	2.2	2.2	1.5 - 3.5
CH <sub>4</sub>	0.3 - 0.8	0.5 - 2	0.3 - 5.0
C <sub>2</sub> + slip	0 - 2	0 - 2	0 - 0.10
N <sub>2</sub>	45 - 50	45 – 50	42 - 50

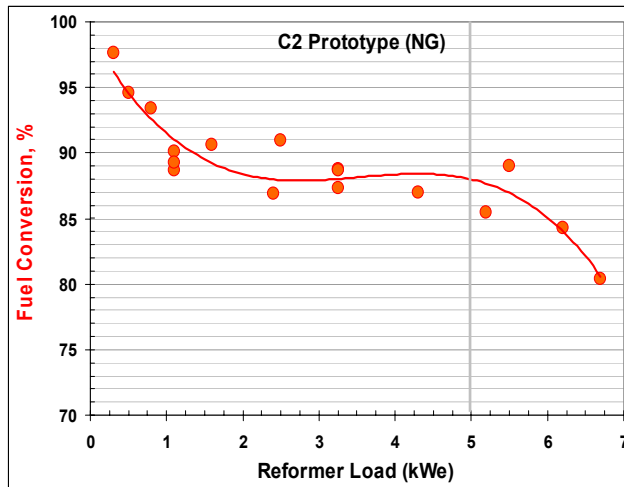
### **C2 Reformer Parametric Evaluation**

Effects of three process parameters, i.e., feed preheat, air/fuel feed ratio (or O<sub>2</sub>/C ratio) and throughput (or kWe load), on reformer performance were studied. The O<sub>2</sub>/C feed ratio was varied in the 0.5-0.7 range, which corresponded to the fuel equivalence (FE) ratio of 2.6-3.6. The performance change was measured with and without the feed preheater. While the C2 reformer was designed for 5 kWe, its load was varied between 0.3 and 6.7 kWe to evaluate the maximum operating limit.

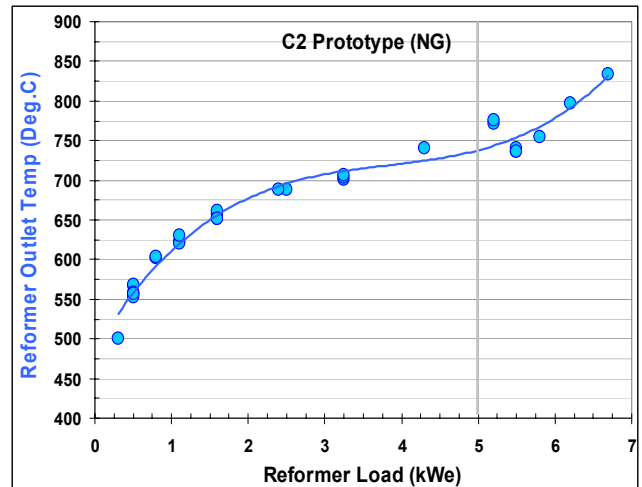
**Figure 50 (a and b)** show the effect of load variation on fuel (NG) conversion and heat release at constant process conditions, i.e., no feed preheat, 0.66 O<sub>2</sub>/C feed ratio and 3.0 fuel equivalence (FE). Between 1-5 kWe, the averaged fuel conversion was in the 87-90% range. At loads below 1 kWe, the gas residence time increased which led to an improved reformer performance yielding high (93-98%) fuel conversion. At loads above 5 kWe, the performance dropped (80-85% conversion) due to shorter bed contact times.



# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 50(a). Fuel conversion with load**



**Figure 50(b). Reformer outlet temperature with load**

The measured reformer outlet temperatures also showed a consistent trend with load. A lower fuel conversion implied less efficient endothermic methane reforming. Therefore, as the fuel conversion dropped, the reformer outlet temperature showed a 100°C increase between 5-6.7 kWe loads. Between 0.3-5 kWe the reformer outlet temperature showed a gradual increase from 500 to 750°C, which was consistent with the earlier test data at bench- and C1-scales

In real systems, the reformer performance and temperature could be properly controlled by adjusting the feed condition. Further, the reformer outlet gas temperature can be used as an effective parameter to control the reformer operation with adequate sensitivity over the 0.3-6.7 kWe load range.

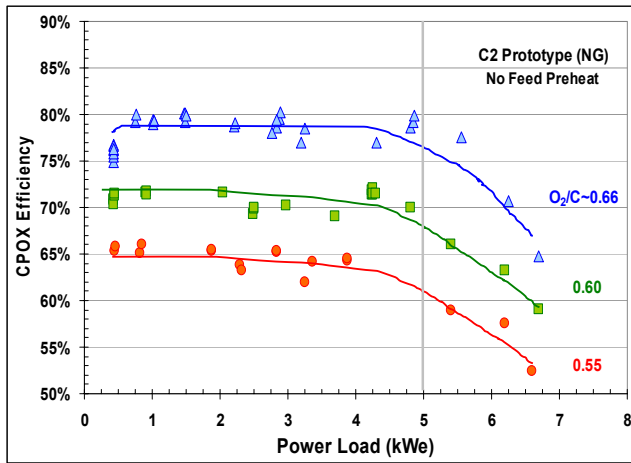
The measured CPOX performance improvement with increasing air levels ( $O_2/C$ ) is shown in **Figure 51(a)**. The test data were collected with no feed preheat and between 0.3-6.7 kWe load range. The reformer efficiency gained almost 15 points (65 to 79%) with a 20% increase in the air level (0.55 to 0.66  $O_2/C$ ). Nearly constant reformer efficiencies were noticeable up to 5 kWe load for all air levels. Though the air level effect remained beyond 5 kWe, the CPOX efficiency trended downward at all  $O_2/C$  ratios tested.

As compared in **Figure 51(b)**, a more than 10-point gain in the reformer efficiency was measured for all  $O_2/C$  levels at higher feed preheat (300 °C). However, the reformer performance showed a steady decline over the entire load range. The data establishes the operating envelop to obtain reasonable reformer performance in the load range tested.

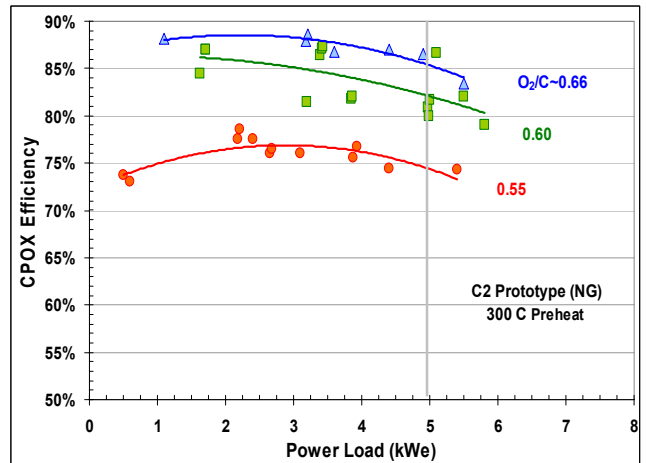
## C2 Reformer Durability

The 5 kWe scale C2 CPOX durability demonstration completed 2,800 hrs of stable operation with automated controls. As shown in **Figure 52[a]**, good reformer activity was maintained over the entire test duration. The reformer power level was varied between 0.3 and 6.7 kWe. After the initial burn-in period, the reformer performance showed a slow decline of 2% per 500 hours of operation.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



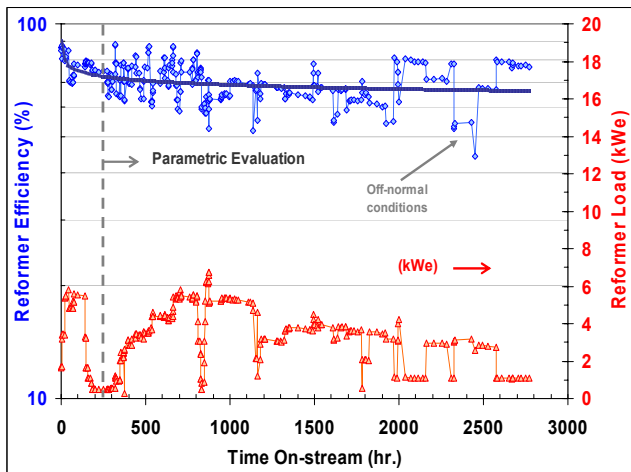
**Figure 51(a). Air level effect on performance**



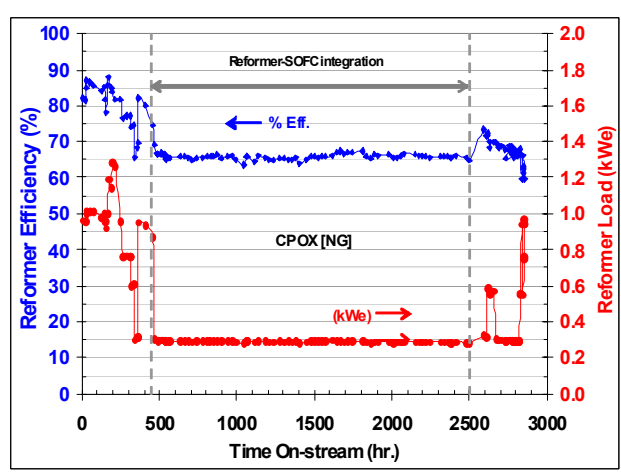
**Figure 51(b). Preheat effect on performance**

Between 2,300-2,500 hrs (Figure 52(a)), a material compatibility test was conducted to study carbon deposition at “off-normal” reformer operation. The process conditions were adjusted to yield low fuel conversions and high hydrocarbon slips. During this period, the off-normal reformer efficiency was decreased below 55% with about 8-11 mole-% methane slip. The reformer performance immediately recovered after the process conditions were re-adjusted to reflect the normal operation.

In comparison, durability demonstration of C1 prototype completed 2,900 hrs of operation before the breakthrough of methane and C<sub>2</sub>+ hydrocarbon species (**Figure 52(b)**). The reformer efficiency dropped more than 10 percentage points in the final hours of operation. A “manual mode” of operation with upset conditions during startup/shutdown may have accelerated the catalyst deactivation. As expected, a well-controlled C2 reformer operation did not show the same behavior past 2,800 hours of operation.



**Figure 52(a): 5 kWe C2 reformer durability**



**Figure 52(b): 1 kWe C1 reformer durability**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## **Material Compatibility**

Separate tests were conducted using multiple coupon sets to study the carbon deposition tendency at normal and off-normal CPOX operating conditions in waterless mode. In each 100-hr test, new coupon sets were installed and exposed to a steady stream of reformed natural gas. The reformat stream was produced on-line by the C2 reformer. The reformer process conditions were adjusted to yield a desired operating condition. The coupons were removed after each test and visually inspected for carbon deposition. Table 5 summarizes the test conditions and observations.

**Table 5. Material Performance During Reformat Exposure**

Test ID	Operation	CH <sub>4</sub> Slip (dry mole%)	Exposure Temp (°C)	Observation
1A	Normal	2 - 4	250	No discoloration, No carbon deposition
1B	Normal	2 - 4	450	Slight discoloration; No carbon deposition
2A	Normal	2 - 4	550	Slight discoloration; No carbon deposition
2B	Normal	2 - 4	625	Slight discoloration; No carbon deposition
3A	Off-normal	8 - 11	450	Slight discoloration; No carbon deposition
3B	Off-normal	8 - 11	650	Carbon deposition on nickel-rich coupons

The “normal operation” test data experimentally confirmed no carbon deposition on all test materials and flow lines in the 250-625°C range. The Inconel and stainless steel flow sections removed from the CPOX outlet location also showed no carbon deposition after 2,600 hours of reformat exposure in the 200-800°C range.

Carbon formation was observed only at high temperatures (650°C) and high hydrocarbon slips. While thermodynamic equilibrium predicts carbon tendency at low temperatures, high temperatures are known to kinetically favor the carbon formation, especially, in the presence of high hydrocarbon slip. The test data shows that 8-11 mole % hydrocarbon slip in the reformed gas and higher than 600°C temperatures will lead to carbon deposition in the downstream system components. These tests have helped to establish reformer operating conditions needed to avoid carbon formation in the waterless CPOX reformer.

## **Summary**

A “waterless” 5 kWe CPOX fuel reformer scheduled for use in the Phase I demonstration test at CPG was extensively characterized with Alliance, OH, pipeline natural gas (NG). Reformer operation and material tests showed carbon-free performance during startup, normal operation and shutdown. A 20:1 load turndown was demonstrated for the C2 reformer (exceeding the original 5:1 target design), thereby increasing the operational flexibility of SOFCo’s fuel cell module. Effects of air flow and feed preheat temperature were examined to establish control boundaries. A good performance match between the C1- and C2-scale reformers demonstrated that SOFCo’s design/scale-up strategy was successful.

The C2 reformer with instrumentation was fabricated and closely integrated into the C2 power module. The SOFCo design incorporates a removable catalyst housing for maintenance and replacement. Final installation and shipment to CPG for testing was completed in December 2004. Initial reformer light-off in the C2 system at CPG was performed in August 2005.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## **ARTICLE 3. Subtask 5 System Development**

### ***C1 AND C2 DEVELOPMENT***

**OBJECTIVE:** Design, build and test a C1 development article to provide CPG-SOFCo with early system and fuel cell operating experience prior to testing the Phase 1 (C2) deliverable. In parallel, design, build and deliver the C2 power module to CPG for integration of the balance of plant and performance of the Phase I demonstration test.

**APPROACH:** The C1 development unit provided a platform for testing an integrated fuel cell system with the general characteristics of the Phase 1 demonstration unit, C2. C1 testing with two 48-cell stacks was completed in September 2004 and verified Phase I system design operating conditions, control strategies and material compatibility.

The C2 development unit built from the C1 experience and was the intended test unit to demonstrate the DOE SECA Phase I performance milestones. C2 was a 4-stack arrangement using 10-cm cells and interconnects with 70 cells per stack. The fuel cell stacks were expected to deliver more than 5 kWe gross power. Greater thermal integration and compactness of this system as compared with C1 was achieved by packaging the stacks, manifold, heat exchanger, combustor and reformer inside the hot enclosure.

**EXPERIMENTS:** SOFCo delivered the C1 test unit to CPG in October 2003. Stack simulators were installed, balance of plant and controls connected, with system operation and controls checkout following. In June 2004, two 48-cell stacks were installed and operational tests initiated in July. Operational tests continued into September 2004, at which time the unit was shut down and attention at CPG focused on C2 balance of plant and controls development. SOFCo delivered the C2 unit with 4 stack simulators to CPG in December 2004. Early operational checkout of the C2 power module components (startup burner, recuperator and insulation system) was completed in January 2005 using CPG air and fuel delivery components (not the C2 BOP components) and manual control to demonstrate thermal performance of the power module. Following successful initial testing, the system was shut down at the end of January. CPG continued fabrication and installation of BOP hardware, electricals and controls software. Hot functional testing resumed in early August to allow CPG checkout of BOP component operation, controls and software during hot operation. Tests with stack simulators were completed in October with preparations underway to install the four, 70-cell stacks for C2 fuel cell Phase I test performance runs. [REDACTED]

**RESULTS AND DISCUSSION:** The following summarizes operation and test results from the C1 and C2 test phases.

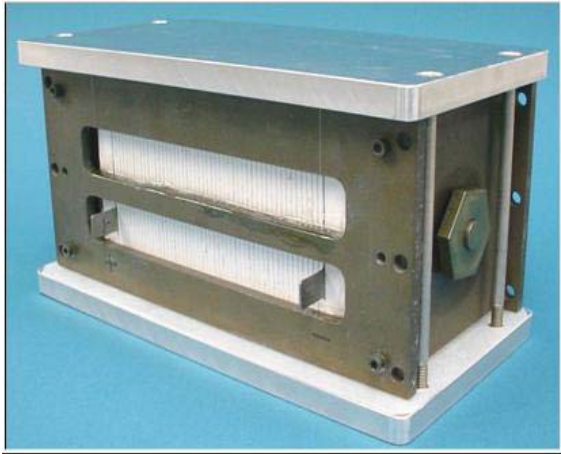
### **C1 Program**

The C1 unit was tested with two 48-cell stacks. An early operational check that involved CPOX startup damaged one of the two stacks. However, C1 operation and controls development successfully continued with one stack. More than 500 watts of power were produced with fuel cell system operation and controls exercised at steady state and transient modes. All system components successfully performed including the reformer, heat exchangers, combustor and insulation system. CPG was able to implement and verify control accuracy for all of the control loops. Overall, the C1 system met its objective by providing the CPG-SOFCo team with the experience of designing, fabricating and testing a fuel cell system. CPG gained valuable

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

experience regarding controls operation and tuning that were important for C2 design and implementation.

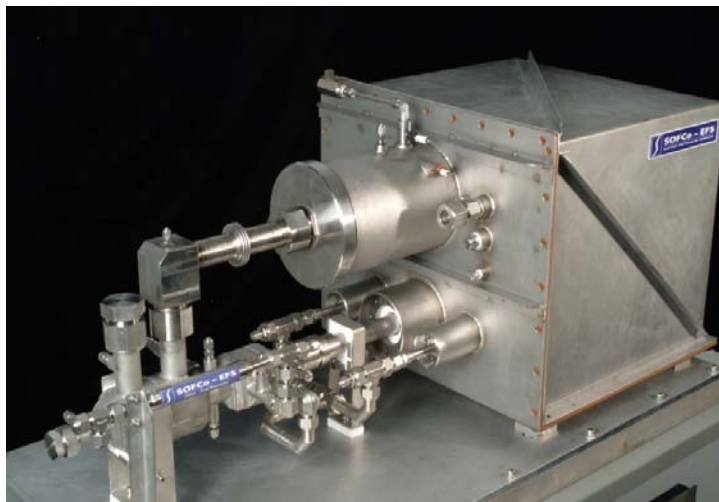
**Figures 53 and 54** show the C1 stack and the two C1 stacks installed in the C1 power module (**Figure 55**) prior to installation of the upper half of the power module enclosure. **Figure 56** shows a representative power trace during C1 operation and checkout in which power was sequentially stepped to zero-power and returned to about 400 watts. Performance and control of the fuel reformer were also verified during C1 operation.



**Figure 53. C1 stack and cage assembly**



**Figure 54. Stacks (2) installed and insulated in C1 hot box**



**Figure 55. C1 power module**

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

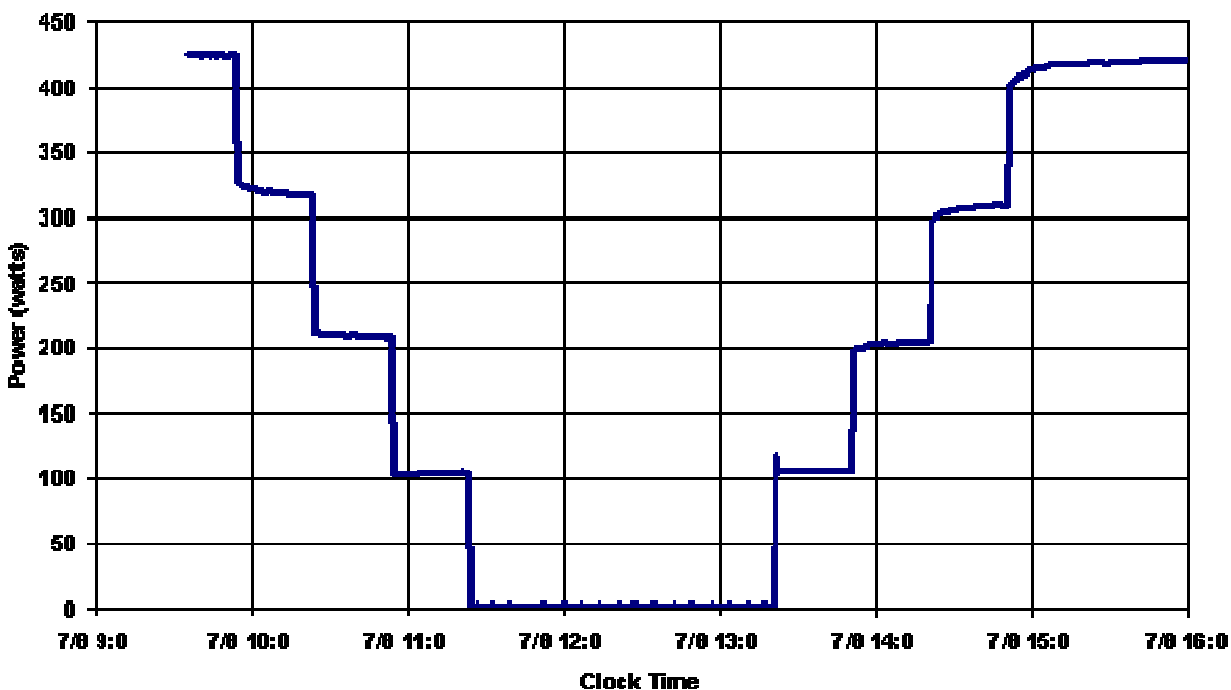


Figure 56. Example C1 performance during operation and controls checkout

### C2 Program

Discussion of the C2 program includes test operations completed from the initial January light-off of the startup burner and power module heat up through controls tuning in preparation for stack installation, [REDACTED]

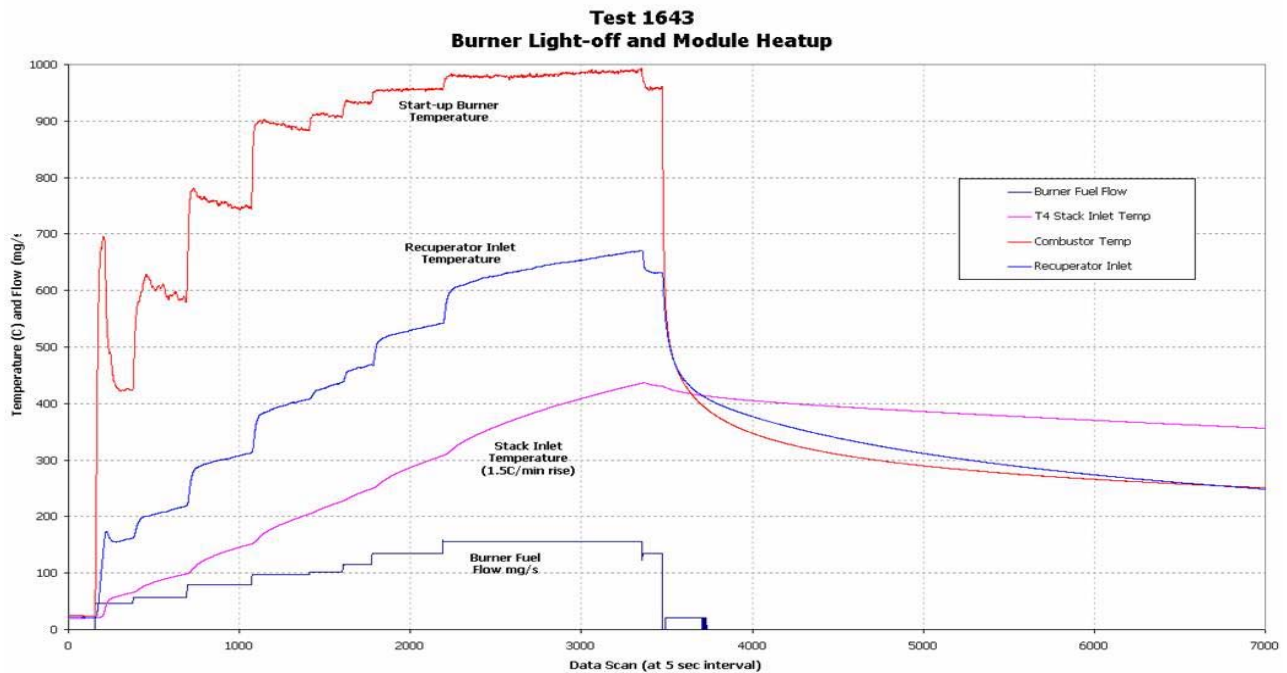
SOFCo delivered the C2 Power Module to CPG the week of December 13. Stack simulators (4) were installed. CPG progressed with preliminary BOP and controls fabrication and debug with initial emphasis on startup burner operation and control. After demonstrating startup burner light-off capability, an early “first manual heat up” of the C2 power module was performed during the week of January 18. In this system configuration separate air blowers (not the final blower) were connected to the recuperator and startup burner, as well as natural gas connection to the startup burner. **Figure 57** shows the configuration of the system on January 18 used for these tests.

Operational and system checkout tests progressed well in January and demonstrated the ability of the startup burner to heat the power module stack inlet to about 700°C as required for fuel cell stack operation. Operations of the startup burner, recuperator heat exchanger, insulation system and enclosure pressure seals (and bellows) were found acceptable. **Figure 58** shows selected temperature and fuel flows for test 1643 performed on January 18. Test 1643 progressed at a heat up rate of about 1.5°C/min to a stack inlet temperature of about 437°C before over-night shutdown. Maximum startup burner fuel flow was 155 mg/s (about 7200 w) and was manually controlled to limit the startup burner temperature to about 1000°C for this first high-temperature startup test.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 57. Power module and BOP configuration at CPG for first power module heat up**

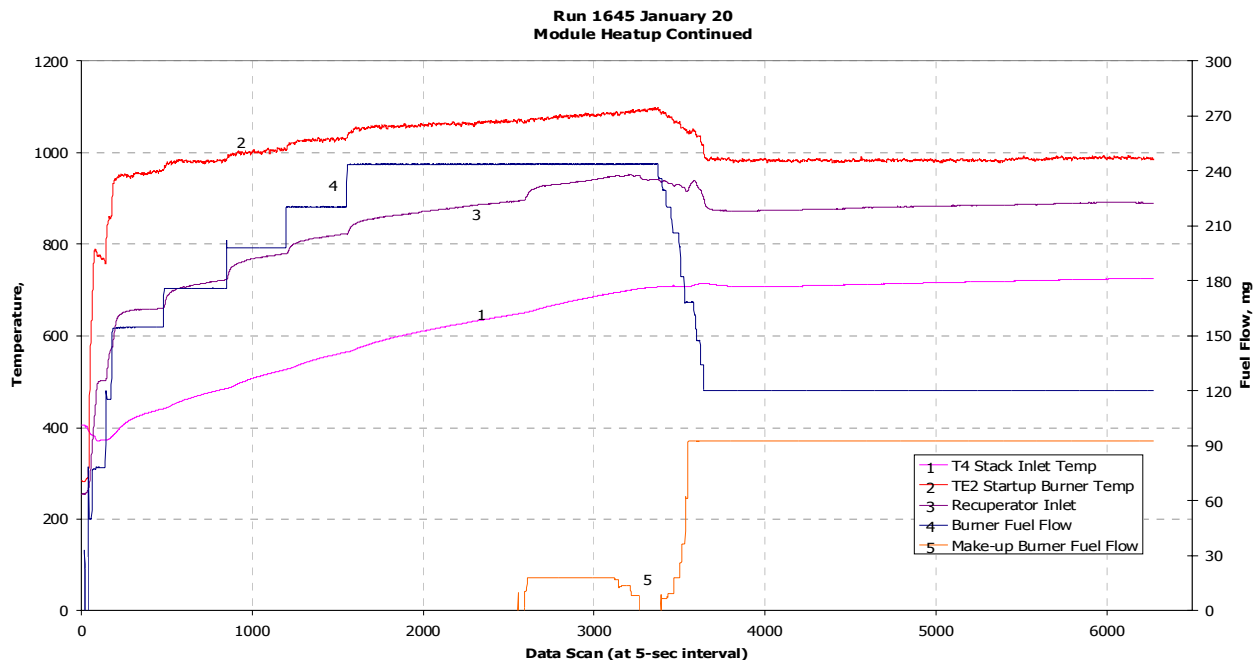


**Figure 58. Selected temperature and fuel flows for test 1643 performed on January 18**

**Figure 59** shows selected temperature and fuel flows for test 1645 performed on January 20. Heat up progressed at a rate of about 1.2°C/min to about 710°C before balancing heat input between the startup burner and makeup fuel line to limit the startup burner compartment temperature to about 1000°C. The operating temperature limit of the startup burner and surrounding cavity had not been determined so a limit of about 1000°C was set. With the stack

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

simulators installed and the reformer not operational (reformer controls not yet established in January), it was realized that the reformer (and reformate combustion in the stack simulators) would become a source of heat input at a stack inlet temperature above 400°C, thereby limiting the startup burner heat rate. For reference, the reformer would be operational prior to the stack inlet temperature reaching 400°C so as to provide a reducing gas on the stack anode for subsequent heat up to the stack operating temperature of >700°C. As a result, the makeup fuel line at the combustor inlet was used to provide supplemental energy input in the absence of an operational reformer. As shown, the startup burner fuel flow of 120 mg/s and the makeup fuel flow of 93 mg/s provided a combined energy release of 9.9 kW (33,800 Btu/hr) and maintained the startup burner cavity at just under 1000°C.



**Figure 59. Test 1645 showing temperatures and fuel flows (startup burner and**

Following the January 20 test, Cummins progressed through BOP component fabrication, assembly, controls checkout and debug. No hot testing of the unit was recorded by CPG until startup burner operation and controls testing resumed on August 3, test 1651.

For reference, a list of the tests with important operating measurements is provided in Table 6. Shown in the table are values that document the average, maximum and minimum values in the data sets for the recuperator inlet temperature and the total fuel flow to the system. As the tabulated data indicates, the manual and automatic control features maintained recuperator inlet temperatures below the 950°C limit except in tests:

- 1645 (average of 951°C over a 20 minute period with a maximum value of 952°C),
- 1656 (average of 952°C over a 24 minute period with a maximum value of 960°C),
- 10030 (intermittent excursions above 950°C with a maximum value of 970°C during a 20 second interval that had an average value of 960°C)
- 10040 (average of 951°C over a 90 minute period with a maximum value of 958°C),
- 10041 (average of 960°C over a 118 minute period with a maximum value of 972°C).



## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

Whereas 950°C was the target “not to exceed” value for control purposes, the temperatures and times noted above are not considered to be damaging based on the heat exchanger manufacturer’s feedback.

Of interest in the test summary listing, however, are the two tests (1656 and 09066) with combined CPOX, startup burner and makeup fuel flows in excess of 500 mg/s. For reference, with operating stacks the equivalent “makeup” fuel flow rate (unutilized stack fuel flow) would be on the order of 10 to 70 mg/s to limit the recuperator inlet temperature to less than 950°C. Test 09066 experienced a control step change in startup burner and makeup fuel flow on the order of 380 mg/s in a 10 second period. Following this step change, the flow dropped back to a normal value. Although this spike in fuel flow may not have been insignificant, it is small in comparison to similar fuel flow rates with much longer duration in test 1656.

Test 1656 on August 12 appears to be the most severe test in the recorded data sets [REDACTED] during an accelerated- power module heat up – the stack inlet temperature increased in excess of 5°C/min. **Figure 60** shows key temperatures and fuel flow rates during the test. [REDACTED]

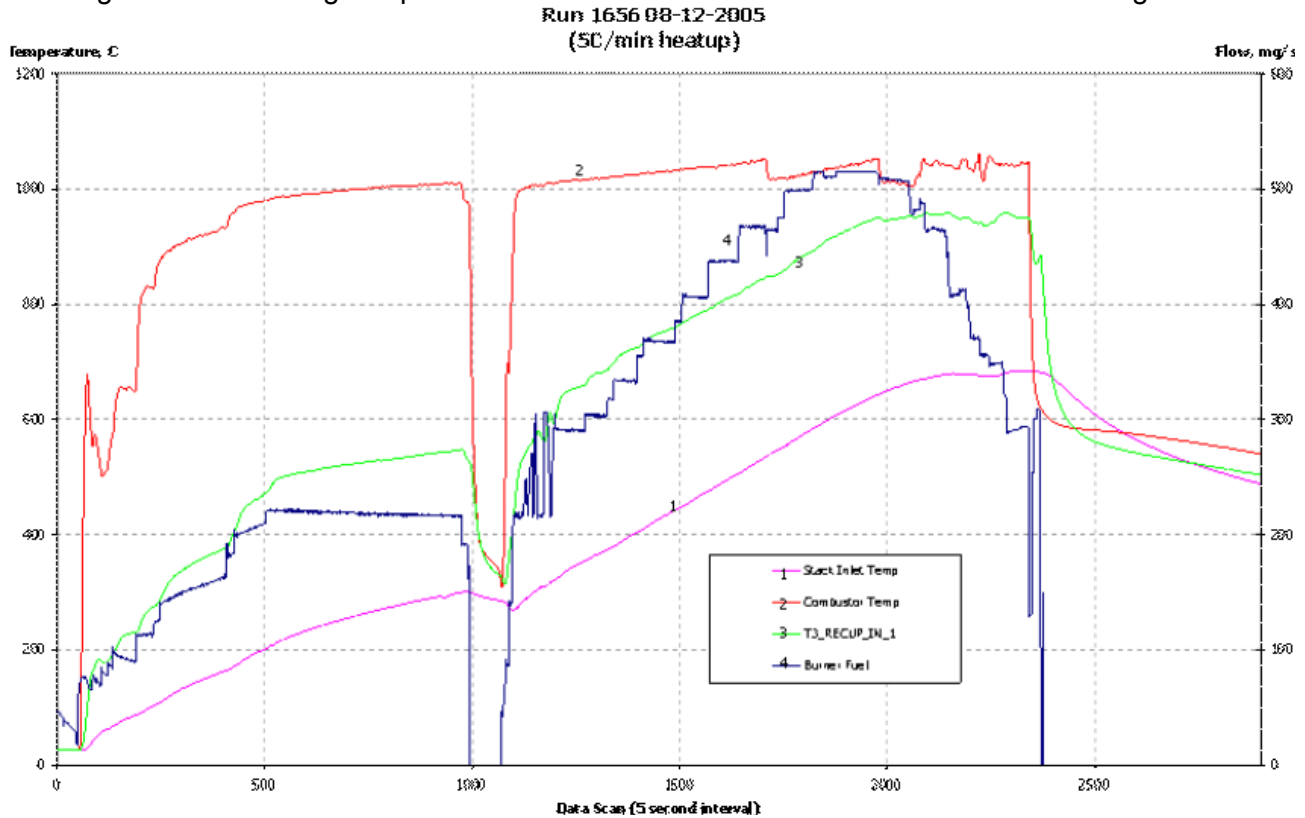
# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

**Table 6. Summary of Test Data Sets and Measured Recuperator Heat Exchanger Inlet Temperature and Fuel Flow Rate**

Date	Test ID	Recuperator Heat Exchanger Hot Gas Inlet Temperature, C			Fuel Flow: CPOX+Burner+Makeup Flow Rate, mg/s		
		Average	Max	Min	Average	Max	Min
1/18/2005	1643	284	671	21	26	159	0
1/19/2005	1644	<b>(Over-night cool down from 1643 end condition to 1645 startup.)</b>					
1/20/2005	1645	845	952	255	215	262	0
8/3/2005	1651	75	219	35	NA	NA	NA
8/4/2005	1654	160	208	43	NA	NA	NA
8/5/2005	1655	460	677	32	149	251	0
8/12/2005	1656	533	960	25	148	516	0
8/16/2005	1657	438	602	122	161	221	38
8/16/2005	1658	630	653	605	214	218	210
8/17/2005	1660	356	805	190	56	313	0
8/17/2005	1661	769	840	673	256	282	206
8/22/2005	1662	87	218	26	26	63	0
8/22/2005	1663	213	398	36	40	110	0
8/24/2005	1665	385	781	31	55	232	0
9/2/2005	09020	Recuperator inlet temperature data for these tests was not available/logged at CPG. Data for tests 09020, 09040, 09050 and 09066 are of most interest where the startup burner was operational as exhibited by the fuel flow data column.			87	246	0
9/3/2005	09030				2	2	2
9/3/2005	09031				2	2	2
9/4/2005	09040				2	130	2
9/4/2005	09041				2	3	2
9/5/2005	09050				4	121	2
9/5/2005	09051				3	4	2
9/6/2005	09066				37	521	2
9/14/2005	09140	600	629	518	102	213	61
9/14/2005	09141	359	618	325	38	103	2
9/19/2005	09190	674	781	62	124	139	24
9/19/2005	09191	787	790	780	130	133	129
9/20/2005	09200	925	940	768	177	274	92
9/20/2005	09201	936	938	934	167	175	133
9/27/2005	09270	935	948	899	124	212	109
9/28/2005	09280	936	939	933	117	123	111
9/28/2005	09281	936	940	932	117	123	110
9/29/2005	09290	929	940	897	117	125	111
9/29/2005	09291	913	927	897	116	118	113
9/30/2005	09300	925	929	921	115	117	113
10/2/2005	10020	936	940	932	117	119	114
10/2/2005	10021	935	940	931	117	120	114
10/3/2005	10030	936	970	912	114	281	86
10/3/2005	10031	936	941	932	86	90	82
10/4/2005	10040	940	958	915	113	201	76
10/4/2005	10041	941	972	933	116	209	102
10/5/2005	10050	936	943	898	124	197	87
10/5/2005	10051	937	940	934	175	181	169
10/6/2005	10060	937	940	931	174	211	142
10/6/2005	10061	903	941	892	168	176	147
10/7/2005	10070	831	941	515	133	193	2

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

Testing continued through September and into October as identified in **Table 6**. During that



**Figure 60. Test 1656 (August 12) showing temperatures and startup burner + makeup**

time, various tests at hot conditions were explored to demonstrate control capability for the SECA Phase I test requirements, including reformer operation. Understanding of the power module and BOP operation was also gained with stack simulators installed. [REDACTED]

**CONCLUSION:** The C1 development unit testing at CPG provided system operational capability to verify control techniques and validate component design for C2 implementation. The C2 Phase I prototype SOFC module was delivered to CPG in December 2004. Initial operational checks of the heat exchanger, startup burner and power module insulation system (January 2005) were determined to be acceptable. BOP operational checks and controls tuning was initiated in July and continued into October. [REDACTED]

[REDACTED] SOFCo and CPG derived substantial fuel cell power system design and operational experience from the SECA Phase I program. Of important success to SOFCo and SECA program contribution was SOFCo's development and demonstration of a compact, waterless CPOX. The fuel processor was demonstrated in the SOFCo laboratory and in the C2 power module with fuel supplied from pipeline natural gas.

## II. Performance Improvements and Cost Reduction

In parallel with the baseline work directed at cells, stacks, reforming and the SOFC module sub-system for the C2 demonstration, SOFCo performed development activities aimed at improving stack performance and reducing the cost of key materials. The primary areas of focus in these

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

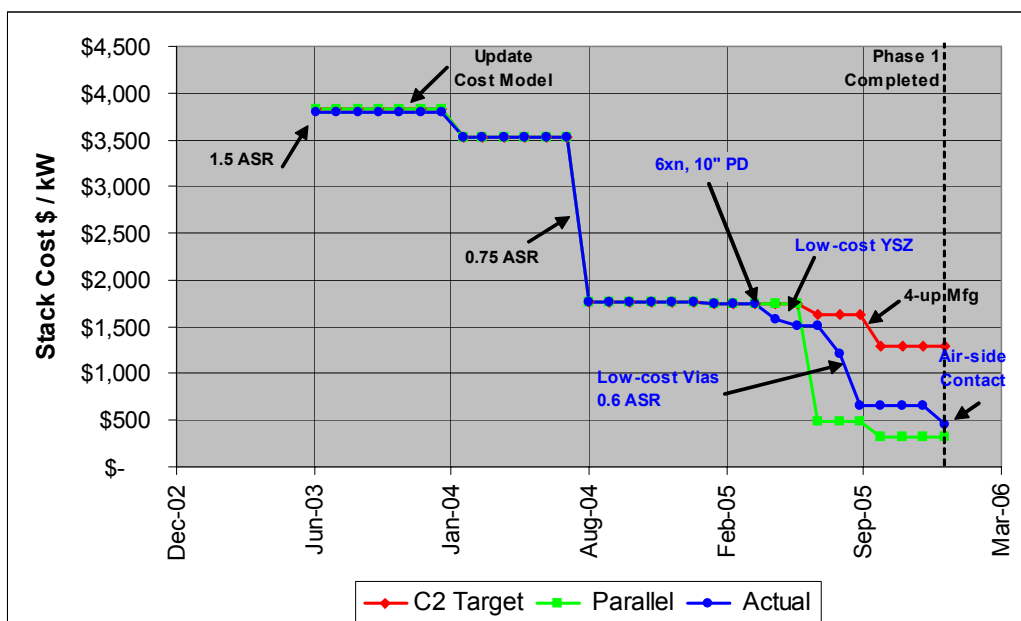
parallel activities are summarized in Table 7. The following sections summarize the work performed in each area. It should be noted that SOFCo successfully developed a low-cost Ni cermet composition for the fuel-side contact ink early in the Phase I period, and that this material was adopted for the baseline stacks. As such, this topic is not discussed below.

**Table 7. Summary of Parallel Path Improvements**

Parameter	Baseline for C2 Stacks	Parallel Path Improvements	
		Planned	Achieved
Performance (ASR)	Cells: 0.65 $\Omega$ -cm <sup>2</sup> Stacks: 0.75 $\Omega$ -cm <sup>2</sup>	Cells: 0.45 $\Omega$ -cm <sup>2</sup> Stacks: 0.50 $\Omega$ -cm <sup>2</sup>	0.60 $\Omega$ -cm <sup>2</sup>
Interconnect material	High-cost 3YSZ	Low-cost 3YSZ	Yes
Interconnect conductors	Pt-YSZ cermet	Air-side vias: Perovskite Fuel-side vias: Ni cermet	Yes Yes
Fuel-side contact ink	Pt-Au-YSZ	Ni Cermet	Yes
Air-side contact ink	Pt-Au-YSZ	Perovskite Oxide	No
Air-side bond layer	Pt-PSM	Perovskite Oxide	Yes
Stack footprint	10 cm x 10.5 cm	15 cm x 16 cm	Yes

Achieving the improved cell (and stack) performance and elimination of the high-cost materials from stacks were determined to be critical for meeting the Phase I cost target of \$350/kW for the stacks (when manufactured at high volume). **Figure 64** provides further insight into the impact of these changes on the projected stack costs, and includes the expected timing for completing the development activities and implementing the improvements. The red curve provides an estimate for the cost of the 10cm baseline stacks to be used for the C2 demonstration; the projected cost for these stacks at high volume was about \$1300/kW. The green curve shows the impact of implementing the improvements and the planned timing for completing key development activities. Reducing the stack ASR to 0.5  $\Omega$ -cm<sup>2</sup> and eliminating the high-cost materials was projected to reduce the stack cost to about \$325/kW. The blue curve shows the actual results achieved in the parallel path effort. As described in the following sections, SOFCo achieved all of the planned outcomes with two exceptions: 1) short stack ASR was reduced to 0.6  $\Omega$ -cm<sup>2</sup> rather than the target ASR of 0.5  $\Omega$ -cm<sup>2</sup>; and development of an acceptable air-side contact ink was not completed. Based on the demonstrated improvements, a stack cost of about \$450/kW (at high volume) was projected.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 64. Parallel path cost reduction roadmap**

## Subtask 2 – Advanced Cell Development

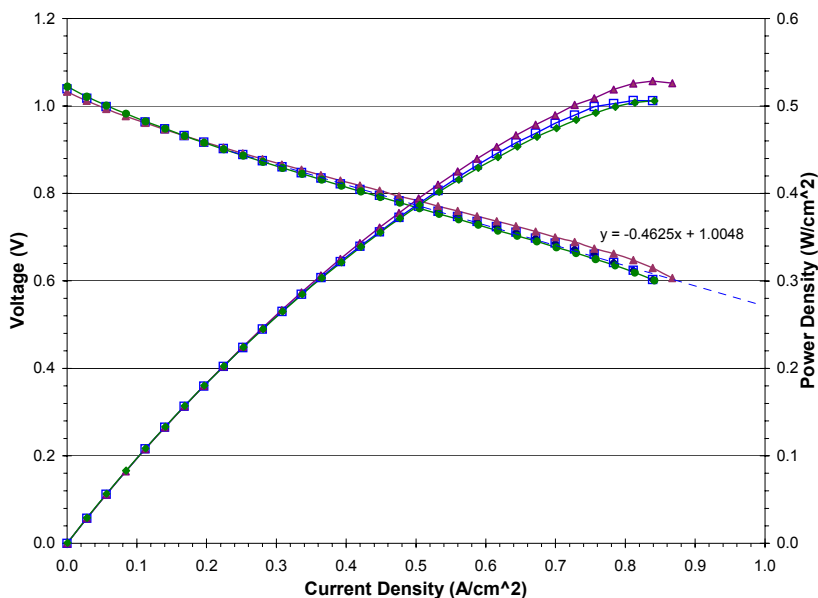
**OBJECTIVE:** Internally develop and/or select from the external supply base, a cell having an 825°C ASR of less than 0.45 Ω-cm<sup>2</sup>. In addition, demonstrate short stacks with such cells having an 825°C ASR of 0.5 Ω-cm<sup>2</sup> or less. For any internally developed cell, establish a fabrication capability within SOFCo’s prototype facility to support development and testing.

**APPROACH:** SOFCo has continued to evaluate cells from external suppliers that are of suitable design for use in SOFCo stacks, basically traditional electrolyte-supported cells utilizing various ScSZ electrolytes. Externally supplied cells have not met ASR targets, and as a result, SOFCo has pursued development of its own cell. To achieve the ASR targets, SOFCo is utilizing a ScSZ electrolyte and mixed electronic and ionic conducting (MEIC) cathodes having low over-potential.

**EXPERIMENTS:** SOFCo’s approach continues to use the single-cell test stands to screen the performance of full-size cells prior to introducing such cells into short stack tests. The test stands are able to achieve representative fuel utilization as exhibited during short stack testing. Initial ASR calculations are performed by taking the slope of the VI curve between approximately 0.6 and 0.8 volts to compare cell performance. A more accurate ASR calculation can then be performed based on SOFCo’s fuel distribution model of the single-cell test stand combined with the VI scan obtained for the cell, and the noted fuel utilization levels. The standard conditions for the single-cell tests reported herein are fuel flows to achieve 30-60% fuel utilization, 350-500 mA/cm<sup>2</sup> current densities, and using a humidified (3% H<sub>2</sub>O) mix of 50% hydrogen and 50% nitrogen. With the addition of an electrochemist to the cell team, SOFCo resumed the use of button cell testing and impedance spectroscopy to more fully explore electrode performance and the source of ASR contributions for the internally developed cells.

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

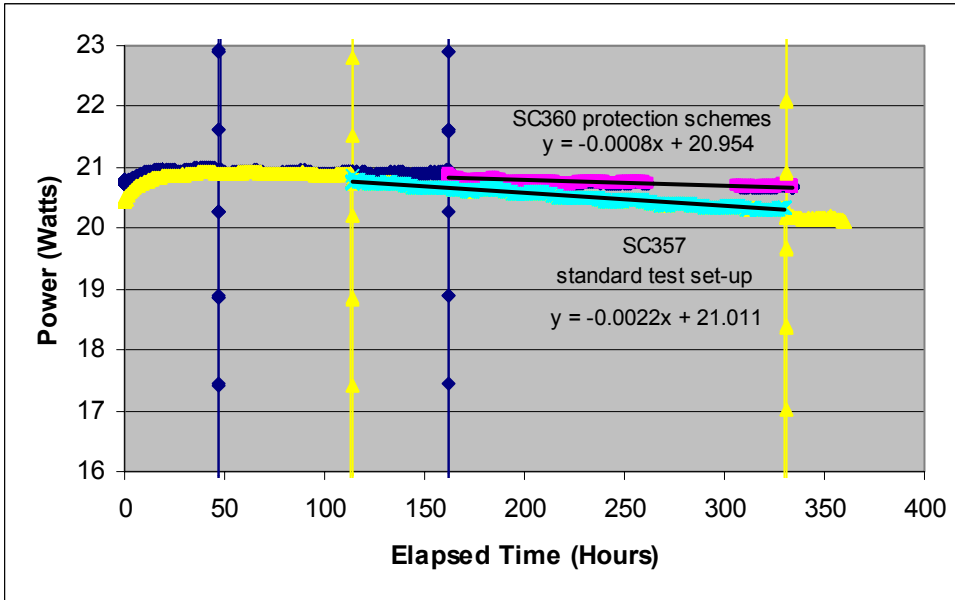
**RESULTS AND DISCUSSION:** SOFCo initially focused internal cell development efforts on [REDACTED] mixed electronic and ionic conducting (MEIC) cathodes. The performance of these cells (~90 micron thick ScSZ electrolyte) at 825°C (SOFCo's average stack temperature) was an ASR of ~0.45  $\Omega\text{-cm}^2$  as measured by the slope method. **Figure 65** shows the VI curves for three such cells – achieving over 500 mW/cm<sup>2</sup> at a fuel utilization of ~90%. The model ASR determination for these three cells averages less than 0.40  $\Omega\text{-cm}^2$ . Near the end of the Phase 1 period, SOFCo did eventually receive an externally supplied cell that met this 0.45 ASR target. The cell [REDACTED] matched the performance of SOFCo's own cell; it appears that a similar ScSZ electrolyte [REDACTED] cathode composition was used.



**Figure 65. Typical performance of SOFCo developed ScSZ electrolyte-supported cells at 825°C tested under humidified 50:50 H<sub>2</sub>:N<sub>2</sub> fuel**

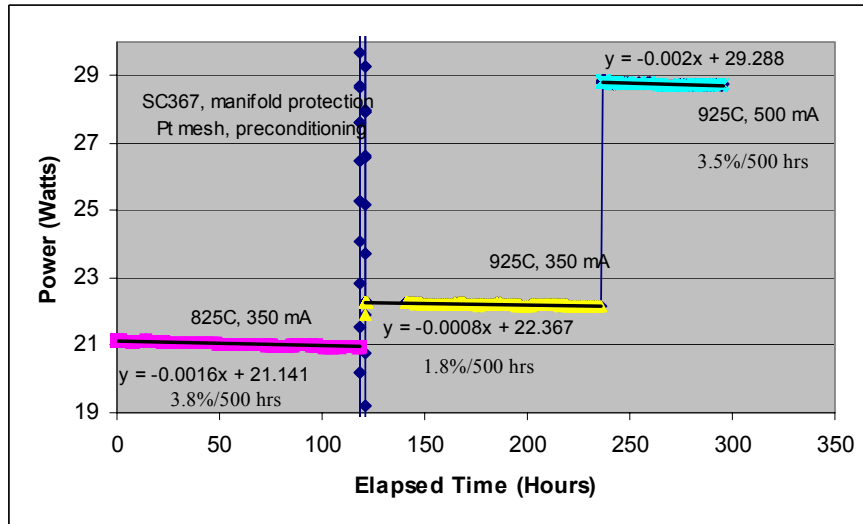
Testing of cells containing MEIC [REDACTED] cathodes presents special challenges, given the more severe impact of chromium contamination of such cathodes. To minimize the impact of chromium contamination on these cells, SOFCo has devised a single cell test that incorporates a dense disk of a conductive ceramic between the cell and the Inconel 800 manifold and an alumina sheath along the gas flow path of the Inconel manifold. The impact of these test design features is highlighted in **Figure 66**, where the measured cell degradation rate was reduced from 5.3% to 1.9% per 500 hours. During 2004, PNNL performed aging studies of ScSZ electrolytes under a CRADA. Those results suggest a ~2% per 500 hours of degradation over the initial 1000 hours of operation resulting from electrolyte aging. It should be noted that future advanced cell designs will likely incorporate thin electrolytes in which the electrolyte aging phenomenon will be minimized.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 66. Degradation of SOFCo cells with and without manifold protection schemes**

There is some concern about the long-term prospects for such MEIC cathodes, based in part on limited investigations at PNNL on LSF20 cathodes under the Core Technology Program. To further investigate degradation behavior, testing was expanded to include a broader range of operating conditions to more fully understand the operating window for the cathode compositions being utilized. As shown in **Figure 67**, the high-end of the operating temperature range for SOFCo stacks (925°C) does not appear to accelerate degradation of the [ ] cathode. Once again, these degradation rates are in line with rates expected solely from electrolyte aging. However, post-test SEM/EDS analysis of cells with this [ ] cathode has revealed some undesirable interfacial reactions. **Figure 68** shows that some strontium



**Figure 67. Degradation results for SOFCo cells subjected to more extreme conditions expected within stacks**

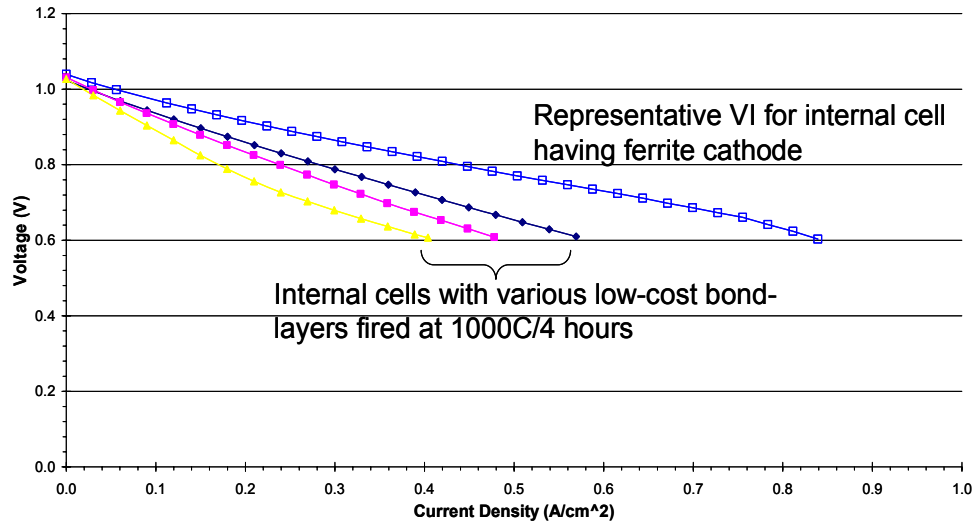
# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

migration has occurred [REDACTED], which likely is a source of increase in the over-potential of these cathodes.

[REDACTED]

Further internal cell development using the [REDACTED] cathodes was de-emphasized in late 2005 because of the interfacial stability issues (Sr migration) and the observation that the cell performance was damaged by application of the prime candidate low-cost bond-layer to the cell. As shown in

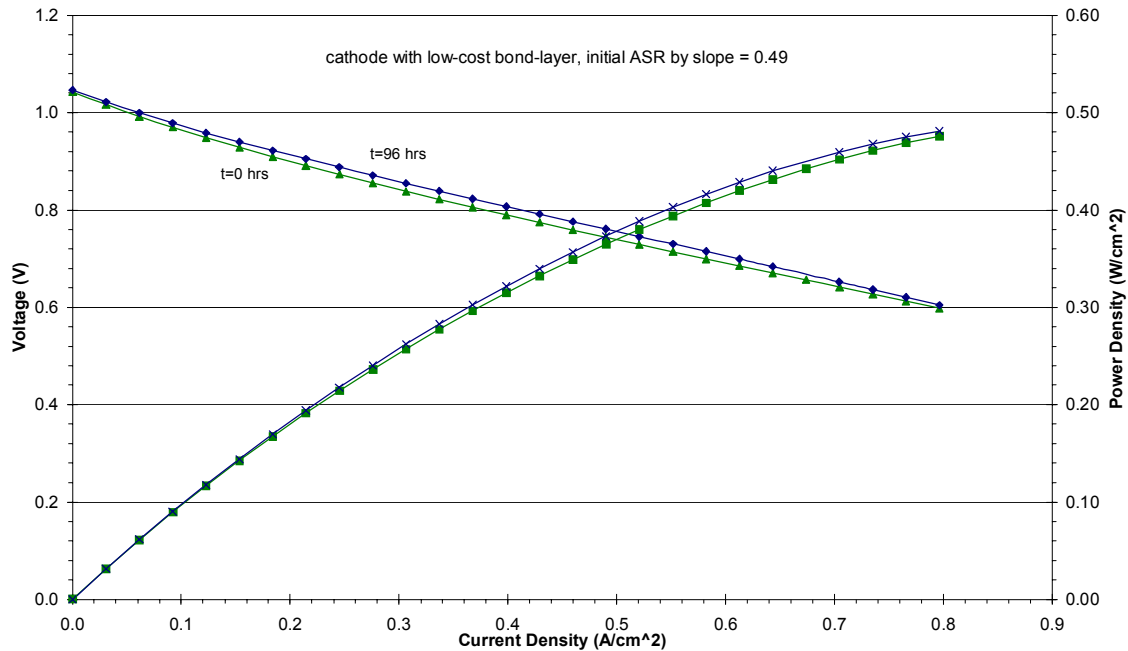
**Figure 69**, the application of the low-cost bond layer to cells using the [REDACTED] cathode resulted in increased cell ASR. This observation also eliminated the possibility of utilizing the one externally supplied cell [REDACTED] which met the ASR requirements.



**Figure 69. Performance of cells using a [REDACTED] cathode with the addition of the SOFCo low-cost bond layer material**



# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

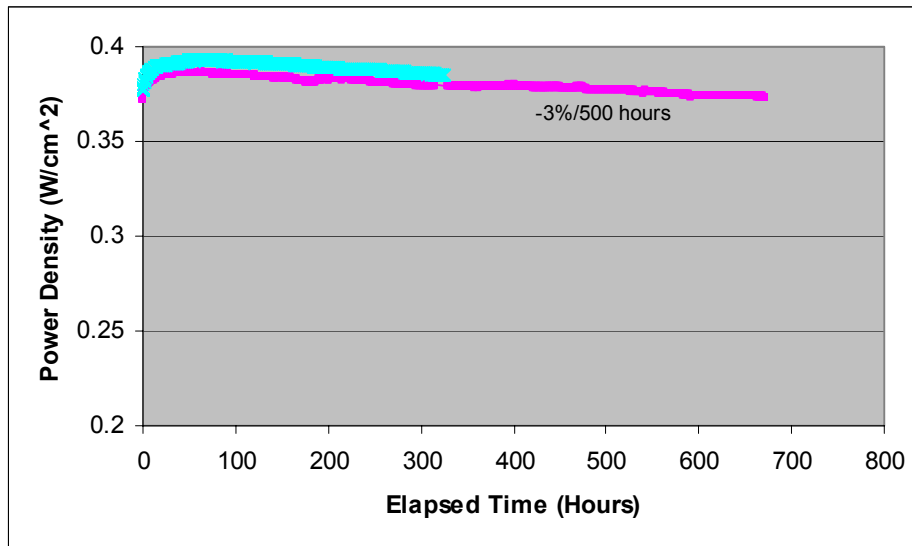


**Figure 70. 825°C performance of SOFCo cells using new MEIC cathodes (with low-cost bond-layer also present)**

To achieve the combined need to meet an 825°C ASR of less than 0.45  $\Omega$ -cm<sup>2</sup>, and be able to utilize the selected low-cost perovskite air-side bond-layer, SOFCo began investigating another class of MEIC cathodes. These cathodes have not yet received the level of investigation by the SOFC community that the [ ] cathodes have, but there is sufficient data available to merit their application to SOFCo cells. In fact, the chemistry of such cathodes would be expected to exhibit better compatibility with the low-cost perovskite bond-layer SOFCo has been attempting to implement. As shown in **Figure 70**, cells with the new cathode have an 825°C ASR of under 0.5  $\Omega$ -cm<sup>2</sup>, even with the low-cost bond-layer present. The electrochemical model evaluation for multiple cells with these cathodes suggests that the ASR is ~0.45  $\Omega$ -cm<sup>2</sup>. Degradation data for these initial cathode compositions is shown in **Figure 71**. The observed degradation is attributed to some phase instability with the particular cathode composition used for the cells tested, as determined by x-ray diffraction analysis. It should be noted that exploration of the new cathode material was initiated toward the end of Phase I, and was continued under internal SOFCo funding.

**CONCLUSIONS:** SOFCo has successfully demonstrated electrolyte-supported cells having an ASR of less than 0.45  $\Omega$ -cm<sup>2</sup>. However, early cells used a MEIC [ ] cathode that appears to have some undesired long-term performance issues. As a result, SOFCo switched internal cell development to a new class of MEIC cathodes. In doing so, SOFCo has been able to meet the combined requirement of ~0.45  $\Omega$ -cm<sup>2</sup> ASR for an electrolyte supported cell at 825C, while also utilizing the low-cost perovskite bond-layer. As discussed in the next section, these cells (with the low-cost bond layer) were successfully tested in short stacks. Further work will be required to optimize the composition and processing of the new cathodes, and to achieve the desired performance in stacks.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 71. Single cell degradation data (825°C) for SOFCo cells using an initial MEIC cathode composition**

## **Subtask 2 – Low-Cost Materials for Air-side Connection**

**OBJECTIVE:** To replace the platinum based cathode-side bond layers and contact ink between the cathode bond layer and interconnect (generally referred to as air-side bumps) with low-cost materials, while minimizing their contribution to stack ASR.

**APPROACH:** The current baseline SOFCo stacks have utilized a platinum-cermet bond layer that is applied to the cathode of the cells and a platinum-cermet contact ink (bump ink) to provide for the air-side connection of cells to the interconnects during stack assembly. It is necessary to replace these materials with lower cost materials to meet the SECA Phase 1 cost targets. Highly conductive oxide perovskite materials have been selected to meet this objective. A number of laboratory-scale evaluations have been performed to verify that the low-cost bond layers and bumps have sufficiently low resistance so as to have a negligible impact on overall stack ASR. Once low resistances were verified, candidate bond layers were transitioned into single-cell testing to verify that they do not pose any degradation to cell performance. Next, extensive interconnect-cell subassembly experiments were performed to determine a cell cathode-side to interconnect assembly method that provided good electrical continuity. In parallel, an [ ] interconnect-to-cell connection was developed to reduce the stress on the cathode-side bumps during service. Finally, the low-cost cathode-side bumps and bond layer and the [ ] anode-side connection were tested in short stacks.

**EXPERIMENTS:** Various testing of selected low-cost air-side bond layer materials and [ ] anode-side connection concepts were performed.

## **RESULTS AND DISCUSSION:**

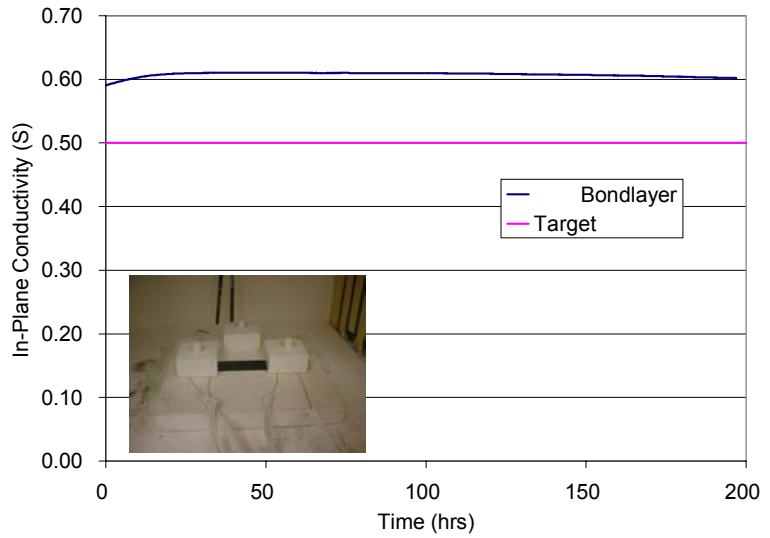
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# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

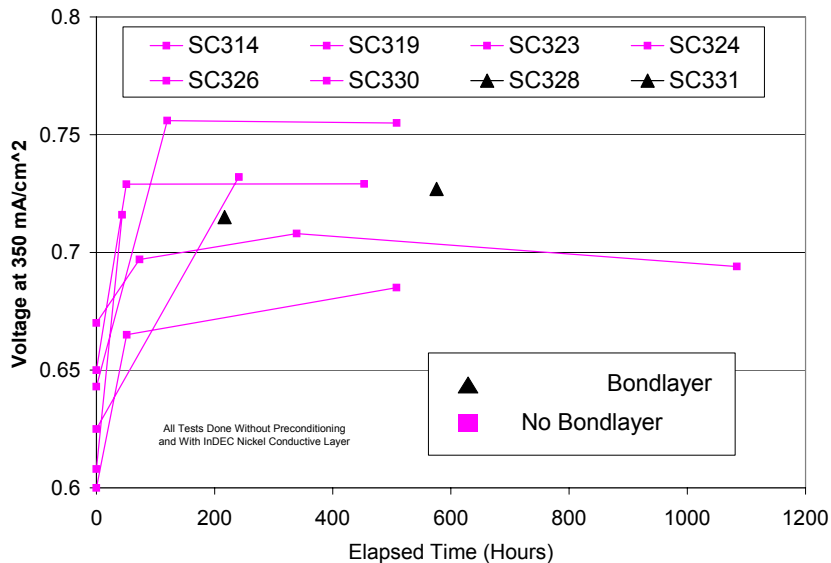
This [ ] anode-side connection was subsequently successfully demonstrated in tall stack testing (as previously discussed). The [ ] anode-side connection has now been adopted as SOFCo's standard practice, and was intended to be used in the construction of the C2 stacks.

**Low-Cost Cathode-Side Connection:** Laboratory-scale resistance testing of the candidate low-cost air-side bond layer on an internally developed cell indicates acceptably high in-plane conductivity at 825°C, as shown in **Figure 73**.

Single-cell testing was accomplished using cells with the candidate low-cost air-side bond layer fired at 1000°C. There appears to be no impact on performance due to the addition of the low-cost air-side bond layer, as shown in **Figure 74**. Additional testing was performed with internal single cells using the new MEIC cathode onto which the candidate bond layer was applied (and fired at 1000°C). Again, there appears to be no impact on cell performance (see **Figure 75**) due to the addition of the bond layer or due to the simulated stack conditioning cycle.

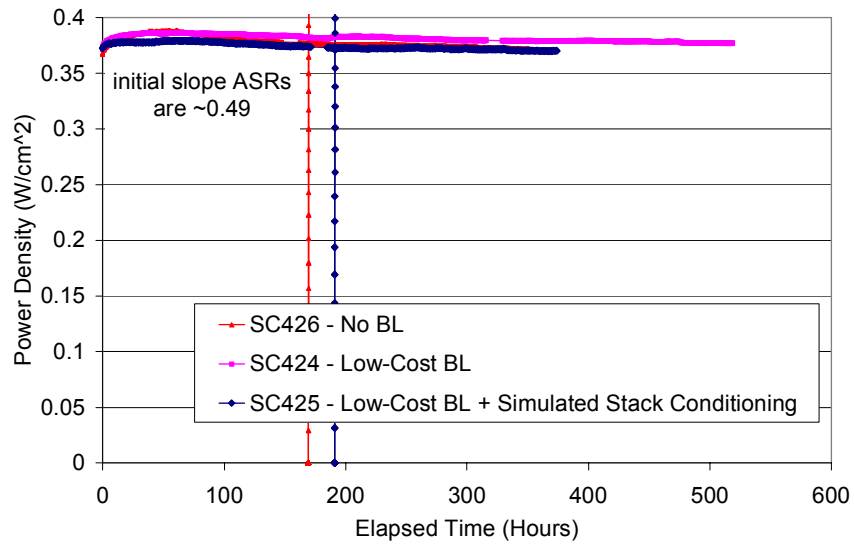


**Figure 73. Low-Cost Cathode Bond Layer Resistance**



**Figure 74. Cathode bond layer in single cell tests**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



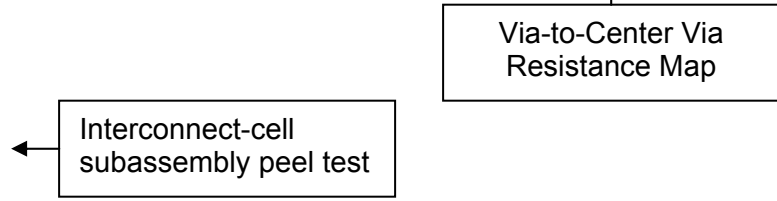
**Figure 75. Performance of low-cost cathode bond layer with**

Laboratory testing of interconnects-cell subassemblies was accomplished. The interconnects were printed on the air-side with a low-cost perovskite adhesion layer and fired. The cells similarly were printed with low-cost perovskite bond layers and fired at 1000°C. The cell and interconnect were then joined with low-cost perovskite bumps and fired at 1000°C. Good electrical continuity and bond strength were achieved for tests with cells having 3YSZ electrolytes, as shown in **Figure 76**.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	
Fuel inlet	1	2.9	2.6	2.4	2.3	2.2	2.3	2.2	2.1	2.2	2.2	####	2.2	2.1	2.2	2.3	2.3	2.4	2.6	2.9	
	2	2.3	2.3	2.2	2.1	2.1	2.1	2.1	2.1	2.1	2.0	2.2	2.1	2.1	2.0	2.2	2.1	3.5	2.3	2.4	
	3	2.7	2.2	2.1	2.1	2.0	2.0	2.0	2.1	2.1	2.0	2.0	2.0	2.0	2.0	2.0	2.1	2.1	####	2.3	2.6
	4	2.1	2.0	2.1	2.1	2.0	1.9	2.0	2.0	1.9	3.3	2.0	1.9	1.9	####	1.9	2.0	2.0	2.0	2.2	
	5	2.5	2.2	2.0	1.9	1.9	1.9	2.0	1.9	1.9	1.9	1.9	1.9	1.8	1.9	1.9	2.0	2.0	2.0	2.2	2.5
	6	2.1	####	2.0	2.0	1.9	1.9	1.8	1.8	1.8	1.8	####	####	1.8	1.8	1.9	1.9	1.9	2.0	2.1	
	7	2.4	2.2	2.0	2.1	1.9	2.0	1.9	1.7	1.7	1.8	1.7	1.8	1.8	1.8	1.8	1.8	1.9	2.0	2.1	2.2
	8	2.1	2.0	2.0	1.8	1.8	1.8	1.7	1.7	1.6	####	1.6	####	1.7	1.6	####	1.8	1.9	1.9	2.0	
	9	2.2	2.0	1.9	####	1.9	1.7	####	1.8	1.5	1.5	1.5	1.7	1.7	1.7	1.7	1.8	1.9	1.9	2.1	2.2
	10	2.0	1.9	1.8	####	1.7	1.7	1.6	1.5	1.4	0.8	1.4	1.5	1.6	2.2	1.8	1.8	1.9	2.0	####	
	11	2.2	2.0	1.9	1.9	1.8	1.7	1.7	1.6	1.5	1.5	1.5	1.7	1.7	1.8	1.8	1.8	1.8	2.0	1.9	2.2
	12	2.0	1.9	1.9	1.9	1.8	1.8	1.7	1.8	1.6	1.6	1.7	1.8	1.8	1.8	1.8	1.8	1.9	1.9	2.1	
	13	2.3	2.0	2.0	1.9	####	1.8	1.8	1.8	1.8	1.8	1.9	1.9	1.9	1.8	1.9	1.9	####	2.0	2.1	2.4
	14	2.1	####	2.0	1.9	2.0	2.0	2.0	2.0	1.9	2.0	2.0	1.9	2.0	1.9	2.5	2.0	2.0	2.1	2.3	
	15	####	2.1	2.0	2.0	1.9	2.0	2.0	2.0	1.9	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.1	2.2	2.4	
	16	####	2.1	2.1	2.0	2.0	2.0	3.6	2.0	2.1	2.0	2.1	2.0	2.1	2.1	2.1	2.1	2.1	2.2	2.3	
	17	2.5	2.3	2.3	####	2.2	2.1	2.1	2.1	2.0	2.1	2.1	2.1	2.2	2.0	####	2.1	2.2	2.2	2.4	2.6
	18	2.4	2.3	2.3	2.2	2.2	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.2	2.2	2.2	4.1	2.4	2.9	
	19	3.0	2.7	2.6	2.4	2.4	2.3	2.3	2.2	2.2	2.2	2.2	2.2	2.3	2.4	2.4	2.5	2.5	2.6	2.7	3.0



**Figure 76. Low-cost cathode-side connection test**

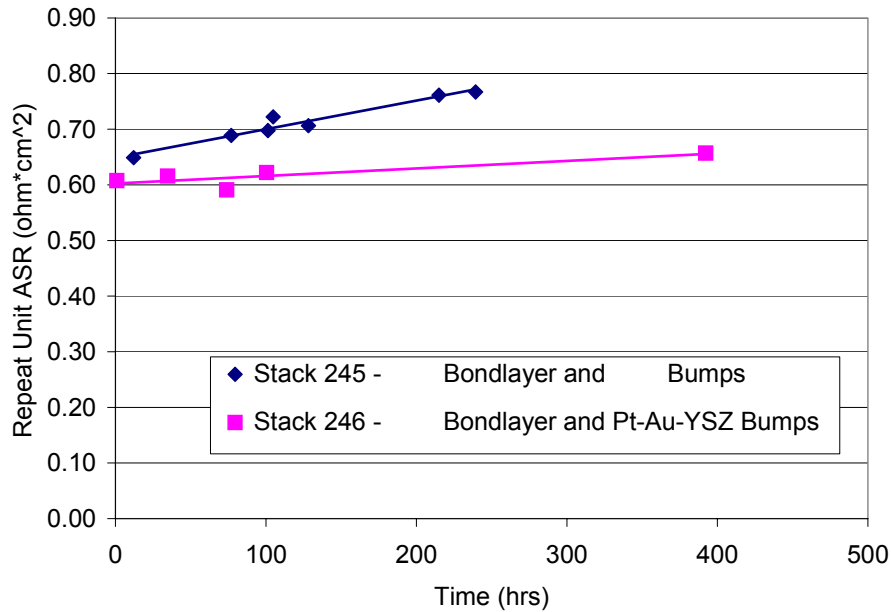
Similar tests using cells with thinner ScSZ electrolyte-supported cells did not achieve acceptable electrical continuity and bond strength. These poorer results are likely due to the increased thermal expansion differential and decreased thickness for these cells.

In spite of the attachment problems observed in the above test, short stack testing was accomplished using internally developed ScSZ electrolyte-supported cells with the candidate low-cost air-side bond layer co-fired with the cathode. These cells had an initial ASR of  $0.47 \Omega\text{-cm}^2$ . Based on historical data with the standard stack construction (i.e., stack 232), the initial non-cell stack ASR contribution is expected to be about  $0.10 \Omega\text{-cm}^2$ . The non-cell stack ASR is expected to increase by  $0.03 \Omega\text{-cm}^2$  due to the measured decreased conductivity of the low-cost bond-layer relative to the standard Pt-based bond-layer. Therefore the expected stack repeat unit ASR was on the order of  $0.60 \Omega\text{-cm}^2$ .

Two low-cost air-side material stack tests were accomplished; these tests were performed using SOFCo internal funding. For both tests, the cell-to-interconnect connection was accomplished as part of the normal stack build. The first test used the low-cost air-side materials for both the bond-layer and contact ink (bump) and was fired in-situ at  $1000^\circ\text{C}$  for 4 hours before stack operation. The second test used the low-cost air-side material for the bond-layer only (the bump was Pt-based) and did not experience the  $1000^\circ\text{C}$  conditioning. As shown in **Figure 77**, the second test had the expected initial performance ( $0.61 \Omega\text{-cm}^2$ ) and a power degradation rate (5% per 500 hours) that is slightly higher than that typically observed in short stack tests. It is noted that the stack did not experience the initial ASR improvement that has been observed for stacks with Pt-based bond-layers. The initial improvement in those stacks possibly results from triple phase boundary enhancement due to Pt migration. The first test with low-cost air-side bump material had poorer than expected initial performance and degradation. This possibly results from the increased contact resistance of a fractured low-cost bump compared to a fractured Pt-based bump (it is expected that the bumps fracture for both systems). To

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

improve the integrity of the air-side contacts, development of an [ ] air-side connection may be necessary.



**Figure 77. Performance of stacks with low-cost air-side**

## CONCLUSION:

Low-cost air-side contact materials have been developed with sufficiently high conductivity and good chemical compatibility. The low-cost bond layer was successfully demonstrated in combination with the new MEIC cathode on internally developed cells at three levels: single cell tests, repeat unit tests and a short stack test. However, attachment of the ScSZ cell to the 3YSZ interconnect with low-cost materials (i.e., bump ink) has not been successfully accomplished. Acceptable stack testing results were accomplished by continuing to use the Pt-based bumps to connect the low-cost bond-layer and interconnect. In the long term, a low-cost [ ] layer may be a better solution for accomplishing this connection.

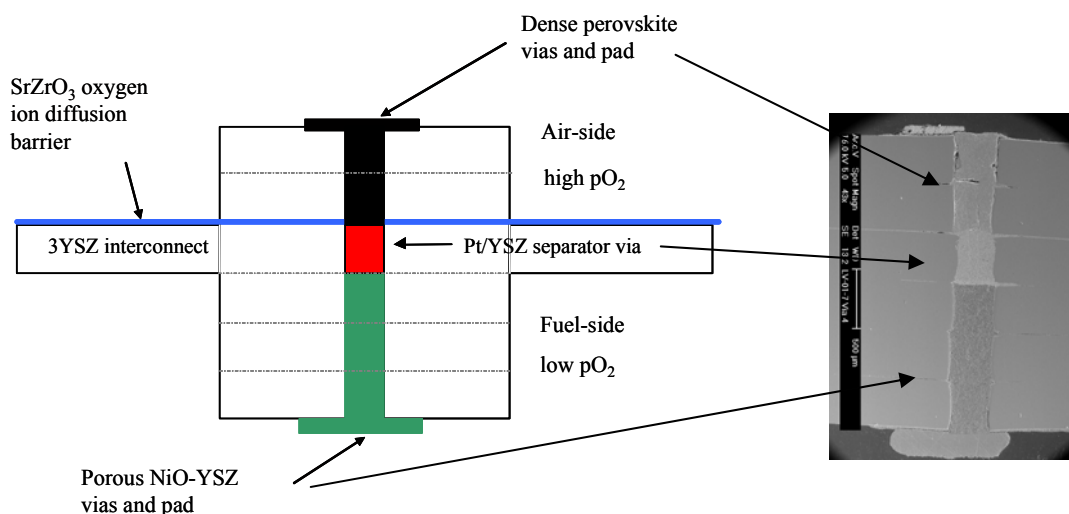
## Subtask 2 – Low-cost Via Materials for Interconnects

**OBJECTIVE AND APPROACH:** The baseline interconnects used throughout the Phase 1 program to demonstrate SOFCo's stack technology have used platinum-YSZ cermet vias. The initial selection of platinum cermet vias was based on the stability of platinum in both reducing and oxidizing environments. Although costly, the use of the platinum cermet allowed for an accelerated development of the manufacturing process for SOFCo's novel multi-layer ceramic interconnect and the successful demonstration of a promising stack technology. A successful SECA Phase 1 program required elimination of the precious metal vias to meet the cost targets. SOFCo focused on developing a design that is based on separate via compositions within the layers making up the air- and fuel-flow fields, and the separator layer, as illustrated in **Figure 78**. Highly conductive perovskite oxide phases have been evaluated for the air-side vias, and nickel-cermet for the fuel-side vias. The separator layer via will continue to contain some platinum, with the target to reduce the Pt content in the SECA Phase 2 program. It should be noted that SOFCo expects to achieve the SECA cost targets with the small amount of platinum retained in the separator layer.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

**EXPERIMENTS:** Substantial laboratory-scale studies of candidate low-cost via materials were performed during Phase I of the SECA program that resulted in the selection of a highly conductive perovskite oxide and a nickel cermet, for the air- and fuel-flow layer vias, respectively. Bulk resistance measurements indicated that these materials can provide for lower cost interconnects with acceptably low ASR contributions. For these low-cost materials to work well within interconnects during stack operation, the air-side perovskite material must be kept at a sufficiently high oxygen partial pressure, and the fuel-side nickel cermet material must be kept at a sufficiently low oxygen partial pressure to prevent the formation of high resistance phases. The efforts during the first two years of the program were aimed at finalizing the selection of materials. Efforts during the final two years were directed at implementing these materials into multi-layer interconnect fabrication, and optimizing the via composition and processing parameters. Substantial effort was required to achieve the desired as-fired microstructures and eliminate the presence of resistive phases at via interfaces. Early tests were conducted using coupons and single interconnects. Subsequently, short stack tests were performed to evaluate performance of the low-cost via materials under representative operating conditions. Toward the end of the Phase I program, a series of short stack tests were conducted to validate that acceptable performance of the low-cost via materials had been achieved.

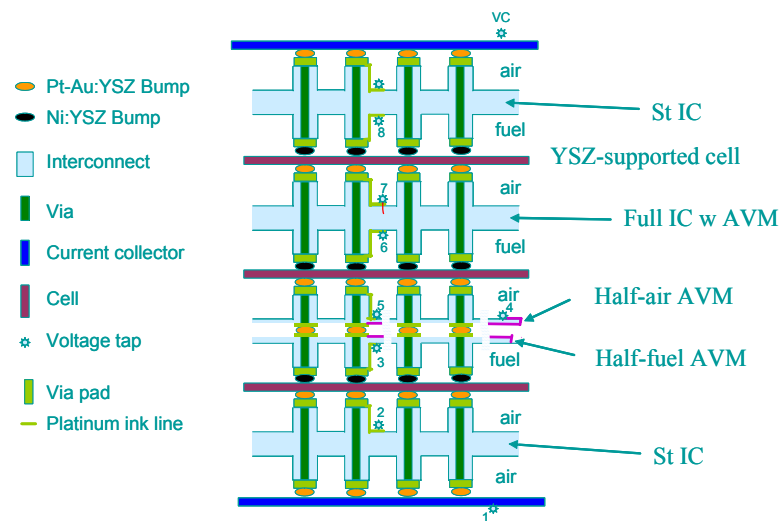
**RESULTS AND DISCUSSION:** During the first two years of the Phase I effort, SOFCo fabricated a variety of test coupons (e.g., single layer structures containing vias), half-air and half-fuel interconnects, and full interconnects. As the work progressed, efforts shifted to fabricating full alternate via material (AVM) interconnects, with 10-30 interconnects being released per month into the SOFCo prototyping line to support the development effort. The resulting interconnects were inspected against the standard interconnect specifications. In the final year of Phase I (i.e., 2005), approximately 50% of the AVM interconnects passed inspection, and were deemed suitable for stack testing. As shown in **Figure 78**, via fill quality was good and defects associated with the vias (e.g., voids, etc.) were generally not observed. This result was encouraging, given the limited quantity of parts being processed.



**Figure 78. Schematic of multi-layer interconnect section showing low-cost vias and SEM micrograph showing a cross-section through a column of vias**

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

In late 2003, SOFCo constructed the first stack containing interconnects with low-cost via materials. Performance of this stack was very poor, as the ASR was  $\sim 100 \Omega\text{-cm}^2$ . Post-test inspection of the interconnects revealed significant delamination between the 3YSZ layers and poor via fill quality. After making changes to the fabrication processes, interconnect quality improved sufficiently to permit assembly and testing of a second stack in 2004. The configuration of this stack (Stack #165) is illustrated in **Figure 79**, along with the associated instrumentation. This stack was constructed using 3YSZ electrolyte-supported cells and contained two standard interconnects, one full AVM interconnect, one half-air interconnect and one half-fuel interconnect. The results for the first 100 hours of testing are provided in **Figure 80**. Overall, the stack ASR started at about  $4.5 \Omega\text{-cm}^2$  and improved to  $<3 \Omega\text{-cm}^2$  by the end of the test period. The ASR of the full AVM interconnect started at about  $7 \Omega\text{-cm}^2$  and improved significantly over the test period, reflecting the slow reduction of the NiO in the fuel-side vias to a conducting Ni phase. The half-fuel interconnect had been pre-reduced prior to stack assembly. As a result, the resistance started low, but was observed to increase with time. The half-air interconnect showed a stable, low resistance during the test period; the resistance was comparable to that measured for standard interconnects. Post-test examinations were performed to understand the source of high resistance for the full interconnect, and the degrading resistance for the half-fuel interconnect. As shown in **Figure 81**, SEM examinations revealed that a highly porous region devoid of Ni existed at the interface between the separator vias (Pt-YSZ) and the fuel layer vias (Ni-cermet). It appears that diffusion of Ni from the fuel-side vias into the Pt was the source of the observed depletion zone.



**Figure 79. Schematic showing arrangement for Stack**

After modifying the via compositions to address the Ni migration issue, additional interconnects were fabricated. With adjustment of the via composition, high via electrical continuity and low resistance was observed in these interconnects. Of the 12 interconnects characterized, 11 showed 100% via continuity and the remaining one had 99.7% continuity (of the more than 400 columns of vias formed in the 6 layers comprising the interconnect). After reduction, the Ni-cermet vias in the half-fuel interconnects had an average resistance of  $0.17 \Omega$  per via, which is similar to that measured for the Pt-YSZ vias.



10 kW SOFC Power System Commercialization  
SECA Semi-Annual Report 41244R08

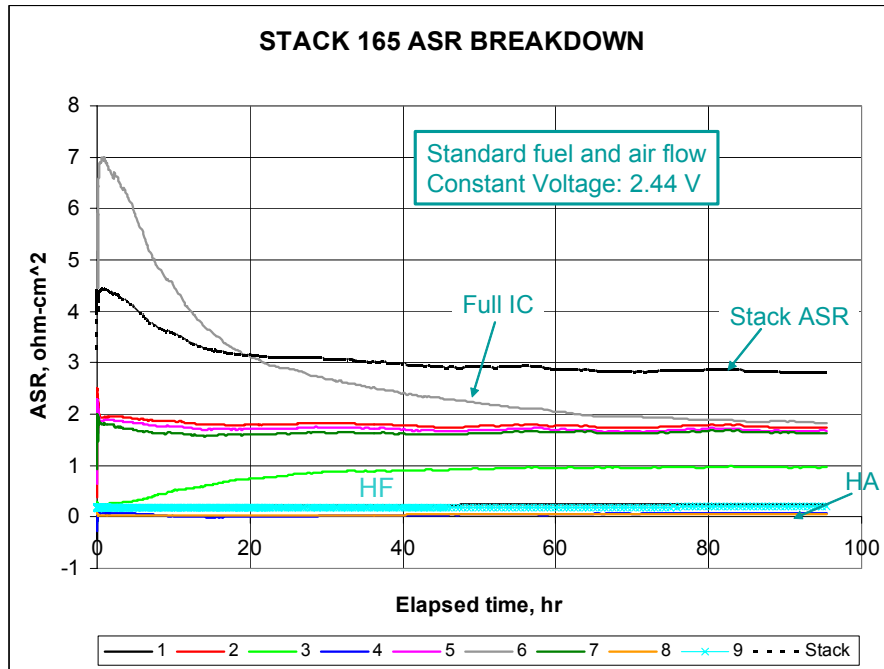


Figure 80. Resistance contributions for AVM interconnects during initial testing period for Stack #165

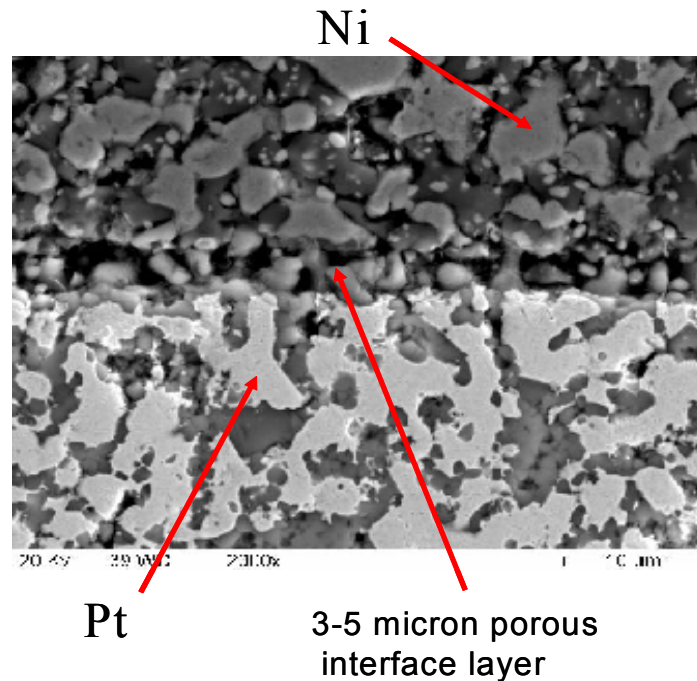


Figure 81. SEM micrograph showing the interface between the separate and fuel-side vias

Subsequently, short stack #211 was constructed using three AVM interconnects (one half-air, one half-fuel, and one full interconnect) and two 3YSZ electrolyte-supported cells. The initial

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

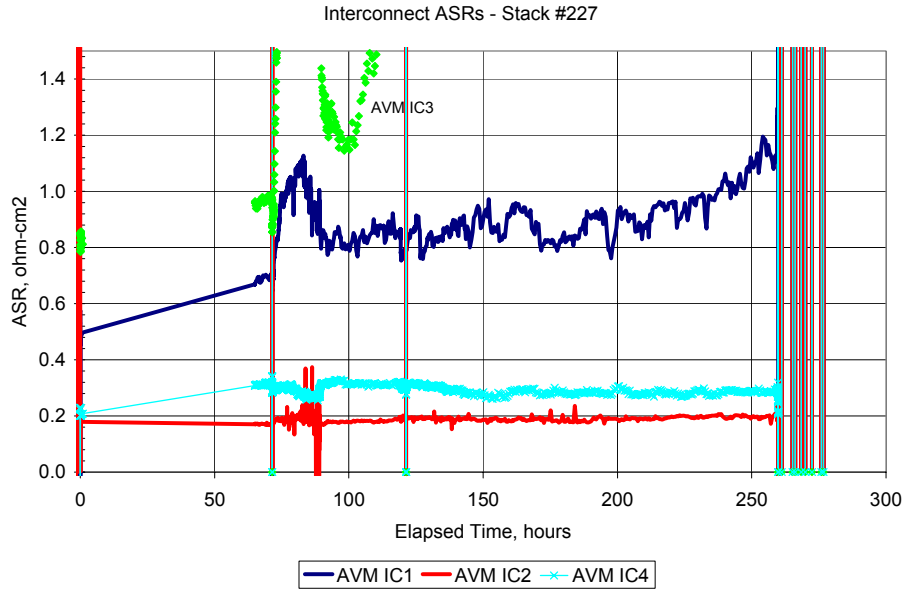
ASR was  $0.06 \Omega\text{-cm}^2$  for the half-air interconnect,  $0.00 \Omega\text{-cm}^2$  for the half-fuel interconnect, and  $0.33 \Omega\text{-cm}^2$  for the full interconnect. For the first 90 hours, the stack was tested under conditions of low fuel utilization and low current density. After 90 hours, the ASR was  $0.03 \Omega\text{-cm}^2$  for the half-air interconnect,  $0.01 \Omega\text{-cm}^2$  for the half-fuel interconnect, and  $0.4 \Omega\text{-cm}^2$  for the full interconnect. The current and fuel utilization of the stack were then increased to  $150 \text{ mA/cm}^2$  and 50%, respectively. The stack was held under these conditions through 500 hours of testing. The full AVM interconnect showed a direct response to stack fuel utilization. After the fuel utilization change, the ASR of the full AVM interconnect dropped to  $0.34 \Omega\text{-cm}^2$  and continued to decrease to  $0.28 \Omega\text{-cm}^2$  at 220 hours, and then slowly increased to  $0.33 \Omega\text{-cm}^2$  again at 500 hours. After 500 hours, the test conditions were changed again to gather more information, and then the stack was shut down. Compared to the first full AVM interconnect tested in 2004 (initial ASR was  $6.8 \Omega\text{-cm}^2$  and decreased to  $1.9 \Omega\text{-cm}^2$  at 500 hrs), the testing results of full AVM interconnect in early 2005 showed significant progress with the low-cost via materials.

The post-test analysis of short stack #211 with low-cost via materials was completed in April 2005. The via resistance for all three interconnects was measured. The results showed that via continuity remained high after testing. The continuity was 99.7% for the half-air interconnect (no change before and after testing), 98% for the half-fuel interconnect (100% before testing), and 95.7% for the full interconnect (no pre-test measurement available because the as-fabricated vias were not conductive). All three interconnects from stack #211 were cross-sectioned and examined by SEM. A new phase was observed at the air-side via/separator via interface in the half-air interconnect; analyses suggested that the phase was probably a mixture of Ni and NiO. A new phase was also observed at the air-side via/separator via interface in the full interconnect. However, EDS analysis indicated that Ni metal was present because oxygen was not detected. This result indicates that the oxygen partial pressure at air-side via /separator via interface in these interconnects may be too low, resulting in possible decomposition of the perovskite oxide at the interface during stack operation.

Additional AVM interconnects were fabricated during Q2 2005, and another short stack test was initiated in June. In stack #227 four full AVM interconnects were tested (number IC1 through IC4). AVM IC2 was from a previous batch of parts, and was similar to the interconnect that was shown to have an ASR of  $\sim 0.3 \Omega\text{-cm}^2$  (from stack #211). Instrumentation in stack #227 allowed for the separate measurement of the ASR for each interconnect, as shown in **Figure 82**. For AVM IC 2, the initial ASR was  $0.18 \Omega\text{-cm}^2$  and ended up at  $0.2 \Omega\text{-cm}^2$  after 260 hours of testing. The performance of the more recent AVM interconnects showed a wide variation, with initial ASR values ranging from  $0.2$  to  $0.8 \Omega\text{-cm}^2$  and higher degradation for IC1 and IC3. Because of the high degradation for two of the interconnects, stack #227 was shut down after  $\sim 260$  hours testing. While these short stack tests showed that the AVM interconnects were approaching the initial ASR target of  $0.2 \Omega\text{-cm}^2$ , the repeatability needed improvement.

Through careful examination of data recorded during AVM interconnect fabrication and subsequent stack assembly and testing, three potential issues were identified. First, the interconnects from the recent lot used a fuel-side via ink made from a new batch of powder. A quick evaluation was performed and confirmed that this ink resulted in higher than expected resistance. Second, stack #227 used a reduction procedure for the fuel-side via materials during testing that may have adversely impacted the resistance of the Ni cermet. Third, one of the as-sintered AVM interconnects was cross-sectioned and examined. Preliminary results show voids in some of the air-side vias, which may be one of the factors affecting interconnect quality and repeatability.

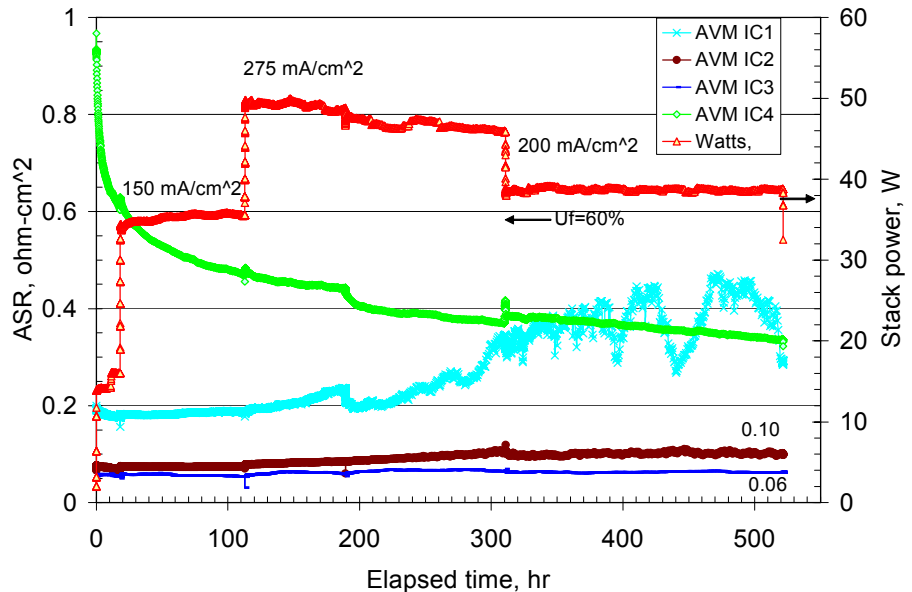
# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 82. Short stack #227 containing four AVM interconnects**

Late in Q2 2005, another batch of AVM interconnects was fabricated using larger diameter vias and slight modifications to the compositions of the fuel-side and separator via materials. Four interconnects from this batch were used to assemble short stack #234 in July 2005; the test was terminated in August after the stack operated for 500 hours. Relevant performance data for the stack is shown in **Figure 83**. The maximum current density for this short stack was 275 mA/cm<sup>2</sup>. Before shut-down, the stack was operated at a 200 mA/cm<sup>2</sup> current density with 60% fuel utilization; the ASR values for the four interconnects were 0.29, 0.10, 0.06, and 0.32 Ω-cm<sup>2</sup> (from bottom to top of the stack) at the end of operation. It was believed that the high ASR for the top AVM

interconnect may have been due to a problem associated with stack assembly. After shut-down, the top interconnect assembly with the current collector was modified and the short stack was retested. The ASR of the top interconnect dropped to 0.17 Ω-cm<sup>2</sup>. Due to the unstable behavior of the bottom interconnect, it is suspected that there may have been an electrical contact

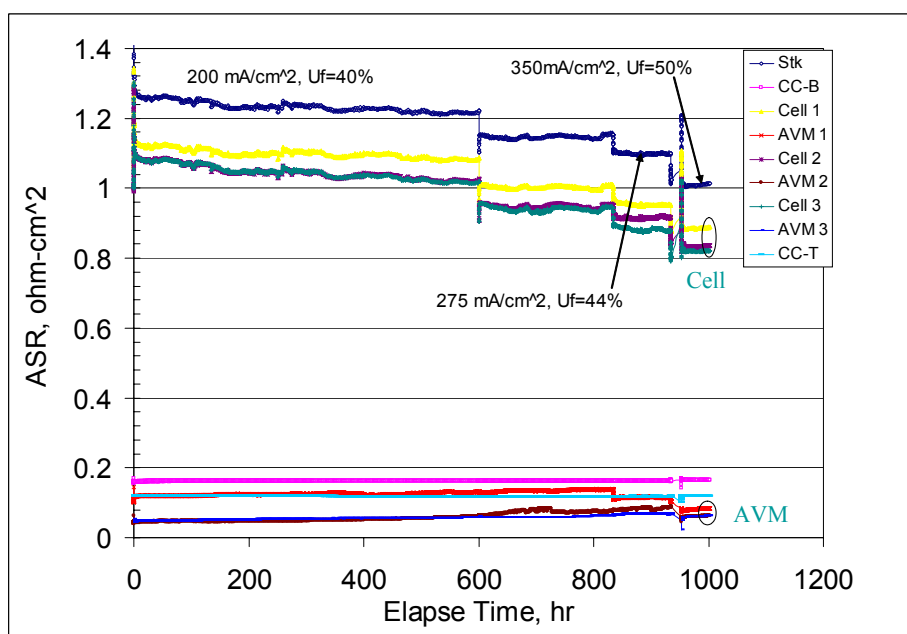


**Figure 83. Short stack #234 containing four AVM interconnects**

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

issue (possibly with the current collector). As a result, a decision was made to modify the connection to the metal current collectors in future stack tests using AVM interconnects.

Toward the end of the Phase I period, two final short stacks were assembled and tested; these tests were performed outside of the SECA program using SOFCo internal funding. ScSZ electrolyte-supported cells were used for both of these stacks, as opposed to 3YSZ cells, to permit testing at higher current densities expected for C2 stacks. Stack #242 was constructed using 4 AVM interconnects that included two slight variations in via composition. The stack was tested for more than 1200 hours and yielded positive results. Two interconnects of one type showed stable ASR values  $<0.1 \Omega\text{-cm}^2$  (the Phase I target) during the test period, while the two interconnects of the second type had ASR values between  $0.15\text{-}0.2 \Omega\text{-cm}^2$ . Stack #243 was constructed using additional AVM interconnects that were similar to the ones showing the best performance in Stack #242. This stack showed very good performance (**Figure 84**) over the 1000 hour testing period, achieving an ASR  $<1 \Omega\text{-cm}^2$  at a current density of  $350 \text{ mA/cm}^2$  and a 50% fuel utilization. The AVM interconnects had an average ASR of  $0.07 \Omega\text{-cm}^2$ . This final test met the Phase I performance targets for the low-cost via materials.



**Figure 84. Performance data for Stack**

**CONCLUSION:** Modified fuel-side and separator-layer via compositions developed during the final year of Phase I resulted in significantly lower ASR contributions and improved stability for the AVM interconnects. In particular, stack #227 was a significant outcome for SOFCo, in that three out of four AVM interconnects in this stack met the June 2005 milestone ( $\text{ASR} < 0.2 \Omega\text{-cm}^2$ ). Moreover, two of the interconnects met the final ASR target ( $\leq 0.1 \Omega\text{-cm}^2$ ) and showed good stability. SOFCo subsequently assembled and tested two additional short stacks that yielded positive performance data. In particular, Stack #243 was operated for 1000 hours and showed stable resistance values; the average ASR for the AVM interconnects at the end of the test was  $0.07 \Omega\text{-cm}^2$ . Based on these results, SOFCo met the Phase I milestone for the low-cost via materials and is prepared to implement these materials into future interconnect production development efforts.

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

### Subtask 3 Alternative Stack Configurations

**OBJECTIVE:** Scale-up interconnects from 10 to 15-cm, demonstrating that it is feasible to increase the interconnect footprint without degrading part quality. Evaluate the impact of alternative stack configurations on stack cost.

**APPROACH:** This task includes design of 15-cm interconnects and evaluation of the impact of alternative aspect ratios on manufacturing cost.

**EXPERIMENTS:** Work on this task started in 2003 and continued through mid 2005. Fabrication trials were performed for 15-cm interconnect using a distributed manufacturing approach for interconnect prototyping, with support provided externally for some operations. The external support was needed due to certain specialized equipment and expertise not yet available at SOFCo. These operations included tape casting, punching, and excising. The balance of the operations was performed by SOFCo, which consisted primarily of via fill, lamination, various firing steps and inspection. Worked aimed at alternative configurations was limited to design studies in 2004 and early 2005. Final interconnect design and component fabrication trials were not performed due to funding limits.

**RESULTS AND DISCUSSION:** SOFCo completed fabrication trials for 15-cm interconnects in late 2004 and demonstrated successful scale-up of the 10-cm design. A decision was made not to proceed with stack assembly and testing in 2005, as it appeared that SOFCo would not continue to pursue the 15-cm design. Rather, SOFCo decided to develop a stack based on an 11-cm x 21-cm footprint. After generating a preliminary design for the new interconnect in Q1 2005, work on footprint scale-up was suspended for budgetary reasons.

**CONCLUSION:** SOFCo successfully demonstrated fabrication of 15-cm interconnects. After making good progress aimed at the next-generation stack design in Q1 2005, work on footprint scale-up was suspended.

### Task 6: Manufacturing Process Development

**OBJECTIVE:** Replace the expensive 3YSZ powder currently used for interconnect fabrication with a low-cost 3YSZ powder.

**APPROACH:** The distributed manufacturing approach continued to be used by SOFCo for interconnect prototyping with support provided externally for some operations. The external support was needed due to certain specialized equipment and expertise not yet available at SOFCo. These operations included tape casting, punching, and excising. The balance of the operations was performed by SOFCo, which consisted primarily of via fill, lamination, various firing steps and inspection. In addition, SOFCo continued to expand its laboratory space and equipment to allow increased throughput of interconnect prototyping. With the expanded capabilities, SOFCo was able to perform fabrication trials for interconnects using low-cost 3YSZ powder.

**EXPERIMENTS:** A small quantity of low-cost 3YSZ powder was acquired in late 2004 and used for fabrication of interconnects. The quantity of powder was too small to produce a statistically significant number of parts for evaluation. However, no major difficulties were encountered in prototyping of this experimental lot up to the lamination step. Additional shipments of low-cost 3YSZ were received in Q2 2005 and fabrication trials were performed.

**RESULTS AND DISCUSSION:** SOFCo continued to contact and evaluate vendors for support of distributed manufacturing operations currently beyond SOFCo's capabilities. As mentioned

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

above, these operations included tape casting, punching, and excising. An issue associated with the distributed manufacturing approach, particularly with SOFCo's low-production needs, is that the vendors did not always perform the services consistently and timely, which often led to quality issues and scheduling conflicts. Therefore, SOFCo continued to search out qualified vendors who have a long-term vision for SOFC applications.

During Q1 2005, SOFCo completed fabrication of a small batch of 15-cm interconnects using tape cast by an outside vendor with the low-cost 3YSZ. A number of these parts met the quality requirements for interconnects, indicating that there were no apparent issues associated with implementation of an alternative 3YSZ powder.

Following the initial fabrication trial, SOFCo ordered a larger batch of low-cost 3YSZ (~200 kg) from the supplier and requested that this powder be produced using their production line, as opposed to their R&D facility. Unfortunately, the transition from the R&D line to production took longer than expected. As a result, SOFCo did not receive the next shipment of powder until late in Q2 2005. Even then, the powder did not meet the SOFCo specification, as the surface area was too high. Although this powder was rejected, a small quantity was retained for evaluation. Tape was cast without difficulty and a small batch of 10-cm interconnects was processed without any significant issues. A second shipment of 3YSZ powder was provided, and although quality of the powder was improved, it still did not meet the surface area specification. As before, SOFCo fabricated a small batch (10 pieces) of 10-cm interconnects; while the shrinkage during co-firing was excessive, no other process issues were identified.

In July 2005, SOFCo received 180 kg of low-cost 3YSZ powder meeting the required specifications. A tape casting run was performed, and this tape was used to process one final batch of interconnects. Interconnect fabrication was completing, with a number of parts meeting the specification. As a result, a decision was made to transition all future interconnect fabrication development activities to the low-cost 3YSZ material.

**CONCLUSION:** Initial fabrication of interconnects using a less expensive 3YSZ powder did not indicate any manufacturing issues that need to be addressed. A small batch of 15-cm interconnects was successfully fabricated using powder produced by the supplier on their R&D line. Subsequently, SOFCo received 3YSZ powder from the supplier from their production line that did not meet specifications. However, fabrication of 10-cm interconnects was completed using this powder without experiencing any problems. SOFCo performed one final fabrication trial using 3YSZ powder which met the quality specification. Successful fabrication of 10-cm interconnects served to confirm that the low-cost 3YSZ powder is suitable for use in the production of SOFCo's multi-layer interconnects.

### **Task 7: Balance of Plant**

#### **Summary Overview**

During this period the low cost BOP system was tested and developed with the C2 complete system. The second half of 2005 began with running the C2 Hot Box with Simulator stacks. Significant testing has been accomplished with the complete BOP and C2 Hot box system. At this time all BOP hardware is functioning as required for system operation. The current configuration has been adapted to match evolving stack and hot box requirements. It is currently capable to provide the required performance from start-up, NOC, Peak power, and shutdown for the SECA phase 1 test. The system has demonstrated stable operation for hundreds of hours at the NOC operating point. System modeling, vibration isolation, and enclosure design has continued during this reporting period. The test cell instrumentation, programming, and calibration are complete for the phase 1 test.

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

However, during development testing a potential failure in the heat exchanger system has been detected in the data. Careful testing indicated that a visual inspection of the recuperator was required. The inspection highlighted an over temperature failure in the recuperator exhaust inlet. This condition has put a hold on further testing.

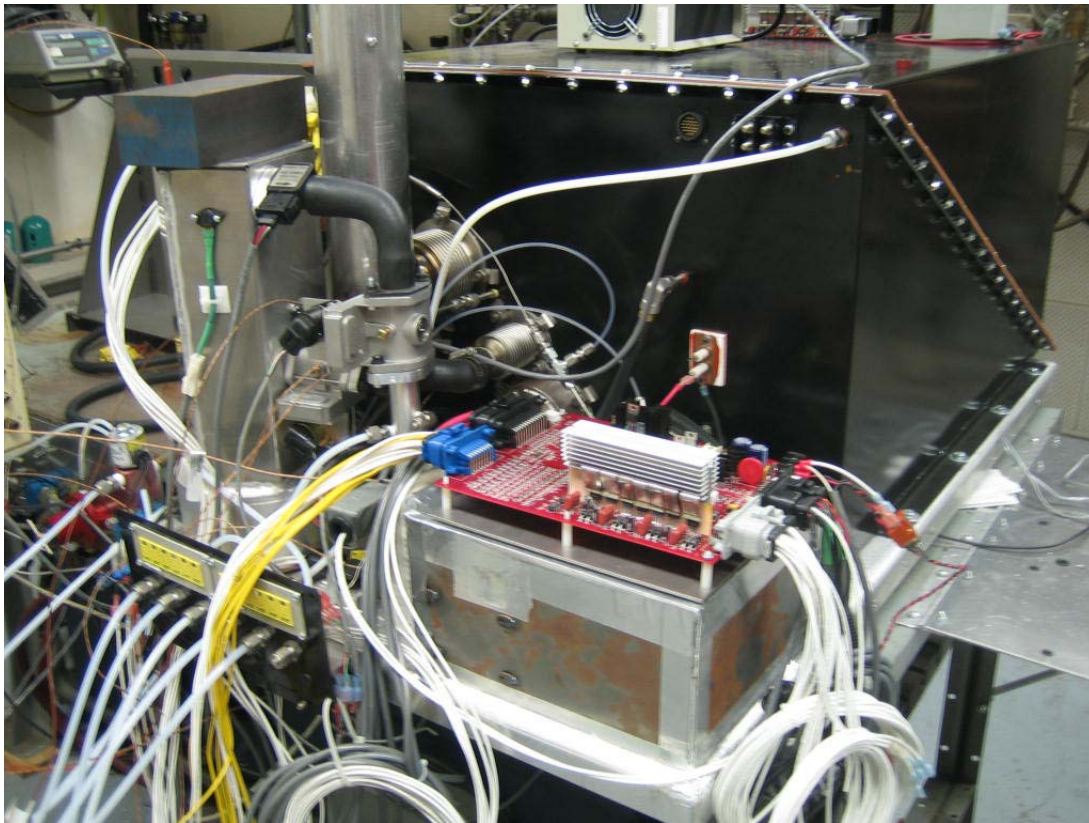
**OBJECTIVE:** Develop and test the air and fuel balance of plant subsystems as a complete C2 operational unit. C2 Development to begin this period.

**APPROACH:** CPG designed, specified, procured, and fabricated the necessary balance of plant hardware for the C2 test unit based on SOFCo ASPEN modeling predictions.

**EXPERIMENTS:** Final verification of the BOP design and hardware was completed this reporting period.

**RESULTS AND DISCUSSION:** Development testing during the period lead to further design changes and implementations. These changes have brought the low cost C2 BOP up to the required performance for the phase 1 test.

**CONCLUSION:** During this reporting the BOP for C2 is operational and capable of performing the required tasks for the SECA phase 1 test. Due to the failure of the recuperator heat exchanger the official phase 1 test has been postponed.



**Figure 85**

**C2\_SIM Development Testing at CPG.**

**C2 Design**

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

**OBJECTIVE:** To identify and develop the BOP, support, and enclosure systems for the C2 test unit. All component, System, and Test setup designs are performed in Pro-Engineer. For communications purposes the Pro-Engineer material is translated into AutoCAD (.dwg) drawings and Adobe (.pdf) file types.

**APPROACH:** The second half of 2005 focused on the development of the BOP for the C2 unit. The main goal was to identify and develop the fuel and air systems portion of the BOP. All component, P&ID, System, and Test setup designs are performed in Pro-Engineer. For communications purposes the Pro-Engineer material is translated into AutoCAD (.dwg) drawings and Adobe (.pdf) file types. The plan was to have all hardware performance verified by the end of July 2005 to begin testing of the C2 hot box with actual stacks. This included fabrication and implementation of the mechanical BOP, instrumentation, data acquisition, and the procurement for the C2\_Stack system.

**EXPERIMENTS:** Verification testing of the new stepper motor actuated butterfly control valves was performed in this reporting period. This included flow range, resolution, and stability demonstrations.



### RESULTS AND DISCUSSION:

#### BOP Development

- 1) Actuator changes to Linear Stepper drives (and CAN) was implemented to actuate butterfly air control valves. Development testing verified the performance of the Low cost Automotive linear stepper motor drives. Over 600 hours of development and system testing has proven these components to provide the required performance. Improved packaging and physical construction improvements are planned to improve performance, cost, durability, and aesthetics.

#### Vibration isolation and Enclosure development continued.

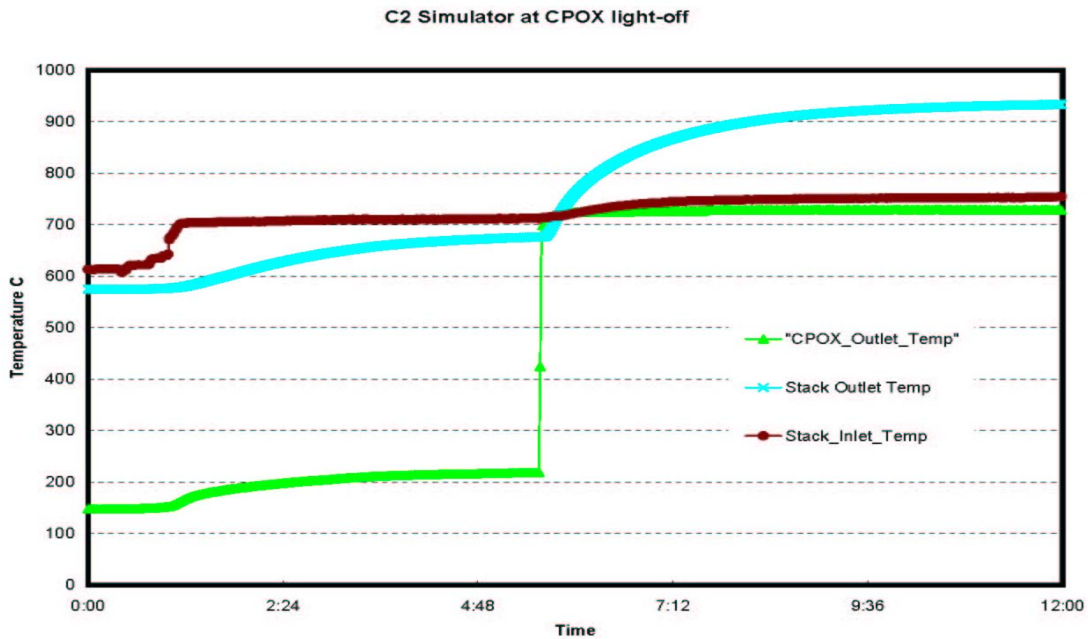
- 1) Vibration isolation hardware has been implemented into the C2 base plate. Future development of the system to manage the expected application inputs is planned.
- 2) The enclosure base has been fabricated and the enclosure panel designs have been completed.

#### Test Cell preparation for Phase I testing.

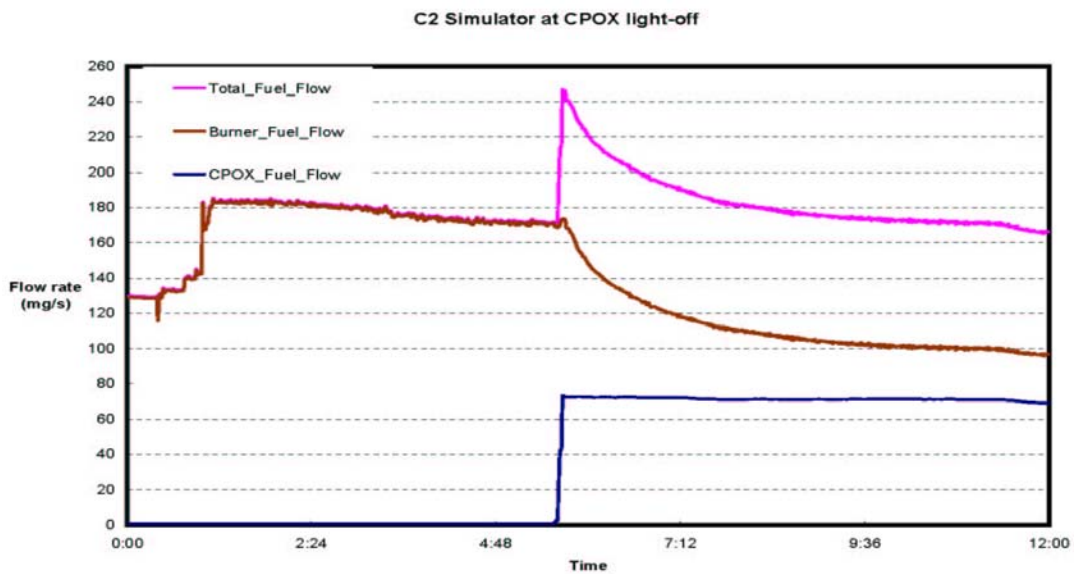
- 1) NG system boost pump implementation. Due to DAS flow meter requirements (30 psi) a natural gas boost pump was plumbed into the test cell. The device takes the 10 in-wg line pressure up to 50 psi. Development testing at the 600 hour mark began to indicate that this device was leaking air into the fuel stream. Investigation into the feasibility of utilizing this boost method is required prior to further usage. Our CNG system is still available to provide high pressure NG for testing.
- 2) Labtech DAS system fabrication and calibration. A complete DAS system has been fabricated during this reporting period. System calibration and programming is completed.
- 3) Web access development for DOE test monitoring. Web access within our corporate system has been challenging. However, we have implemented a beta test web system.
- 4) Labtech system development for acquiring C2 system digital control output variables and measurements. The C2 control software has the additional code implemented to allow a



# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 87 CPOX Light-off**



**Figure 88 C2 System Temperatures**

secondary Labtech DAS system to request and receive control variables and measurements. The system is functional with the current data set. Revisions to the data set are implemented as required. This data is not part of the Web accessible data set.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



## **C2\_SIM Development Testing.**

- 1) The CPOX was operated through various load levels to provide the expected energy released with actual stacks. **Figure 87** shows a light-off sequence and the CPG stable controls.
- 2) The temperatures within the C2 system are shown in **Figure 88**:

## **Task 8: Controls and Power Electronics (CPG) Development**

### **WBS Task 8.1 Control System Design**

#### **8.1.1 Thermal/Fluid Stack Controls**

##### **Objective**

The design and development of the control system that is responsible for the thermal management of the stack and balance of plant system. In addition this control is responsible for the mass flow control of the various fluids through the system and is tasked with insuring the stack receives the proper air and fuel flow to satisfy the load demand. This control is resident in the MCU.

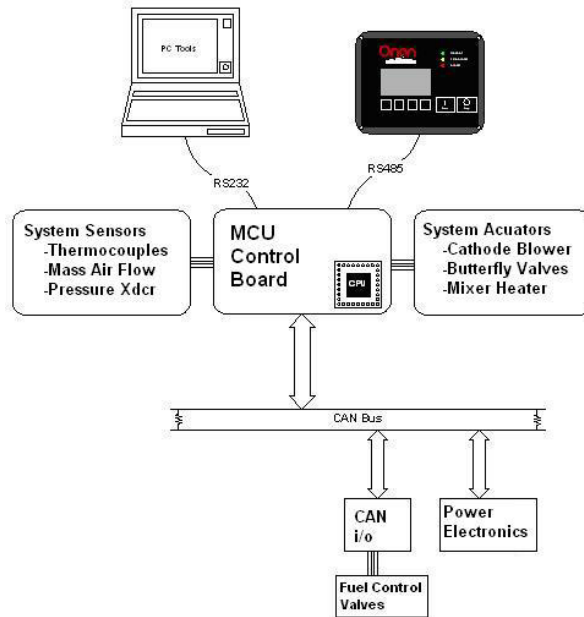
##### **APPROACH:**

The C2 control system hardware consists of three parts; the master control unit (MCU) which contains the control algorithms and processes the analog sensor signals, a distributed network of daughter boards strung along a CAN network, and a low cost LCD based human machine interface (HMI). The MCU also supplies a number of outputs that can be used digitally or pulse width modulated (PWM) for actuator control purposes, each output is rated for 12Vdc and 7.5Amps. The MCU is a completely new design for the C2 system and was completed the end of 2004. The daughter boards connected to the CAN network are used to interface the MCU to the more sophisticated actuators in the system, such as the various flow control valves. The use of the daughter boards and the CAN network allows for a great deal of flexibility in the type of actuators that can be used, and allows these various actuators to be changed while the basic control software remains unchanged.

The C2 control system is a departure from the previous C1 system in that it incorporates a new low cost single board control. The previous C1 system consisted of a more expensive multi-board system that was modified from a commercial Cummins Inc. product. The C2 control is a completely new design dedicated to controlling the fuel cell system.

The fuel cell control software consists of basically three parts; they are the Thermal/Fluid closed loop controls, the control modes or states, and system diagnostics. The Thermal/Fluid closed loop controls are the control loops that are necessary to control the temperatures of the various subsystems, such as the stack inlet air temperature or the inlet exhaust temperature to the recuperator. The control modes correspond to what set points the various subsystems need to be controlled to, in order to satisfy the task at hand, such as; system warm up, hot idle or shut down states. While the diagnostic routines, are those routines that serve a diagnostic and self protection type function.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08



**Figure 89**  
**Control System**  
**Hardware Architecture**

**Controls:** During the second half of 2005 the controls work concentrated on the completion of the integration of the controls with the mechanical hardware. In addition, the control loops required to control stack inlet and outlet temperatures were tuned, tested and verified as completed. At the end of August 2005 the various control loops and control algorithms had been tuned and test. The controls were ready for the phase 1 durability testing to begin in the fall 2005.

The control loops were tuned by performing a series of system ID experiments on the hot system simulator to determine the system dynamics and gains. Simple dynamic models were then fitted to the data gained from these experiments, and the models were used to tune the controls. The control simulation and tuning was done with Simulink modeling software.

In addition to the control system tuning and validation, during the month of July of 2005 software in the MCU to export the control sensor signals to the external data acquisition system was thoroughly tested and verified as complete. This feature was added to the MCU to reduce the number of redundant sensors in the system and was pivotal in the control system tuning.

An overview of the control algorithms for the C2 system are summarized below;

**Stack temperatures:** The stack temperatures are controlled by controlling the Cathode inlet air and outlet temperatures; this also effectively controls the stack average temperature. The stack Cathode inlet air temperature is cooled by regulating the amount of Cathode air flow that is bypassed around the recuperator heat exchanger. In addition the inlet air temperature can be further regulated by controlling the energy transferred across the recuperator by controlling the inlet exhaust temperature to the recuperator. For more detail in how the recuperator inlet temperature is controlled see the section entitled Recuperator exhaust inlet temperature below.

The stack Cathode outlet temperature, and thus stack delta T, is controlled by the amount of Cathode air mass flow that is driven through the system. Increasing the Cathode mass flow rate decreases the Cathode outlet temperature, and vice versa.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

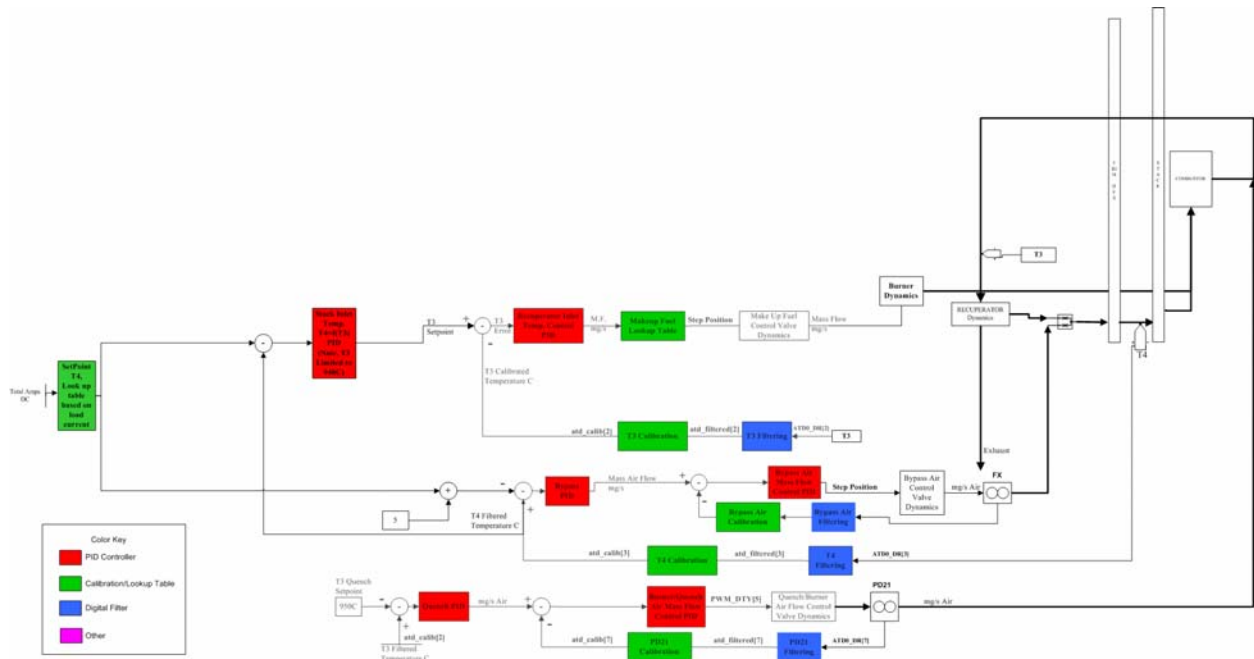
**Cathode Mass Flow;** In the C2 design the Cathode mass air flow is regulated by a closed loop routine that controls the blower speed in concert with an automotive based mass air flow sensor (MAF).

**CPOX outlet temperature;** The CPOX outlet temperature is controlled by a closed loop routine that regulates the CPOX air flow.

**Recuperator exhaust inlet temperature;** In the C2 design the recuperator exhaust inlet temperature is regulated with a feed back loop by controlling the burner air flow and the system makeup fuel. The control over recuperator inlet temperature is a means by which the thermal energy input to the balance of plant can be precisely controlled. Consequently, controlling the recuperator inlet temperature is an integral part of the start up process and is critical in maintaining hot idle conditions. If during stack loaded operation the recuperator inlet temperature needs to be reduced, quench air passed through the startup burner is utilized. If during stack loaded operation the recuperator inlet temperature needs to be raised, make up fuel is added to the exhaust flow.

## RESULTS and DISCUSSION:

During the months of July and August 2005 the control loops necessary for the completion of the phase 1 testing were completed. These loops included the routines necessary for the control of stack inlet and outlet temperatures. The stack inlet temperature control loops included stack inlet control via bypass mass flow control, and stack inlet temperature control via recuperator inlet temperature control. The recuperator inlet temperature control was realized by another inner control loop that controlled the make up fuel flow, and burner/quench air flow. In addition the stack outlet temperature control was implemented via total Cathode mass air flow control. The stack inlet temperature control diagram can be seen below;

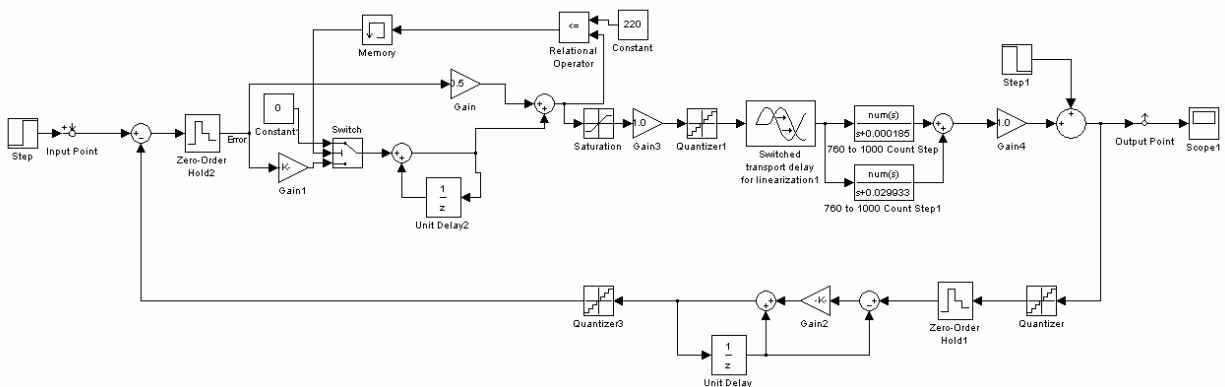


**Figure 90**  
**Stack Inlet Temperature Control Loops.**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

In order to tune the system a series of step command experiments were performed on the various mass flow controls, while the resulting temperature transients were recorded by a data acquisition system. The data gathered was then used to construct a dynamic model of the open loop plant dynamics. This dynamic model in turn was used to simulate the closed loop system within Simulink. The gains derived from this modeling were used directly in the actual control system and the resulting performance matched the simulated results.

For example, to tune the recuperator inlet temperature control loop, an experiment was performed where by a step change was made to the make up mass flow of fuel and the resulting recuperator inlet temperature response was recorded by the data acquisition system. A dynamic system model was fit to this data and the resulting model was used to tune the closed loop control system via a Simulink software model. The closed loop Simulink model can be seen in the following figure;



**Figure 91**  
**Simulink Model of Recuperator Temperature Control**

The model derived in this way was accurate enough such that the gain derived could be used directly in the actual system without further tuning. In addition, other control techniques, such as feed-forward, could be tested before implementation with the actual hardware.

The above tuning example was repeated for each individual control loop, starting with the inner most loops and then working outward to the farthest outer control loop. In addition, these tests were also repeated at a number of operating conditions in order to verify the veracity of the modeling.

## CONCLUSION:

- Completed implementation and tuning of control software for new control system.
- Completed software interface to new hardware.
- A/D control, counter timer control, serial communications etc.
- Implemented new more efficient binary search routine for lookup tables.
- Implemented new improved first order filter algorithm.
- Implemented CAN based control of new stepper motor controls.
- Implement control algorithms.
- Implemented control software for exportation of controls data to the system data acquisition system.

## 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

- Integrated the controls with the actuators and sensor hardware.
- Tuned, tested and completed the control loops required to perform the phase 1 system testing milestone scheduled for fall 2005.

### **Load Controls**

#### **Objective:**

Develop the hardware and software to control fuel cell stack loading, ensure proper fueling depending on electrical load, and supply an interface between the stack controls and the power electronics.

#### **APPROACH:**

The load management is resident in the inverter control DSP. The inverter control DSP acts as the master by controlling the battery boost directly, while the fuel cell controls act as the slave insuring that the stack receives the proper flow of fuel and air to meet the load demand.

#### **RESULTS and DISCUSSION:**

During the first half of 2005 a new CAN interface was built to enable the MCU to control the fuel cell boost. This will allowed the MCU to control the stack loading until the new power electronic system is ready for integration.

#### **CONCLUSION:**

The system interface to the load controls was completed during the second half of 2005, and was ready for phase 1 testing.

Mass flow control device development

#### **Objective:**

Design and development of alternative low cost mass flow controls based on automotive style actuators to replace the current lab type mass flow controls.

#### **APPROACH:**

The approach that has been taken for C2 is to use off the shelf automotive hardware to control and measure the various system mass flows. Each mass flow controller consists of an automotive throttle valve controlled by the MCU via a CAN daughter board interface. The mass flow is measured by a MAF sensor that is connected directly to the MCU and the mass flow is controlled by a PID loop resident in the MCU.

#### **RESULTS and DISCUSSION:**

The flow control valves are controlled by butterfly valves driven by stepper motors. The stepper motor driver boards were designed and built during the first half of 2005. The driver boards contain a small microcontroller that communicates with the MCU, via a CAN bus. The small micro also controls the stepper motor drive transistors, and maintains step count and home position control.

#### **CONCLUSION:**

The new mass flow controller development was completed during the first half of 2005.

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## WBS Task 8.2 Power Electronics Design

### 8.2.1 Fuel Cell Boost Converter

#### Objective:

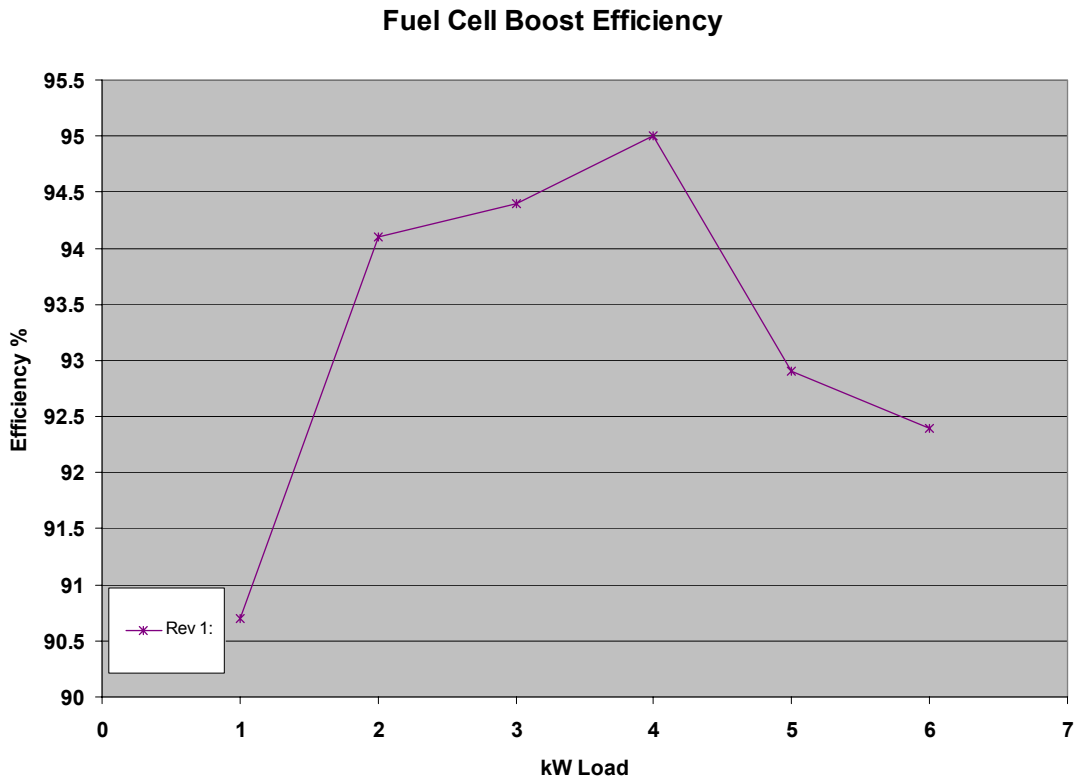
Design and development of a DC-DC boost converter stage for fuel cell output to output inverter conversion. Provides appropriate DC bus voltage for AC inversion from the low voltage fuel cell source, and protects fuel cell from transient loads.

#### **APPROACH:**

The fuel cell boost converter is a low cost, and efficient, inductor based design. The boost is controlled by a cost effective analog control, with external control inputs for enable, soft-start, and current limit signals. These external inputs allow the operation of the boost to be control via a master power management control.

#### **RESULTS and DISCUSSION:**

During the first half of 2005, a lot of work had been done to package the boost into a production prototype that can be economically integrated with the fuel cell system. This was accomplished with only a slight degradation in overall efficiency. The boost efficiency can be seen, in the following diagram, to reach a maximum of 95%.



**Figure 92  
Fuel Cell Boost Efficiency**

In addition, a new CAN daughter board has been designed and built to allow the MCU to control the boost via the system CAN communications bus.

# 10 kW SOFC Power System Commercialization

## SECA Semi-Annual Report 41244R08

Thermal testing under the expected fuel cell load conditions and Cathode mass airflows was performed during the second half of 2005 and there were no issues found. The temperatures of the IGBT module and inductor were within normal operating parameters.

### **CONCLUSION:**

The fuel cell boost has been repackaged to allow system integration and production. In addition a new CAN interface board has been designed and built to allow the boost to be controlled via the CAN communication serial bus. Thermal verification testing was completed and the fuel cell boost was ready for phase 1 fuel cell testing to commence.

### **8.2.2 Battery Boost/Charger/Inverter**

#### **Objective:**

Design and development of bi-directional battery boost-charger and inverter, to provide load modulation and load peaking for fuel cell and battery state-of-charge maintenance as well as DC to AC conversion for external loads.

#### **APPROACH:**

These components are presently under development at CPG as part of an on going development project for a new variable speed diesel generator set. The components design for this generator will be modified for use with the fuel cell system.

#### **RESULTS and DISCUSSION:**

At this time the power electronic system development at CPG has been delayed, as a backup in case the power electronic system is not available for use in the phase 1 endurance testing, a system has been constructed to allow the fuel cell to startup off of an AC source and to invert the fuel cells DC voltage into an AC source for the purposes of electrical loading. This system was built during the first half of 2005 and is available for phase 1 testing this fall.

#### **RESULTS and DISCUSSION:**

An AC inverter will be available for phase 1 system testing.

#### **CONCLUSION:**

The back up system built to allow the fuel cell to startup off of an AC source and to invert the fuel cells DC voltage into an AC source for the purposes of electrical loading will be utilized for the phase 1 system test.



10 kW SOFC Power System Commercialization  
SECA Semi-Annual Report 41244R08

**References**

**Not applicable**

**BIBLIOGRAPHY**

**Not applicable**

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## LIST OF ACRONYMS AND ABBREVIATIONS

10cmxN – 10cm by yet to be determined measurement  
2-D – Two dimensional  
3-D – Three dimensional  
3D Solid Model - Commercially available software  
A/F – Air/Fuel  
ABAQUS – Commercially available software  
AC – Alternating Current  
AC – Alternating current  
AMPS –  
Amps – Unit of current  
Arms – Amps root mean square  
ASC – Anode-supported Cell  
ASPEN – Commercially available software  
ASR – Area Specific Resistance  
ASTM- American Society for Test and Materials  
ATR – AutoThermal Reforming  
AutoCAD – Commercially available software  
BOP – Balance of Plant  
C – Celsius  
C1 – Phase 1 Development Prototype  
C2 – Final Phase 2 deliverable Prototype  
CAN – Controller Area Network (ISO11898)  
cBOP – cold Balance of Plant  
CFD – Computational Fluid Dynamics  
cm – centimeter  
Cm<sup>2</sup> – squared centimeters  
CO – Carbon Monoxide  
CPG – Cummins Power Generation  
CPOX – Catalytic Partial Oxidation Catalyst  
CTE – Coefficient of Thermal Expansion  
dB(A) – Decibels A weighted  
DC – Direct Current  
DOE – U.S. Department of Energy  
ESC – Electrolyte-supported Cell  
EZ-Thermal™ – Commercially available software  
FEA – Finite Element Analysis

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

## LIST OF ACRONYMS AND ABBREVIATIONS (cont'd)

FLUENT – Commercially available software  
FMEA – Failure Mode Effects Analysis  
Gate A, B, C – Levels of managerial evaluations  
GHSV – Gas Hourly Space Velocity  
hBOP – hot Balance of Plant  
HD-5 – California Air Resources Board Emission Certification Propane Fuel  
hrs – Hours  
HX – Heat Exchanger  
Hz – Hertz  
I/O – Input/Output  
ID – Inside Diameter  
K – degrees Kelvin  
kg/hr – kilogram per hour  
kPa – Kilo Pascal  
kW – kilowatt  
kWe – kilowatt electrical  
L – Liter  
lbs/hr – pounds per hour  
LPG – Liquid Propane Gas  
m – meters  
M/A-COM – Division of Tyco Electronics, Subcontractor to MTI  
Mathworks - Commercially available software  
MLC – Multi-Layer Ceramics  
msec – millisecond  
MTI – McDermott Technology, Inc.  
mxn – to be determined cell footprint dimension  
NETL – National Energy Technology Lab  
NG – Natural Gas  
Ohm – unit of resistance  
P&ID – Process and Instrumentation Drawing  
PCU – Power Cell Unit  
PF – Post Fired  
PNNL – Pacific Northwestern National Lab  
POC – Proof-of-Concept  
POX – Partial oxidation  
Ppm – parts per million

# 10 kW SOFC Power System Commercialization SECA Semi-Annual Report 41244R08

Ppmv – parts per million volume

## **LIST OF ACRONYMS AND ABBREVIATIONS (cont'd)**

PPT – Product Preceding Technology

ProEngineer - Commercially available software 3-dimensional design modeler

Q1..Q4 – quarters of calendar year

QD – Quiet Diesel (Cummins-Onan TM product)

S/C – Steam to Carbon

SECA – Solid State Energy Conversion Alliance

SEM – Scanning Electron Microscope

SI – LeSystemme International d'Unites

SMAF – Small Mass Air Flow (Sensor)

### ***SOFC – Solid Oxide Fuel Cell***

SOFCo – Solid Oxide Fuel Cell Company

SR – Steam Reforming

US – microseconds

V – Volts

VAC – Volts Alternating Current

VDC – Volts Direct Current

V-I – Voltage - Current

VPI -- Value Package Introduction, a Cummins proprietary process

WBS – Work Breakdown Structure

YSZ – Yttrium Stabilized Zirconia