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**MONOLITHIC SOLID OXIDE FUEL CELL
TECHNOLOGY ADVANCEMENT FOR
COAL-BASED POWER GENERATION**

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SECTION 1
INTRODUCTION

1. INTRODUCTION

This report describes the work performed under Contract DE-AC21-89MC26006, for the U.S. Department of Energy (DOE)/Morgantown Energy Technology Center (METC) program. The report covers the period October through December 1991.

The program is conducted by a team consisting of AiResearch Los Angeles Division of Allied-Signal Aerospace Company and Argonne National Laboratory (ANL). The objective of the program is to advance materials and fabrication methodologies to develop a monolithic solid oxide fuel cell (MSOFC) system capable of meeting performance, life, and cost goals for coal-based power generation. The program focuses on materials research and development, fabrication process development, cell/stack performance testing and characterization, cost and system analysis, and quality development.

SECTION 2

BACKGROUND ON MONOLITHIC SOLID OXIDE FUEL CELLS

2. BACKGROUND ON MONOLITHIC SOLID OXIDE FUEL CELLS

The monolithic solid oxide fuel cell (MSOFC) is a recent design concept in fuel cell technology. The MSOFC is made up of all solid components, permitting fabrication into unique shapes unachievable in fuel cell systems having liquid electrolytes. The MSOFC design places the thin ceramic components of the fuel cell in a lightweight, compact structure of very small cells. The monolithic design eliminates inactive structural support, increases active surface area, and lowers voltage losses caused by internal resistance. This results in a MSOFC with high efficiency, excellent performance, and power per unit mass or volume unequaled in any other solid oxide fuel cell configuration.

2.1 DESIGN CONCEPT

In a MSOFC, fuel and oxidant produce a dc current by combining electrochemically across the solid oxide electrolyte at an operating temperature of 1000°C. Oxygen in the oxidant fed to the cathode accepts electrons from the external circuit to form oxide ions that are then conducted through the dense electrolyte to the anode. At the anode, oxide ions combine with hydrogen in the fuel to form water, liberating electrons as shown in Figure 2-1. Cells of the MSOFC are connected by an interconnect to build voltage. The interconnect carries electrons from the anode of one cell to the cathode of the next in electrical series.

Figure 2-1. Solid Oxide Fuel Cell Chemistry

There are two versions of the MSOFC, coflow and crossflow. In the coflow version, the monolithic fuel cell consists of a honeycomb-like array of adjacent fuel and oxidant channels that resemble corrugated paperboard (Figure 2-2). Oxidant channels are lined with a porous cathode, and fuel channels are lined with a porous anode. A gastight layer of electrolyte is located between the anode and cathode. An interconnect, which is also gastight, connects the cells in electrical series.

Figure 2-2. Coflow Monolithic Solid Oxide Fuel Cell

The typical thickness of the layers is 25 to 100 μm , and the distance from cell to cell is 1 to 2 mm. As shown in Figure 2-2, the MSOFC is made of two types of laminated structures, each composed of three ceramics—anode/electrolyte/cathode and anode/interconnect/cathode. The laminated structure containing the electrolyte is appropriately corrugated. The corrugations form the gas seals at the edges of the monolithic structure (which is extremely important because the gas seal has been a stumbling block in solid oxide fuel cell planar design).

In the crossflow version, the fuel and oxidant channels are formed by corrugated anode and cathode layers and are oriented at right angles to each other (Figure 2-3). The anode/electrolyte/cathode and anode/interconnect/cathode composites are flat and stacked alternately between the corrugated anode and cathode layers. The crossflow version results in the power density being reduced by a factor of about 2, compared with that of the coflow version; however, the crossflow version offers a simpler means of ducting gases in and out of the monolithic fuel cell.

2.2 DESIGN FEATURES

Small cells of lightweight self-supporting ceramics are the key to the MSOFC potential. The small size of individual cells in the monolithic structure increases the active surface area. For example, a coflow MSOFC with channels about 1 mm in diameter has a ratio of active surface area to volume of about $9.4 \text{ cm}^2/\text{cm}^3$ (Figure 2-4). This is about seven times the ratio for tubular solid oxide fuel cells of 1 cm in diameter.

More importantly, the small cell size in the MSOFC reduces voltage losses due to internal electrical resistance. Because current is carried in-plane by the electrodes in both conventional and monolithic designs, a decrease in the cell size decreases the

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Figure 2-3. Crossflow Monolithic Solid Oxide Fuel Cell

Figure 2-4. Monolithic Configuration, Providing a Maximum Active Surface Area per Unit of Volume

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internal electrical resistance. In MSOFC's, the major source of internal voltage loss is the resistance that electrons encounter in the cell ceramic components. Inside the operating fuel cell, an oxidation reaction (the electrochemical reaction of hydrogen fuel) releases electrons on the anode side of the barrier that separates oxygen channels from fuel channels. Flowing along the anode, electrons travel around the fuel-channel wall and across the interconnection to the cathode of the adjoining oxygen channel (Figure 2-2).

The electrons then flow along the cathode around the oxygen channel to the cathode/electrolyte interface, where they are consumed in a reaction (with oxygen) that produces an oxide ion. The oxide ion is transported through the plane of the electrolyte, reacts with hydrogen on the fuel side of the electrolyte to produce electrons, and the process continues. In this way, the electron current traverses the multicell monolithic structure. Because individual cells in the monolithic fuel cell are small, electron/ion paths are short and resistive losses are low. The lower resistance of the monolithic fuel cell improves cell efficiency by about 10 percent. As a result of lower internal resistance, the MSOFC's can be operated at higher current densities than conventional fuel cells while achieving the same output voltage.

The power density for the MSOFC is higher than that of conventional solid oxide fuel cells and other electrical power-generating technologies. The high power density results from the higher surface area, higher current density, and lower weight. The lower weight in the monolithic design is primarily the result of eliminating inert supports, which is possible because the thin ceramic layers are self-supporting. Because of the shorter current paths involved in the monolithic design, electrodes can be thinner, which reduces weight further.

Because the monolithic design employs thin electrodes, diffusion losses are minimized. The cell voltage is determined by the pressures of the reactants and products at the electrode/electrolyte interface. Thus, any reduction in the pressure at the reaction site represents a loss in voltage, and therefore efficiency. A loss in voltage due to reduced pressure is given by

$$[RT/nf] \ln (P_{\text{eff}}/P_{\text{gas}})$$

where P_{eff} is the pressure at the interface

P_{gas} is the pressure of the bulk gas stream

P_{eff} is dependent on the diffusion of reactants and products through the electrodes. In tubular designs, the thick support tube (30 percent porosity) and thick electrodes represent large diffusional barriers. The monolithic design minimizes diffusion losses by eliminating the thick support tube and employing thin electrodes.

In addition to gas diffusion limitations, a potential barrier exists to charge transport across phase boundaries between the electrodes and the electrolyte. This is associated with the electrode reactions. The voltage loss due to this barrier is the activation potential. At the operating temperature of the MSOFC, reaction rates are very

rapid, so it is not necessary to provide a very thick electrode to ensure sufficient active surface area.

Practical cell operation is always strongly exothermic. Cooling is required in order to maintain a desired cell operating temperature. The MSOFC can be cooled by providing excess oxidant beyond that required by the cell reaction. The temperature rise of this stream will carry off heat. The MSOFC can also be cooled by heat-absorbing reactions within the cell. If the fuel is a relatively complex molecule, it can be broken down or reformed in the cell. These reactions are endothermic and can contribute substantially to cell cooling. Examples are the steam reforming of methane and other hydrocarbons such as those found in natural gas, coal gas, and those produced in the decomposition of methanol.

Fabrication of the MSOFC requires incorporation of four thin cell components into the monolithic structure. The MSOFC is fabricated by forming thin ceramic components as green (unfired) layers, incorporating them into the monolithic structure, and cofiring the structure at elevated temperatures to form the required sintered body. Since the fabrication of the fuel cell requires only a single high-temperature processing step, the MSOFC has potential for low-cost manufacturing.

Table 2-1 summarizes the key features and the benefits offered by the MSOFC design.

TABLE 2-1	
FEATURES AND BENEFITS OF MONOLITHIC DESIGN	
Feature	Benefit
Small cell size	<ul style="list-style-type: none">• Increased active surface area• Low internal resistance• Improved efficiency
Corrugated structure	<ul style="list-style-type: none">• No high-temperature seals• Robust structure
Self-supporting structure	<ul style="list-style-type: none">• Lightweight• Compactness• High power and energy densities
Single high-temperature processing step	<ul style="list-style-type: none">• Potential low-cost manufacturing

SECTION 3
PROGRAM OVERVIEW

3. PROGRAM OVERVIEW

Figure 3-1 is a work breakdown structure (WBS) for the program, showing six major tasks and 25 subtasks. Solid line boxes indicate tasks in which significant results have been achieved. Dashed line boxes indicate tasks that have been started, but for which no reportable results are available. Unboxed tasks are those that have not yet been started.

The following paragraphs summarize the progress made on various tasks during this quarter.

3.1 STUDY OF INTERCONNECT/CATHODE INTERACTION (TASK 2.2)

An interconnect formulation ($\text{La}_{0.8} \text{Ca}_{0.22} \text{Dr}_{0.9} \text{Co}_{0.1} \text{O}_3$) has been identified which densifies when fired alone, but does not densify when fired in a composite tape. Two approaches are being explored for minimizing the interaction between the interconnect and electrodes. The first approach is to fabricate a two-material cell comprised of only electrolyte and interconnect with no LaMnO_3 ; this allows for more severe conditions to be used to densify the interconnect. In the second approach, the interaction between the interconnect and electrodes is being studied in an attempt to discover possible corrective measures that can be taken using the normal four-material approach.

3.2 STUDY OF LONG-TERM CATHODE STABILITY (TASK 2.3)

Efforts this quarter were expended in several areas. The performance of CE/E/CE composites was studied to determine their electrochemical characteristics. The conductivity of three Ca and Co doped LaCrO_3 samples was determined. Three doped LaCrO_3 /YSZ/doped LaCrO_3 symmetric composites (referred to as I/E/I) were studied to evaluate the possibility of making a two-material MSOFC.

3.3 FABRICATION DEMONSTRATION (TASK 3.1)

Primary activities of the MSOFC fabrication effort during the quarter focused on batch modifications, fabrication of single-cell modules and stacks, bonding studies, and evaluation of interconnect preparation procedures.

Electrolyte (E), anode (A), and cathode/electrolyte (CE) batch formulations were modified by replacing portions of the zirconia with alumina. The objective was to toughen the fuel cell components in order to decrease the magnitude and frequency of defects in fabricated samples. The addition of alumina did not affect the phases present in the electrolyte and anode. In the CE, the lanthanum zirconate phase (seen in the baseline formulation) was suppressed by the formation of a manganese aluminate phase. The addition of alumina caused some differences in anode sintering behavior, whereas little change in total shrinkage occurred in the modified CE.

Different modified batches were incorporated into the fabrication of MSOFC single cells and single-cell stacks. Experiments were carried out to find the best combination of modified layers and baseline layers when structural integrity of fabricated samples

MSOFC TECHNOLOGY ADVANCEMENT FOR COAL-BASED POWER GENERATION

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Figure 3-1. Work Breakdown Structure for MSOFC Technology Advancement for Coal-Based Power Generation Program

was considered. Visual and optical microscopic examination indicated that a combination of baseline A, baseline E, and modified CE gave the best results.

Significant progress was made in the fabrication of modules with electrode cover and the development of a creep-sintering technique. Electrode-covered modules were successfully processed with minimal observable defects. The bowing of fabricated samples during firing was minimized by using high solid loading anode for anode corrugations and covers. The creep-sintering technique was developed to fabricate single-cell stacks with improved flatness. The technique involved varying constraint weight during processing: relatively low weight used for the low temperature binder extraction and high weight for the high temperature sintering regime. The use of this technique has yielded flat modules, electrode-covered modules, and single-cell stacks suitable for the second step bonding.

Extensive characterization was performed on bonded two-cell stacks. The results indicated no damage or cracking of the interconnect or bonded samples during the bonding step. Bonded stacks showed excellent strength (sectioned without delaminating) and thermal cyclability (3 cycles to 900° to 1000°C). The anode and anode/interconnect had excellent bonding contact, whereas the cathode/interconnect needed improvement. Modifications of bonding materials and procedures resulted in improved cathode/interconnect bonding contact.

Characteristics of interconnect powders prepared under different conditions were evaluated. The results indicated that the sintered microstructure of the interconnect could be improved by breaking down the agglomerates in the as-received powder into their individual particles.

3.4 TESTING ON COAL GAS (TASK 4)

Six single-cell disks were tested this quarter. Four were tested to attempt improvements in the current collector attachment. One was to experiment with a new possible removable seal, and another was to obtain baseline data for tri-layers made with the new Al₂O₃-toughened CE layer, discussed in para. 5.1.2.3.

One single-cell stack and two two-cell stacks were tested this quarter. The single-cell stack showed very good performance and was held under an endurance test for nearly 200 hr.

3.5 STACK PERFORMANCE MODELING (TASK 5.1)

The mathematical model of the fuel cell is complete. The model simulated the electrochemical, electrical, thermodynamic, and chemical equilibrium processes occurring in the cell.

3.6 EVALUATION OF NDE METHODS (TASK 6.1)

This subtask is essentially complete. A draft copy of the final topical report is currently being reviewed.

3.7 QUALITY CONTROL OF SOURCE MATERIALS (TASK 6.3)

SSC, Inc. is determining the exact stoichiometry of a previously received lot of lanthanum chromite powders, which sintered to 99-percent theoretical density.

SECTION 4

MATERIALS RESEARCH AND DEVELOPMENT (TASK 2)

4. MATERIALS RESEARCH AND DEVELOPMENT (TASK 2)

4.1 STUDY OF CATHODE/INTERCONNECT INTERACTION -TRI-LAYERS (TASK 2)

4.1.1 Objective

Densification of the interconnect under relatively moderate firing conditions (1350°C in air) remains the main challenge in fabricating the MSOFC. While the interconnect ($\text{La}_{0.8}\text{Ca}_{0.22}\text{Cr}_{0.9}\text{Co}_{0.1}\text{O}_3$) densifies to high density (90 to 95 percent) when fired alone, it does not densify when fired in contact with electrodes; interaction with the electrodes apparently inhibits its densification. Two approaches are being explored for obtaining a dense interconnect. One approach is to fabricate a two-material cell comprised of only electrolyte and interconnect materials. Because such a cell contains no LaMnO_3 , more severe conditions (higher temperature or more reducing gas atmosphere) can be used to densify the interconnect. The second approach is to manage the interaction better between interconnect and electrodes to obtain high density in the usual four-material cell.

4.1.2 Results

4.1.2.1 Two-Material MSOFC

In the two-material MSOFC, the cathode and anode of cathode-electrolyte-anode composites are made from interconnect material; these layers must remain porous during firing. At the same time, the interconnect and electrolyte layers must densify nearly completely. In addition, strong bonds must be formed between the electrolyte and electrodes and between the interconnect and electrodes. The electrolyte composites made using interconnect material ($\text{La}_{0.8}\text{Ca}_{0.22}\text{Cr}_{0.9}\text{Co}_{0.1}\text{O}_3$) as the electrodes had both dense electrolyte and porous electrodes. However, the bonding between the electrolyte and electrodes was very weak, as seen by the fact that the electrodes detached from the electrolyte in several cases.

To improve bonding between the electrodes and electrolyte, electrolyte composites were tape cast with electrodes that consisted of 10-, 20-, or 30-wt% partially stabilized zirconia (6 mol% Y_2O_3) mixed with the interconnect material. All composites were fired for 1/2 hr at 1400°C in air. Bonding in the 10- and 20-percent composites was weak, and the electrodes detached from the electrolyte when samples were broken for microscopic examination. Bonding was stronger in the 30-percent composite, and the electrodes did not detach. Figure 4-1 focuses on one of the electrolyte/electrode boundaries in the 30-percent composite, and shows that the electrolyte is dense while the electrode is porous, as desired. However, the boundary between electrolyte and electrode is sharp, suggesting that the bonding might be weak and that it could be improved further. To improve bonding, more zirconia was added to the electrodes, in the amount of 35- and 40-wt% partially stabilized zirconia. Figure 4-2 shows that distinct boundaries still exist between the electrolyte and electrodes of the 40-percent composite, suggesting that more zirconia might be necessary. However, addition of too much zirconia might degrade the electrochemical performance of the composites, so more

Figure 4-1. Interface of the 30-Percent Composite Showing Dense Electrolyte and Porous Electrode

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Figure 4-2. 40-Percent Composite with Dense Electrolyte and Porous Electrode

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zirconia will be added to the electrodes only if the electrochemical performance of the 30-, 35-, and 40-percent composites proves to be satisfactory.

There must also be a strong bond between the electrodes and the interconnect. To test the bonding between interconnect and electrodes, five-layer composites (LC/E/Zy/E/LC) were tape cast, where LC is 100 percent $\text{La}_{0.8}\text{Ca}_{0.22}\text{Cr}_{0.9}\text{Co}_{0.1}\text{O}_3$, E is 65-wt% $\text{La}_{0.8}\text{Ca}_{0.22}\text{Cr}_{0.9}\text{Co}_{0.1}\text{O}_3$ /35-wt% TZ-3Y, and Zy is 70-wt% TZ-8Y/30-wt% TZ-3Y. These composites were fired at 1400°C in air for 1/2 hr. All composites were intact after firing, suggesting that bonding is sufficiently strong to proceed with fabrication of a two-cell stack. Microscopy of these composites is being done to examine interconnect/electrode interface and the density of the interconnect.

4.1.2.2 Four-Material MSOFC

It is thought that the interconnect does not completely densify in the usual four-material MSOFC, because the liquid that aids sintering migrates into the electrodes before densification is complete. To improve densification, an attempt is being made to increase the amount of liquid phase present during sintering, so that migration of some liquid does not significantly impact densification. In the past, attempts to increase the amount of liquid phase included the addition of CaCrO_4 , which melts at $\sim 1200^\circ\text{C}$ according to the phase diagram. Prof. Harlan Anderson reported that CaCrO_4 is not useful for this purpose, however, because it forms Cr_2O_3 , which coats the grain boundaries and inhibits densification. Prof. Anderson also reported that compositions closer to the 1022°C eutectic would be more useful for this purpose. To prevent migration of the liquid phase before sintering can proceed, the liquid phase should not form until the sintering temperature has been reached ($\sim 1200^\circ\text{C}$). According to the phase diagram, the "941" composition exhibits a local maximum in melting point (1288°C in oxygen), and may be ideal. For this reason, the "941" composition has been prepared for use as a sintering aid.

4.1.2.3 Future Work

If the electrochemical performances of the two-material anode-electrolyte-cathode composites prove to be satisfactory, small ($\sim 5 \times 5 \text{ cm}^2$) two-cell stacks will be made, consisting of the following repeating unit: three-layer electrolyte composite/interconnect corrugation/flat interconnect. If fabrication is successful, the performance of the cells will be measured. If problems exist with debonding, compositions of the various layers will be adjusted (e.g., more zirconia could be added to the electrodes if they detach from the electrolyte).

The "941" $\text{CaO-Cr}_2\text{O}_3$ composition will be tested as a sintering aid in interconnect composites (anode/interconnect/cathode).

The use of a thick (200 to 500 μ) interconnect layer will be tested. If the interconnect is thick enough, part of it should densify despite interactions with the electrodes.

4.2 STUDY OF SINGLE LAYERS

The densification of perovskite lanthanum chromites has been extensively studied. Recently, Mori¹ and others have fully densified calcium-doped lanthanum chromites at relatively low temperatures (1200° to 1400°C) by adding excess calcium. Their powder processing involved calcining above 1000°C followed by grinding and reheating steps to obtain a single-phase solid solution of $\text{La}_{1-x}\text{Ca}_x\text{CrO}_3$ with $0.2 \leq x \leq 0.4$.

This study is focused on lower temperature powder preparation (800°C) and sintering (1050 to 1300°C) of $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$. (The powders intentionally are slightly chromium-rich to avoid the possible appearance of excess lanthanum compounds which degrade the mechanical integrity of the sintered component; this is also found to affect sinterability.) At calcine temperatures of 800°C or less, CaCrO_4 is found to coexist with $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$ for $x \geq 0.2$. However, CaCrO_4 is soluble at higher temperatures and will dissolve into the matrix when sintered above its solubility limit. Since CaCrO_4 melts incongruently, its use as a liquid-phase sintering aid is restricted. However, the Ca-Cr-O system includes other compositions that may react and melt with the CaCrO_4 . In particular, the eutectic composition, $\text{Ca}_{1.09}\text{Cr}_{.91}\text{O}_{4.8}$ and a calcium-rich ternary compound, $\text{Ca}_3(\text{CrO}_4)_2$ are being investigated. Attempts will be made to use these compositions to densify calcium-doped lanthanum chromites.

4.2.1 Experimental Procedure

4.2.1.1 Powder Processing

All powders were prepared using the Argonne-modified Pechini method. Metal nitrates or carbonates were dissolved into water and nitric acid and then mixed with an ethylene glycol/citric acid mixture having a mole ratio of 1:1:1. The solution was boiled to drive off water and nitrates and allowed to gel. The gel was then put in a drying oven at ~120°C. As the gel dried, it foamed and charred. The char was milled and reheated to about 400°C to remove residual carbon. The resulting powders were then milled and calcined to various temperatures as needed.

4.2.1.2 Milling Effects on Agglomerate Size

One of the drawbacks of using the technique described above is that large agglomerates are formed which cause poor packing and ultimately large pores in the sintered product. To improve this, the effect of milling time on particle size is presently being studied. Both lanthanum chromate and lanthanum chromite powders will be analyzed using the Horiba for particle size distribution and the scanning electron microscope (SEM) for studies of their microstructure.

4.2.1.3 Quench Study

$\text{La}_{.59}\text{Ca}_{.4}\text{CrO}_4$ powder prepared at 400°C and containing CaCrO_4 was isostatically pressed into 1.2-cm pellets at 210 MPa. Green densities approached 50 percent of the calculated theoretical density. The pellets were heated at a rate of 8°C/min and quenched in water from the temperatures: 490°, 1075°, 1175°, 1230°, 1275°, 1350°.

and 1400°C. One pellet was held at 1400°C for 2 hr and then furnace-cooled, and another was held at 1400°C for 3 min and then air-quenched.

Microanalysis and energy dispersive analysis were performed on fractured surfaces using a JEOL scanning electron microscope (SEM).

Similar quench runs and SEM analyses will be performed in the future on compositions having optimum sintering behavior. These specimens will also be candidates for future microprobe experiments.

4.2.1.4 Thermal Analysis

Thermogravimetric and differential thermal analyses (TG/DTA) were performed on $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_4$ $0.1 \leq x \leq 0.4$, CaCrO_4 and $\text{Ca}_{1.09}\text{Cr}_{0.91}\text{O}_{4.8}$ powders using a Netzsch TGA/DTA system. The furnace heating rate for each run was 10°C/min and peak temperatures were 1200° and 1600°C. Both alumina and platinum-type crucibles were used.

Future TG/DTA experiments are projected for $\text{Ca}_3(\text{CrO}_4)_2$ and $\text{Ca-Cr-O/La}_\mu\text{Ca}_\nu\text{CrO}_3$ mixtures to determine melting points and other reaction temperatures.

4.2.1.5 X-Ray Diffraction Study

Calcined powders were analyzed for phases present using X-ray diffraction on either a Sintag or a General Electric diffractometer.

X-ray densities were calculated from d-spacing measurements on the Sintag. The Sintag was calibrated in the relevant d-spacing range using a silicon wafer.

The solubility of CaCrO_4 in $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$ was also studied by X-ray diffraction using the Sintag. Disk specimens for $x = 0.1, 0.2$, and 0.3 were sintered for 2 hr at temperatures of 1100°, 1200°, 1300°, 1400°, and 1500°C. More work is being done to determine the solubility limits. In addition, solubility data for compositions ranging from $x = 0.35$ to 0.60 are presently being accumulated.

An additional experiment was performed on powder and disk specimens of $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$, $x = 0.3$ and 0.4 to see if CaCrO_4 would segregate at temperatures at which the solubility limit of Ca in LaCrO_3 was exceeded. The disks and powders were heated to 970°C for 4 hr and analyzed by X-ray diffraction to ensure the presence of CaCrO_4 . Powder and pellets were then heated to 1500°C for 2 hr to dissolve the CaCrO_4 into the matrix and again analyzed for the phases present. Finally, the specimens were annealed at 850°C for 100 hr and analyzed for phases present.

4.2.1.6 Depth Profiles on $\text{La}_{1-x}\text{Ca}_x\text{CrO}_3$

Auger electron spectroscopy (AES) depth analysis was conducted on compacted powders and sintered pellets of $\text{La}_{.89}\text{Ca}_{.1}\text{CrO}_3$. The powders were uniaxially pressed to 70 MPa after calcination at 800°C for 8 hr. The sintered pellets were prepared by isostatically pressing the powders to 210 MPa after calcination at 400°C. Sintering was carried out at 1500°C for 2 hr.

The AES instrument had a sputtering area of about 10 mm² and electron beam area of about 3 mm². Sensitivities for each element were back-calculated from bulk measurements. As a consequence of analyzing rough surfaces, only a qualitative estimate of the sputter depths could be obtained. This was acceptable as a first approximation, however, a microprobe technique that is able to analyze a smaller area must be used in future experiments to clearly define the depth profile of an individual particle.

4.2.1.7 La_{0.69}Cr_{0.3}O₃ / Ca-Cr-O Reaction Study

Powders of the compositions CaCrO₄, Ca_{1.09}Cr_{0.91}O_{4-δ}, Ca₃Cr₂O₈ and CaCO₃ were prepared as above. The calcination temperatures varied between 400° and 800°C to avoid melting. As a result, the Ca₃Cr₂O₈ composition consisted of a CaO-CaCrO₄ mixture rather than the pure Ca₃(CrO₄)₂ phase. Hence, the notation Ca₃Cr₂O₈ is used. The effect of using this as opposed to single-phase Ca₃(CrO₄)₂ will be shown later.

To observe the melting characteristics of the four Ca-Cr-O compositions, 1.2-cm-dia disks were pressed from each composition and placed on 2.5-cm-dia disks of La_{0.69}Ca_{0.3}CrO₃ and La_{0.675}Ca_{0.3}CrO₃. The reaction couples were then heated to 1100°C and in a separate run to 1230°C, held for 2 hr and furnace-cooled. Later, 1.2-cm disks of Ca_{1.09}Cr_{0.91}O_{4-δ} and Ca₃Cr₂O₈ were placed on 2.5-cm disks of CaCO₃, heated to 1230°C and furnace-cooled.

4.2.1.8 Non-Stoichiometric Mixtures

A non-stoichiometric compound, La_{0.675}Ca_{0.3}CrO₃, was prepared using the modified Pechini technique described above. It was calcined at 800°C for 8 hr and mixed with each of the powders: CaCO₃, Ca₃Cr₂O₈ and Ca_{1.09}Cr_{0.91}O_{4-δ} in reagent grade methyl alcohol. After milling, the powders were dried, and isostatically pressed into 1.2-cm disks at 210 MPa. Two disks of each composition were then sintered at 1020°C for 2 hr and at 1400°C for 1 hr. The following reactions were used to estimate the stoichiometry of the final products:

4.2.2 Results and Discussion

4.2.2.1 Phase Relations

Two phases appear when lanthanum chromite powders (x > 20 percent Ca) made from the Pechini method are calcined at 800°C for 8 hr. Calcium chromate (CaCrO₄) and a lanthanum calcium chromate solid solution (La_{0.99-x}Ca_xCrO₄) first appear at the burnout of carbon and crystallization of the char above 400°C. Thermal analysis shows the transformation of lanthanum chromate to lanthanum chromite

between 700° and 800°C is apparent from the large endothermic peak shown in Figure 4-3. This peak shifts to lower temperatures as the Ca content increases. The DTA also shows that a liquid phase forms in all the $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$ composition at approximately 1030° to 1080°C.

By comparing Figure 4-3 and the $\text{CaO-Cr}_2\text{O}_3$ phase diagram², shown in Figure 4-4, it became evident that the liquid phase formed at ~1030°C in the $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3/\text{CaCrO}_4$ mixture was actually the eutectic associated with the CaCrO_4 phase. However, CaCrO_4 melts incongruently forming a eutectic liquid, $\text{Ca}_{1.09}\text{Cr}_{.91}\text{O}_{4-\delta}$ and a higher melting point solid, CaCr_2O_4 . This indicated that CaCrO_4 is ineffective as a sintering aid for $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$.

4.2.2.2 Reaction Mechanism

Using this information, and the results from Auger spectroscopy, X-ray diffraction, and from SEM analysis, a reaction mechanism is proposed which explains the sintering behavior up to and including the solubility temperature of CaCrO_4 in these powders. Figure 4-5 illustrates the mechanism.

When the powders are calcined below the solubility limit, CaCrO_4 appears in X-ray patterns of $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$. However, at this stage, SEM micrographs do not seem to show the presence of distinct CaCrO_4 crystallites. This might be further studied with back-scatter techniques in subsequent SEM analyses.

The Auger results suggest the formation of a calcium-rich layer upon initial sputtering of the powders (Figure 4-6 and 4-7). This gives evidence of the CaCrO_4 coating proposed in the first step of the mechanism. At ~1030°C, incongruent melting occurs. The liquid formed is very reactive towards the excess chromium in the $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$ as is later discussed in the reaction studies section. It is tentatively believed that calcium in the liquid at the coating/particle interface is absorbed, leaving behind a composition which acts as a barrier to lanthanum transport.

Particle rearrangement also takes place in this temperature range which suggests that there is liquid present at particle contact points.

Although not shown in the model, it becomes apparent that the Ca-Cr-O phase also begins to segregate from the $\text{La}_{.99-x}\text{Ca}_x\text{CrO}_3$ (into large masses), as shown in the SEM photomicrograph of Figure 4-8, for a quenched $\text{La}_{.59}\text{Ca}_{.4}\text{CrO}_3$ specimen. The specimen was heated at 1400°C for 3 min and was air-quenched. It appears that the dendritic CaCrO_4 structure has segregated from the $\text{La}_{.59+\delta}\text{Ca}_{.4-\delta}\text{CrO}_3$ matrix. Elemental analysis using EDS shows no measurable amount of lanthanum within the CaCrO_4 . Mixtures of CaCrO_4 and lanthanum carbonate are presently being made to further study the solubility of lanthanum in CaCrO_4 .

At higher temperatures (now being determined for each composition) the CaCrO_4 completely dissolves into the matrix. This became evident when all the compositions containing CaCrO_4 second phase (up to 40-percent calcium) were X-rayed after sintering at 1500°C. In addition, the CaCrO_4 was observed to reappear after the pellets had been annealed at 800°C for 100 hr, suggesting that these reactions are reversible.

Figure 4-3. DTA Runs of $\text{La}_{.79}\text{Ca}_{.2}\text{CrO}_3$ (LCC-20), $\text{La}_{.69}\text{Ca}_{.3}\text{CrO}_3$ (LCC-30) and $\text{La}_{.59}\text{Ca}_{.4}\text{CrO}_3$ (LCC-40) in Pt. Heating Ramp Rate $10^\circ\text{C}/\text{min}$

Figure 4-4. Equilibrium Phase Diagram for the CaO-Cr₂O₃ System.
After Ford and White²

Particles after calcining at
800 C are coated with CaCrO_4 .

Figure 4-5. Proposed Sintering Mechanism for The $\text{La}_{0.99-x}\text{Ca}_x\text{CrO}_3$ System

Figure 4-6. Auger Analysis of Pressed Pellet of $\text{La}_{0.69}\text{Ca}_{0.30}\text{CrO}_3$

Figure 4-6. Auger Analysis of Pressed Pellet of $\text{La}_{0.69}\text{Ca}_{0.30}\text{CrO}_3$

$\text{La}_{0.69}\text{Ca}_{0.3}\text{CrO}_3$ Sintered 1400 C / 2 hr

Figure 4-7. Auger Analysis of Sintered Surface of $\text{La}_{0.69}\text{Ca}_{0.3}\text{CrO}_3$

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Figure 4-8. A SEM Micrograph of $\text{La}_{0.59}\text{Ca}_{0.4}\text{CrO}_3$ Air-Quenched from 1400°C

4.2.2.3 Reaction Studies

The reaction studies, although providing only qualitative results, give more insight to the reactions of the Ca-Cr-O compositions with $\text{La}_u\text{Ca}_v\text{CrO}_3$ or other substrates. It was observed that when a CaCrO_4 disk was heated on either alumina or platinum substrates, to temperatures as high as 1500°C , a liquid exsolved out and spread over the substrate leaving a porous skeleton of the disk. However, at 1030°C , $\text{Ca}_{1.09}\text{Cr}_{0.91}\text{O}_{4.8}$ melted completely in a platinum TG/DTA crucible but not in alumina. Interactions between Ca-Cr-O compositions and various substrates were then studied.

The first experiment consisted of 1.2-cm disks of CaCrO_4 , $\text{Ca}_{1.09}\text{Cr}_{0.91}\text{O}_{4.8}$, $\text{Ca}_3\text{Cr}_2\text{O}_8$ and CaCO_3 , on 2.5-cm disks of $\text{La}_{0.69}\text{Ca}_{0.3}\text{CrO}_3$. After heating at 1100°C for 2 hr, the CaCrO_4 retained its size and shape, but had lost liquid into the $\text{La}_{0.69}\text{Ca}_{0.3}\text{CrO}_3$ and had the characteristic porous, skeletal appearance. The $\text{Ca}_{1.09}\text{Cr}_{0.91}\text{O}_{4.8}$ had contracted, deformed, and liquified at the interface with the substrate. The $\text{Ca}_3\text{Cr}_2\text{O}_8$ remained unchanged in size and shape but changed from a bright yellow

(CaO + CaCrO₄) to a dark green color (perhaps formation of Ca₃(CrO₄)₂.) No evidence of melting could be seen, however the CaCO₃ pellet transformed into unsintered CaO.

The main contrast between the 1100°C and the 1230°C run is that at 1230°C, the Ca_{1.09}Cr_{0.91}O_{4-δ} had completely melted into the substrate and the Ca₃Cr₂O₈ had clearly melted, contracted, and densified areas of the La_{0.69}Ca_{0.3}CrO₃ substrate beneath it.

When the same Ca-Cr-O pellets were set on La_{0.675}Ca_{0.3}CrO₃ substrates and heated to 1230°C for 2 hr, the Ca_{1.09}Cr_{0.91}O_{4-δ} and the Ca₃Cr₂O₈ experienced poor melting. Although this experiment needs to be repeated, it seems to indicate that the Ca-Cr-O compositions have a greater difficulty melting in the presence of excess chromium. As in the case of the alumina setters, the calcium in the system may be leached out preferentially, leaving an unreacted solid.

To prove this, Ca_{1.09}Cr_{0.91}O_{4-δ} and Ca₃Cr₂O₈ pellets were set on CaCO₃ disks and heated them to 1230°C. At about 1030°C, the Ca_{1.09}Cr_{0.91}O_{4-δ} began to melt and spread over the surface of the substrate forming a clear sessile drop. The Ca₃Cr₂O₈ also began to melt near 1230°C. It soaked into the CaO substrate and the pellet on top quit melting only after the liquid had soaked the zirconia setter beneath the CaO. Apparently, the liquid phase is very reactive to solids which will readily dissolve calcium ions such as, alumina, zirconia, and chromium excess lanthanum chromite.

4.2.2.4 Non-Stoichiometric Mixtures

It was believed that by mixing a calcium-rich, congruent melting powder of Ca-Cr-O with chromium excess La_uCa_vCrO₃ (u + v < 1) reaction liquid phase sintering will occur. The composition of the mixture would be such that it ultimately formed a stoichiometric product.

Compositions based on the three chemical equations (1), (2), and (3) were prepared. The eutectic composition Ca_{1.09}Cr_{0.91}O_{4-δ} and Ca₃(CrO₄)₂ qualified as congruent melting liquid phases. Calcium carbonate served as a control end member. Disks of the mixtures from equations (1) and (2) were first heated to 1020°C for 2 hr and then heated to 1400°C and annealed for 1 hr. An identical heating schedule was used later for mixtures of equation (3).

The mixture from equation (2) sintered to 90 percent of full density whereas the other compositions showed little improvement over the conventional compositions. SEM micrographs of the fractured specimen are shown in Figures 4-9 and 4-10. Figure 4-9 shows well-sintered grains that have grown as large as 5 microns. Figure 4-10 is a back-scatter photo that shows the chemical homogeneity. Although the calculated reaction results in a lanthanum-rich product, the ceramic appears to be chemically homogeneous. Presently, the chemical stability is being studied by heating the pellet in a humid environment. To date, the pellet has shown no evidence of degradation. X-ray diffraction will be performed on the specimen following the humidity tests.

Figure 4-9. SEM Micrograph of Sintered $\text{La}_{0.618}\text{Ca}_{0.4}\text{CrO}_3$ (Calculated Composition). Heated at 1020°C/2 Hr and 1400°C/ Hr

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Figure 4-10. Sintered $\text{La}_{0.618}\text{Ca}_{0.4}\text{CrO}_3$. SEM Elemental Back-Scatter Image

4.2.3 Conclusions

According to German³, the solubility of the liquid phase in the solid must be low and the solubility of the solid in the liquid must be high for optimum liquid phase sintering. Many of our results show just the opposite. This may be a result of exceeding the optimum heating ranges. Continuing work will center on determining the solubility limits of CaCrO_4 in LaCrO_3 and La in CaCrO_4 . From this work, optimum densification of $\text{La}_{1-x}\text{Ca}_x\text{CrO}_3$ may be identified by determining the temperature region of low liquid and high solid solubility for a given composition.

4.2.4 References

1. M. Mori, N. Sakai, T. Kawada, H. Yokokawa, M. Dokiya, "Low-Temperature Air-Sinterable Lanthanum Calcium Chromite with Chromium Deficit for SOFC Separator", *Denki Kagaku*, 58 (1990) 528.
2. W. Ford, J. White, *Trans. Bri. Cer. Soc.*, 48, 417, (1949).
3. R. German, Liquid Phase Sintering, Plenum Press, N. Y., p. 182, (1985).

4.3 LONG-TERM CATHODE STABILITY

The objectives of this subtask are to determine the effect of long-term operation of the MSOFC on the cathode performance and to develop a unified theory of interfacial and bulk phenomena in the fuel cell. Densification, manganese migration, and other physical and chemical changes of the cathode may be a problem in long-term fuel cell operation. Determining and understanding the mechanisms of interfacial reactions and degradations will allow the cathode to be modified to improve its overall performance.

Efforts this quarter were expended in several areas. The performance of two CE/E/CE composites was examined to determine their electrochemical characteristics and to obtain data useful for the modeling effort. The work completed at Argonne in the electrochemical modeling effort was transferred to Professor L. Lobban at the University of Oklahoma. The conductivity of three Ca and Co doped LaCrO_3 samples was determined. Three doped LaCrO_3 /YSZ/doped LaCrO_3 symmetric composites (referred to as I/E/I) were studied to evaluate the possibility of making a two-material MSOFC.

The interfacial resistance, determined by ac impedance, of four CE/E/CE composites at 1000°C as a function of oxygen partial pressure is shown in Figure 4-11. The results from the ED033 composites were included in the previous report and are given here for comparison. In general, the two 1:5:1 composites performed much better than the 1:1:1 composites. Assuming all composites were fabricated similarly with the only significant difference being in the relative thickness of the layers, then the 1:1:1 composites with the thicker cathode layers should have performed better. Since the reverse is true, one could conclude that the electrochemical properties of the cathode layers are quite different. An indication of this was seen when one of the cathodes in each of the two 1:1:1 composites delaminated during dc polarization studies on the composites.

Figure 4-11. Interfacial Resistance of CE/E/CE Composites at 1000°C as a Function of Oxygen Partial Pressure (Circles are 1:5:1 Composites and Squares are 1:1:1 Composites)

The interfacial resistance of the CE/E/CE composites can be compared to that of composite 11930 which was fabricated earlier than the ED composites and studied in this quarter. The interfacial resistance as a function of oxygen partial pressure of 11930 and two other 1193 composites studied previously is given in Figure 4-12. All three composites performed similarly and much better than the ED composites studied above.

In a continuing effort to obtain data useful to the electrochemical modeling effort, dc polarization studies were conducted on the CE/E/CE with the results given in Figure 4-13. The oxygen and air curves clearly show two reaction mechanisms that were postulated and discussed in a previous report. One of these mechanisms is dominant at low-current densities and the other at high-current densities. Also, the transition region between the two mechanisms is dependent on the partial pressure of oxygen. It is interesting to note that the air curve and the lowest oxygen partial pressure curve show the onset of a limiting current.

The work completed at Argonne on the electrochemical model of the solid-oxide fuel cell was transferred to the University of Oklahoma. The model has been developed to the point of requiring a considerable amount of parametric studies and reaction mechanism variation. It was felt that the program would be better served by transferring the modeling work because of the type of effort involved in these tasks and amount of time required to perform these tasks. Argonne is continuing to work with Professor Lobban and his graduate student to complete the development of the model.

The conductivity of three Ca and Co doped LaCrO_3 samples was measured. A comparison of their conductivity in oxidizing (air) and reducing ($\text{H}_2/\text{H}_2\text{O}$) environments at 1000°C is given in Table 4-1. It can be seen that their conductivity is more than adequate for the MSOFC. The temperature dependence of the conductivity of the three samples in air is given in Figure 4-14. The stability of these samples in reducing environments was only partially examined: One of the samples (04291-1) was left in a reducing environment for 24 hr without any significant loss of conductivity. It should be noted that its conductivity in air changed from 43.9 to $37.8 \Omega^{-1}\text{cm}^{-1}$ after the reducing cycle.

TABLE 4-1		
CONDUCTIVITY OF Ca AND Co DOPED LaCrO_3 INTERCONNECT SAMPLES AT 1000°C		
Sample	Air Atmosphere $\text{H}_2/\text{H}_2\text{O}$ Atmosphere Conductivity in $\text{ohm}^{-1}\text{cm}^{-1}$	
04291-1	37.8	2.65
03121-1	57.7	0.476
09171-1	46.8	2.33

Figure 4-12. Interfacial Resistance of CE/E/CE Composites at 1000°C as a Function of Oxygen Partial Pressure

Figure 4-13. Polarization Curves of CE/E/CE Composites 11930 at 1000°C and the Indicated Oxygen Partial Pressure in Atmospheres

Figure 4-14. Conductivity of Ca and Co doped LaCrO₃ Interconnect Samples in Air as a Function of Temperature

Ac impedance was used to examine the electrochemical performance of three I/E/I composites in reducing ($\text{H}_2/\text{H}_2\text{O}$) and oxidizing (air) environments. The results of these studies are given in Table 4-2. The first two samples tested were fabricated similarly with no YSZ in the electrode layers. The performance of these composites with air on both sides was somewhat on the high side, but in general acceptable. In contrast, the composite's performance in a reducing environment was very poor. Since the conductivity of the LaCrO_3 drops off precipitously in reducing environments, a small increase in the bulk resistance was to be expected, but the dramatic increase in both the bulk and interfacial resistances in reducing environments was surprising.

TABLE 4-2						
BULK AND INTERFACIAL AREA SPECIFIC RESISTANCE OF I/E/I COMPOSITES AT 1000°C (CALCULATED VALUES GIVEN IN PARENTHESES)						
Sample	Air Atmosphere		$\text{H}_2/\text{H}_2\text{O}$ Atmosphere (all values in ohm-cm ²)		Air & $\text{H}_2/\text{H}_2\text{O}$	
	Interfacial	Bulk	Interfacial	Bulk	Interfacial	Bulk
1st I/E/I (no ZrO_2)	0.256	0.274	3.10	0.875	3.49 (3.36)	0.515 (0.575)
2nd I/E/I (no ZrO_2)	0.184	0.484	2.91	1.10	3.58 (3.09)	0.801 (0.792)
IEI-8 (35 wt%)	0.109	1.17	1.40	2.79	1.63 (1.51)	2.11 (1.98)

The electrochemical performance of the I/E/I composites with an oxidant on one side and a fuel on the other can be predicted from the single atmosphere results. This is shown in the last two columns of Table 4-2 with the calculated values given in parentheses below the measured values. Note that the interfacial values are shown as the total (sum of cathode and anode interfacial effects) values for the oxygen partial pressure gradient work and are individual electrode values for the single atmosphere work.

I/E/I composites were fabricated with considerable amounts of YSZ in the electrode layers in an effort to improve their electrochemical performance and bonding of electrode layers to the electrolyte layer. Besides improving the physical bond between the electrode and electrolyte layers, YSZ was added to spread out the three-phase area and increase the effective electrochemically active area. A composite with 35 wt% YSZ in the electrodes (IEI-8) was tested and the results are given in Table 4-2. As expected, there was a dramatic decrease in interfacial resistance of the composite in both reducing and oxidizing environments. Unfortunately, there was also a significant increase in the bulk resistance of the composite. Some increase in the bulk resistance was expected, but the size of the increase was much higher than it should have been.

Work on the I/E/I composites will continue with electrodes containing between 30 and 40 wt% YSZ. Post-test analyses of composite IEI-8 will be conducted to look at the electrode microstructure and for the presence of significant quantities of $\text{La}_2\text{Zr}_2\text{O}_7$ that may increase the bulk resistance. Also, the bulk conductivity of the doped LaCrO_3 used in the electrodes will be measured.

SECTION 5
FABRICATION (TASK 3)

5. FABRICATION (TASK 3)

5.1 FABRICATION DEMONSTRATION (TASK 3.1)

5.1.1 Objective

The objective of this task is to fabricate MSOFC cells and stacks of increasing power output to demonstrate the viability of the fabrication process.

5.1.2 Fabrication of Cells and Stacks

MSOFC cells and stacks are being made using a fabrication sequence based on the tape-calendering technique. This technique is explained in the October-December 1989 Quarterly Report, AiResearch Document 89-63136(3), pages 3-1 through 3-3. Primary activities during the last quarter included batch preparation and characterization, fabrication of single cells and multicell stacks, bonding studies, interconnect development, and shrinkage matching.

5.1.2.1 Batch Preparation and Characterization

Batch preparation efforts for the last quarter were concentrated in two areas: providing a tape supply for cell and stack fabrication, and investigating formulation modifications to improve the structural performance of the baseline materials. Several batches were prepared last quarter using the established procedures. The prepared batches were characterized in terms of weight-loss during firing, sintered density, and total firing shrinkage (diametral).

Four baseline anode batches were made to replenish the tape supply. The average properties of these batches are summarized in Table 5-1. Figure 5-1 shows the properties of these anode batches along with those prepared since May 1991. The properties of these batches fall within the ranges expected for baseline materials.

TABLE 5-1				
AVERAGE SINTERED PROPERTIES OF ANODE BATCHES				
Baseline Anode Batch	Diametral Shrinkage, Percent	Weight Loss, Percent	Density, g/cc	TD*, Percent
A11191-1	17.7 ± 0.9	23.1 ± 0.1	4.38 ± 0.18	71.5 ± 3.0
A11201-1	17.6 ± 1.0	22.9 ± 0.1	4.48 ± 0.09	73.1 ± 1.5
A11201-2	19.6 ± 0.5	22.6 ± 0.2	4.33 ± 0.13	70.7 ± 1.9
A11211-1	19.3 ± 1.3	22.9 ± 0.2	4.43 ± 0.11	72.2 ± 1.8
*Theoretical density

Figure 5-1. Properties of Baseline Anode Batches Made Since May 1991

Other batches prepared in the last quarter were made in an effort to improve the structural performance of the baseline batches. The primary objective was to decrease the magnitude and frequency of defects in modules and stacks. The approach was to toughen the baseline materials. Toughening was made by replacing portions of the zirconia components with alumina in each of the anode (A), electrolyte (E), and cathode-electrolyte intermediate (CE) formulations. The addition of alumina has been shown to increase the strength and toughness of stabilized zirconia in previous work ^{1,2}.

Modified anode (A10241-1), electrolyte (E10241-1) and intermediate (CE11071-1 and CE11071-2) batches were prepared based on this toughening approach. Table 5-2 shows the average sintered properties for these batches.

TABLE 5-2				
AVERAGE PROPERTIES OF MODIFIED BATCHES				
Modified MSOFC Batch	Diametral Shrinkage, Percent	Weight Loss, Percent	Density, g/cc	TD*, Percent
A10241-1	16.5 ± 0.5	23.5 ± 0.1	4.15 ± 0.06	69.9 ± 1.1
E10241-1	13.2 ± 0.1	17.8 ± 0.1	5.43 ± 0.05	99.9 ± 0.9
CE11071-1	22.4 ± 0.8	24.6 ± 0.1	5.14 ± 0.18	85.5 ± 3.0
CE11071-2	22.2 ± 0.4	24.8 ± 0.1	5.05 ± 0.17	83.9 ± 2.8
*Theoretical density				

Using X-ray diffraction, the phase compositions of the modified materials were evaluated and compared to the baseline materials. Table 5-3 shows the phase compositions of the baseline and modified batches. In the anode layer, all of the alumina (in both the baseline and the modified material) reacts with nickelous oxide to form a nickel aluminate phase; the zirconia remains as a separate phase in the anode. In the electrolyte layer, the zirconia and alumina remain as separate phases. In the intermediate layer, the lanthanum zirconate phase, which is seen in the baseline, is suppressed by the formation of a manganese aluminate phase in the modified material.

The effect of the presence of Mn_2AlO_4 phase on the electrochemical properties of the intermediate (CE) is being evaluated.

5.1.2.2 References

1. Allied-Signal, unpublished results.
2. O. Yamamoto, et al. "Electrical and Mechanical Properties of Zirconia-Alumina Composite Electrolyte," in Proceedings of the Second International Symposium on Solid Oxide Fuel Cells, Commission of the European Communities, 1991, pp. 437-444.

TABLE 5-3		
PHASES PRESENT IN BASELINE AND MODIFIED MSOFC BATCHES		
	Baseline	Modified
Anode (A)	NiO ZrO ₂ NiAl ₂ O ₄	NiO ZrO ₂ NiAl ₂ O ₄
Electrolyte (E)	ZrO ₂	ZrO ₂
Cathode/electrolyte (CE)	ZrO ₂ LaMnO ₃ La ₂ Zr ₂ O ₇	ZrO ₂ LaMnO ₃ Mn ₂ AlO ₄

5.1.2.3 Cell and Stack Fabrication

Fabrication of flat single cell modules and stacks with minimum observable defects for subsequent second-step bond was this effort's primary objective. More than 200 single-cell modules and stacks were made in the last quarter to support second-step bond development and two-cell stack building.

The fabrication of single-cell modules concentrated on the 2.5-in. and 1.3-in. green footprint single-cell modules. The emphasis of the work was to minimize the degree of bowing in fabricated samples while maintaining gas-channel shape retention and structural integrity. Flatness could only result from high constraint weight; however, the higher weight caused corrugation collapsing as the tapes softened during binder extraction. Firing runs with different constraint weights were carried out and the results were used to select a tradeoff constraint weight for each module size.

During the last quarter, several modifications were made in the processing of single-cell modules. Significant progress was made in the fabrication of modules with electrode cover and in the use of a creep-sintering process. Single-cell modules of a 1.3-in. footprint area were fabricated with either an A or CE cover layer bonded on in the green state. Several anode and CE batches with different solids loading levels were used as cover layers to determine the most suitable combination. Evaluation of initial results indicated that the high shrinkage of the anode layer caused significant bowing of fabricated samples. Therefore, a higher solids loading anode (A07030-1) was used for the anode corrugation as well as anode cover. This construction resulted in flat anode-covered modules with minimal observable defects. Using A07030-1 as the anode corrugation in CE-covered modules produced similar results. The ability to fabricate electrode-covered modules improves the two-step stack-building process by lessening the number of second-step bonds.

To obtain flat stacks with well-defined gas channels, varying the constraint weight during processing was attempted; low weight was used for low-temperature binder extraction and high weight was used for the sintering temperature regime. This

variation can be performed by splitting the processing run into two steps. In the first step, the green sample is constrained with a low weight and fired to a temperature lower than sintering temperature but higher than binder burnout temperatures. The sample can then support more weight. In the second step, the sample is then fired to the sintering temperature with a higher constraint weight. This higher load assists in flattening the sample during sintering, a type of creep-assisted sintering. Two maximum temperatures were investigated for the first step. The first, 600°C, is just above the binder burnout temperature, while the other, 1300°C, is just below the sintering temperature. Going up to 1300°C in the first step proved better because the resulting stacks had fewer defects. The constraint weight used in the second step is double that used in the first step. This creep-sintering process has yielded flat modules and electrode-covered modules with minimal observable defects.

The single-cell stack fabrication effort last quarter focused on evaluating the effect of altered electrode cover layer thicknesses and the impact of Al₂O₃-toughened components on single-cell stacks.

A series of single-cell stacks, consisting of varied electrode cover layer thicknesses, were fabricated and processed. The objective of this effort was to evaluate cover thicknesses for stack flatness improvement. The standard single-cell stack cover layer green thickness is 13 mils. Table 5-4 shows the combinations of anode and cathode cover layer thicknesses evaluated.

TABLE 5-4		
SINGLE-CELL STACKS WITH VARIOUS COVER THICKNESSES		
Stack ID	Anode Thickness, mils	Cathode Thickness, mils
EB037-1	13	13
EB037-2	13	26
EB037-3	26	13
EB037-4	26	26

Visual and optical microscopic examination of the sintered samples indicated that fabricating single-cell stacks with 13-mil electrode cover layers gave the best results. All stacks that were fabricated with either a 26-mil anode or cathode cover layer possessed severe defects and delaminations. Although the cathode covers layers remained extremely flat in stacks constructed with 26-mil cathode covers, the anode layers within these stacks sintered to a concave shape, causing delamination and severe defects within the stacks.

An effort was made this quarter to reduce the magnitude and frequency of the defects (particularly the V-cracks commonly seen in CE corrugations) found in single-cell stacks. The approach taken to reduce these defects was to incorporate toughened

components in single-cell stacks. Experiments were carried out to find the best combination of toughened layers and baseline layers when structural integrity and electrochemical performance were considered in single cells and single-cell stacks.

To find the best structural combination, matrices of single-cell discs (green dia = 1.25 in.) and single-cell stacks (green dimensions = 1.3 in. x 1.3 in.) were fabricated. Table 5-5 shows the matrices and the yields for each combination of cell and stack.

TABLE 5-5				
RESULTS OF CELLS AND STACKS FABRICATED WITH BASELINE AND MODIFIED COMPONENTS				
Component			Results	
A	E	CE	Single-Cell Disk (Yield)	Single-Cell Stack (Observation)
Baseline	Baseline	Baseline	8/10	Defect Type II
Baseline	Modified	Baseline	0/4	Defect Type IV
Modified	Baseline	Baseline	4/4	Warped
Modified	Modified	Baseline	4/4	Warped
Baseline	Baseline	Modified	4/4	No observable defects
Baseline	Modified	Modified	4/4	Defect Type III
Modified	Baseline	Modified	4/4	Warped
Modified	Modified	Modified	3/4	Warped, slumped
Defect Type II: Defect (hairline crack) barely identifiable at 70X and not found within electrolyte layer				
Defect Type III: Defect (hairline crack) easily identifiable at 70X and not found within electrolyte layer				
Defect Type IV: Defect (hairline crack) found in or through electrolyte layer				

From these results, it was concluded that a combination of baseline A, baseline E, and modified CE was structurally the best combination considered. Several 1.3-in. (green footprint area) single-cell stacks have been successfully processed and subsequently fabricated into multicell stacks using the second-step bonding technique.

During last quarter, a number of baseline single-cell stacks were also processed using the creep-sintering technique. Optical microscopic examination of baseline single-cell stacks after processing showed the presence of a few common defects. However, the degree of bowing was dramatically reduced compared to single-cell stacks processed by the conventional sintering method. In addition, single-cell stacks constructed with baseline anode, baseline electrolyte and Al₂O₃-toughened CE were fabricated and processed using the creep-sintering technique. Visual and optical

examination of the stacks after processing showed the presence of very few defects and the absence of bowing. Several of these stacks have been selected for multicell bonding and testing. Additional stacks are being processed to confirm this preliminary analysis and assess the reproducibility.

5.1.2.4 Bonding Studies

During the last quarter, a number of two-cell stacks were fabricated by the two-step cofiring process. The stacks were made from single-cell modules or single-cell stacks of a 1-in. and 2-in. footprint area (sintered area). After firing, all the stacks showed bonding without delamination.

Extensive evaluation and characterization were performed on bonded samples. The key results are summarized below:

- (a) The bonding step did not cause any damage or cracking to the interconnect or the bonded single-cell modules or stacks.
- (b) Bonded stacks showed excellent strength. Several samples were sectioned without delaminating.
- (c) The anode/anode and anode/interconnect had excellent bonding contact, whereas the cathode/interconnect needed improvement.
- (d) Bonded stacks were thermally cycled (3 cycles to 900° to 1000°C) and remained bonded.

Modifications of bonding materials and procedures have resulted in improved bonding contact. Examples of improved bonding are shown in Figure 5-2.

Development of a bonding formulation last quarter focused on materials for the cathode-electrolyte intermediate (CE) to interconnect (I) bond. Since the poor contact of the CE-I bond appeared to be due to the high shrinkage and low toughness of the bonding tape, evaluation of CE bond formulations concentrated on solids loading, powder reactivity, and use of a toughening agent. Four batches were prepared according to the matrix shown in Table 5-6. Each formulation was used in a CE-I bond with both corrugation/flat plate and flat plate/flat plate bond geometries. Figure 5-3 shows a typical area of the flat plate/flat plate bonds.

All formulations yielded good bonds as shown in the photographs. The low solids loading formulations (Nos. 1 and 2) have better bonding tape homogeneity, making them preferable.

5.1.2.5 Interconnect Studies

During the last quarter, tapes were prepared with lanthanum chromite powders treated in a variety of ways. The tapes were evaluated to determine any processing or powder preparation conditions that could influence densification.

(a)

(b)

F-64418

Figure 5-2. Micrographs of Bonding between Interconnect
and Electrode Corrugations (80X Magnification)
(a) Anode Corrugation/Interconnect
(b) Intermediate Corrugation/Interconnect

TABLE 5-6			
CE BOND FORMULATIONS			
Formulation No.	Solids Loading	Reactivity	Toughening
1	Low	High	No
2	Low	High	Yes
3	High	Low	No
4	High	Low	Yes

Tapes were initially prepared using the as-received powder (surface area = 2.91 m²/g) and powder that was ball-milled and calcined (surface area = 1 to 2 m²/g). The measured surface area is average surface area for spherical agglomerates of the lanthanum chromite. In most cases, the as-received powder when mixed with the binder and plasticizer could not be rolled into a tape. Tapes made of lanthanum chromite, which was milled and calcined, could easily be made into tapes; however, only a maximum density of 94-percent theoretical density was achieved by sintering at 1400°C for 1 hr. The as-sintered and fractured surfaces of this material, shown in Figure 5-4, indicate that there is a lot of enclosed porosity residing at the grain boundaries.

The tape composed of the as-received powder that would not roll was heated to remove the binder so that the powder could be used in subsequent batches. The powder had a surface area of 18.5 m²/gm after it was debindered. The high surface area indicated that the soft agglomerates were broken down during shear mixing when the plastic mass was being formed. This powder was calcined at temperatures above 1000°C for 5 hr and then made into tapes. Sintering of this tape resulted in densities of 94-percent theoretical density, similar to those of the previously prepared material (ball-milled and calcined). The as-sintered surface of this material, shown in Figure 5-5, is more uniform and shows very little porosity in the microstructure. The fracture surface shows a similar distribution in the amount of porosity in the microstructure as compared to the sample shown in Figure 5-4; however, the size of the porosity is much larger in the sample in Figure 5-5.

This result indicates that the as-sintered microstructure can be improved by breaking down the agglomerates in the as-received powder into their individual particles. There is no change, however, in the sintered density of the material.

5.1.2.6 Shrinkage Studies

The main thrust of this effort last quarter was to measure the shrinkage profile of toughened electrode compositions. The objective was to determine the effect of the toughening additive (alumina) on sintering shrinkage profiles of the anode and intermediate layers.

(a)

(b)

(c)

(d)

Figure 5-3. Cross-Section of Bonding Between Flat Cathode/
Electrolyte Intermediate (CE) (Top) and Interconnect
(I) (Bottom) (Magnification = 64X)

- (a) Bond Formulation 1
- (b) Bond Formulation 2
- (c) Bond Formulation 3
- (d) Bond Formulation 4

As-Sintered

Fracture

F-64414

Figure 5-4 As-Sintered and Fracture Surfaces of Lanthanum
Chromite Sintered 1400°C for 1 hr. Powder from
Lot 0260, Ball-Milled and Calcined

As-Sintered

Fracture

F-64415

Figure 5-5 As-Sintered and Fracture Surfaces of Lanthanum Chromite Sintered 1400°C for 1 hr. Powder from Lot 0260, Previously made into Tape in As-Received State

Figures 5-6 and 5-7 compare the sintering shrinkages of toughened anode (Figure 5-6) and CE (Figure 5-7) with their baseline counterparts. As can be seen from Figures 5-6 and 5-7, the addition of alumina causes some differences in anode sintering behavior, whereas little change in total shrinkage occurs in the modified intermediate. Since the effect of alumina addition on shrinkage is minimal in the CE, the incorporation of the toughened intermediate into single-cell and stack fabrication is expected to have an insignificant effect on stack structural integrity. This has been shown to be the case, as reported in para. 5.1.2.3.

5.2 DEFECT ANALYSIS (TASK 3.3)

5.2.1 Objective

The objective of this task is to analyze defects in MSOFC cells and stacks and correlate identified defects to material and fabrication parameters. Correlation can be used to suggest fabrication modifications to minimize defects.

5.2.2 Analysis

Defect analysis in the last quarter was focused on module characterization using the simplified defect classification discussed in the last report, AiResearch Document 91-94749. An effort was also initiated to correlate the observed defect frequency to the baseline batch combinations used in single-cell modules.

A preliminary analysis has shown that the batch combinations might affect the resulting defect frequency. Figure 5-8 exemplifies this for 2.5-in. green footprint modules. (The letters along the bottom of the figure denote the particular batch used for that layer within the module.) Figure 5-8 shows how changing module composition by one batch can affect the frequency of the type of defect observed. The reasons for this effect are not known at this time because of complications caused by other factors such as processing parameters. Further compilation and analysis are expected to lead to a better understanding of this observation on defect frequency.

5.3 MATERIAL AND PROCESS SPECIFICATIONS (TASK 3.4)

5.3.1 Objective

The objective of this task is to develop baseline material and process specifications for the fabrication of the MSOFC. The specification will document the characteristics of starting materials and process parameters and identify acceptable ranges for each variable.

5.3.2 Specification Development

During this reporting period one new process specification was completed: AF561 – Fuel-Cell Single-Cell Performance Testing. The method of conducting electrical performance testing of single-cell discs under actual operating conditions is discussed. Sample preparation, test rig assembly, experimental procedure, and data collection are described in detail.

TOUGHENED ANODE SINTERING SHRINKAGE COMPARISON

— A05201-1, std. form. A10241-2, toughened

Figure 5-6. Sintering Shrinkage Profiles of Baseline and Toughened Anodes (A)

TOUGHENED INTERMEDIATE SINTERING SHRINKAGE COMPARISO

— CE08291-1, std. form. - - - CE11071-1, toughened

Figure 5-7. Sintering Shrinkage Profiles of Baseline and Toughened Cathode/Electrolyte Intermediates (CE)

GREEN PLAN AREA: 2.5" x 2.5"
THRU ST1768/EE228

Figure 5-8. Defect Frequency for Single-Cell Modules Fabricated with Different Batches

Statistical process controls (SPC) are being incorporated into the process specifications as the controls are developed. Currently, data on sintered components are being collected so that the acceptance limits can be further refined for the SPC controls that have been established . On an interim basis, 95-percent confidence limits ($\pm 2\sigma$) are being used that are based on recent measurements of each powder.

SECTION 6

TESTING ON COAL GAS (TASK 4)

6. TESTING ON COAL GAS (TASK 4)

6.1 PERFORMANCE TESTING (TASK 4.1)

6.1.1 Objective

The objective of this task is to assess the electrochemical performance characteristics of MSOFC cells and stacks under conditions of pressure, temperature, gas composition, gas flow rate, and current density. Test results will be used to provide inputs to support materials, fabrication, and quality improvements.

6.1.2 Single-Cell Testing

The testing of single cells concentrated on three efforts: (1) evaluating better methods to apply current collectors to single-cells, (2) evaluating a removable seal for single cell testing, and (3) assessing the electrochemical performance of Al_2O_3 -toughened single cells discussed in para. 5.1.2.3.

Since the present platinum paste method is unreliable and time-consuming, two alternate methods to attach current collectors were evaluated. Both methods involved applying current collectors to single cells using the same materials utilized in multicell stack bonding (discussed in para. 5.1.2.4). The first method involved dissolving anode and cathode bonding tapes in alcohol to form a slurry. The slurry of each electrode material was then used to apply current collectors to the appropriate electrode surfaces of single cells. Single cells ED045-26, ED040-45 and ED040-36 were prepared in the manner discussed above and performance tests were conducted on each cell. Each of the cells was fabricated from baseline materials and constructed with an A:E:CE layer thickness ratio of 2:1:1. The cells were heated to 1000°C under helium before testing commenced. Performance tests were conducted at 1000°C with 97 percent H_2 /3 percent H_2O as fuel, and air as oxidant. The test results are summarized below:

- (a) ED045-26— During initial performance testing, severe voltage instabilities occurred at a current of 500 ma. The sample was allowed to stabilize for 48 hr, and a performance test was performed without voltage instabilities. The performance curve for ED045-26 is presented in Figure 6-1. The area specific resistance (ASR) obtained was 0.96 ohm-cm^2 . The open-circuit voltage (OCV) was stable at 1.05 v.
- (b) ED040-45— During performance testing, severe voltage instabilities were experienced at a current of 50 ma. The polarity was reversed at 400 ma for 20 sec in an attempt to improve performance. However, voltage instabilities were encountered at 100 ma. The open-circuit voltage (OCV) was stable at 1.02 v.
- (c) ED040-36— Voltage instabilities occurred at 50 ma during performance testing. The voltage instability was overcome by performing an electrolysis at 800 ma for approximately 5 min. Figure 6-2 shows the polarization curve generated after reverse polarization. The resulting ASR was 1.4 ohm-cm^2 . The OCV was stable at 0.98 v.

Figure 6-1. Performance Curve of Cell ED045-26

T = 1000°C

Fuel = 97 Percent H₂/3 Percent H₂O

Oxidant = Air

Figure 6-2. Performance Curve of Cell ED040-36 After Electrolysis at 0.8 amp for 5 Min
T = 1000°C
Fuel = 97 Percent H₂/3 Percent H₂O
Oxidant = Air

An additional method was evaluated to attach current collectors to cells which involved adding platinum powder to the slurry mixtures of each of the electrode materials. The slurry/Pt powder was mixed until a homogeneous mixture resulted. Current collectors were attached to the electrode surfaces of the single-cell ED040-35 using the slurry/Pt mixture. Single-cell ED040-35 was prepared using this method and a performance test was conducted. Performance testing was performed with the conditions mentioned earlier. The results are presented below.

- ED040-35—Voltage instabilities were encountered at 400 ma during performance testing. The sample was allowed to stabilize overnight under H₂; however, voltage instabilities still persisted at 400 ma. The OCV was stable at 1.015 v.

One possible reason for the voltage instabilities observed during performance testing appears to be a poor contact between the platinum screen current collector and the electrode surface.

Presently, current collectors are applied to single-cell samples using the standard platinum paste method. However, it was determined that increasing the metallizing temperature (from 800° to 900°C) minimizes the problems of current collector detachment encountered in the past.

FiberFrax LDS moldable insulation caulk was evaluated as a candidate to provide a removable (noncement) material for sealing single-cell discs for performance testing. Single-cell FD040-4 was prepared for testing and applied to the test rig using LDS moldable caulk as a sealant. The initial OCV obtained for FD040-4 (with 97 percent H₂/3 percent H₂O as fuel, and air as oxidant) was 0.85 v. This OCV is lower than the typical value of 1.05 to 1.07 v obtained for single cells. A performance test was not performed due to a slow degradation of the OCV over time, indicating the presence of a significant leak. Upon observation of FD040-4 after cooling to room temperature, large gaps between the LDS moldable material and the single cell were observed.

A performance test was also conducted to determine the electrochemical performance of single cells that have been toughened by the addition of Al₂O₃ (discussed in para. 5.1.2.3). Single-cell ED065-8 was fabricated with baseline anode and electrolyte layers and an Al₂O₃-toughened cathode-electrolyte layer, using a 2:1:1 construction. The results of the performance test are discussed below.

- ED065-8—The initial performance curve for ED065-8 is shown in Figure 6-3. The OCV was stable at 1.01 v. The initial ASR was 1.2 ohm-cm². This ASR is slightly higher than the typical value of 0.7 to 0.8 ohm-cm² obtained for strictly baseline 2:1:1 single cells.

Additional performance tests are necessary to confirm the preliminary electrochemical performance characteristics obtained for single cells possessing a toughened cathode-electrolyte layer.

Figure 6-3 Performance Curve of Cell ED065-8
T = 1000°C
Fuel = 97 Percent H₂/3 Percent H₂O
Oxidant = Air

6.1.3 Stack Testing/Life Testing

A total of three stacks, one single-cell stack and two two-cell stacks were performance tested during this quarter. Results for each stack are discussed below.

- (a) EB023-02 — Single-cell stack EB023-02 was held under an endurance test for nearly 200 hr. Open-circuit voltage and area-specific resistance were checked on a regular basis. Performance for the stack is shown in Figure 6-4. As can be seen, OCV's were relatively stable, while AST gradually increased (higher resistances) over a period of days ranging from 1.7 to 2.4 ohm-cm², with the exception of the first day under test. It is not fully understood why the initial curve for day 1 is lower than the following days. It is speculated that the nickel in the anode was still being reduced, but this would be difficult to verify at operating conditions. The curves generated for Figure 6-4 were all taken with 20 percent H₂/80 percent He (humidified), and air at 1000°C. Stack EB023-02 was tested with a variety of fuel concentrations and humidities, to determine their effect on performance. By running the stack without humidity, and 20 percent H₂ /80 percent He, an OCV of 0.965 was obtained.
- (b) EG196-04 — Two-cell stack EG196-04, tested in mid-October, performed much poorer than expected due to a manifold leak, which was confirmed by a post-test analysis. As a result, it was decided to perform off-line experiments to study new cements and hopefully correct problems that were observed.
- (c) EG259-02 — Two-cell stack EG259-02 was tested in mid-December. In an attempt to eliminate external variables during performance testing, an alternate manifolding procedure was utilized for the testing of EG259-02. The alterations involved prefiring a majority of the manifold components using Aremco 516 zirconia-based cement before the stack was inserted into the manifold. The stack was then inserted into the manifold and sealed using Aremco 571 magnesia-based cement. The 516 cement was not used to seal the stack in the manifold since it was determined that it may induce cracks in the stack as a result of expansion during curing. The stack's OCV was 1.527 v, and the ASR was 15.94 ohm-cm² (7.97 ohm-cm² per stack). A post-test analysis is being conducted to assess the impact of the manifolding alterations. The results will be discussed in the next report.

Figure 6-4. Performance of AiResearch MSOFV Single-Cell Stack EB023-02

SECTION 7

QUALITY DEVELOPMENT (TASK 5)

PERFORMANCE AND COST ANALYSIS

7. PERFORMANCE AND COST ANALYSIS, TASK 5

7.1 STACK PERFORMANCE MODELING, TASK 5.1

7.1.1 Objective

The objective of this task is to develop a mathematical model of a crossflow MSOFC to obtain a better understanding of the interactions among the various design and operating variables, and their effects on the overall performance of the stack.

7.1.2 Results

A mathematical model of the MSOFC called FCELL is complete. The model simulates the electrochemical, electrical, thermodynamic, and chemical equilibrium processes occurring in the cell. A single cell within the MSOFC stack is modeled. This cell is assumed to be sufficiently deep within the stack so that end effects may be neglected. The horizontal plane of the cell is divided into a two-dimensional grid of rectangular areas or elements. Each element has a single, centrally located node. The elements are sufficiently small that the nodal values of temperature, pressure, concentration, etc. can be used over the entire element for the calculation of local current densities and heat fluxes. The mass balance, energy balance, and expressions for the electrochemical and electrical phenomena are set up and solved for each element.

The inputs to the program include the geometry of the fuel cell, the cell operating voltage, and the inlet fuel and oxidant temperatures, pressures, flow rates, and compositions. The model can operate with any fuel that contains any or all of the following constituents: hydrogen, water, simulated diesel fuel ($C_{16}H_{34}$), carbon monoxide, carbon dioxide, methane, oxygen, or nitrogen. Air or pure oxygen can be used as the oxidant. If desired, the program can operate in a mode in which the user specifies the cell current, fuel utilization, or maximum desired cell temperature in place of the cell voltage or one of the inlet flow rates. In this mode of operation the program uses an additional iteration loop to determine the cell performance based on the input values. The program outputs include the temperature, composition, Nernst potential, and current at each node. Also, a number of performance parameters are calculated such as the fuel and oxidant utilization, total cell power, fuel efficiency, and average temperatures.

SECTION 8
QUALITY DEVELOPMENT (TASK 6)

8. QUALITY DEVELOPMENT, TASK 6

8.1 EVALUATION OF NONDESTRUCTIVE EVALUATION (NDE) METHODS, TASK 6.1

8.1.1 Objectives

The objectives of this subtask are to: (1) evaluate, develop, and implement promising NDE methods that can be used to improve the yield of reliable ceramics for MSOFC's during processing and in final fabricated parts, and (2) select and develop NDE methods best suited to ensure the quality of commercial-scale fabricated MSOFC's.

8.1.2 Results

This subtask is complete. A draft copy of the final report entitled, *Nondestructive Characterization Methods for Monolithic Solid Oxide Fuel Cells* is currently being reviewed.

8.2 QUALITY CONTROL OF SOURCE MATERIALS (TASK 6.3)

8.2.1 Objective

The objective of this task is to evaluate the characteristics and quality of source materials for use in the fabrication of the MSOFC. Variations in material characteristics and their effect on MSOFC fabrication will be quantified, and the acceptable quality (material requirements) of source materials will be specified.

8.2.2 Evaluation of Lanthanum Chromite

SSC, Inc., the supplier of the lanthanum chromite powders, is analyzing its powders to determine the exact stoichiometry of Lot 147, which sintered to 99-percent theoretical density. Once the stoichiometry is determined, a future lot will be prepared and evaluated with this stoichiometry.

END

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