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**Quarterly
Technical Status Report
October-December 1992**

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

93-65838

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**Department of Energy
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MASTER

Allied-Signal Aerospace Company

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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 1992.

The program is conducted by a team consisting of AlliedSignal Aerospace Systems and Equipment (ASE) (formerly the AiResearch Los Angeles Division of Allied-Signal Aerospace Company), Argonne National Laboratory (ANL), and AlliedSignal Research and Technology. 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

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 MATERIALS RESEARCH AND DEVELOPMENT (TASK 2)

Densification of the interconnect under relatively moderate firing conditions remains a challenge in fabricating the MSOFC. Although the interconnect densifies to high density 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 that consists of only electrolyte and interconnect materials. These materials are currently formed into five layers symmetrically configured around the ZrO_2 electrolyte. The outer two layers comprise $\text{ZrO}_2/\text{LaCrO}_3$ mixtures to improve bonding. The layer closest to the electrolyte called the bond coat contains greater wt% ZrO_2 than LaCrO_3 , whereas the opposite is true for the outer layer. The best performance achieved to date is 1.91 ohm-cm^2 , which is inadequate for fuel cell stack criteria. It was anticipated that thinner electrolyte and bond coat layers would further improve performance. However, the thinner electrolyte does not properly densify at 1400° or 1500°C .

The second approach is based on the idea that the interconnect does not densify in the four-material cell because the liquid that aids sintering migrates into the neighboring electrode layers before the onset of densification. It is thought that better densification would result if the sintering aid did not melt until after the onset of densification.

3.2 FABRICATION (TASK 3)

During this period, new anode batches were prepared and characterized to support stack and large-plate fabrication efforts. Thin-electrolyte cells were fabricated using the new anode batches. Additionally stack building concepts are being developed that reduce the number of interfaces to be bonded with bonding tapes in the two-step bonding stack fabrication process.

Also during this period, a conceptual design review was held to discuss a methodology to meet the 100-w stack milestone in FY 1993 by the end of September 1993. Team members from Argonne National Laboratories, AlliedSignal Research and Technology, and AlliedSignal Systems and Equipment were in attendance.

IG-02458-E

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

3.3 TESTING ON COAL GAS (TASK 4)

During this quarter, a performance test was conducted on a single cell with an anode formulation modified to improve the match in coefficients of thermal expansion. Also during this peiroad, testing was conducted on a thin electrolyte disk over the temperature range of 700° to 1100°C.

3.4 PERFORMANCE AND COST ANALYSIS (TASK 5)

An electrical analysis was conducted to investigate the resistive losses associated with an alternate MSOFC configuration comprising anode, electrolyte, cathode, and interconnect plates, and anode and cathode bars. The analysis showed the dependence of area specific resistance on bar height, bar thickness, and number of bars per inch.

3.5 QUALITY DEVELOPMENT (TASK 6)

A single-cell stack design based on this thin-electrolyte technology was analyzed for stress levels. High stress levels were found in the anode layer of the A/E/C/ trilayer. Better matching of the anode and cathode CTE's was found to reduce all stress levels to acceptable levels.

SECTION 4

MATERIALS RESEARCH AND DEVELOPMENT (TASK 2)

4. MATERIALS RESEARCH AND DEVELOPMENT (TASK 2)

4.1 TASK 2.2: CATHODE/INTERCONNECT INTERACTION

Strontium- and cobalt-doped and calcium- and cobalt-doped lanthanum chromite powders with various A-site to B-site ratios were prepared via the liquid mix method. Pellets comprising each interconnect powder were pressed and fired for 1 hr in air at 1400° and 1500°C. The density was measured and is presented below:

Composition	A/B ratio	Density, g/cm ³ (1400°C)	Density, g/cm ³ (1500°C)
La _{0.8} Sr _{0.22} Cr _{0.9} Co _{0.1} O ₃	1.02	5.20	6.16
La _{0.8} Sr _{0.20} Cr _{0.9} Co _{0.1} O ₃	1.00	5.55	6.22
La _{0.8} Sr _{0.18} Cr _{0.9} Co _{0.1} O ₃	0.98	5.31	6.22
La _{0.8} Sr _{0.22} Cr _{0.9} Co _{0.1} O ₃	1.02	5.85	6.12

These results are consistent with the report of Chick *et al.*¹ Al composite tapes were also made with each powder and used in assembling cofired modules comprising Al/I corrugation/AEC. Problems with curling of the Al tapes during binder removal, and collapsing of the interconnect corrugations during curing show that the organic content needs to be adjusted. The Al tapes tend to curl toward the interconnect, suggesting that it would be better to use the interconnect layer by itself rather than the two-layer composite containing the anode.

An effort was initiated to identify chemical changes occurring at the electrode/interconnect interface as a result of the intimate contact between these materials at high bonding temperatures. Because large numbers of unique cations are present in contiguous materials, there is a significant probability for chemical change, either through interdiffusion and substitution, or through compound formation. These chemical interactions may result in physical, as well as crystallographic changes. For example, in considering a cathode material consisting only of La_{0.69}Sr_{0.3}MnO₃ in contact with interconnect material consisting of La_{0.79}Ca_{0.2}Cr_{0.9}Co_{0.1}O₃, the following might occur: Sr²⁺ and Cr³⁺ from the cathode and interconnect, respectively, could react to form Sr₃Cr₂O₈ or Mn³⁺ and Ca²⁺ could form CaMn₂O₄. These interactions might result in either a change in the coefficient of thermal expansion, or chemical instability in one of the components due to excess La³⁺. While this is a simple example, it points out only a few of the many interactions and consequences that are possible.

A preliminary plan of experimental study has been developed. In these experiments, bilayers of doped and undoped lanthanum chromite/lanthanum manganite and lanthanum chromite/zirconia will be sintered at 1400°C and then held at either 1400°C or 1000°C for different times and cooling rates. The interactions at the interface and in the bulk layers of the components will be determined by energy-dispersive X-ray

1. "Factors Affecting Air-Sintering of Chromite Interconnections", L.A. Chick and J.L. Bates, Fourth Annual Fuel Cells Contractors' Review Meeting, Morgantown, W.Va, July 14-15, 1992.

analyses, ion mapping, and X-ray diffraction. Tape casting of films of sample materials has begun.

4.2 TASK 2.6: ANCILLARY MATERIALS DEVELOPMENT

Problems encountered in the assembly and use of the conventional fixture for testing MSOFC stacks have led to the conclusion that effort should be devoted to developing new, more resilient test fixtures. Analysis of the failure modes of the stack and/or the test fixture suggests the following approach:

(a) Examination of the as-built stacks

The stacks should be checked for reproducibility, strength, thermal cyclability, alignment, quality of internal seals, etc., using relatively simple tests such as visual inspection, simple tension, thermal cycling, and sectioning and microscopy.

(b) Compliant Test Fixture Designs

To reduce stresses during testing of the stack, the test fixture should be weakened (by using a small cross-section or a low-modulus material) so that it will take up most of the strain from a thermal expansion mismatch. A test fixture with reduced stiffness would also be expected to ease stress concentrations resulting from any warpage of the stack.

(c) Thermal expansion/contraction measurements for the assembled stacks

Although CTE's of individual components may be known, the simple linear combination of the values to estimate the CTE of the stack may not be accurate due to elastic rearrangement of the components, possible reactions, or other adjustments to the thickness of each layer.

During this quarter, some 1-in. by 1-in. stacks of the conventional design built by AlliedSignal were examined at ANL, who observed that the edges and corners of successive layers are sometimes misaligned; such an arrangement leaves uneven lines for sealing, requiring an increase in bulk of the sealant, which may complicate mechanical behavior of the stack during heatup and testing. The stacks were successfully thermally cycled between room temperature and 1000°C.

Hairline cracks were observed in the cathode corrugation/electrolyte interface area at the edges of the cells. These cracks were not found on the interior sections of the cells, indicating that the cracks are probably due to stress from mismatch, an effect that decreases towards the center of the composite structure.

Thin, flat plates of partially stabilized zirconia (6 mol% Y_2O_3 , TZ-3Y) were fabricated for use in testing as a possible manifold material. TZ-3Y was thought to be suitable because its thermal expansion should be close to the thermal expansion of fuel cell components. Samples of TZ-3Y and fuel cell components were made for thermal expansion measurements.

To test the suitability of TZ-3Y as a manifold material, its thermal expansion coefficient and that of fuel cell components fabricated at ANL were measured. The results obtained to date are given below. Because the thermal expansion changes with temperature, it is given at low temperature as well as high temperature.

<u>Material</u>	<u>Thermal Expansion</u>	<u>Thermal Expansion</u>
	<u>(°C x 10⁶)</u> <u>(200 to 300°C)</u>	<u>(°C x 10⁶)</u> <u>(900 to 1000°C)</u>
Anode	11.0	12.0
Electrolyte	9.2	11.0
Cathode	10.0	12.0
Manifold (TZ-3Y)	10.5	10.5

The thermal expansion of the TZ-3Y falls in the range of values for the other components, although their thermal expansion values are not well matched.

Compliant test fixture designs incorporating thin (tape-cast) manifold materials were drawn up and tested. One such design, incorporating an inclined plane geometry, is especially attractive since it does not involve precise test fixture machining to match stack dimensions and can be used for short as well as tall stacks. Preliminary tests with this design were conducted successfully. The next stack tests will be conducted using such a fixture.

SECTION 5
FABRICATION (TASK 3)

5. FABRICATION (TASK 3)

5.1 OBJECTIVE

During this reporting period, efforts focused on fabrication of anode material with a better match in CTE and thin-electrolyte cells. Additionally, a conceptual design review was held to discuss technical approaches to achieving the 100-w stack milestone by September 1993.

5.2 FABRICATION

During this reporting period, three new anode batches were prepared to support cell and stack fabrication efforts. These batches contained 50 vol% solids loading. The properties of both the starting materials and the batches were characterized. Characterization of starting materials involved mainly particle size distribution and surface area analysis. Characterization of the anode batches involved determination of firing weight loss, linear firing shrinkage, and fired density/porosity. The characterization data are being compiled. The microstructures of fired anodes were also examined using SEM. No unusual features were observed.

A number of thin-electrolyte single cells were fabricated this quarter for use in stack building. All the fabricated cells had a footprint area of 4 by 4 in. (10 by 10 cm). The cells were made by progressively rolling the green electrolyte/anode bilayer (to produce thin (5 to 10 μ) electrolyte), cofiring it at elevated temperatures in air, and then applying the cathode layer by slurry painting. The bilayer rolling procedure is shown in Figure 5-1. Thin electrolyte/anode bilayers prepared in this manner were flat and showed no cracking or delamination. The bilayers had a linear firing shrinkage of about 17.5 percent. To date, the bilayer fabrication yield was 100 percent. Figure 5-2 shows, as an example, a photograph of two typical bilayers fabricated in this quarter.

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Figure 5-1. Bilayer Rolling Procedure

Figure 5-2. Photograph of Thin Electrolyte/Anode Bilayers

5.3 CONCEPTUAL DESIGN REVIEW

A conceptual design review of the technical approach for producing a 100-w MSOFC was held in December. Table 5-1 lists the meeting attendees.

The meeting centered on discussions of the electrical analysis, pressure drop calculations, stress analysis, and a baseline drawing. Also during the meeting Argonne presented test manifold concepts. The result of this meeting was an action item to develop 3-month plan to evaluate the best building blocks for use in the 100-w stack.

5.3.1 Electrical Analysis

An electrical analysis was conducted to investigate the resistive losses associated with an alternative MSOFC configuration comprising anode, electrolyte, cathode, and interconnect plates, and anode and cathode bars. The schematic for this configuration is shown in Figure 5-3, later in this discussion. An introduction, covering the need for the analysis, and discussion of the method and results are presented below.

TABLE 5-1			
MEETING ATTENDANCE ROSTER			
Location: ASE		Agenda: 100-w MSOFC stack conceptual design review	
Participant's Name	Title	Company	Telephone
J. Esopa	MSOFC project test engineer	ASE	(310) 512-4334
R. Gibson	MSOFC program manager	ASE	(310) 512-3310
J. Guiheen	MSOFC project materials engineer	ASE	(310) 512-4328
J. Hart	MSOFC assistant program manager	ASE	(310) 512-3715
C. Horne	MSOFC project materials engineer	ASE	(310) 512-4757
G. Hossepian	Project engineer	ASE	(310) 512-1932
J. Kennedy	Space systems product line manager	ASE	(310) 512-3304
M. Kimble	MSOFC project electrochemical engineer	ASE	(310) 512-2281
M. Krumpelt	Chemist	Argonne	(708) 252-8520
T. Kueper	Postdoctoral associate	Argonne	(708) 252-5486
R. Kumar	Chemical engineer	Argonne	(708) 252-4342
A. MacKnight	Space systems senior project engineer	ASE	(310) 512-3307
N. Minh	MSOFC project senior engineering specialist	ASE	(310) 512-3515
C. Stancliffe	Space systems chief engineer	ASE	(310) 512-3679
J. Yamanis	Group manager	ASR&T	(201) 455-5052

5.3.1.1 Introduction

Imperative to achieving high power densities in the monolithic solid oxide fuel cell (MSOFC) is maintaining low electrical resistive losses throughout the fuel cell stack. Large resistive losses may contribute to nonuniform current fluxes with a concomitant effect on overall stack performance. Two methods of minimizing resistive losses are to select highly conductive electrical materials and to design the fuel cell stack in a manner to minimize these losses. Material selection in this case is governed primarily by the sintering and shrinkage/expansion properties and to a lesser degree by conductive properties. Hence, specifying highly conductive materials, regardless of the other material properties, is not a viable method of minimizing the resistance.

The fuel cell stack can be designed to better distribute the potential gradients across the elements, resulting in a lower area specific resistance (ASR). A region of normally high electrical resistive losses occurs in the anode bar/interconnect/cathode bar. The objective of this work is to minimize ASR across this particular region by optimizing the design. The design parameters investigated are the bar heights and

thicknesses (for each electrode) and the number of bars per inch of interconnect. The methodology for determining the optimal design will be described next.

5.3.1.2 Method

A schematic of the MSOFC, Figure 5-3, shows the location of the anode bar/interconnect/cathode (A/I/C) bar in regard to the anode/electrolyte/cathode (A/E/C) regions. The focus of the current ASR minimization is the A/I/C region, which assumes that the contribution to overall ASR due to the A/E/C regions and across the A/E/C-A/I/C bonds is negligible. What is desired is to calculate the potential gradient throughout this A/I/C region, which can then be used to determine the ASR. Since the conductivity of the anode, interconnect, and cathode is fairly constant with temperatures near 1000°C, it is necessary to analyze only a repetitive unit of the A/I/C assembly, rather than the whole unit itself. A repeating unit of the A/I/C arrangement can be defined geometrically as shown in Figure 5-4. By determining the potential gradients throughout this repeating unit only, an ASR can be calculated which represents the entire A/I/C assembly.

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Figure 5-3. Schematic of the Monolithic Solid Oxide Fuel Cell

The finite element method is well suited to analyzing a structure such as that shown in Figure 5-4. This method allows each region of the structure to be analyzed as

Cathode bar
Interconnect
Anode bar

B-25760

Figure 5-4. Three-dimensional Representation of the Repeating Unit

many discrete nodes at which the Laplacian of the potential is applied. The current flux, j (A/cm²), through the element is given as:

$$j = -\sigma \nabla E \quad (1)$$

where σ is the conductivity of the material in S/cm and E is the potential in v. Differentiating the current flux, assuming constant conductivity, gives:

$$0 = \frac{\partial^2 E}{\partial x^2} + \frac{\partial^2 E}{\partial y^2} + \frac{\partial^2 E}{\partial z^2} \quad (2)$$

which is solved at each nodal point as defined by the repeating unit discretization. A Fortran program was written to define the material properties, discretize the repeating unit, define the boundary conditions, and generate the three-dimensional nodal array. Required input to the program includes the following:

- (a) Anode conductivity, σ_a
- (b) Cathode conductivity, σ_c
- (c) Interconnect conductivity, σ_i
- (d) Bar height, h
- (e) Bar thickness, t_b
- (f) Interconnect thickness, t_i
- (g) Number of bars per inch of interconnect

Output from this program was structured into a suitable format that was then used as input to a finite element solver. This computer program, Images-Thermal, uses the Gaussian elimination method to solve the Laplacian of the potential, Equation (2), at the defined node points. Boundary conditions are a constant current density orthogonal to the upper plane of the cathode bond and to the bottom of the anode bond. Note that to maintain conservation of energy, the current density was defined as positive going into the bottom of the anode bond and negative coming out of the cathode bond. Since only the maximum voltage difference across the repeating unit is desired, the magnitude of the current density can be selected arbitrarily (10,000 amp/cm² for this work). Equation (1) can then be applied at the bottom surface of the anode bond and the upper surface of the cathode bond. A potential value also needs to be defined at any single nodal point (1000 v). All other boundaries can be treated as "insulated" due to symmetry of the repeating unit in the A/I/C assembly. Postprocessing of the results allows the potential distribution to be obtained. For calculating the ASR, the largest voltage drop across the element was used as given by:

$$ASR = \frac{\Delta V}{j(\#bars/in.)t_b} \quad (3)$$

The various input parameters investigated are listed in Table 5-2. The cathode and anode bars are assumed to be the same geometrically, and the same number of bars per inch has been applied to both anode and cathode.

TABLE 5-2	
PARAMETER RANGES FOR THE A/I/C ASSEMBLY	
Property	Value
Number of bars/inch	5, 10, 15, and 20
t_b	10, 30, and 50 mils
t_i	10 mils
h	10, 30, and 50 mils
σ_a	578 S/cm
σ_i	26.6 S/cm
σ_c	90 S/cm

5.3.1.3 Results

Table 5-3 lists the resulting ASR's for the various configurations given in Table 5-2. Note that the cases with a 50-mil-thick bar at 20 bars/inch cannot be executed since this arrangement results in a design with no flow channel. Table 5-4 contains the ASR's when all three layers are assumed to have the same conductivity as the interconnect. This arrangement represents an interconnect bar/interconnect layer/interconnect bar

TABLE 5-3

**ASR PREDICTIONS FOR VARIOUS
ANODE BAR/INTERCONNECT/CATHODE BAR ARRANGEMENTS OF THE A/I/C**

Case	#bars/in	Bar	Bar	ASR (x100) ohm-cm ²
		Thickness (mil)	Height (mil)	
1	5	10	10	7.56
2	5	10	30	7.42
3	5	10	50	8.12
4	5	30	10	4.33
5	5	30	30	3.51
6	5	30	50	3.57
7	5	50	10	2.77
8	5	50	30	2.08
9	5	50	50	2.08
10	10	10	10	1.95
11	10	10	30	2.34
12	10	10	50	2.92
13	10	30	10	0.85
14	10	30	30	0.91
15	10	30	50	1.09
16	10	50	10	0.44
17	10	50	30	0.48
18	10	50	50	0.60
19	15	10	10	0.95
20	15	10	30	1.31
21	15	10	50	1.74
22	15	30	10	0.36
23	15	30	30	0.47
24	15	30	50	0.61
25	15	50	10	0.18
26	15	50	30	0.25
27	15	50	50	0.34
28	20	10	10	0.61
29	20	10	30	0.91
30	20	10	50	1.23
31	20	30	10	0.21
32	20	30	30	0.31
33	20	30	50	0.42

TABLE 5-4

**ASR PREDICTIONS FOR VARIOUS
INTERCONNECT/INTERCONNECT LAYER/INTERCONNECT BAR ARRANGEMENTS**

Case	#bars/in	Bar		ASR (x100) ohm-cm ²
		Thickness (mil)	Height (mil)	
1	5	10	10	12.76
2	5	10	30	19.18
3	5	10	50	25.87
4	5	30	10	8.04
5	5	30	30	9.07
6	5	30	50	10.79
7	5	50	10	5.99
8	5	50	30	5.93
9	5	50	50	6.76
10	10	10	10	4.05
11	10	10	30	7.43
12	10	10	50	11.09
13	10	30	10	1.86
14	10	30	30	2.77
15	10	30	50	3.94
16	10	50	10	1.09
17	10	50	30	1.58
18	10	50	50	2.29
19	15	10	10	2.20
20	15	10	30	4.58
21	15	10	50	7.10
22	15	30	10	0.84
23	15	30	30	1.58
24	15	30	50	2.42
25	15	50	10	0.44
26	15	50	30	0.90
27	15	50	50	1.41
28	20	10	10	1.48
29	20	10	30	3.33
30	20	10	50	5.22
31	20	30	10	0.52
32	20	30	30	1.12
33	20	30	50	1.76

assembly. The ASR's from Tables 5-3 and 5-4 are shown graphically in Figures 5-5 and 5-6, respectively.

5.3.1.4 Discussion

In Figure 5-5, the ASR shows some general trends with the bar thickness and height, as well as with the number of bars/inch. The ASR decreases as the number of bars/inch increases. Except for the five bars/inch cases, the ASR also decreases as the bar height decreases and as the bar thickness increases. Over the parameter ranges investigated for this work, the lowest ASR results with 15 bars/inch, 50-mil-thick bars, and a bar height of 10 mils. This configuration results in an ASR of $0.0018 \Omega\text{-cm}^2$.

Figure 5-6 show trends similar to those in Figure 5-5. The optimal arrangement for the interconnect bar/interconnect layer/interconnect bar also occurs at 15 bars/inch, 50-mil-thick bars, and 10-mil bar heights. The predicted ASR from this configuration is $0.0044 \Omega\text{-cm}^2$.

5.3.1.5 Summary

Area specific resistances were calculated for different geometric dimensions across the anode bar/interconnect/cathode bar assembly. The optimal arrangement is predicted to be 15 bars/inch of interconnect with both anode and cathode bars being 50 mils thick and 10 mils high. A low ASR value of $0.0018 \Omega\text{-cm}^2$ resulted from this arrangement. The same arrangement was found optimal for a case where the bars are assumed to be made out of the interconnect material having the same electrical conductivity as the interconnect layer. An ASR for this case of $0.0044 \Omega\text{-cm}^2$ was predicted.

5.3.2 Pressure Drop Calculations

Pressure drop calculations were made for the plate and bar design configuration. The pressure drop was calculated for various bar heights assuming a bar thickness (width) of 50 mils and a fin density of 10 fins/inch. The pumping power required to make up the pressure loss was also calculated. Based on previous system studies, a maximum allowable pumping power of 5 percent of total cell power was established. A bar height of 27 mils was the minimum height that could be used in this configuration (50 mils thick, 10 fins/inch) to stay within the maximum pumping power restriction. Plots of pressure drop vs bar height and pumping power vs bar height are shown in Figures 5-7 and 5-8.

5.3.3 Stress Analysis

A single-cell design based on thin-electrolyte technology was analyzed for stress levels. High stress levels were found in the anode layer of the A/E/C trilayer. Better matching of the anode and cathode CTE's was found to reduce all stresses to acceptable levels.

anode cond. = 578 S/cm
inter cond. = 26.6 S/cm
cathode cond. = 90 S/cm

Figure 5-5. ASR Variations for Different Anode Bar/Interconnect Layer/Cathode Bar Arrangements

inter cond. = 26.6 S/cm

Figure 5-6. ASR Variations for Different Interconnect Bar/Interconnect Layer/Interconnect Bar Arrangements

Figure 5-7. Pressure Drop vs Bar Height

Figure 5-8. Pumping Power vs Bar Height

5.3.4 Baseline Stack Drawing

A stack drawing was prepared based on the following baseline assumptions:

- (a) 4 by 4 in. footprint
- (b) Options for thin- or thick-electrolyte technology
- (c) Electrode bar geometry
- (d) Approximately five cells required to yield 100 w
- (e) 5 screen Pt mesh current collector

Drawing PA 169476, is based on these assumptions. A manufacturing development test plan will be prepared and will be implemented beginning in January 1993 to identify the best building blocks for use in the final 100-w stack design.

The crossflow design calls for a building block of A/E/C trilayer. Trilayers based on thin (0.000197 in.) or thick (0.0025 in.) electrolyte will be used, depending on which approach proves to be better, based on manufacturing development testing. Cathode bars connect the cathode layer to the interconnect layer. Likewise anode bars connect the anode layer to the interconnect.

A bar density (bars/inch) of 10 is used. The bars may be in either the green or the sintered state. The better approach will also be evaluated during manufacturing development testing. The dimensions in the drawing are based on sintered bars. Reinforcing bars are used to seal the edges of the fuel and oxidant flow passages.

Current collectors comprising five overlapping Pt wire mesh screens with a center uptake section are bonded to interconnect plates at the top and bottom of the stack. The current collector design will also be studied as a part of the manufacturing development test. The best approach will be incorporated into the final 100-w stack design.

5.3.5 Test Manifold Design

During the meeting, Argonne presented their test manifold design; the objective of the design process was to redesign both the test fixture and the gas supply and exhaust manifold to eliminate destructive stresses on the test stack during thermal cycling and stack operation.

Three problems with the current manifold design were identified. First, the bottom and top plates and the side bars are much thicker than the fuel cell element, making the test fixtures inflexible. Second, the side bars must be machined precisely to the same height as the test stack, with slightly different stacks requiring different side bar dimensions. Third, the coefficients of thermal expansion of the fixture, cement, and stack must be carefully matched.

An alternative design which eliminates the cover plate and precise machining and employs thin manifold sections was proposed. This design is compared to the current design in Figures 5-9 and 5-10.

Current Design of Test Fixture

$$a_M E_M \gg a_S E_S$$

$$\epsilon_S \sim \epsilon_{TH}$$

STACK SUBJECTED TO
ALL MISMATCH STRAIN

Alternative Design To Reduce Stress

$$a_M E_M < a_S E_S$$

$$\epsilon_S \sim 0$$

B-25763

Figure 5-9. Alternative Design Reduces Stress

PRESENT DESIGN

ALTERNATIVE DESIGN

- THIN MANIFOLD SECTIONS
- NO COVER PLATE
- NO PRECISE MACHINING NEEDED

B-25764

Figure 5-10. Comparison of Present and Alternative Design Configurations

SECTION 6
TESTING ON COAL GAS (TASK 4)

6. TESTING ON COAL GAS (TASK 4)

A performance test was performed on a single cell with the anode modified to improve its thermal expansion coefficient match. The objective of the test was to evaluate the effect of the modification on anode, thus cell, performance. The test was carried out at 1000°C with humidified hydrogen as fuel, and air as oxidant. The open-circuit voltage (OCV) of the cell was 1.06 v, and the area-specific resistance (ASR) of the cell was 0.8 ohm-cm². This ASR was consistent with the performance expected for cells with baseline anodes. Thus, it appeared that the modification had no significant effects on the anode performance.

A thin-electrolyte cell (EE388-03) was tested this quarter. The test was carried out over the temperature range of 700° to 1100°C with humidified hydrogen fuel and air oxidant. The performance curves of the cell are shown in Figure 6-1. The cell exhibited excellent open-circuit voltages (OCV's) and low area specific resistances (ASR's), as summarized in Table 6-1.

Figure 6-1. Performance Curves of Thin-Electrolyte Cell
Fuel = Humidified Hydrogen (3% H₂O), Oxidant = Air

SECTION 7

PERFORMANCE AND COST ANALYSIS (TASK 5)

7. PERFORMANCE AND COST ANALYSIS

During this reporting period, an electrical analysis was conducted on conceptual designs. A detailed description of this work was presented previously in para. 5.3.1.

SECTION 8
QUALITY DEVELOPMENT (TASK 6)

8. QUALITY DEVELOPMENT, TASK 6

8.1 STRESS ANALYSIS OF STACKS (TASK 6.2)

A single-cell stack design based on the thin-electrolyte technology discussed previously, was analyzed for stress levels. High stress levels were found in the anode layer of the A/E/C trilayer. Better matching of the anode and cathode CTE's was found to reduce all stress levels to acceptable levels.

DATE

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END