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Proceedings of the Fuel Cells '94 Contractors Review Meeting

Clifford P. Carpenter II
Manville J. Mayfield

August 1994

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

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Manville J. Mayfield

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In Memoriam

This Proceedings is dedicated to the memory of Manville J. Mayfield, who lost his life in the crash of USAir Flight 427 on September 8, 1994.

Friends and Coworkers
Office of Product Technology Management

Foreword

The Morgantown Energy Technology Center (METC) of the U.S. Department of Energy held the Fuel Cells '94 Contractors Review Meeting from August 17 to 18, 1994.

METC annually sponsors this conference to provide a forum for energy executives, engineers, scientists, and other interested parties to discuss advances in fuel cell research and development projects, to exchange ideas with private sector attendees, and to review relevant results in fuel cell technology programs. Two hundred and three attendees from industry, academia, and Government participated in the 2-day meeting.

The U.S. Department of Energy, Office of Fossil Energy, organized the conference to showcase the partnerships within the Government and with industry by seeking active participation and involvement from the Office of Energy Efficiency and Renewable Energy, the Electric Power Research Institute, the Gas Research Institute, and the Advanced Projects Research Agency. In addition to sessions on fuel cells for stationary electric power generation, sessions on the U.S. Department of Energy's Fuel Cell Transportation Program and the Department of Defense/Advanced Projects Research Agency's Fuel Cell Logistic Fuel Program were presented. The poster session held a prime time slot to facilitate discussion between industry and researchers. Also, the slides used in the presentations were included with the agenda and abstracts for reference during the conference. Attendees replied with favorable comments on all of these format changes.

This meeting contained 23 technical presentations in six lecture sessions and one poster session. METC used camera-ready manuscripts from the authors to produce these proceedings. METC neither refereed nor edited the manuscripts.

Clifford P. Carpenter II
Conference Technical Coordinator
Power Generation Technology Branch

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Session 2

Solid Oxide Fuel Cells

2.1

Tubular Solid Oxide Fuel Cell Development Program

CONTRACT INFORMATION

Cooperative Agreement	DE-FC21-91MC28055
Contractor	Westinghouse Electric Corporation 1310 Beulah Road Pittsburgh, Pennsylvania 15235-5098 (412) 256-2125
Contractor Program Manager	Mr. Emerson R. Ray
METC Project Manager	Ms. Diane T. Hooie
Period of Performance	April 1, 1991 to November 30, 1995

Schedule and Milestones

FY1994 Program Schedule

	O	N	D	J	F	M	A	M	J	J	A	S
Market/User Assessment Studies	—	—	—	—	—	—	—	—	—	—	—	—
Reference Design	—	—	—	—	—	—	—	—	—	—	—	—
Base Cell Technology	—	—	—	—	—	—	—	—	—	—	—	—
100 Cm Porous Support Tube (PST) Cell Dev.												
100 Cm Air Electrode Supported (AES) Cell Dev.	—	—	—	—	—	—	—	—	—	—	—	—
Alternate Process Development	—	—	—	—	—	—	—	—	—	—	—	—
100 to 200 Centimeter Cell Development	—	—	—	—	—	—	—	—	—	—	—	—
Process Scale-up/Optimization/ Cost Reduction	—	—	—	—	—	—	—	—	—	—	—	—
Facility Construction/Field Test Gen. Support	—	—	—	—	—	—	—	—	—	—	—	—
Long Cell Test Station Design and Construction	—	—	—	—								
Cell and Bundle Testing	—	—	—	—	—	—	—	—	—	—	—	—
Generator Development	—	—	—	—	—	—	—	—	—	—	—	—
Joint Field Unit Module Fabrication	—	—	—	—	—	—	—	—	—	—	—	—
Joint Field Unit Gen./System Fab./Test	—	—	—	—	—	—	—	—	—	—	—	—

OBJECTIVES

This paper presents an overview of the Westinghouse Solid Oxide Fuel Cell (SOFC) development activities and current program status. Our goal is to develop a cell that can operate for 50,000 to 100,000 hours. Progress toward this goal will be discussed and test results presented for multiple single cell tests which have now successfully exceeded 48,000 hours of continuous power operation at temperature. Curves are presented which demonstrate the improved cell performance and life obtained to date.

BACKGROUND INFORMATION

Solid oxide fuel cells readily conduct oxygen ions from an air electrode (cathode), where they are formed, through a solid electrolyte to a fuel electrode (anode). There they react with carbon monoxide (CO) and hydrogen (H_2) contained in the fuel gas to deliver electrons and produce electricity. Solid oxide fuel cells are highly efficient devices that convert chemical energy into electricity at atmospheric or elevated pressures and at a temperature of approximately 1000°C. They produce electricity using a variety of fossil fuels such as coal derived fuel gas, reformed natural gas, or reformed distillate fuel. The temperature of the exhaust gases from the cells varies between 600 to 900°C, a temperature which is attractive for cogeneration applications or for use in bottoming cycles for all-electric power plants.

The state-of-the-art tubular SOFC (Figure 1) developed by Westinghouse features a porous air electrode made of strontium-doped lanthanum manganite. A gas-tight electrolyte of yttria-stabilized zirconia (approximately 40 microns thick) covers the air electrode, except in a strip about 9 mm wide along the entire active cell length. This strip of exposed air electrode is covered by a thin, dense, gas tight layer of magnesium-doped lanthanum chromite. This layer, termed the cell interconnection, serves as the electric contacting area to an adjacent cell or to a power contact. The fuel electrode is a nickel-zirconia cermet and covers the electrolyte surface except in the vicinity of the interconnection.

For operation, air is introduced to the inside of the fuel cell through an air injector tube. The air, discharged from the injector tube near the closed end of the cell, flows through the annular space formed by the cell and its coaxial injector tube. Fuel flows on the outside of the cell. Typically, 85% of the fuel is electrochemically utilized (reacted) in the active fuel cell section. The gas-impervious electrolyte does not allow nitrogen to pass from the air side to the fuel side, hence the fuel is oxidized in a nitrogen free environment, averting the formation of NOx. At the open end of the cell, the remaining fuel is reacted with the air stream exiting the cell, thereby providing additional useful heat.

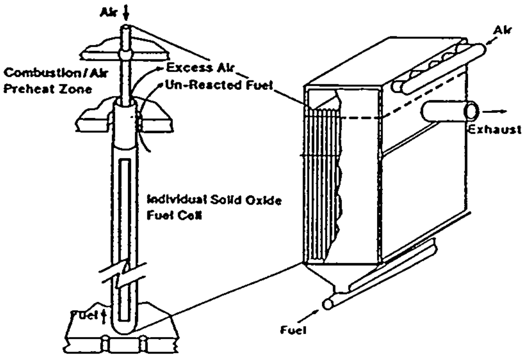

Figure 1. SOFC Module

Because of the high generator temperature and because water vapor evolves on the anode side of the cell, reformation of natural gas and other hydrocarbon fuels can be accomplished within the generator, thus eliminating the need for an external reformer.

To construct an electric generator, individual cells are "bundled" into an array of series-parallel electrically connected cells forming a mechanically stress forgiving structure that is a basic generator building block. The individual bundles are connected in series to build generator voltage and to form submodules. The parallel electrical connection of the cells within a bundle enhances generator reliability. Submodules are further combined in series to form the generator module.

The performance of the basic module design has been demonstrated by the operation of nine generators, including those operated under field test conditions, presented in Table 1.

Table 1.
Westinghouse SOFC Generators

System	Generator Rating	Number of Generators	Total Cells	Cell Length	Date
DOE	5 kW	1	324	30	1986
Osaka Gas	3 kW	2	288	36	1987
Tokyo Gas	3 kW	1	144	36	1987
GRI	3 kW	1	144	36	1988
DOE	20 kW	1	576	50	1990
The UTILITIES	25 kW	1	1152	50	1992
JGU	25 kW	1	1152	50	1992
SCE	20 kW	1	576	50	1994
ARPA*	30 kW	1	576	50	1995

*in production.

Scaleup to multi-hundred kW and multi-MW commercial modules is the next step, and is now underway. For larger applications, modules rated at several MW are planned. These modules, shown in Figure 2, would then be arranged in the power block concept shown in Figure 3. Any number of power blocks could then be installed to satisfy the specific requirement for large plants. Conceptual design studies have confirmed the practicality and economic feasibility of the power block concept in applications ranging up to several hundred megawatts.

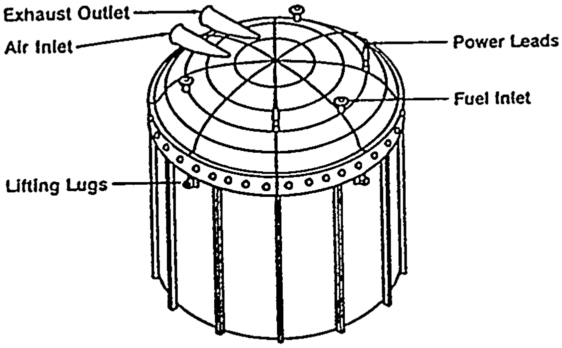

Figure 2. SOFC Module

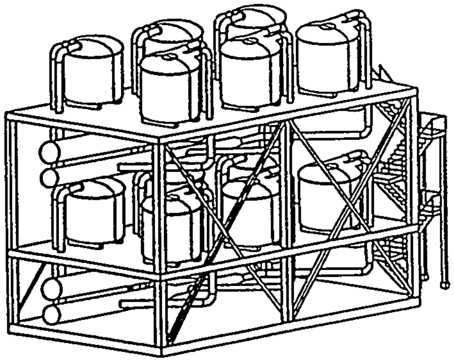

Figure 3. SOFC Power Block

PROJECT DESCRIPTION

The development of the SOFC technology is being advanced under a five-year cooperative agreement between DOE-Morgantown Energy Technology Center, Ms. Diane T. Hooie, Program Manager, and Westinghouse. This program, extending from 1991 through 1995, is focused on continuing to develop, improve, and scale up the tubular SOFC technology. The primary objective of the program is the design, fabrication, and testing of multi-hundred kilowatt and multi-megawatt SOFC generators, both at Westinghouse and at customer sites. Testing at customer sites will provide operational feedback, field experience, and greater awareness of SOFC technology.

The Westinghouse goal is to develop a cell that can operate for 50,000 to 100,000 hours. To date, Westinghouse has produced and operated for extended periods several thousand cells and has systematically improved cell performance, life, and voltage stability (see Figure 4). In August 1994 two test cells each surpassed 48,000 hours of power operation. These cells are the longest operating SOFCs in the world. Other long running cells have been shut down intentionally for detailed examination in order that improvements can be made in future cells. Still other advanced cells have been under test for up to three years with degradation rates as low as 0.5% per 1000 hours.

Successful commercialization of SOFC technology will depend on the ability to produce cost competitive systems that can operate for five to ten years before replacement of the SOFC module(s). Westinghouse is focusing its efforts on developing a thorough understanding of the parameters that limit cell and generator lifetime in order that new processes and/or materials, as may be needed, can be developed. Furthermore, a detailed understanding of the cost of manufacturing cells and generators is being pursued so that more cost effective processes and procedures can be implemented, and mass production facilities designed.

To enhance economic viability, Westinghouse is working to reduce the manufacturing costs of the cells and generators and to increase the power output per cell. Current programs are aimed at producing longer cells and increasing power output per unit cell length so as to contribute to improved power plant economics. Over the past few years Westinghouse has focused on increasing the active length of the SOFC from 30 cm (in pre-1986 cells) to 100 cm with the results being a corresponding increase in power output per cell.

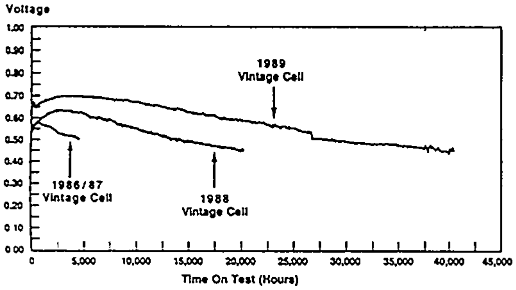

Figure 4. SOFC Development Progress Increased Cell Stability

The early technology SOFCs had a porous support tube (PST) with 2 mm wall thickness (defined as a thick-wall PST) that provided the support structure for the SOFC. Although sufficiently porous to allow oxygen ion diffusion to the air electrode, the PST still impedes diffusion and thus reduces performance. Efforts were successful in first reducing the PST wall thickness and ultimately in eliminating it altogether (see Figure 5). To compensate for the PST, the air electrode thickness was increased to provide structural support for the cell. This new air electrode supported (AES) cell has improved cell efficiency and power production. The combined effects of increasing the cell length from 30 cm to 100 cm and eliminating the PST results in a six-fold increase in cell power output. Longer cells of 1.5 to 2 meters long are currently being produced and evaluated to further improve SOFC economics, particularly for large central station electric utility power plants. Studies of advanced cells are also being pursued.

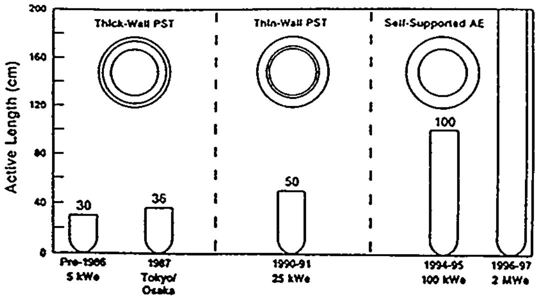

Figure 5. Evolution of Solid Oxide Fuel Cell Design

Construction of a 28,000 sq. ft. Pre-Pilot Manufacturing Facility (PPMF), located near Pittsburgh, provided the opportunity to move the technology from a laboratory environment to a manufacturing environment, and is a major step in the commercialization program of Westinghouse. It enables the processes and quality control programs to be put in place that will be required to commercialize the technology during the 1990's. A larger capacity facility is being designed and equipment installation will begin late in 1994.

Fabrication of the multi-kilowatt generator (MKG), rated at 20 kW, and containing five hundred and seventy-six 50 cm long cells was the first generator module completed at the PPMF. Testing of the MKG was successfully started in November 1990 and continued for 1750 hours on pipeline natural gas, 900 hours on hydrogen, and an additional 750 hours on naphtha. NOx levels were measured at <0.5 ppm in the generator exhaust. The design, fabrication, and testing of the MKG was a primary objective of the development contract with DOE during the late 1980's and early 1990's. Subsequently, the MKG generator module has been replicated and used in 25 kW SOFC customer test units (see Figure 6), the subject of a companion report by Dr. Stephen Veyo.

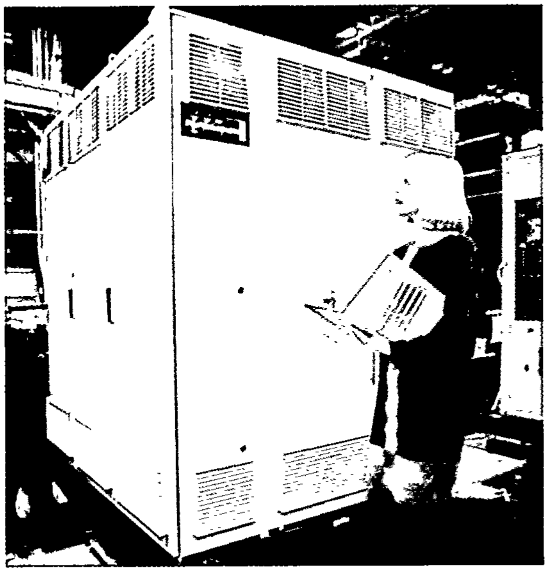

Figure 6. 20 kW SOFC Generator System

The next larger SOFC generator, rated at 100 kW to 300 kW is in the design stage. It will incorporate 150 cm AES cells and internal reforming of natural gas. This module design will represent a sub-module of the Megawatt Class generator system. A number of sub-megawatt class generator systems are planned and will undergo field tests at various other sites around the world.

RESULTS

In parallel with the SOFC technology development program, Westinghouse has performed numerous application studies to ensure that its development programs will result in a product that can satisfy the needs of various market sectors in the late 1990's and beyond. These studies help to define design requirements and establish the performance, economic, and development goals that the SOFC technology must achieve to reach commercialization. These studies are an important part of the Westinghouse Commercialization Plan for the SOFC technology.

SOFCs, with their high temperature exhaust, high efficiency, low emissions, fuel flexibility, and modularity, can be used in a broad spectrum of applications. Electric utilities have identified applications for the SOFC technology that include multi-megawatt size distributed electric power plants located near load centers, repowering of existing units, and large central station all-electric power plants. The gas utilities' interest in the SOFC technology is primarily for on-site power generation for commercial and small industrial applications.

SUMMARY

A number of significant developments and accomplishments have been realized during the past two years of the tubular SOFC program:

- Single and multi-cell SOFCs have operated for more than 48,000 hours in cell tests at power.
- Improved cell voltage stability (as low as 0.5% per 1000 hours degradation rate) has been achieved in single cell tests.
- Two 25 kW SOFC customer test units were delivered in 1992.
- A 20 kW SOFC system has been manufactured and delivered to Southern California Edison (SCE).
- The porous support tube has been eliminated, thus reducing cell cost and increasing power output per cell.
- Fuel flexibility was demonstrated in a 20 kW SOFC module that operated on natural gas for 1750 hours, naphtha for 750 hours, and hydrogen for an additional 900 hours.
- Economically competitive system performance was confirmed in application studies of numerous types of SOFC plants: natural gas fueled cogeneration units of 50 kW to 20 MW and distributed power plants of 10-20 MW; 300 MW central stations fueled by natural gas and coal-derived fuel gas; and an existing steam turbine plant repowered by SOFCs.
- A contract award has been received from DOD-ARPA concerning a program to test a tubular SOFC generator on logistic fuels (No. 2 fuel oil and JP-8).

FUTURE WORK

The existing Cooperative Agreement between Westinghouse and the Department of Energy was initiated in April, 1991 and will continue through November 1995. This five year program includes considerable financial contributions from Westinghouse, from domestic and foreign utilities, and from other agencies, in addition to the U.S. Department of Energy. Under this program Westinghouse will continue development of the tubular solid oxide fuel cell technology and generator systems and plans for field testing of multi-hundred kilowatt and multi-megawatt rated generators during the mid to latter-1990's, The ultimate objective of this program is to develop tubular SOFC technology to the point of acceptable risk for private sector commercialization.

Development of the tubular solid oxide fuel cell technology is being supported by the Westinghouse Electric Corporation, the United States Department of Energy (DOE-METC), and various utility and industry sources. The Cooperative Agreement between Westinghouse and DOE is administered by the Morgantown Energy Technology Center (METC).

2.2

Tubular Solid Oxide Fuel Cell Demonstration Activities

CONTRACT INFORMATION

Contract Number	DE-FC21-91MC28055
Contractor	Westinghouse Electric Corporation 1310 Beulah Road Pittsburgh, Pennsylvania 15235-5098 (412) 256-2125
Contractor Project Manager	Mr. Emerson R. Ray
Principal Investigator	Dr. Stephen E. Veyo
METC Project Manager	Ms. Diane T. Hooie
Period of Performance	April 1, 1991 to November 30, 1995

CONTRACT INFORMATION

Experimental field units are an integral part of the Cooperative Agreement between Westinghouse and the United States Department of Energy (DOE) for the development of tubular solid oxide fuel cell (SOFC) technology. The design and construction of the host systems for SOFC customer field units has been funded with customer and Westinghouse monies. DOE has participated in the customer test unit programs by providing the cells and in part, the stacks or generator modules. Test units have been operated by the customer entirely at customer expense with data shared with Westinghouse and DOE. Cells and generator modules are returned to Westinghouse at the completion of the customer test program.

OBJECTIVES

The development of a viable fuel cell electrical power generation system involves not only the development of cell and stack technology, but also the development of the overall system concept, the strategy for control, and the ancillary subsystems. The design requirements used to guide system development must reflect a customer focus in order to evolve a commercial product. In order to obtain useful customer feedback, Westinghouse has practiced the deployment with customers of fully integrated, automatically controlled, packaged solid oxide fuel cell power generation systems. These field units have served to demonstrate to customers first hand the beneficial attributes of the SOFC, to expose deficiencies through experience in order to guide continued development, and to garner real world feedback and data concerning not only cell and stack parameters, but also transportation, installation, permitting and licensing, start-up and shutdown, system alarming, fault detection, fault response, and operator interaction.

BACKGROUND INFORMATION

The deployment of customer test units began in 1986 when our first experimental field unit was exercised by The Tennessee Valley Authority (TVA). It was continued in 1987-88 with the two hydrogen and CO fueled 3 kW units deployed with the Osaka Gas Company, and the Tokyo Gas Company. Since 1992, Westinghouse has deployed several 20 kW class natural gas fueled SOFC generator modules (stacks) integrated into systems. The first of these was a 25 kW SOFC system deployed with The UTILITIES, a consortium of The Kansai Electric Power Company, Inc., the Tokyo Gas Company, and the Osaka Gas Company, and the second was a 25 kW cogeneration system deployed with the Joint Gas Utilities (JGU), a consortium of the Tokyo Gas Company, and the Osaka Gas Company. Most recently, a 20 kW SOFC system was deployed with the Southern California Edison Company. Table 1 provides a summary of the characteristics of Westinghouse SOFC field units to date.

Table 1

Time Year	Customer	Stack Rating (kW)	Stack Number	Cell Type	Cell Length (mm)	Cell Number	Oper (Hrs)	Fuel
1986	TVA	0.4	1	TK-PST	300	24	1760	H_2+CO
1987	Osaka Gas	3	1	TK-PST	360	144	3012	H_2+CO
1987	Osaka Gas	3	1	TK-PST	360	144	3683	H_2+CO
1987	Tokyo Gas	3	1	TK-PST	360	144	4882	H_2+CO
1992	UTILITIES-A	20	1	TN-PST	500	576	2595	PNG
1992	UTILITIES-B1	20	1	TN-PST	500	576	1576	PNG
1993	UTILITIES-B2	20	1	TN-PST	500	576	7064	PNG
1992	JGU-1	20	2	TN-PST	500	576	817	PNG
1994	SCE-1	20	1	TN-PST	500	576	1200+	PNG
				Future Work				
1995	JGU-2	25	1	AES	500	576	TBD	PNG
1994	SCE-2	25	1	AES	500	576	TBD	PNG
1995	ARPA	30	1	AES	500	576	TBD	DF-2/JP-8
1996	????	100	1	AES	1500	1152	TBD	PNG

PNG = pipeline natural gas
TK-PST = thick wall porous support tube
TN-PST = thin wall porous support tube
AES = air electrode supported

The generator module used in the 20 kW class SOFC customer field units is a near replica of the Multi-kW Generator (MKG) developed by Westinghouse for DOE. This module utilizes 576 cells of 500 mm active length arranged in four quadrants. Each quadrant contains eight bundles of eighteen cells. This construction is shown in Figure 1.

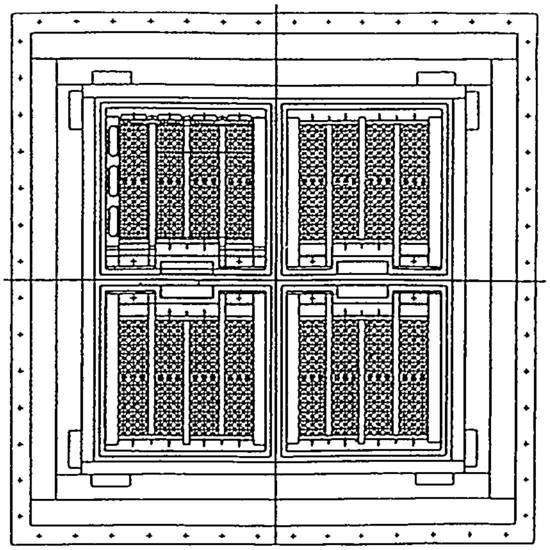

Figure 1. SOFC Generator Cross Section

PROJECT DESCRIPTION

The UTILITIES system and the SCE system are packaged dc generation systems that use the 20 kW class (MKG) SOFC stack within an enclosure. The UTILITIES system as originally installed incorporated two adjacently placed independent system units whereas the installation at SCE uses a single system. Each enclosure envelope houses the SOFC stack with integral reformers, the exhaust gas heat recovery heat exchanger (recuperator), the air supply system (centrifugal blower with motor and an electric air heater for start-up and low power operation), the fuel supply system pipeline natural gas (PNG) compressor and valves for isolation and flow modulation), a power distribution module containing instrumentation power supplies, a silicon controlled rectifier (SCR) controller for the air heater, and variable frequency motor drives, and a controls module containing a programmable logic loop controller and a high speed data acquisition system. A dissipater for the dc power is housed in a separate rack panel cabinet. System parasitic power is supplied from the grid. Drawings describing the system and a flow schematic may be found in Reference 2, last year's proceedings of this conference and in References 1, 4 and 5.

The SOFC power system is uncomplicated and simple in configuration. Only nine analog control loops are required. Start-up is accomplished using electric power for air heating and hydrogen as stack fuel until thermal conditions and adequate water evolution at the anode permit a transition to PNG fuel. The start-up process is semi-automatic requiring operator verification for state transition. After the transition to PNG fuel is accomplished, operation is fully automatic with no operator required. The transition from dormancy at ambient temperature to fully automatic operation on PNG takes eight hours. In automatic operation, the system requires set point values for delivered dc current, stack temperature, fuel utilization, and air utilization. Fuel and air flow are automatically adjusted in proportion to the dc current. The control system will automatically shut the system down in response to either an operator command or the deviation of several critical parameters outside of the permitted value range. In case of a severe fault such as control computer failure or a ground fault current or a combustible gas leak, the unit will automatically transit to EMERGENCY STOP, a state wherein the system is isolated from and purged of combustible gas, the dc current path from the SOFC is interrupted, and the only energy permitted to enter the system is control and data acquisition system power.

The 25 kW SOFC cogeneration system developed for the JGU is a similarly packaged system that used two 20 kW class (MKG) stacks connected in electrical series, delivered ac power to dissipative resistors, and employed an exhaust gas heated heat recovery steam generator placed between the high temperature and low temperature sections of a partitioned exhaust gas to process air recuperative heat exchanger. More details concerning this system may be found in references 2, 3 and 6.

RESULTS

Assembly of The UTILITIES 25 kW SOFC system was completed in November 1991 and after factory start-up and operational tests, the unit was shipped via overland truck and air freight to the Rokko Island Test Center for Advanced Energy Systems (near Kobe, Japan), operated by The Kansai Electric Power Company. Testing was initiated in February 1992, following installation and three weeks of pre-operation on-site software and hardware verification directed toward licensing.

System start-up was accomplished easily, demonstrating that automatic operation at nominally 1000°C is possible in less than eight hours after initiating the start-up process from ambient temperature. Subsequent shutdown was also achieved according to process design with the stack cooled to less than 100°C in six hours. Observed and predicted power versus current were in good agreement.

Module A operated for 2529 hours at an average output of 16.8 kW dc, achieving a maximum output of 17.7 kW dc. The system was started from ambient temperature and cooled to ambient temperature four times and as a result of various system difficulties, endured nine partial cool-downs. Testing spanned three distinct periods: from 11/29/91 to 12/13/91 at the Westinghouse factory, 2/25/92 to 5/1/92 and 6/13/92 to 7/12/92 at the Rokko Island test site. Test operation in Japan was interrupted by the need to repair a failed electric air heater which prevented a restart following an automatic stop induced by an instability in the dissipater control circuits. The final shutdown was also induced by current stability problems with the dissipater. A restart was not possible because of damaged cells and Module A was returned to the factory.

Module B operated for 1576 hours at an average output of 17.4 kW dc and achieved a maximum power of 19.2 kW dc. The system was started from ambient temperature twice and experienced two shutdowns to ambient temperature and three partial cool-downs. The final system shut down occurred during a period of operational probing for maximum output. Subsequent post- operation investigation showed that the unit was operated at excessively high fuel utilization, due possibly to an internal fuel maldistribution caused by blocked fuel distribution ports (as observed), possibly exacerbated by cell leaks. Analysis of data suggested that only one quadrant had problems. Module B was returned to the factory where a limited repair of the suspect quadrant was attempted. A subsequent restart in October 1992 was aborted because of excessive internal temperatures due to cracked cells.

Module B was rebuilt (Module-B2) with new cells and returned to Rokko Island without factory start-up. Installation and start- up took place during the last days of March 1993. Start-up was successful, but hampered by a lower than expected cell voltage and an instability in the output of the natural gas mass flow meter. Field modification of the control software during operation permitted completion of an extended start-up process. Module B2 operated for 1843 hours on PNG spanning the period from 4/1/93 to 6/17/93. Nominal output during this period was 17 kW dc. Shutdown was caused by a failure of the air supply system. Drive belts for the blower failed, and the motor bearings failed the day after belt replacement. Operation was vexed by inadequate ambient temperature compensation in the air and PNG flow meters.

A new blower and motor assembly were installed in July 1993. The system was restarted on Aug 5, 1993 without incident and operated continuously until November 10, 1993 when an automatic shutdown was induced as part of a MITI licensing inspection. Restart was accomplished without incident. On March 10, 1994 the unit was shut down voluntarily after having exceeded 7000 hours of operation on desulfurized PNG. The last 5200 hours of operation were without forced outage, the only interruption in operation occurring as part of the MITI inspection. A plot of dc power versus hours of operation is shown in Figure 2. The only item of routine maintenance required was periodic replacement of an automotive dry type air filter.

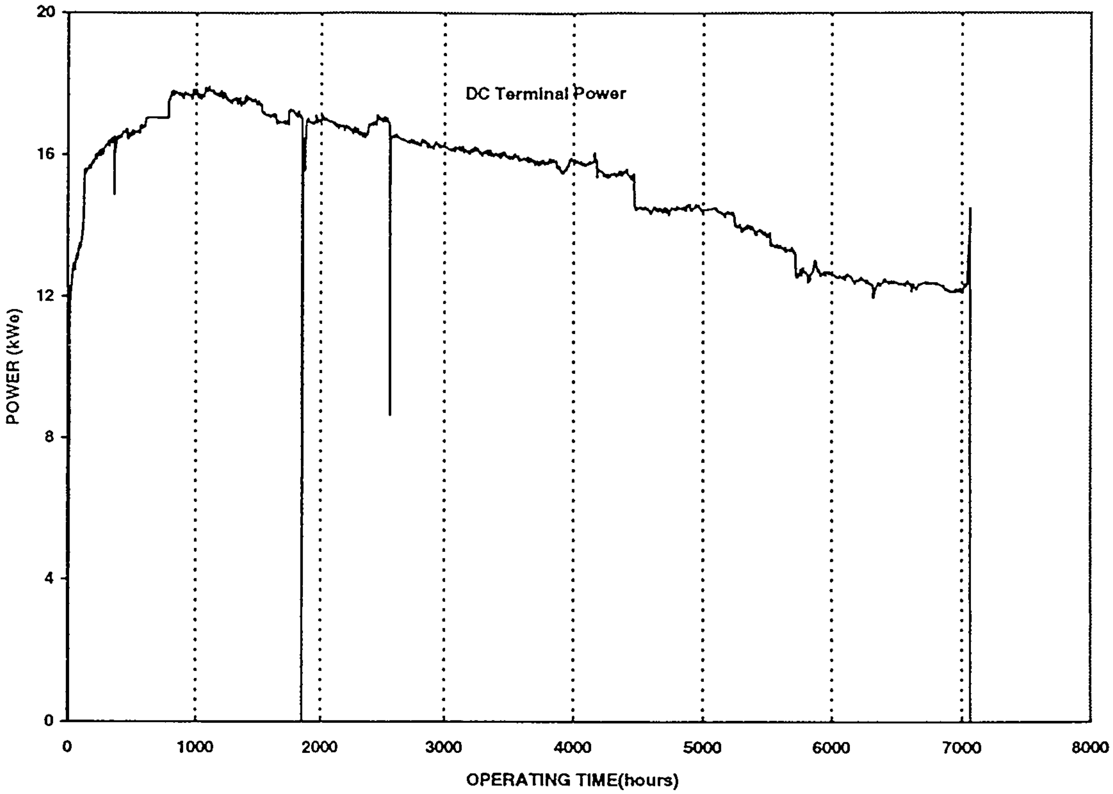

Figure 2. Power Versus Time for Module B-2 of The UTILITIES 25 kW SOFC System

The UTILITIES 25 kW SOFC system has been returned in its entirety to Westinghouse.

The JGU 25 kW SOFC cogeneration system operated in 1992 for a total of 817 hours, mostly during the factory test. The startup in Japan in October 1992 was not successful. The unit has been returned to Westinghouse for modification and repair.

A 20 kW system unit was delivered to the Highgrove Generating Station of SCE in Grand Terrace (near San Bernadino), CA in March 1994, and in late May 1994 an MKG type SOFC stack was installed. The unit was successfully started on June 7, 1994 and as of the end of July has accumulated 1200 hours of operation on pipeline natural gas. The maximum output achieved to date is 18.9 kW, with normal operation slightly in excess of 18 kW. Shortly after startup, the unit endured several short duration outages, the most memorable caused by the inadvertent cut-off of all power to SOFC systems. To date the only system problem of note appears to be a hysteresis like bi-stable behavior in the natural gas mass flow meter.

CONCLUSIONS

The UTILITIES SOFC systems and the SCE system can operate in thermally self-sustaining mode with no fuel or electrical power consumed for temperature maintenance.

Stack V-I performance is in general agreement with expectations based upon single cell tests.

Projected overall maximum system efficiency for 25 kW class SOFC machines, on a net ac/HHV basis, is about 33%.

Measured NOx emissions are less than 1 ppm.

At a prepared site with cranes and rigging, unloading and physical installation can be accomplished easily in one day.

SOFC systems can be started within eight hours, maneuvered across the load range, and stopped easily.

Operation of the SOFC systems in a non space conditioned shelter showed that instrumentation did not operate satisfactorily over the temperature range claimed by suppliers.

The UTILITIES 25 kW SOFC system is the first high temperature fuel cell power system to have been certified by MITI as in compliance with applicable regulations for fossil fueled power plants in Japan.

FUTURE WORK

Westinghouse will at a future date replace the SOFC stack in the Southern California Edison Company 20 kW class system. The replacement stack will utilize air electrode supported (AES) cell technology (no structural support tube) rather than the zirconia porous support tube type cell presently installed.

The JGU system has been returned to Westinghouse facilities where it is presently undergoing modification to enable system operation with a single stack rather than two stacks. In addition, one of the stacks from the JGU unit is being rebuilt with AES-SOFCs, re-using almost all internal components. After a factory test, the JGU system will be returned to Japan for verification testing, expected before the end of the first quarter of 1995.

In early 1995, a new AES-SOFC stack will be installed in the SCE system under the sponsorship of the Advanced Research Projects Agency of the Department of Defense with the contract being administered by the NASA Lewis Research Center. In addition to the new stack, a logistics fuel processor will be added, which will enable the SCE system to operate on diesel and jet fuel as well as natural gas.

In mid 1996 Westinghouse expects to deliver the first 100 kW SOFC system using the AES cell technology.

ACKNOWLEDGMENT

Westinghouse gratefully acknowledges the support of The United States Department of Energy--Morgantown Energy Technology Center for the continued development of the tubular SOFC. In addition, Westinghouse acknowledges the past, present, and future financial and programmatic support for the field unit test program of our commercial customers: the Tennessee Valley Authority; The Kansai Electric Power Company; the Tokyo Gas Company; the Osaka Gas Company; the Southern California Edison Company; and the Southern California Gas Company.

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2.3

Solid Oxide Fuel Cells Materials Research

CONTRACT INFORMATION

Contract Number (FTPA)	13822
Contractor	Pacific Northwest Laboratory Operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO 1830 P.O. Box 999 Richland, Washington 99352 (509) 375-3938
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Principle Investigators	Dr. Jeffry Stevenson Dr. Larry Pederson Greg Coffey
METC Project Manager	Diane Hooie
Period of Performance	April 1, 1988 to June 30, 1994

Schedule and Milestones

FY934Program Schedule

	S O N D J F M A M J J A S
Materials Development and Property Evaluation	--
Materials Synthesis	
Materials Processing and Fabrication	--
Electrochemical Processes	--

ABSTRACT

Protonic conductivity in the strontium ytterbium cerates was shown to have a large grain boundary component. Protonic conductivity in these materials requires the presence of water, which reacts with oxygen vacancies to yield hydroxyl groups that support proton conduction. Transference numbers were determined for protons, oxygen ions, and electrons as a function of temperature. Protonic conduction dominated in hydrated cerates up to 450°C. At higher temperatures, oxygen ion and electronic conductivity became progressively more important. Hydrogen could be pumped across the cerate membrane by applying a dc voltage. At 800°C, pure hydrogen fluxes greater than 1 sccm/cm^2 could be driven across the membrane.

OBJECTIVES

The purpose of this research is to develop alternative materials for solid oxide fuel cell (SOFC) interconnections and electrodes with improved electrical, thermal, and electrochemical properties. Another objective is to develop synthesis and fabrication methods for these materials whereby they can be processed in air into SOFCs. The approach is to (1) develop modifications of the current, state-of-the-art materials used in SOFCs, (2) minimize the number of cations used in the SOFC materials to reduce potential deleterious interactions, (3) improve thermal, electrical, and electrochemical properties, (4) develop methods to synthesize both state-of-the-art and alternative materials for the simultaneous fabrication and consolidation in air of the interconnections and electrodes with the solid electrolyte, and (5) understand electrochemical reactions at materials interfaces and the effects of component composition and processing on those reactions.

BACKGROUND

Because of attractive potential applications, mixed conductors in the lanthanum strontium cobalt chromium iron oxide system and protonic conductors in the strontium ytterbium cerate system are being studied. Mixed conductors, materials that conduct both ions and electrons, are potentially useful as semipermeable membranes in gas separation, as electrodes in solid oxide fuel cells, and as membrane reactors. Protonic conductors can be used as electrolytes in solid oxide fuel cells, as hydrogen sensors, in gas purification systems, and in steam electrolyzers.

Although proton conduction in inorganic solids was first reported more than two decades ago,[1] only the ABO_3-type perovskites have been shown to be sufficiently stable under conditions of high temperature and both oxidizing and reducing environments needed for practical applications such as solid electrolytes in solid oxide fuel cells.[2,3] Sites for proton conduction are believed to be created when water reacts with oxygen vacancies. Oxygen ion and electronic conduction also occurs, becoming progressively more important at elevated temperatures.[2-4]

PROTONIC CONDUCTION IN SrYbCe OXIDES

Fine and homogenous $SrCe_{1-x}M_xO_{3-\alpha}$ (M=dopant ions) perovskite powders by calcination of combustion derived precursors at <950°C. The precursors of these perovskites were obtained using exothermic redox decomposition of mixtures containing strontium acetate, ceric ammonium nitrate, ytterbium nitrate and glycine at 250°C. The perovskite powders prepared by this technique have been sintered to about 98% theoretical density at 1500°C in 12 hrs.

Four-point conductivities were determined in both dry air and in water-saturated air in the approximate temperature range of 400 to 800°C. Experimental results were fit to the expression:

$$\sigma T = C\exp(-E/kt) \qquad (1)$$

where the pre-exponential, C, is a charge carrier density and material-dependent constant, E is the total activation energy for ionic and electronic migration, k is the Boltzmann constant, and T is the absolute temperature. Activation energies and pre-exponentials determined from the conductivity data are given in Table 1. Based on these results and the data of Sherban and Nowick,[5] the optimal concentration of Yb dopant may be x=0.05 for the $SrCe_{1-x}M_xO_{3-\alpha}$ system. Overall conductivities are slightly higher than obtained previously,[12] which is attributed to higher sintered densities achieved in the present study.

Transference numbers as a function of temperature were determined by the emf response to known hydrogen and oxygen gradients. Hydrogen ion conduction accounted for essentially all of the conductivity below 400°C, as shown in Figure 1. Above that temperature, oxygen ion and electronic conduction, by inference, grew progressively more important. These results are consistent with those reported previously by Scherban and Nowick.[5]

The kinetics of water uptake by $SrCe_{0.95}Yb_{0.05}O_{3-\alpha}$ was studied over a wide range of temperatures and water partial pressures using thermogravimetry. Water adsorption typically followed a two-stage process, as is shown in Figure 2. The first, rapid uptake stage is attributed to hydration of the grain boundaries, whereas the much slower, long-term hydration is attributed to hydration of the bulk grains, which averaged 15 µm in diameter in these fully dense samples. In previous studies of water uptake by the cerates, complete hydration of the bulk was reported be reached within a few hours at temperatures even below 600°C; no contribution from grain boundaries was recognized.[6] Changes in resistance as a function of time following introduction of water followed similar trends to water uptake kinetics, indicating that grain boundaries contribute substantially to the observed conductivity. Protonic conductivity from grain boundary phases in the barium cerates have been observed previously, and attributed to a separate grain boundary phase.[16] In the strontium cerates, no microscopic evidence for a grain boundary phase or segregation could be found.

By applying a voltage across the $SrCe_{0.95}Yb_{0.05}O_{3-\alpha}$ sample, hydrogen could be pumped through the electrolyte, even at high temperatures where the transference number for hydrogen was smaller than 0.5. Hydrogen flux rates greater than 1 $sccm/cm^2$ could be pumped through a 2 mm thick disk at 800°C. Introduction of deuterium oxide in the place of water resulted in deuterium labelling of the molecular hydrogen product, even at temperatures too low to result in bulk phase hydration. This behavior again is consistent with grain boundaries contributing a significant share of the total protonic current.

In summary, fully dense, phase-pure $SrCe_{0.95}Yb_{0.05}O_{3-\alpha}$ has been prepared, which is suitable for use as a proton-conducting electrolyte, as a hydrogen pump, and as a hydrogen sensor. Thermogravimetric, permeation, and electrical property characterization results are consistent with the conclusion that the grain boundaries contribute substantially to the protonic current in these materials.

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Table 1. Activation Energies, E, and Pre-Exponentials, C, for Conductivity of $SrCe_{1-x}Yb_xO_{3-\alpha}$ Measured in Dry Air and Water-Saturated Air

Atmosphere	Yb Content	E(eV)	C (S/cm)
dry air	x=0.05	0.79	3.3×10^4
water-saturated	x=0.05	0.62	3.4×10^3
dry air	x=0.10	0.85	7.0×10^4
water-saturated	x=0.10	0.64	3.8×10^3
dry air	x=0.15	0.88	3.6×10^4
water-saturated	x=0.15	0.67	2.7×10^3

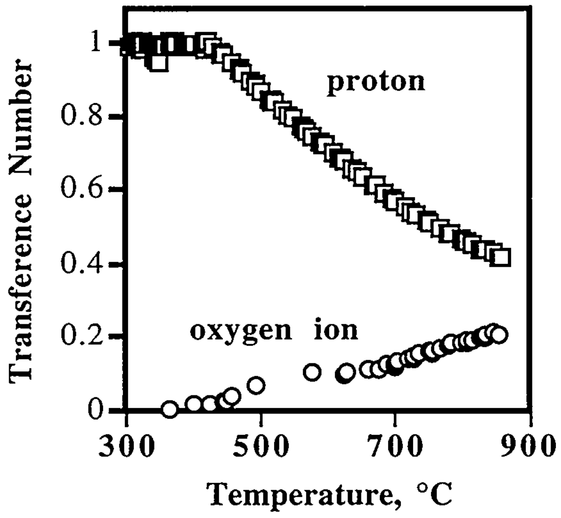

Figure 1. Transference numbers as a function of temperature for $SrCe_{0.95}Yb_{0.05}O_3$.

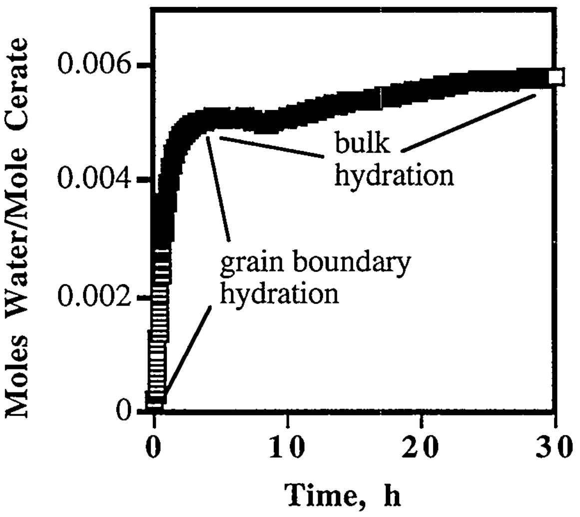

Figure 2. Kinetics of water uptake at 800°C for $SrCe_{0.95}Yb_{0.05}O_3$ by thermogravimetry.

2.4

Advances in Research for Solid Oxide Fuel Cells

CONTRACT INFORMATION

Contract Number	49638
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Co-Investigator	Timothy Kueper and Rajiv Doshi
METC Project Manager	William C. Smith
Period of Performance	October 1, 1993 to July 1, 1994

OBJECTIVE

Solid oxide fuel cells are attracting considerable interest among industrial organizations wanting to position themselves in a potentially important technology of the future. More than a dozen new organizations worldwide have begun SOFC development in the last few years.

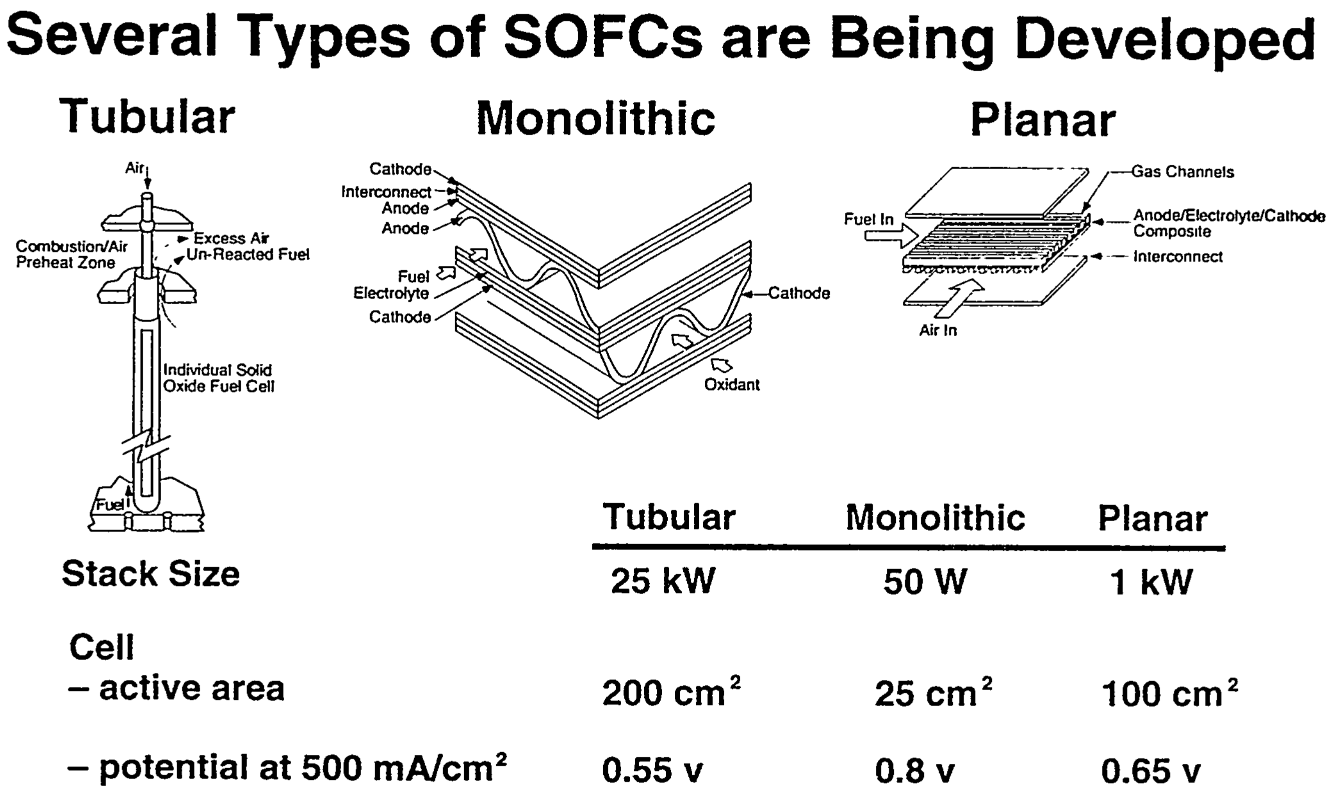
BACKGROUND INFORMATION

Most of this R&D activity is in the planar technology, because it represents a good compromise between the proven but IR-limited tubular configuration and the high-performance but difficult-to-fabricate monolithic structure. Table 1 lists many but not all the organizations engaged in research on the three types of SOFCs, and Fig. 1 summarizes the status of this technology.

Table 1. R&D Activity on SOFCs

Tubular	Monolithic	Planar
Westinghouse Russia	Allied Signal MHI	Cerametec Z-Tek TMI Siemens Dornier Sanyo Murata Tonen Tepco Fuji MHI

The planar configuration, in addition to being more easily fabricated, can also be adapted to metallic bipolar plates. However, to limit the effects of oxygen on the metal, the fuel cell operating temperature needs to be lowered from the typical



Several Types of SOFCs are Being Developed

Tubular **Monolithic** **Planar**

	Tubular	Monolithic	Planar
Stack Size	25 kW	50 W	1 kW
Cell			
– active area	200 cm^2	25 cm^2	100 cm^2
– potential at 500 mA/cm^2	0.55 v	0.8 v	0.65 v

1000°C to about 800°C; and to compensate for the lower conductivity of the electrolyte at 800°C, the thickness of the electrolyte needs to be reduced.

PROJECT DESCRIPTION

Challenges

The challenges of developing the planar cell configurations are finding high-temperature edge and manifold seal materials that will make very flat ceramic trilayers of sufficiently large area, and minimize contact resistances in stacks of cells. Also, decreasing the operating temperature requires development of reliable thin-film fabrication methods for the electrolyte, and finding a metal with good oxidation resistance and a thermal expansion coefficient well matched with the different cell components. Finally, toughness and a thermal stress tolerance of stacks need to be improved.

RESULTS

Advances

Several industrial developers in Japan, Europe, and the U.S. are operating SOFC stacks of about one-half of a kilowatt power output. Typically, these stacks have an active area of 100-150 cm^2, and 10-20 cells running at 300-600 mA/cm^2 current density [1]. The cell potentials are still not stable enough but decline by about 10% in the first 1000 h of operation.

Several types of cell degradation mechanisms have been identified: coarsening of nickel-based anodes, migration of chromium from the interconnect material into the cathode, increasing contact resistance between the air electrode and metallic interconnect plates, and failure of seals.

Most of the planar stacks are made by tape casting and sintering the electrolyte, and then screen printing electrodes on both sides. The trilayers are then stacked with lanthanum chromite or metallic bipolar plates by using proprietary contact cements. Alternative fabrication methods are to calender anode/electrolyte bi-layers or deposit thin electrolyte films onto sintered anodes.

Argonne National Laboratory has been exploring new sealant and bipolar plate materials. Advances in these specific areas are discussed elsewhere in this report.

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Poster Session

P1 Alternative Materials for Solid Oxide Fuel Cells

CONTRACT INFORMATION

Contract Number (FTPA)	13822
Contractor	Pacific Northwest Laboratory Operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO 1830 P.O. Box 999 Richland, Washington 99352 (509) 375-3938
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Principle Investigators	Dr. Timothy R. Armstrong Dr. Jeffry W. Stevenson
METC Project Manager	Diane Hooie
Period of Performance	April 1, 1989 to June 30, 1994
Schedule and Milestones	

FY94 Program Schedule

	S O N D J F M A M J J A S
Materials Development and Property Evaluation	--
Materials Synthesis	
Materials Processing and Fabrication	--
Electrochemical Processes	--

OBJECTIVES

The purpose of this research is to develop alternative materials for solid oxide fuel cell (SOFC) interconnections and electrodes with improved electrical, thermal, and electrochemical properties. A second objective is to develop synthesis and fabrication methods for these materials whereby they can be processed in air into SOFCs. The approach is to (1) develop modifications of the current, state-of-the-art materials used in SOFCs, (2) minimize the number of cations used in the SOFC materials to reduce potential deleterious interactions, (3) improve thermal, electrical, and electrochemical properties, (4) develop methods to synthesize both state-of-the-art and alternative materials for the simultaneous fabrication and consolidation in air of the interconnections and electrodes with the solid electrolyte, and (5) understand electrochemical reactions at materials interfaces and the effects of component composition and processing on those reactions.

BACKGROUND INFORMATION

Solid oxide fuel cells (SOFCs) continue to develop as promising, clean, and efficient technology for the direct conversion of hydrogen and fossil fuels to electrical energy. The standard materials utilized in state-of-the-art designs are: the air electrode, La(Sr)MnO_3; the fuel electrode, ZrO_2•Ni cermet; the interconnection, La(Mg)CrO_3 or La(Sr)CrO_3; and the electrolyte, Y_2O_3-stabilized ZrO_2. The required combination of these different materials in state-of-the-art SOFCs poses several materials fabrication and performance related problems.

New alternative materials with improved properties that can be more easily fabricated into fuel cells are needed for the further development of cost efficient and improved performance SOFCs. An important key is the development of a SOFC interconnection material that can be sintered in air to a high density at temperatures less than 1500°C. It should have a good thermal expansion match with, and intimately bond to, both the air and fuel electrodes, without deleterious interactions at the interfaces. At the present time this goal has not been attained with the state-of-the-art $La_{1-x}Sr_xCrO_3$ interconnections.

PROJECT DESCRIPTION

The overall approach for this research and development is to:

- Minimize the number of cations in the electrode, electrolyte, and interconnection by developing yttrium compounds, such as Y(Ca)CrO_3 as the interconnection, and Y(M')MnO_3 as the air electrode.

- Develop advanced synthesis and fabrication processes for air sintering, below 1500°C, of chromite interconnections through (1) the use of sintering aids; and (2) the synthesis of sub-micrometer powders.

- Establish methods for the simultaneous processing and consolidation of air-sinterable powders.

- Electrochemically evaluate interface reactions (in reproducible and controlled laboratory tests) for both the alternate and state-of-the-art materials and cell components developed under this program.

- Evaluate the chemical reactivity and interdiffusion effects that take place between the various fuel cell components: electrolyte/cathode, interconnect/cathode, and interconnect/anode.

This paper summarizes a comprehensive study that assessed the effect of ambient oxygen partial pressure on the stability of air-sinterable chromites and the sintering behavior of doped lanthanum manganites.

RESULTS

Stability of Chromite Interconnects

The interconnect in a solid oxide fuel cell must be electrically conducting and simultaneously stable in both oxidizing and reducing environments at high temperature. These extreme conditions restrict the viable candidate materials to lanthanum or yttrium chromites. While chromites do maintain their perovskite structure when exposed to the reducing environment on the anode side of an operating SOFC, they experience a loss of lattice oxygen which results in a decrease in electrical conductivity and an increase in volume.[1,2]

The effect of atmosphere on the physical dimensions of doped lanthanum chromites was studied by measuring the linear expansion of sintered specimens as a function of ambient oxygen partial pressure (P(O_2)) at several relevant temperatures. After initially equilibrating the specimens in air, the P(O_2) was decreased stepwise using a CO_2/Ar-4% H_2 buffered gas system. The compositions studied were $La_{0.85}Ca_{0.15}CrO_3$ (referenced hereafter as LCC-15), $La_{0.80}Ca_{0.20}CrO_3$ (LCC-20), $La_{0.75}Ca_{0.25}CrO_3$ (LCC-25), and $La_{0.70}Ca_{0.30}CrO_3$ (LCC-30). Figures 1 and 2 show linear expansion as a function of P(O_2) at three temperatures for LCC-20 and LCC-30, respectively. As the P(O_2) was decreased, the physical dimensions of the sintered specimens remained essentially constant until a critical P(O_2) was reached, after which the specimens expanded with decreasing P(O_2). Increases in temperature caused the onset of expansion to occur at higher values of P(O_2). Also, the magnitude of the linear expansion at a given P(O_2) increased with increasing temperature. Similar behavior was observed for LCC-15 and LCC-25.

Figure 3 shows linear expansion at 1000°C for all four compositions. Increasing Ca content caused the onset of expansion to occur at higher P(O_2) and, for a given value of P(O_2), resulted in an increase in expansion. At 10^{-18} atm P(O_2), linear expansion ranged from 0.12% for LCC-15 to 0.37% for LCC-30. This decrease in stability with increasing Ca content suggests that, for interconnect applications, Ca doping of lanthanum chromite should be no greater than is necessary in order to obtain adequate sinterability and electrical conductivity.

The observed expansion can be correlated to the formation of oxygen vacancies as reported by Anderson.[1,2] A likely explanation lies in the fact that the removal of oxygen anions from the perovskite lattice eliminates the electrostatic attraction between those anions and the surrounding cations and causes increased electrostatic repulsion between those unshielded cations. The net result is an increase in lattice cell dimensions, and, therefore, an increase in specimen volume.

Microstructures of sintered LCC specimens were examined by SEM after exposure to oxidizing environments (control), reducing environments, or sequential exposure to reducing and oxidizing conditions. Control specimens (annealed in air at 1000°C) exhibited no discernible phase segregation or impurity phases at the grain boundaries. Similarly, specimens exposed to a highly reducing atmosphere (10^{-18} atm P(O_2) at 1000°C) showed no discernible phase segregation. However, the highest doped chromite, LCC-30, did exhibit a change in the fracture mechanism. Prior to reduction, fracture was intergranular in nature, whereas in reduced specimens an intragranular fracture mechanism was observed, suggesting that the grains were weakened as a result of the substantial oxygen deficiency and lattice expansion that these samples experienced.

Other microstructural features were noted in the specimens which experienced sequential exposure to reducing and oxidizing environments. In particular, in LCC-30 (after reduction and re-

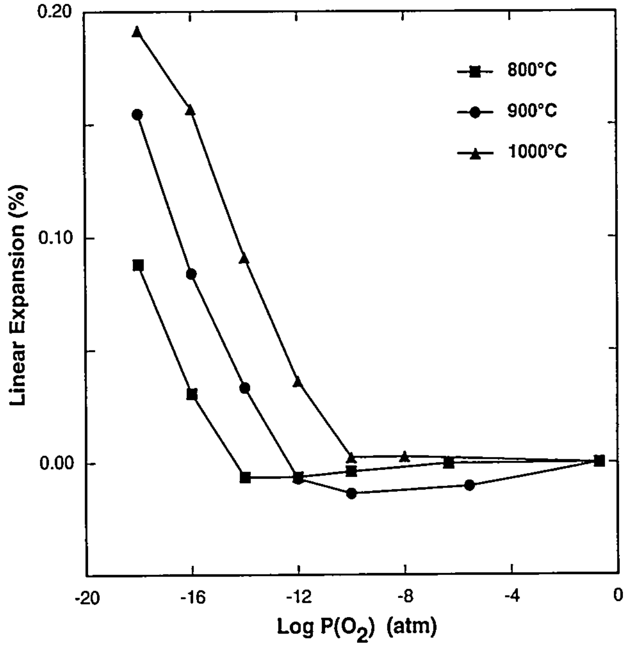

Fig. 1. Linear Expansion vs. Log P(O_2) for LCC-20 at the Indicated Temperatures

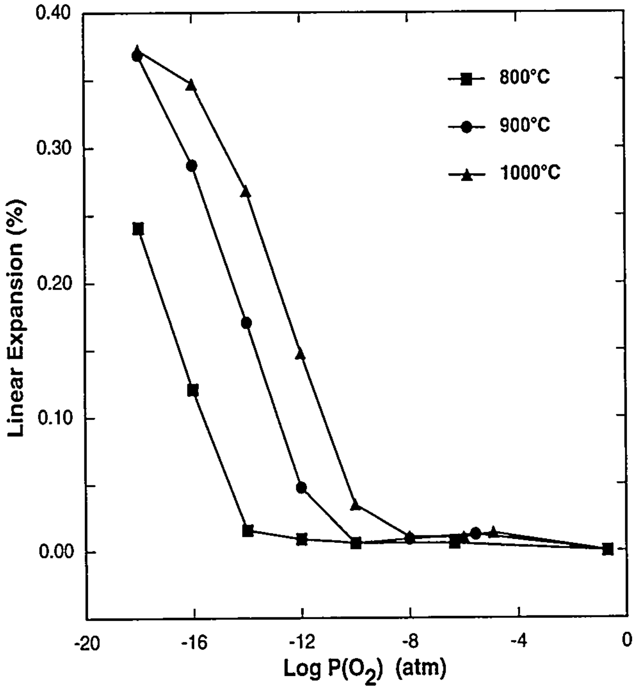

Fig. 2. Linear Expansion vs. Log P(O_2) for LCC-30 at the Indicated Temperatures

oxidation at 1000°C) a Ca and Cr enriched second phase (possibly calcium chromate or an amorphous phase of similar composition) was apparent. Also, while the fracture mechanism after reduction was found to be intragranular in nature, upon re-oxidation the mechanism was intergranular, probably due to the presence of a weak interfacial phase (i.e., calcium chromate or an amorphous phase) at the grain boundaries.

Further evidence for the presence of calcium chromate was found by thermogravimetric analysis of LCC-30 during oxidation-reduction cycles. As expected, a weight loss was measured during reduction due to the removal of lattice oxygen. Upon re-oxidation, a temporary net weight gain occurred in which the specimen weight exceeded its original oxidized weight. With further annealing at temperature in air, the specimen weight decreased to its original value. This net weight gain is consistent with the formation of calcium chromate, in which the Cr cations are oxidized to the +6 valence state. The disappearance of the weight gain with time may result from the decomposition of the calcium chromate phase as the Ca and Cr ions are re-incorporated into the primary perovskite structure, during which process the hexavalent Cr ions would be reduced to the tri- and/or tetravalent state.

Sintering of Doped Lanthanum Manganite

SOFC cathodes must endure severe environmental conditions during operation. For example, the cathode material must be stable at 1000°C in air with high electronic conductivity and a thermal expansion compatible with the YSZ electrolyte. Chemical interaction with the electrolyte and interconnect materials must be minimal. Also, the cathode material must have a highly porous microstructure so that molecular oxygen can diffuse through the cathode to the cathode/electrolyte interface. Given these requirements, it is not surprising that the number of likely candidate materials for SOFC cathodes is very limited. The current preferred material for SOFC applications is lanthanum manganite, $LaMnO_3$, doped with an appropriate amount of Ca or Sr. This material offers adequate electrical conductivity, reasonable thermal expansion match to YSZ, and stability in the SOFC cathode operating environment.[2-5]

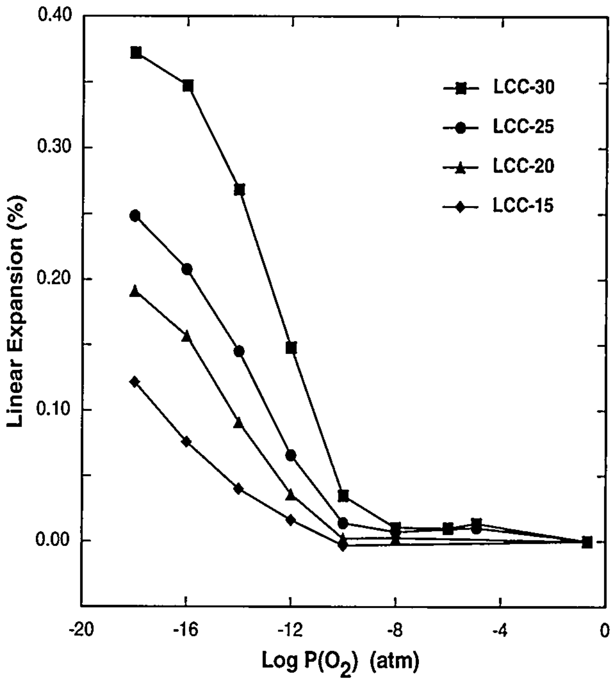

Figure 3. Linear Expansion vs. Log $P(O_2)$ for the Indicated Compositions at 1000°C

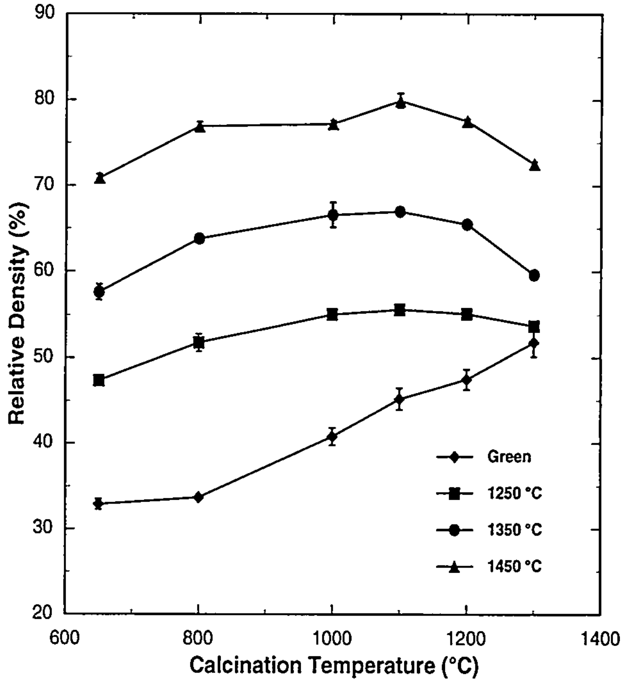

Figure 4. Sintered Density vs. Calcination Temperature for LSM-24

The following compositions were selected for this sintering study due to their close thermal expansion match to YSZ: $(La_{0.84}Sr_{0.16})_xMnO_3$ [referenced hereafter as LSM-16], $(La_{0.76}Sr_{0.24})_xMnO_3$ [LSM-24], $(La_{0.80}Ca_{0.20})_xMnO_3$ [LCM-20], and $(La_{0.70}Ca_{0.30})_xMnO_3$ [LCM-30], with x (i.e., A/B cation ratio) = 0.95, 0.98, 1.00, 1.02, and 1.05. These compositions were synthesized using the glycine-nitrate process,[6] after which they were "pre-calcined" at 650°C for 0.5 hour. The manganite powders were then calcined at a variety of temperatures for 1 hour to determine the effect of calcination temperature on sintering behavior.

The effect of calcination temperature on sintered densities of LSM-24 and LCM-30 (sintered for one hour at the indicated temperatures) is shown in Figures 4 and 5, respectively. Green densities of the dry-pressed compacts are also shown in these figures. The green densities of compacted pre-calcined powders were very low. These low packing densities were caused by extensive particle agglomeration resulting from the synthesis method, which yields nanometer-sized particles which are partially sintered into an open, high surface area network. The pre-calcined manganite powders typically had a bimodal distribution consisting of approx. 15 vol.% 1-2 μm agglomerates and approx. 85% 10-100 μm agglomerates. Coarsening of the powders by calcination reduced the powder surface areas and allowed for improved particle packing densities, so that green density increased with increasing calcination temperature.

Sintered densities were also affected by the calcination temperature. For low calcination temperatures, sintered densities increased with increasing calcination temperature, presumably due to the higher green densities. Sintered densities reached a maximum for powders calcined between 800 and 1200°C, but usually remained below 90% of theoretical due to the agglomerated condition of the powders. For powders calcined above 1200°C, sintered densities tended to decrease substantially, presumably due to the formation of hard agglomerates and the decreasing driving force for densification which occurs with increasing particle size. Figures 4 and 5 also show that, for any given calcination temperature, sintered density increased

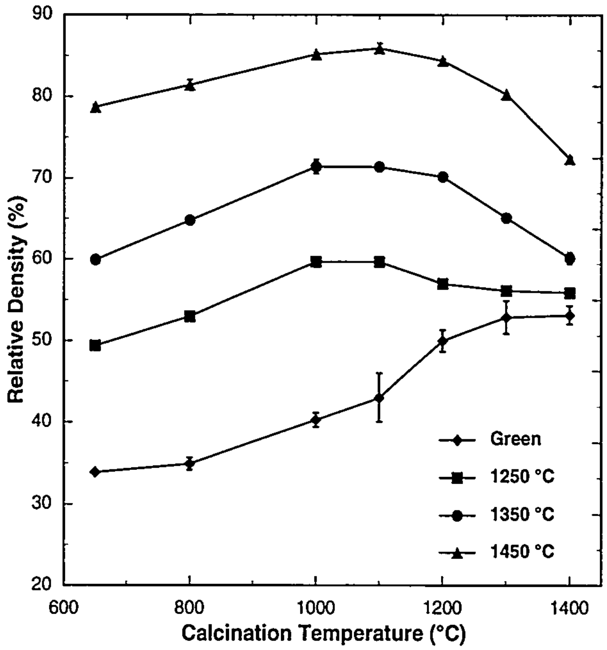

Fig. 5. Sintered Density vs. Calcination Temperature for LCM-30

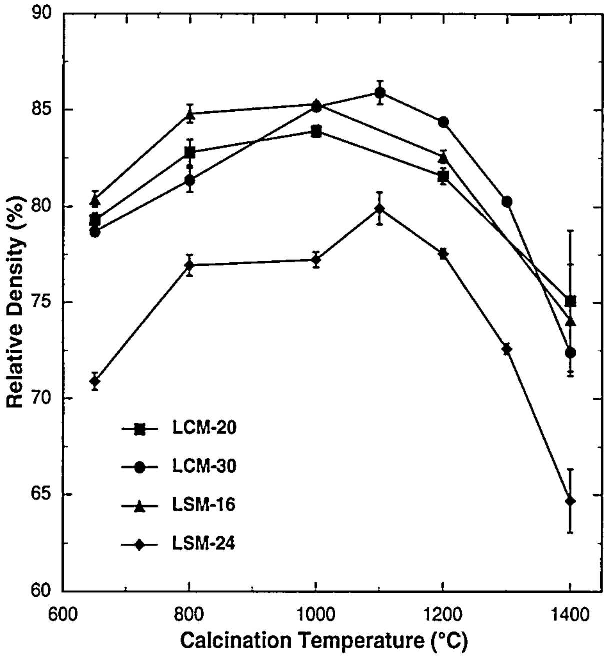

Fig. 6. Sintered Density vs. Calcination Temperature for the Indicated Compositions Sintered at 1450° C

with increasing sintering temperature. Similar results were obtained for LSM-16 and LCM-20. For example, Figure 6 shows the densities for all four lanthanum manganite compositions sintered at 1450°C for 1 hour. With the exception of LSM-24, which exhibited lower densities than the other three compositions, sintered density exhibited little dependence on dopant type and content.

It was found in this study that the sintering behavior of lanthanum manganite is highly dependent on the ratio of A-site cations (La, Sr, Ca) to B-site cations (Mn) present in the material. Compositions were prepared with A/B cation ratios of 0.95, 0.98, 1.00, 1.02, and 1.05. Since it could be safely assumed that all ions present in that structure occupied regular lattice sites,[7] those five ratios would correspond to $(La_{1-y}M'_y)_{0.95}MnO_3$, $(La_{1-y}M'_y)_{0.98}MnO_3$, $(La_{1-y}M'_y)MnO_3$, $(La_{1-y}M'_y)Mn_{0.98}O_3$, and $(La_{1-y}M'_y)Mn_{0.95}O_3$ (M' = Ca or Sr), respectively, in the case of single phase materials. While some of the compositions were indeed determined by XRD analysis to be single phase perovskites, small amounts of additional phases were discovered in others (see below). (For convenience, an oxygen stoichiometry of 3 (rather than $3 \pm \delta$) is assumed, since oxygen nonstoichiometry in highly doped manganites tends to be relatively small).[8]

Figures 7 and 8 show sintered densities for LSM-24 and LCM-30, respectively, as a function of the A/B cation ratio. These figures demonstrate that A-cation deficiency (i.e., A/B ratio < 1) resulted in a significant increase in density. Similar behavior was observed in LSM-16 and LCM-20. This pronounced effect of A/B cation ratio on densification behavior was also observed in shrinkage plots obtained by measuring the linear shrinkage of green compacts as a function of increasing temperature. These plots showed an enhanced shrinkage for A cation deficient manganites relative to stoichiometric and B cation deficient manganites. The shape of these shrinkage plots was consistent with a solid state sintering mechanism; i.e., there were no signs of sudden shrinkage events due to liquid phase formation, as has been reported in lanthanum and yttrium chromites.[9,10]

XRD analysis was performed on specimens

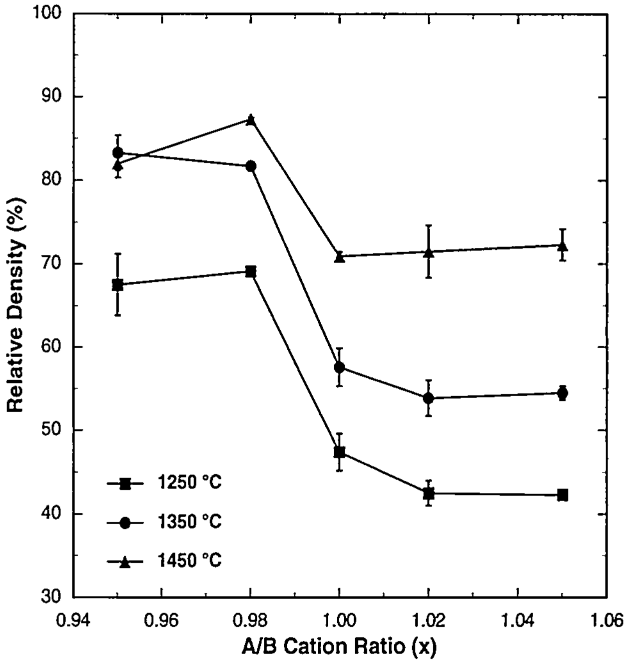

Fig. 7. **Sintered Density vs. A/B Cation Ratio for LSM-24**

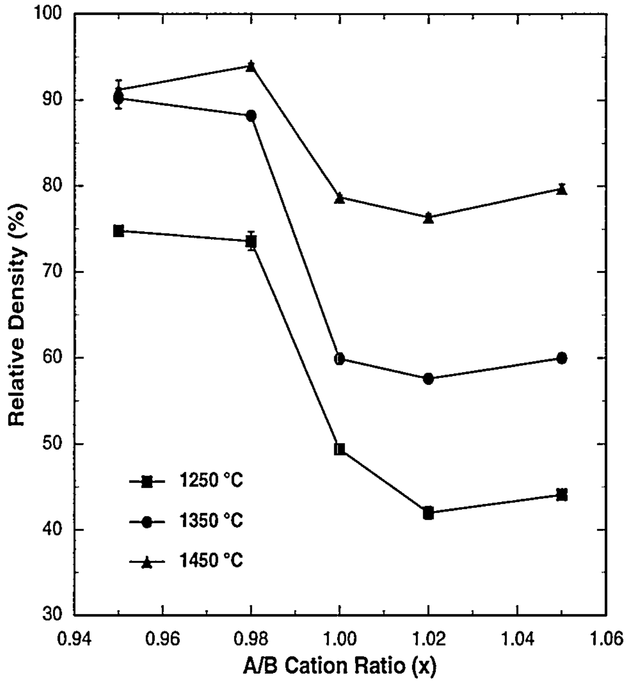

Fig. 8. **Sintered Density vs. A/B Cation Ratio for LCM-30**

Table I. Minor Phases in Lanthanum Manganite (Approx. Wt.% Shown In Parentheses)

Composition	x=0.95	x=0.98	x=1.00	x=1.02	x=1.05
LSM-16	--	--	--	La_2O_3 (3)	La_2O_3 (5), $La(OH)_3$ (0.5)
LSM-24	Unidentified Spinel (1)	--	--	La_2O_3 (2), $La(OH)_3$ (1)	La_2O_3 (3), $La(OH)_3$ (1)
LCM-20	Mn_3O_4 (0.5)	--	--	--	La_2O_3 (2)
LCM-30	Mn_3O_4 (0.5)	--	--	--	CaO (0.5)

sintered at 1450°C to determine the stability of the perovskite phase as a function of the A/B ratio. The results of this analysis are shown in Table I. La_2O_3 was found in some of the compositions with an A/B ratio greater than 1.00. Since La_2O_3 readily absorbs atmospheric moisture to form $La(OH)_3$ (thereby degrading the strength of the sintered material) it is necessary to avoid the presence of La_2O_3 in lanthanum manganite, so that, overall, an A/B cation ratio in the range 0.98-1.00 appears to be optimal for the synthesis of single phase lanthanum manganites. The addition of pore formers (e.g., organic materials which can be burnt out at low temperatures) may be helpful in obtaining the degree of porosity required for the SOFC cathode application.

In an attempt to find the reason for this difference in sintering behavior, slow XRD scans were performed on LSM-24 powders with A/B ratios of 0.98 and 1.00. This analysis showed that, prior to sintering, both powders consisted primarily of the desired perovskite phase, except for approx. 5 wt.% $SrCO_3$ as a second phase. Also, the surface areas and degree of agglomeration of the manganite powders were determined to be essentially independent of A/B cation ratio.

Since the value of the A/B ratio was the only detectable difference in these powders, the enhanced sinterability of manganites when A/B = 0.98 instead of 1.00 can best be explained by examining the defect chemistry of these materials. Since neither interstitial ions nor wrong site ions are expected to be present in significant numbers,[7] preparation of these materials with A/B < 1.00 should produce an excess concentration of A-site vacancies within the structure. Similarly, manganites with A/B > 1 would have excess B-site vacancies within the structure. The higher densities found in the A cation deficient materials indicate that the diffusion of A-site cations is the limiting factor in the mass transport of ions during densification (as suggested by van Roosmalen et al.[11] for undoped lanthanum manganite). Thus, when excess A-site vacancies are introduced, diffusion of A-site cations occurs more readily, resulting in higher sintered densities. Since the introduction of excess B-site vacancies (A/B > 1.00) had little effect on sintering behavior, the diffusion of B-site cations is apparently not a limiting factor in the densification process.

FUTURE WORK

Future SOFC efforts in FY 1994 will continue to focus on the optimization of state-of-the-art and alternative component materials and fabrication procedures. Specific short-term areas of emphasis include an investigation into the effect of ambient oxygen partial pressure on the stability and mechanical properties of Ca and Sr doped lanthanum chromite and Ca doped yttrium chromite. The viability of composite cathodes will be evaluated by studying the sintering, electrical, and thermal properties of physical mixtures of YSZ and Sr and Ca doped lanthanum manganite. This program will conclude in FY 1994.

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P2

Support Research for Solid Oxide Fuel Cells

CONTRACT INFORMATION

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Principal Investigator	Michael Krumpelt
Co-Investigator	Timothy W. Kueper
METC Project Manager	William C. Smith
Period of Performance	October 1, 1993 to July 1, 1994

OBJECTIVE

The purpose of this work is to develop technology for a planar solid oxide fuel cell (SOFC) which uses a thin electrolyte and operates at about 800°C. The two major tasks of this effort are: (1) develop a conductive bond layer between the electrodes and the interconnect and (2) integrate a metallic interconnect into the stack which is well matched with the other cell components with regard to thermal expansion and is adequately protected from corrosion.

BACKGROUND INFORMATION

The fabrication of planar SOFCs involves the assembly of separately fired cell components, which allows versatility and simplicity in the processing methods used for each component. However, several new design issues need to be addressed in the assembly of a planar stack.

The form of the bond layer between stack components is one area which needs design modification. Since, in general, fuel cell components will not be perfectly flat, a conductive bond layer is needed between layers which are to be joined. Without such a layer, electrical contact will tend to be restricted to points distributed over the cell area, causing current concentrations and high resistance. Most methods of applying ceramic material in bond layer form tend to involve large consolidation shrinkages, which cause shrinkage cracking. This cracking results in unacceptable losses in the strength and conductivity of the bond layer. In this research, metals are used in an altered-bond-layer fabrication process to improve the conductivity, and the use of dense foils is explored to reduce bond layer shrinkage.

The bonding procedure also represents an opportunity to form seals at the cell edges, which are necessary to prevent the fuel and oxidant gases from mixing when external manifolding is used. The sealing material needs to form a dense bond between the electrolyte and the interconnect, penetrating any porous electrode materials present at the cell edge. Together with the electrical bond layer which has been applied over the cell area, these seals will determine the mechanical strength of the stack. Our approach is to apply a glass-ceramic sealant as a thin, dense, fiber-like gasket during the initial assembly of the stack. This eliminates potential problems with slurry application and porosity after firing.

The planar SOFC design also allows the choice of ceramic or metal interconnect materials. The standard ceramic interconnect material, doped lanthanum chromite, is difficult to fabricate and expensive, and may have a stability problem when used over extreme oxygen partial pressure ranges. Metals are an obvious choice for a high-strength, highly conductive interconnect material, particularly when low operating temperatures are considered. Factors which complicate the use of metals in SOFCs include their oxidation behavior and their high thermal expansion. These issues are addressed in this research.

PROJECT DESCRIPTION

The first objective of this research is to produce bond layers which allow planar SOFC components to be joined with high strength and conductivity. Our approach is to use metal pastes and foils; such materials offer a higher bulk conductivity than ceramic bonding materials, improved crack resistance, and improved intimacy of the bond. While metal slurries are the most straightforward to apply, foil brazes have also been used in this research. The conductivity, stability, and strength of different metal bonding systems have been evaluated. Edge seal formation using glass-ceramic sealants and a compression-bonding technique has also been studied in combination with the bond layer research.

The second objective of this research is to identify metal interconnect materials with suitable oxidation resistance and a thermal expansion which is matched well to the planar SOFC components. Our approach is to select low-expansion metals with the best high-temperature oxidation resistance, test these materials in fuel cell operating conditions, and explore oxidation protection methods where necessary. Testing of the candidate metals includes dilatometry and thermal cycling while these materials are bonded to cell components. The corrosion characteristics of the metal are also determined under typical fuel cell conditions for the interconnect. Where improvement is needed, coating procedures, including sol-gel and electrophoretic deposition, are explored.

RESULTS

Bond Layers

Initial experiments served to test the concept of a metallic slurry as a bond layer. Bonds were made between anode/electrolyte/cathode trilayers and between ceramic and metal interconnect materials. Initially, platinum paste was used for this purpose. No measurable electronic resistance could be attributed to these bond layers, and their mechanical properties were excellent (the bonds tended to hold in situations where ceramic bond layers had previously fractured). The platinum paste bonds were also tested in combination with edge seals formed from compression-bonded glass fibers (materials developed in an ongoing program at ANL) and again had excellent electrical and mechanical properties. Success of the bond formed depended on the heating rate. When the heating rate was greater than 2.5°C/min, the glass fibers were capable of flow, forming a seal through the electrode porosity and allowing the metallized surfaces to bond. The measured contact resistance in this case was less than 0.1 ohm-cm^2. When the heating rate was greater than 6°C/min, bonded specimens sometimes fractured within the glass. Thus, we concluded that moderate heating rates in forming metal-paste contact layers and glass fiber edge seals will yield successful compression bonds.

Experiments were then conducted with bonding materials that are potentially cost-effective, including silver-based braze foils. One advantage of a foil is that it starts out dense, eliminating the inherent consolidation shrinkage of paste materials. These foils also have a low yield strength, which allows stress relief to occur with reasonably low temperature rates. Thus, these materials have the potential to relieve thermal-expansion-mismatch stresses between components.

Initial testing consisted of firing the braze foils between candidate metal plates in air for 8 h. The brazing temperature was varied to study the tradeoff between the lack of softening behavior at low temperatures and excessive oxidation at high temperatures. Strengths were excellent after the 900°C treatment and 1000°C treatment. Resistance of the bond layer was less than 0.1 ohm-cm^2 when either the 900°C or 1000°C treatment was used. In ongoing research, behavior of the braze foil with a metallized and non-metallized SOFC trilayer is being studied at firing temperatures of 800-1000°C.

Metal Interconnect

Commercially available alloys were tested for expansion behavior and oxidation resistance. One candidate metal had adequately matched thermal expansion to survive thermal cycling with stack components; its measured expansion coefficient of 10.4 x 10^{-6}/°C is slightly lower than that of the zirconia electrolyte and electrode materials (Figure 1). Partial stacks were formed by

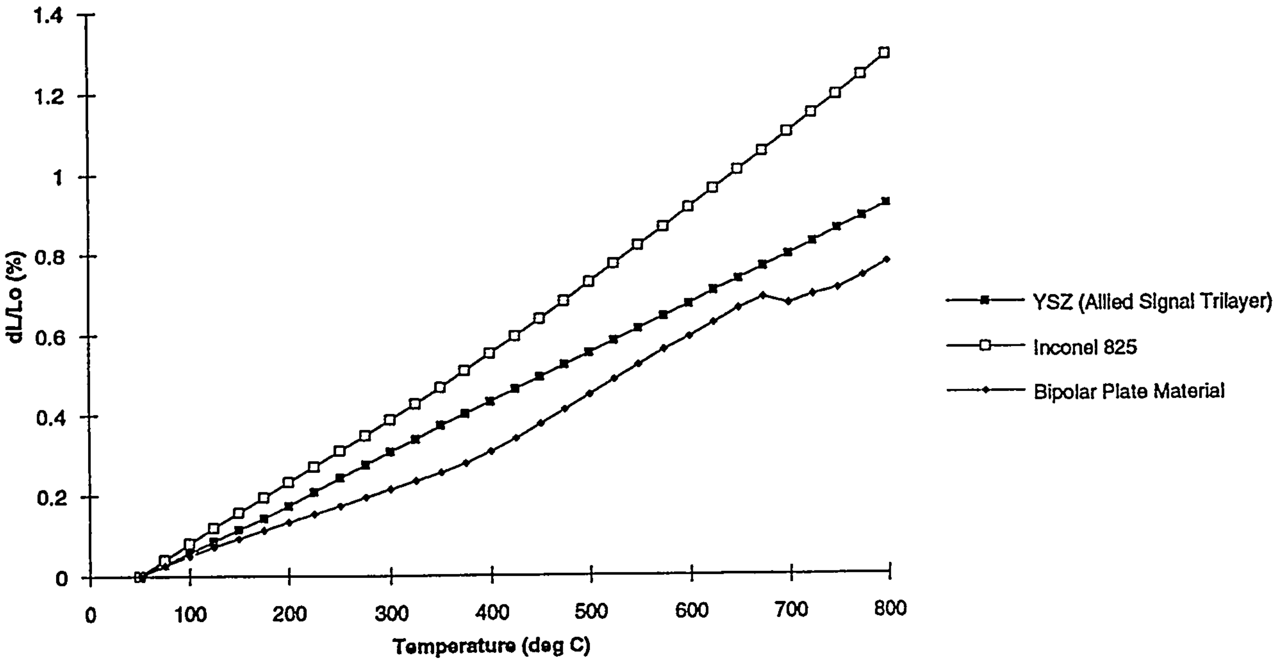

Fig. 1 Thermal Expansions of the Candidate Bipolar Plate Material, YSZ Trilayer, and a Typical Oxidation-Resistant Alloy (Inconel 825).

bonding this metal to SOFC trilayers sealed by platinum paste and glass edge sealants. These structures survived thermal cycling to 800°C without fracture, indicating a good thermal expansion match. Previously tested metals which had expansion coefficients of about 17 x 10^{-6}/°C were found to de-bond when subjected to similar tests.

Literature values of corrosion resistance for this candidate metal indicate that, when no protective coating is used, a depth of attack of 100 μm is reached after 500 h in air at 700°C. This corrosion rate would probably be excessive for electrical contact areas, depending on the conductivity of the oxide layer that is formed.

Tests were performed on the metal in the unprotected state to check its oxidation behavior at temperatures of 800 to 1000°C and to determine the conductivity of the resulting oxidation layers. Air treatments at 800°C consisted of 5 cycles of 8 h each. The candidate metal was examined afterward and had a dull gray appearance; no spallation was observed. Scanning electron microscopy showed that the oxide layer was less than 10 μm thick. Application of platinum paste contacts was performed to check the resistance of the oxidation layer, and it was found to be roughly 8 ohm-cm^2. A single 1000°C treatment for 8 h was also performed on the unprotected metal. In this case, spallation of the oxidation layer occurred. The spalled layer was 15 μm thick and consisted of dense oxides of the more reactive metals in the alloy, with roughly 1 μm grain size. We concluded that the unprotected alloy, while structurally sound at 800°C, has inadequate oxidation resistance for electrical contact areas or for 1000°C operation, and some protection method will be required in these cases.

Since electrical contact areas will also be subjected to the application of a bond layer, the oxidation protection behavior of potential bonding materials (platinum paste and platinum sputtering, as well as silver-based braze foils) was examined. Also tested was electrophoretic deposition of fine ceramic powders. In general, the application of platinum bonding materials resulted in greatly improved oxidation resistance, but more practical materials are needed.

Silver-based braze foils gave plate-to-plate contact resistances of less than 0.1 ohm-cm^2 after 8-h treatments in air at 1000°C. The appearance was excellent (the braze created a smooth coating at the candidate metal surface, although a dark layer of oxidized braze material was present above this). Some evidence of increased spallation was seen in unprotected areas adjacent to the braze. Thus, while this method seems to provide adequate protection for electrical contact areas, the effect on spallation of unprotected areas must be studied further.

The use of ceramic powders for oxidation protection was also studied. Electrophoretic deposition has the potential of creating very thin, dense oxide layers, which would be ideal for oxidation protection without a large resistance. This technique has the added advantage of being capable of patching its own cracks selectively upon successive coatings, since deposition is most rapid in areas where the conductive substrate is exposed. A candidate alloy was coated using electrophoretic deposition of yttria powder from acetyl acetone solution (10 to 20 V, 10 to 60 s), and the coated alloy was sintered at 1200°C in a reducing atmosphere. For these initial tests, the yttria coating was only partially adherent, and full density was not achieved for this powder size. After a further 1000°C treatment in air for 8 h, spalling of the metal was inhibited, but only in small areas of good yttria adhesion. While these results are encouraging, the starting powder characteristics must be improved in order for this method to be effective.

Along with electrical contact surfaces, the interconnect surface where corrosion may be a problem is the seal around the edges of the stack. Glass sealants were found to react to some extent with corrosion products of the candidate interconnect material. When thin (200 μm diameter) fibers of glass were used as the seal material, the reaction tended to prevent good compression bonds from forming; in some cases complete de-bonding of the layers occurred. However, when thicker glass fibers were used, better seals were obtained. The improvement was probably a result of better compressibility of the greater volume of glass and a greater extent of spreading over the metal surface, which distributed any stresses more evenly.

FUTURE WORK

Further characterization of the bonding methods and oxidation protection methods is needed. The stability of the bonding/oxidation protection materials in the presence of the other cell materials under atmospheres expected during operation must be determined. A performance study in fuel cell operating conditions is also needed to determine the conductivity of the interfaces formed. Large-area cell components will be characterized for warpage amounts and then bonded together and tested, so that the relationship between surface irregularity and optimum bond layer thickness can be assessed.

Bond layers formed from other materials will also be studied. In particular, conductive glasses need to be explored as a bond material which may give good coverage of the metal interconnect and reduced corrosion in the active cell area.

Further study of oxidation protection techniques for the metal interconnect will be conducted. Two techniques which may prove valuable are (1) electrophoretic deposition with very fine particle size and (2) laser ablation of ceramic coating materials.

Multiply Manifolded Molten Carbonate Fuel Cells

P3

CONTRACT INFORMATION

Contract Number	49946
Contractor	Argonne National Laboratory 9700 S. Cass Avenue Argonne, IL 60439
Contractor Project Manager	Kevin M. Myles
Principal Investigators	Michael Krumpelt and Michael F. Roche
Co-Investigators	Howard K. Geyer and Stanley A. Johnson
METC Project Manager	Bruce Harrington
Period of Performance	October 1, 1982 to Open

OBJECTIVE

This study consists of research and development activities related to the concept of a molten carbonate fuel cell (MCFC) with multiple manifolds (1). The objective is to develop an MCFC having a higher power density and a longer life than other MCFC designs. The higher power density will result from thinner gas flow channels; the extended life will result from reduced temperature gradients. Simplification of the gas flow channels and current collectors may also significantly reduce cost for the multiply manifolded MCFC.

BACKGROUND INFORMATION

The new design employs a number of gas inlet and exit manifolds distributed across the plane of the MCFC. Because the gas flow in this new design is two dimensional, existing one-dimensional models did not adequately describe its features. Therefore, a two-dimensional model was developed (2) that relied on first principles and employed overpotential relationships previously developed by Wilemski *et al* (3) of Physical Sciences, Inc. (PSI).

Validation of the two-dimensional model on a single cell was addressed first by analyzing a cross-flow cell and comparing the results with those calculated previously using the PSI one-dimensional model (3). In the cross-flow cell, the gas flows entered and exited along the cell edges, and a 1 m x 1 m square represented the entire cell. Profiles of temperatures (for anode, cathode, and electrolyte matrix), Nernst potentials, and current densities in the cross-flow cell were generated using the two-dimensional model. These profiles were essentially identical to those previously generated by the PSI one-dimensional model.

In the distributed-manifold design, the gas flows enter and exit at the corners of a smaller square that is repeated a number of times to form a 1 m x 1 m cell. Issues addressed by the initial modeling study (2) included the effects of corner manifolding on a 1 m x 1 m cell and on smaller squares (e.g., 0.072 m x 0.072 m) or rectangles (e.g., 0.144 m x 0.072 m) that could be employed to build up a distributed-manifold MCFC. With the rectangular designs, the corner manifolding simulated either co-flow or counter flow.

PROJECT DESCRIPTION

This project is one of four MCFC projects that are being pursued at Argonne National Laboratory. The other three projects are described elsewhere in these proceedings. This project is linked to one of the other projects (development of an

MCFC with high power density) because high-power-density MCFCs will require multiple manifolding for maximum volumetric power density.

In recent modeling studies, we have further explored the effects of the number of gas manifolds and the type of gas flow (counter, cross, corner, or co-flow) on MCFC power and temperature gradients.

Flow visualization has also been employed to determine the gas distribution in the flow channels of the multiply manifolded MCFC. The flow visualization technique that provided the most useful data employed injection of ammonia-saturated air at selected points near the gas inlet to develop an ammonia-sensitive paper placed in the gas flow channels of a plastic mock-up (25 cm x 25 cm) of the multiply manifolded MCFC.

In addition, we have begun tests of MCFCs (about 100 cm^2 in area) that incorporate the corner flow characteristics of the multiply manifolded MCFC and also use dimpled flow fields of the type envisioned in the original patent (1).

RESULTS

A computer modeling study was conducted in which pressure drops, anode and cathode utilizations, and areas of inlet and outlet manifolds were held constant as the number of inlet and outlet manifolds in the MCFC was increased. This study showed that the multiply manifolded MCFC provided a significant reduction in stack height and cell temperature gradients. These were decreased by 37% and 48%, respectively, when a 4 x 4 array of 625 cm^2 cells was substituted for each 1 m^2 cell in the MCFC stack. This study also showed that arrays much larger than a 4 x 4 array of 625 cm^2 cells (e.g., a 10 x 10 array of 100 cm^2 cells) would provide little additional benefit in reduction of stack height and cell temperature gradients.

Another modeling study compared the performance of multiply manifolded MCFCs having four different types of flow (counter, cross, corner, and co-flow) of the anode and cathode gases in fuel cells employing 70% air and 30% carbon dioxide at 1 atm as the cathode gas. For the same average temperature, the current and power were, respectively, 50% and 20% higher for co-flow than for counter flow, which yielded the lowest current and power of the four types of flow. Corner flow mimicked cross flow and yielded intermediate levels of current and power. Co-flow was also superior to counter flow with a simulated system gas (8% O_2, 8% CO_2, bal. N_2 and steam) at 3 atm as the cathode gas, but the difference between the two was smaller in this case.

Flow visualization was employed to confirm and extend the modeling studies. The flow visualization studies, in conjunction with computer modeling, showed that the corner-flow characteristics of the multiply manifolded MCFC could be converted to counter, cross, or co-flow as desired by suitable modification of the gas flow channels. The flow pattern depended on the length-to-width ratio of the manifolding and the design of the gas flow channels. Thus, the multiply manifolded MCFC provides a high degree of design flexibility. As noted previously, it also yields a higher power density and lower temperature gradients than previous designs.

Differential pressures were measured during the flow visualization studies, and the data were used as inputs for computer modeling of flow maldistribution in an MCFC stack. Calculations based on the differential pressure measurements of one flow channel design indicated a flow maldistribution of under 1% for the anode gas in an MCFC stack having a height of one meter. This degree of maldistribution is acceptable.

A corner-flow MCFC (83 cm^2) that employs a flat current collector/bipolar plate and uses dimpled anodes and cathodes to form the anode and cathode gas-flow fields is now in operation. Its initial performance is as good as that of previous MCFCs having a more conventional design. Thus, many of the concepts in the original patent (1) have been demonstrated.

FUTURE WORK

The design of the gas flow channels is an important feature of the multiply manifolded MCFC. Meacham's patent (1) proposes dimpling of the electrodes to form the gas flow channels and a flat metal sheet as the current collector and bipolar plate. More conventional designs employ flat electrodes and a combination of metal sheets (perforated, corrugated, and flat) to form the gas flow channels, current collectors, and bipolar plate. Both types of design are being evaluated in this study. For the evaluations, MCFCs with a variety of flow channel designs for the anode and cathode gases will be tested. In addition, the size of the MCFCs is being increased from about 100 cm^2 to 625 cm^2 to provide a better simulation of the basic unit of the multiply manifolded MCFC.

ACKNOWLEDGMENTS

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P4

Lithium Ferrate and Lithium Cobaltate Cathodes for Molten Carbonate Fuel Cells

CONTRACT INFORMATION

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Principal Investigators	Michael Krumpelt and Michael F. Roche
Co-Investigators	Ira D. Bloom, J. Ernesto Indacochea, and Gene H. Kucera
METC Project Manager	Bruce Harrington
Period of Performance	October 1, 1982 to Open

OBJECTIVE

The objective of this research is to develop cathodes for the molten carbonate fuel cell (MCFC) having a performance approaching that of the lithiated nickel oxide cathode and a significantly greater life, particularly in pressurized MCFCs. To meet this objective, cathodes containing either doubly doped lithium ferrate or lithium cobaltate are being developed.

BACKGROUND INFORMATION

The use of dopants such as Co, Ni, Mn, and Cu to improve the electronic conductivity of lithium ferrate cathodes for MCFCs was first described in a patent assigned to ANL investigators (1). This patent also included the use of lithium cobaltate as an MCFC cathode.

Subsequently, measurements were conducted to determine the properties of singly doped lithium ferrate cathodes under a broad range of MCFC operating conditions (2). These measurements showed that cobalt-doped lithium ferrate was the best of the singly doped lithium ferrate cathodes, but it only performed well over a very narrow range of MCFC cathode gas compositions. Our research then focused on the use of two dopants in lithium ferrate cathodes (3). This doubly doped lithium ferrate cathode now has a performance approaching that of the lithiated nickel oxide cathode.

In addition, we have continued to investigate the lithium cobaltate cathode. Its performance also approaches that of the lithiated nickel oxide cathode. Others (4) have also been investigating the lithium cobaltate cathode and have shown that it exhibits a performance approaching that of the lithiated nickel oxide cathode.

PROJECT DESCRIPTION

This project is one of four MCFC projects that are being pursued at Argonne National Laboratory. The other three projects are described elsewhere in these proceedings. In this project, we are optimizing the composition, microstructure, and loading density of the doubly doped lithium ferrate cathode and the lithium cobaltate cathode.

Experiments in this project include measurements of the resistivities, Seebeck coefficients, and microstructures of the doubly doped lithium ferrate cathode and lithium cobaltate cathode. In addition, their performance in MCFCs is being determined, and post-test analyses on the MCFCs are being conducted.

This project is linked to another project (development of an MCFC with high power density) because such MCFCs will require a cathode having a longer life than the present cathode, lithiated nickel oxide.

RESULTS

Molten carbonate fuel cells with either doubly doped lithium ferrate or lithium cobaltate cathodes now exhibit a voltage of 915 mV at 1600 A/m^2 when employing a cathode gas (58 mol% O_2-42 mol% CO_2) that simulates operation at 3-4 atm with standard oxidant (air plus 27 vol% CO_2). Under the same conditions, the voltage of an MCFC employing the lithiated nickel oxide cathode is 950 mV. The voltages of MCFCs employing doubly doped lithium ferrate and lithium cobaltate cathodes will be increased to 950 mV at 1600 A/m^2 by further cathode optimization. For example, studies are underway to determine the effects of increasing the cathode loading densities from their present value (about 100 mg/cm^2) to the loading density currently being employed with the lithiated nickel oxide cathode (150 mg/cm^2).

The lithium cobaltate cathode, like the lithiated nickel oxide cathode, required no special doping to achieve a high performance because its resistivity was low (1 Ω-cm *vs* 0.1 Ω-cm for lithiated NiO). Double doping was required to achieve an equivalent resistivity and performance from the lithium ferrate cathode. Cell tests of about 2000 h at high oxygen and carbon dioxide pressures have indicated the potential for long life in MCFCs having cathodes prepared from either doubly doped lithium ferrate or lithium cobaltate.

FUTURE WORK

Future experiments will be directed toward a detailed comparison of the relative performance, life, and life-cycle cost of the doubly doped lithium ferrate, lithium cobaltate, and lithiated nickel oxide cathodes in pressurized MCFCs. These experiments will be conducted by Argonne National Laboratory and an industrial partner.

ACKNOWLEDGMENTS

This study is being conducted under the auspices of the U.S. Department of Energy, Contract No. W-31-109-Eng-38. Joseph R. Stapay and Kevin Byrne provided technical assistance in the laboratory.

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P5

Carbonate Fuel Cell Analysis

CONTRACT INFORMATION

Contract Number DE-AC21-93MC30054

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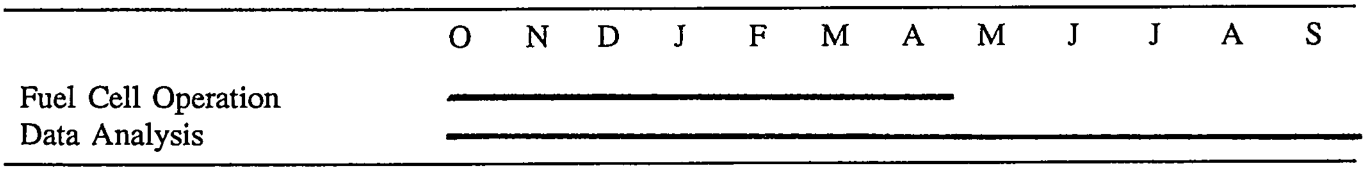
METC Project Manager Cliff Carpenter

Period of Performance August 1993 to February 1995

Schedule of Milestones

FY94 Program Schedule



OBJECTIVES

Power plant system studies indicate that carbonate fuel cell systems have the potential of offering high efficiency, competitive cost and environmentally superior power plants operating on coal. The overall objective of the project was to demonstrate carbonate fuel cell compatibility with coal-derived gases; the specific objective of this project was to assess the possible effect of contaminants on the performance and life of a carbonate fuel cell. The approach taken was to test a 20kW fuel cell stack using actual coal-gas, to monitor its performance relative to a standard and to perform a detailed post-test analysis of the stack to determine long term operating issue(s), if any.

PROJECT DESCRIPTION

This project was a continuation of an EPRI sponsored program. Under the EPRI program, a truck transportable skid mounted test facility was designed and constructed. The fuel cell stack was also built and tested under EPRI sponsored activities. The fuel cell test facility was interfaced with Destec Energy Corporation's LGTI coal gasification plant in Plaquemine, Louisiana. The Destec coal gasification process uses a pressurized, entrained flow, slagging, slurry-fed gasifier with a continuous slag removal system. The sweet syn-gas has an energy value of ~245 Btu/cu.ft.. A slip stream of the sweet syn-gas was interfaced with the carbonate fuel cell test facility. The facility has the capability to test 20kW fuel cell stacks on either natural gas or coal derived syn-gas.

STACK TEST

A 20kW carbonate fuel cell stack was designed and constructed at ERC's manufacturing subsidiary, Fuel Cell Manufacturing Corporation (FCMC). The stack, shown in Figure 1, is comprised of 54 cells of 2 ft. x 3 ft. size. This stack was the first large size stack produced at the new FCMC facility. Following assembly and preconditioning, the stack underwent 100 hours of testing at ERC on natural gas to establish a baseline prior to shipping to the gasifier site. The stack was tested at a 32kW power output at 800 amps.

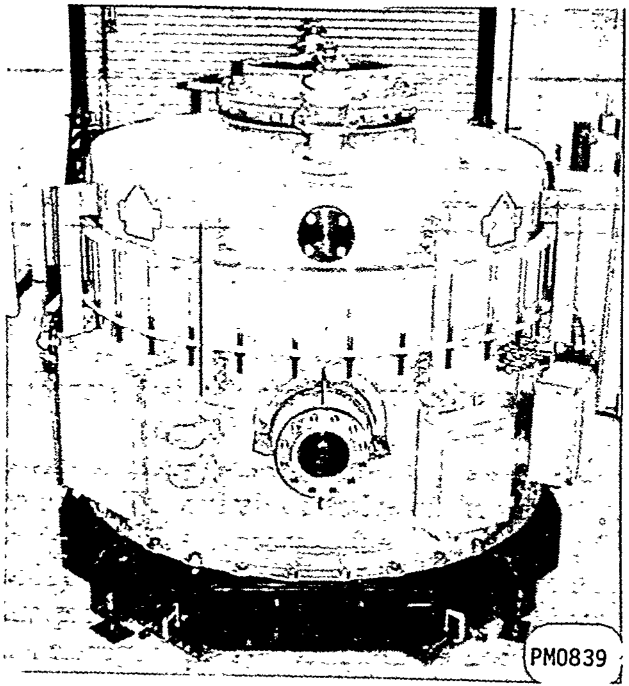


Figure 1. DFC Stack for Coal-Gas Testing
This Stack was the First Large Size Stack Built by Fuel Cell Manufacturing Corporation (FCMC)

The enclosed stack was truck transported to the gasifier test site along with the necessary instrumentation and installed on a foundation at the test facility. A schematic of the Destec test facility is depicted in Figure 2. The syn-gas slip stream passed through a scrubber, an MDEA acid gas removal process, and an absorber column prior to reaching the test facility. As the gas enters the facility it is preheated (E-101) and passes through zinc and alumina absorbtion beds (R-101). The gas is further heated (E-104) before entering the fuel cell anode. Anode exhaust is sent to the burner (H-101) where it is combusted with air and additional fuel and then enters the fuel cell cathode. Cathode exhaust is vented to the atmosphere. This facility was designed and built under EPRI sponsorship.

The installed fuel cell stack is shown in Figure 3. The stack is placed in an enclosure specially designed for outdoor operation. This is the first carbonate fuel cell tested in an outdoor industrial environment.

The stack was initially operated at Destec on natural gas to allow a direct comparison with initial performance during the qualification test at ERC, and for later comparison with syn-gas performance. As shown in Figure 4, stack performance on natural gas at the Destec facility was comparable with the performance obtained at ERC prior to shipping. There was no effect on stack performance as a result of the thermal cycle or transportation to the Destec facility.

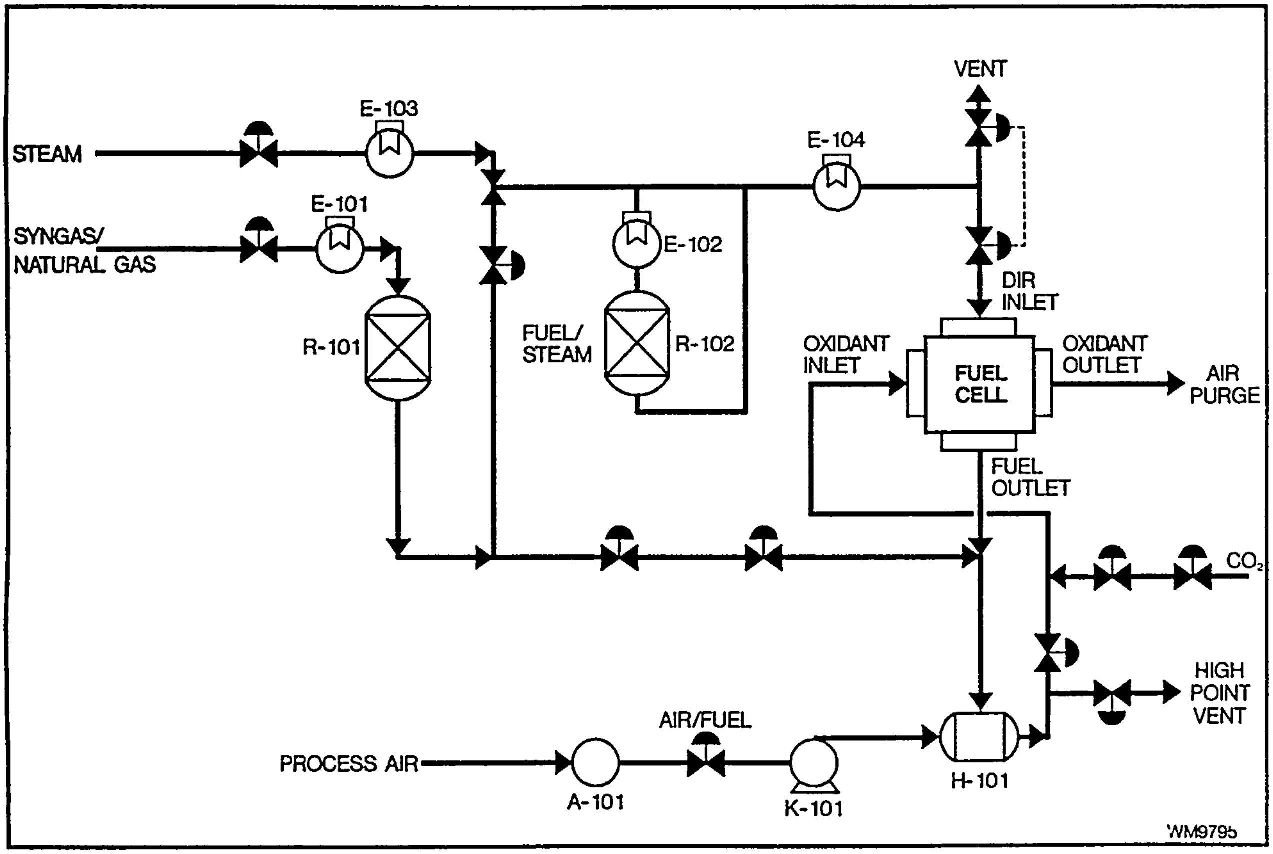


Figure 2. Destec Test Facility Schematic
The Truck Transportable Skid Mounted Test Facility was Interfaced with Destec Energy Corporation's LGTI Coal Gasification Plant

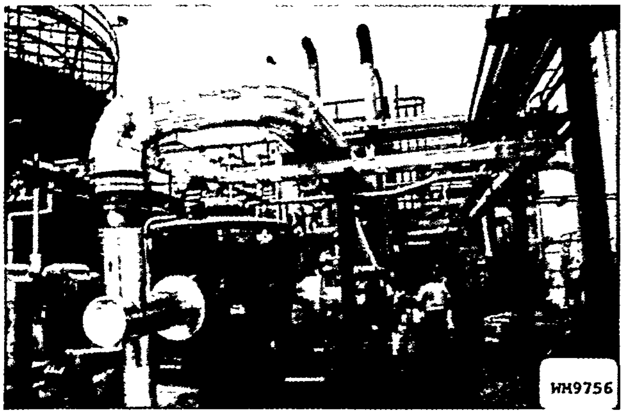


Figure 3. DFC Stack at the Destec Site
This Stack is the First to Operate in an Outdoor Industrial Environment

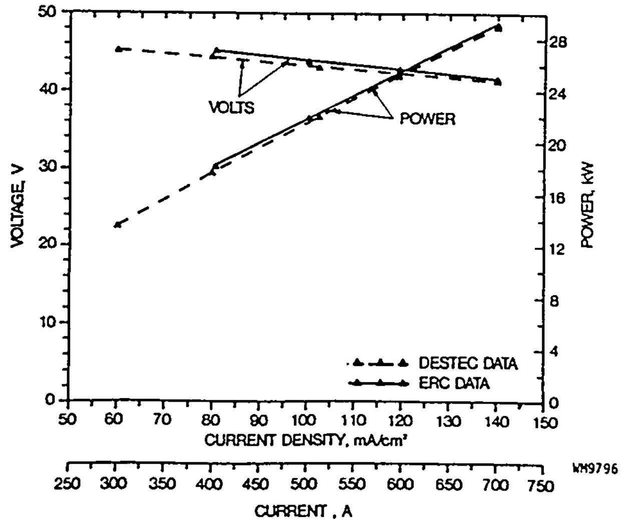


Figure 4. Stack Performance Comparison on Natural Gas
Performance was Unaffected by Thermal Cycle and Shipping

Following the initial test on natural gas, the stack was restarted on syn-gas for a 4,000 hour endurance run. This was the first time in the world that a fuel cell stack of any type was operated on actual syn-gas at a coal gasifier site. An ERC laboratory scale stack has been operated in Germany at an RWE facility on simulated fluid bed Winkler type gasifier product gas including contaminants for ~11,000 hours.

Stack performance on natural gas and syn-gas is compared in Figure 5. The performance difference of 26 mV per cell was expected due to the lower energy content of the syn-gas. The stack successfully completed the 4,000-hour endurance goal, logging a total of 4,022 hot hours with over 1,800 hours operation on load with syn-gas. Stack operating history is shown in Figure 6. The facility suffered several off-design events, including a breakthrough of sulfur contaminants. Burner related trips also interrupted fuel cell operation. Even after possible exposure to high sulfur levels and frequent off-load periods, stack performance remained relatively stable. This is illustrated in Figure 7, a performance comparison of a typical cell group. There is little change in beginning-of-life (BOL) and end-of-life (EOL) performance.

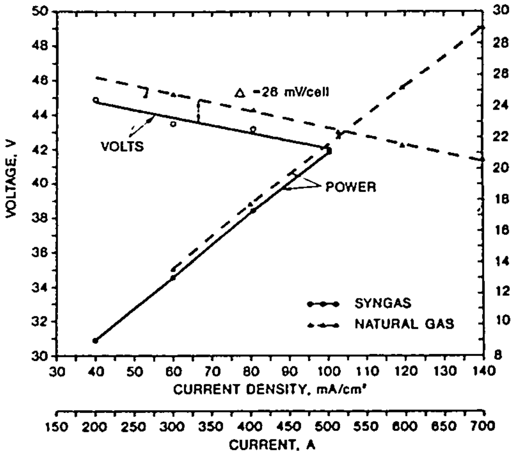


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Figure 5. Performance Comparison on Natural Gas and Syn-Gas
The Lower Energy Content of the Syn-Gas Resulted in the Expected Stack Performance Difference

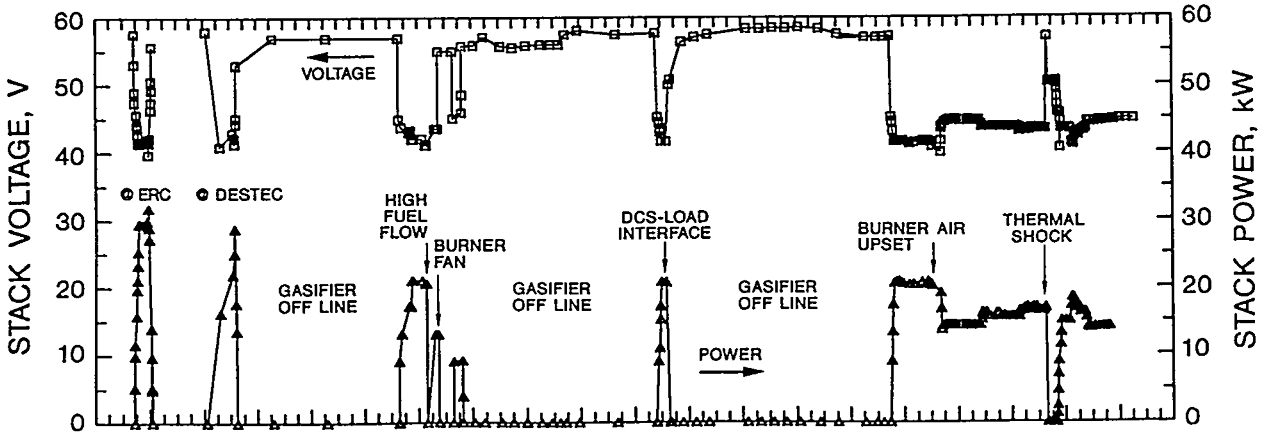

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Figure 6. DFC Stack Operating History
4,000-Hour Test Provided First-Hand Coal-Gas Operation Experience

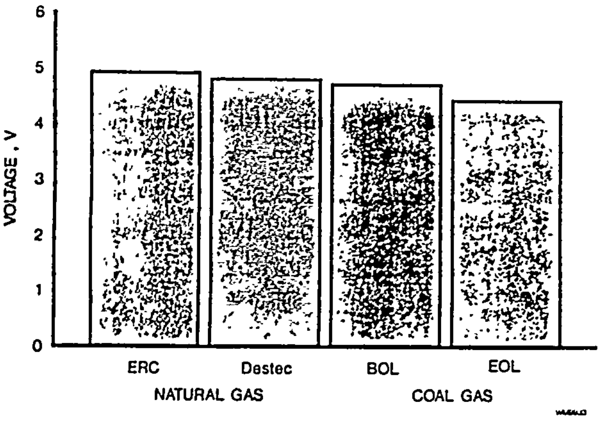

Figure 7. Performance History of a Representative Cell Group
Performance Remained Fairly Stable Throughout the 4,000-Hour Endurance Test

POST-TEST ANALYSIS

The objectives of the post-test analyses are:

- Fuel cell stack operability in an outdoor, industrial environment
- Materials stability in coal-gas
  - Material degradation
  - Contaminant accumulation
  - Electrolyte loss

The stack was returned to ERC for detailed post-test analyses now in progress. This report provides the results available to date.

A detailed disassembly of the stack and the external hardware were completed. The stack enclosure provided complete protection from outdoor conditions. No evidence of corrosion or any form of degradation was observed.

The stack hardware (manifold, compression system, insulation) remained in very good condition and appeared to have performed as designed. Detailed analysis of the anode side hardware did not show any possible indications of metal dusting. Some evidence of soot formation was noticed. This may have resulted from off-design operations encountered during the test. Particulates were found in the cathode side piping. The metallurgy of this material suggests its origin in materials upstream of the fuel cell. This observation suggests that the balance-of-plant equipment selection should be an important consideration.

A thorough metallographic analysis of the fuel cell active materials was also completed. Cell hardware corrosion rates were in-line with natural gas data, as shown in Figure 8. Electrolyte loss to the cell hardware also appeared normal. This data projects that the baseline cell hardware will meet the 40,000 hour life goal. The anode, cathode and matrix also appeared normal. No degradation due to coal-gas operation was observed.

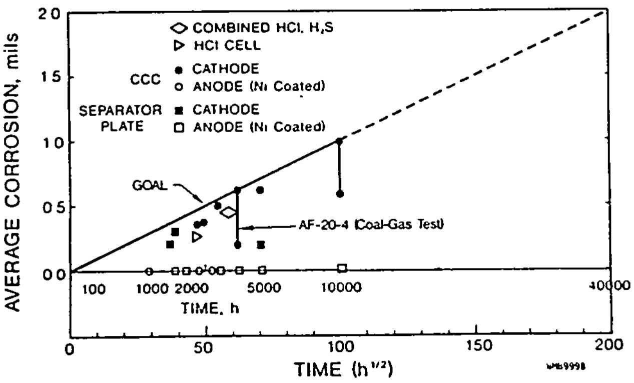

Figure 8. Post-Test Corrosion Data
Corrosion of Current Collector within Acceptable Limit

Possible contaminant accumulation in cell components and electrolyte inventory are being analyzed. These results will be available at a later date.

CONCLUSION

The operability of carbonate fuel cells on coal-gas was demonstrated. The carbonate fuel cell performance and stability were unaffected by coal-gas. Stabilization of coal-gas and proper balance-of-plant materials are important considerations for operation with coal-gas.

Molten Carbonate Fuel Cell with High Power Density

P6

CONTRACT INFORMATION

Contract Number	49946
Contractor	Argonne National Laboratory 9700 S. Cass Avenue Argonne, IL 60439
Contractor Project Manager	Kevin M. Myles
Principal Investigators	Michael Krumpelt and Michael F. Roche
Co-Investigators	Ira D. Bloom, Howard K. Geyer, and Stanley A. Johnson
METC Project Manager	Cliff Carpenter
Period of Performance	October 1, 1982 to Open

OBJECTIVE

The objective of this research is a doubling of the current density of the molten carbonate fuel cell (MCFC) from the present value of 1600 A/m^2 to 3200 A/m^2 and a similar increase in the volumetric power density.

BACKGROUND INFORMATION

Pressurized operation will be required to achieve an acceptable MCFC voltage (above 0.75 V) at a current density of 3200 A/m^2. The present MCFC cathode, nickel oxide, because of its relatively short life in pressurized MCFCs, cannot be used in the high-power-density MCFC. Instead, lithium ferrate cathodes (doubly doped) and lithium cobaltate cathodes will be employed because they are less sensitive to pressure. These cathodes are being developed in another MCFC project at Argonne National Laboratory (1). The distributed manifold concept (2) will also be employed in the high-power-density MCFC since it will provide the highest volumetric power density and lowest temperature gradients, according to our computer modeling studies (3). The modeling studies have also shown that co-flow of the anode and cathode gases will provide the highest volumetric power density at any MCFC temperature. Consequently, co-flow of the anode and cathode gases will be employed in the high-power-density MCFC.

PROJECT DESCRIPTION

This project is one of four MCFC projects that are being pursued at Argonne National Laboratory; the other three projects are described elsewhere in these proceedings. This project is linked to two of the other projects (one on the multiply manifolded MCFCs, the other on lithium ferrate and lithium cobaltate cathodes for MCFCs) because high-power-density MCFCs will require both multiple manifolding and the advanced cathodes.

Experiments in this project include computer modeling of MCFC performance, tests of candidate MCFCs at high current densities, and post-test examination of these MCFCs. Currently, MCFCs having an area of 25 cm^2 are being tested; their area will be increased to 100 cm^2 in the near future. The best components identified in the 100 cm^2 tests will then be tested in full-scale subunits of the multiply manifolded MCFC (625 cm^2).

RESULTS

An MCFC (25 cm^2) having a doubly doped lithium ferrate cathode was operated for about 1400 h at a current density of

3200 A/m^2. The cathode gas, which contained 70% oxygen and 30% carbon dioxide, simulated operation with air-carbon dioxide mixtures at about 4 atm. The MCFC voltage, initially above 0.7 V, declined to about 0.6 V toward the end of the test because of increasing cell resistance. The increased cell resistance, associated with corrosion of current collectors in the MCFC, appears to be the main impediment to extended operation at high current density.

FUTURE WORK

Future experiments will continue to focus on operation of the MCFCs at high current density. The experiments will employ lithium ferrate and lithium cobaltate cathodes, and the relative merits of various flow configurations will be evaluated in MCFCs having areas of up to 625 cm^2. As the experiments progress, we will also address the problem of increased cell resistance.

ACKNOWLEDGMENTS

This study is being conducted under the auspices of the U.S. Department of Energy, Contract No. W-31-109-Eng-38. Joseph R. Stapay and Kevin Byrne provided technical assistance in the laboratory.

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P7

Pitting Corrosion of Aluminized Seals in Molten Carbonate Fuel Cells

CONTRACT INFORMATION

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Principal Investigators	Michael Krumpelt and Michael F. Roche
Co-Investigator	Ira D. Bloom
METC Project Manager	Cliff Carpenter
Period of Performance	October 1, 1982 to Open

OBJECTIVE

The objective of this research is to gain a better understanding of the corrosion of the aluminized Type 316 stainless steel (SS) employed in the seal areas of the molten carbonate fuel cell (MCFC). The seals are formed between the aluminized Type 316 SS surface and the electrolyte (generally a mixture of molten alkali carbonates and lithium aluminate). These "wet" seals are used at the periphery of the MCFC and around the gas manifolds that deliver fuel and oxidant to the anode and cathode of the MCFC. They contain the anode and cathode gases within the MCFC. Of particular interest in this study is measurement of the pitting corrosion of the aluminized Type 316 SS in the seal areas of the MCFC.

BACKGROUND INFORMATION

Corrosion of Type 316 SS is known to be most severe on the anode side of an MCFC (1, 2). It is particularly severe in areas exposed to exhaust anode gas (CO_2 with a high concentration of H_2O and a low concentration of H_2). To minimize this corrosion, the Type 316 SS current collector is ordinarily protected by some type of nickel plating. In addition, the Type 316 SS in the seal areas at the MCFC periphery and around its gas manifolds is usually protected by some form of aluminizing process which can provide protection for at least 10,000 h of operation (3).

Pitting corrosion affects nearly all metals, including aluminum and its alloys (4). Its occurrence in MCFCs has not been investigated to date.

PROJECT DESCRIPTION

This project is one of four MCFC projects that are being pursued at Argonne National Laboratory. The other three projects are described elsewhere in these proceedings. This project is most closely associated with the project to develop multiply manifolded MCFCs, which require a larger number of seals than other types of MCFC.

Experiments in this project include potentiostatic measurements of pitting potentials and determinations of pit morphology for Type 316 SS coupons (bare and aluminized) in molten alkali carbonate electrolytes at 650°C. Static immersion tests of the coupons in the same electrolytes will also be conducted, and the effects of contaminants such as chloride and sulfide ions will be determined. In these experiments, the gas phase will consist of simulated exhaust anode gas.

RESULTS

Measurements are in progress to determine the pitting potential range of Type 316 SS coupons when exposed to exhaust anode gas in molten Li_2CO_3-K_2CO_3 and Li_2CO_3-Na_2CO_3 electrolytes at 650°C. A potentiostatic technique is being employed. To date, the typical duplex corrosion layer (5) has been observed, but the pitting potential range has not yet been determined.

Aluminized Type 316 SS coupons have been received from a commercial supplier. Two different types of coating were employed. These coupons will be annealed and will then be subjected to a variety of corrosion tests.

FUTURE WORK

Potentiostatic measurements will be conducted to determine the pitting potentials of the aluminized Type 316 SS coupons. Immersion tests will also be conducted in which the coupons will be exposed to molten alkali carbonate electrolytes under exhaust anode gas for an extended period of time. The effects of contaminants such as chloride and sulfide ions will be determined in both the potentiostatic and immersion tests.

ACKNOWLEDGMENTS

This study is being conducted under the auspices of the U.S. Department of Energy, Contract No. W-31-109-Eng-38. Joseph R. Stapay and Kevin Byrne provided technical assistance in the laboratory.

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P8

Processing of $LaCrO_3$ for Solid Oxide Fuel Cell Applications

CONTRACT INFORMATION

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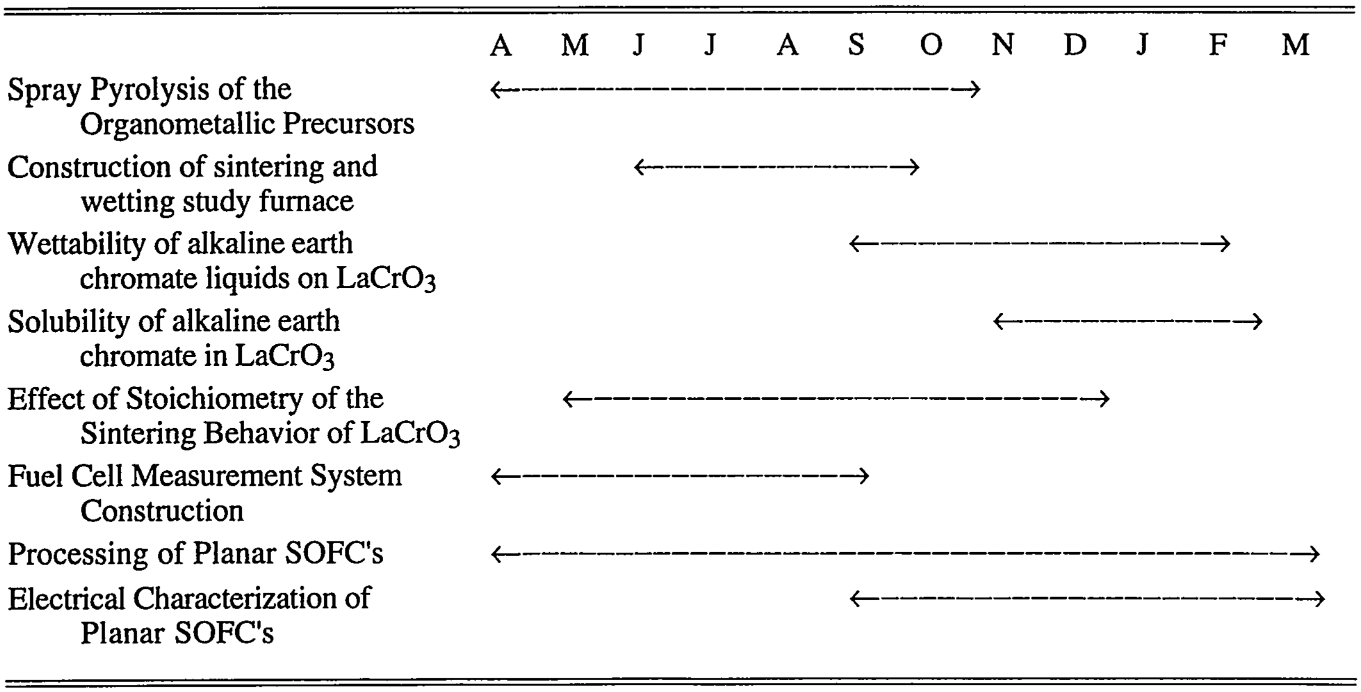
METC Project Manager	Bruce Harrington
Period of Performance	April 16, 1994 to April 15, 1995

Schedule and Milestones

FY94 Program Schedule



OBJECTIVES

The University of Missouri-Rolla is performing a 5 year research program dedicated towards the development of $LaCrO_3$-based interconnect powders which densify when in contact with anode and cathode materials for solid oxide fuel cells (SOFC). During a previous DOE-METC program (DE-FG21-89MC26015) we developed and characterized potential cathode and interconnect materials.[1] During the course of this program we investigated compositions within the pseudo-ternary $LaCrO_3$-$LaMnO_3$-$LaCoO_3$ system. Results indicated that some of the compositions in this system exhibited the requisite electrical conductivity and stability towards reduction, and could be sintered in air at temperatures less than 1450°C.

In this research program we have expanded our studies on the processing and sintering of $LaCrO_3$ to make dense interconnects using $LaCrO_3$-based oxides at temperatures less than 1500°C in an air atmosphere and in contact with both anode and cathode oxides. The specific objectives of this research program are to:

- Develop a novel technique which reproducibly yields $LaCrO_3$-based powders with the desired particle characteristics
- Fully understand the liquid phase sintering mechanism
- Clearly identify the reason why $LaCrO_3$ does not densify in the presence of electrolyte and cathode materials
- Systematically solve this problem through judicious control over the liquid phase.
- Incorporate materials developed in this program into planar cells and measure their performance.

BACKGROUND INFORMATION

One of the primary factors limiting the commercialization of SOFC's is the inability to fabricate $LaCrO_3$-based interconnects cheaply, and in contact with other SOFC components. The very properties which make $LaCrO_3$ a good interconnect material, also make it very difficult to sinter. Microstructural evolution ↔ property relationships of the $LaCrO_3$-based interconnect need to be understood. This was recently confirmed again at a DOE-sponsored SOFC workshop held May 11, 1994 in Oak Ridge, TN. At this meeting the densification behavior of $LaCrO_3$ was identified as the top "show-stopper" limiting the development of SOFC's. Indeed, the inability to co-sinter $LaCrO_3$-based compounds in air at $T<1550°C$ with other SOFC components leads to unacceptable interfacial reactions, and resultant losses in efficiency. Many researchers have addressed this problem, yet progress has been hindered due to recognized difficulties in repeatable processing. Phase development, stoichiometry, particle size/distribution, agglomerate/ aggregate formation, volatility of components, and their dependence on dopant levels and sintering temperature, time and atmosphere are very difficult to systematically take into account.

The results of our work and other investigators[2-10] have lead us to the conclusion that the densification of $LaCrO_3$-based oxides is controlled by the ability for mass transport (sintering) to occur under conditions which minimize Cr oxide volatilization (Cr activity, a_{Cr}). We demonstrated that this could be accomplished by sintering at temperatures >1750°C and oxygen activities of about 10^{-10} atm[2] but clearly these sintering conditions are not compatible with the stability of other SOFC components. Another approach to reduce a_{Cr} is to lower the sintering temperature with liquid fluxes, which was initially pursued by Argonne National Labs[8]. Although the high mobility of the fluoride fluxes detrimentally affected the other cell components, their work showed that liquid phase sintering with a more compatible transient phase would be an attractive approach.

In our earlier DOE-METC program we showed that additions of Ca and Co to $LaCrO_3$ and $YCrO_3$ allowed densification to take place in air at temperatures below 1450°C. However further studies showed that interactions during cofiring between the transient liquid phase (mixture of CaO-CoO and Cr_2O_3) and the electrolyte and cathode prevent densification of the $LaCrO_3$.

Recent sintering studies on Ca-and Sr-doped $LaCrO_3$ suggest that the ability to densify these compounds in air at T<1550°C may be related to the presence of a transient liquid phase formed from the melting of chromates (CaO-CoO-Cr_2O_3 system)[1,12]. If this is true, then in order to co-sinter this system with other SOFC components, detailed knowledge concerning the composition, source, stability range, and wetting behavior of the liquid phase is required. With this information we can vary the parameters which will allow for the densification of the interconnect when cofired with the other SOFC components. Successful completion of the proposed objectives will be a major step towards the commercialization of SOFC.

PROJECT DESCRIPTION

This project is divided into four primary objectives:

#1 Reproducible Synthesis of $LaCrO_3$ Powders
#2 Sintering of $LaCrO_3$-based Powders
#3 Co-sintering of $LaCrO_3$-based Powders with Cathode and Electrolyte Materials
#4 Characterization of Planar SOFC's

Results from these studies are presented in the following sections.

$LaCrO_3$ SINTERING STUDIES

RESULTS

Studies over the past year have focused on understanding the role $CaCrO_4$ plays in the densification behavior of $(La,Ca)CrO_3$. A-site excess and deficient materials were compared through the use of SEM, EDS, XRD and DTA.

Experimental Procedure

Powder specimens were prepared having the compositions: $(La_{0.6}Ca_{0.4})_xCrO_4$, where x = 0.95, 0.98, 0.99, 1.00, 1.01, 1.02 and 1.05. A large amount of Ca (≈40 at%) was added into the system so that $Ca_m(CrO_4)_n$ could be easily observed and analyzed. Samples were synthesized and processed in the low temperature monazite-type form to control the distribution of secondary phases. Above 700°C, $(La,Ca)CrO_4$ transforms into $LaCrO_3$, $CaCrO_4$ and other unidentified minor phases which, at higher temperatures for a $(La,Ca)CrO_3$ solid solution and a $Ca_m(CrO_4)_n$ liquid phase[13].

Powder synthesis was performed using a modified liquid mix process[14] which is schematically illustrated in Figure 1. This process is identical to the procedure we are using to prepare organometallic precursors for spray pyrolysis (SP). This study was not performed on powders prepared using spray pyrolysis as it was initiated prior to completion of the SP system. Drying was

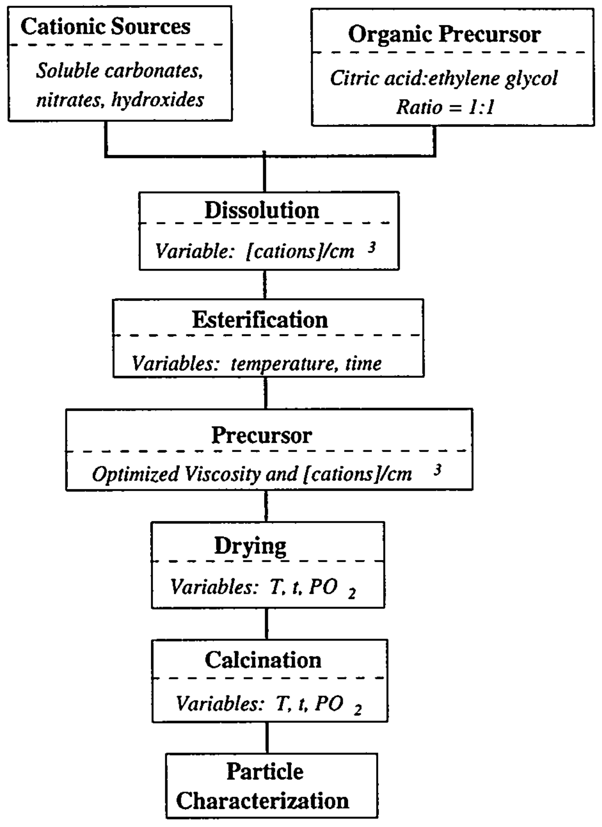


Figure 1. Experimental Overview

performed in an oven at 120°C, which initiated charring. Prior to calcination the char was milled in ethyl alcohol for 8 h to reduce agglomeration. Calcination took place in flowing argon for 8 h at 550°C. Phase analysis was performed using XRD (Scintag, Model 2000).

Calcined powders were uniaxially pressed into 12.7 mm x 3.2 mm disks at 500 MPa. Bulk green and sintered densities were determined using geometric and liquid displacement techniques. For the sintering studies, disks were heated at 5°C/min to temperatures ranging between 1300 and 1450°C for 2 -100 h.

Several specimens with compositions of $(La_{0.6}Ca_{0.4})_{0.99}CrO_3$ (LC99) and $(La_{0.6}Ca_{0.4})_{1.02}CrO_3$ (LC102) were air-quenched to room temperature from various temperatures during a sintering run. The furnace heating rate was 4°C/min, and isothermal holds were made at 1030°C and 1350°C for 2 h. Microanalysis on quenched and sintered samples was performed by SEM and EDS.

The melting characteristics of the liquid phase in LC99 and LC102 were compared with $CaCrO_4$ using DTA (STA Model 409, Netszch).

Results and Discussion

Porosity and Shrinkage

The sintering behavior of $(La_{0.6}Ca_{0.4})_xCrO_3$ compositions were studied by measuring the shrinkage, density and porosity of the samples sintered under various temperature/time profiles. A plot of shrinkage with respect to the (La+Ca)/Cr cation ratio under the sintering conditions of 1400°C/2h is shown in Figure 2. The total shrinkage increases rapidly as (La+Ca)/Cr increases from 0.95 to 0.99. The maximum shrinkage is obtained when the cation ratio is between 0.99 and 1.01.

Density measurements were difficult to correlate, because the amount of exsolved $Ca_m(CrO_4)_n$ changed throughout the process, making the theoretical density difficult to track. However, the open porosity, shown in Figure 3, directly correlates to the densification behavior. We see the open porosity decreases steadily as (La+Ca)/Cr increases from 0,95 to 1.0., and approaches zero between 1.01 and 1.05.

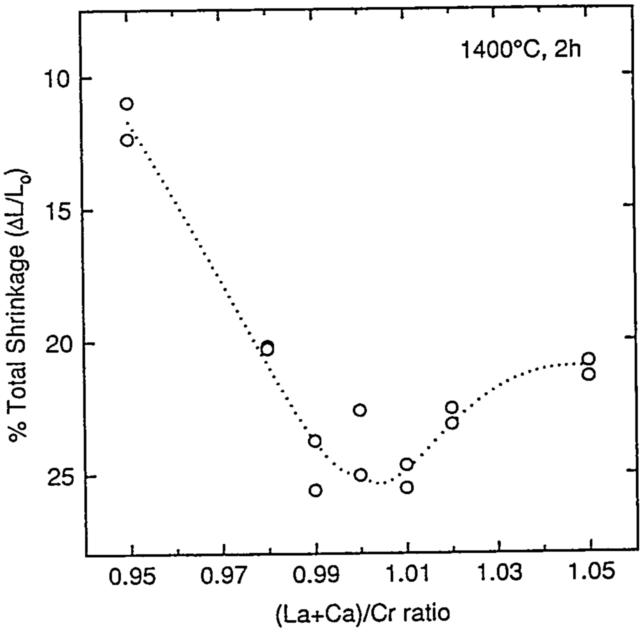


Figure 2: Percent total shrinkage vs (La+Ca)/Cr ratio of $(La_{0.6}Ca_{0.4})_xO_3$

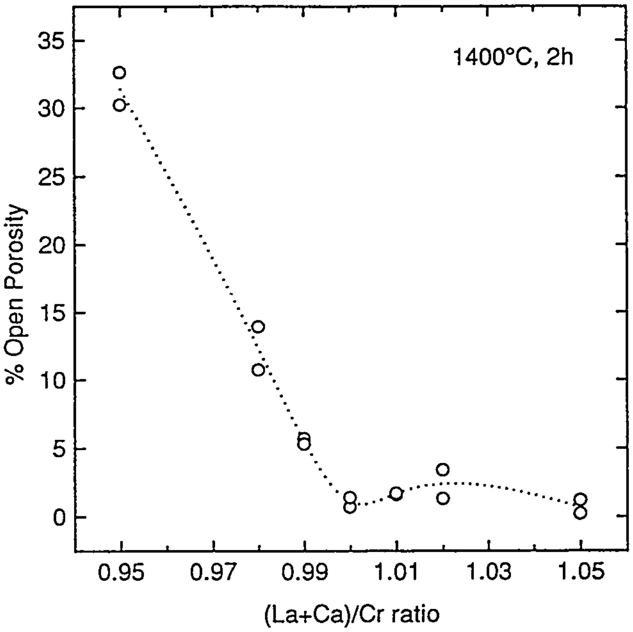


Figure 3: Percent open porosity vs (La+Ca)/Cr ratio of $(La_{0.6}Ca_{0.4})_xO_3$

Sintering Behavior

Evidence of the liquid phase was readily observed in quenched specimens. Figure 4 is an SEM micrograph showing the recrystallized liquid as a dark, glass-like mass enveloping the lighter $(La,Ca)CrO_3$ particles. Similar microstructures were common in all of the bulk $(La_{0.6}Ca_{0.4})CrO_3$ compositions under similar conditions. According to EDS, the bulk quenched liquid is composed of Ca, Cr and O. XRD confirmed the presence of mainly $CaCrO_4$ and sometimes other $Ca_m(CrO_4)_n$ phases.

The sintering of LC99 and LC102 were compared by SEM analysis on a series of quenched samples. Figures 5-7 show SEM micrographs of fracture surfaces which show the behavior of the two compositions. Each specimen pair was air-quenched from similar temperatures during the sintering process.

Figure 8 is a cross section of LC102, which shows a dark layer which has formed on the free surface. The liquid has spread into a uniform layer along the free surface after being squeezed out of the grain boundaries as the grains coalesced. The surface layer was identified by XRD as $Ca_5(CrO_4)_3OH$. Figure 9 shows a back-scatter image of the cross section of $Ca_m(CrO_4)_n$ crystals on the surface of LC99 and Figure 10 is a top view of a polished specimen. Surrounding this mass is a large porous region with a radius of ≥100 µm. EDS indicated that these crystals are Ca-poor, although XRD only showed the presence of $CaCrO_4$.

Annealing experiments demonstrated the difficulty in dissolving large amounts of Ca into $(La,Ca)CrO_3$. Specimens containing 40at% Ca with (La+Ca)/Cr cation ratios of 0.95 to 1.05 contained residual, re-crystallized liquid in the grain boundaries after long term annealing. For example, Figure 11 is a back scatter image of LC105 sintered and annealed at 1300°C for 100 h. According to EDS, the dark cubic phase contains Ca and O.

Melting Behavior

The characteristic of liquid phase formation with respect to stoichiometry, are further illustrated by comparing the DTA curves of $CaCrO_4$, LC102 and LC99, as shown in Figure 12. In the upper set of curves, the onset of melting takes place at nearly the same temperature for each composition. In particular, the $CaCrO_4$ and LC99 curves display a similar, single-step melting process. The double peaks corresponding to LC102 indicate that the associated liquid phase melts in two stages.

The crystallization peaks in the lower section occur at lower temperatures which reflects supercooling of the liquid. The $CaCrO_4$ composition crystallizes first, and the other two liquids crystallize ≈10°C lower. The liquid in LC102 again shows a double peak which suggests a two step recrystallization process.

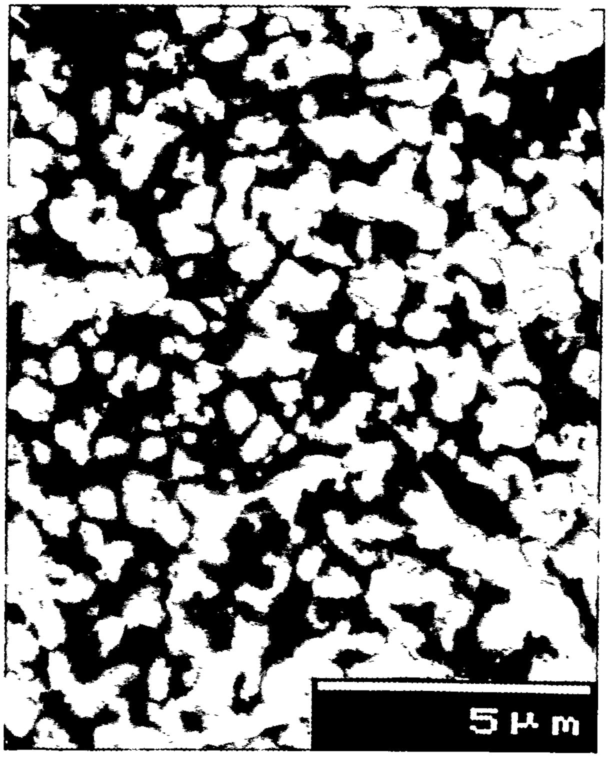


Figure 4: Polished surface of LC95 heated to 1350°C

Discussion

The large amount of exsolved $Ca_m(CrO_4)_n$ in the system made it difficult to correlate the sinterability of $(La_{0.6}Ca_{0.4})_xCrO_3$ compositions directly by shrinkage or density measurements. For example LC99 had a shrinkage comparable to LC101, which contradicts the results of Sakai[15] and Chick[16]. Possibly, enough liquid was present to overcome some of the effects of stoichiometry. Obviously, the amount of total Ca added to these

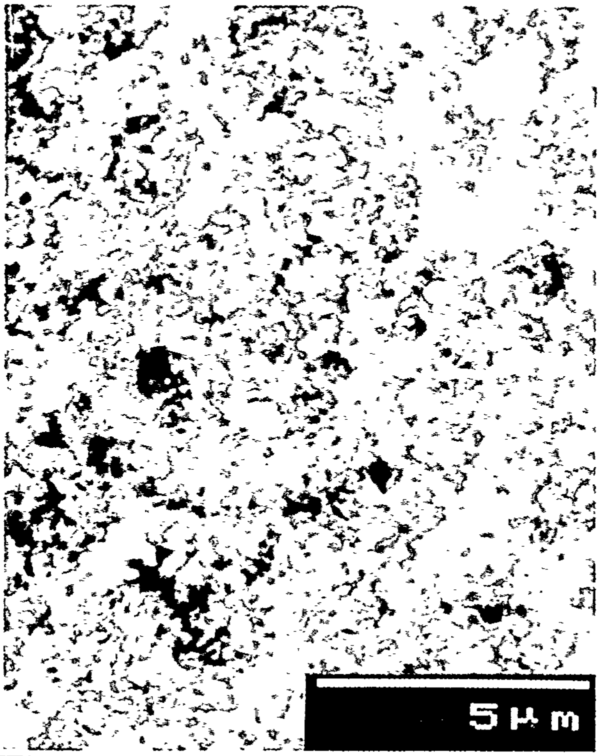


Figure 5a: Fracture surface of LC99 heated to 740°C

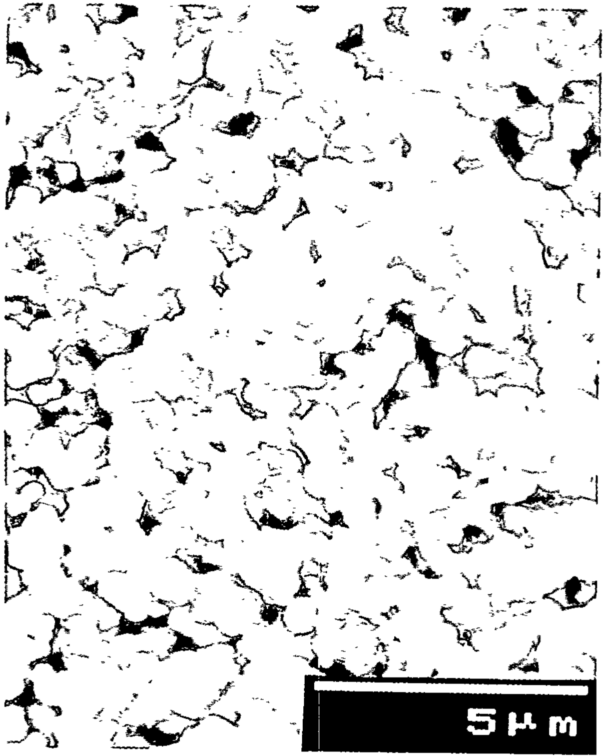


Figure 6a: Fracture surface of LC99 heated to 1030°C

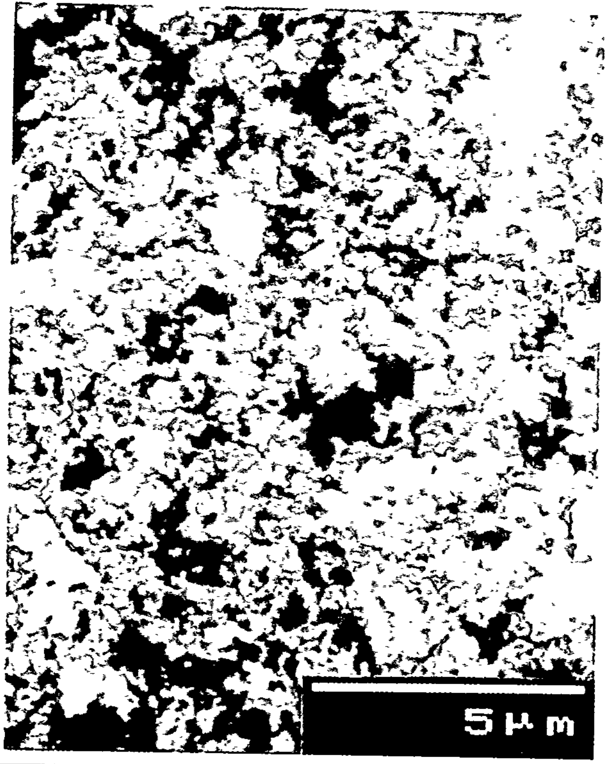


Figure 5b: Fracture surface of LC102 heated to 740°C

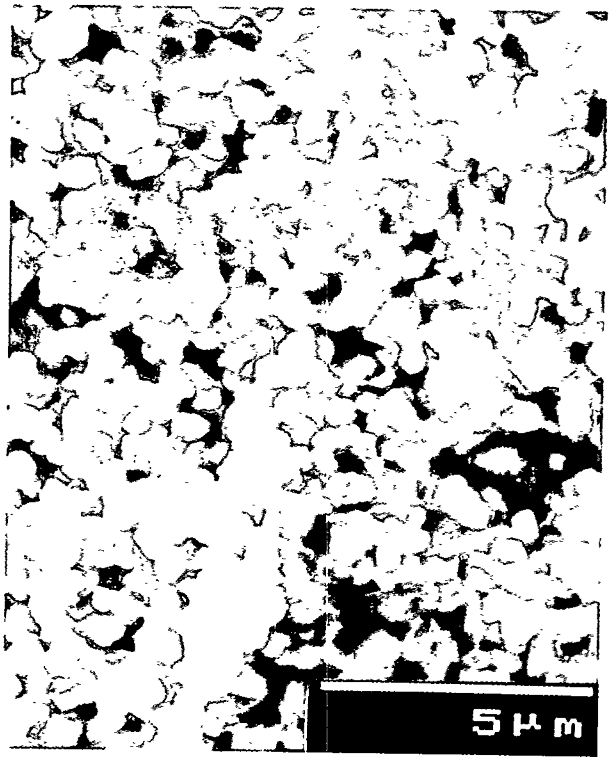


Figure 6b: Fracture surface of LC99 heated to 1030°C

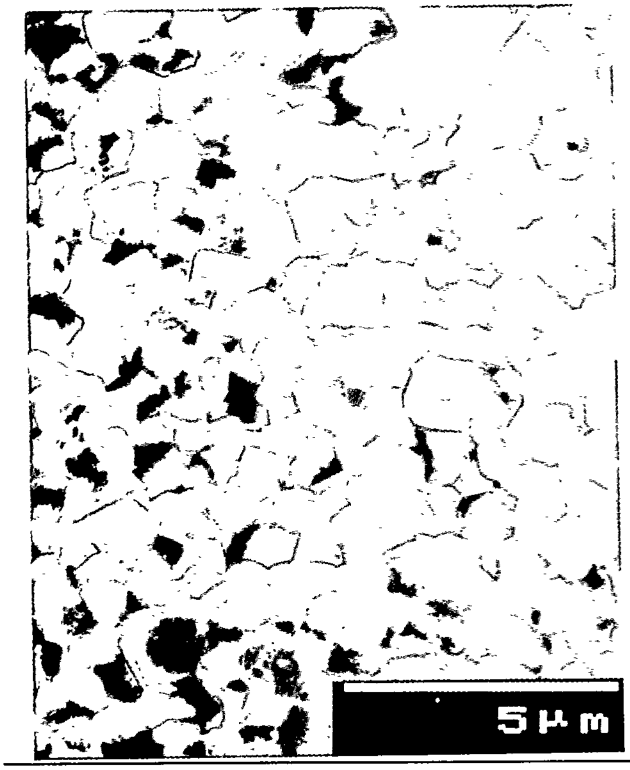


Figure 7a: Fracture surface of LC99 heated to 1350°C

Figure 8: Fracture surface of LC102 heated to 1350°C

Figure 7b: Fracture surface of LC102 heated to 1350°C

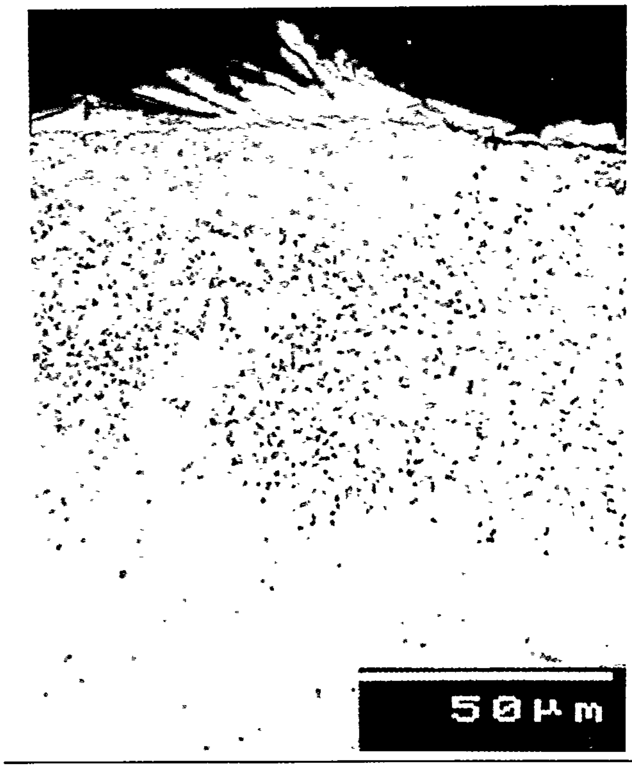


Figure 9: Fracture surface of LC102 heated to 1350°C

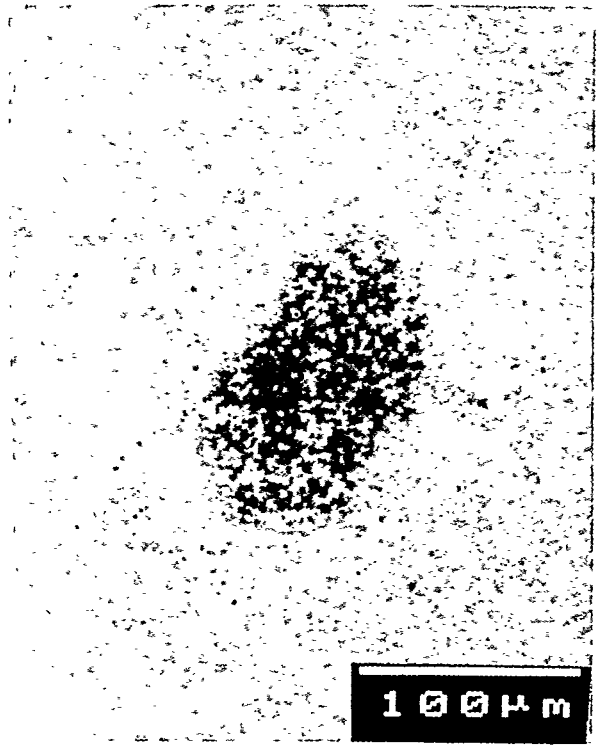


Figure 10: Top surface of LC102 heated to 1400°C

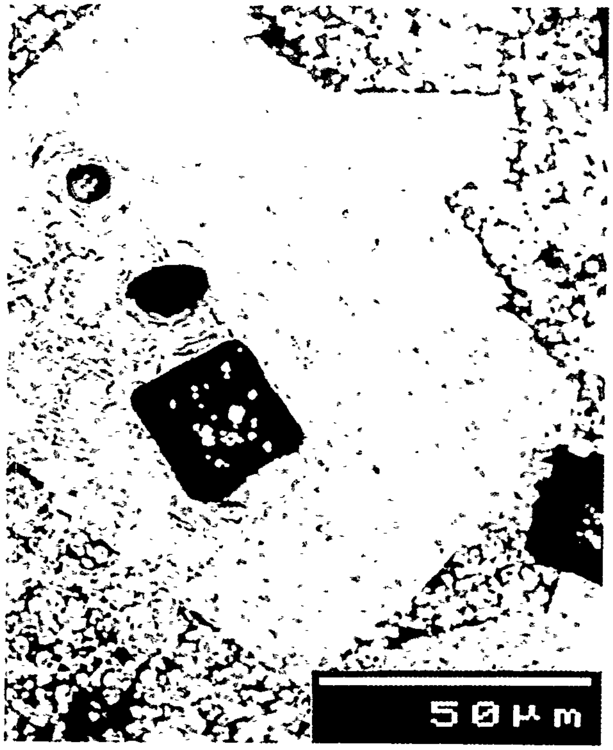


Figure 11: Polished surface of LC105 heated at 1300°C-100h

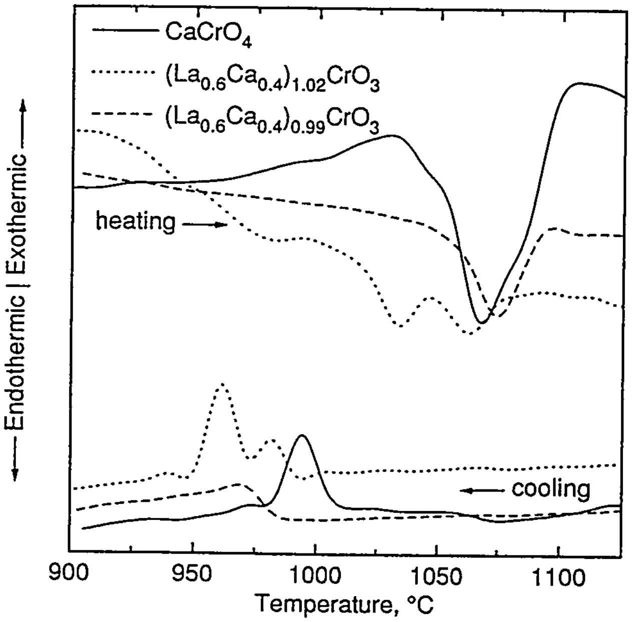


Figure 12: DTA comparison of various compositions.

compositions exceeded optimum values and should be reduced if these compositions are to be used as an interconnect. This is evident from the annealing experiment. A former study[9] suggested that < 25at% Ca in (La,Ca)CrO_3 approaches an optimal composition.

Notable differences in the densification became evident from porosity measurements. The A-site deficient compositions show a higher open porosity than their A-site excess counterparts.

Microscopic analysis of a series of quenched specimens provided a visual investigation of the sintering. Initially, specimen pairs of A-site excess and deficient compositions followed the same general steps of densification. Both transformed to the perovskite structure below 740°C as evidenced by XRD. Grain growth has occurred so that the primary particles can be resolved.

From previous work it is known that $CaCrO_4$ loses oxygen and begins to form a solid solution with $LaCrO_3$. Excess Ca which is not incorporated into solid solution segregates along with Cr and O to the free-surface in both A-site excess and A-site deficient materials.

Upon melting $Ca_m(CrO_4)_n$, all compositions experienced rapid grain growth. The recrystallized liquid is identified in either the figures as darker, irregular masses; the lighter equiaxed grains are (La,Ca)CrO_3. It appears that $Ca_m(CrO_4)_n$ segregates from (La,Ca)CrO_3. Continued heating was accompanied with further grain growth and spreading of the liquid.

From these series of micrographs, the sinterability of the two compositions appear to be quite similar. In both cases a liquid phase forms and spreads, grain growth occurs at similar rates, and the final grains sizes are comparable. This did not. allow any explanation to the observed differences in shrinkage and porosity. However, differences became apparent by examination of the specimen free surfaces.

A comparison of Figures 8 and 9 illustrate the difference between the liquid phases in the final sintering stages of A-site excess and deficient compositions. The liquid on the free surface of the A-site excess specimen has spread into a uniform 3 µm layer, demonstrating good wetting characteristics. This is desirable for optimum liquid phase sintering. In contrast, the $Ca_m(CrO_4)_n$ phase in the A-site deficient material formed a 100 µm diameter island on the surface. The large volume of porosity surrounding this island is a result of the segregation of the liquid from the matrix. This $Ca_m(CrO_4)_n$ phase appears to be more crystalline, and it is believed that it contained Ca-Cr-O solids throughout sintering, because $CaCr_2O_4$ whiskers were observed to grow from some of the A-site deficient compositions while at the sintering temperature.

The divergent behavior of the $Ca_m(CrO_4)_n$ compositions towards densification may be understood by reference to the CaO-Cr_2O_3 phase diagram shown in Figure 13. Assuming (La,Ca)CrO_3 retains its stoichiometry with respect the A and B cations, any nonstoichiometry within a given system is presumed to appear in second phases. Therefore, when Ca dissolves into the (La,Ca)CrO_3 matrix, it is accompanied with stoichiometric amounts of Cr and O. This process of dissolution consequently induces a change in composition of the $Ca_m(CrO_4)_n$ phase in nonstoichiometric systems.

The change in composition of $Ca_m(CrO_4)_n$ while heating is indicated by the dashed lines on the phase diagram. The lines are estimated and their curvature will depend upon the kinetics of dissolution of $CaCrO_4$ into (La,Ca)CrO_3. The initial composition of $Ca_m(CrO_4)_n$ at low temperature depends upon the A/B ratio and the amount of exsolved $CaCrO_4$. A different A/B ratio in (La,Ca)CrO_3, or mole fraction of $CaCrO_4$ will shift the initial composition of the secondary phase along the composition axis. For example if (La,Ca)CrO_3 was initially prepared as A-site excess, and no $CaCrO_4$ was exsolved, the secondary phase would be CaO.

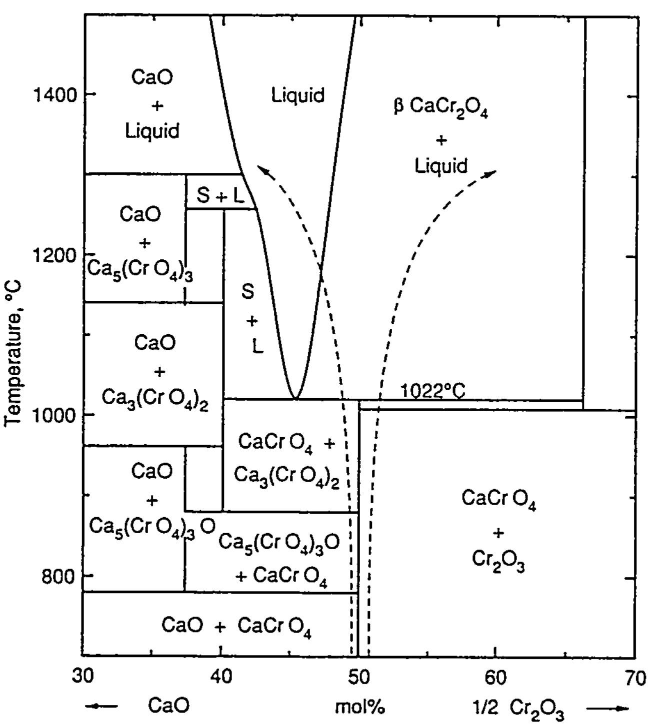


Figure 13: Phase diagram of the CaO - Cr_2O_3 system.

Upon heating of A-site excess (La,Ca)CrO_3, the initial composition of the exsolved phase starts to the left of the $CaCrO_4$ composition line. As the temperature is increased the Ca solubility also increases, and Ca, Cr and O begin to dissolve into the perovskite matrix. The Ca content of $Ca_m(CrO_4)_n$ will increase towards the CaO end member. At 1022°C, a liquid phase forms with some amount of solid β-$CaCr_2O_4$. As heating continues, the liquid composition changes towards CaO and the solid B-$CaCr_2O_4$ disappears. At some point the melt becomes 100% liquid. The SEM study showed that good wetting behavior is exhibited by this liquid phase system throughout the sintering process.

In A-site deficient (La,Ca)CrO_3, the Ca content of $Ca_m(CrO_4)_n$ decreases towards the

Cr_2O_3 end member with increasing temperature and Ca solubility. At some point, a solid network is formed between $\beta-CaCr_2O_4$ and the $(La,Ca)CrO_3$ grains and densification by particle rearrangement stops. The remaining liquid eventually segregates from the matrix, forming islands on the free surface. Regions surrounding these areas are typically porous.

DTA results also indicated deviating behavior in the opposing liquid phases. The phase diagram indicates two melting peaks should be exhibited, one near 1022°C, the eutectic temperature, and one at a higher temperature upon complete melting of the liquid. Conversely, the DTA curve of Cr-rich $Ca_m(CrO_4)_n$ would contain only one broad peak, beginning at the eutectic temperature since there would be no other reaction expected.

Conclusion

The present results from our studies on the densification behavior of $LaCrO_3$-based interconnects has clearly delineated the effects of stoichiometry. A liquid phase forms in both A-site excess and deficient compositions, however the final stages of sintering differ due to changes in the composition and resultant wetting behavior. The melt in the A-site excess material becomes 100% liquid, and retains its wetting behavior, while the A-site deficient material does the opposite.

SOFC PERFORMANCE STUDIES

In this portion of the research we are focusing on the microstructure ↔ property relations in solid oxide fuel cells (SOFC's) to better understand the mechanisms involved in cell performance. The aim is to fabricate SOFC's with controlled microstructures utilizing $La_{1-x}Sr_xMnO_3$ (LSM), yttria stabilized zirconia (YSZ), and Ni-YSZ composites as the cathode, electrolyte, and anode, respectively. The specific objectives of this research project are to:

→ Fabricate SOFC components with controlled microstructures (i.e. grain size and porosity of the cathode/anode).

→ Gain a better understanding of the mechanisms involved in improving cell performance via electrochemical and impedance techniques, and correlate these results to the microstructure.

This aspect of the project is divided into three phases:

I. Construct a system for measuring planar SOFC's

II. Processing and forming of trilayer structures of anode/electrolyte/cathode

III. Electrical characterization of single cells by electrochemical and impedance techniques.

Phase I has been completed whereas Phase II and III are presently ongoing and will overlap during the course of this research.

I. System Construction

The system built for measuring cell performance is completed and shown in Figure 14. The apparatus is a two tube design capable of operating at temperatures up to 1100°C. The oxygen activity of the oxidant and fuel is controlled by mass flow controllers and the pO_2 is monitored using a ZrO_2 sensor. The electrical characterization, I-V, current density, and interfacial characterization, will be carried out using a computer controlled Schlumberger Impedance/Gain-Phase Analyzer (1286) and Electrochemical Interface (1260).

The apparatus was calibrated using a YSZ electrolyte with Pt anode/cathode. Results of open circuit potential were in good agreement with theoretical values.

II. Cell Fabrication

At present, powder preparation and forming techniques are underway to fabricate planar cells. Various powder preparation techniques have been investigated, Pechini liquid mix, Glycine - Nitrate, and mixed oxide, to vary the starting powder characteristics. A tape casting, laminating, and

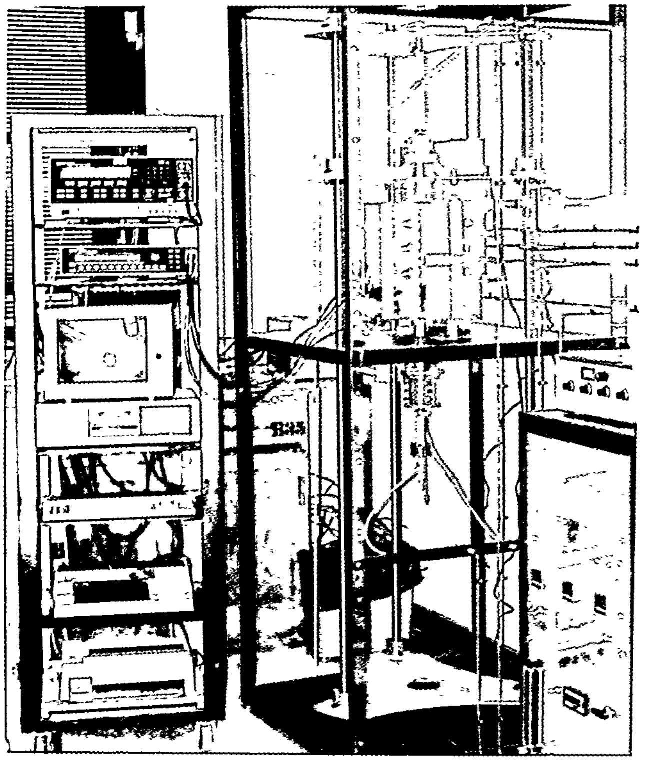


Figure 14: Cell measurement system.

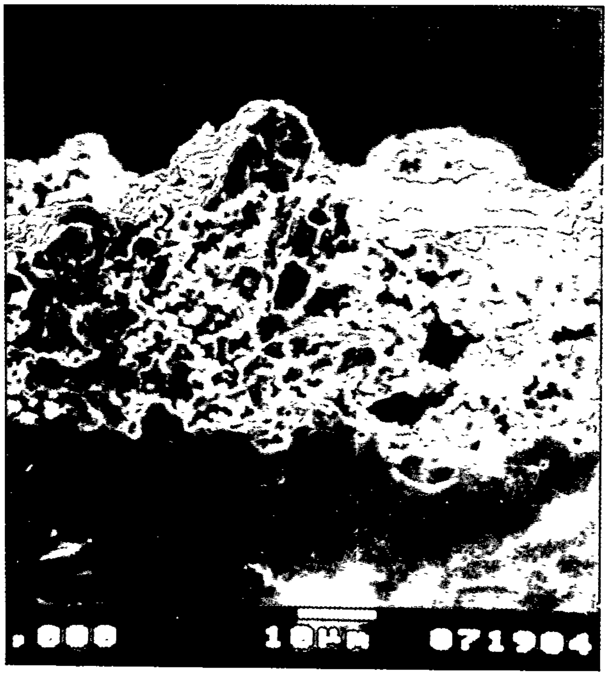


Figure 15: YSZ: Anode couple.

screen printing fabrication scheme is currently being used to prepare trilayer structures.

The YSZ electrolyte and Ni-YSZ anode are currently co-fired at 1400°C. Commercially available YSZ can be sintered to near theoretical density and has a final grain size of approximately 5 µm. The anode is deposited by screen printing and is made porous by introducing a fugitive phase which is burnt out during densification. The final microstructure (i.e. Ni grain size, pore size, pore size distribution, and % porosity) is dictated by the powder and fugitive phase characteristics and amount of fugitive phase added. A typical fracture surface of a YSZ/ Ni-YSZ couple is shown in Figure 15.

After co-firing the electrolyte and anode, a LSM paste is screen printed onto the electrolyte followed by a 1100°C heat treatment. Different powder preparation techniques enable various LSM microstructures to be fabricated. Distinctly different microstructures have been attained as shown in LSM/YSZ couples, Figures 16 and 17.

The overall goal in fabricating planar cells is to systematically vary the microstructure (i.e. grain size, % porosity) of the cathode and anode to better

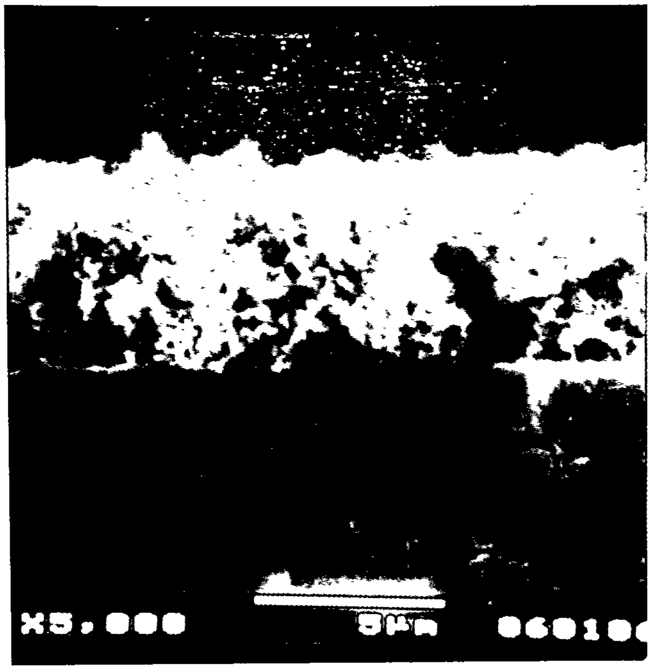


Figure 16: LSM anode - fine scale, high TPB.

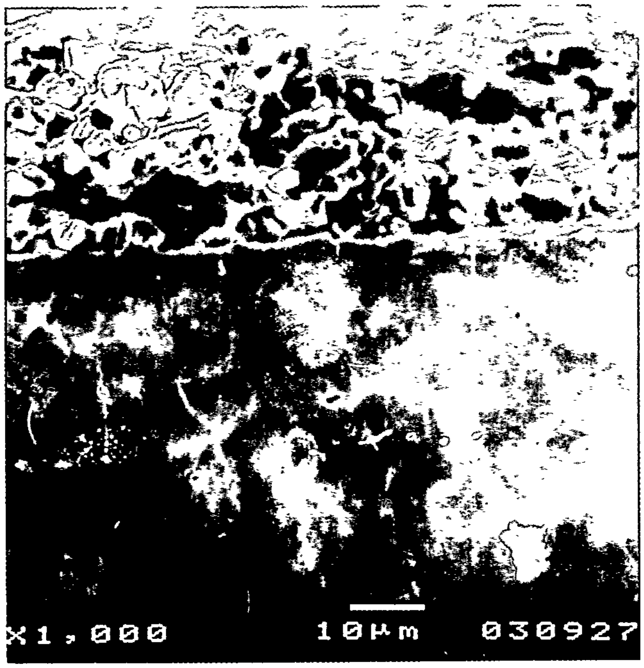


Figure 17: LSM anode - fine scale, low TPB.

understand the factors effecting cell performance. Some questions which should be answered are:

- → What effect does the triple phase boundary (pore, YSZ, LSM) have on performance?
- → Can a dense cathode be used if there is sufficient ionic conductivity at a low overpotential?
- → Is long term stability sensitive to the microstructure of the electrodes?
- → Is there a critical thickness for the anode and cathode?
- → Would a composite structure of fine grain material next to the electrolyte with coarse grain structure composing the remainder of the electrode be more efficient?
- → Can results be reproduced?

III. Electrical Characterization

A Pt/YSZ/Pt cell has been used to calibrate the system with results that correlate well with theoretical values. At present electrochemical and impedance techniques are being explored through literature review and experimental studies.

Some typical experimental studies which will be performed include:

- → I - V, current density and open circuit potential as a function of temperature, time and pO_2
- → Impedance spectra as a function of temperature, time and pO_2

FUTURE WORK

With respect to the major thrusts of this program:

Sintering of $LaCrO_3$-based Powders

Specific Tasks:

A. Reproducible Green State Synthesis via Tape Casting

- ♦ Develop the tape casting system & characterize the dispersive behavior of the spray-pyrolyzed powders in the binder system.

B. Further Characterization of the Chromate Liquid Phase

- ♦ Composition
- ♦ Volume fraction
- ♦ Viscosity as a f(T, composition)
- ♦ Temperature stability
- ♦ Wettability on $LaCrO_3$-based powders (contact angle measurements)

Co-sintering of $LaCrO_3$-based Powders with Cathode and Electrolyte Materials

Goal: Understand why LCC and YCC do not densify when in contact with ZrO_2 or LSM

Specific Tasks:

A. Identify the Underlying Cause

- Contact angle measurements of liquid phase on LSM and ZrO_2
- Diffusion couples
  - → dense & porous specimens
  - → microstructural analysis of the interface (microprobe)

B. Develop Approaches to Solve the Co-Sintering Problem

- If Ca^{2+} segregation (surface diffusion, preferential migration):
  - → decrease the Ca activity gradient (Δa_{Ca}) by using Ca-doped $LaMnO_3$ and ZrO_2
- If wicking:
  - → alter pore size distributions to diminish the driving force - this knowledge will come out of consideration of the viscosity of the melt & the contact angles - the pore size needed to prevent wicking can then be calculated.
  - → alter wetting behavior via dopants to the liquid phase (changes in viscosity - incorporation of ions that bond well to Cr^{3+} compared to Mn^{3+} or Zr^{4+})

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P9 Sealant Materials for the Solid Oxide Fuel Cell

CONTRACT INFORMATION

Contract Number	**49638**
Contractor	**Argonne National Laboratory** **9700 South Cass Avenue** **Argonne, IL 60439** **(708) 252-4342**
Contractor Project Manager	**Kevin M. Myles**
Principal Investigators	**Michael Krumpelt and Romesh Kumar**
Co-Investigators	**Ira Bloom and Kevin L. Ley**
METC Project Manager	**William C. Smith**
Period of Performance	**April 1, 1991 to June 30, 1994**

OBJECTIVE

The objective of this work is to develop sealing materials for solid oxide fuel cells (SOFCs). A suitable sealant must form strong, dense bonds with SOFC components, be chemically stable and mechanically compatible with the components at the operating temperature and over the temperature range of interest, be chemically stable in the operating environment of the SOFC (i.e., H_2 and H_2O on the anode side, O_2 on the cathode side), and must be electronically insulating.

BACKGROUND AND PRIOR WORK

Advanced flat-plate, SOFC stacks under development offer high electrochemical performance because of their short current paths. However, in such a planar SOFC stack, the individual cells must be sealed at the edges, and the stack itself must be sealed to the gas supply and exhaust manifold.

In general, the sealant must be compatible with the cell components and the cell environment. If excessive reactions between the sealant and cell components or gases occur, they can lead to cell failure. This effort seeks to develop materials that are chemically stable toward the cell gases and that form dense, adherent bonds to cell and stack components.

For use in the planar SOFC, the sealant must be electrically insulating. Since it would contact both the anode and cathode, any electronic conductivity in the sealant will produce undesirable shunt current flow.

The thermal properties of the sealant must match its application. At the cell edge, the sealant must be compatible with the thermal expansion behavior of, at most, two ceramics. For example, in the cathode compartment, the sealant must be matched to the behavior of yttria-stabilized ZrO_2 (YSZ) and $La(Sr)MnO_{3-x}$ (LSM). Rigid sealants are appropriate here. Along the stack-to-manifold junction, the thermal requirements are different. Because of the number of different ceramics present (anode, cathode, electrolyte, interconnect, and the manifold) and the long sealing distances, sealants which can tolerate some thermal mismatch may be necessary.

Borate-based sealants were chosen for several reasons: they are electrical insulators, they possess the desired combination of viscosity and expansion coefficient in the temperature region of interest, and they contain no mobile ions. A borate-based material was preferred over a silica-based one since, in addition to eliminating any possible SiO_2 reduction, target viscosities in the desired temperature range (800–1000°C) could be achieved without loss of compositional flexibility.

Last year, the properties of one glass-ceramic sealant, Material K, were reported. Material K was found to produce extremely adherent and durable bonds to coupons of YSZ, NiO/YSZ, and LSM. The glass-ceramic also bonded very well to disks of alumina. No obvious signs of interfacial reactions, such as phase formation, were observed between the glass-ceramic material and any of the bonded substrates. Sealant/substrate composites were thermally cycled twice between room temperature and 1000°C to gauge the thermal compatibility of the sealant with substrate materials. No delamination or cracking was observed.

The sealing ability of the glass-ceramic was tested in an electrochemical cell consisting of a 500-μm-thick YSZ disk with platinum screen electrodes on either side. A 1-mm-thick O-ring gasket of the glass-ceramic was used as the seal. The sealing operation itself consisted of pre-shrinking the gasket at 800°C and then using the gasket to bond a zirconia tube to the disk at 1000°C. The cell was then tested for electrochemical performance. Flowing, humidified 5% H_2 in helium served as the fuel gas, and the stagnant furnace air as the oxidant. The cell EMF was measured at 800, 900, and 1000°C. All EMF values were 99% of theoretical. During the test, the cell was thermally cycled between 400 and 1000°C five times and between room temperature and 1000°C once. The EMFs after thermal cycling were within 1–2 mV of previous values.

PROJECT DESCRIPTION

Argonne is continuing development of a family of glass- and glass-ceramic sealants. The advantage of using these glass-based materials over rigid ceramics as seals is that expansion-mismatch-induced stresses between components occur over only a fraction of the cooling cycle, hence reducing the stresses produced. The temperature at which stresses start to build corresponds to the temperature at which the viscosity exceeds 10^{12}–10^{13} Pa-s and the glass becomes essentially a solid. This temperature is approximately the same as the glass-transition temperature and should be as low as possible. At the same time, the viscosity of the sealant at the fuel cell operating temperature should be greater than 10^3 Pa-s (below which glass flows readily) and, hence, should not be excessively fluid.

The first test of a sealant is its ability to bond to the fuel cell components: YSZ electrolyte, porous NiO/YSZ anode, and LSM cathode. The next step is to determine the interactions between the fuel cell components and the sealants as a function of time at 1000°C and under humidified-hydrogen atmospheres. The bonding ability of various candidate sealing materials to these components is determined qualitatively. Sealant materials that form strong bonds are subsequently examined by microscopy to determine porosity and extent of chemical interaction.

Promising materials are characterized further to evaluate both the thermal expansion behavior and viscous properties over the temperature regimes of interest. Since candidate sealing materials have to be electronically insulating, electrical properties are also evaluated using standard four-point dc techniques.

The final stage of the sealant development is evaluation by external companies. Presently, two glass-ceramic materials are in this stage of evaluation.

RESULTS

Several additional sealants from the borate-based family have been made to determine the feasible range of coefficients of thermal expansion (CTEs) and viscosity versus temperature. The developed sealant materials have CTEs in the range of about 8–12 × 10^{-6}/°C, which spans the CTE values needed for SOFC applications. To illustrate the behavior and flexibility of these materials, a dilatometric curve of Material K (a glass-ceramic) is given in Fig. 1 along with that obtained from commercial YSZ. As can be seen from the figure, there is excellent thermal matching up to temperatures where the sealant exhibits viscous flow.

The viscosity vs. temperature profiles for four sealant compositions (the glass-ceramic K and the glasses 42, 43, 47) were determined by a sagging beam method [1, 2]. These profiles are given in Fig. 2 and show the range of viscosities feasible by changing composition and microstructure. From the figure, the glass-ceramic material (Material K) is much more viscous and its viscosities are not as sensitive to temperature as glass (Materials 42, 43, and 47). The viscosity curves for the three glasses illustrate the range feasible by changing composition only. Adding the crystalline phase makes the slope of the curve less steep.

These sealants were found to form bonds at temperatures where their viscosity is 10^6 Pa-s or less. From Fig. 2, such temperatures are 1000 and 675°C for Materials K and 43, respectively.

Four-point dc measurements were used to determine the electrical properties of the glass-ceramic at three different oxygen partial pressures (0.21, ~10^{-10}, and ~10^{-19} atm), in the temperature range of 800 to 1000°C (see Fig. 3). The electrical conductivity at a given temperature was found to be independent of oxygen partial pressure in the range studied, confirming that the conductivity is ionic and not electronic.

Structures similar to the cell edge and stack-to-manifold junction were bonded using these sealants to verify their usefulness. Two types of cell edge structures were assembled. One consisted of ceramic interconnect, anode, and electrolyte. The other was made up of interconnect, cathode, and electrolyte. Both types were thermally cycled 7–10 times in air. In addition, the anode-containing structures were thermally cycled 10 times in humidified 5% H_2 in helium. No cracking was observed in either structure type.

The stack-to-manifold seal was found to be strongly affected by geometry. In the electrolyte-edge-sealed-to-interconnect configuration (A), cracking reproducibly occurred in the electrolyte. In the interconnect-edge-sealed-to-electrolyte configuration (B), structures could be sealed without cracking. Since the electrolyte is the less expansive and stronger material present, the first observation is contrary to expectation; the interconnect should crack first.

Finite-element analyses employing macro- and micro-models were used to aid understanding of the results. In configuration (A), the electrolyte component invariably cracked in a characteristic mode. Modeling indicated that tensile stresses exceeding the fracture strength of the electrolyte developed through the full thickness of the electrolyte normal to the sealant line, leading to crack propagation. In configuration (B), modeling revealed that the tensile stresses present were localized at the corners of the interconnect and in the sealant line. The analyses also showed that the angle between the plates has a minor influence on the overall stress patterns developed at stack-to-manifold junctions.

FUTURE WORK

Testing of glass- and glass-ceramic sealants by industrial fuel cell developers is underway. Preliminary results are encouraging. Argonne is working with these developers to tailor sealant materials to their specific SOFC materials and cell geometries and is determining materials interactions under cell operating conditions.

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[1] Hagy, H. E., J. Am. Ceram. Soc. 46(2) (1963) 93.

[2] Strnad, Z., **Glass-Ceramic Materials**, Glass Science and Technology 8, Elsevier, New York, 1986, pp. 162-165.

This work is supported by the U.S. Department of Energy under contract no. W-31-109-Eng-38.

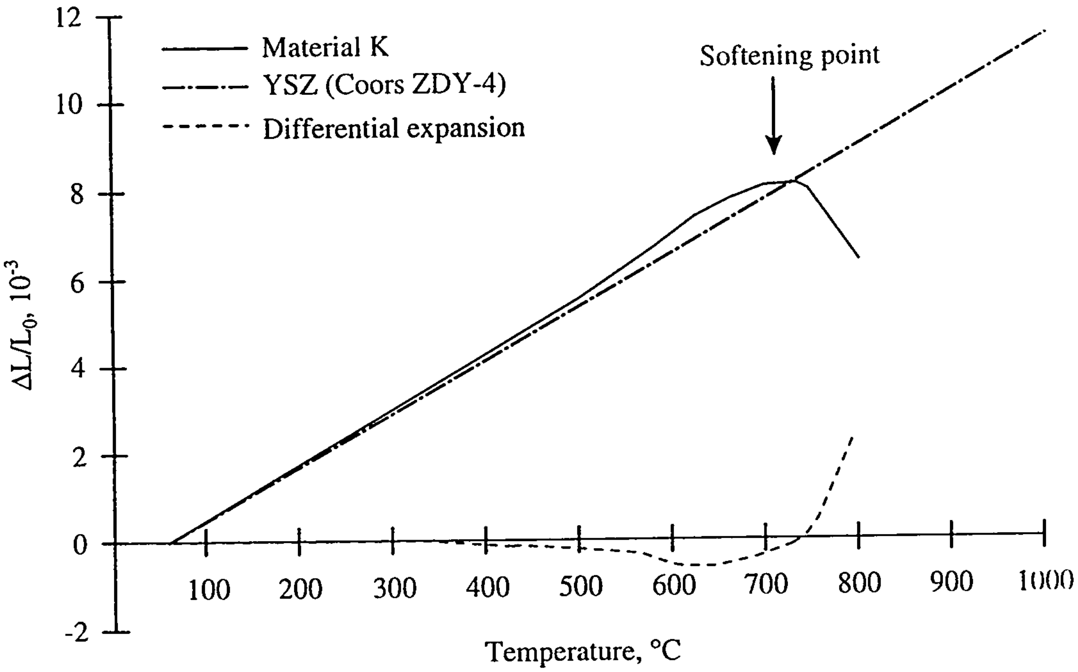

Fig. 1. Thermal Expansion Behavior of Material K

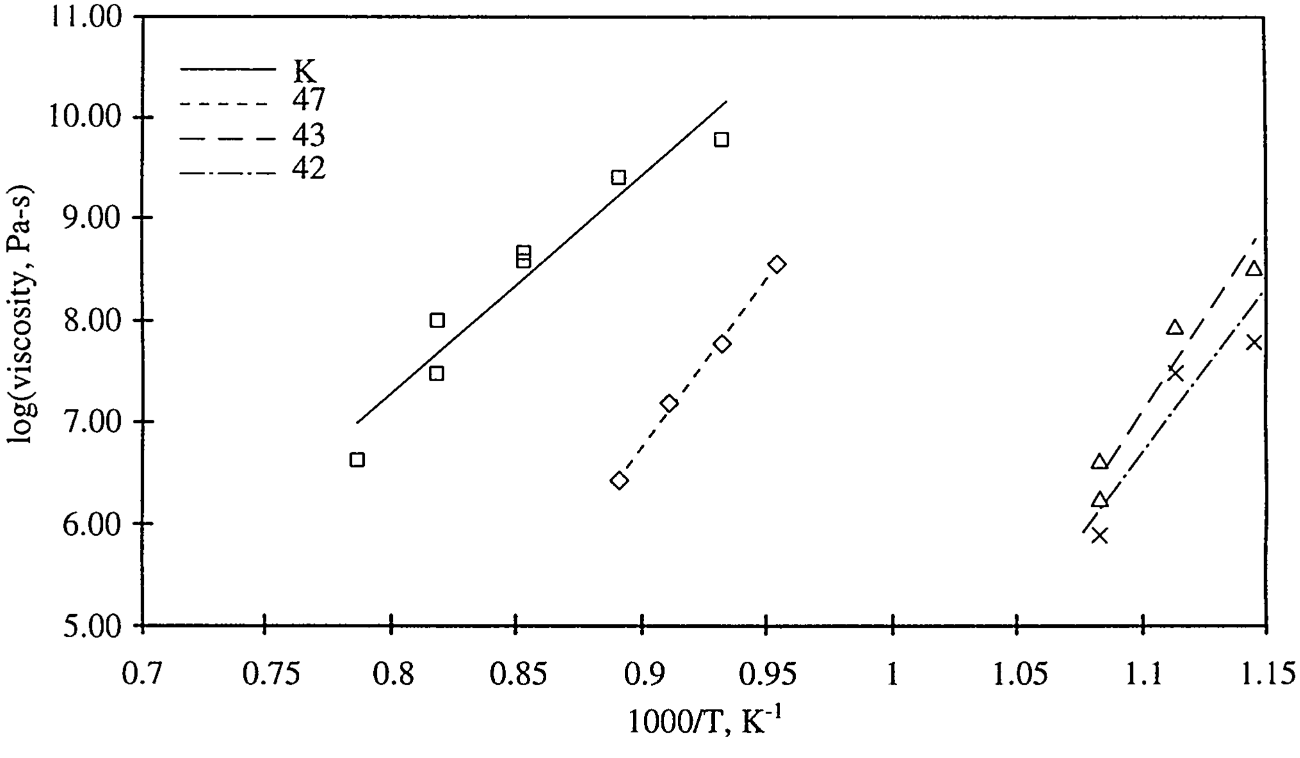

Fig. 2. Arrhenius Plot of Viscosity vs. Temperature for some Sealant Materials.

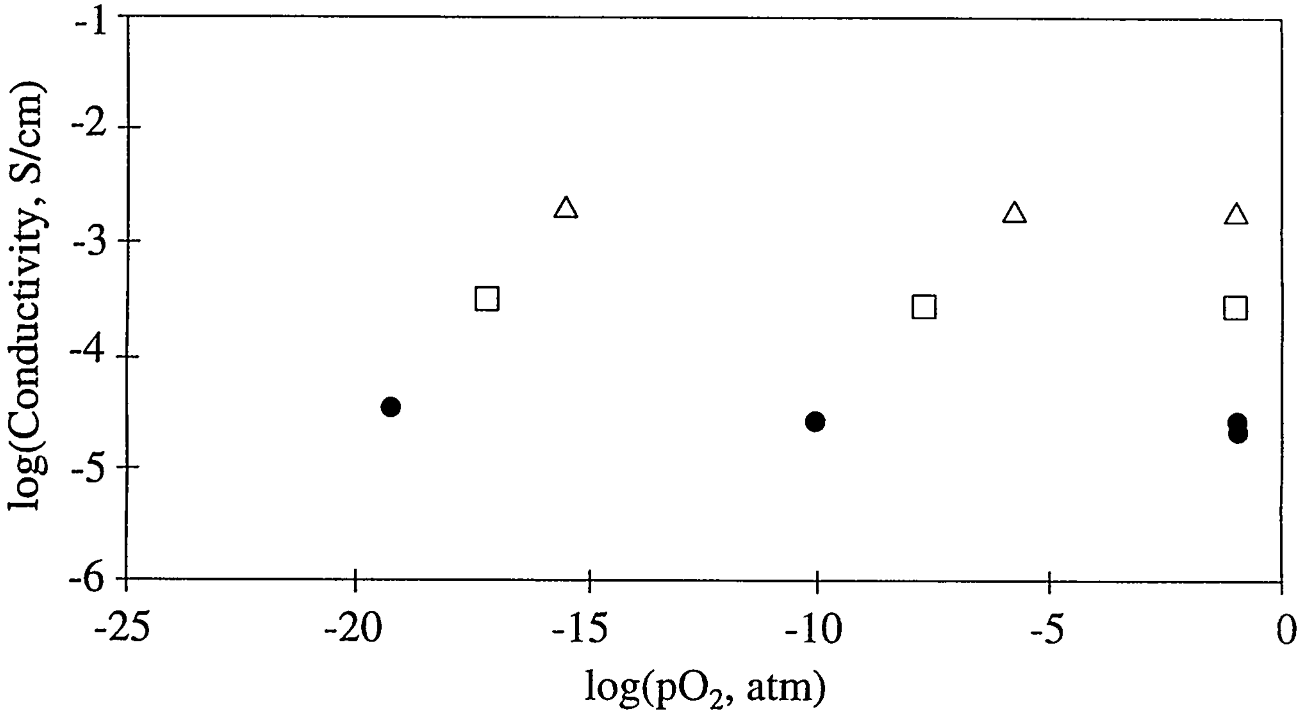

Fig. 3. Dependence of Conductivity of Material K on Oxygen Partial Pressure.

P10 Trace Species Removal for Fuel Cell Application

CONTRACT INFORMATION

Contract Number:	DE-FG058-92ER81330
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Principal Investigators:	A. Pigeaud A. Gelb, PSIT-PowerServe
METC Project Manager:	Norman T. Holcombe
Period of Performance:	7/27/92 to 5-24/1994

ABSTRACT

The development of advanced, potentially very clean, coal-fired energy conversion requires integration with reliable and efficient, cleanup systems which can remove almost all gaseous contaminants and metallic vapor species from high-temperature, high-pressure gas streams down to sub-ppm concentration levels. In the thermal as well as electrochemical conversion systems, the removal of polluting trace elements to very low levels is becoming a mandatory necessity both in terms of the decay that contaminants such as: HCℓ, metal sulfides, arsine, alkali, metals, zinc, cadmium, lead and mercury vapors, can have on energy conversion equipment, and that can also seriously affect the environment if emitted into the atmosphere.

The objective of current work at ERC has therefore been to develop a novel, high temperature polishing concept using a liquid-metal-alloy 'getter' for scrubbing hot coal-derived fuels. A patent has been applied for this invention which basically proposes to scrub hot reducing gases with a reactive liquid metal alloy by pumping it at elevated temperatures over a high efficiency, gas/liquid-film, contacting device in counter-current fashion, This is to remove a number of contaminants and hazardous air pollutants (or HAP's) from hot fuel gas containing very low levels. Results from equilibrium thermogravimetric analysis are quite promising with respect to arsenic and a number of other metallic species. Reactor design for the scrubber has been completed and is currently being fabricated.

P11 Macrocyclic Redox Promoters for Direct Methanol Fuel Cells

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Don Maricle
International Fuel Cells, Windsor CT

CONTRACT INFORMATION

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Principal Investigator	Shantha Sarangapani
METC Project Manager	Venkat K. Venkataraman
Period of Performance	Sep. 1991 - Oct.1993

ABSTRACT

Direct methanol fuel cell (DMFC) powered vehicles are foreseen as the choice of the future, because of its environmentally benign combustion products, potential for high efficiency and above all for its unlimited supply within United States. The major obstacle in the development of methanol fuel cell is the methanol electrode, particularly, a catalyst for the oxidation of methanol. No stable and active catalysts exist that can oxidize methanol for long periods of time without the loss of efficiency.

Our research has identified certain transition metal macrocyclic complexes as promoters for Pt during methanol oxidation. Half-cell studies of some of these promoters show 100-150 mV improvement over the control at 8 times less platinum loading compared to the control. These results were reported in the last DOE Joint Contractors Meeting. We now present an update of these results, together with the results of full cells employing our catalyst promoters.

INTRODUCTION

It is well understood at this point that a promoter metal capable of oxygen transfer to the platinum or that could oxidize the adsorbed residues from methanol oxidation ideally should improve the performance of the methanol oxidation electrode. The best catalysts for this reaction must have the ability to adsorb water at potentials close to the operating potentials for methanol oxidation.

Dissociative water adsorption on Pt alone occurs at about +0.8V vs. NHE at 50°C and this is why Pt alone is not a good catalyst for methanol oxidation. **So far, macrocyclic complexes of well defined stereochemistry and redox potentials have not been explored as promoters for methanol oxidation in the presence of Pt. Considerable amount of work has been done in the area of oxygen electocatalysis and lithium batteries using metallomacrocycles as electrocatalysts.**

The advantages of our approach are that a)the promoter species at a given potential is well defined unlike the case of oxides of ruthenium, tin or molybdenum where complications exist in exactly identifying the oxidation state of metal. b) The intimately bound metallo macrocycle undergoing reversible, well defined redox changes under a Pt layer would allow facile redox reaction in comparison to promoter oxides simply mixed with carbon. c) If a good model catalytic system is discovered through this effort, possibilities of achieving multimetal sites bound to carbon using highly stable ligand systems exist. Thus more efficient, tailor-made systems could be constructed.

OBJECTIVES

The objectives of the program were to:

- Synthesize macrocyclic complexes
- Prepare catalysts by depositing Pt & macrocyclic on carbon
- Conduct physical & electrochemical characterization of complexes
- Conduct electrochemical studies of promoters
- Conduct half-cell screening studies
- Conduct experiments using polymer electrolyte membrane fuel cells

ICET Inc. has identified redox promoter species that maintain a well defined oxidation state with affinity for CO like species in the potential range for methanol oxidation. Thus the activity of Pt is maintained through the scavenging action of the redox promoter. Half cell polarization studies in aqueous 1M H_2SO_4/1M CH_3OH were used to screen the performance of the promoted platinum on carbon electrodes against a control.

RESULTS

We present some of the salient features of our work from both the half cell and full cell studies.

The half cell polarization results in **Figure 1** show the methanol oxidation performance of a promoted electrode with only 5.8% Pt compared to a control that contained 9.36%.

We have also identified the fact that ligands around selected metal atoms play an important role in controlling the promoter action. **Figure 2** illustrates the behavior of various metallo macrocycles in different ligand environments. Further testing of some macrocycle promoted electrodes may be necessary due to the highly hydrophobic nature of these complexes which results both in fabrication problems as well as "breaking in time."

The nature of metals in a constant ligand environments appears to affect the promoter action as illustrated in **Figure 3**.

The full cell tests were run in standard 25cm^2 single cell hardware, using the fuel cell test stand shown in **Figure 4**.

The performance of pure platinum black versus platinum black promoted by one of ICET's promoters is shown in **Figure 5**. At 100ASF clearly there is a 100mV improvement in the presence of the promoter.

In **Figure 6**, the performance of ICET's promoter with 20% platinum on carbon is compared to LANL data on Pt-Ru alloy black and IFC platinum black data. Clearly the promoted platinum on carbon performance is very close to that realized by Pt-Ru alloy black, where the noble metal loading is 2.2mg/cm^2. In the ICET electrode the noble metal loading is 0.24mg Pt/cm^2.

CONCLUSIONS

- Polarization data from half-cell studies show the presence of a strong ligand effect. It appears that the catalytic properties of promoters can be significantly altered through the choice of proper ligands.

- While central metal atom of the chosen complex has influence on the performance of the catalysts for a given ligand, it is clear that the chosen metal atom has to be matched with a proper ligand to get maximum performance.

- The stability of the complexes appear to be good as observed from the results of half-cell and full cell studies. This has been confirmed from full cell experiments. Life time studies are under way.

- One of the promoters has shown excellent performance in methanol fuel cell studies.
  This cell's performance is almost equal to that of a Pt black control and a Pt-Ru black electrode; the black electrodes have a noble metal loading of more than an order of magnitude higher than our promoted anode.

- ICET's promoters perform well with Pt black electrodes also. An improvement of 100 mV is noticeable at 100 mA/cm^2.

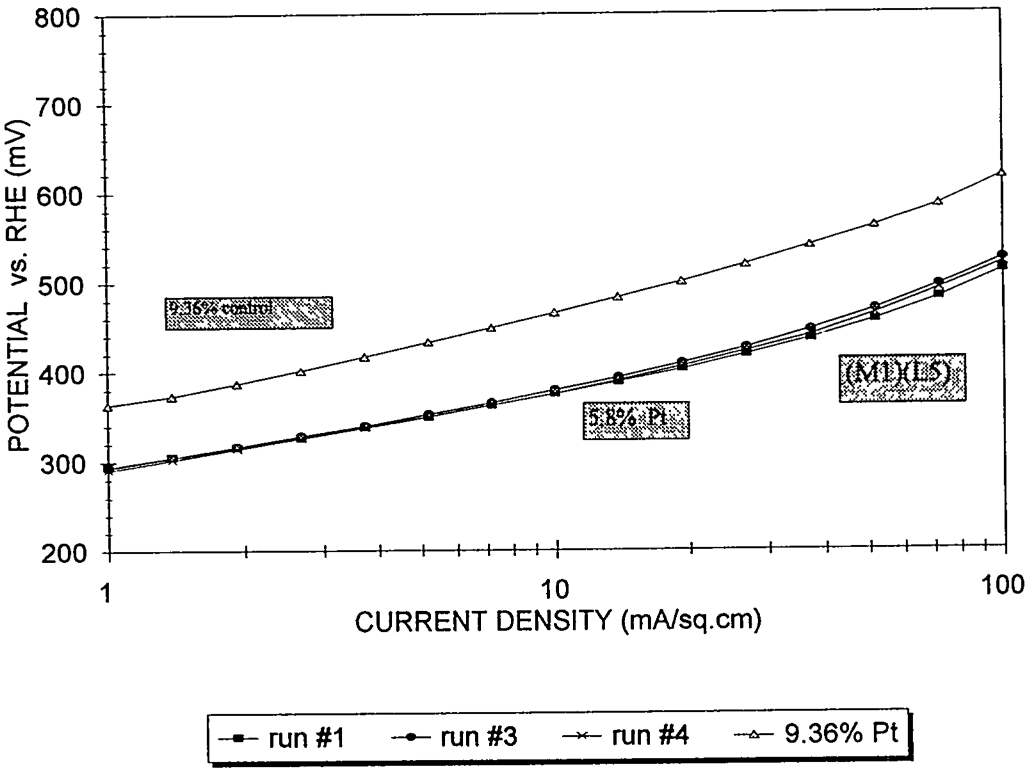


Figure 1. Half cell poarization study of a promoted electrode vs. control. 1M H_2SO_4; 1M CH_3OH. 80°C

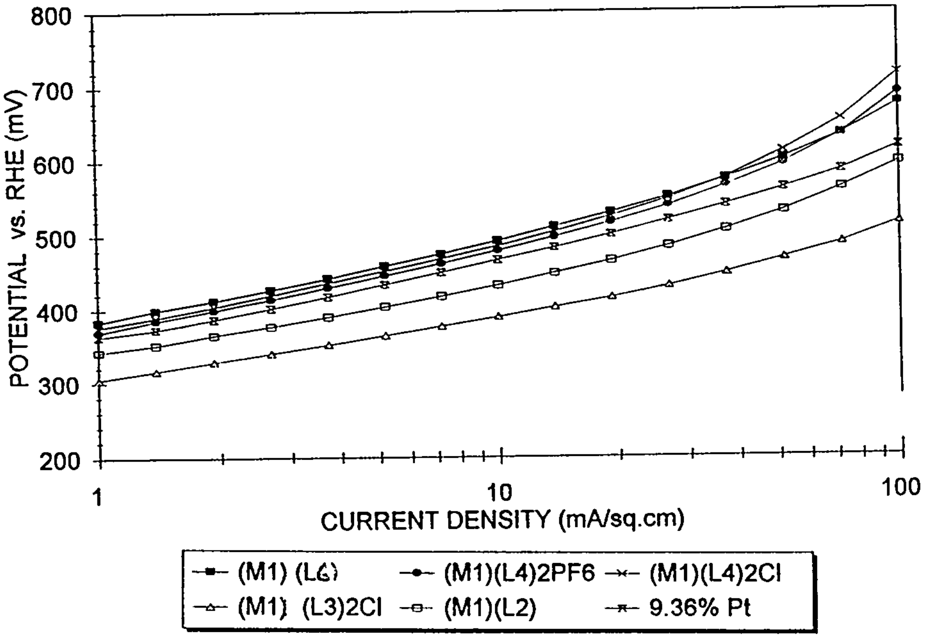


Figure 2. Half cell polarization behavior of various promoted electrodes showing the effect of ligand environment. Testing conditions as in figure 1.

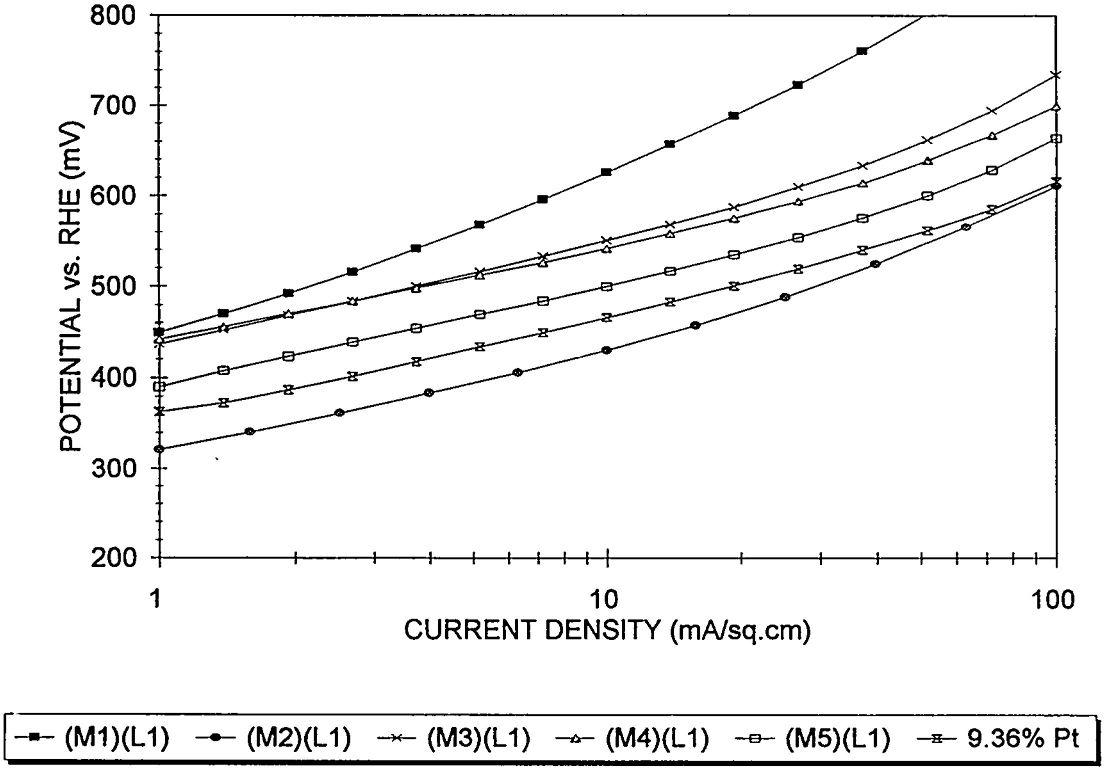
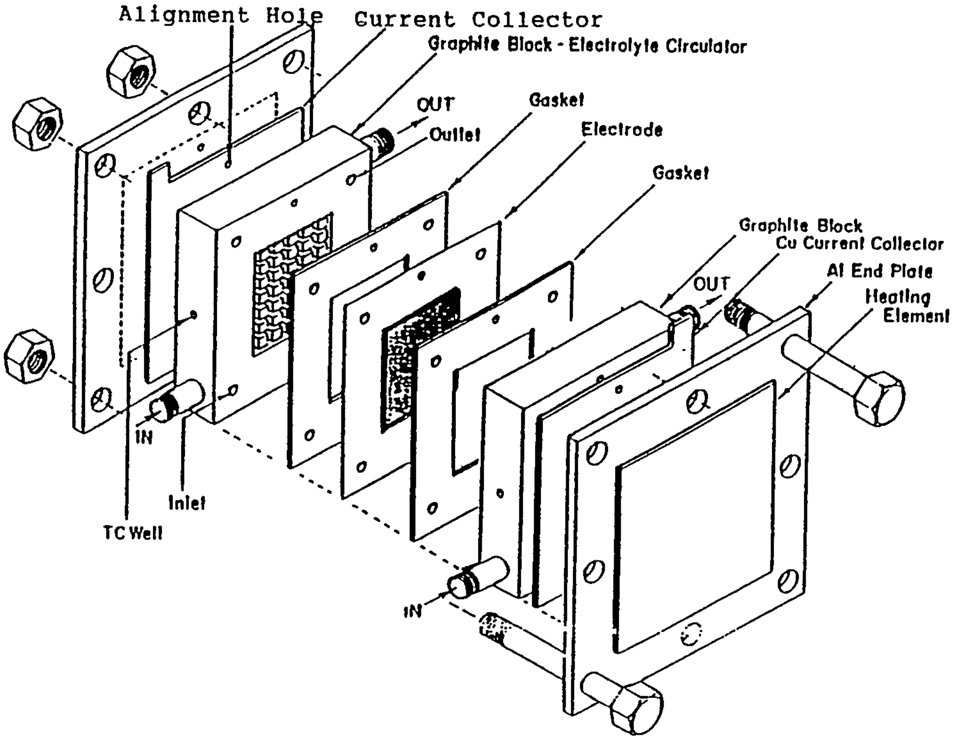
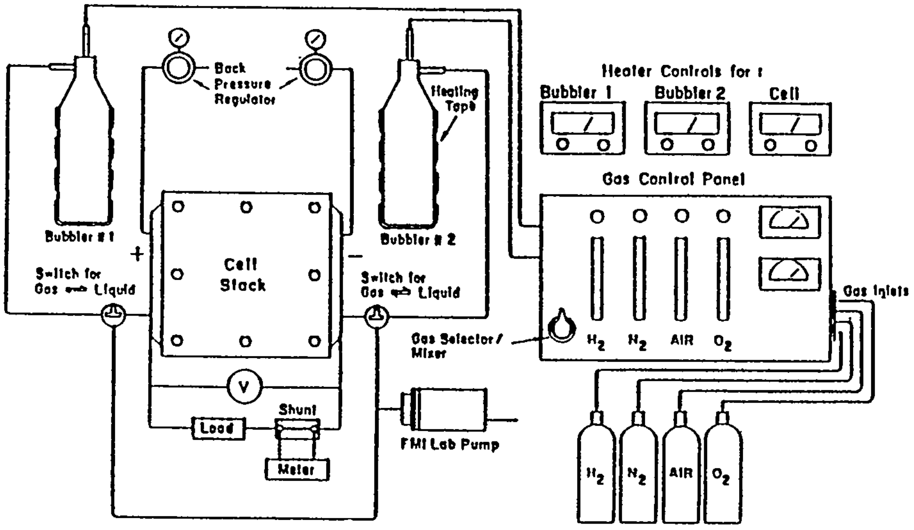


Figure 3. Influence of different metals in the same ligand environment



Single cell hardware



FUEL CELL TEST STAND

Figure 4. Single cell hardware and the fuel cell test stand used for the study.

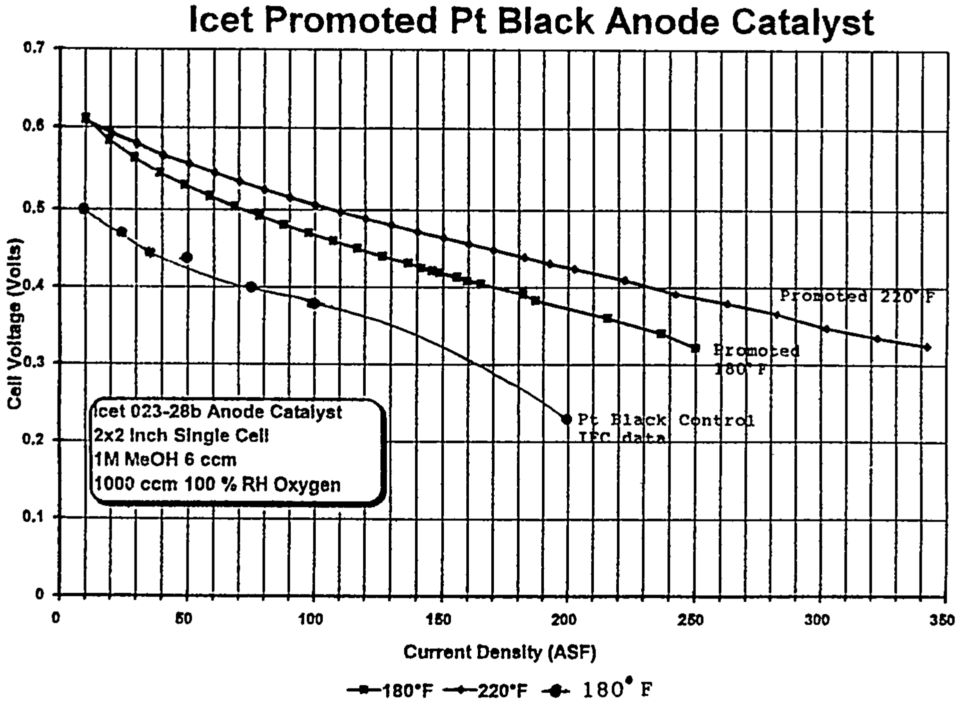


Figure 5. Performance of a promoted Pt black electrode in a full cell vs. pure Pt black

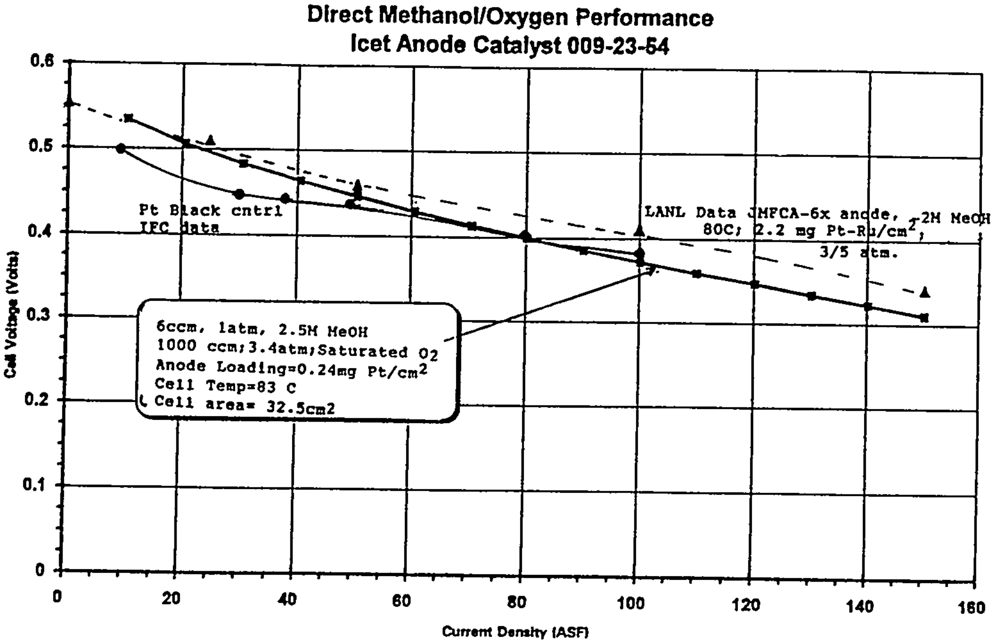


Figure 6. Comparison between ICET's promoted electrode (20% Pt on carbon), pure Pt black and Pt-Ru alloy black(LANL data)

P12

Perovskite Solid Electrolytes for SOFC

CONTRACT INFORMATION

Contract Number	DE-FG02-90ER80910
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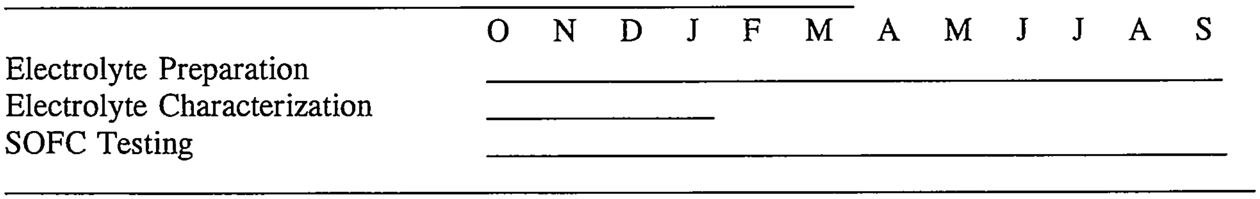
Principal Investigator	Anthony F. Sammells
METC Project Manager	Kamalendu Das
Period of Performance	July 23, 1990 to November 19, 1994
Schedule and Milestones	

FY94 Program Schedule



OBJECTIVES

The objective of this program is to experimentally determine the utility of selected perovskite related solid electrolytes for application in SOFC's operating at intermediate temperatures (600-750°C). Specific technical objectives addressed in this program include:

- Gaining useful insight into those crystallographic and thermodynamic parameters which influence both activation energy (E_a) for ionic transport and the population of ionic charge carriers.
- Synthesizing selected perovskite related solid electrolyte powders expected to possess i) low E_a for ionic conduction and ii) an intrinsically high population of ionic charge carriers.
- Preparing sintered solid electrolyte disks and determine their ionic conductivity.
- Incorporating solid electrolytes demonstrating acceptable ionic conductivity into small research size SOFC's operating at intermediate temperatures and determine their electrochemical performance.
- Determining the long-term stability of selected solid electrolyte materials incorporated into SOFC's.

BACKGROUND INFORMATION

Because of the relatively high activation energy E_a for ionic conduction in yttria stabilized zirconia (YSZ) corresponding to $\cong 0.8$eV, it is necessary to operate fuel cells incorporating this material at 1000°C where this oxygen anion conducting solid electrolyte possesses a sufficiently high ionic conductivity an ionic conductivity of 2 x 10^{-1}Scm^{-1}.

As a consequence of the above, there are strong incentives to identify alternate solid electrolytes possessing sufficiently high ionic conductivities to facilitate practical fuel cell operation at intermediate temperatures. Research towards this goal is addressing development of an approach for clarifying the dependency between E_a for ionic conduction in perovskite related lattice and parameters relating to i) metal oxide lattice dimensions, ii) the nature and sizes of ionic lattice constituents, iii) thermodynamic parameters relating to perovskite stability and iv) the intrinsic lattice polarizability or "flexibility" towards facilitating ionic mediation. Such an approach is currently evolving at Eltron Research, Inc. (Eltron) for the systematic selection of perovskite related solid electrolytes with the potential for achieving ionic conduction significantly higher than currently found with YSZ based materials.

PROJECT DESCRIPTION

Perovskite solid electrolytes being selected for preparation into sintered disks and tubes for subsequent fabrication into SOFC's were based in part upon rationale currently under evolution at Eltron which is showing trends[1-7] between activation energy (E_a) for ionic conduction and the respective parameters: 1) the average perovskite metal-oxygen energy and associated enthalpy of formation ΔH_f, 2) the lattice free volume (FV) obtained by subtracting the ionic volumes of lattice constituents in a unit cell from the overall unit cell volume, 3) the parameter $r_{critical}$ (r_c) which corresponds to the radius of an opening between two A site cations and one B site cation, through which the mobile ion must pass, and 4) the lattice polarizability α.

The expression for ionic conductivity (σ) in a solid electrolyte, as derived from random walk theory, may be given by:[8,9]

$$\sigma T = A \exp(-\Delta H_m/KT) \qquad (1)$$

where

$$A = (Z\lambda^2 e^2/6\upsilon_o K)C(1-C)\upsilon_o \exp(\Delta S_m/K) \qquad (2)$$

and $-\Delta H_m$ (equivalent to E_a) and ΔS_m are, respectively, enthalpies and entropies for activation, C is the fraction of available sites occupied by mobile ions, λ the jump distance, Z the number of jump directions, υ_o the molar volume, K the Boltzmann constant and e the electronic charge.

Examining relationship (1) above, we see that overall ionic conductivity σ is influenced both by an exponential and a preexponential term A. Performing a study on the influence of perovskite lattice parameters and ionic constituents upon E_a only addresses the exponential component of this relationship. The preexponential term A is related to the number of charge carriers present in the solid-state lattice.

As a consequence, experimental investigations also include perovskite related lattice where a high population of oxygen vacancies were intrinsic, thereby contributing to the possibility of high ionic conductivity. This general approach resulted in our investigating materials based upon the brownmillerite structure $A_2B_2O_5$, which may be considered to be derived from the perovskite structure by removing 1/6 of the unit cell oxygen atoms.

Both stoichiometric and non-stoichiometric perovskite related solid electrolyte compositions are being studied.

RESULTS

Work over the last year has focused on scale-up of perovskite-type solid electrolyte fuel cells. Two approaches were taken: planar and tubular. For planar fuel cells, tape cast thin solid electrolyte disks were prepared. For tubular fuel cells, extruded solid electrolyte tubes were fabricated.

Tape Cast Perovskites

Initial tape casting experiments performed during this period, on perovskite type solid electrolytes was performed on yttria-stabilized zirconia (YSZ). To ensure that appropriate tape-casting procedures were being performed before working with $BaCe_{0.9}Gd_{0.1}O_{3-x}$ as a model perovskite solid electrolyte. The tape-casting apparatus used for all experiments during this period was a Model 22351 tape caster from EPH Engineering Associates, Inc. The apparatus is based on a two doctor blade mechanism as schematically shown in Figure 1.

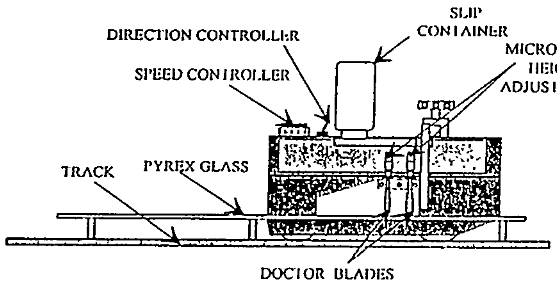


Figure 1. Tape-casting apparatus used for the preparation of thin solid electrolyte disks of $BaCe_{0.9}Gd_{0.1}O_{3-x}$.

YSZ powder, solvent and dispersants were mixed together and ball-milled for 24hr. Binder and plasticizer were then added to the mixture and ball-milled slowly for an additional 24hr. The slip was then cast with the front doctor blade set at 0.8mm and the rear doctor blade at 0.6mm. The slip was dried for 24hr and then cut into disks. Two individual disks were pressed together at 10,000psi for 10min. Resulting green disks were then sintered for 4hr at 1650°C in the atmosphere to give 85-93% dense material.

After optimizing tape-casting procedures for YSZ, related procedures were adapted for tape-casting $BaCe_{0.9}Gd_{0.1}O_{3-x}$. This proceeded from stoichiometric mixtures of $BaCO_3$, CeO_2 and Gd_2O_3. The initial slip formulation was based on that used for the related compound $BaTiO_3$, previously reported in the literature.[10,11] This slip strategy however was found unsatisfactory for preparing tapes.

Successful sintered tapes of $BaCe_{0.9}Gd_{0.1}O_{3-x}$ were prepared using a binder capable of being dissolved into organic solvents so as to avoid reaction between $BaCe_{0.9}Gd_{0.1}O_{3-x}$ and water. This was achieved using Ethocel (medium grade, Dow Chemical). The binder solution was prepared by first dissolving Ethocel in 100ml 1-propanol and further diluting with 50ml cyclohexane. This procedure yielded a semiclear gel which when mixed with additional slip constituents, gave a slip of good viscosity for subsequent tape casting.

This strategy yielded single-phase $BaCe_{0.9}Gd_{0.1}O_{3-x}$ disks (Figure 2) possessing sintered densities of 89%. Typical thicknesses were 300-400μm with disk diameters being 35-40mm.

Techniques for incorporating tape-cast thin solid electrolyte disks into intermediate temperature fuel cells were addressed. The two major issues were ensuring adequate seals were present between the sintered tape-cast disk and the housing in order to completely separate fuel and oxidant zones within the fuel cell and overcoming any mechanical weakness associated with sintered solid electrolyte disks.

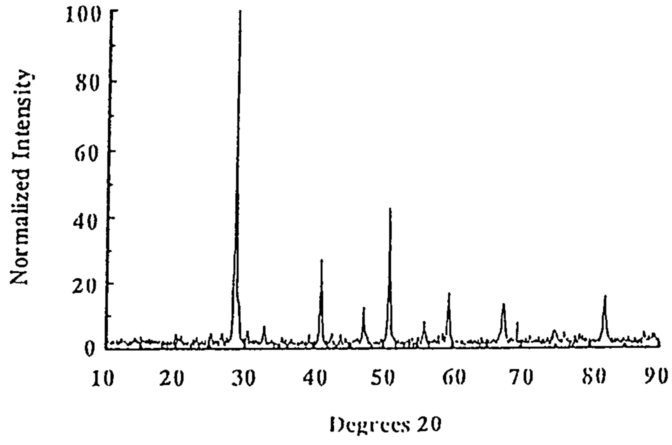


Figure 2. XRD pattern of a sintered thin disk of $BaCe_{0.9}Gd_{0.1}O_{3-x}$ prepared by tape-casting using a non-aqueous binder (Ethocel) solution.

Extruded Solid Electrolyte Tubes

As a parallel effort to tape-casting solid electrolyte thin disks for scaling-up intermediate temperature fuel cells, we have begun to prepare solid electrolyte tubes using extrusion techniques.[12] This approach required that the ceramic powder be prepared with organic additives to give the resulting precursor mixture plasticity. Following extrusion the resulting green solid electrolyte tubes were then sintered to yield the final product. Solid electrolyte extrusion was performed using a Bonnot Model M-1 B-B Gun Extruder at Eltron shown schematically in Figure 3. The extrusion die had an inner and core diameter yielding green solid electrolyte tubes of dimensions 13mm outer diameter and 10mm inner diameter.

The mixture used for extruding $BaCe_{0.1}Gd_{0.1}O_{3-x}$ tubes possessed the following respective components:

Constituent	Amount
$BaCe_{0.9}Gd_{0.1}O_{3-x}$	115g
Solvent (1-propanol)	80ml
Binder (Ethocel)	55ml
Binder (B7-MEK)	55ml
Dispersant (Maphos 56)	5.5ml
Plasticizer (polyethylene glycol 400)	8.2ml

$BaCe_{0.9}Gd_{0.1}O_{3-x}$ powder (prepared by the oxide method), solvent and dispersant were mixed together and ball-milled for 12hr. Plasticizer and binders were then added and ball-milled an additional 12hr. The mixture was poured into a thin layer and allowed to dry. After drying, the mixture had a putty-like texture. This mixture was kneaded for several minutes and subsequently extruded yielding geometrically stable green tubes. These tubes were air-dried for one day and then sintered at 1650°C for 4hrs. This yielded sintered tubes with densities 90-95% of theoretical.

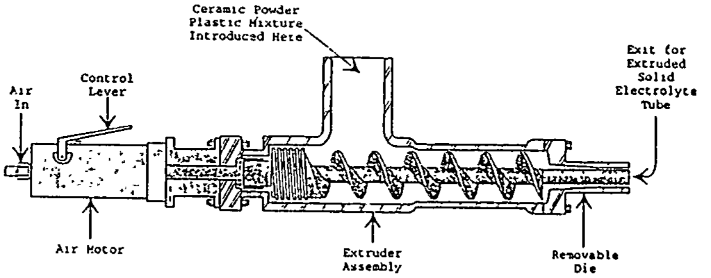


Figure 3. Schematic illustration of extruder used at Eltron for preparing sintered tubes of solid electrolytes.

Although these tubes were judged adequate for subsequent incorporation into intermediate temperature fuel cells, we required higher green strengths to prepare longer tubes. To obtain higher green strengths, the extrusion mixture was modified to include a binder/plasticizer/lubricant which would flow under pressure but harden at normal pressures. We used a commercial material (Ceracer 640, Shamrock Technologies) corresponding to a modified polyethylene wax. The first step in the process was to mix this precalcined powder with a solvent, 1-propanol. The powder to solvent ratio was 2:1 by weight. The dispersant, an aliphatic phosphate ester (Maphos 56, PPG Industries, Inc.), was added to this mixture, 2% by weight relative to the ceramic powder. This mixture was mixed thoroughly in a polyethylene container on a ball-mill for 24 hours. After mixing, two complementary binders were added. The first was a methyl methacrylate resin (Acryloid® Resin, B-7 MEK, Rohm and Haas). This particular binder is a 30% solid resin dissolved in methyl ethyl ketone. This binder was added so as to obtain 3% by weight of the solid resin to ceramic powder. The second binder was the modified polyethylene wax as discussed above. This binder was added in a 3-4% weight ratio relative to the ceramic powder. The slip was then dried by heating at 100°C for 3 hr.

The mixture was as follows:

Constituent	Amount
$BaCe_{0.9}Gd_{0.1}O_{3-x}$	98.0g
Solvent (1-propanol)	60ml
Dispersant (Maphos 56)	4.6ml
Binder (B7-MEK)	14.5g
Binder/Lubricant (Ceracer 640)	3.77g

Although no attempts were made to control extruder temperature during the extrusion process, we observed that superior green tubes were formed when the extruder assembly was ~120°C. If the temperature was too low, the mixture did not flow. If the temperature was much higher, the mix became too soft and green tubes became deformed.

After drying, solid electrolyte tubes were sintered. The first step in the sintering process was binder burn-out. This was achieved by heating the tube at 1°C/min from room temperature to 600°C. The tube was then heated at 3°C/min until the desired sintering temperature was reached. $BaCe_{0.9}Gd_{0.1}O_{3-x}$, sintering was performed at 1650°C for 4hr. Sintering was performed in a box furnace (Deltech Inc.) in the atmosphere. After sintering, tubes were cooled to room temperature at a rate of 3°C/min. X-ray diffraction indicated that the solid electrolyte tube was single-phased. Sintered densities were typically 93-95% of theoretical. Additionally, the green strength of tubes was much improved, allowing us to sinter longer tubes, ~10cm.

In addition to preparing tubes of $BaCe_{0.9}Gd_{0.1}O_{3-x}$, we studied two other materials, $Ba_2GdIn_{0.8}Ga_{0.2}O_5$ and $BaCe_{0.9}Sm_{0.1}O_{3-x}$. The former solid electrolyte was first prepared and studied by us earlier in this program.[2] The latter solid electrolyte was chosen, as it was recently shown to have a higher ionic conductivity than $BaCe_{0.9}Gd_{0.1}O_{3-x}$.[13]

Powder of $Ba_2GdIn_{0.8}Ga_{0.2}O_5$ was again prepared using the oxide method with the extrusion mixture being prepared under identical conditions to previously discussed for $BaCe_{0.9}Gd_{0.1}O_{3-x}$. These tubes did not appear to have as high a green strength as the $BaCe_{0.9}Gd_{0.1}O_{3-x}$ and as a consequence were allowed to dry in air for two weeks before sintering. Sintering was performed by packing green tubes in $Ba_2GdIn_{0.8}Ga_{0.2}O_5$ powder within an alumina tube and sintering at 1450°C for 4 hr. Sintered tubes prepared using this procedure possessed cracks and were not mechanically robust.

Fuel cells from $BaCe_{0.9}Gd_{0.1}O_{3-x}$ solid electrolyte tubes were fabricated. Machinable alumina components, coated with a ceramic glaze were used as holders for sintered solid electrolyte tubes. Typical tube lengths were 3-4cm. A schematic illustration of the experimental arrangement for a tubular type fuel cell is shown in Figure 4. A small amount of flint glass powder, was placed in the bottom of the holder. A pyrex ring was placed on top of this powder and the solid electrolyte tube placed on the ring. A spring was used to apply a constant low pressure (2-3lbs) to ensure that the molten flint glass flowed around the pyrex ring and holder. This assembly was brought to a sealing temperature of 850°C. Subsequent leak testing was performed by passing CH_4 inside the tube and N_2 outside the tube. The degree of leakage was measured by determining the CH_4 concentration in the N_2 stream. For these fuel cells, a CH_4 concentration of 10-20ppm was observed in the N_2 stream.

Previous to cell assembly, electrodes were prepared on the tube surfaces. For the cathode, silver was applied to the outer surface of the tube using silver epoxy (Epotek P-10) and current collection was obtained by attaching a stainless steel wire to the electrode, also using silver epoxy. Nickel served as the anode and was applied onto the inner surface using a nickel epoxy (Transene Nickel Bond Type 60) and current collection was again obtained using a stainless steel wire attached with the same epoxy. Fuel

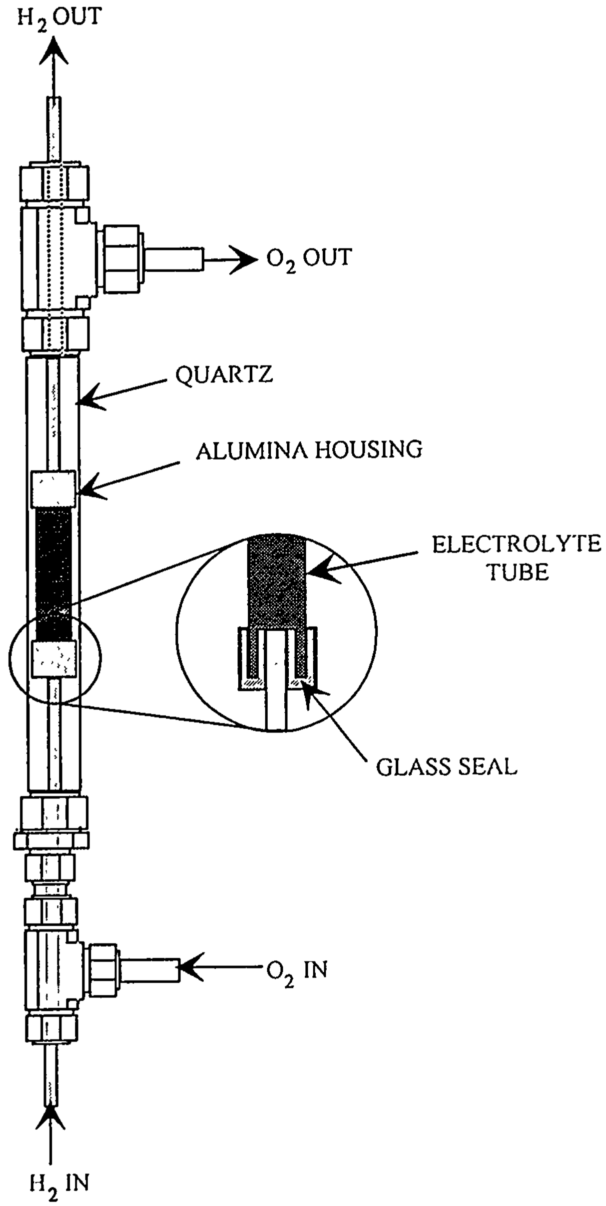


Figure 4. Experimental arrangement for incorporating solid electrolyte tubes into a fuel cell.

cells prepared using this general procedure demonstrated stable performance for over 500hrs, at which time they were voluntarily terminated. There was no evidence of physical deterioration of the solid electrolyte. This result indicates that preparing tubular fuel cells of this material is possible and that long-term operation is feasible.

Techniques for Preparation of Fine Metal Oxide Powders

Both tape-casting and extrusion techniques yielded ceramic $BaCe_{0.9}Gd_{0.1}O_{3-x}$ components compatible for incorporation into intermediate temperature fuel cells. However there were incentives to achieve higher sintered densities from these ceramic materials. One strategy for this was to start with $BaCe_{0.9}Gd_{0.1}O_{3-x}$ powder possessing a finer particle size. To this end, we have begun to develop and adopt techniques for preparing fine solid electrolyte precursor powders, with emphasis being placed upon spray pyrolysis and combustion techniques.

In spray pyrolysis, a precursor solution of desired metal nitrates or other anions is sprayed into a hot furnace. The solution evaporates and the anion decomposes yielding fine metal oxide particles. A variation on this is a method known as drip pyrolysis in which the precursor solution was dripped into the hot furnace where reaction products were collected within a tubular alumina substrate.

Initial attempts at preparing fine metal oxide particles were performed using the drip pyrolysis technique. Here a mixture of 1.66g Gd_2O_3, 35.8g $Ce(NO_3)_3$ and $BaCO_3$ were dissolved in 485ml water and 15ml concentrated HNO_3. This solution was then dripped slowly into an alumina tube held at 1000°C. This procedure caused alumina tubes to eventually crack and therefore this technique was abandoned in favor of the spray pyrolysis technique.

Spray pyrolysis was performed using a precursor solution identical to that used for drip pyrolysis. The apparatus is shown schematically in Figure 5. This consisted of a spray nozzle (Aztec Model 2000s) and a mullite tube equipped with a baffle system to trap the finest particles. The baffle system consisted of a series of stainless steel mesh screens placed inside an alumina tube through which the air stream, and fine particles, exited the reactor. This type of baffle system proved effective since we were able to collect

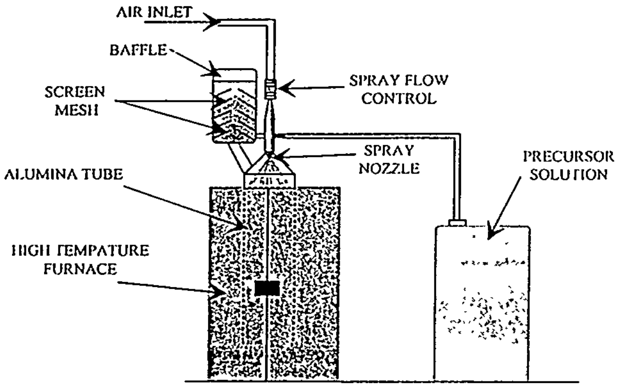


Figure 5. Illustration of the spray pyrolysis apparatus used to prepare fine particles of metal oxides used for further ceramic processing.

82% of the expected precursor powder based on the concentration and volume of solution sprayed into the tube. The mullite tube was kept at 700-1000°C in the furnace zone. It was observed that highest temperatures gave the best results.

A modification[14] to the citrate strategy for preparing fine precursor particles was also used which has been found suitable for processing barium containing materials. 0.54g ($1.5x10^{-3}$ mol) of Gd_2O_3 was ground thoroughly with 12g ($6x10^{-2}$ mol) of citric acid in a mortar and pestle and placed into a beaker. The solid mixture was heated on a hot plate to 150-160°C until the citric acid melted. 5.91g ($3x10^{-2}$ mol) $BaCO_3$ with 10ml H_2O was added to the melt. Next, 11.7g ($2.7x10^{-2}$ mol) $Ce(NO_3)_36\cdot(H_2O)$ dissolved in 10ml H_2O were added. With this latter addition, a precipitate formed, presumably $Ba(NO_3)_2$. The above procedure was repeated using 6.90g ($2.0x10^{-2}$ mol) Ce_2CO_3 mixed in 20ml of H_2O in place of the $Ce(NO_3)_3\cdot 6(H_2O)$. However, precipitation occurred again. This procedure was not pursued further.

Another recent combustion synthesis technique is the glycine-nitrate technique.[15,16] First attempts were made at preparing $BaCe_{0.9}Gd_{0.1}O_{3-x}$ followed this technique closely but we were unsuccessful as precipitation occurred before combustion. We therefore modified this technique using ethylene diamine as a chelating agent to increase the solubility of the Ba^{2+} salts in the gel.

As an example, 23.52g (0.09 mol) of $Ba(NO_3)_2$, 39.1g (0.081 mol) of $Ce(NO_3)_3\cdot 6H_2O$ and 4.06g ($9x10^{-2}$ mol) of $Gd(NO_3)_3$ were dissolved in 100ml of H_2O in a 1.5l pyrex beaker. 10ml ethylene diamine, 10ml concentrated HNO_3 and 23.7g (0.32 mol) of glycine were added. The beaker was covered with several sheets of 100 mesh stainless steel. The solution was heated on a hot plate. The solution thickened and became dark. Combustion occurred before any precipitation was noticed. Therefore, the addition of ethylene diamine served to increase solubility of the Ba^{2+} in the gel.

Figure 6 shows the X-ray diffraction pattern of the as-prepared powder. The pattern shows some extra peaks indicating that the resulting powder as inhomogeneous. The powder was placed into an alumina crucible and calcined at 1200°C for 12hr. The X-ray diffraction pattern of the calcined powder (Figure 7) shows that the material is now single-phased. Therefore, the use of a glycine-nitrate combustion technique modified by the addition of ethylene diamine appears to be adequate to obtain fine particles of Ba-containing materials.

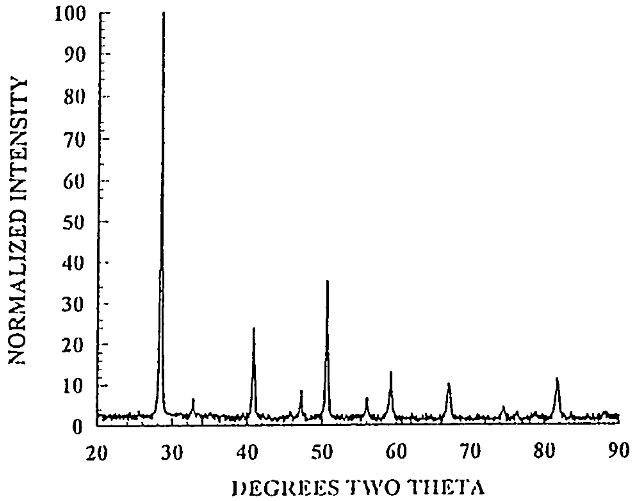


Figure 6. X-ray diffraction pattern (Cukα radiation) of $BaCe_{0.9}Gd_{0.1}O_3$ powder prepared by a modified glycine-nitrate combustion technique.

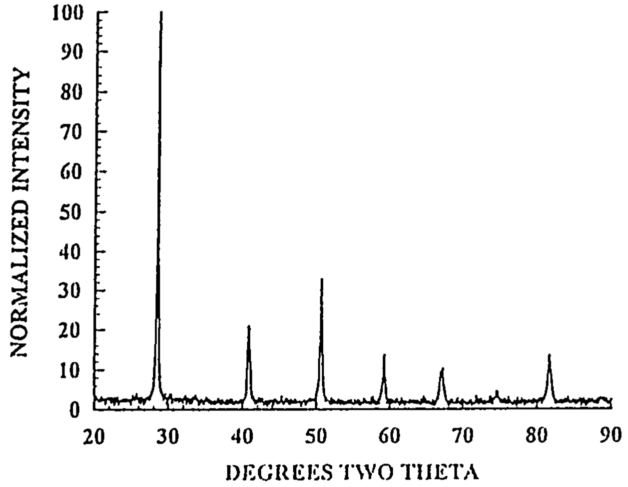


Figure 7. X-ray diffraction pattern (Cukα radiation) of $BaCe_{0.9}Gd_{0.1}O_3$ powder prepared by a modified glycine-nitrate combustion technique and calcined at 1200°C for 12 hours.

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P13

A Practical Method of Hydrogen Purification For Use in Fuel-Cell Systems

CONTRACT INFORMATION

Related SBIR Contract Numbers	DE-FG03-91ER81229, DE-FG03-91ER81228, and DE-FG03-92ER81419
Contractor	Bend Research, Inc. 64550 Research Road Bend, OR 97701-8599 (503) 382-4100
Contract Project Manager	David J. Edlund
Principal Investigator	David J. Edlund
METC Project Manager	Venkat K. Venkataraman, Jim Longanbach, and Thomas Dorchak
Period of Performance	March 31, 1992 to July 31, 1995

ABSTRACT

BACKGROUND AND OBJECTIVES

These programs are directed at the development of a practical and economical composite-metal membrane for use in high-temperature hydrogen-separation/purification processes. One potential application of this technology is for the purification of hydrogen, made by hydrocarbon reforming or partial oxidation reactions, for use in fuel cells to produce electricity. Although this application falls outside the scope of the referenced SBIR contracts, the technology has advanced under these programs to the point where initial feasibility tests have been conducted by Ballard Power Systems using a high-pressure methanol/steam reformer.

Figure 1 shows the general features of an integrated fuel-cell system in which hydrogen is produced in a hydrocarbon reformer, purified, and then fed to the fuel cells. Generally speaking, the hydrogen-purification method should meet the following requirements:

- produce high-purity hydrogen;
- be compact in size and weight;
- have low parasitic power requirements;

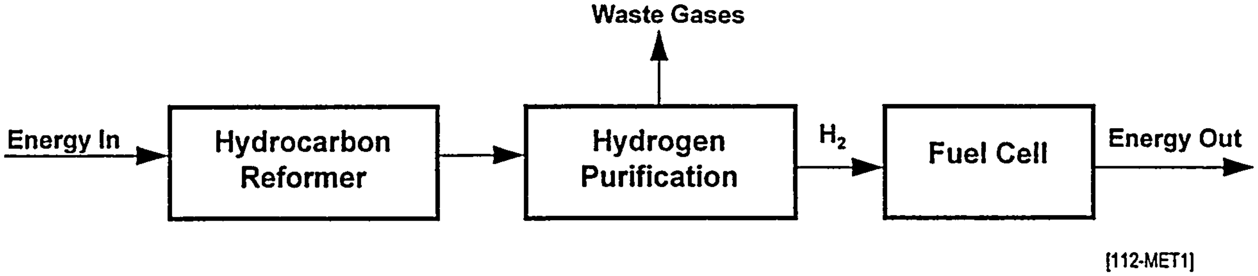

Figure 1. Simplified Representation of a Fuel-Cell System Showing the Three Key Subsystems

- exhibit rapid load-following capability; and
- be affordable.

The metal-membrane technology under development at Bend Research has the potential to meet these requirements.

PROJECT DESCRIPTION

The unique structure of the composite-metal membrane is shown in Figure 2. These metal membranes require only a minimal use of expensive palladium alloys as the coating-metal layer (the permselective layer). The membrane obtains mechanical strength from the base-metal/mechanical support layer, and the intermediate oxide layer prevents intermetallic diffusion between the coating-metal layer and the mechanical-support layer. Thus, current membranes are sufficiently durable for operation at temperatures up to about 550°C and transmembrane pressures of 600 psig. Lifetime tests at 400°C and 500°C have shown no decline in hydrogen flux after more than 6 months of continual operation.

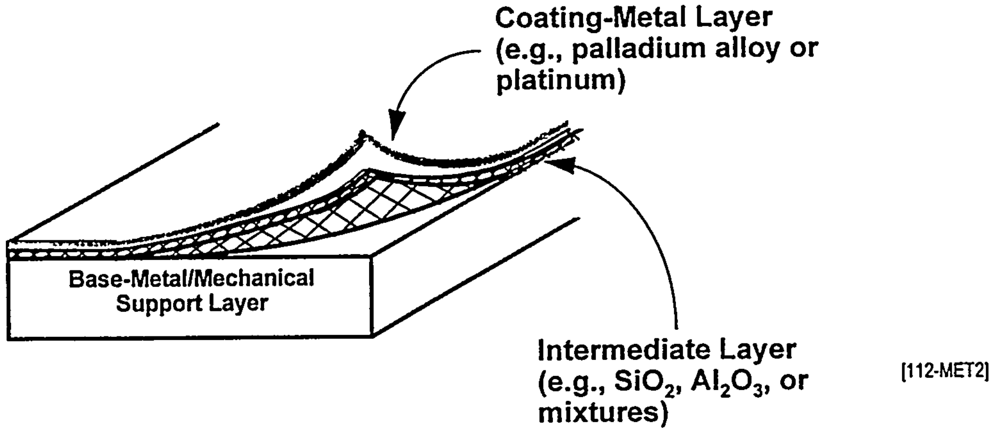

Figure 2. Key Features of the Bend Research Composite-Metal membrane

The proposed integrated fuel-cell system incorporating a metal-membrane hydrogen purifier is shown in Figure 3. The hydrocarbon/steam feed stream is converted in the reformer to hydrogen with some impurities (largely CO_2, CO, H_2O, and some CH_4). The reformate stream exits the reformer at 300°C to 600°C and 100 psia to 600 psia, depending on the selected operating conditions for the reformer. This hot, high-pressure reformate stream then flows into the membrane module, where hydrogen selectively permeates the membrane to yield a high-purity hydrogen permeate stream. The raffinate stream, now depleted of hydrogen, may be directly vented, catalytically combusted (to remove small amounts of CO and H_2) then vented, or fed back to the fuel stream used to heat the reformer.

The pure hydrogen exiting the membrane module is sent to the fuel-cell stack. Recuperative heat exchanging provides two benefits: 1) it provides the necessary cooling of the hydrogen fed to the fuel cell, and 2) it preheats the hydrocarbon/steam feed stream to the reformer.

RESULTS

Work under these three SBIR programs has focused on developing a high-flux and low-cost composite-metal membrane, and developing and demonstrating a membrane module. In addition, tests at Ballard Power Systems (funded by Ballard) were conducted to demonstrate a prototype module on a methanol/steam reformate stream. Specifically, the following primary results have been obtained:

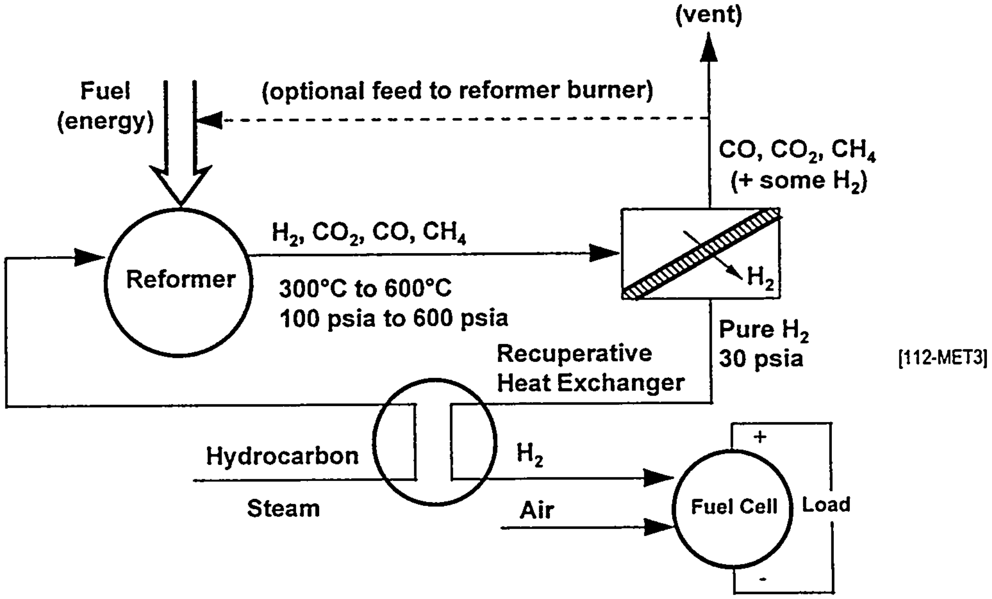

Figure 3. Proposed Integrated Fuel-Cell System Incorporating a Metal-Membrane Module to Yield High-Purity Hydrogen

- The membrane composition has been improved, resulting in a several-fold increase in hydrogen flux versus earlier-generation membranes, while the overall cost of membrane materials has decreased seven-fold. Thus, the current-generation membrane delivers a hydrogen flux of 100 SCFH/ft^2 at 300°C and 100-psig hydrogen feed pressure, and 200 SCFH/ft^2 at 500°C and 100-psig hydrogen feed pressure. Figure 4 shows the hydrogen flux as a function of the partial pressure of hydrogen in the feed stream (the permeate hydrogen is at ambient pressure) at 300°C, 400°C, and 500°C.

- Prototype plate-and-frame membrane modules containing up to 0.4 ft^2 of membrane have been designed, fabricated, and operated for up to 3 weeks. This represents a scale-up factor of about 100-fold relative to the laboratory-scale membrane samples that had been the focus of our testing. The design of these modules is shown in Figure 5. A patent application on the module design is now being filed. The projected selling price of membrane modules is expected to be about \$300/ft^2.

- A prototype plate-and-frame module containing 0.2 ft^2 of membrane was operated at 300°C on a 600-psia reformate stream from a methanol/steam reformer. Significantly, no detectable amounts of CO were found in the hydrogen permeate stream.

In addition, we have begun a collaboration with Teledyne Wah Chang (Albany, OR) to develop and market metal-membrane modules for a wide range of applications. It is anticipated that Teledyne Wah Chang will become the exclusive manufacturer of the membrane modules.

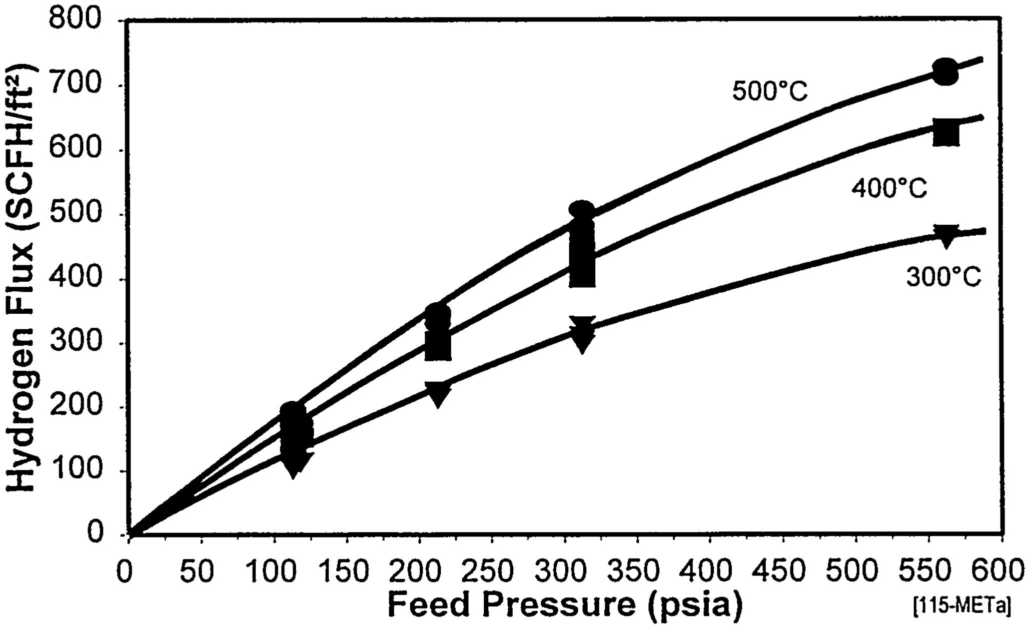

Figure 4. Flux Data for a Composite-Metal Membrane Comprising a Coating Layer of Pd-25Ag Alloy 25 μm Thick

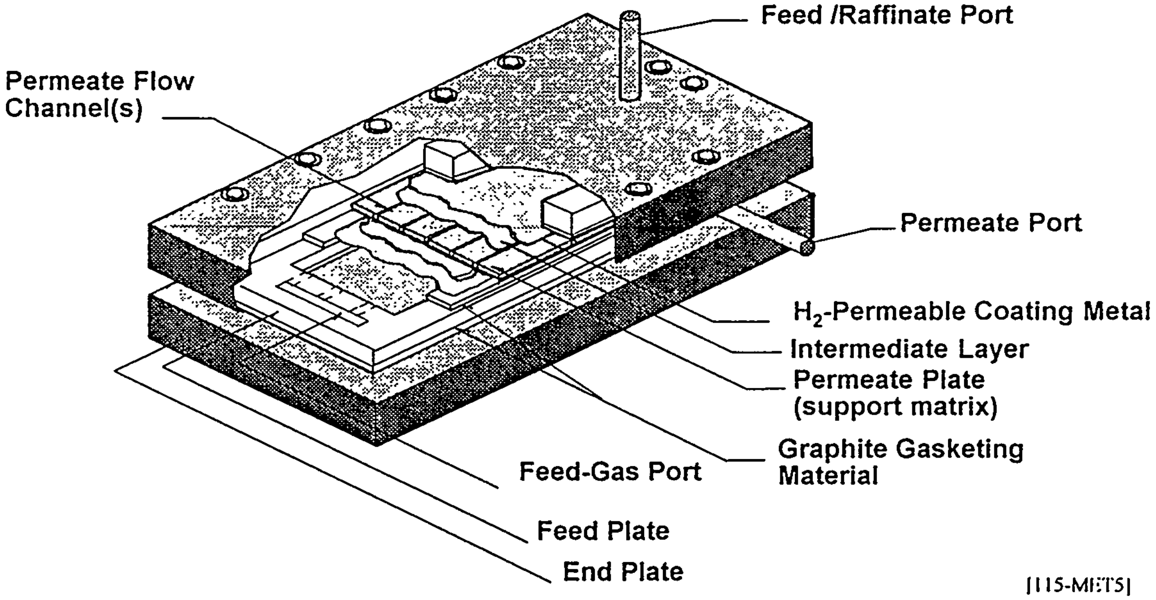

Figure 5. Cutaway View of a Prototype Plate-and-Frame Membrane Module

FUTURE WORK

We must still demonstrate that membrane modules can be scaled-up to contain between 5 ft^2 and 100 ft^2 of membrane area, and show that the membrane has sufficient lifetime when operated under actual conditions (i.e., start-up and shut-down using reformate feed streams). Although Teledyne Wah Chang is providing some assistance (both through cost-sharing and technical expertise) in working toward these goals, we are seeking strategic partners with interest in commercializing fuel-cell systems to provide additional financial and technical support to meet these objectives in the near term.

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P14 Investigation of Wetting Phenomena and Oxidation/Lithiation Behavior of Nickel in Molten Carbonate

CONTRACT INFORMATION

Contract Number	DE-AC21-90MC27394[MCP-9-IIT]
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Contractor Project Manager	T. Benjamin (M-C Power Corp.)
Principal Investigators	J. R. Selman
METC Project Manager	M. C. Williams
Period of Performance	Oct. 1, 1992 - Aug. 31, 1994

OBJECTIVES

Performance improvement and quantitative understanding of the long-term performance decay which may take place in MCFC stacks. are the key points for the success of molten carbonate fuel cells (MCFC) for the coming century. By considering these points:

1. Meniscus formation on metal electrodes in molten carbonate is investigated. The objective is to clarify the phenomena in the boundary region among electrode, gas and molten carbonate under cathode gas conditions.
2. Oxidation of nickel foil in-situ under an oxidant atmosphere is relevant for an understanding of the in-situ oxidation process to which nickel sinters are subjected in some MCFC stacks. The potential of the nickel foil is monitored while the oxidation and partial dissolution of the foil is followed optically and recorded on videotape. Additionally, the cathodic dissolution and the particle behavior are discussed to understand and quantify the conditions that may lead to high ohmic shorting of MCFC.

BACKGROUND

The surface tension at an inert metal electrode, as determined by the height of the meniscus at a partially submerged electrode. goes through a maximum as the applied potential is varied from cathodic to anodic (with respect to the standard oxidant reference electrode).

The basic requirements for MCFC cathode materials are stability. conductivity and suitable pore structure. The above performance and stability requirements are satisfied by lithium-doped NiO electrodes except for the solubility in the electrolyte. The dissolution of the lithiated-NiO cathode into the electrolyte and the subsequent precipitation of metallic nickel in the electrolyte tile are coupled processes. Because of their complexity they must be analyzed together, from both experimental and theoretical points of view.

Experience with laboratory cells and scaled-up cells in MCFC stacks indicates that formation of dispersed nickel particles precipitated in the electrolyte matrix, from cathode to anode side, can degrade cell performance and shorten cell life. NiO dissolution, nickel precipitation and shorting phenomena are connected and must be evaluated together, to arrive at possible solutions for the overall performance and stability problem. The causative mechanisms include dissolution, migration, deposition and electronic conduction processes. All these processes are coupled to electrochemical reactions and transport processes within the MCFC.

Slow dissolution of the NiO cathode and reductive deposition of the NiO as metallic nickel can cause formation of a conducting path in the electrolyte tile and lead to a short circuit. An attack on this problem is made more difficult by the fact that the various MCFC developers are not in agreement that shorting actually occurs, or even can occur, in their stacks. Most developers agree that NiO dissolution takes place, even though it can be mitigated significantly by increasing the basicity of the electrolyte in the cathode or in the electrolyte matrix. There is also reason to give credence to the suggestions made by some developers that the integrity and uniformity of the electrolyte matrix plays a significant role in the occurrence or non-occurrence of actual shorts. Table 1 is an attempt to summarize the known and unknown factors in the dissolution/shorting issue.

Two simultaneous reactions occur at the NiO cathode. The overall reaction is the result of a corrosion couple in which NiO is anodically dissolved and oxygen is cathodically reduced. NiO dissolves into carbonate melt by an acidic

Table 1. Known and unknown factors in NiO dissolution and shorting of MCFC cells or stacks.

DISSOLUTION
KNOWN FACTORS
ACIDIC / BASIC DISSOLUTION BEHAVIOR
PARTIAL PRESSURE OF CO_2
ELECTROLYTE ADDITIVES (ACIDITY BASICITY)

UNKNOWN FACTORS
EFFECT OF LITHIATION PROCESS
EFFECT OF CURRENT DENSITY
EFFECT OF CRACKS AND VOIDS IN THE MATRIX
LONG TERM EFFECT OF ADDITIVES
CURRENT COLLECTOR / CATHODE / MATRIX INTERACTION
MOTION AND DISSOLUTION OF PARTICLES IN THE MATRIX

SHORTING
KNOWN FACTORS
MATRIX THICKNESS
CO_2 PRESSURE

UNKNOWN FACTORS
CONDUCTING LINK BETWEEN CATHODE AND ANODE
CRITICAL AMOUNT OF NICKEL IN THE TILE
STRUCTURAL DEFECTS IN THE MATRIX
ADDITIVES
MOTION AND DISSOLUTION OF PARTICLES IN THE MATRIX
EFFECT OF TILE REACTIONS (${H_2O}$ formation)

flux mechanism. As long as oxygen partial pressure is low. the rate of dissolution is proportional to CO_2 pressure. There is some indication that dissolved nickel is present as Ni^{2+}. The first step in this acidic dissolution mechanism for the formation of Ni^{2+} ions is, therefore

$$NiO = Ni^{2+} + 1/2\ O_2 + 2e^- \qquad (1)$$

which is coupled with the main reduction reaction:

$$1/2\ O_2 + CO_2 + 2\ e^- \rightarrow CO_3{}^{2-} \qquad (2)$$

Overall this is a mixed-potential corrosion process, e.g., slow dissolution reaction in the anodic sense with the oxygen reduction reaction in the cathodic sense.

$$NiO + CO_2 \rightleftarrows Ni^{2+} + CO_3^{2-} \qquad (3)$$

The dissolution level of nickel oxide may be reduced by operating the cathode at low CO_2 partial pressure but too low a CO_2 partial pressure causes a negative effect on cell performance as Eq. 2 suggests. It has also been reported that the solubility of NiO in carbonate environment is considerably reduced by the addition of Mg^{2+}, Ca^{2+}, Sr^{2+} and Ba^{2+} cations [Ref. 1]. The effect of additives on the oxidation behavior of NiO in carbonate melts is being investigated in this project. The effect of additives on NiO solubility becomes less if the additives have a tendency to segregate toward the anode.

The dissolution-deposition process also causes structural changes in the cathode due to loss of material and electrolyte displacement toward the matrix. However, decrease of cathode performance is not so much due to the depletion of active cathode material, as to the accumulation of metallic nickel in the tile.

PROJECT DESCRIPTION

Wetting angle phenomena and cathode dissolution kinetics in the molten carbonate are relevant to understand performance and life-time requirements

In recent experiments at I.I.T. related to above phenomena, formation of a meniscus (Fig. 1) at an inert electrode partially immersed in molten carbonate, is observed in order to determine the reaction zone. The current at a certain immersion depth with inert WE under cathodic load (polarization) was measured for different temperature and gas conditions.

Observation of surface changes on a nickel electrode during in-situ oxidation and lithiation is also monitored and evaluated by using electrochemical methods (open circuit potential change and cyclic voltamogram) in order to clarify electrolyte behavior during wetting and oxidation. The investigation makes use of the apparatus previously used by Matsumura and Selman [Ref. 2]with minor modifications. This system has four main parts (Fig. 2a). They are: an electric furnace containing a test cell; electrochemical measurement equipment; optical observation equipment; and a gas supply unit. The test cell and electrodes (Fig. 2b) are located in the middle of the furnace.

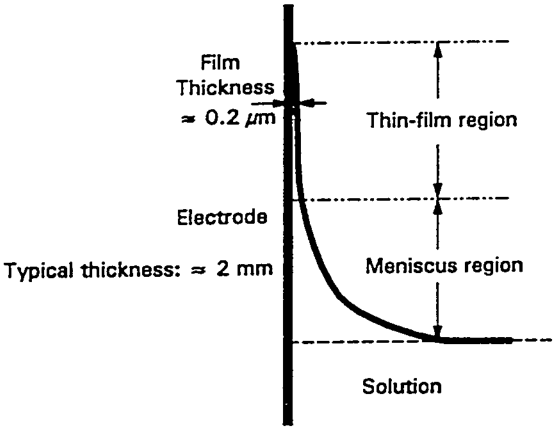


Figure 1. Schematic of Meniscus Formation in Molten Carbonate

RESULTS

Wetting has been studied at a gold foil electrode, under various oxidant atmosphere conditions, and at a nickel electrode. under standard oxidant atmosphere.

Effect of Polarization on Wetting Properties

Depth of Electrode. The current through the WE with cathodic potential applied between RE and WE was measured at different immersed depths of the WE. Under constant temperature (650^{0}C), constant gas conditions (O_2 CO_2/N_2= 15/30/55 mole %) and constant applied potential between RE and WE (WE-RE = -200 mV), the current through the WE was monitored while its depth of immersion was changed form 10 mm to 15 mm and from 15 mm to 10 mm. The current change is shown schematically in Figs. 3(a) and (b). While the immersion was changing from 10 mm to 15 mm, the current showed a temporary increase. When changing the immersion from 15 mm to 10 mm, the current showed first a temporary decrease. then relaxed to almost the same value as before immersion.

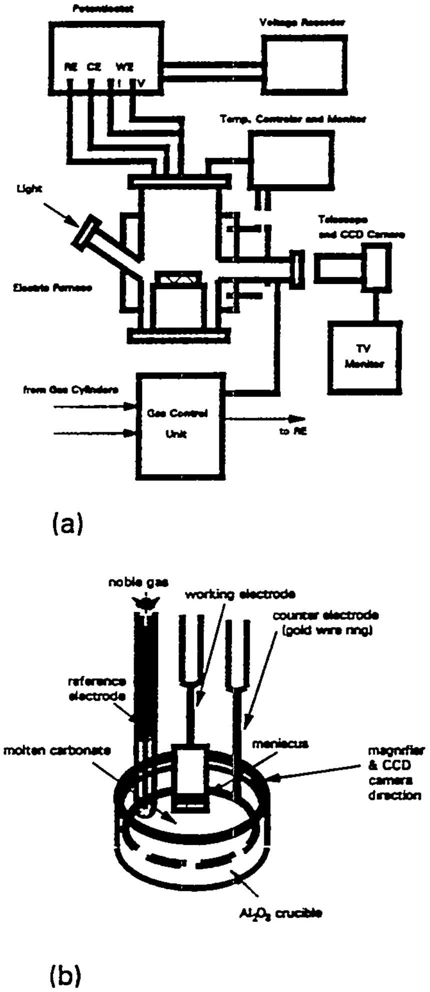


Figure 2. Apparatus Used for Wetting Angle Measurements: (a) Controlled Atmosphere Chamber with Optical and Electrochemical Equipment; (b)Detail of Cell and Electrode Configuration Showing Semi-Immersed Foil.

Fig. 4 shows the influence of the immersed depth on the current. It seems that the current depends on the immersion depth but the influence becomes less with increased immersion. At more than 10 mm immersion, the characteristics are independent of immersion. This suggests that electrochemical reaction takes place mainly near the electrolyte surface.

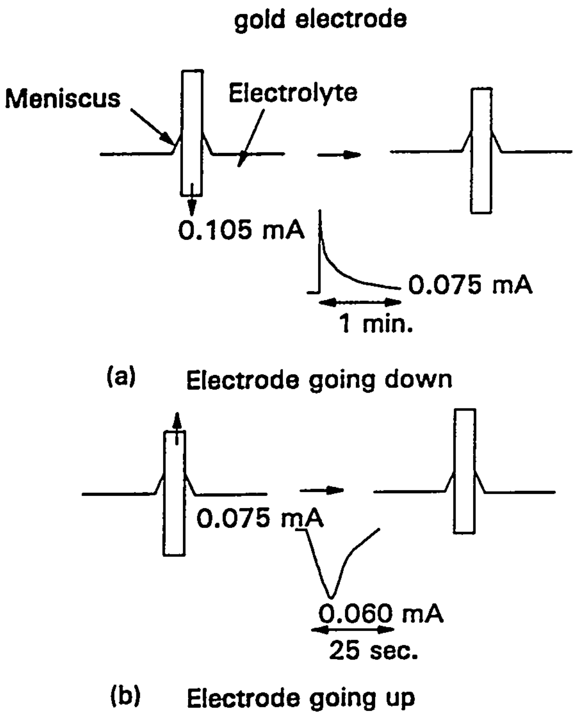


Figure 3. Schematic of Current Behavior with Immersed Depth

The transient phenomena may be explained as follows. In case (a), where immersion was changed from 10 to 15 mm, the electrolyte forming the meniscus, which may be expected to include more dissolved oxygen and carbon dioxide gas, is dragged into the melt. The dissolved species immediately react at the immersed electrode surface, which causes an increase of current, which relaxes to zero. On the other hand, in case (b), where the electrode was brought back to its original position, it seems that the electrolyte forming the meniscus is stretched upward somewhat due to the interaction forces between electrode surface and electrolyte. Therefore, the ionic resistance between the reaction site and the bulk melt increases temporarily, and hence the current is decreases temporarily.

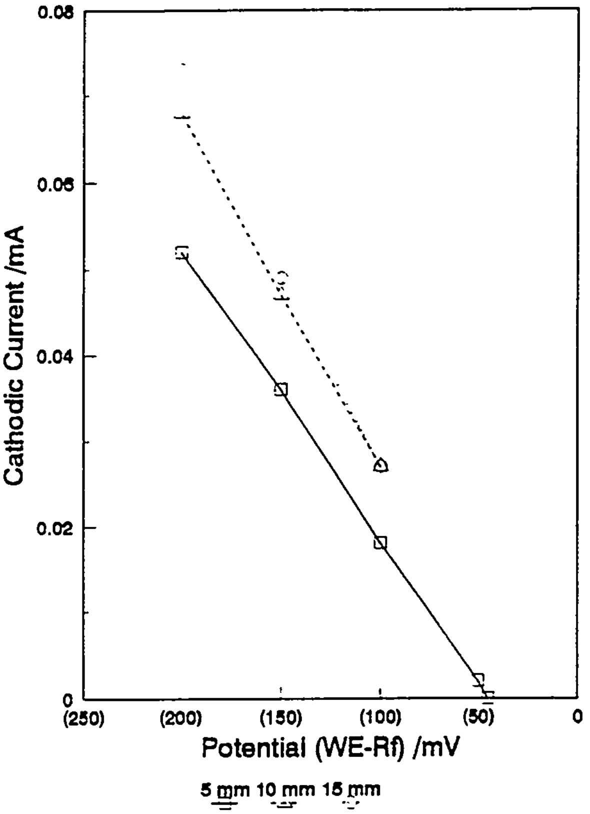


Figure 4. Effect of Electrode Immersion on Current-Potential Behavior

Temperature Effect. In continued measurements, the current through the WE at 10 mm immersion under applied cathodic potential was measured for different temperature conditions. The current depends on temperature and increases with temperature at constant applied potential. This increase in current is caused by a decrease of reaction overpotential and/or electrolyte resistance with temperature. However, over a range of temperatures, it has been found that the adsorbed ions may, with equal likelihood, be cathodically active species such as peroxide and superoxide. The behavior of working electrode at three different temperatures is shown in Fig. 5. The quasi-parabolic response at 600°C is relatively shallow, and the data near the electrocapillary minimum appear to be remarkably similar to those for low-oxygen, high CO_2 oxidant compositions, which are in equilibrium with melts that are rich in O^{2-} and to a lesser extent in O_2^{2-}.

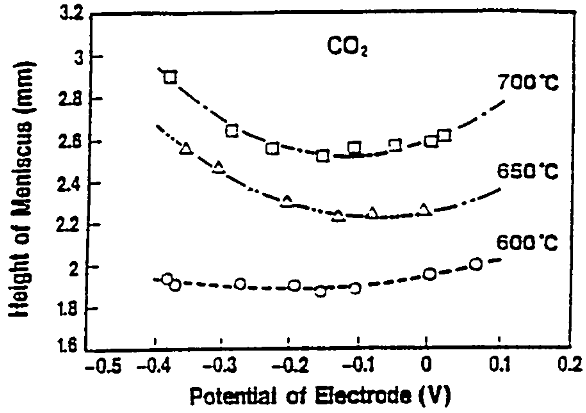


Figure 5. Meniscus Height Measurements at a Gold Foil Electrode Semi-immersed in Li-K Carbonate at Three Temperatures. The Effect of Polarization is shown.

Effect of Gas Composition. The current through the WE at 10 mm immersion under applied cathodic potential was measured for different gas conditions at 650°C. Fig. 6 shows the voltage vs. current characteristics for each gas condition. These two characteristics show similar behavior under different gas compositions. The results agree with previous work. This means that the gas solubility and its transport to the reaction site does not play a role in the electrochemical reaction occurring on the electrode surface.

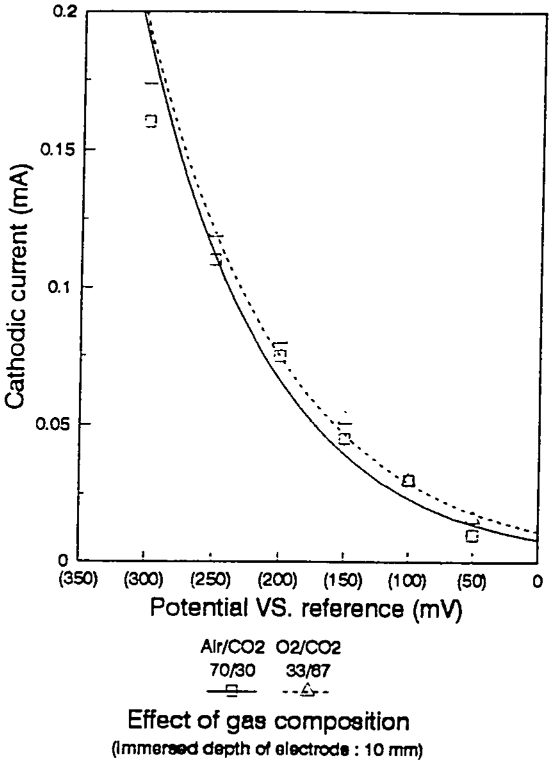


Figure 6. Effect of Gas Composition on Current-Potential Behavior

Conclusions

From these results, it is possible to infer the reaction mechanism of this electrode system as follows. Electrochemical reactions on the immersed part of the electrode are controlled by the concentration of the reactant species (O^{2-} or CO_2) near the surface. Electrochemical reactions on the non-immersed part of the electrode are controlled by the mobility of the reaction product (CO_3^{2-}).

There is no evidence for the formation of a thin electrolyte film above the meniscus at a gold electrode and for its contribution to the electrochemical reaction. If there would be such a thin film it must contribute to the electrochemical reactions, therefore current change should be in the opposite direction to the previously defined case.

It was also possible to determine the cross section of the meniscus by means of back lighting. Although the geometrical scale of the shape could not be measured because it was not clear, it was confirmed that the contact angle of electrolyte to gold electrode is almost 60 degrees. Therefore, a clear boundary line between electrode and electrolyte could be observed, however, a thin film above the meniscus could not be observed. This means that a smooth gold electrode is not well wetted by molten carbonate even under cathode gas conditions.

At an inert (gold) electrode surface tension, as determined from the height of the meniscus at a partially submerged gold electrode, goes through a maximum when the applied potential is varied from cathodic to anodic with respect to the standard oxidant reference electrode. In the absence of oxygen, the minimum is very shallow and displaced in the positive direction by as much as 100 mV. The parabolic response to potential can be analyzed in part on the basis of principles which are well-known from aqueous solution thermodynamics ("electrocapillary effect" and Lippmann-Helmholtz equation). The analysis leads to the conclusion that some oxygen ions present in the melt are specifically (i.e., chemically) adsorbed on the metal surface. In our earlier publication on these measurements, the adsorbed ions were tentatively identified as oxide ions. [Ref. 2] This would be consistent with the slow kinetics of oxide neutralization:

$$O^{2-} + CO_2 \rightarrow CO_3^{2-} \qquad (4)$$

The optical record of the oxidation of the nickel foil shows three distinct phases, with several remarkable and unexpected aspects. First, intense gas evolution is found to occur during phase II (main plateau). Second, throughout the initial phases, fine particles are seen to spall from the foil and to be convected away from it, down the meniscus. Third, this particle motion takes place at relatively high apparent speed. Apparently, the electrolyte undergoes intense convection during the oxidation process.

In summary, the stages of nickel oxidation/lithiation can be analyzed, with reference to (Fig. 7), as taking place in three stages having different characteristics.

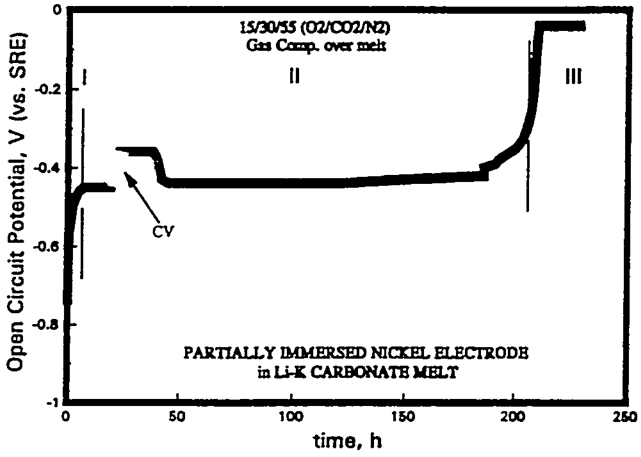


Figure 7. Potential-Time Behavior of Nickel Partially Immersed in Li-K Carbonate under Standard Oxidant Gas, at Open Circuit

Stage I. Initial NiO Surface Formation

Reaction in this region is very rapid (a few hrs.), then drops off to low rates.

Stage II. Oxidation-Lithiation of Interior Layer

The oxidation process continues, and lithium incorporation into the lattice starts to take place. The oxide layer formed in the previous stage causes limitations on diffusion of oxide species and lithium ions.

Stage III. Equilibrium

Stage I provides an oxide film at the surface. This film initially limits diffusion of reactants. In stage II, there is a gradient of oxide and lithium ions between the electrode surface and interior part of the electrode which is not initially oxidized or lithiated. Lithium incorporation into lattice seems slower than oxide diffusion. The presence of lithium in the lattice increases the diffusion coefficient of nickel ion in the NiO lattice because of the increased concentration of vacancies (Figs. 8 and 9).

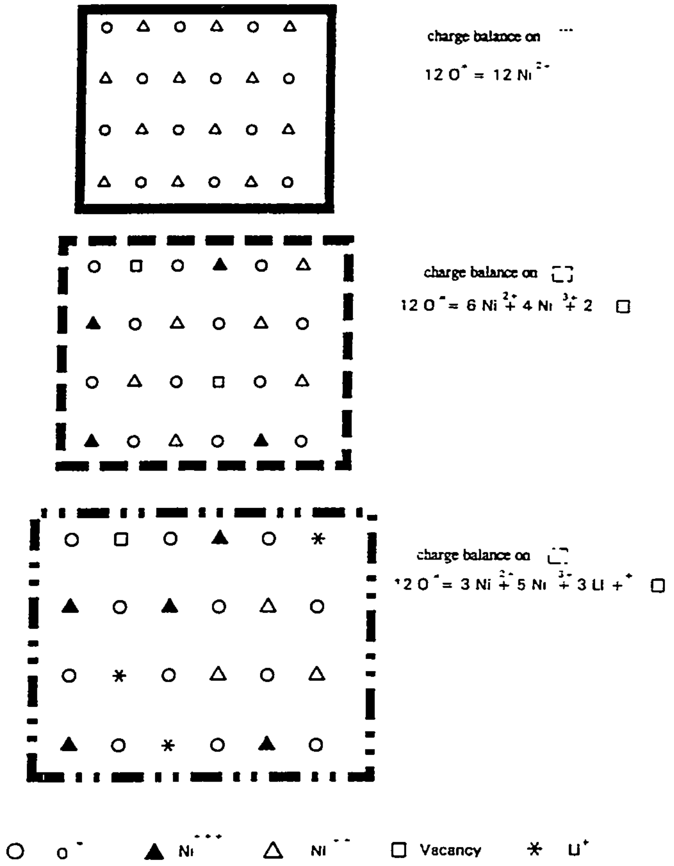


Figure 8. Schematic of Solid-State Oxidation-Lithiation Process: Cases: There is no Defect Formation, Defect Formation and Finally Doping with Lithium

Visual Observations

During the nickel oxidation experiment it is visually observed that most of the corrosion products are dissolved into the melt during the early stages of the oxidation/lithiation process.(Possibly, NiO particles lose contact with the inner nickel substrate and pieces drop into the electrolyte). These solid corrosion products are mostly collected at the surface of the ceramic tubes immersed into melt.

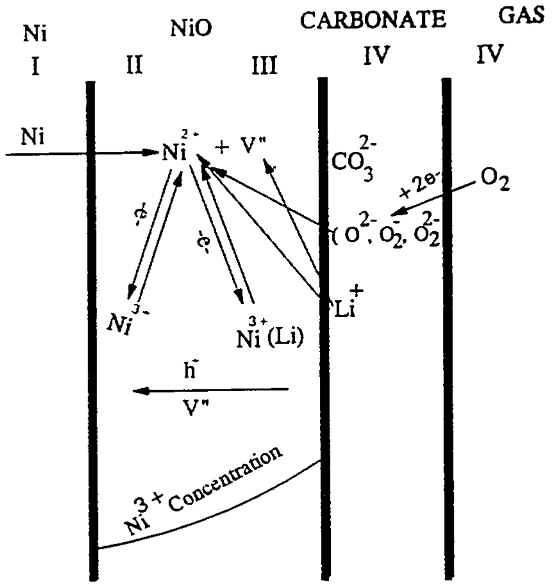


Figure 9. Schematic of Oxidation-Lithiation Process for Different Stages

It is observed that the oxidized electrode has different layers. NiO formed on nickel consists of a black outer layer and a green inner layer which represents different oxidation levels. After oxidation, the electrode could easily be separated into two pieces, starting from the middle where there were a lot of white spots. Another interesting observation was the presence of a few black particles. These could be NiO deposits but there is also a possibility that they are carbon formed during CV application, at high negative potentials. The particles form a homogeneously distributed thin film at the surface of the crucible. Even when the nickel electrode was exactly in the middle of the crucible, a black colored film was formed at part of the crucible surface in contact with or covered by carbonate.

CV of Nickel Electrode at Oxidation/Lithiation

Cyclic voltammograms recorded while the sample was undergoing oxidation/lithiation and they confirm the potential at which lithiation takes place. The likelihood of certain electrochemical reactions can also be calculated from thermodynamic information (free energies of reactants and products). Cyclic Voltammetry was applied before the oxidation process and during lithiation. These measurement were used to distinguish between different electrode reactions. When the lithiation process is completed, the OCP of the NiO electrode reaches the oxygen equilibrium potential. In Fig. 10 CV for a partially immersed Ni electrode under a pure CO_2 oxidant gas (with a trace amount of oxygen), and in Fig. 11 under 1/2% (O_2/CO_2) gas environment is shown.

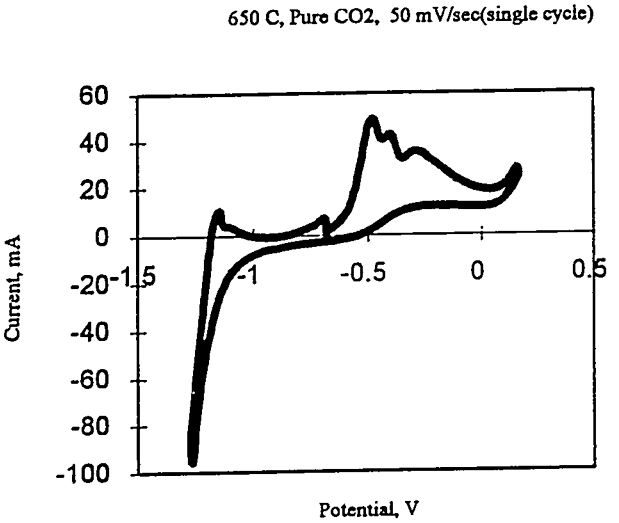


Figure 10. Cyclic Voltammogram of Nickel Electrode in Molten Carbonate

In Fig. 10, although no oxygen was present in the system, some oxygen leakage may have caused the rest potential to be around -650 mV during time in which cyclic voltammetry was applied in the anodic direction. At positive potentials the sharp increase in current is attributed to the anodic oxidation of carbonate. A peak observed around -0.4 V is assigned to lithiation of NiO electrode. The formation of trivalent nickel increases the conductivity of NiO electrode. This also increases the current corresponding to this reaction. In the reverse sweep, the current suddenly drops to potentials more negative than -0.35 V because of the reduced conductivity. However, additional work is necessary to confirm this interpretation of potential-current behavior.

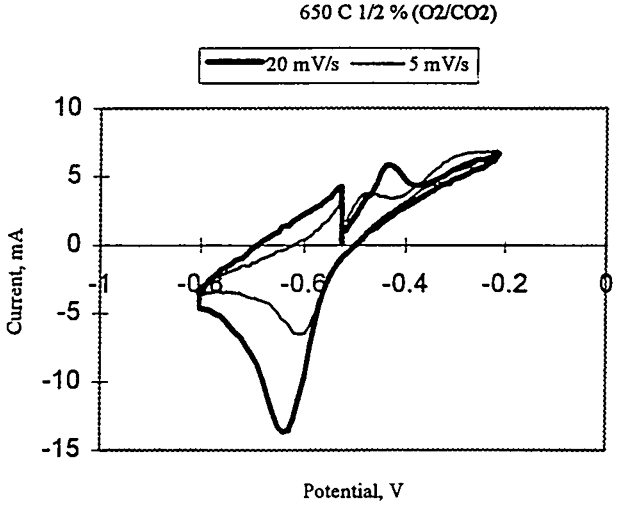


Figure 11. Cyclic Voltammogram of Nickel Electrode in Molten Carbonate

FUTURE WORK

The primary experimental concern in this fuel cell system is that of designing and operating the cell in such a manner that the rate of cathode degradation is minimized and cell endurance is assured. Experiments in which nickel is exposed to low-level oxidant atmospheres and reducing atmospheres are now under way.

Understanding the phenomena involved in these oxidation/lithiation and dissolution processes requires modeling and a series of experiments with different controlling parameters and materials are necessary. AC impedance will also be used to clarify the effect of additives. These experiments are now in progress (using nickel, iron, SS 310 and 316 as electrode material, and conventional carbonate mixtures with additives as electrolyte).

REFERENCES

[1]. J.B.J. Veldhuis, S.B. van der Molen, R.C. Makkus and G.H.J. Broers, Ber. Bunsenges. Phys.Chem. 94 (1990) 947

[2]. M. Matsumura and J.R. Selman, J.Electrochem. Soc., 139 (1992) 1255-1261

Session 3

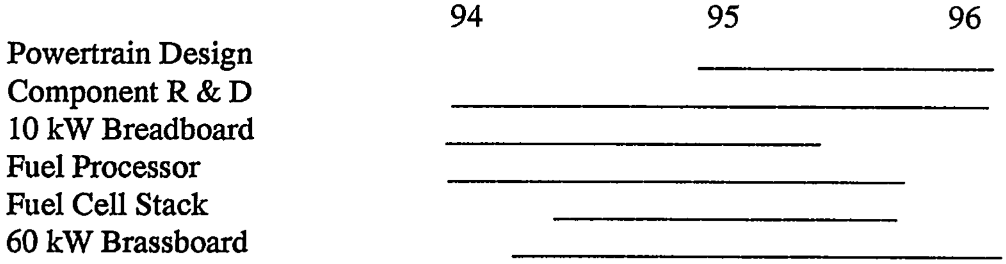
DOE's Fuel Cell Transportation Program

3.2 Proton Exchange Membrane Fuel Cell for Transportation

CONTRACT INFORMATION

Contract Number	DE-AC02-90CH10435
Contractor	General Motors Corporation 3044, West Grand Blvd., Detroit, Michigan 48202
Contractor Project Manager	Swathy Swathirajan
Principal Investigator	Matthew H. Fronk
DOE/OTT Project Manager	Robert A. Kost
Period of Performance	January 1991 to June 1994

PROGRAM SCHEDULE (Phase II)



OBJECTIVES

The objective of the first phase of the program was to demonstrate the feasibility of a 10 kW fuel cell powerplant fueled by methanol. This powerplant is based on a methanol fuel processor and a proton exchange membrane (PEM) fuel cell stack. In the second phase, the powerplant will be scaled up to 30 kW and a 60 kW brassboard system with a 30 kW battery will be built. In the final phase, the fuel cell powerplant will be scaled to 45 - 60 kW and a proof-of-concept fuel cell vehicle will be built and tested.

BACKGROUND INFORMATION

Proton Exchange Membrane (PEM) fuel cells are extremely promising as future power plants in the transportation sector to achieve an increase in energy efficiency and eliminate environmental pollution due to vehicles. PEM fuel cells are favored for light duty vehicle applications because of the low operating temperature (<100 C) and a higher power density compared to the phosphoric acid fuel cell. PEM fuel cells use a solid polymer electrolyte with carbon-supported platinum electrodes. The efficiency of the powerplant is about 60 % with hydrogen, and about 40 % when a methanol fuel processor is used on-board. Trace amount (3 -10 ppm) of CO is likely to be a regulated emission from a methanol fuel processor based fuel cell stack.

GM is currently involved in a multiphase program with the US Department of Energy for developing and testing a proof-of-concept hybrid vehicle based on a PEM fuel cell power plant and a methanol fuel processor. Other participants in the program are Allison Engine Co., Los Alamos National Labs, Dow Chemical Co., Ballard Power Systems and DuPont Co. In the just completed phase I of the program, a 10 kW PEM fuel cell power plant was successfully built and tested to demonstrate the feasibility of integrating a methanol fuel processor with a PEM fuel cell stack.

PROJECT DESCRIPTION

Phase I effort included several tasks such as system conceptual design study, component R & D, fuel cell stack and methanol fuel processor development, powersource ancillaries, sensors and controls development, and finally, integration and evaluation of a 10 kW powerplant. The system conceptual design study included a modeling study of the projected performance of the fuel cell powerplant and the use of this data to predict vehicle performance using a vehicle simulation code. Component R&D involved studies of the membrane-electrode assembly, improvements in membrane performance, designing new electrode structures and processes to reduce the platinum loading in the fuel cell by an order of magnitude, and the design of catalytic reactors for performing various heterogeneous chemical reactions involved in the fuel processor.

Fuel processor activities focused on the design, fabrication, assembly and testing of a fuel processor that consisted of a steam reformer, shifter and a preferential oxidizer (PROX). The fuel cell stack was fabricated at Ballard and the performance and reliability of the stacks were tested using both synthetic reformate as well as reformate from the fuel processor. An advanced stack was designed to handle catalyst poisoning problems at the anode due to the presence of CO and CO_2 in the fuel mixture. Other tasks involved the design and testing of ancillary components and the integration of the 10 kW powerplant.

RESULTS

Figure 1 is a schematic of the powerplant under development at the GM/LANL joint development center. The fuel processor output is a mixture of hydrogen, CO_2 and trace amounts of CO. This processed fuel is fed to the fuel cell stack that generates d.c. power, heat and water. Heat and water are partially recovered for system operation. The tests used two 5 kW Ballard stack modules. The stack power density is about 0.2 kW/kg or l. A high performance fuel processor is under development at LANL and automotive packaging now appears feasible. The goal is to increase the power density of the entire power plant to 2 - 3 kg/kW and 3 - 4 l/kW.

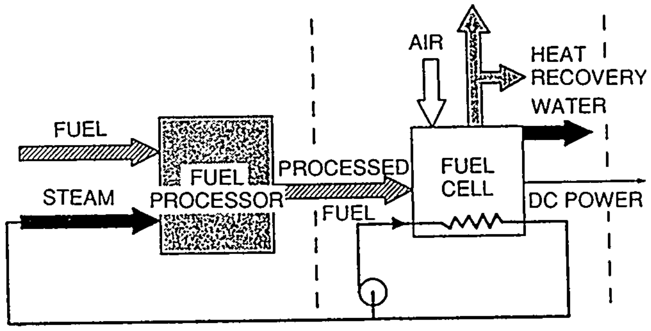


Figure 1. PEM fuel cell powerplant

Fuel Processor. The steam reformer is a packed-bed reactor operating at 275 °C and converts nearly 99.5 % of the methanol using CuO/ZnO catalyst . The residual methanol is converted in the first stage of the shift reactor that operates at 250° C. The second stage operates at 200°C and reduces the CO content from 1.5 % to 0.5 % using the water gas shift reaction. In the PROX reactor that uses a Pt/alumina monolith and operates at 180°C, the CO content is reduced to less than 10 ppm at steady-state by reaction with oxygen. However, as seen in Figure 2, the CO output of the reactor depends on catalyst zone length. This is because the residence time of the reactant gas mixture must be optimized to prevent the reaction between CO_2 and hydrogen to form more CO. This can be done by controlling the rate of flow of the gases through the reactor or optimizing the length of the reactor.

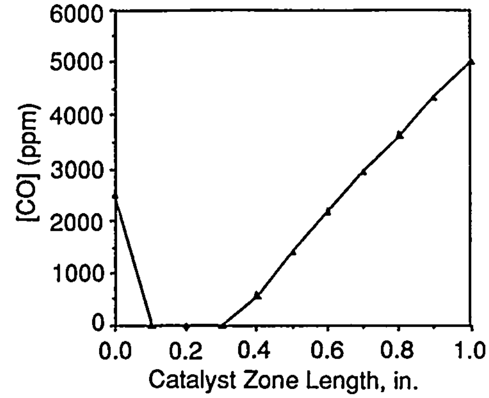


Figure 2. PROX reactor response

Fuel Cell Stack. Some of the issues of the fuel cell stack are water and heat management, high platinum loading and CO-contaminated fuel management. The fuel and air that enter the power section must be humidified to prevent membrane drying. This is handled by a separate humidification section. Thermal management of the stack requires several cooling plates for water circulation. The current approach to water and heat management therefore add weight and volume to the PEM fuel cell stack and alternative approaches are clearly needed to reduce stack weight and volume. On the issue of the high cost of catalyst, several recent developments are quite promising. In a recent GM invention, a Nafion slurry coating was used to reduce the platinum loading to less than 0.1 mg/cm^2. This method was also used to tailor the carbon supports and the platinum loadings to the differing needs of the fuel cell reactions.

Since pure hydrogen is not used in the stack, some problems arise due to the presence of CO_2 in the fuel mixture, as seen in Figure 3. Even in the absence of CO, the cell voltage at 1000 ASF drops by nearly 300 mV due to the presence of CO_2 in the fuel mixture. This is much higher than the 20 mV loss when hydrogen is mixed with 25 % nitrogen. CO_2 interacts with adsorbed hydrogen on platinum to form adsorbed COOH, which reacts with more hydrogen to form a COH poison that blocks the hydrogen oxidation reaction. Two approaches were used to tackle this problem. In the first approach, the fuel mixture was mixed with 2% air, which led to a 200 mV improvement in cell voltage. In a second

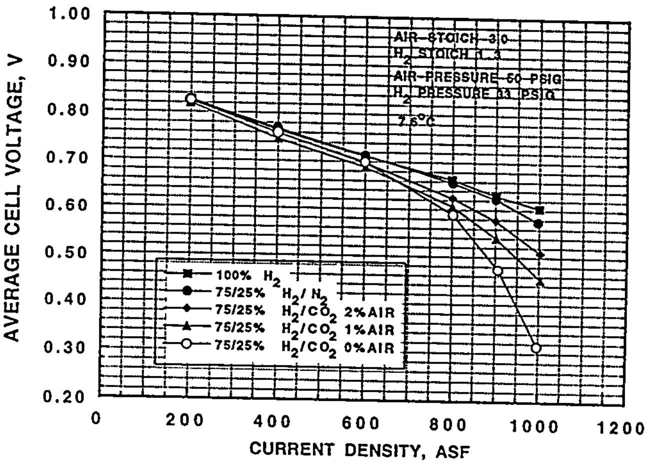


Figure 3. Effect of CO_2 in the fuel mixture

approach, the anode catalyst was changed to a Pt-Ru alloy. Ru has two effects. Being oxidized, it does not adsorb hydrogen, thus preventing the interaction of CO_2 with electrode sites. Second, even if CO is formed on an adjacent Pt site, Ru helps oxidize the poison since it adsorbs a water residue that can convert CO to CO_2.

System Integration. Several ancillary components are needed to make the fuel processor and the fuel cell stack function effectively. Some of these components are fuel and water pumps, vaporizers, filters, heat exchangers, compressor, expander, water condenser, combustor, pipes, valves and connectors, sensors, controllers and a battery pack. Some important system issues are integration of the various components, freezing, low grade heat from the stack and combustor fuel control.

FUTURE WORK

In the phase II follow-on effort, the primary objective is to develop and test a 60 kW brassboard system (30 kW fuel cell power plant and a 30 kW battery pack). In addition to fuel processor and fuel cell stack components and systems development activities, control systems, sensors and ancillary equipment needed for integrating the brassboard system will also be developed and tested. Major challenges include the development of a compact and a quick starting methanol fuel reformer with good transient characteristics and low CO emissions, and a low cost fuel cell stack with a high power density on reformate/air operation at relatively low pressures.

3.3 Phosphoric Acid Fuel Cell for Buses

CONTRACT INFORMATION

Contract Number	DE-AC02-91CH10447
Contractor	H-Power Corporation 60 Montgomery Street Belleville, NJ 07109 (201)450-4400
Contractor Project Manager	Arthur Kaufman
DOE Project Manager	Robert Kost, EE/OTT
Period of Performance	October 1, 1991 to October 31, 1994

ABSTRACT

A methanol-fueled, phosphoric acid fuel cell (PAFC) powered bus has been built under Phase II of a cost-shared Government program. (The Project is sponsored by the U.S. Department of Energy [DOE], the U.S. Department of Transportation [DOT], and California's South Coast Air Quality Management District [SCAQMD].) The current project is utilizing the technology of Phase I, in which liquid-cooled PAFC technology was shown to be suitable for use in transit bus applications. The objective of the Phase II project is to integrate and demonstrate a PAFC/battery power source in actual bus use. This project will produce three 30-foot test-bed buses and the conceptual design for a 40-foot bus. H-Power Corporation is the prime contractor for Phase II, while Bus Manufacturing U.S.A., Inc. (BMI) is the bus fabricator. The Contractor team also includes Transportation Manufacturing Corporation (bus configuration liaison; 40-foot bus design); Fuji Electric Co., Ltd. (fuel cell hardware); Soleq Corporation (power electronics); and Booz, Allen & Hamilton Inc. (bus system analysis and testing).

The methanol-fueled PAFC-powered bus offers attractive incentives in the areas of reduced noxious emissions (CO typically 50-100 times lower and NO_X typically 2000-3000 times lower than the 1998 federal heavy-duty diesel exhaust emission standards and virtually no hydrocarbons or particulate matter, based on current PAFC system technology); domestically-produced alternative fuel usage; higher propulsion system efficiency (higher vehicle mileage per unit of fuel energy content in relation to corresponding diesel power plants that operate on either diesel or methanol fuel); and greatly reduced noise pollution.

These fuel cell buses are being built so as to be fully consistent with the design requirements of heavy-duty transit buses in service today. These include performance criteria (acceleration; hill-climbing; top speed) as well as comfort (e.g., heating/air-conditioning) and

safety features. The test-bed bus incorporates seats for 25 passengers and the driver. In addition, in compliance with Americans with Disabilities Act (ADA) requirements, a wheelchair lift is provided along with means to accommodate two wheelchairs.

All of the primary energy for bus propulsion and accessories is supplied by methanol via the nominal 50kW PAFC system. A Ni-Cd surge battery, linked to the fuel cell output via a step-up d.c.-d.c. converter, provides incremental power for acceleration and hill-climbing. The battery is charged by the fuel cell during low-to-moderate loads and also accepts charge from braking energy. Power source specifications and the power/energy management concept have been optimized through the use of system simulation models.

The key hardware within the fuel cell subsystem (FCS) for the first test-bed bus (TBB-1) - methanol reformer and fuel cell stack - was tested successfully in 1993. The FCS was integrated during the latter part of that year; and subsequent testing, evaluation, and control optimization yielded results that meet the initial performance specifications for the FCS (115V at 480A, equivalent to a gross power output of 55.2 kW; system efficiency of 42.6% with respect to the lower heating value of methanol, based on net power delivered to the d.c.-d.c. converter).

TBB-1 was successfully integrated in March 1994, and it has been demonstrated at a variety of meetings and conferences starting in April. This test-bed bus is currently being prepared for a test program that will be conducted by H Power and Booz, Allen & Hamilton starting in August 1994.

TBB-2 and TBB-3 are currently under construction. Both are expected to be integrated before the end of 1994.

Session 4

DOD/ARPA's Fuel Cell Logistics Fuel Program

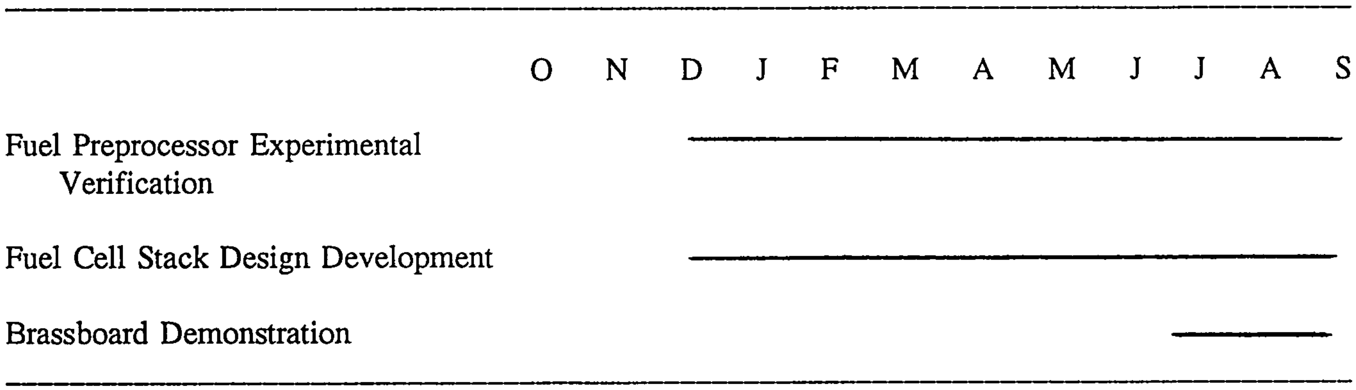
4.2 MCFC Development for Logistic Fuel Applications

CONTRACT INFORMATION

Contract Number	NAS3-27021
Contractor	Energy Research Corporation 3 Great Pasture Road Danbury, CT 06813 (203) 792-1460
Contractor Project Manager	Sandors G. Abens
Principal Investigators	Pinakin Patel, Martin Piwetz, George Steinfeld
NASA Project Manager	Paul Prokopius
Period of Performance	December 6, 1993 to November 30, 1995

Schedule and Milestones

FY94 Program Schedule



OBJECTIVES

Energy Research Corporation (ERC) is conducting a program for the development of fuel cell power plants for fixed and bare-base DoD applications under the ARPA "Fuel Cell Power Plant Initiative." ERC will adapt its direct carbonate fuel cell power plant technology in combination with a logistic fuel processing system for this dual use power plant application. This development is expected to improve the efficiency, fuel flexibility, emissions, and thermal and acoustic signatures of future fixed-base and mobile power plants.

Bare-base power systems are required to rapidly establish a sustainable operation at a location vital to various military missions. These power systems may be used globally, in all seasonal environments, and in any of the climatic extremes listed in MIL-STD-210B. The power systems must be rugged, portable, reliable, affordable, and simple to maintain.

The key features of the fuel cell power plant design include:

- Multifuel operation capability on logistic fuels (DF-2 and JP-8) and natural gas,
- An adiabatic logistic fuel preprocessor for providing pre-reformed fuel which is compatible with direct internal reforming fuel cell operation,
- High thermal efficiency - leading to 40% reduction in fuel consumption as compared with diesel engine gensets,
- Rugged fuel cell stack construction employing metallic cell hardware allows transportation and handling under military conditions, and
- Low thermal and acoustic signature, as well as negligible pollutant emissions.

The principal objective of the current two-year program is to develop and demonstrate a brassboard logistic fuel preprocessor operating with a 32kW Direct Carbonate Fuel Cell stack. Preprocessing of logistic fuels and militarization of the direct fuel cell stack are the key research areas. A key goal is to design a simple, reliable fuel system with minimum steam requirements. Hydrodesulfurization followed by adiabatic preconversion is planned to provide a methane-rich stream to be utilized in the internally reforming direct fuel cell stack. In parallel, a direct fuel cell stack design compatible with military requirements will be defined and evaluated.

BACKGROUND

Direct Fuel Cell Power Plant. The Direct Fuel Cell power plant is based on ERC's internal reforming carbonate fuel cell technology. This fuel cell concept is based on the chemical reactions shown in Figure 1. Methane fuel is reformed in the anode compartment of the fuel cell to produce CO and H_2. Both H_2 and CO are oxidized by the carbonate ion (CO_3^{--}) to H_2O and CO_2, discharging two electrons. The carbonate ions are formed on the cathode side of the fuel cell by reaction of O_2 and CO_2 plus two electrons from the external circuit. Since methane reforming is endothermic, it serves to remove heat produced in the fuel cell, minimizing fuel cell cooling air requirements. Because methane conversion in the fuel cell is achieved by waste heat, the direct fuel cell is inherently more efficient than conventional fuel cells operating on externally reformed fuel.

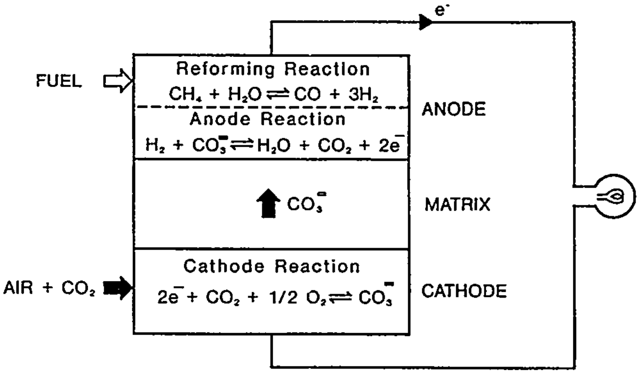


Figure 1. Direct Fuel Cell Concept

Individual fuel cell stacks can be grouped in modules identical to the one used for commercial natural gas power plants, with the exception of the design changes developed under the stack militarization task. This allows a dual use application for both commercial and military systems. Figure 2 depicts ERC's 1MW fuel cell modules which incorporate eight carbonate fuel cell stacks. Two of these modules are required for a 2MW power plant.

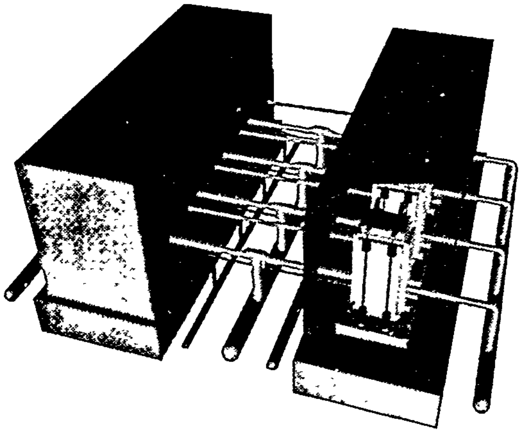


Figure 2. Engineering Model of a 1MW Carbonate Fuel Cell Module
Two modules containing four stacks each are required in a 2MW plant

The Direct Fuel Cell power system concept is shown in Figure 3. Oxygen and carbon dioxide are supplied to the cathode by a burner which combusts the residual fuel in the anode tailgas with excess air. Stack exhaust heat is used to raise the steam needed for reforming the fuel to hydrogen.

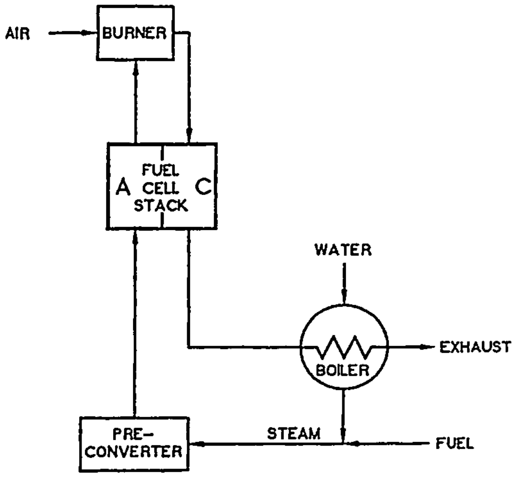


Figure 3. Direct Fuel Cell System Concept

Using computer codes available at ERC for the 2MW DFC power plant, thermal-to-electric efficiency has been estimated at 58% and 57% for DF-2 and JP-8, respectively, and at 61% on natural gas, all on a lower heating value basis. Heat recovery to produce hot water at 150°F can be provided, increasing the combined electric and thermal efficiency of the plant to 80-85%.

Logistic Fuels Characteristics. Logistic fuels such as diesel fuel (DF-2) and jet fuel (JP-8) provide a high energy density fuel offering transportability for mobile applications and back-up for fixed-base power plants which normally operate on natural gas. Such multifuel capability provides independence and versatility for military installations in the event of commercial power interruption or outage caused by natural disaster, accident, sabotage, vandalism or attack.

A logistic fuel processing front-end can be added to an existing DFC natural gas plant with minor changes in the remainder of the plant, and a water recovery system can be added to make the power plant water self-sufficient. Table 1 summarizes some of the characteristics of logistic fuels which are of interest for processing them for use in fuel cells. The higher sulfur content and lower H/C ratio makes processing of DF-2 somewhat more challenging compared to JP-8.

The logistic fuel preprocessor conceptual block diagram is shown in Figure 4. The two main processes are desulfurization and preconversion to methane.

Desulfurization. The key step in processing logistic fuels for fuel cell applications is desulfurization. Sulfur compounds in these fuels, which can be present up to 0.5 wt% according to Federal Specification VV-F-800D for Diesel Fuel, must be reduced below 1 ppm to maintain the catalytic activity of downstream catalysts. While commercial suppliers are already preparing to reduce sulfur levels to 0.05 wt% (500 ppm) in diesel fuel to meet clean air regulations, this level is still too high for fuel cell applications, and must be reduced further.

Table 1. Properties of Logistic Fuels JP-8 and DF-2

Both Fuels Have Similar Characteristics, But DF-2 Poses a Greater Processing Challenge

PROPERTY	JP-8	DF-2
Molecular Formula (avg)	$C_{12.4}H_{21.2}$	$C_{14.6}H_{24.8}$
Molecular Weight	170	200
H/C Ratio (Molar)	1.80	1.70
Sulfur Mass %	0.3 (max.)	0.5 (max)
Specific Gravity	.805	.850
Gross Heating Value Btu/lb	19,780	19,570
Gross Heating Value Btu/gal	132,600	138,600

Source: U.S. Army, Fort Belvoir Research Development and Engineering Center

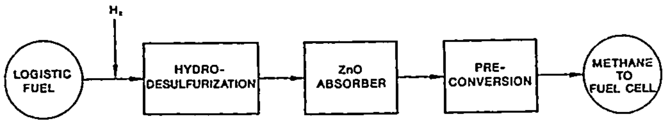


Figure 4. Logistic Fuel Processing for DFC

The desulfurization approach is based on utilizing advanced hydrodesulfurization catalysts to convert the sulfur compounds to hydrogen sulfide in the presence of hydrogen. The H_2S is removed by absorption in a ZnO bed. This hydrotreating step also lowers the aromatics content of the fuel, making it easier to preconvert to methane without carbon formation. A commercially available HDS catalyst was tested in a previous program[2] for phosphoric acid fuel cell applications.

Preconversion. Following the desulfurization step, an adiabatic preconversion step is used to convert the higher hydrocarbons in DF-2 and JP-8 to methane by the following reactions:

$C_nH_m+nH_2O$--->$nCO\ (n+m/2)H_2$ endothermic
CO +3H2 --->CH_4 +H_2O exothermic
CO +H_2O --->CO_2 +H_2 exothermic

The overall process can be operated adiabatically and has been shown in a previous DoD-funded project to be feasible for operating carbonate fuel cells with diesel-like fuels[1].

The endothermic steam reforming of hydrocarbons can be considered to be irreversible for all hydrocarbons above methane which are converted directly to carbon oxides and hydrogen with no intermediate products. This reaction is followed by the exothermic methanation and water-gas shift reactions. Calculations based on typical exit conditions for a diesel preconverter, as well as earlier subscale testing, indicate that a high methane content can be achieved. This makes operation on diesel fuel or jet fuel appear to the fuel cell as operation on natural gas, with a small variation in the gas composition. This facilitates the use of the same fuel cell design for logistic fuels as for natural gas, maximizing dual use and building on technology already developed for the commercial sector.

PROJECT DESCRIPTION

The current phase of the program is organized into the following tasks:

Task I: Analytical optimization and bench-scale experimental verification of the fuel preprocessor

Task II: Development and demonstration of a 32 kW militarized DFC stack design

Task III: Development of a brassboard logistic fuel system and the demonstration with the 32 kW DFC stack

Task IV: Preliminary design of a 2 MW logistic fuel fixed-base power plant

Task V: Preliminary design of a logistic fuel power unit for bare-base deployment

After initial analysis and bench-scale verification of the fuel preprocessing system, which started in early 1994, a brassboard system will be constructed. In parallel, approaches for fuel cell stack weight and volume reduction will be developed and verified at subscale and subsequently incorporated in a 32kW stack using full area cells. The brassboard fuel feed system and the 32kW stack will be tested with DF-2 diesel and JP-8 jet fuels. The results will be utilized to derive preliminary power plant designs for fixed and bare-base applications. Maximum power rating for a single fuel cell generator set, as well as a multiple-skid, transportable power plant will be defined, and a preliminary design will be provided considering the bare-base requirements specified by DoD. A preliminary fixed-base power plant design will be provided at the 2MW power level.

The detailed design requirements of bare-base deployment will be defined in a predesign mission analysis to evaluate design options that are consistent with bare-base deployment needs. Transportation requirements for C-130 transport will define the maximum power rating of the bare-base power plant which is currently anticipated to be in the range of 300 kW. A skid mounted unit containing fuel processing, fuel cell stacks and power conditioning sections of the plant will be packaged in a single ruggedized unit for transportability and field operation. Trade-offs will be considered between a single maximum rating unit transportable in one shipment by C-130 versus a larger modular power plant transportable on multiple skids which can be readily assembled in the field.

Team members for this project include ERC and its wholly-owned subsidiary, Fuel Cell Manufacturing Corporation; Haldor Topsoe, Inc. and its parent company, Haldor Topsoe A/S; and Fluor Daniel Corporation.

RESULTS AND CONCLUSIONS

The main activities on the project so far have been on Task I (fuel processor verification) and Task II (DFC stack design). Bench-scale testing with diesel fuel shows that desulfurization can be achieved by the proposed processes. Initial work on preconversion is currently in progress. Construction of the subscale DFC stack to be used for preliminary performance testing is underway also.

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ACKNOWLEDGEMENT

This work is being performed by Energy Research Corporation and its subcontractors as part of the ARPA Fuel Cell Power Plant Initiative. Technical and management direction is being provided by the NASA Lewis Research Center under Contract NAS3-27021.

4.3 SOFC Development for Logistics Fuel Applications

CONTRACT INFORMATION

Contract Number	NAS3-27022
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Principal Investigators	Stephen E. Veyo, W-STC Martin M. Piwetz, HTI W. Elgin Moss, SCE
NASA Project Manager	Paul Prokopius
Period of Performance	Dec 22, 1993 to Dec 31, 1995

ABSTRACT

Westinghouse has undertaken a two year project under sponsorship of the Advanced Research Projects Agency (ARPA) to develop and demonstrate the technology enabling use of logistics fuels, diesel fuel (DF-2) and jet turbine fuel (JP-8), in Solid Oxide Fuel Cell (SOFC) Power Systems for fixed military bases and bare base deployment. The project is administered by the National Aeronautics and Space Administration (NASA). The project team includes as major subcontractors Haldor Topsoe, Inc.(HTI) and Southern California Edison (SCE). The project will culminate in the demonstration at SCE's Highgrove Fuel Cell Test Center of an SOFC power plant fueled by logistics fuels. The SOFC generator module (stack) will employ the air electrode supported (AES) tubular SOFC developed by Westinghouse under a cooperative agreement with the Department of Energy (DOE), Morgantown Energy Technology Center (METC). The AES-SOFC stack will have a maximum output of 30 kWe. The Logistics Fuel Processor (LFP) is under development by Haldor Topsoe, Inc. and will provide a sulfur free, methane rich fuel stream to the SOFC. HTI will construct a brass-board LFP as a non-integrated subsystem that will be coupled to an SOFC system now in place at the SCE site. Prior to the SOFC/LFP demonstration, the fuel cell stack presently in use at the SCE site will be replaced with a new one employing the AES-SOFC. SCE will install and operate the SOFC/LFP system.

Concomitantly, Westinghouse and HTI will provide conceptual designs for two deployable SOFC/LFP power systems of practical size (200 kW and 2 MW) utilizing the SOFC technology being developed under the Westinghouse/DOE-METC cooperative agreement and the results of the LFP development work done under this program.

Preliminary LFP development results indicate that diesel fuel can be desulfurized to the less than one ppm sulfur content required.

Preliminary system design results indicate that without intimate integration of the SOFC and the LFP, process water will be required for fuel reformation in the amount of approximately four gallons per hour per 100 kW of nominal electrical generating capacity .

Future LFP development effort will complete the experimental definition of process parameter values for the desulfurizer and fuel reformer thereby permitting design and fabrication of the brass-board hardware and the conceptual designs for deployable LFP subsystems.

Future SOFC and system related effort will conclude the definition of requirements and produce conceptual designs for deployable SOFC generator modules (stacks) and integrated SOFC/LFP systems.

Testing of the brass-board SOFC/LFP is expected to begin at the SCE site in the second quarter of 1995.

Diesel and jet fuels are commonly available storable liquid fossil fuels. Successful demonstration of diesel and jet fuels with SOFC power plants, coupled with the already demonstrated use of pipeline natural gas, proves dual fuel capability and therefore should expand the potential attractiveness of the SOFC to electric utility and industrial customers in the commercial sector. If the SOFC/LFP can successfully meet military requirements, then there exists the potential for accelerated technology development because of dual civilian/military use.

Session 5

Molten Carbonate Fuel Cells

5.1 Update: Commercialization of the Direct Fuel Cell

B.S. Baker, C.R. Bentley and D.R. Glenn
Energy Research Corporation
Danbury, CT and Washington, DC

The commercialization of the Direct Fuel Cell (DFC), ERC's trade name for its internal reforming carbonate fuel cell technology, moved into an important phase this past year. Activities were characterized by extensive testing both at ERC and user sites. The next period requires a continuation of a product improvement program to further refine all aspects of the stack, module and plant system, while concurrently affecting the transition from a technology-driven effort to a full scale prototype power plant hardware demonstration.

R&D: THE BASIS FOR PRODUCT DESIGN

The ERC program has been dedicated to completing the component and stack, stack module and system designs for power plant demonstration projects. However, other important milestones were also met this past year that add to the DFC's commercial success potentials. These included the successful operation of a 30 kW stack on coal gas and a laboratory-scale stack test fueled directly by ethanol.

Two full size (125 kW) stacks were built this past year by Fuel Cell Manufacturing Corporation (FCMC) under Dept. of Energy (DOE) contracts for testing at ERC's Danbury facility (See Figure 1). The last of these stacks is now being tested in the subscale 125 kW power plant facility which incorporates many features of the 2 MW Santa Clara Demonstration Project (SCDP). A more comprehensive treatment of the R&D accomplishments relating to the SCDP, other fuels options and overall technology progress is being presented in other papers at this meeting.

PREPARING FOR THE COAL OPTION

Fuel Cell Engineering Corp. (FCE) was selected for an award in the Fifth Round solicitation of the Clean Coal Technology program. The selection is part of an IGCC project with Duke Energy Corp. as the prime contractor. The project scope includes a British Gas Lurgi (BGL) gasifier complex integrated with a combined cycle turbine system and ERC-based carbonate fuel cell technology in an FCE-designed1.25 MW power plant submodule. ERC has been involved in several studies of coal gasification fuel cell systems using the BGL gasifier. It is favorable for the DFC because the gas contains a relatively high methane content thereby providing some cooling capability to the fuel cell. This project will provide the experience to commercialize a phased capacity addition product to supply larger size modules in conjunction with coal and natural gas-fueled baseload installations.

As a first step to fielding an integrated gasification-fuel cell plant, ERC supplied a 20 kW stack, from FCMC, and Haldor Topsoe Inc. built a mobile Balance of Plant (BOP) for coal gas testing. All funds for this construction activity were provided by the Electric Power Research Institute (EPRI). The coal gas tests were conducted at Destec

Energy Inc., Plaquemine, Louisiana where their coal gasification system provides coal gas for a turbine-power generation system. Figures 2 to 4 show the initial stack data at ERC and in Louisiana on natural gas, connected to the gasification system at the Destec facility, and the performance over the 4,000hour test period on the carbon monoxide rich coal gas.

Despite numerous facility-related operating discontinuities during the 4,000 hour test, including an errant supply of water infiltrating the stack's cells and several thermal cycles, the unit recovered and exhibited resilience in generating power under adverse conditions whenever coal gas was available. This experience will be applied to the designs in the Clean Coal V project. This test, the first time a fuel cell of any kind was operated on real coal gas, demonstrated the dual fuel capability of the Direct Fuel Cell.

Additional coal gas testing of ERC stacks took place in Germany with an 11,000 hour test on simulated coal gas including impurities. More recently, tests on two stacks began there on real coal gas evaluating both hot and cold gas cleanup systems.

ALTERNATIVE FUELS

Under contracts from the National Rural Electric Cooperative Association (NRECA) and the Tennessee Valley Authority, ERC operated a lab-scale DFC stack on ethanol. These tests results will be available soon in an NRECA report. The overall corn to electricity option is shown conceptually in Figure 5.

ERC also operated its DFC on upgraded Exxsol, a sulfur-free diesel-like fuel, using a methane producing fuel processor operating at temperatures below that of the fuel cell. This process unlike steam reforming or partial oxidation does not require an external energy input.

COMMERCIALIZATION: THE ULTIMATE OBJECTIVE

The two ERC subsidiaries, FCMC and FCE, are the spearheads for producing, selling, installing and servicing the coming commercial units.

• Manufacturing

Fuel Cell Manufacturing Corporation (FCMC) is a privately funded carbonate fuel cell stack and module production facility. Located in Torrington, CT, the facility was designed to provide capability for 2 MW per year of stack production on a single shift basis. The plant has recently been increased to 63,000 sq.ft. to accommodate stack module assembly. Employment has grown to 46 people in support of the production of components for the Santa Clara Demonstration Project (SCDP) and other activities.

Process trials began in the summer of 1991. Laboratory processes developed at ERC were transferred to production scale equipment. Batch processes were converted to continuous processes in many cases, and material handling and process control systems implemented. During this period, fuel cell active area was increased from 4,000 to 6,000 sq.cm. Limited production of full-sized components was achieved and shipments began in July of 1992. A chronological summary of events is shown in Figure 6.

After successfully completing the

startup of the initial scope of work, additional operations have been vertically integrated to improve quality and reduce cost. Some of the in-house production processes currently used are shown in Table 1.

Table 1
FCMC Production Processes

- Milling
- Blending
- Tape Casting
- Sintering
- Electrolyte Filling
- Heat Treatment
- Sheet Metal Forming
- Shearing
- Packaging
- Inspection
- Stacking
- Manifolding
- Instrumentation
- Insulation
- Welding
- Shipping

A stacking machine for tall stacks is shown in Figure 7.

Technology transfer from ERC has been successfully proven in 2, 8, 20 and 125 kW stacks; the most recent of these being a prototype for the stacks to be supplied for the 2 MW Demonstration Project. Thousands of active components have now been manufactured and this volume has provided the opportunity for successful process optimization. Statistical process control has led to yield improvement at virtually every manufacturing operation. The most recent, a 258 cell stack, showed maximum OCV variations of ±0.2%. In addition, equipment modifications and methods improvements have resulted in enhanced output capability. The present facility is capable of producing about 10 MW/year based on multi shift operation and certain operations can yield up to 50 MW/year with existing equipment.

While production of the stacks and stack modules for the SCDP continues, the parallel focus is on developing plans for full scale commercial manufacturing. This involves long range cost improvement activity, vendor development and facilities planning.

• Power Plant Engineering

FCE is the principal corporate contact for plant engineering, construction/installation, sales and after market service functions. With this charter, FCE staff have regular interface meetings with the Fuel Cell Commercialization Group (FCCG) members/buyers and the committees formed to assist in the commercialization process. A major emphasis has been on fielding the first 2 MW demonstration, and producing a conceptual design and first cost estimate of the commercial unit, and a model contract format that would reduce legal time (hence costs) to negotiate contracts with buyers.

The DOE-SCDP-EPRI co-funded 2 MW demonstration program celebrated its groundbreaking in early April. The progress of this project is the subject of another paper at this meeting.

FCE, working together with team members, Fluor Daniel and Jacobs Applied Technology, produced the first complete conceptual design for a commercially viable 2.7 MW DFC power plant in May. The plant retains the sought high degree of modularity, fewer stacks and a footprint that meets the "tennis court" criteria set very early in the program. (A comparison with SCDP is shown in Figure 8). The unit incorporates two modules each comprised of four 375 kW (BOL nominal output) DFC stacks arrayed in a round module container and interconnected within the enclosure. The BOP has a heat recovery steam generator module that integrates all thermal management functions to minimize high temperature service piping runs resulting in materials cost savings and improved efficiency. The power conditioning module is state-of-the-art with VAR control capability to dynamically adjust power quality to match grid requirements. Initial cost estimates based on the conceptual design for this plant and presented to the FCCG are about $1,200/kW (1990 dollars), somewhat higher than desired but not inconsistent with other DFC studies conducted earlier.

Over the past year, the company has prepared and refined a formal Commercialization Plan describing the organizations involved in the commercial unit's evolution, both within and outside of ERC, the applications and market capture potentials for the 2.7 MW unit and descriptions of the remaining technical, financial and business issues. This new program roadmap is intended to update materials that were the foundation for ERC's selection in 1990, by the American Public Power Association's National Review Team.

The plan will be a valuable asset to assist raising the needed additional capital investments to construct commercial production facilities, install the necessary infrastructure to define the support systems to assemble, ship, install and service the commercial units, and staff factory and field teams.

ACKNOWLEDGEMENTS

ERC and its subsidiaries would like to acknowledge the support of DOE and EPRI in the areas of stack design, construction and testing including facility design; SCDP for the 2 MW BOP design and construction; NRECA and TVA for alcohol fuel activities; Destec Energy Inc. and RWE AG for coal gas testing and FCCG for guidance relative to utility needs in North America.

LIST OF FIGURES

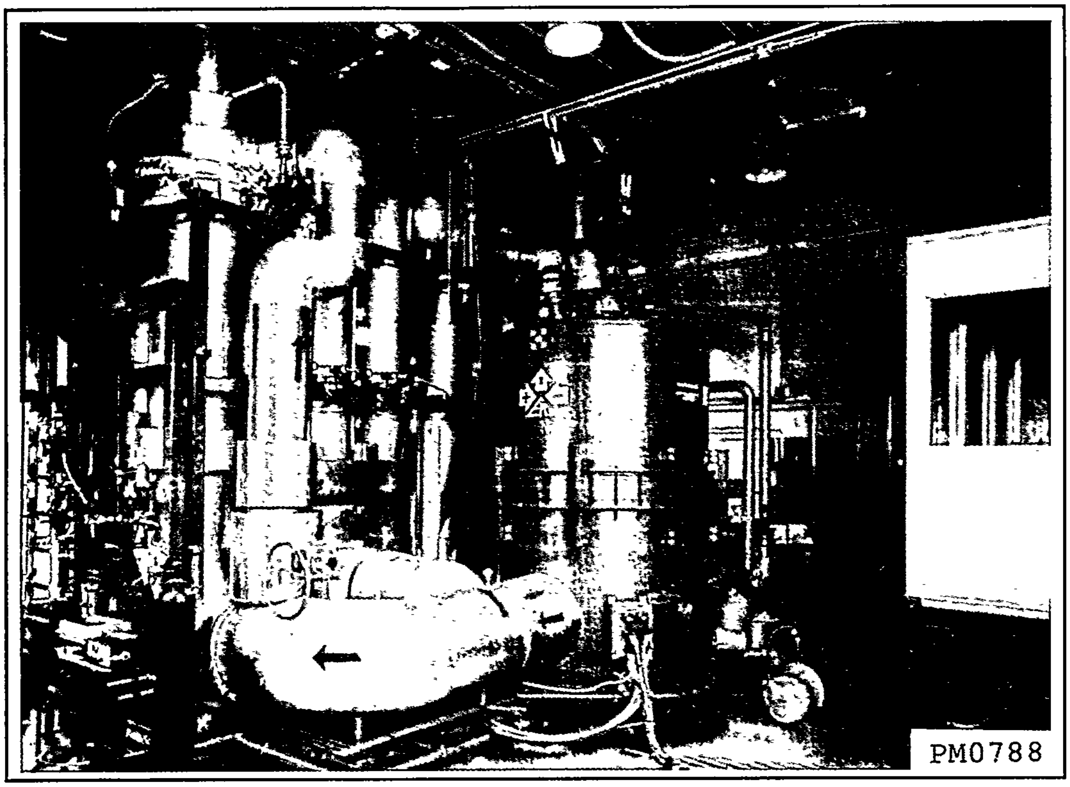


Figure 1. DFC Test Complex

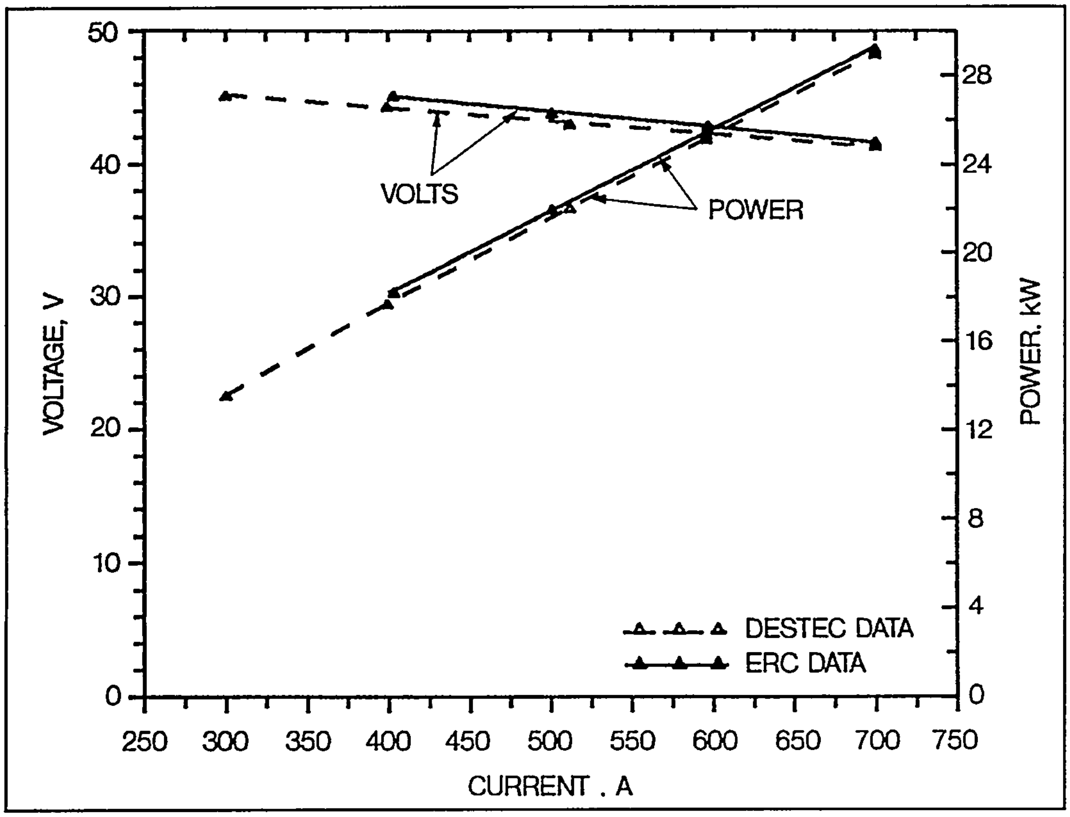


Figure 2. Comparison on Natural Gas Fuel at Different Sites

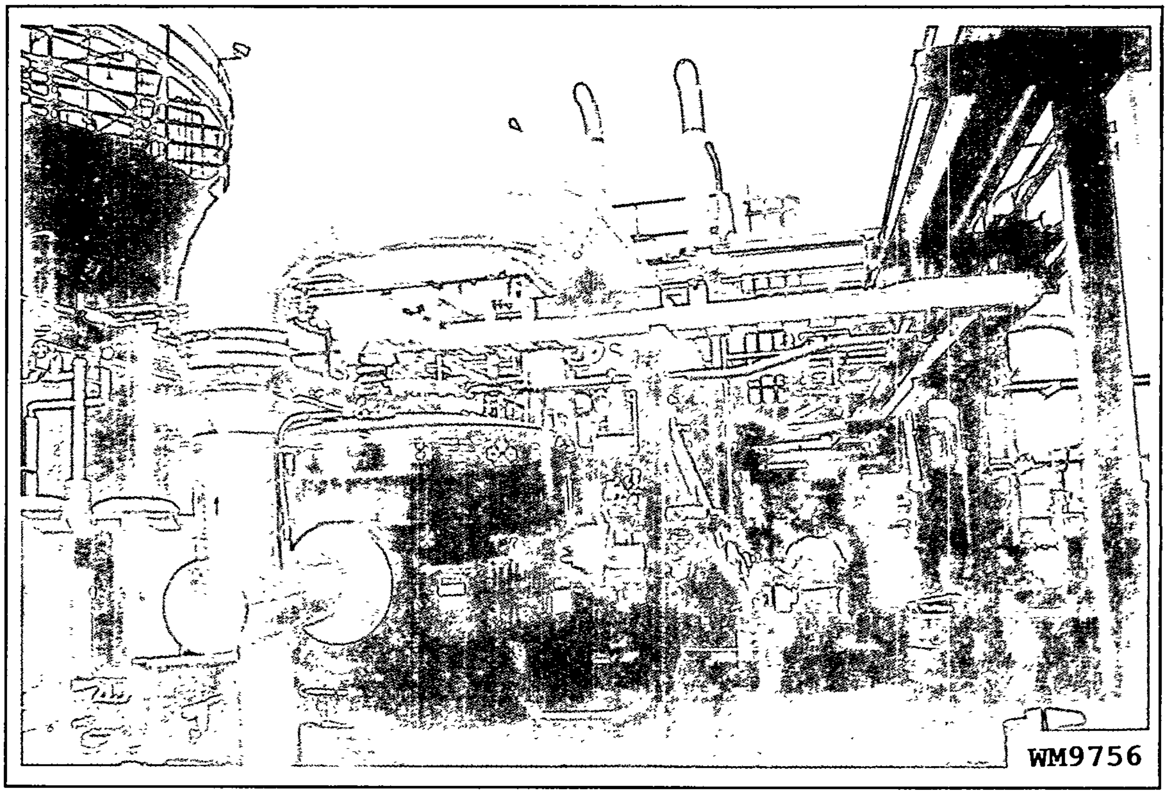


Figure 3. Stack Installed at Destec

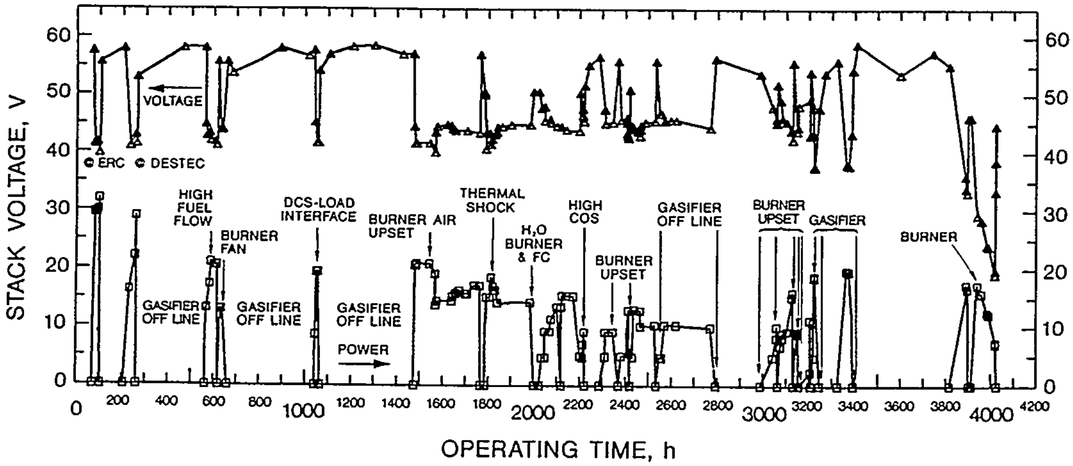


Figure 4. Coal-gas Performance History

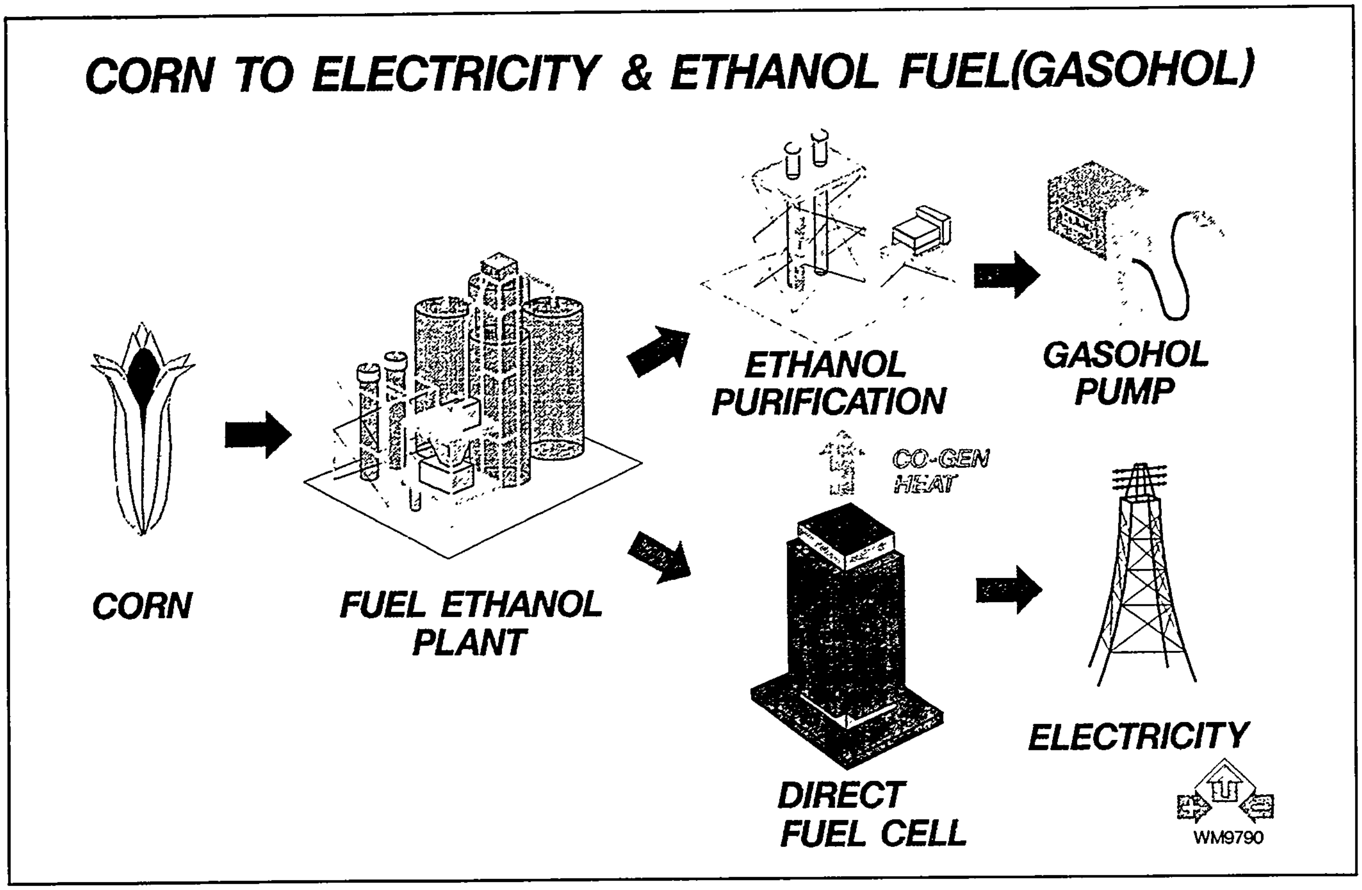
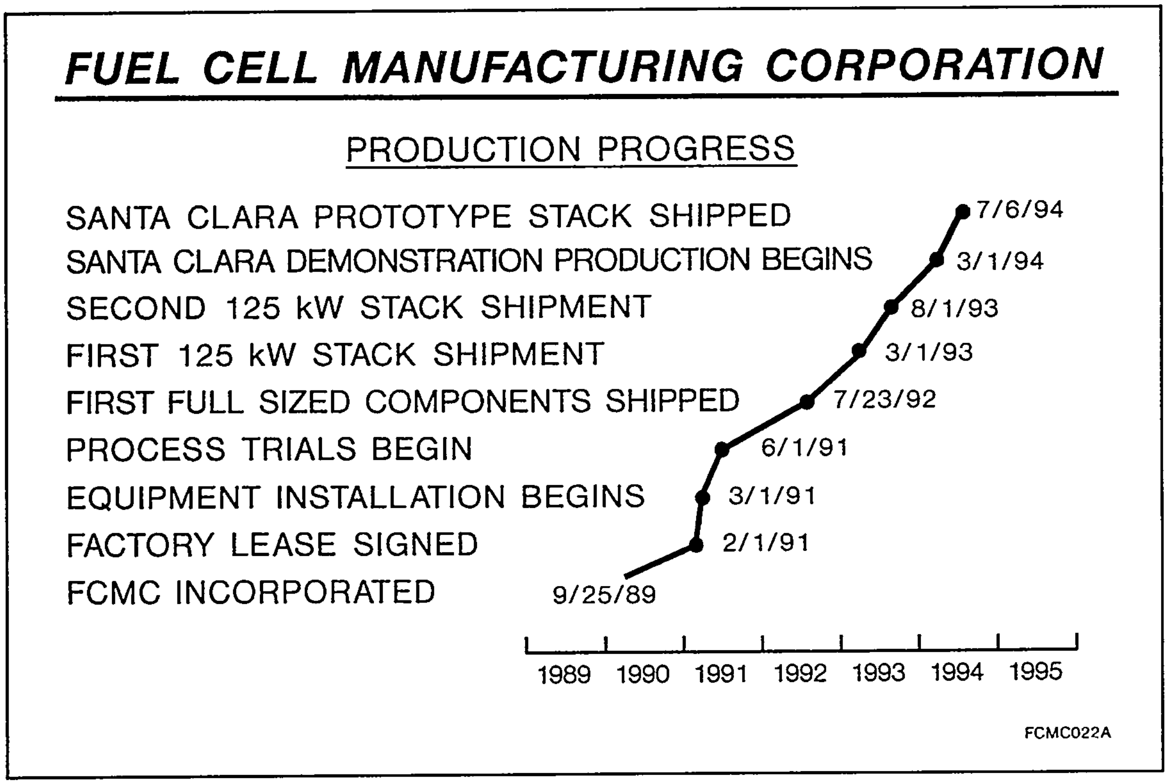


Figure 5. Corn to Ethanol Approach


FUEL CELL MANUFACTURING CORPORATION

PRODUCTION PROGRESS

SANTA CLARA PROTOTYPE STACK SHIPPED
SANTA CLARA DEMONSTRATION PRODUCTION BEGINS
SECOND 125 kW STACK SHIPMENT
FIRST 125 kW STACK SHIPMENT
FIRST FULL SIZED COMPONENTS SHIPPED
PROCESS TRIALS BEGIN
EQUIPMENT INSTALLATION BEGINS
FACTORY LEASE SIGNED
FCMC INCORPORATED

Figure 6. Chronology of FCMC Activities

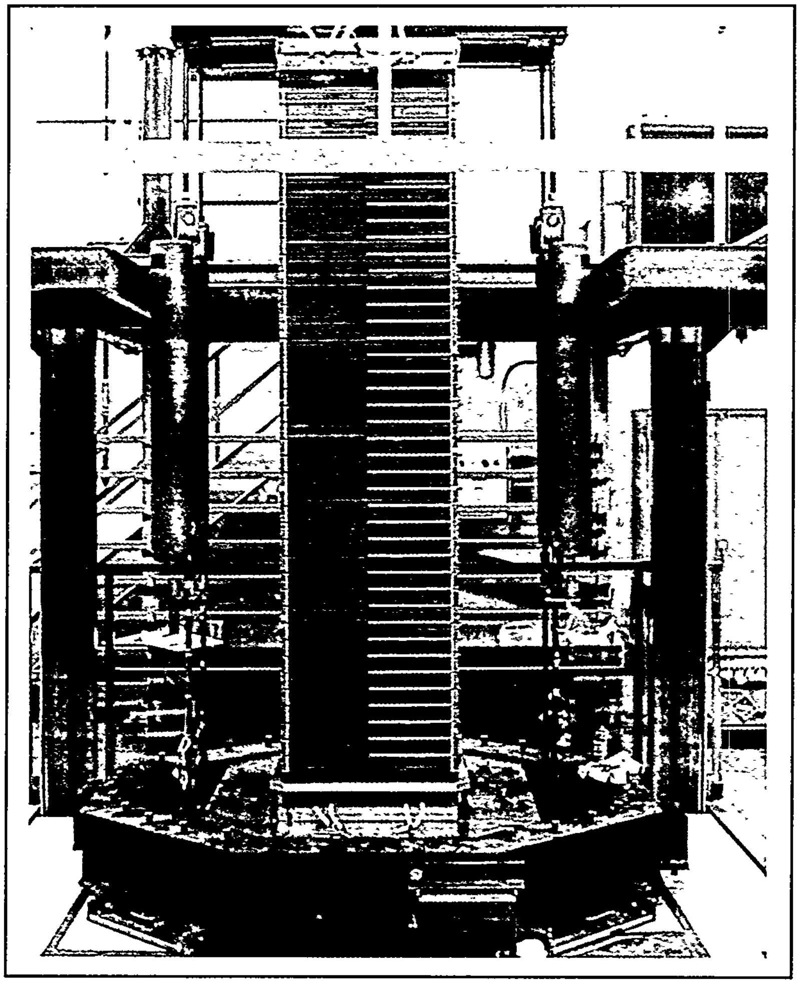


Figure 7. Stacking Equipment at FCMC

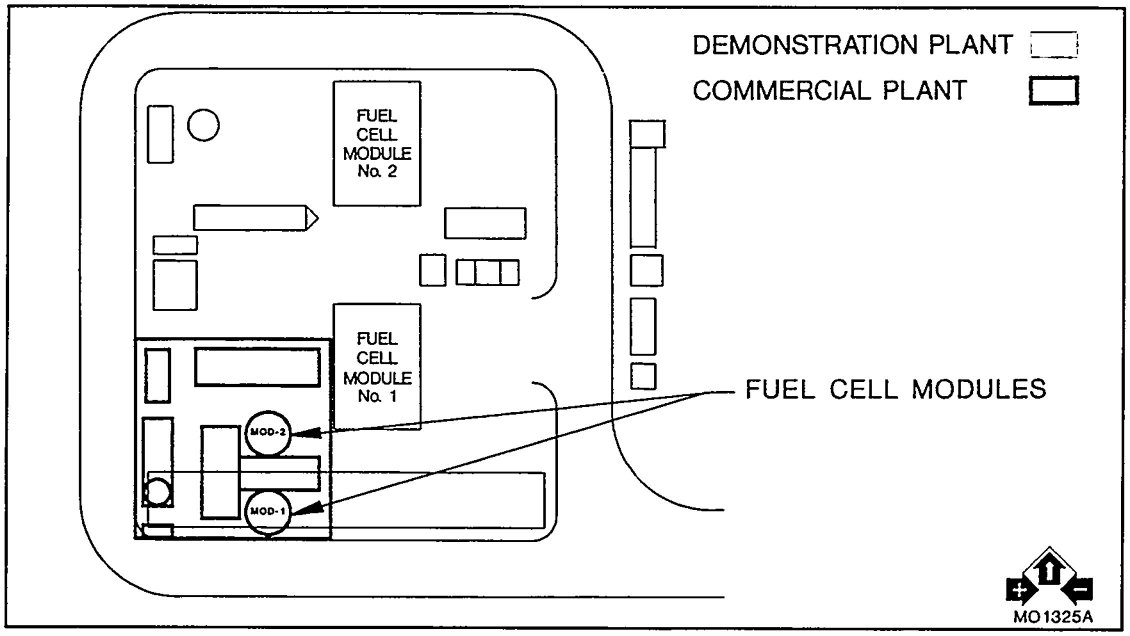


Figure 8. Size Comparison of SCDP and First Commercial Prototype

5.2a Overview of MCFC Commercialization Strategy

CONTRACTOR INFORMATION

Contract Name	DE-AC21-90MC27394
Contractor	M-C Power Corporation 8040 S. Madison Street Burr Ridge, IL 60521 (708)986-8040
Contractor Project Manager	Joseph A. Scroppo
Principal Investigators	Rene M. Laurens Vincent J. Petraglia
METC Project Manager	Mark C. Williams
Period of Performance	September 1, 1991 to March 31, 1995

Introduction

M-C Power Corporation (MCP) was founded in 1987 with the sole purpose of commercializing the IMHEX® molten carbonate fuel cell (MCFC), invented by the Institute of Gas Technology (IGT). IMHEX uses internal manifolding to distribute both anode and cathode fuel and exhaust gases. The design is similar to that used in commercial flat plate, internally manifolded heat exchangers, hence the name IMHEX. Like its commercial counterpart, this design allows the use of a single vertical force to maintain all manifold and edge-of-stack seals while simultaneously maintaining electrical cell-to-cell continuity. The design has been successfully operated with 11-square foot commercial size cells in 20-cell short stacks. Full commercial height stacks of 250 cells will be tested at Unocal later this year and in San Diego mid next year.

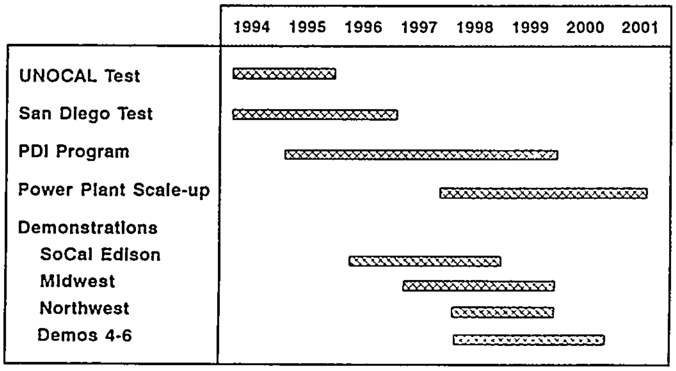
M-C Power's commercialization plan encompasses five key strategies:

- Achieve development program goals,
- Establish strategic alliances with other firms,
- Secure contingent orders for our market-entry units,
- Construct new manufacturing facility to meet future production growth, and
- Reduce product costs for both market-entry and future production.

Development Program

Our development program is being funded by DOE, GRI, EPRI and a host of industrial firms. The program encompasses a five year period with market entry planned for 1999. Figure 1 identifies the five major activities for the development program. The progression of activities includes the testing of single commercial size, 250-cell stacks at Unocal and San Diego in the 1995-6 time frame followed by the testing of a multiple stack, one-megawatt power plant at Southern California Edison (SCE) in 1997. The SCE demonstration is the culmination of a 50-month Product Design & Improvement (PDI) project now being negotiated with DOE.

Table 1. Development Program Schedule



The PDI project is the foundation of our development program. It not only provides the funding needed to advance our technology from the testing phase to the demonstration phase, but it also provides for the first demonstration of our technology at the megawatt class market-entry size. Up to five additional demonstrations are planned. We are currently negotiating with two companies, one in the Midwest and one in the Northwest, which are interested in hosting a demonstration unit. We believe that several demonstrations covering a broad spectrum of applications and territories is required to convince the power generation industry of the viability of the IMHEX technology.

Our development program also includes an activity to scale up the IMHEX power plant from the megawatt size planned for market-entry to the multi-megawatt size more suited for the emerging distributed power generation market of 1-20 megawatts. Fuel cells in general, and IMHEX in particular, have significant advantages over other methods in this power category because both high efficiency and low emissions can be maintained in small size power plants.

Strategic Alliances

Because M-C Power is a small start-up company and because both the development and market-entry phases of our commercialization program encompass so many different facets, we elected to carry out our commercialization program by forming strategic alliances that use the expertise of others. This approach not only takes advantage of the specific capabilities of other companies more experienced in a particular field, but also allows M-C Power to concentrate our limited capabilities on the development of a reliable, cost-competitive IMHEX stack.

The first such alliance formed was the TEAM assembled to carry out the development phase of our program. This Team comprises four members, each with a distinct responsibility;

- M-C Power, development of the IMHEX stack
- IGT, advancing IMHEX technology,
- Bechtel, system process design, and
- Stewart & Stevenson Services, for the package design of a unitized (skid-mounted) power plant.

Another strategic alliance formed early by M-C Power is the Alliance to Commercialize Carbonate Technology (ACCT). ACCT membership includes over 60 utility and industrial companies which monitor and guide our development program. One of the principal services of ACCT is to help guide the design of our market entry power plant and to stimulate demand for this plant. ACCT is discussed in more detail later in this paper.

A clear path to commercial success requires a detailed knowledge of the target market and the type of power plant required to capture this market. For this reason, M-C Power executed an agreement with Stewart & Stevenson to fabricate, sell, and service IMHEX power plants in this market. Stewart & Stevenson in the premier fabricator of unitized plants for distributed power generation throughout the world. Their current product is based on aeroderivative turbines in 20-50 MW sizes. Stewart & Stevenson believes that IMHEX can complement their product line in the smaller capacity ranges of 1-20 MW. Our agreement with Stewart and Stevenson allows the assignment of territorial distributorships, which the Team can offer as inducements to companies interested in joining our Development Program.

Because of the close thermal integration of the reformer and stack in an MCFC power plant, it is important that the fabrication of both elements be continuously and closely coordinated as advances in technology occur. M-C Power is therefore negotiating an agreement with Ishikawajima-Harima Heavy Industries (IHI) which will permit M-C Power to fabricate flat plate reformers specifically designed by IHI for MCFC power plants. M-C Power has successfully tested a small 20-kW reformer and the data obtained is being used by IHI to design and fabricate a 250-kW reformer for use in the San Diego Test. Once the technology is successfully scaled to commercial size, M-C Power will execute a license to fabricate and sell IHI's reformer in the U.S.

Because the IMHEX stack and the IHI reformer can be tailored to fit a broad range of power production by adding or subtracting plates and stacks, it is likely that market-entry size will be determined by another key system component. Therefore, we are working with various equipment vendors so that they can become more familiar with the equipment needs and market potential of IMHEX power plants. This will allow the team to tailor the design. Early involvement prior to commercialization is an important aspect of this relationship. Current plans are to develop such relationships with manufacturers of our bipolar plate, recycle gas blower, inverter, etc.

Cost Reduction

Cost reduction is an ongoing effort for all commercial products and is especially important for new emerging products. It is generally believed that fuel cells can achieve market-entry at a price of about $1500/kW. Further cost reduction to a price of about $1000/kW will be needed to capture a significant portion of the distributed power generation market. At this price, and with its high efficiency and low emissions, IMHEX will have distinct advantages over turbines and other methods of power generation in the 1-20 MW size category.

The objective of the Development Program is to identify and address those factors which can lead to a reliable, cost-competitive IMHEX power plant. In order to guide this program and focus our efforts on future costs, M-C Power has developed a comprehensive Manufacturing Cost Program that predicts each cost related to the manufacture of IMHEX stacks at M-C Power, both in our current and future manufacturing facilities. This program predicts costs as a function of production rate for the,

1. type and amount of each active component (anode, cathode, electrolyte, and matrix)
2. type and design of each non-active component (bi-polar plate, current collector, etc.)
3. type and design of each non-repeat part (end plates, holding bars, etc.)
4. type and capacity of each process used (tape casting, electrode sintering, etc.)
5. type and capacity of assembly and testing procedures used (manual or automatic)
6. labor required (both direct and indirect)
7. space required (for both manufacturing and office)
8. capital required (for both machinery and buildings)

The most important use of the Manufacturing Cost Program is to guide our stack development efforts. Because the program is interactive, questions regarding any and every manufacturing step can be quickly analyzed.

- Value of using thinner components? Different components?
- Try other manufacturing processes? Accelerate current processes?
- Optimum number of cells per stack? Stacks per power plant?
- What is the optimum production capacity per manufacturing line? Per factory?

Answers to such questions provide M-C Power the means needed to direct our development efforts towards the most cost-effective stack production methods possible.

The Manufacturing Cost Program, when combined with our market analyses, also allows M-C Power management to forecast future business scenarios and determine at what production rate and product cost is M-C Power a viable profitable business. Our present forecasts indicate that it will be the attainable growth in production capacity, not market limitations, that control our future business. In the near term, during the market-entry years, product costs will be labor and capital intensive. In later years, at higher production rates and overall lower costs, product costs will be materials intensive.

The Manufacturing Cost Program has only recently been established as a viable working tool at M-C Power. And it is presently limited to predicting future stack costs. Our goal for the near future is to extend the program, or build a similar program, that our Team can use to predict costs for the entire power plant, including each item in the balance of plant.

Conclusion

M-C Power has made considerable progress in the last several years due principally to our reliance on Team members for product development and on a large number of companies for their participation in our Development Program. This is our most important business strategy and we will continue this strategy until and throughout commercialization.

5.2b The Alliance to Commercialize Carbonate Technology: A Revitalized Industry Commitment to IMHEX® Fuel Cell Commercialization

John T. Nimmons, J.D.
Managing Director, ACCT

Background

The Alliance to Commercialize Carbonate Technology, or "ACCT", is a key element in the commercialization strategy for IMHEX molten carbonate fuel cell (MCFC) technology. It is a working alliance *among* prospective buyers and users of MCFC's, and *between* those organizations and the team that is designing and developing, and will manufacture and distribute, IMHEX power plants.

ACCT organizations include privately- and publicly-owned electric, gas, combination, and other utilities, natural gas pipelines, independent power producers, institutional and industrial users, and representatives of the IMHEX development team led by M-C Power Corporation. ACCT's Steering Committee presently includes PSI Energy, Southern California Edison, San Diego Gas & Electric, Southern California Gas, Tenneco Gas, and M-C Power.

ACCT was formed in 1991 to stimulate the commercialization of MCFC's using technology pioneered by the Institute of Gas Technology, and now under commercial development by M-C Power and other IMHEX team members (identified below). ACCT's activities since than have reflected M-C Power's step-by-step approach to MCFC commercialization, based on lessons learned from earlier fuel cell efforts by others.

M-C Power and ACCT recognized that the success of specialty fuel cells used in America's space program has led many to underestimate the difficulties of developing larger, more versatile, and less costly fuel cells for utility and commercial applications. They were aware of the risk of raising expectations too far, too fast, before the technology's performance, reliability and cost-effectiveness could be demonstrated to users, and they resolved to follow a more gradual path in bringing IMHEX technology to the market.

From the start, ACCT's leaders have tempered their enthusiasm over the promise of IMHEX technology, with realism about the technical and business challenges facing all fuel cell commercialization. They have followed a deliberate and incremental approach, intended to foster industry interest in the technology at a pace that tracks its development progress.

Thus ACCT's early efforts during the technology development phase were directed mainly toward keeping participants informed of M-C Power's progress on stack development, and identifying preferred design and performance characteristics through a series of informal user surveys and discussions. ACCT companies also began work with M-C Power to focus development activities on lower-cost product options, so that early power plant sales will be able to sustain low-volume production while fuel cell markets grow.

Through these interactions, ACCT's leaders gained increasing confidence in M-C Power's cell stack technology. They also watched M-C Power assemble a formidable team that complements its own capabilities in cell stack development and manufacturing with the expertise of world leaders in the design, construction, and marketing of complete power plants. The IMHEX development team now includes The Bechtel Corporation, whose industrial process and power plant experience promise an efficient, well-integrated product; Stewart & Stevenson Services, Inc., whose fabrication capabilities and service infrastructure can help ensure an economic power plant and effective distribution channels; and the Institute of Gas Technology, whose research talents allow focused resolution of difficult technical issues and evaluation of advanced fuel cell components.

ACCT Transition Initiative

Progress in these areas convinced ACCT's leaders that M-C Power is serious about working with prospective utility and industry customers, and can attract venture partners with the experience and credibility needed to move IMHEX power plants into commercial markets. In keeping with ACCT's incremental approach of building industry commitment in step with IMHEX development progress, ACCT's Steering Committee therefore embarked on a major new initiative early in 1994 to expand member involvement in the IMHEX commercialization program.

In formulating this initiative, the Steering Committee agreed with the IMHEX team that, as efforts shift from technology development to commercialization, ACCT can be most effective as a strong, user-driven organization, operating at arms' length from the IMHEX team but in a close collaborative relationship with it. Thus ACCT's main tasks over the past six months have been to reexamine and enhance the benefits it can offer to its utility and industry members, and to define the most constructive form of collaboration between those members and the IMHEX team.

Following the Steering Committee's announcement of its initiative at ACCT's March annual meeting, two *ad hoc* Task Forces were formed to address these questions. They included representatives of utility leaders in fuel cell commercialization, as well as utilities and industries which have only recently recognized the potential value of fuel cells in their competitive portfolios. What these Task Force-members share in common is a willingness to contribute considerable time and energy to redesign ACCT to serve their companies' needs in the commercialization of MCFC's.

Task Force Results & Future Prospects

The result of their efforts has been to more clearly define ACCT's objectives, the benefits available to its members, the nature of its collaboration with the IMHEX team, and the structure needed to advance its purposes.

ACCT Task Force members have affirmed the following organizational objectives:

- To increase industry understanding of MCFC technology and applications, and the regulatory and business environment in which they will compete;
- To help focus and support IMHEX development and demonstration efforts in areas of critical interest to member organizations:
- To provide opportunities for members to shape commercial power plant design, participate in demonstrations, and share early MCFC experience;

- To build members' confidence that power plant performance, reliability, and durability can be achieved at competitive costs; and

- To develop a framework for early power plant purchase commitments.

ACCT's Task Forces have also established a variety of benefits available to members. These include, among others, access to technical and program information; attendance at general membership and working committee meetings; and participation in power plant definition, demonstrations, targeted MCFC applications studies, and industry negotiations for early commercial orders. Valuable additional benefits in the form of dues rebates, purchase credits, distribution rights, and reserved units are also proposed for members who provide additional support for ACCT's activities.

Apart from the very tangible benefits ACCT now proposes to offer its members, ACCT's Steering Committee and Task Forces have explicitly recognized that the organization's highest value may lie in providing a mechanism through which members can help the IMHEX team concentrate its efforts on matters of critical interest to them as customers. These include building successful demonstration units and streamlining product development efforts to produce a basic introductory power plant on a reasonable schedule, at a competitive price. These are the focus of ACCT's collaboration with the IMHEX team, and will be addressed through joint participation in the standing committees and issue-oriented task forces, whose structure is illustrated below.

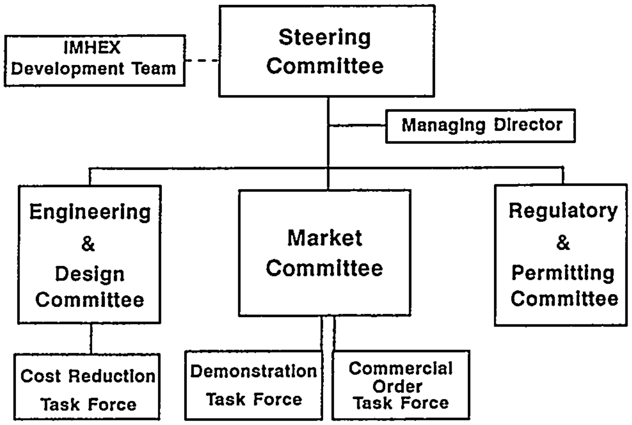


Figure 1. ACCT Structure

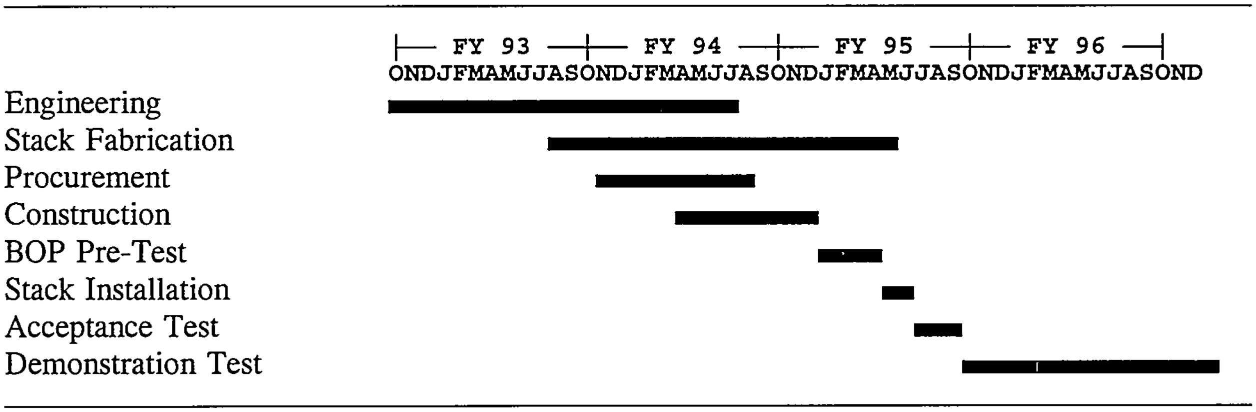
ACCT's Steering Committee and members of its *ad hoc* Task Forces, with M-C Power's strong encouragement and support, have shown impressive dedication over the past six months redesigning ACCT as a stronger, more effective users' organization. They have clarified the organization's objective, enhanced the benefits available to its members, and positioned ACCT to help the IMHEX team understand its customers' needs through an ongoing collaborative structure. Together, ACCT companies and the IMHEX team are now much better poised to cooperate in making the commercialization process a true success.

5.3 Status of Santa Clara MCFC Product Development Test

CONTRACT INFORMATION

Cooperative Agreement Number	DE-FC21-92MC29237
Contractor	Fuel Cell Engineering Corporation 3 Great Pasture Road Danbury, CT 06813 (203) 790-6922
Contractor Project Manager	Anthony J. Leo
Principal Investigators	Anthony J. Leo (FCE) Ashok K. Kush (FCE) Thomas P. O'Shea (Santa Clara Demonstration Project)
METC Project Manager	Thomas J. George
Period of Performance	October 1, 1992 to December 31, 1996

Schedule and Milestones



OBJECTIVES

The objective of the 2MW Santa Clara Demonstration Project is the demonstration of the carbonate fuel cell technology at full scale. Additional objectives of the project include the demonstration of specific advantages of the carbonate fuel cell power plant, such as high efficiency, low emissions, reactive power capability, and high reliability and availability. The project will also provide a design basis for pre-commercial early production power plants.

BACKGROUND INFORMATION

The project is an outgrowth of the ongoing carbonate technology development work and commercialization efforts at Energy Research Corporation (ERC, parent company to Fuel Cell Engineering Corporation). Carbonate technology development activities at ERC have progressed to the point where 125kW stacks have been tested in ERC's integrated test facility, and subscale stacks have been tested in utility and industrial sites around the world. ERC is working with the Fuel Cell Commercialization Group (FCCG) in a collaboration to commercialize the carbonate fuel cell technology in the late 1990's. A commercialization path has been identified which includes the demonstration of the technology in full size power plants prior to the production of pre-commercial units. The Santa Clara Plant will be the first of these demonstrations, and the project's utility participants have been drawn from the FCCG membership. The participants in the Santa Clara Demonstration Project (SCDP) are as follows: City of Santa Clara, City of Los Angeles Department of Water and Power, City of Vernon Light and Power Department, Electric Power Research Institute, National Rural Electric Cooperative Association, Sacramento Municipal Utility District, and Southern California Edison Company.

A critical step in ERC's commercialization effort was the formation of two subsidiaries in 1990: The Fuel Cell Manufacturing Corporation (FCMC) and the Fuel Cell Engineering Corporation (FCE). FCMC will manufacture carbonate stacks and multi-stack modules, initially from it's pilot manufacturing facility in Torrington, CT and eventually from a commercial manufacturing facility. FCE is responsible for power plant design, integration of all subsystems, sales/marketing, and client services.

FCE began work on the Santa Clara demonstration power plant design in 1990, and submitted a preliminary design to the SCDP in August, 1991. A series of SCDP and EPRI supported studies were conducted to optimize the design over the next year, and Final Engineering began in September, 1992. The design of the multi-stack modules used in the DC power section of the plant was begun earlier under EPRI and ERC support.

The DOE supported portion of the project began in October, 1992 and included the development of environmental information and the development of the final design of the Santa Clara multi-stack module. DOE support is now focused on stack and multi-stack module manufacturing. DOE funding is through the Morgantown Energy Technology Center.

Funding for the design and supply of the BOP and the overall plant construction, testing, and operation is being supplied by SCDP and FCE. Additional funding for some of the BOP pre-testing activities is being provided by the California Energy Commission (CEC).

PROJECT DESCRIPTION

The 2MW plant is the first application of a commercial-scale carbonate fuel cell power plant on a U.S. electric utility system. The site for the plant is at 1255 Space Park Drive in the City of Santa Clara. The site is owned by the City's Electric Department and is immediately adjacent to the Scott Receiving Station, a 115/60kV switching station on the City electrical system. The demonstration plant will occupy a portion of the site, measuring approximately 150 by 150 feet.

The power plant will use 16 fuel cell stacks, each rated at 125 kW. The stacks will be delivered to the site in truck-shippable 4-stack submodules, which will be installed into the two 1MW modules to comprise the DC section of the plant.

The design specifications for the power plant are summarized in Table 1. The plant will be tested for a total of 10,000 hours, consisting of a 1000 hour acceptance test and a 9000 hour endurance test phase.

Table 1. Design and Performance Criteria

Nominal Capacity	2 MW
Plant Rating, net AC	1.8 MW
Heat Rate (LHV) at Rated Power	6850 Btu/kWh
Max Emissions at Plant Rating SO_x NO_x CO_2	 0.003 lb/MWh 0.0004 lb/MWh 845 lb/MWh
Noise	60 dB(A) at 100 ft
Availability	90%
Startup Time, Cold Start to Rated Power	40 hours
Ramp Time, Standby to Rated Power	30 minutes
Reactive Power	+/- 1.67 MVAR

The project is currently in the detailed engineering phase, with engineering activities focused on procurement and construction support. The following sections describe the results of the power plant BOP and Stack Module design work, and discuss the status of power plant procurement and construction.

RESULTS

Balance of Plant Design and Procurement

The power plant process is ERC's simplified design, which had been developed in earlier system optimization studies. Process design in the current program has been focused on developing the detailed process design for the specific SCDP plant, and developing complete equipment specifications to the point where procurement could proceed.

A simplified process flow diagram is shown in Figure 1. Natural gas is preheated using the system exhaust gases and treated to remove odorants and impurities before being mixed with steam for use in the fuel cells. The fuel is internally reformed to hydrogen in the fuel cell stacks, and 75% of the hydrogen is consumed in the power generation reactions of the fuel cell anodes, producing water and CO_2. The anode exit stream consists of this steam and CO_2, carbon monoxide from the reforming and shift reactions, plus the 25% residual hydrogen. The exit stream is sent to a catalytic oxidizer, H-101, where the hydrogen and carbon monoxide are reacted with excess air.

This quenched flue gas stream contains the oxygen and CO_2 required by the fuel cell cathodes, and it is sent back to the fuel cells as the cathode feed stream. The quench air used in H-101 is adjusted to provide the desired temperature for use in the fuel cell cathodes. After the cathode reactions consume some of the oxygen and CO_2, the cathode exit gases are sent to the heat recovery system. The exit gases provide heat to the fuel treatment system, the fuel/steam heater (E-101), water boiler/superheater (E-102) and the fuel preheater. These heat exchangers are configured into a packaged heat recovery unit, which also contains the start-up burner (H-102), the steam generator deaerator, and the system exhaust stack.

Other subsystems in the plant include the water treatment system, instrument air compressor, and backup power generator. The layout of the plant is shown in Figure 2. The plot plan shows the location of the major equipment shown in the process flow diagram, plus additional facilities such as the control room, visitors center, etc.

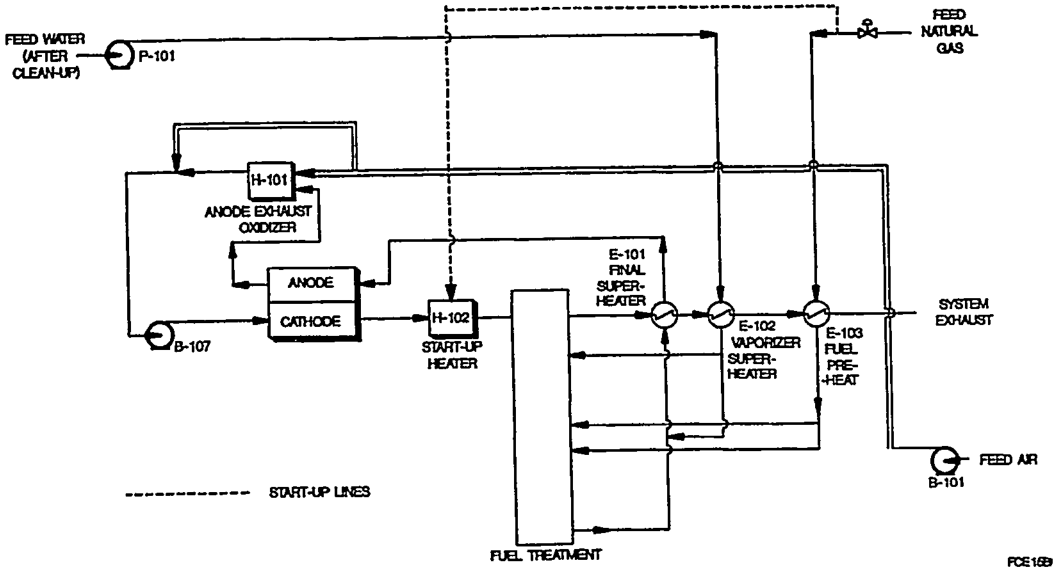


Figure 1. Simplified Process Flow Diagram for 2MW SCDP Power Plant

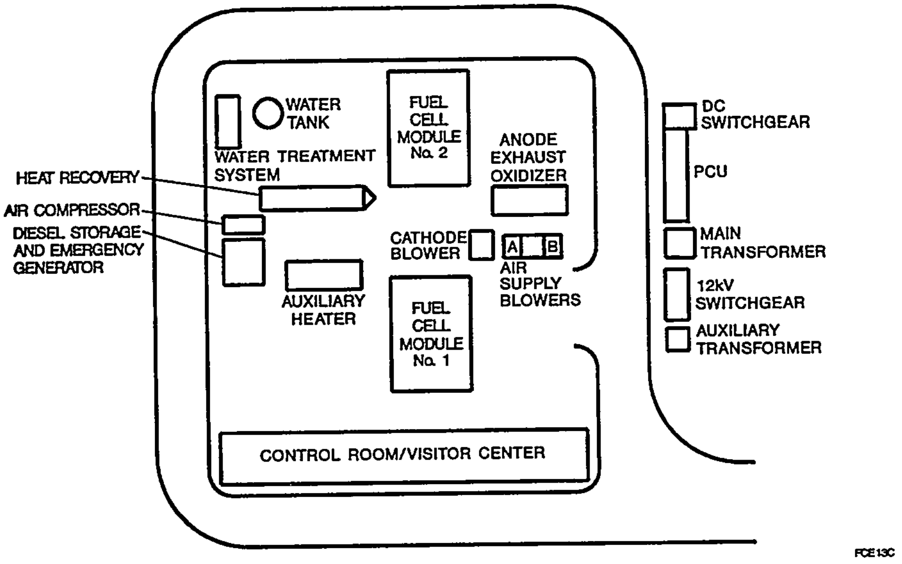


Figure 2. 2MW SCDP Power Plant Layout

The process and mechanical design of the plant is now essentially complete, with remaining engineering activities focused on procurement and construction support. All major BOP equipment items are on order. Equipment has begun to arrive at the site, and most equipment should be received by the end of September of this year.

Multi-Stack Module Design and Fabrication

The DC power section of the plant will consist of two 1MW Stack Modules. Each of these Modules will contain two Submodules, which are the truck shippable units which enclose the fuel cell stacks. Each Submodule will consist of four stacks and associated piping, instrumentation, and electrical components in a sealed enclosure.

The structural enclosure for the stack Submodules will be shop fabricated and shipped to the FCMC facility in Torrington, CT. There, the fuel cell stacks will be installed in the enclosure and all piping, electrical, and instrumentation work within the enclosure will be completed. All piping connections will terminate at the enclosure sides with weld connections which will be sealed during shipment to the site. All electrical and instrument connections are brought to terminal boxes on the outside of the enclosure. This design approach provides for installation of the submodules at the Santa Clara site with a minimum of field labor. The four submodules will be truck shipped to the site, where two units will be installed in each of the two 1MW modules.

An illustration of the Module arrangement is shown in Figure 3, and Figure 4 shows a rendering of the completed plant with the Modules installed. Each Module will consist of two Submodules with associated gas distribution piping, instrumentation connections, and electrical connections.

As with the BOP, the Stack Module design is essentially complete, with ongoing engineering work supporting procurement activities. The stacks are being fabricated at FCMC, and the Submodule enclosures, piping and instrumentation components are on order.

Regulatory and Permit Status

All necessary permits for the project have been received. At the beginning of the DOE supported portion of the project, FCE submitted environmental information to DOE to support the submittal of an Environmental Assessment for the National Environmental Policy Act (NEPA) approval. Prior to this, the Bay Area Air Quality Management District had granted the project an exemption from Air Quality permitting requirements. Also, under the California Environmental Quality Act (CEQA) a Negative Declaration has been issued, indicating that no significant environmental impact is anticipated, and a Conditional Use Permit has been issued. Local construction, water discharge, and fire department permits have been obtained as necessary during the plant design and construction phases.

Plant Construction Status

Balance of plant construction began in April, 1994, and is scheduled to be complete by January, 1995. The site civil work is almost complete, and the construction focus is shifting to the receipt and installation of mechanical equipment and piping.

FUTURE WORK

Upon completion of BOP construction, the plant start-up phase will begin. This includes BOP testing, stack installation, and power plant initial start-up.

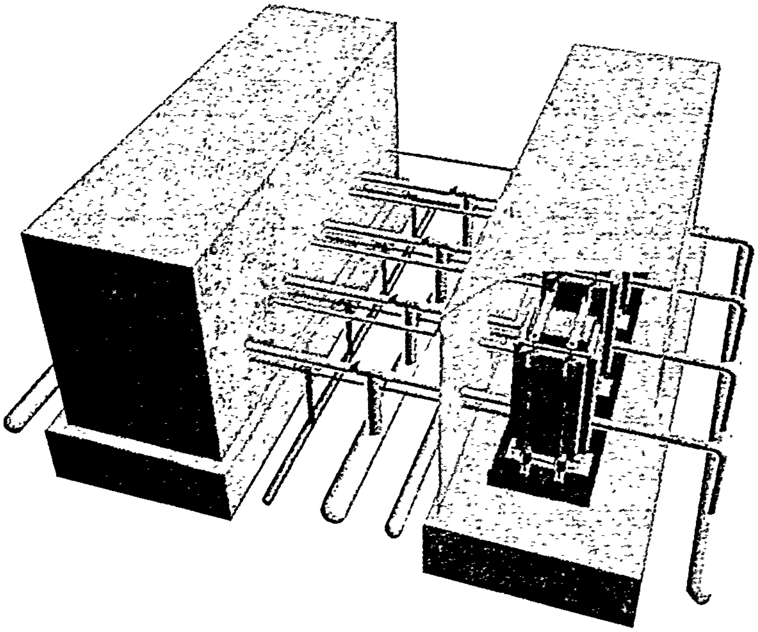


Figure 3. Configuration of 1MW Fuel Cell Stack Module

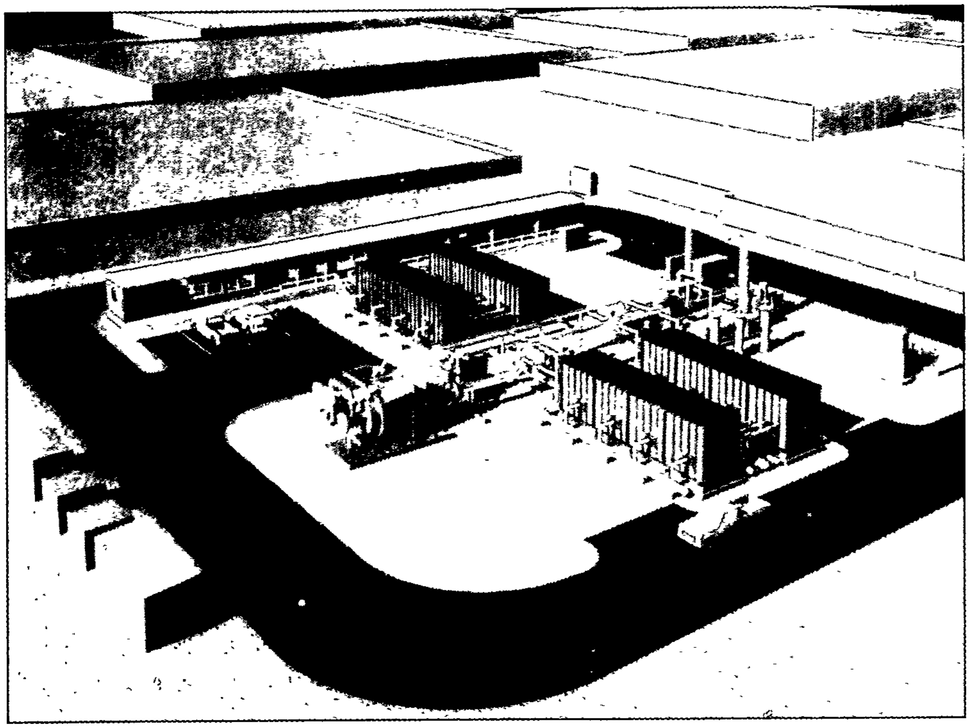


Figure 4. Rendering of 2MW SCDP Power Plant

The 1000 hour acceptance test will then be conducted, followed by the 9000 hour endurance test phase. The acceptance test phase will measure the plant performance against the design specifications for heat rate, power quality, emissions, noise level, etc. During the endurance phase the plant performance and operational characteristics will be monitored, and the integrity and wear of plant components and subsystems will be assessed. Training of staff from the participating utilities in the operation and maintenance of the plant is also planned in this phase of the project in order to prepare them for eventual introduction of fuel cells into their generating systems.

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Serfass, J.A., Glenn, D.R., and Claussen, R.W.; Preparing for Commercialization: The Collaboration Matures. In *1992 Fuel Cell Seminar Program and Abstracts*, p. 183. Nov/Dec 1992.

5.4

Status of 250 kW Product Development Test at San Diego Gas & Electric

R. A. Figueroa, Program Manager, R&D
San Diego Gas & Electric

J.A. Scroppo, Program Manager
M-C Power Corporation

INTRODUCTION

San Diego Gas & Electric (SDG&E) is participating in the product development test (PDT) program with the Department of Energy (DOE) and M-C Power, Incorporated, in cost sharing the development of M-C Power's internally manifolded heat exchanger (IMHEX) carbonate fuel cells. The results of this work will be a test of a 250 kW net power plant to be installed and operated by SDG&E. SDG&E's responsibilities include host site selection, development and plant construction and operation.

The primary purpose of this paper to present an update of the status of the host site for the PDT 250 kW net power plant and progress of manufacturing and supply of key plant components.

BACKGROUND

The initial host site for the PDT test was the Kaiser Permanente Medical Center in San Diego, California. By the end of 1993 several milestones had been completed including: preliminary P&IDs, preliminary plant layout, exemption from the San Diego Air Pollution District (APCD), and site engineering was 50 percent complete. The site engineering provided additional information on a number of existing mechanical, electrical, and signal underground conduits and soil conditions which were the basis for a preliminary site development cost study.

Kaiser Engineering and Turner Engineering were selected to perform the site development cost estimate. The bids from these contractors were within ten percent from each other and their cost estimates were in excess of $800,000.

The estimate included retaining walls, foundation pads for the fuel cell equipment, partial demolition of an existing structure, relocation of an existing oxygen storage facility, and relocation of underground conduits. In addition, there were strict requirements for the plant enclosure to comply with Kaiser's architectural environment.

This cost estimate was much greater than the anticipated site development costs of $250,000 based on engineering judgment, construction experience of large utility type equipment and costs incurred at the UNOCAL site for a similar size plant.

At this time, SDG&E initiated parallel efforts to identify other potential sites within the medical center and contacted alternate potential hosts, the U.S. Navy and VA Hospital, in the event that the site cost issues were not resolved with Kaiser.

The Navy and VA hospital had been approached during the initial host site selection and both had indicated interest, but at that time there were constraints which precluded them from being selected and Kaiser was the selected as the primary host site. This time however, the Navy indicated strong interest in being selected as the host site and we initiated the process for identifying available potential sites that would be compatible with the work already accomplished.

After reviewing the alternate locations at the medical center it was determined that the site development costs would not be significantly reduced by shifting the site within Kaiser. An alternate approach was for Kaiser to cost-share the site development by funding about $500,000. Kaiser management however, was unable to make this commitment and regrettably agreed to forego being the host to the PDT test.

NEW HOST SITE

Our focus shifted to a hand full of sites at several Navy facilities. The selected candidate site is within the Naval Air Station (NAS) at Miramar, located about 12 miles northeast from San Diego civic center. NAS Miramar has an existing steam generating plant which provides steam in the range of 16,000 pounds per hour, with two of four boilers, to meet the needs of facilities within the base.

The electric and steam outputs from the PDT test will supplement a small portion of both the electric and steam needs of the NAS Miramar base.

The host site is located near the intersection of Mitscher Way and Miramar Way which is about a half mile west of highway 15 at the Miramar Way off ramp. The site is about 40 ft x 115 ft in area and is located behind a telephone switch building, about 100 yards north of the steam plant, and nearly 100 feet from all utility interconnection points.

The site approval process from the Navy involves several command centers. We've met with Commanding Officers from the various command centers and all have endorsed the project. We're in the final site approval stage which involves a modification of SDG&E's contract with the Navy. This modification is required in order to obtain the only permit necessary from the NAS Miramar Commanding Officer to proceed with construction.

HOST SITE STATUS

Most of the work completed for the Kaiser site will be applicable to the Navy site. Previously, Kaiser had the responsibility for site engineering and design. For the navy site, SDG&E will take this responsibility including site and plant construction.

To-date, we've completed a hazardous material and safety code assessment and soil analysis for the site. Bechtel has modified the plant layout. With this information we've completed a preliminary site engineering and design effort which includes foundations, buildings, and security fencing.

We filed and have received from the San Diego APCD an exemption for environmental permits for the PDT plant. We have also filed for a categorical exclusion from NEPA.

Utilities available at the site within 100 feet include city water, natural gas, main steam supply, condensate return, 12 KV substation (4160 V), and sewer drain.

PLANT COMPONENT STATUS

All of the major component specifications and bids completed for the Kaiser site are being reviewed and more than likely they will be used without changes to place orders. Stack components are being manufactured. Approximately 15 percent of the components are complete. All repeat components have been ordered.

As a result of the work performed at the UNOCAL project we've been able to make improvements on materials and components for the PDT plant. One such change is the stack pressure vessel. The vessel for the PDT plant will be smaller and lighter.

CONCLUSIONS

In conclusion, there was some site rework necessary, but most of these changes are completed. The process modifications have been minimal to accommodate the new site characteristics. The overall schedule impact on the PDT project has been about four to six months delay with minimal budget impact.

5.5 Status of 120-kW Direct Fuel Cell Product Improvement and Testing

CONTRACT INFORMATION

Contract Number:	DE-AC21-90MC27168
Contractor:	Energy Research Corporation 3 Great Pasture Road Danbury, CT 06813 (203) 792-1460
Contract Project Manager:	Mohammad Farooque
Principal Investigators:	R. Bernard, J. Doyon, L. Paetsch, P. Patel, A. Skok and C. Yuh
METC Project Manager:	Thomas T. George
Period of Performance:	September 1990 to October 1994

1. SCHEDULE AND MILESTONES

A simplified schedule and the major milestones of the cost-shared program are shown in Figure 1. The program will be completed in October, 1994. All major milestones of the program have been met.

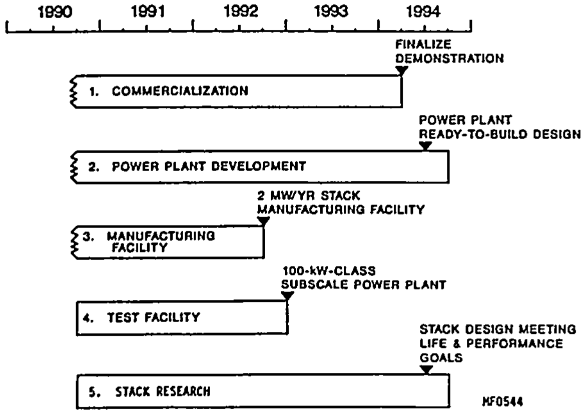

Figure 1. Overall Schedule and Major Milestones:
All Major Milestones of this Program have been Attained

2. OBJECTIVES

ERC's carbonate fuel cell goals are commercialization of MW-Class natural gas/logistic fuel units in the near-term and 100MW-Class coal gas/natural gas dual-fuel units long-term. As a major milestone towards achieving this objective, ERC's subsidiary Fuel Cell Engineering Corp. is constructing a 2MW natural gas power plant at the City of Santa Clara, CA for demonstration in the 1995-1996 time frame. The on-going program effort has prepared this efficient energy conversion technology for the demonstration.

3. BACKGROUND

The carbonate fuel cell promises highly efficient, cost-effective, environmentally superior power generators for pipeline natural gas, coal-gas fuels, biogas, and other hydrocarbon fuels. ERC has been engaged in the development of this unique technology from the late 1970's, primarily focusing on the development of the Direct Fuel

Cell (DFC) technology, pioneered by ERC[1-4] since the early 1980's. The DFC design incorporates the unique internal reforming feature which allows utilization of a hydrocarbon fuel directly in the fuel cell without requiring any external reactor and associated heat exchange equipment. This permits direct operation on pipeline natural gas and a variety of coal-derived fuels. The DFC provides upgrading of waste heat to chemical energy; therefore, contributes to higher overall efficiency for energy conversion.

ERC plans to offer commercial DFC power plants in various sizes, initially focusing on the MW-scale units. ERC plans to offer standardized, packaged MW-scale DFC power plants operating on natural gas or other hydrocarbon-containing fuels for commercial sale in the later part of this decade. After gaining experience from the early MW-scale power plants and maturing of the technology, ERC expects to introduce larger power plants operating on natural gas and/or coal-gas in the beginning of the 21st century.

A major step toward commercialization is the demonstration of a MW-size power plant. ERC is near the end of a DOE-sponsored private-sector cost-shared multi-year program for preparing the technology for this demonstration. Design of the power plant has been finalized and construction initiated. Stack manufacturing and performance have been validated; stack construction initiated.

4. PROJECT DESCRIPTION

ERC is completing this multi-year DOE program (private-sector cost shared) to prepare the carbonate fuel cell power plant for full-scale demonstration in 1995-96 and commercialization thereafter. Planned accomplishments of this program have been attained, achieving progress in all fronts for commercialization efforts. Area-wise status of ERC's commercial product development to date is summarized below.

- **Stack Design and Experience**

  - Stack performance consistent with electrical efficiency for demonstration power plant design attained at the power plant operation condition (>760 mV/cell at 133 mA/cm^2).

  - Over 25 short stacks, three 20kW-Class stacks and four 100kW-Class stacks built and tested (over 80,000 hours of cumulative testing). The 100kW-Class stacks tested at design pressure and utilizations in an integrated power plant system (CO_2 recycled from anode to cathode side).

  - Stacks endurance tested for up to 10,000h; post-test results project stack life >5 years.

- **BOP Design and Experience**

  - Modularized power plant design incorporated in Santa Clara Demonstration Project (SCDP).

  - BOP development and testing in progress for the past 4 years. This includes over 10,000h of operation providing design information and hands-on operator training, low-Btu converter for CO_2 recycle, operation on a variety of pipeline natural gas fuels, and remote monitoring of testing at customer sites.

  - Over 200,000kWh power generated, 35,000kWh power generated in grid-connected operation.

  - 50% efficiency in small-scale (125kW) complete power plant mode, highest ever shown in any single cycle power generating equipment.

- Extremely low NO_x and SO_x, and low noise emissions demonstrated in subscale power plant tests.

- **Manufacturing Capability/Experience**

  - Low cost commercial stacks projected for a variety of market scenarios (independently verified by a DOE/EPRI-appointed auditor).

  - Semi-continuous manufacturing facility operational for the past two years. 10MW/yr capacity on a multishift basis (100% privately financed).

- **Transportation Experience**

  - Two stacks transported by road to California; one stack transported by road to Louisiana; six stacks transported by air/road to Europe; three tall stacks transported within Connecticut (from manufacturing facility to test site).

- **Multifuel Capability**

  - Operated stacks on slip streams of entrained and fluid bed gasifiers; operation on methanol, ethanol, and diesel-like fuel demonstrated; 30kW stack test planned on diesel fuel.

- **Demonstrations**

At User Sites

- First demonstration of a 2MW power plant at the City of Santa Clara site (construction in progress).

- Two 7kW integrated system natural gas demonstrations at Elkraft Power Company, Denmark, 1990, 1994, grid-connected operation.

- 20kW integrated system demonstration on natural gas at PG&E, 1991, grid-connected operation.

- 70kW integrated system demonstrated at PG&E, 1992, grid-connected operation.

- 20kW stack operated on coal gas from an entrained Destec Gasifier, 1993-94 (4000h on-line).

- Lab-scale stacks operated 11,000 hours on simulated Rheinbraun coal-gas[6], including major contaminants, 1993-94 (Germany); two labscale stacks on test on fluid bed gasifier slip stream.

At ERC

- Demonstrations of 125kW subscale power plant for upto 1,800 hours and two planned thermal cycles representing many power plant features incorporated in the 2MW SCDP.

- **Commercialization Initiative**

- Fuel Cell Commercialization Group (FCCG), an organization of potential utility buyers, is actively involved with the commercialization of ERC's 2MW fuel cell power plants.

- Worldwide commercialization network established, providing market access in Europe and Asia.

- **Commercialization Team**

- ERC responsible for coordination and financing.

- Established Fuel Cell Engineering (FCE) subsidiary for power plant development, marketing, and sales.

- Established Fuel Cell Manufacturing Corporation (FCMC) subsidiary for stack manufacturing.

- Continuous A&E collaboration with Fluor Daniel for the past 15 years.

- Collaboration with Jacobs Applied Technology, a leading packager, for commercial product modularization.

- Working with FCCG and its Design, System Planning, Information Transfer and Model Contract Committees.

- Initiated regional distribution alliances with interested organizations.

This paper highlights the progress in stack technology during the past year. Advances in commercialization, demonstration and coal tests are presented in papers presented separately in this conference.

5. RESULTS/ACCOMPLISHMENTS

Over the past year, significant progress has been achieved in defining the full-height stack design for the demonstration power plant, verification life of fuel cell components, and stack cost reduction. Progress in these areas are discussed next.

5.1 Full-Size Stack Design Validation

In the DFC power plant, the carbonate fuel cell stacks will be arranged in truck-transportable modules, where each module will contain a number of identical carbonate fuel cell stacks. The stack module for 2-MW demonstration will be 11.3m x 2.6m x 3.7m, containing four full-height stacks. The stacks will consist of 258 cells of approximately 6,000 cm^2 cell area, with a nominal rating of 125kW. A photograph of such a stack is shown in Figure 2. The recent efforts at ERC were dedicated to getting the full-height stack ready for field demonstration. The design has been finalized, manufacturability verified, and performance validated in integrated system tests; details of these accomplishments are discussed next.

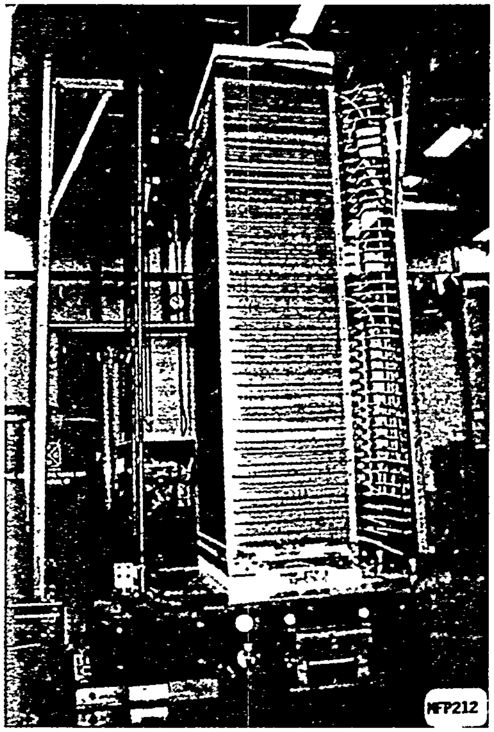

Figure 2. Photograph of the Full-Height DFC Stack:
258-Cell 6,000 cm^2 Cell Area Stacks Will be Used in the Demonstration Unit

ERC first assembled a tall stack simulator to investigate and resolve tall stack design issues and subsequently tested four tall stacks to verify/improve design and manufacturing. The issues which required significant development efforts include assembly fixture, manifold system, flow distributor, compression system, stack enclosure, internal reformer, and transportation design. The path followed for development of these key components is presented in Table 1. Stacks tests AF-100-1 and AF-100-3 were performed during 1992-1993 time period and AF-100-2 and AF-100-4 tests were conducted during the past year (1993-94).

Table 1. Tall Stack Design Issue and Verifications Road Map:
Stack AF-100-4 Verified the Demonstration Stack Design

ISSUE	DESIGN ITERATION/VERIFICATION				
	SIMULATOR	AF-100-1 (70kW)	AF-100-3 (125kW)	AF-100-2 (125kW)	**AF-100-4 (130kW)**
STACK ASSEMBLY	X	X	*		
LIGHTWEIGHT MANIFOLD SYSTEM	X	X	X	*	
FLOW DISTRIBUTOR	X	X		X	*
STACK COMPRESSION SYSTEM		X	X	*	
STACK ENCLOSURE	X		*		
INTERNAL REFORMER		X		X	*
TRANSPORTATION		X	*		
"X" Denotes Design Iteration; "*" Design Finalized					

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The objective of Stack AF-100-2 test was to establish the power plant tall stack baseline design. Stack AF-100-2 was tested during the fourth quarter of 1993 for 1800 hours which included: 870 hours at 125 mA/cm^2 operation, two thermal cycles and diagnostic testing (stack test lifegraph shown in Figure 3). The test produced ~134MWh electricity from the pipeline natural gas.

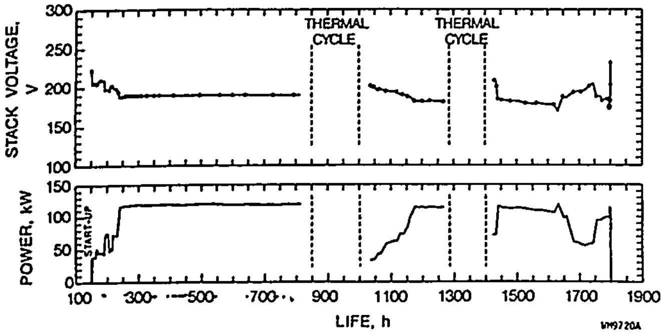


Figure 3. AF-100-2: Operating History:
1800-Hour Operation Included 870 Hours of Rated Operation and Two Planned Thermal Cycles

The stack characteristics were checked after each thermal cycle. The performance of the stack at different test stages are reported in Figure 4. Less than ~3% change in heat rate was observed during the testing period.

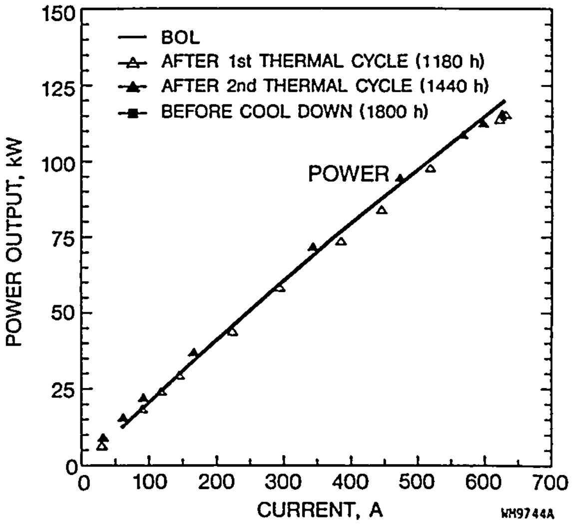


Figure 4. 125kW Stack Performance (Pipeline Natural Gas):
No Significant Performance Loss During Testing

The cell performance, as measured by the group voltages was very uniform (±1.5%), except for Group 2 which evaluated a non-standard cell design. This represents improvement in performance uniformity over ±2.5% for first two full-height stack tests and appears to have resulted from better fuel flow distribution and improved stack components tolerance achieved for Stack AF-100-2. Without a flow correction, about 5% flow variation in fuel flow from cell-to-cell is expected (Figure 5). The flow distributor design used in Stack AF-100-2 predicted flow variation to within 2%. The Stack AF-100-2 indicated a slight voltage variation along the vertical height, the trend of which was consistent with the predicted flow pattern. Subsequently, the distributor design was further refined for the 2MW demonstration power plant stack predicting a fuel flow variation of less than 1%. This improved design was incorporated in Stack AF-100-4.

Maintenance of constant stack compression pressure throughout the stack life is very important for performance as well as transportation considerations. Simplicity and compactness are also required for packaging and cost-effectiveness. A compact system providing only <0.2 kg/cm^2 compression pressure variation was verified in Stack AF-100-2 (Figure 6). The old design used in Stack AF-100-1 occupied too large a space and resulted in a large compression pressure variation.

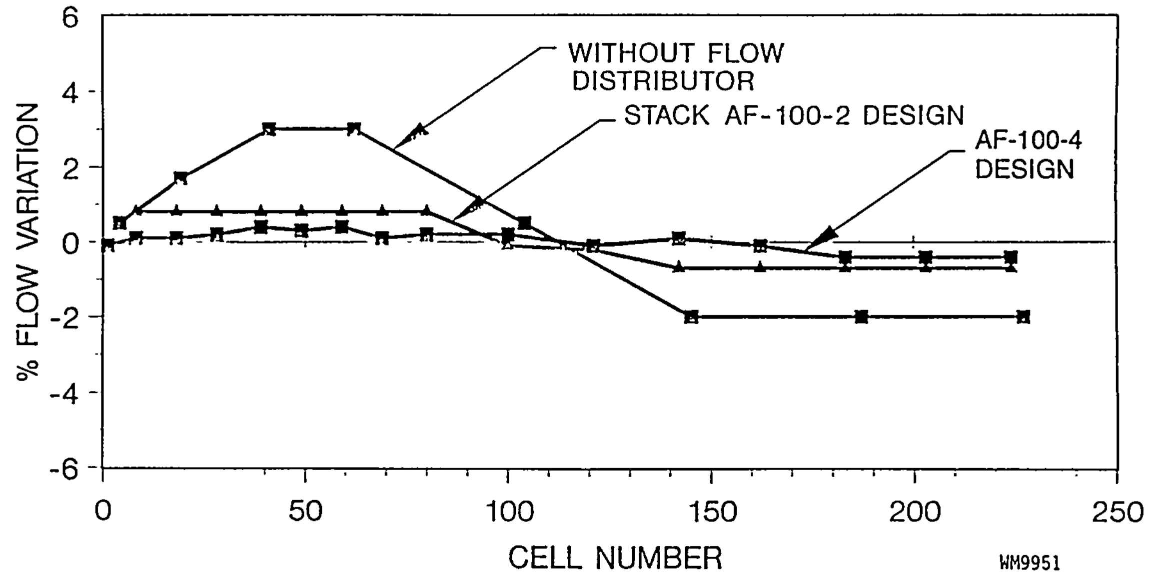


Figure 5. Improvement in Fuel Side Flow Distribution Using Flow Distributor: Less than 1% Fuel-Side Cell-to-Cell Flow Variation is Expected

The full-height stack design incorporates both the Indirect Internal Reformer (IIR) and Direct Internal Reformer (DIR) concepts. Stack AF-100-2 test showed that the Fuel-Turn/Oxidant-Out corner of the stack appeared to operate warmer than other parts of the stack, primarily because of local higher current density and minimal local cooling. This part of the stack was designed intentionally to operate warmest to maximize methane conversion in the internal reforming section.

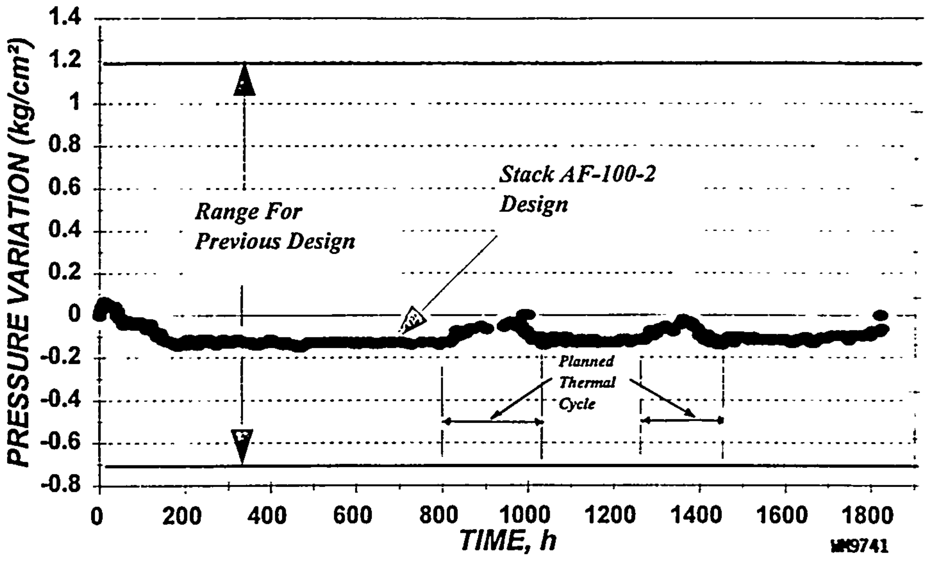


Figure 6. Full-Height Stack Compression Pressure Lifegraph:
Stack Compression Pressure within 0.2 kg/cm^2 was Maintained

The internal reformer design was modified to achieve uniform heat removal. This modified internal reformer design was verified in 2kW and 20kW stack tests and then incorporated in tall stack design.

The improved internal reformer and flow distributor designs were implemented in the latest tall Stack AF-100-4. This stack is identical to the demonstration stack design. Stack AF-100-4 has completed about 700 hours hours of testing to date, including one planned thermal cycle. The stack test lifegraph is shown in Figure 7; the test is continuing to gather additional operating data.

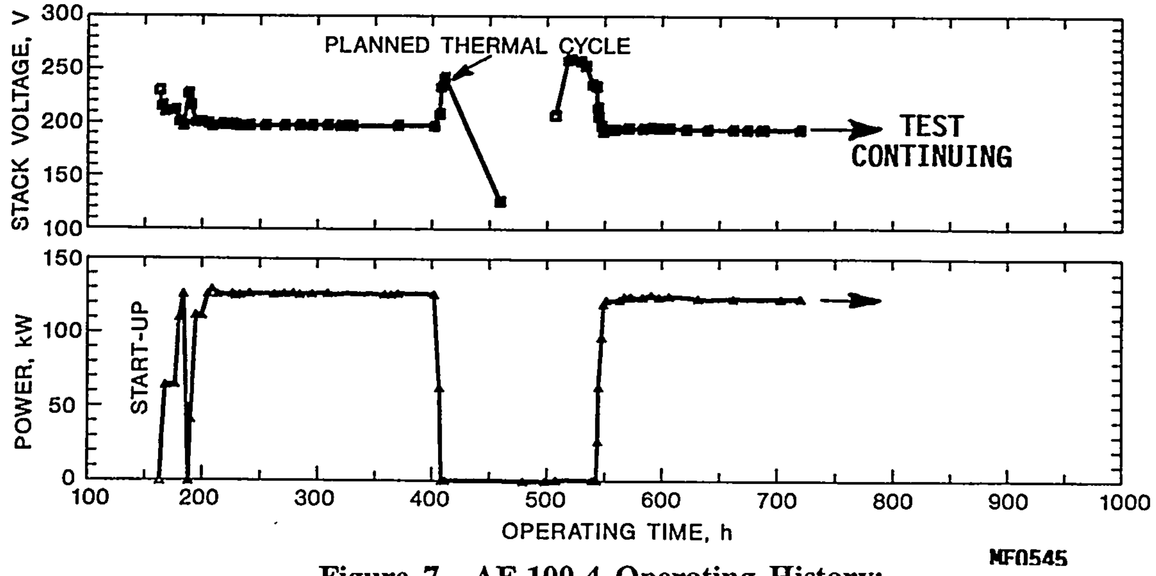
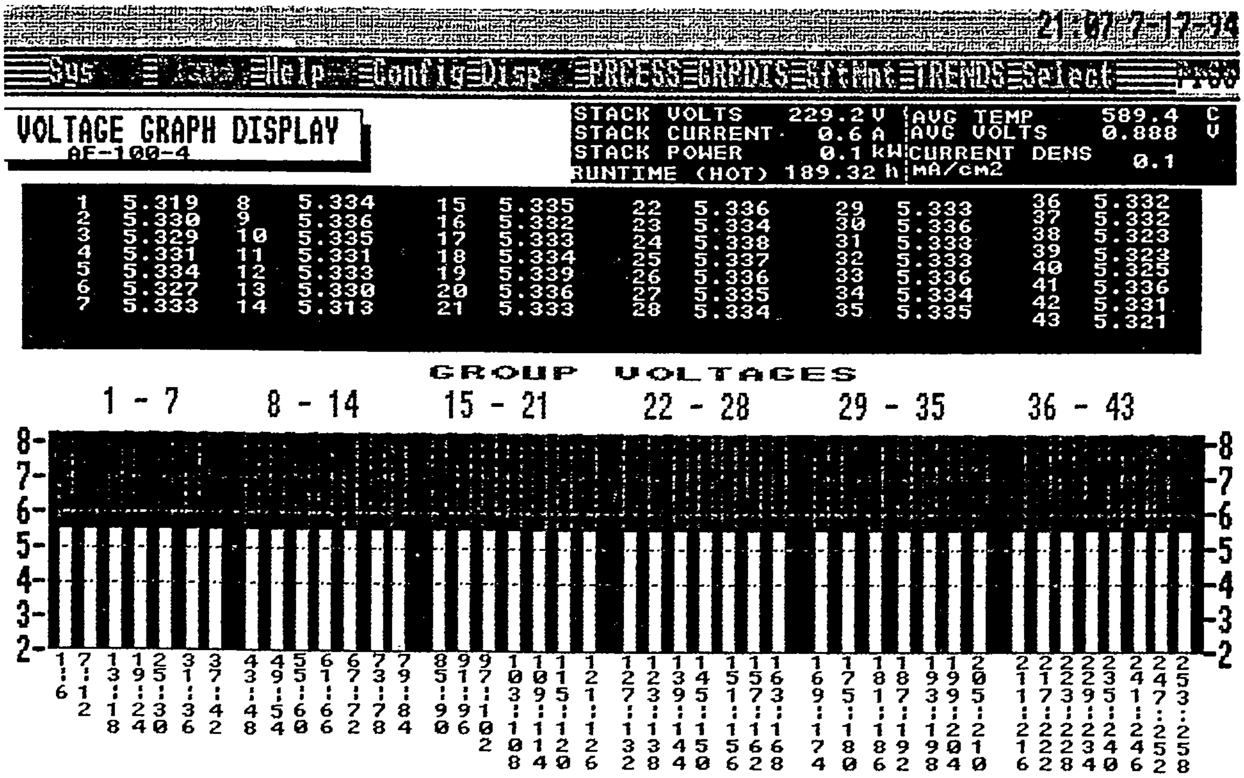


Figure 7. AF-100-4 Operating History:
Demonstration Stack Design Validated; Parametric Investigation will Continue for Design Data

The stack open circuit voltage distribution, in 6-cell groupwise shown in Figure 8, indicated only 0.2% variation. This demonstrates excellent manufacturing reproducibility. The stack has been operated with pipeline gas producing 130kW at ~74% fuel utilization. This stack represented a efficiency of 55% (LHV) from natural gas to electricity in the stack. The stack voltage variation at the 130kW power operation is shown in Figure 9. We believe this is the highest carbonate fuel cell power produced to date. Excellent performance distribution was observed.

The stack was thermally cycled and the stack performance as well as group-to-group group performance remained essentially unchanged. Thermal profile of the Stack AF-100-4, reported in Figure 10, shows the improvement achieved by the modified internal reformer design. The oxidant gas is used for heat removal from the stack. Oxidant exhaust temperature approached the warmest cell temperature to within 20°C, as opposed to 100°C observed in Stack AF-100-2 test. The NO_x and SO_x emissions from the test were measured. The emission levels were found to be negligible (Table 2). This represents a clean power production unmatched by any other fossil fuel based power conversion technology.



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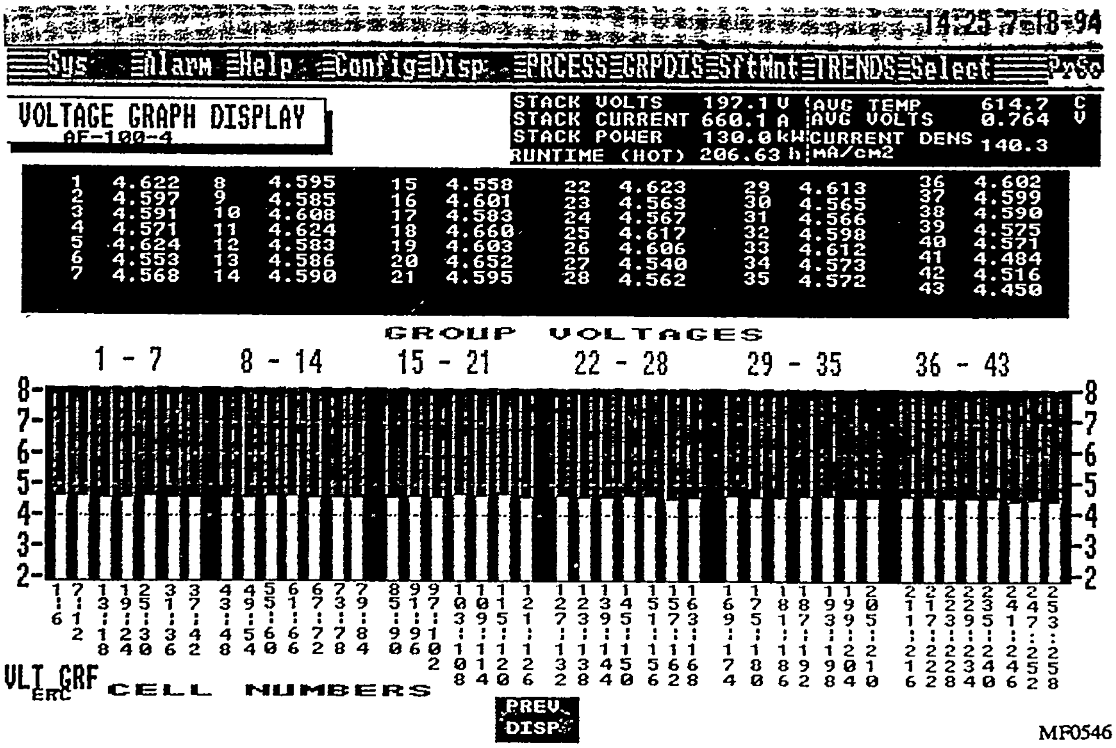
VOLTAGE GRAPH DISPLAY
AF-100-4

STACK VOLTS       229.2 V  AVG TEMP        589.4  C
STACK CURRENT       0.6 A  AVG VOLTS       0.888  V
STACK POWER         0.1 kW CURRENT DENS
RUNTIME (HOT)   189.32 h   mA/cm2            0.1

 1  5.319   8  5.334  15  5.335  22  5.336  29  5.333  36  5.332
 2  5.330   9  5.336  16  5.332  23  5.334  30  5.336  37  5.332
 3  5.329  10  5.335  17  5.333  24  5.338  31  5.333  38  5.323
 4  5.331  11  5.331  18  5.334  25  5.337  32  5.333  39  5.323
 5  5.334  12  5.333  19  5.339  26  5.336  33  5.336  40  5.325
 6  5.327  13  5.330  20  5.336  27  5.335  34  5.334  41  5.336
 7  5.333  14  5.313  21  5.333  28  5.334  35  5.335  42  5.331
                                                      43  5.321
```

Figure 8. Open Circuit Uniformity:
Excellent Manufacturing Reproducibility



```
14:25 7-18-94
Sys  Alarm Help  Config Disp  PRCESS GRPDIS SftMnt TRENDS Select  PxSc

VOLTAGE GRAPH DISPLAY      STACK VOLTS     197.1 V  AVG TEMP      614.7 C
    AF-100-4               STACK CURRENT   660.1 A  AVG VOLTS     0.764 V
                           STACK POWER     130.0 kW CURRENT DENS  140.3
                           RUNTIME (HOT)  206.63 h  mA/cm2

 1 4.622   8 4.595   15 4.558   22 4.623   29 4.613   36 4.602
 2 4.597   9 4.585   16 4.601   23 4.563   30 4.565   37 4.599
 3 4.591  10 4.608   17 4.583   24 4.567   31 4.566   38 4.590
 4 4.571  11 4.624   18 4.660   25 4.617   32 4.598   39 4.575
 5 4.624  12 4.583   19 4.603   26 4.606   33 4.612   40 4.571
 6 4.553  13 4.586   20 4.652   27 4.540   34 4.573   41 4.484
 7 4.568  14 4.590   21 4.595   28 4.562   35 4.572   42 4.516
                                                      43 4.450
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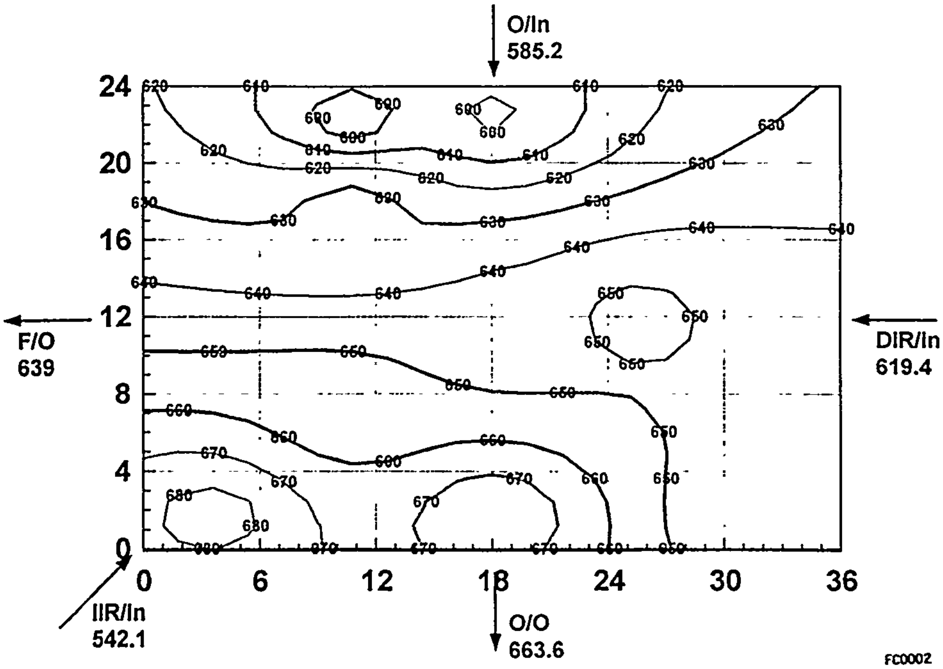
Figure 9. Performance at 130kW Operation (Natural Gas; 74% Fuel Utilization):
Excellent Performance Distribution



FC0002

Figure 10. Stack AF-100-4 Temperature Profile at 137 mA/cm^2 (Fitted from Experimental Data Points; All Temperatures are in °C):
Plate-to-Oxidant Approach Temperature of <20°C was Achieved

Table 2. Verification of Environmental Characteristics:
Low Levels of NO_x and SO_x and Noise Verified

System Test	NO_x, ppm	SO_x, ppm	Sound, db
120kW ERC Test	0.04	<0.01	~60* at 10 m

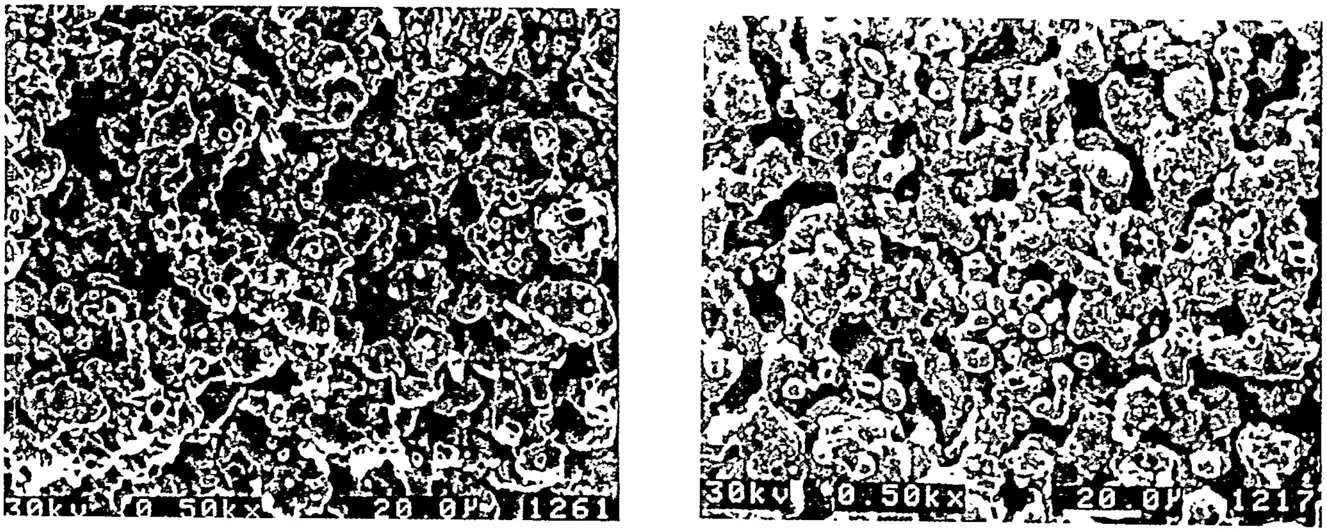
* excludes electrical equipment

The Stack AF-100-4 test is being conducted in an enclosed area. The sound noise level of the mechanical and flow system of the test was measured. The data showed that the noise level is low, comparable to average street level noise, and normal conversation could be carried out around the test area. Additional tests have shown that the noise drops to 60 db at 10 meters when the "open space" condition is achieved.

In summary, this test demonstrates that ERC's basic design meets its goals. The Stack AF-100-4 test validated the design for the demonstration project stacks. Additional tests will continue to gather design information for use in the next generation design.

5.2 Life Verification

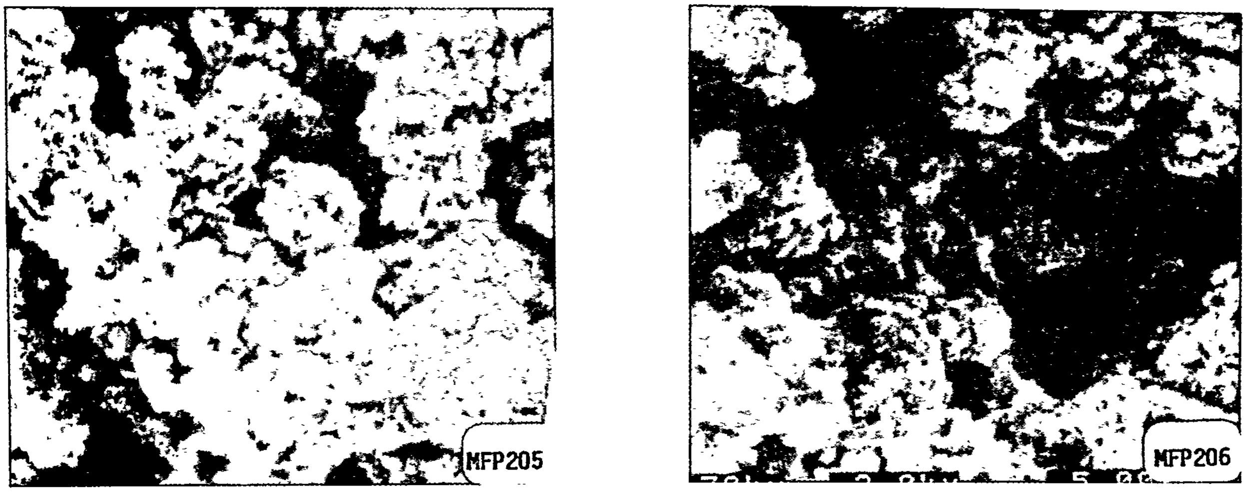
A useful life of 40,000 hours is desired for commercialization. ERC has accumulated more than 80,000h total stack testing time (50,000h in the current program); several stacks were tested for 5,000-10,000 hours. These tests provided important insight into the endurance behavior of cell components. The results are summarized in a recent publication[7]. Additional information available since then is reported here. Metallographic analysis of the anode and cathode tested for 10,000 hours showed little morphological changes (Figures 11 and 12). Very little anode creep was observed. The cathode surface area appears to stabilize at about 0.3-0.5 m^2/g after initial loss during first 2,000 hours. Therefore, the electrodes are expected to perform well for 40,000 hours.



AS SINTERED

AFTER 10,000h

Figure 11. Effect of Endurance on Anode Morphology:
No Significant Change in Anode Morphology after 10,000h Operation



LITHIATED NiO CATHODE (1,000h TEST)

AFTER 10,000h

Figure 12. Effect of Endurance on Cathode Morphology: No Significant Change in NiO Particle Size after 10,000h Operation

The aluminized coating for wet-seal protection has been shown to provide sufficient protection for the substrate stainless steels during a 20,000h testing (Figure 13). The X-ray microprobe analysis indicated sufficiently low aluminum depletion rate. Therefore, the aluminized coating is expected to meet the 40,000h life goal. These results project that the cell components will meet the five year life goal.

5.3 Stack Cost Reduction

ERC together with FCMC is continuously pursuing stack cost reduction activities and has accomplished significant progress during 1993-94. As an example, through a combination of design adjustment, manufacturing modification, and competitive vendor development, the anode cost has been reduced by at least an order of magnitude. This type of activity has also been extended to non-repeating components. The stack end plate weight and cost have also been reduced by a factor of 4. Manufacturing cost reduction has also been achieved by streamlining of the processes and increasing yields. The cost reduction efforts will be pursued with a higher priority in the future.

6. CONCLUSION

Full-height stack design has been validated in fully integrated natural gas system tests. The design is now ready for the full-size MW-class power plant demonstration.

7. FUTURE WORK

The plan for immediate future is Stack AF-100-4 additional testing and post-test analysis for use in next generation stack design.

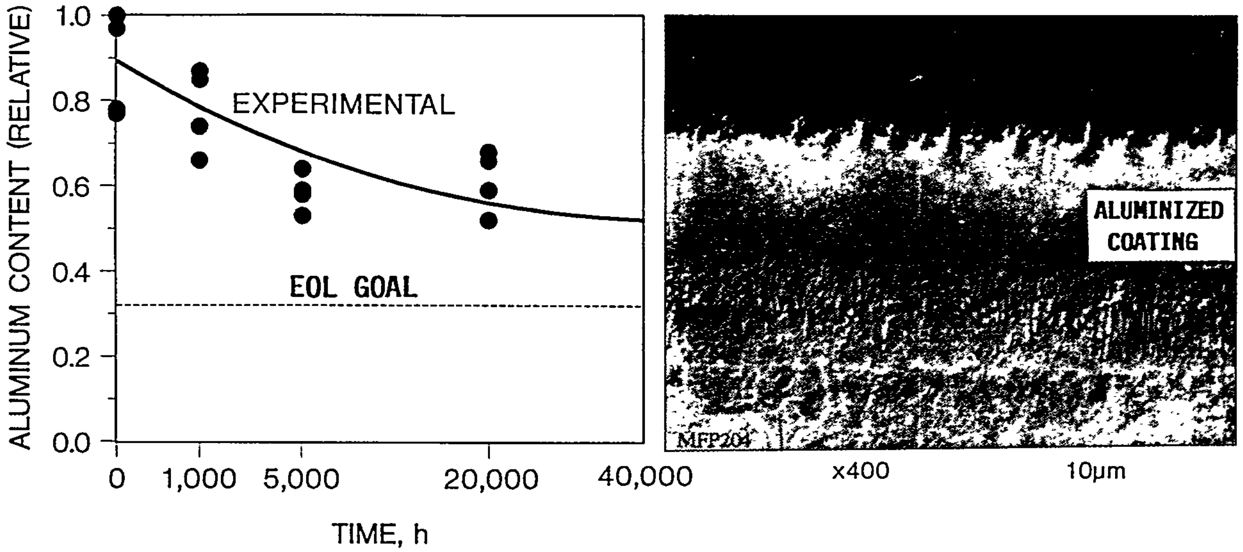


Figure 13. Effect of Endurance on Wet Seal Stability:
Wet Seal Coating Appears Stable after 20,000h

8. ACKNOWLEDGEMENT

ERC would like to acknowledge the support of DOE and EPRI for the stack and technology development and testing.

9. REFERENCES

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[5] M. Farooque, et al., "MCFC Power plant System Verification", Proc. 5th Annual Fuel Cells Contractors Review Meeting, p. 280, Morgantown, WV, August 1993.

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[7] C. Yuh, et al., "Carbonate Fuel Cell Endurance: Hardware Corrosion and Electrolyte Management Status," Proc. 3rd International Symp. Carbonate Fuel Cell Technology, p. 158, PV93-3, The Electrochemical Society, 1993.

5.6 Status of Unocal Product Development Testing

CONTRACT INFORMATION

Contract Number	**MCP-9-UNO**
Contractor	**UNOCAL** **376 S. Valencia Ave.** **Brea, California 92621** **(714) 528-7201**
Contractor Project Manager	**Joseph Scroppo**
METC Project Manager	**Mark Williams**

OBJECTIVE

Molten carbonate fuel cells have been under development for many years and on a small scale they can reliably and efficiently produce power. However, they are costly and to compete successfully with other electrical power production methods the cost must be reduced. The partners in this project are building a 250 Kilowatt molten carbonate fuel cell demonstration plant. The plant will show that the technology exists to build large scale molten carbonate fuel cells and operate them successfully. Information obtained from the plant construction and operation will also be used in the design and construction of future plants and will aid in reducing costs.

This progress report discusses construction of the demonstration plant at Unocal's Fred L. Hartley Research Center in Brea, California.

BACKGROUND INFORMATION

Unocal is a medium sized oil company based in Los Angeles, California. The company has extensive natural gas reserves and is interested in technology that will increase the value of those reserves. We joined the Molten Carbonate Fuel Cell Project team in 1990 along with our partners:

Bechtel
Department of Energy
Electric Power Research Institute
Institute of Gas Technology
M-C Power
South Coast Air Quality Management District
Southern California Edison
Stewart & Stevenson
The Gas Company

Unocal has a long history of involvement in innovative energy technologies. We are the world's largest producer of geothermal power, with plants at the Geysers in Northern California and the Philippines. We are also developing a new geothermal field in Indonesia. We have successfully developed technology for extracting steam from the hot saturated brines in California's Imperial Valley and operated some small commercial plants there. The Imperial Valley properties were recently sold to Magma Power Company

About ten years ago, Unocal developed a successful process for retorting oil shale and demonstrated it by operating a 10,000 barrel per day retort and upgrading plant in Colorado that produced a high quality paraffinic syncrude. The plant discontinued operations in 1990 and the technology for producing syncrude from oil shale must wait for higher crude oil prices before it is commercially viable.

We contributed a site for the completed 250 kilowatt molten carbonate fuel cell demonstration plant as well as funds and staff for on-site assembly and operations. The fuel cell site is located at Unocal's Fred L. Hartley Research Center in Brea, California. Brea is a small city in Southern California, about ten miles north of Disneyland.

The Research Center has 35 buildings occupying 77 acres of a 120 acre parcel and a staff of about 650 people. We have a 3.8 megawatt cogeneration plant providing steam and electrical power along with an ice bank and an absorption chiller for air conditioning. There is about 450,000 square feet of laboratory and office space on-site. A variety of research and development work is conducted on-site including pilot plant work for our refineries and analytical chemistry support for many Unocal operations.

The refinery pilot plant work is particularly noteworthy because we have been drawing on our experience in this area for the fuel cell project. Many aspects of molten carbonate fuel cell demonstration plant operation are similar to refinery pilot plant operation, particularly those dealing with safety considerations, control systems and handling hydrogen rich gases at high temperatures.

In preparation for fuel cell operations, our staff has been actively involved in the final design process with our partners, utilizing Hazops and other process safety management techniques to ensure safe and reliable operation of the plant. We have also been working closely with Bechtel and M-C Power on the site construction details. The fuel cell site is in the northeast corner of our facility, near our refinery process pilot plants. This site was chosen because it has excellent access to all plant utilities, including natural gas, electricity, steam, water, compressed air, and compressed nitrogen. It is also a large relatively open area with good access for heavy equipment which helps to keep construction costs down.

PROJECT DESCRIPTION

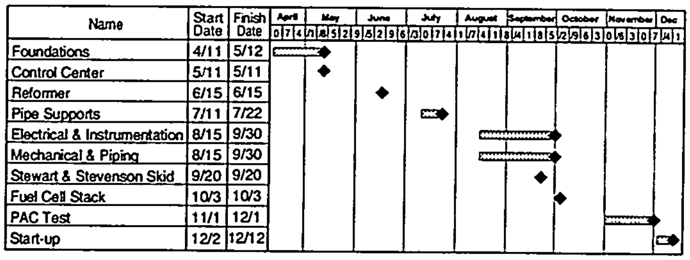
The principal equipment in the demonstration plant is the M-C. Power Molten Carbonate Fuel Cell Stack, a reformer to manufacture hydrogen from natural gas, a fuel treatment skid from Stewart & Stevenson, a control room with a distributed control system and a variety of power conversion equipment, and electrical switch gear. The site needed to contain the plant is about 100 feet by 150 feet. Brea, California is in an active seismic zone so a very robust plant design was required. Special attention had to be paid to equipment foundations and to soil conditions at the site.

Actual site preparation started in March. As we planned the construction program with our partners in late 1993 and early 1994, a question arose as to whether or not we should use a general contractor for the on-site construction work as is normal practice. We have had considerable experience with construction projects like this one and our experience has been that the process works well if the job is well defined and documented and the technology is well known. However, if there is much work that is undefined or if the project involves new technology that contractors have had little experience with, we have found the costs to be very high.

We proceeded with the bidding for the fuel cell construction project with prospective general contractors and we were not disappointed in our expectations. The bids were much higher than we thought they ought to be based on our knowledge of construction costs for pilot plants and other special purpose plants using new technology. With the results of the construction bidding in hand, we consulted with our partners and decided to use a different approach for the construction of the demonstration plant.

The solution to the problem of general contractors bidding high on a project that they don't fully understand,and to cover unknown risks, is to not use a general contractor. The drawback to this approach is that someone must coordinate the construction effort and handle all the details a general contractor normally would handle. Fortunately we had some experience with this type of project management and had experienced staff who could do the work. Unocal agreed to act as the general contractor and to subcontract easily identifiable parts of the project to vendors working and bidding in their area of specialty.

The results of this approach were very good. On the civil work for the site, the best general contract bid was $132,000. When we bid the same work as a complete package to subcontractors, the best bid was $85,000, a savings of almost $50,000. The second portion of the job was for structural steel work and a good comparison is not possible because the scope of the work changed from the time of the original bid to the time that the subcontract was let. We strongly suspect though that by the time we got done writing change orders on this item for a general contractor the cost would be much higher than the subcontractor bid. Bids are not available for the electrical, piping/mechanical, insulation and painting, but we expect good low cost performance from subcontractors in these areas, also.



Name	Start Date	Finish Date
Foundations	4/11	5/12
Control Center	5/11	5/11
Reformer	6/15	6/15
Pipe Supports	7/11	7/22
Electrical & Instrumentation	8/15	9/30
Mechanical & Piping	8/15	9/30
Stewart & Stevenson Skid	9/20	9/20
Fuel Cell Stack	10/3	10/3
PAC Test	11/1	12/1
Start-up	12/2	12/12

M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California

Construction Plan
5/27/94

Figure 1.

Site work is progressing according to the plan shown in Figure 1, and as of mid August, the foundations have been poured, the control center is on-site, the reformer is in place and the pipe supports have been mounted on their foundations. Electrical & instrumentation and piping and mechanical work is under way. Scheduled completion of construction is November, after the arrival of the fuel cell stack. Delivery dates of the major pieces of equipment are as follows:

<u>Major Equipment on Site</u>	<u>Delivery Date</u>
Control Room Building	May 11, 1994
Inverter	May 17,1994
Electrical Switch Gear	May 17, 1994
Bailey Control System	June 6, 1994
KTI Reformer	June 15, 1994
Misc. Piping and Hardware	June 20, 1994

Major Equipment Awaiting Delivery	Delivery Date
Stewart & Stevenson Fuel Processing Skid	September 20, 1994
Molten Carbonate Fuel Cell Stack	October 3, 1994

RESULTS

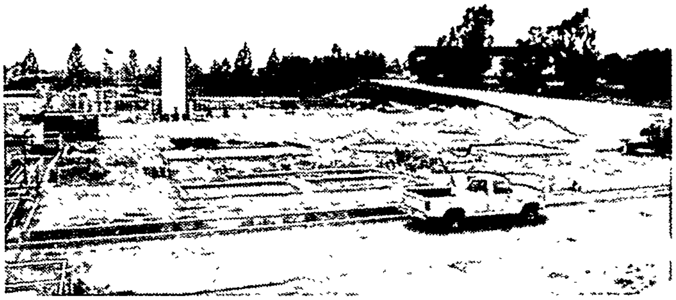
As we have progressed with the construction of the plant, we have documented each stage with photographs. Five photographs taken at various stages of construction follow

M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California

Construction Phase
4/4/94

Figure 2.

Figure 2 shows initial construction and civil engineering work. The site is bordered on two sides by utility trenches. A large liquid nitrogen tank can be seen in the background.



M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California

Construction Phase
5/10/94

Figure 3.

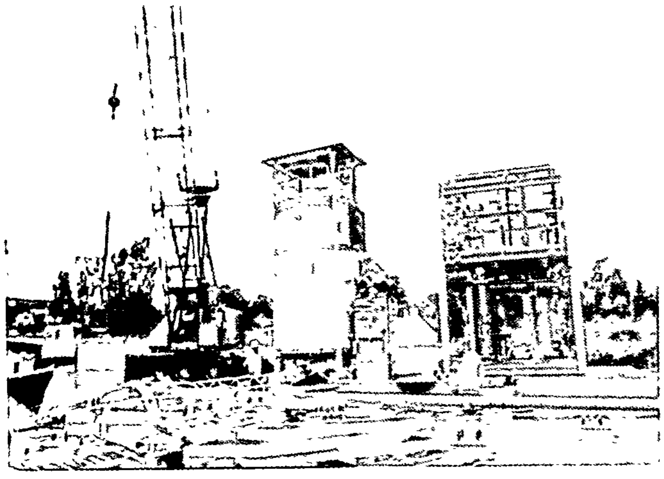
Figure 3 documents the foundations for the major pieces of equipment. The large concrete pad with the circle on top of it is the foundation for the fuel cell stack. The foundation for the control room is in the foreground.

M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California

Construction Phase
5/11/94

Figure 4.

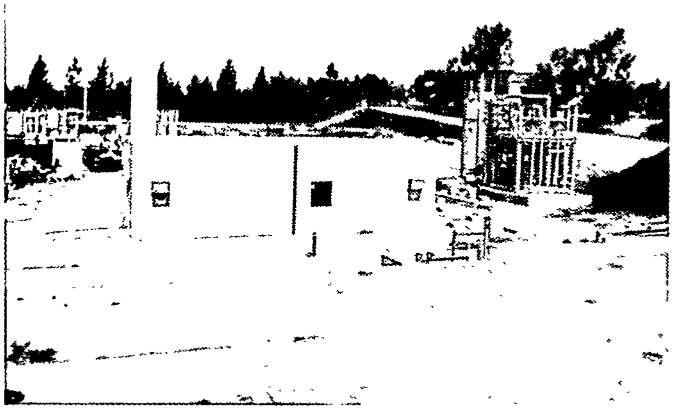
Figure 4 shows the placement of the control room module. It is a prefabricated, concrete building.



M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California
Positioning of KTI Reformer

Figure 5.

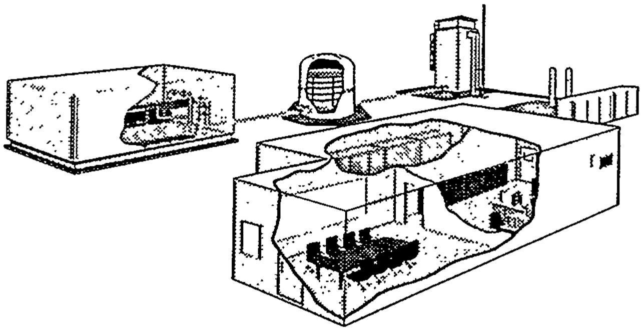
Figure 5 shows the placement of the reformer on its foundation. The small concrete pads in the foreground are for pipe supports



M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California
Current Status of Site

Figure 6.

Figure 6 shows the status of the site in mid July, when this paper was prepared. The liquid nitrogen tank and other equipment visible in the background are not part of the site. They will provide support to the site as well as to other Unocal pilot plant operations.



M-C Power 250 kW Fuel Cell Demonstration Plant
UNOCAL Fred L. Hartley Research Center
Brea, California
Completed Site

Figure 7.

Figure 7 is an artist' s rendering of the completed plant showing all the major pieces of equipment in place.

Upon completion of site construction in November we will commence testing all the components and debugging of the control systems, then proceed with plant start-up. The electricity produced by the plant will be fed into the research center utility system. The plant will also produce 150 psig steam that will be fed into the research center steam system. Natural gas fuel, water and compressed air will be provided from the research center utilities along with any other utilities that might be needed.

We will man the plant 24 hours per day with one person per shift. The operator will be backed up by our cogeneration plant operators and by refining pilot plant operators if necessary. In early

September we will start recruiting and training operators for the facility.

FUTURE WORK

We see this plant not only as the first large scale demonstration of molten carbonate fuel cell technology, but as a test bed for future large fuel cell stacks. As can be seen from the figures, the plant is modular in design so the various components can be easily modified or replaced. Additionally the facility is equipped with an extensive control and data acquisition system so process changes can be easily made and monitored. Chemical analysis of gas streams from the fuel cell can be carried out in our analytical laboratories. We look forward to participating in the further development and commercialization of fuel cell technology.

Panel Discussion

Fuel Cells -- An International Perspective

Fuel Cells - An International Perspective

CONTRACT INFORMATION

Contract Number	DE-AC21-90MC27394
Contractor	Illinois Institute of Technology 10 W. 33rd Street Chicago, IL 60616
Contractor Project Manager	T. Benjamin (M-C Power Corp.)
Principal Investigator	J.R. Selman
METC Project Manager	M.C. Williams
Period of Performance	Oct.1, 1992 - Aug. 31, 1994

ABSTRACT

An overview of recent fuel cell development and commercialization plans in Europe and Japan is presented. In Japan, the Moonlight Program which, starting in 1980, led to the successful demonstration and testing of 1 MW PAFCs and 100 kW MCFCs, is now bing continued as the "new Sunshine Program", with the goal of demonstrating 1 MW MCFC plants having at least 45% (HHV) efficiency, in 1997. Most major Japanese utilities cooperate with the developers in Technology Research Associations, which take care of developing and optimizing the Balance-of-Plant (BOP). In parallel with these major efforts, SOFC development is being pursued, by private programs funded by utilities as well as government programs. Recently, though, SOFC development appears to have lost some vigour compared to the advances in MCFC performance, especially of the DIRMCFC. Moreover, since 1992 a solid polymer (PEMFC) fuel cell development program has been started by the Japanese Government.

In Europe several national MCFC programs are in progress: in the Netherlands, Italy, and Germany, each closely allied with a U.S. developer. However, the major part of European and government and European Community fuel cell funding goes toward SOFC and PEMFC. The PEMFC development is directed at transportation applications and includes a significant activity in developing direct methanol fuel cells as well as methanol-reformate fueled PEMFCs.in developing direct methanol fuel cells as well as methanol-reformate fueled PEMFCs.

Performance of State-of-the-Art MCFC Stacks

	Nominal rating (kW)	Cell area (m^2)	Number cells	Current density (mA/cm^2)	Power density (mW/cm^2)	Operating pressure (MPa)	Life time (hx0.001)
ERC (IIR)	2	0.37	5	160	113	0.1	10
	70	0.37	234	120	93	0.1	2
	120	0.60	246	125	100	0.1	1.8
M-C Power	25	0.93	20	172	129	0.1	2.5
Hitachi	100	1.2	88	150	107	0.7	1.7
IHI	3	1.4	2	175	121	0.1	2.1
	100	1.0	102	150	126	0.7	5.1
MELCO (DIR)	30	0.5	62	150	106	0.1	≥5
MELCO (IIR)	100	0.5	192	150	114	0.1	2.3
ECN	10	0.335	33	150	125	0.4	2.1
(DIR)	1	0.1	10	150	109	0.1	0.7

Note: Values given for current density, power density, and operating pressure correspond, as much as can be surmised, to reported maximum stack performance. Stack lifetime does not necessarily correspond to operation under these conditions, and in most cases reflects "hot time" during a sequence of various operating conditions.

Summary table on FC-commercialization status as of mid-1994; BOP means balance of plant.*

Fuel-Cell Type	Demonstration Sales (Power Range)	Commercial Sales	Steps to Commercialization	Technology Leader(s) [Foreign Associate(s)]	Primary Foreign or US Competitors [Other Important Entries]
PAFC	1989 - 91 (200 kW to 11 MW)	Since 1992	Large Numbers of Sales	IFC (US) [CLC (Europe), Toshiba (Japan)]	Fuji (Japan) [ERC (US), MELCO (Japan), Sanyo (Japan)]
Atmospheric Pressure MCFC	Current (2 MW)	Later than 1997	Operation with BOP	ERC (US) [MELCO (Japan)]	IHI (Japan), [MELCO (Japan), Toshiba (Japan)]
Pressurized MCFC	Current (250 kW)	Later than 1997	Operation with BOP	MC-Power (US) [IHI (Japan)]	IHI (Japan) [BCN (The Netherlands), Hitachi (Japan), IFC (US), MELCO (Japan), MHI (Japan), Sanyo (Japan)]
Tubular SOFC	Current (25 kW and above)	Later than 1997	Operation of 100kW and larger systems with BOP	Westinghouse (US)	[MHI (Japan)]
				Representative Active Developers	
Planar SOFC (at 20 kW or more)	Not determinable		Feasibility demonstration for long-term, thermally self-sustaining operation	Allied Signal (US), Ceramatec (US), Dornier (Germany) and Cookson (UK), Fuji (Japan), Fujikura (Japan), GEC (UK), MELCO (Japan), MHI (Japan), Mitsui (Japan), Murata (Japan), Norcell (Norway), Osaka Gas (Japan), Sanyo (Japan), Siemens (Germany) and ECN (The Netherlands), Statol (Norway), Sulzer (Switzerland), Tonen (Japan), Ztek (US)	

Fuel-Cell Type	Program Goals		Steps to Commercialization	Technology Leader	Representative Participants:
	Motive	Stationary			
PEMFC	Vehicular commercial applications entering 2005-2007 (according to DOE)	Stationary power-supply demonstrations in the 1 to 200 kW range are projected by 2000	Feasibility demonstrations at acceptable costs	Ballard (Canada)	AISI (Japan), Allied-Signal (US), Allison Engine Co. (US), Analytic Power (US), Bestech (US), Daimler-Benz (Germany), Dow (US), ElectroChem (US), Elton Research (US), Energy Partners (US), Energy & Environmental Corp. (US), ERC (US), Fuji (Japan), Giner, Inc. (US), GM (US), H-Power (US), ICET (US), IFC (US), Interfacial Sciences (US), Lynntech (US), MHI (Japan), Physical Sciences, Inc. (US), Sanyo (Japan), Siemens (Germany), Tecogen (US), Westinghouse (US), plus numerous others

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* (Draft Report AFC2WG, Prof. S. Penner, Ed. 1994)

Appendices

Appendix A

Fuel Cells '94 Contractors Review Meeting

August 17-18, 1994

AGENDA

WEDNESDAY, AUGUST 17, 1994

7:30 a.m. Registration/Coffee and Refreshments

8:30 a.m. *Welcome to METC*
William T. Langan
DOE/METC

SESSION 1 -- OPENING SESSION

Chairperson: Manville J. Mayfield

8:45 a.m. 1.1 *DOE/METC Perspective*
Manville J. Mayfield
DOE/METC

9:00 a.m. 1.2 *An EPRI Perspective*
John B. O'Sullivan
EPRI

9:15 a.m. 1.3 *GRI Perspective*
James A. Kimball
GRI

9:30 a.m. 1.4 *DOE/EE&RE Perspective*
Albert R. Landgrebe
U.S. Department of Energy

9:45 a.m. 1.5 *DOD/ARPA Perspective*
Robert L. Rosenfeld
DOD/ARPA

10:00 a.m. BREAK

SESSION 2 -- SOLID OXIDE FUEL CELLS

Chairperson: Diane T. Hooie

10:30 a.m. 2.1 *Tubular Solid Oxide Fuel Cell Development Program*
Emerson R. Ray
Westinghouse Electric Corporation

11:00 a.m. 2.2 *Tubular Solid Oxide Fuel Cell Demonstration Activities*
Stephen E. Veyo
Westinghouse Electric Corporation

11:30 a.m. 2.3 *Alternative Materials for Solid Oxide Fuel Cells*
Timothy R. Armstrong
Pacific Northwest Laboratory

12:00 p.m. 2.4 *Advances in Research in Solid Oxide Fuel Cells*
Michael Krumpelt
Argonne National Laboratory

12:30 p.m. LUNCH

1:30 p.m. POSTER SESSION

P1 *Solid Oxide Material Research*
Jeffry W. Stevenson
Pacific Northwest Laboratory

P2 *Support Research for Solid Oxide Fuel Cells*
Timothy W. Kueper
Argonne National Laboratory

P3 *Multiply Manifolded Molten Carbonate Fuel Cells*
Michael F. Roche
Argonne National Laboratory

P4 *Lithium Ferrate and Lithium Cobaltate Cathodes for Molten Carbonate Fuel Cells*
Michael F. Roche
Argonne National Laboratory

P5	*Carbonate Fuel Cell Analysis* Jennifer Hunt Energy Research Corporation
P6	*Molten Carbonate Fuel Cell with High Power Density* Michael F. Roche Argonne National Laboratory
P7	*Pitting Corrosion of Aluminized Seals in Molten Carbonate Fuel Cells* Michael F. Roche Argonne National Laboratory
P8	*Processing of $LaCrO_3$ for Solid Oxide Fuel Cell Applications* Wayne Huebner University of Missouri-Rolla
P9	*Sealant Materials for the SOFC* Ira Bloom Argonne National Laboratory
P10	*Trace Species Removal for Fuel Cell Application* A. Ed Pigeaud Energy Research Corporation
P11	*Macrocyclic Redox Promoters for Methanol Oxidation-II. Fuel Cell Results* S. Sarangapani ICET, Inc.
P12	*Perovskite Solid Electrolytes for SOFC* Anthony F. Sammells Eltron Research, Inc.
P13	*A Practical Method of Hydrogen Purification for Use in Fuel-Cell Systems* David Edlund Bend Research, Inc.
P14	*Investigation of Wetting Phenomena and Oxidation/Lithiation Behavior of Nickel in Molten Carbonate* M. S. Yazici Illinois Institute of Technology

SESSION 3 -- DOE'S FUEL CELL TRANSPORTATION PROGRAM

Chairperson: Albert R. Landgrebe

2:30 p.m. 3.1 *Transportation Program Overview*
Albert R. Landgrebe
U.S. Department of Energy

3:00 p.m. 3.2 *Proton Exchange Membrane Fuel Cell for Transportation*
Swathy Swathirajan
General Motors Corporation

3:30 p.m. 3.3 *Phosphoric Acid Fuel Cell for Buses*
Arthur Kaufman
H-Power Corporation

4:00 p.m. ADJOURN

5:30 p.m. SOCIAL HOUR AND BARBECUE
Lakeview Resort and Conference Center

THURSDAY, AUGUST 18, 1994

7:30 a.m. Registration

8:20 a.m. *Welcome Back to METC*
Edward L. Parsons
DOE/METC

SESSION 4 -- DOD/ARPA'S FUEL CELL LOGISTICS FUEL PROGRAM

Chairperson: James K. Stedman

8:30 a.m. 4.1 *Overview of ARPA's Fuel Cell Logistics Fuel Program*
James K. Stedman
Institute for Defense Analysis

8:40 a.m. 4.2 *MCFC Development for Logistic Fuel Applications*
Sandors G. Abens
Energy Research Corporation

9:05 a.m.	4.3	*SOFC Development for Logistics Fuel Applications* Stephen E. Veyo Westinghouse Electric Corporation

SESSION 5 -- MOLTEN CARBONATE FUEL CELLS

Chairperson: Mark C. Williams

9:30 a.m.	5.1	*Update: Commercialization of the Direct Fuel Cell* Donald R. Glenn Fuel Cell Engineering Corporation
10:00 a.m.	5.2	*Overview of MCFC Commercialization Strategy* Paul B. Tarman M-C Power Corporation John T. Nimmons Alliance to Commercialize Carbonate Technology
10:30 a.m.		BREAK
10:50 a.m.	5.3	*Status of Santa Clara MCFC Product Development Test* Ashok Kush Fuel Cell Engineering Corp.
11:15 a.m.	5.4	*Status of 250 KW Product Development Test at San Diego Gas & Electric* R. Al Figueroa San Diego Gas & Electric
11:40 a.m.	5.5	*Status of 120-kW Testing/Product Improvement* Mohammad Farooque Energy Research Corporation
12:05 p.m.	5.6	*Status of Unocal Product Development Testing* Donald T. Clark UNOCAL
12:30 p.m.		LUNCH
1:30 p.m.		Panel Discussion *Fuel Cells -- An International Perspective* Moderator: Manville J. Mayfield

John B. O'Sullivan
Electric Power Research Institute

James Kimball
Gas Research Institute

Bernard S. Baker
Energy Research Corporation

Lee Camara
M-C Power Corporation

Walter J. Dollard
Westinghouse Electric Corporation

J. Lueckel
International Fuel Cells

J. Robert Selman
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3:30 p.m.	*Closing Remarks* Manville J. Mayfield
3:45 p.m.	Tour

Appendix B

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