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Proceedings of the Fuel Cells '95 Review Meeting

Tom J. George

August 1995

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

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Session 1

Opening Session

1.2 An EPRI Perspective — 1995

John B. O'Sullivan
Electric Power Research Institute

ABSTRACT

Whereas my views and commentary last year were decidedly upbeat — they will be more moderate this year. Why the change? Well, the much needed rebate program for the 200 kW PAFC units is moving with glacial speed, and both MCFC developers are behind schedule. Although in hindsight, both schedules were clearly optimistic and success oriented, it is not good to have delays during a congressional period that is focused on cost cutting, down selection, and schedule stretch out. And our utilities are also becoming impatient as vendor milestones slip. And lest anyone think that only the MCFC people have problems, the dominant SOFC developer has their next major demonstration in Europe, not in the United States.

What is my major concern — a possible hiatus in this extended period of adequate and constant funding levels that have provided the strong technologic base for these aggressive demonstrations. Are we going to hand the technologic reins over to Europe and Asia at this critical stage? — or — figure out an improved paradigm for R&D in a competitive world!

The three major historic funding entities: DOE, EPRI, and GRI are demonstrably weaker and less certain in 1996. DOD/ARPA and DOC/NIST will not be able to take up the slack with their more focused programs. I believe the utility programs will eventually turn around and become aggressive again as the "new utilities," gas and electric, define their future business roles. It is truly unfortunate if we have all three funding entities withdraw markedly from the field in the same time frame. DOE could be a crucial entity in 1996-1997.

The impact of utility deregulation in the funding of R&D and the difficulty in introducing new and advanced technology into low cost competitive business strategies is being recognized. Two of California's largest utilities are not funding advanced generation R&D. The California Energy Commission funded a study this spring to identify whether this might be a legitimate concern. This study was the likely trigger for a recently initiated GAO study of the same thesis requested by Representative George Brown (Democrat — California).

I can hardly wait to see what I write next year — assuming that there is a DOE Fuel Cell Contractors Meeting and that I am on the agenda for EPRI. Hopefully, the MCFC technologic demonstrations and the plans for future demonstrations of commercial prototypes will be positive and high on my list; to be immediately followed by all the good things happening with planar and tubular SOFC and PEMFCs for stationary applications. Last but not least, the PC-25s will have exceeded a critical point in market penetration and the price will be dropping in concert.

1.3 Fuel Cells '95 GRI Perspective

James Kimball
Gas Research Institute

ABSTRACT

The past 12 months have seen confirmation of the developments and trends we discussed at the 1994 conference. Much publicity has been given to fuel cells for power generation during this period, with the general theme that it is "time to take fuel cells seriously." For on-site generation, the one commercially available unit — the ONSI model PC-25 — has established itself as a robust and reliable piece of equipment. It is recognized, even at its current price, as a good niche product, particularly where clean and uninterruptible electric service is required, and many proposals are pending for energy services to end-users by means of one or more PC-25 units. However, announced sales to date have been disappointingly few. We pointed out a year ago that marketing issues now have supplanted R&D issues, at least for the PC-25. By this time next year, Gas Research Institute (GRI) hopes to see major successes from this product in the market.

The use of modular technologies in general, and fuel cells in particular, for distributed applications — be they for customer on-site generation or for use within the utility distribution company, or "Disco" — is a concept which has also gained greater currency since our last meeting. It is almost accepted wisdom now that these applications, rather than central station generation, are the appropriate ones for fuel cell technologies, and that the advantages of small size and mass-produced units can lead to an effective competitive strategy. The current period of intense scrutiny of institutional arrangements within the electric utility industry is expected to lead to major structural and operational changes, somewhat similar to those which shook the natural gas industry in the 1980's. Fuel cells, along with other modular technologies, will soon begin to look economically attractive to the distribution companies which are likely to survive electric utility restructuring.

Fuel cell technologies now under intense development, molten carbon-based generating equipment in particular, seem likely to reach commercial readiness at just the right time to enter the Disco market. This application should begin around the turn of the century, and achieve significant penetration over the subsequent 15 to 20 years. GRI is currently conducting a major technical, economic, business review of effort it is sponsoring in the MCFC area, to be sure that the right business steps are being taken to match technical development. A GRI-sponsored effort has been successfully completed in the past year to develop a suitable balance-of-plant design for a viable distributed utility MCFC power generating product. GRI will continue its attention to this application throughout the next year, including both technical and institutional issues. We believe fuel cells, and the distributed utility paradigm, will result in significant business opportunities for the natural gas industry.

1.4 DOE Perspective on Fuel Cells in Transportation

Robert Kost
Office of Transportation Technologies
U.S. Department of Energy

ABSTRACT

Fuel cells are one of the most promising technologies for meeting the rapidly growing demand for transportation services while minimizing adverse energy and environmental impacts. This paper reviews the benefits of introducing fuel cells into the transportation sector; in addition to dramatically reduced vehicle emissions, fuel cells offer the flexibility to use petroleum-based or alternative fuels, have significantly greater energy efficiency than internal combustion engines, and greatly reduce noise levels during operation. The rationale leading to the emphasis on proton-exchange-membrane fuel cells for transportation applications is reviewed as are the development issues requiring resolution to achieve adequate performance, packaging, and cost for use in automobiles. Technical targets for power density, specific power, platinum loading on the electrodes, cost, and other factors that become increasingly more demanding over time have been established. Fuel choice issues and pathways to reduced costs and to a renewable energy future are explored. One such path initially introduces fuel cell vehicles using reformed gasoline while on-board hydrogen storage technology is developed to the point of allowing adequate range (350 miles) and refueling convenience. This scenario also allows time for renewable hydrogen production technologies and the required supply infrastructure to develop. Finally, the DOE Fuel Cells in Transportation program is described. The program, whose goal is to establish the technology for fuel cell vehicles as rapidly as possible, is being implemented by means of the United States Fuel Cell Alliance, a Government-industry alliance that includes Detroit's Big Three automakers, fuel cell and other component suppliers, the national laboratories, and universities.

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
Program Engineer	Mr. Christopher Cracraft
METC Project Manager	Mr. Bruce C. Harrington
Period of Performance	April 1, 1991 to November 30, 1996

Schedule and Milestones

FY1995 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. The Westinghouse goal is to develop a cost effective 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 56,000 hours of continuous power operation at temperature. Results of development efforts to reduce cost and increase power output of tubular SOFCs are described.

BACKGROUND INFORMATION

High temperature solid oxide fuel cells (SOFCs) utilizing zirconia-based electrolyte offer a clean, pollution-free technology to electrochemically generate electricity at high efficiencies. SOFCs provide many advantages over traditional energy conversion systems; these include high efficiency, reliability, modularity, fuel adaptability, and very low levels of NOx and SOx emissions. Furthermore, because of their high temperature of operation (~1000°C), these cells can be operated directly on natural gas eliminating the need for an expensive, external reformer system. SOFCs also produce high quality exhust heat (600 to 900°C) which can be used in a gas turbine and/or combined cycle application.

A solid oxide fuel cell essentially consists of two porous electrodes separated by a dense, oxygen ion conducting electrolyte. Oxygen supplied at the cathode (air electrode) reacts with incoming electrons from the external circuit to form oxygen ions, which migrate to the anode (fuel electrode) through the oxygen ion conducting electrolyte. At the anode, oxygen ions combine with H_2 (and/or CO) in the fuel to form H_2O (and/or CO_2) and generate electricity.

While most fuel cell developers have gravitated to a planar configuration, Westinghouse developed a tubular design with a closed end. Any practical fuel cell generator requires the interconnection of many cells. This interconnection coupled with the necessity to keep fuel separated from oxidant creates design problems associated with seals. The Westinghouse tubular configuration facilitates a generator design which is "seal-less" that is, the generator requires no high integrity seals, necessary in other designs. The Westinghouse tubular design is therefore conductive to free thermal expansion and limited temperature gradiants thereby low mechanical stress.

The Westinghouse state-of-the-art tubular SOFC features a doped porous lanthanum manganite air electrode. In order to provide a current path from the air electode to the exterior of the cell, a gas tight, axial interconnection zone about 9 mm wide is fabricated on the air electrode. The interconnection is a doped lantanum chromite. The electrolyte, yttria stabilized cubic zirconia, is deposited over the exposed air electrode, overlaping slightly the interconnection to form a lap joint. The electrolyte is fully dense and gas tight, and about 40 microns thick. The fuel electrode is a porous nickel-zirconia cerment fabricated over the electrolyte surface, except in the vicinity of the interconnection.

For cell operation (Figure 1), oxidant (air or oxygen) is introduced through a ceramic injector tube positioned inside the cell. The oxidant, discharged near the closed end of the cell, flows through the annular space formed by the cell and the coaxial injector tube. Fuel flows on the outside of the cell from the closed end and is electrochemically oxidized while flowing to the open end of the cell generating electricity.

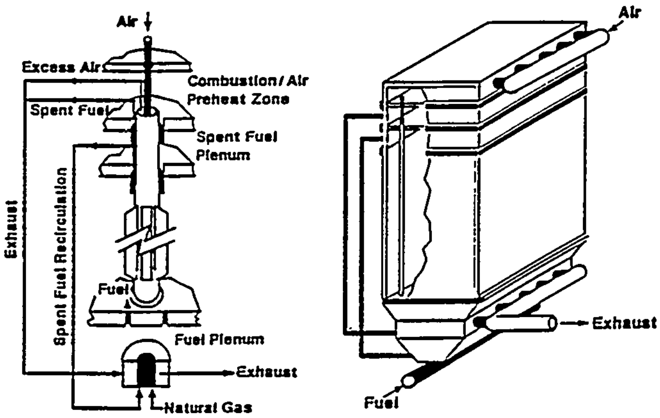


Figure 1. SOFC Generator

At the open end of the cell, the oxygen-depleted air exits the cell and is combusted with the partialy depleted fuel. Typically, 85% of the fuel is electrochemically utilized (reacted) in the active fuel cell section. Part of the depleted fuel is recirculated in the fuel stream and the rest combusted to preheat incoming air and/or fuel to the fuel cell.

During the cell operation 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.

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 development of a viable fuel cell electrical power generation system involves not only the development of cell technology, but also the development and demonstration of the generator module. Table 1 shows that the performance of the basic module design has been demonstrated under field test conditions by the operation of 11 generating systems.

Table 1
Westinghouse SOFC Generating Systems

System	Generator Rating	Number of Generators	Total Cells	Cell Length	Date
TVA	0.4 kW	1	24	30	1986
Osaka Gas	3 kW	1	144	36	1987
Osaka Gas	3 kW	1	144	36	1987
Tokyo Gas	3 kW	1	144	36	1987
JGU-1	20 kW	2	576	50	1992
UTILITIES A	20 kW	1	576	50	1992
UTILITIES B1	20 kW	1	576	50	1992
UTILITIES B2	20 kW	1	576	50	1993
SCE-1	20 kW	1	576	50	1994
JGU-2	25 kW	1	576	50	1995
SCE-2/ARPA	27 kW	1	576	50	1995
EDB/ELSAM*	100 kW	1	1152	150	1996

*in production.

Scaleup to the MW-class commercial modules is the next step, and is now underway. These modules would be arranged in a power block concept. Any number of power blocks could then be installed to satisfy the specific requirement for large plants (Figure 2). Conceptual design studies have confirmed the practicality and economic feasibility of the power block concept in applications ranging up to several hundred megawatts.

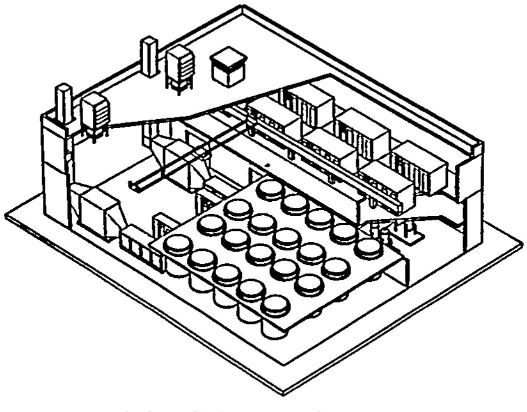


Figure 2. SOFC MWe-Class Power Plant

PROJECT DESCRIPTION

The development of the Westinghouse tubular SOFC technology is being advanced under a six-year cooperative agreement between DOE-Morgantown Energy Technology Center, Mr. Bruce C. Harrington, Program Manager, and Westinghouse. This program, extending from 1991 through 1996, 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. As shown in Table 1 Westinghouse has practiced the deployment with customers of fully integrated, automatically controlled, packaged solid oxide fuel cell power generation systems. Testing at customer sites has and will continue to provide operational feedback, field experience, and greater awareness of SOFC technology.

Successful commercialization of tubular 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.

Two Westinghouse tubular solid oxide fuel cells have been continuously generating power for over 56,000 hours (6.5 years) with degradation rates less than 0.5% per 1000 hours (Figure 3). These cells are the longest operating SOFCs in the world and show that Westinghouse is advancing the tubular SOFC technology toward commercialization. To date, Westinghouse has produced and operated for extended periods several thousand cells and has systematically improved cell performance, life, and voltage stability. Other long running cells have been shut down intentionally for detailed analysis in order that improvements can be made in future cells. Still other advanced cells have been under test for up to three years with similar degradation rates.

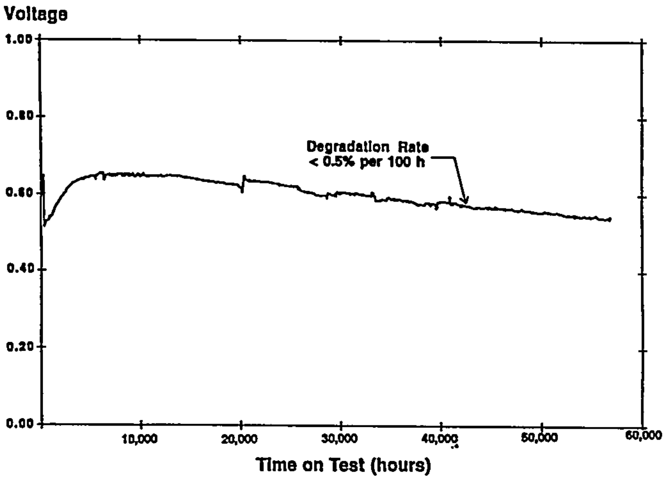


Figure 3. Longest Operating SOFC Test

Increased power output per unit cell length is desirable to improve SOFC power plant economics. The earlier technology tubular SOFC was fabricated on a calcia-stabilized zirconia porous support tube (PST). Although sufficiently porous, this tube presented an inherent impedance to air flow toward the air electrode. In order to reduce such impedance to air flow, the wall thickness of the PST was first decreased from the original 2 mm (the thick-wall PST) to 1.2 mm (the thin-wall PST). The calcia-stabilized zirconia suport tube has now been completely eliminated and replaced by a 1.9 mm wall thickness doped-lanthanum manganite tube in the state-of-the-art technology tubular SOFCs. These latest technology tubular cells are designated as air electrode supported (AES) cells; schematic design of such a cell is shown in Figure 4.

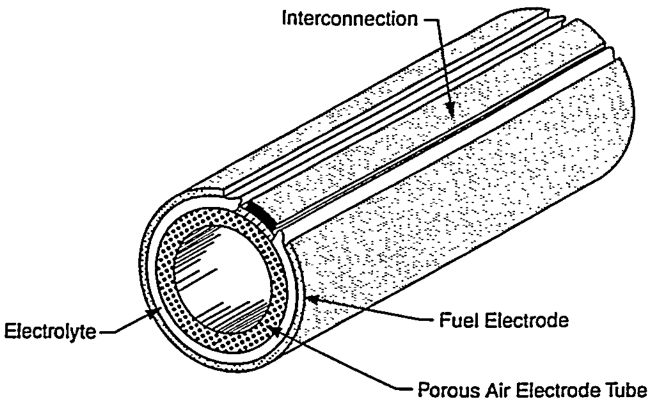


Figure 4. AES Type Tubular SOFC

The voltage-current characteristics of the AES cells (1.6 cm diameter, 50 cm active length) at 1000°C are compared with those of the thick-wall PST and the thin wall PST cells, with 89% H_2 + 11% H_2O Fuel (89% fuel utilization) and air as oxidant (4 stoichs), in Figure 5. The curves in Figure 5 clearly illustrate the significantly improved performance of the AES cells over the PST-type cells. Elimination of one component (ceramic support tube) combined with the higher performance yields a lower cost SOFC ($/W) than the earlier technology PST-type cells.

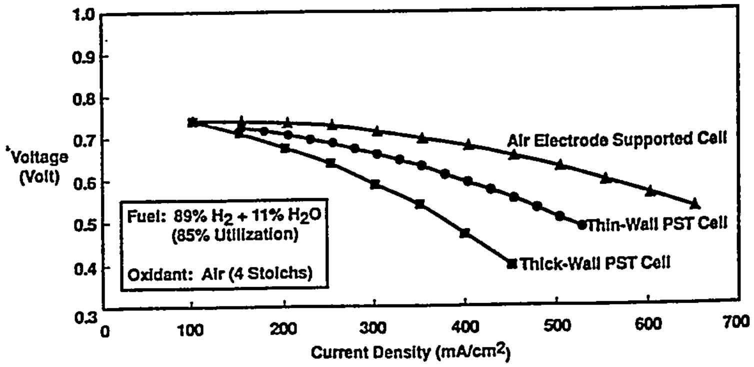


Figure 5. V-I Comparison of thick-wall PST, thin-wall PST, and AES cells.

In addition to eliminating the calcia-stabilized zirconia support tube, the active length of the cells has also been continually increased to increase the power output per cell. A greater cell power output decreases the number of cells required in a given power size generator and thus improves SOFC power plant economics. The active length has been increased from 30 cm for pre-1986 thick-wall PST cells to 150 cm for today's commercial prototype AES cells. Additionally, the diameter of the tube in longer length AES cells has been increased from 1.6 cm to 2.2 cm to accommodate larger pressure drops encountered in longer length cells. Figure 6 shows the power output from different cells illustrating the many-fold increase in power output from the latest technology AES cells.

As described above, Westinghouse development programs are successfully producing longer cells and increasing the power output per unit cell length so as to contribute to improve power plant economics. Also, Westinghouse has development programs in place to reduce the cost of the cells and generators by utilizing alternate fabrication techniques. These development programs will enhance the economic viability of tubular SOFCs for commercialization.

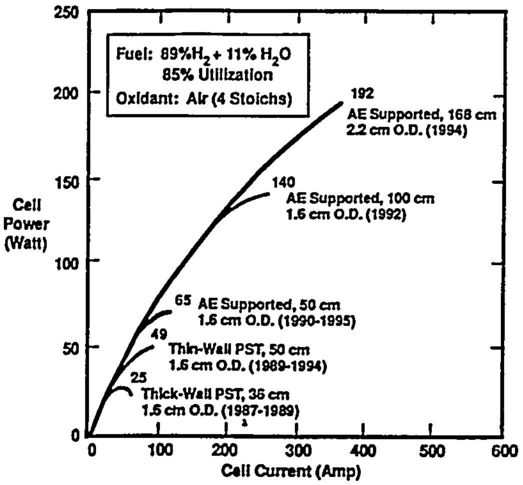


Figure 6. Power Output of Different Type and Length Cells.

The Pre-Pilot Manufacturing Facility (PPMF), located near Pittsburgh, PA, has been dedicated to the tubular SOFC technology. This unique facility has provided the means to move the technology from the laboratory environment to a manufacturing environment, while enabling the processes and quality control programs to be put in place that will be required to commercialize the technology during the 1990's.

This year a new facility will be completed, also located near Pittsburgh, PA, called the Pilot Manufacturing Facility (PMF). This facility represents a scaleup in production and SOFC size capability, and incorporates the refined processes and quality control programs developed at the PPMF. At the PMF, only the latest tubular SOFC technology, utilizing AES cells, will be produced.

The first 100 kWe Field Demonstration System, incorporating 150 cm AES cells will be fabricated and factory tested at the PMF, and then delivered to the Netherlands to a group of utilities consisting of the Dutch EDB consortium and the Danish ELSAM consortium. The system will be installed at NUON, one of the group of five Dutch gas and electricity distribution utilities which belong to the EDB consortium. The successful operation of this unit will represent a major milestone in the commercialization of advanced technology tubular SOFCs for large-scale power generation.

Most recently demonstrated was a 27 kWe field unit, which utilizes 50 cm AES cells and operates on natural gas and logistic fuels. Funding for this unit is provided by ARPA. The unit is being tested by Southern California Edison. Additional details of the operation and performance characteristics is the subject of a companion paper, to be presented by Dr. Stephen Veyo.

SUMMARY

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

- Single and multi-cell SOFCs have operated continuously for more than 56,000 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.
- A 20 kW SOFC was operated by Southern California Edison (SCE) for 6015 hours.
- The JGU 25 kWe SOFC customer unit began operation in 1995.
- The ARPA 27 kWe SOFC began operation at SCE in 1995.
- A pressurized test article operated for 5000 hours at Ontario Hydro Technologies (OHT).

High temperature SOFCs offer many advantages over traditional energy systems. These include high conversion efficiency, reliability, modularity, fuel adaptability, and virtually unlimited siteability due to very low NOx emissions. Furthermore, these cells produce high quality exhaust heat which can be used in a gas turbine and/or combined cycle to further increase the overall plant efficiency. Westinghouse has developed the materials and fabrication processes for these cells, increased the power output per cell, and has successfully employed and tested these cells for power generation in successively larger generators.

When fully commercialized, the SOFC systems are expected to serve a wide range of power and heat applications, such as multi-megawatt size distributed electric power plants located near load centers, repowering of existing units, and large central station all-electric power plants.

FUTURE WORK

The existing Cooperative Agreement between Westinghouse and the Department of Energy was initiated in April, 1991 and will continue through November 1996. This six 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), Mr. Bruce C. Harrington, Project Manager.

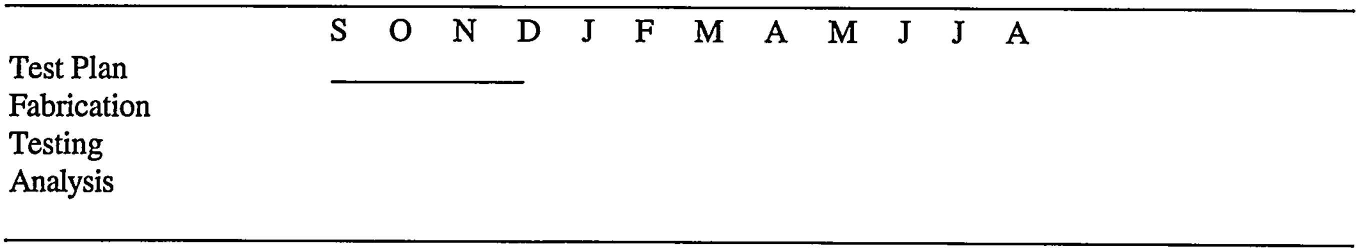
2.2 Tubular Solid Oxide Fuel Cell Demonstration Activities

CONTRACT INFORMATION

Contract Number	DE-FC21-91MC28055 NAS3-27022
Contractor	Westinghouse Electric Corporation 1310 Beulah Road Pittsburgh, PA 15235-5098 (412)256-1901 (telephone) (412)256-2002 (telefax)
Contractor Project Manager	Mr. Emerson R. Ray
Principal Investigators	Dr. Stephen E. Veyo
METC Project Manager	Mr. Bruce Harrington
Period of Performance	April 1, 1991 Nov. 30, 1995

Schedule and Milestones

FY1995 Program Schedule



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 SOFC technology. The design and construction of the host systems for SOFC customer test units has been funded with customer and Westinghouse moneys. 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 at no cost to DOE 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 driven 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

Westinghouse has practiced the deployment with customers of fully integrated, automatically controlled, packaged solid oxide fuel cell power generation systems since 1986 when a 400 Watt system was supplied to the Tennessee Valley Authority. Subsequently, Westinghouse deployed two 3 kW SOFC systems with The Tokyo Gas Company, Ltd. and The Osaka Gas Company Ltd., and most recently several 20 kW class systems. The 20 kW class systems utilize the multi kilo-Watt (MKG) generator module or stack developed under a cooperative agreement with the United States Department of Energy- Morgantown Energy Technology Center (DOE-METC). This generator module contains 576 tubular SOFCs of nominally 16 mm diameter with 500 mm active length arranged in four quadrants. Each quadrant contains eight bundles of eighteen cells. This construction is shown in Figure 1. Originally designed to use cells that employ a porous zirconia support tube (PST), the stack design has most recently been adapted to use the air electrode supported (AES) cell design.

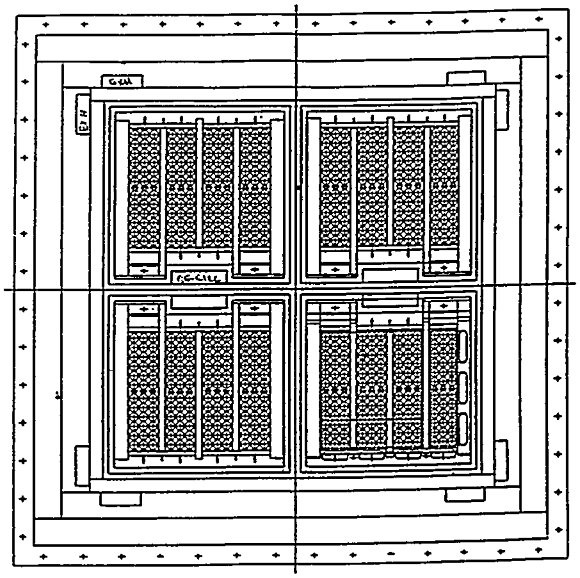


Figure 1. MKG SOFC Generator Cross Section

PROJECT DESCRIPTION

The UTILITIES, a consortium of the Kansai Electric Power Company (KEPCO), Tokyo Gas, and Osaka Gas tested three MKG class SOFC stacks in fully integrated automatically controlled systems. The stacks utilized the porous support tube (PST) type of tubular SOFC. The UTILITIES' systems are packaged dc generation systems that use one 20 kW class (MKG) SOFC stack per system enclosure. The most successful of these achieved 7064 hours of operation generating electric power before planned shutdown in March 1994. The last 5200 hours of operation were without forced outage. Further details along with results may be found in the literature. (See for example references 1,2,4 and 5)

The Southern California Edison Company tested a sibling SOFC system, shown in Figure 2, for 6000 hours before planned shutdown on May 2, 1995. In late May, a Westinghouse field service team exchanged an AES generator module (stack)for the PST type then in place, and restarted the system on May 23 after completing revisions to the SOFC system to permit fueling by either natural gas or reformate from a logistic fuels (diesel or jet turbine fuel) processor.

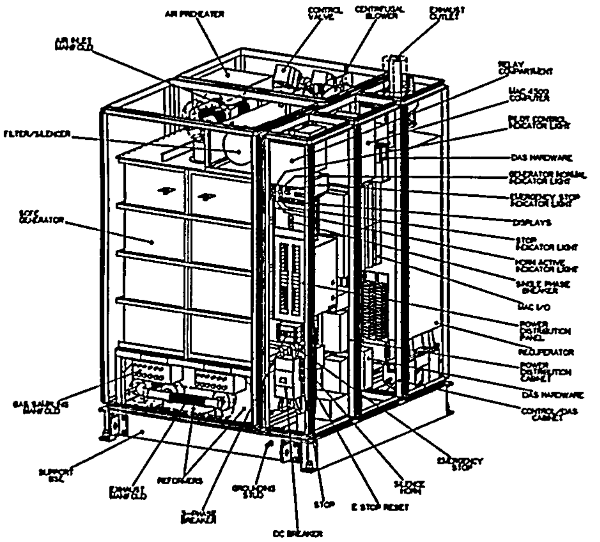


Figure 2. SCE 20 kW SOFC Power System

The Joint Gas Utilities (JGU), a consortium of the Tokyo Gas Company and the Osaka Gas Company, sponsored a 25 kW SOFC cogeneration system that utilized two MKG class SOFC modules in a single packaged enclosure. A description of this unit and its performance can be found in references 3 and 6. The JGU unit did not perform satisfactorily and arrangements were made for its repair. The cogeneration system was modified to accept a single AES-SOFC module in place of a previously used pair of PST type modules. An extended factory test that was initiated on March 22, 1995 was suspended on May 11 after 1200 hours of operation because the integral reformers became deactivated. The reformer catalyst will be replaced and a restart is expected in August 1995.

In addition to providing benchmark endurance data for high temperature fuel cell stacks, the 20 kW systems will guide design and development of the next SOFC field unit of 100 kW class.

The first 100 kW SOFC field unit is scheduled to be delivered to a consortium of Dutch and Danish utilities in late 1996, and will be operated at a test site provided by NUON, one of the sponsoring Dutch utilities. This unit will use the latest in Westinghouse's tubular SOFC technology, AES cells of nominally 22 mm diameter by 1500 mm in active length. System final design is in process, and the first cells are in trial production at a second generation pilot manufacturing facility provided by Westinghouse.

Testing of cells typical of the 100 kW unit has been initiated by Westinghouse. In addition, sibling cells will be tested by the Kansai Electric Power Company in an automated test apparatus purchased by KEPCO from Westinghouse. This test apparatus is capable of exercising up to four cells simultaneously under both steady conditions and transient conditions in a programmed thermal cycle. To date, KEPCO has exercised AES cells typical of the 20 kW class systems.

Table 1 provides a summary of the characteristics of Westinghouse SOFC field units to date.

Table 1.
Record of SOFC Field Unit Programs

Time Year	Customer	Stack Rating (kW)	Stack Number	Cell Type	Cell Length (mm)	Cell Number	Oper. (Hrs)	Fuel	MWH
1986	TVA	0.4	1	TK-PST	300	24	1760	H_2+CO	0.5
1987	Osaka Gas	3	1	TK-PST	360	144	3012	H_2+CO	6.1
1987	Osaka Gas	3	1	TK-PST	360	144	3683	H_2+CO	7.4
1987	Tokyo Gas	3	1	TK-PST	360	144	4882	H_2+CO	9.7
1992	JGU-1	20	2	TN-PST	500	576	817	PNG	10.8
1992	UTILITIES-A	20	1	TN-PST	500	576	2601	PNG	35.0
1992	UTILITIES-B1	20	1	TN-PST	500	576	1579	PNG	25.5
1993	UTILITIES-B2	20	1	TN-PST	500	576	7064	PNG	108.0
1994	SCE-1	20	1	TN-PST	500	576	6015	PNG	93.1
1995	JGU-2	25	1	AES	500	576	1200	PNG	23.2
1995	SCE-2	27	1	AES	500	576	1000+	PNG/DF-2/JP-8	22+
				Future Work					
1996	EDB/ELSAM	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

RESULTS

A 20 kW SOFC system unit was delivered to the Highgrove Generating Station of SCE in Grand Terrace (near San Bernardino), CA in March 1994, and in late May 1994 an MKG type PST-SOFC stack was installed. The unit was successfully started on June 7, 1994. On May 2, 1995, the unit was shut down in order to permit installation of a new stack incorporating the air electrode supported (AES) tubular cell technology.

A record of the terminal power as a function of time for the program period is shown in Figure 3. For a program period of 8000 hours, the unit operated for 6079 hours. Shortly after startup, the unit suffered two unscheduled shutdowns. The first was due to inadequate natural gas pressure into the gas compressor, and the second was due to an inadvertent interruption of UPS sourced power to the unit. For the first 1700 hours of operation, cell performance improved with time and SCE operators exercised the system over a range of operating parameters. The maximum power achieved was 18.9 kW at 85% fuel utilization and a generator control temperature of 1020°C. After about 1700 hours, system performance began to decline. The last 4000 hours of operation was not continuous, but was interrupted by three site power outages at the SCE test site, at 1900 hours into the test, and at 4550, and 5100 hours. The stoppage that occurred at the 5100 hour point coincided with the end of the year holiday season, and as a result no attempt was made to restart until 5700 hours into the program. Unfortunately, the electric air heater used for startup failed at this point and the restart was aborted. The spare heater was installed, but programmatic difficulties within SCE prohibited restart until 6400 hours into the program, in February 1995. From this date until the planned final shutdown, operation went smoothly although the power capability of the stack was declining.

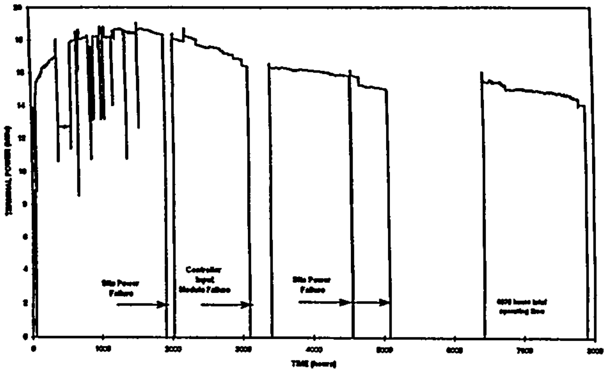


Figure 3. Power vs Time
SCE 20 kW SOFC Power System

In late May 1995, a Westinghouse field service team removed the PST type stack and returned it to Westinghouse manufacturing facilities in Pittsburgh for post test evaluation. A replacement stack utilizing air electrode supported (AES) cells was installed. This new stack was fabricated as part of a project funded by the United States Department of Defense (DOD) Advanced Research Projects Agency (ARPA) administered by the National Aeronautics and Space Agency (NASA) under contract NAS3-27022. Under the NASA contract, the SOFC system was modified so as to be able to accept as fuel either natural gas or reformate from a logistic fuels (DF-2 diesel or JP-8 jet turbine fuel) processor located externally to the SOFC system. The NASA contract also supported the development of a "brassboard" logistic fuels processor. This LFP has been developed by Haldor Topsoe, Inc. under a subcontract.

The ARPA/SCE AES SOFC system was successfully started on May 23, 1995 and will operate on natural gas fuel until such time as the LFP is ready for use. During the period June 12 through June 29 the SOFC system was exercised using natural gas as fuel. Shown in Figure 4 are voltage versus current plots for generator control temperatures of 1020, 1035, and 1050°C. The generator control temperature coincides roughly with the temperature at the centroid of a quadrant of the active stack. In general the 20 kW class of SOFC systems can not operate on natural gas at generator currents less than approximately 100 Amperes because there is concomitantly insufficient fuel pressure for the ejector type spent fuel recirculation pump to recirculate sufficient water vapor and CO_2 to maintain the O:C ratio as required. The system is controlled to maintain generator voltage above a pre-defined limit, which limits the maximum achievable current and power. Figure 4 shows that this limit coincides presently with a terminal voltage of approximately 116 Volts. It can be seen that operation at higher generator temperatures has no benefit at current less than about 140 Amperes, and an increasing benefit at higher current. Figure 5 shows the power capability of the AES-SOFC stack as a function of current at the same three control temperatures. The curves are almost congruent over their common range. A power level of 25 kW was achieved at 215 Amperes and 83% fuel utilization, 18% air utilization with a control temperature of 1050°C at 770 hours of operation. The achievement of higher power levels at lower values of fuel utilization was foiled by insufficient site natural gas pressure. The power curve as shown in Figure 5 is extrapolated from 215 to 240 Amperes.

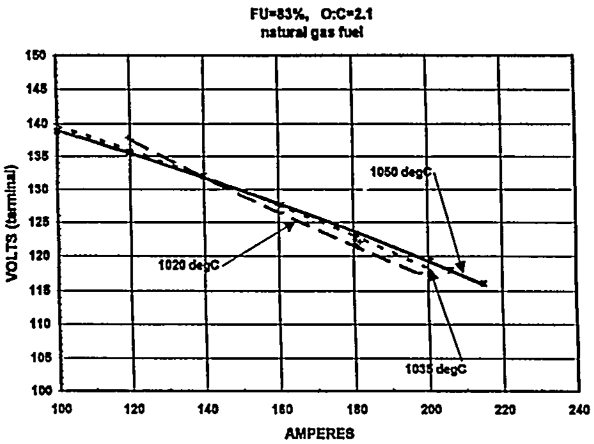


Figure 4. Voltage vs Current ARPA/SCE 25 kW AES-SOFC Power System

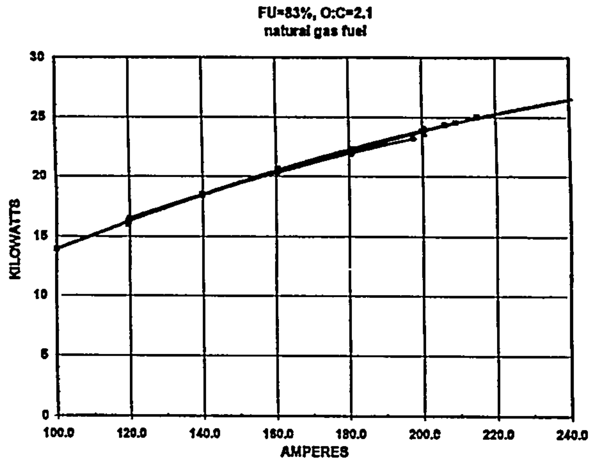


Figure 5. Power vs Current ARPA/SCE 25 kW AES-SOFC Power System

A plot of power versus time of operation for the ARPA/SCE AES SOFC system is shown in Figure 6. One forced outage is shown. This outage was caused by the process air duct not being secured to the blower discharge nozzle following the PST/AES stack exchange operation. Note from Figure 7 that through 1200 hours of operation, the AES-SOFC stack continues to improve in performance.

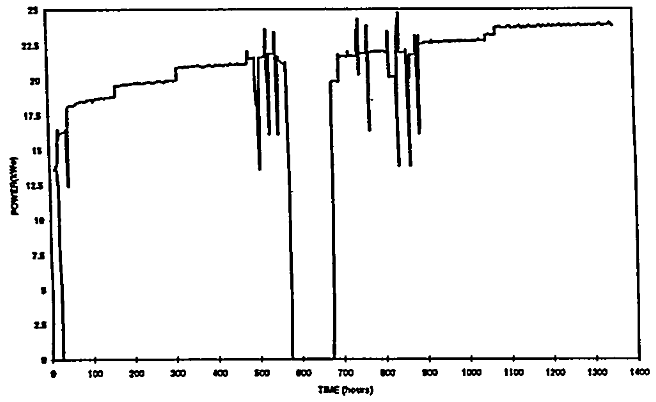


Figure 6. Power vs Time
ARPA/SCE 25 kW AES-SOFC Power System

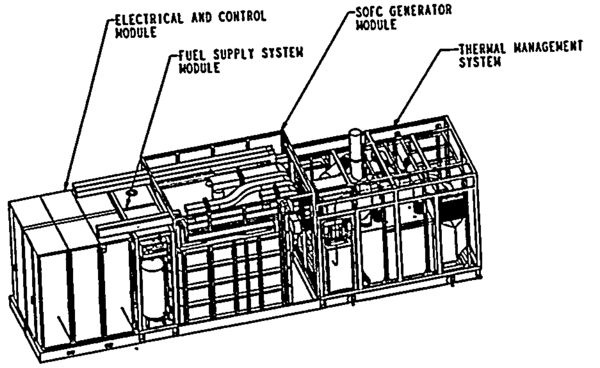


Figure 7. 100 kW SOFC Power System

Factory test of the JGU-AES-SOFC Cogeneration system was initiated on 22 March 1995. One anomaly apparent early in operation was that there was a much more marked spread in the control temperatures for the four quadrants, approximately 50°C, than had ever been observed in the past. Except for this quadrant to quadrant temperature anomaly, the stack appeared to be in excellent condition with good internal voltage balance and good internal temperature distribution.

For approximately 500 hours the cells continued to improve and current was increased. A maximum power of 21.4 kW was achieved at 180 Amperes, and 80% fuel utilization. Cell voltage improvement was observed until about 850 hours into operation, after which it was observed the cells in three quadrants were declining in voltage. Various diagnostic activities were undertaken, and at approximately 1200 hours of operation the unit was shut down since it became clear that the reason for the voltage decline was a combination of deficient fuel flow in the hottest quadrant due to a fuel leak and reformer deactivation as evidenced by a change in the temperature distribution and an increased burden on the air heater to maintain temperature--i.e. cooling of the cells.

After shut down of the JGU unit, cold flow tests were conducted to confirm balance in air and fuel flows and corrective measures taken to ensure fuel flow balance and eliminate a fuel leaks from the reformers. In addition, the reformer catalyst has been replaced and the control algorithm designed to ensure adequate oxygen to carbon atom ratio in the reformer revised.

The JGU 25 kW AES-SOFC Cogeneration system was moved from its site of manufacture, the Westinghouse Pre-Pilot Manufacturing Facility (PPMF) in Monroeville, PA to the new Pilot Manufacturing Facility (PMF) located at the Westinghouse Science and Technology Center in mid July 1995. Factory test of this unit is expected to resume in August 1995, at the PMF.

CONCLUSIONS

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

The MKG class SOFC stack when equipped with AES cells can achieve a power output in excess of 25 kW, a 25% improvement relative to the capabilities of the PST-SOFC stack.

As evidenced by operation of the SOFC system at the Highgrove Generating Station of Southern California Edison, the SOFC system is maneuverable across the range of its control parameters and operation and maintenance by utility personnel is easily accomplished.

As evidenced by operation of the SOFC system at the SCE site, automatic shutdown in response to faults and subsequent recovery to automatic operation is a normalized procedure.

Southern California Edison's operation of an SOFC system for six thousand hours of power generation represents a domestic record for high temperature fuel cells of 20 kW capacity class.

FUTURE WORK

Under the sponsorship of the Advanced Research Projects Agency of the Department of Defense, the 25 kW AES-SOFC system now operating at Southern California Edison with natural gas fuel will be switched to processed logistics fuel. This switch-over in fuel is scheduled to occur in July 1995.

The extended factory test of the JGU 25 kW AES-SOFC Cogeneration System will be resumed in August 1995.

The first field unit to incorporate the 22 mm diameter by 1500 mm long AES SOFC is a 100 kW SOFC Power System shown in Figure 7. This unit is sponsored by a consortium of Dutch and Danish utilities designated as EDB/ELSAM. The unit will be tested at the NUON (one of the sponsoring Dutch utilities) district heating site at Westervoort, The Netherlands.

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, and the United States Department of Defense - Advanced Research Projects Agency for funding the fabrication and test of a 25 kW class AES-SOFC stack on both natural gas and logistic fuels. 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 Gas Company; the Southern California Edison Company, and EDB/ELSAM. A particular acknowledgment and note of gratitude is due to personnel of The Southern California Edison Company for their competent and responsive installation and operation of a 20 kW class SOFC system, setting a domestic record of 6000 hours for high temperature fuel cell system and stack operation.

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2.3 Advanced Materials for Solid Oxide Fuel Cells

CONTRACT INFORMATION

Contract Number (FTPA)	22407
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
Contractor Project Manager	Dr. Timothy R. Armstrong
Principle Investigators	Dr. Timothy R. Armstrong Dr. Jeffry Stevenson
METC Project Manager	Bruce Harrington
Period of Performance	October 1, 1994 to June 30, 1995

Schedule and Milestones

FY 95 Program Schedule

	S	O	N	D	J	F	M	A	M	J	J	A
Interconnect Development		----	----	----	----	----	----	----	----	----	----	----
High Performance Cathode												
Accelerated Testing		----	----	----	----	----	----	----	----	----	----	----
Materials for Reduced Temperature Operation												

OBJECTIVES

The purpose of this research is to improve the properties of the current state-of-the-art materials used for solid oxide fuel cells (SOFCs). The objectives are to:

1. develop materials based on modifications of the state-of-the-art materials;
2. minimize or eliminate stability problems in the cathode, anode, and interconnect;
3. Electrochemically evaluate (in reproducible and controlled laboratory tests) the current state-of-the-art air electrode materials and cathode/electrolyte interfacial properties;
4. Develop accelerated electrochemical test methods to evaluate the performance of SOFCs under controlled and reproducible conditions; and
5. Develop and test materials for use in low-temperature SOFCs.

The goal is to modify and improve the current state-of-the-art materials and minimize the total number of cations in each material to avoid negative effects on the materials properties.

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 amterials for the simultaneous fabricatoin and consolidation in air of the interconnections and electrodes with the solid electrolyte, and (5) understand electochemcial reactions at materials interfaces and the effects of component composition and processing on those reactions.

BACKGOUND INFORMATION

Solid oxide fuel cells have emerged as a clean and efficient technology for the direct conversion of hydrogen and fossil fuels to electrical energy. The key feature of a SOFC is its high energy conversion efficiency. Compared with conventional methods of power generation, SOFCs offer several advantages: substantially higher conversion efficiency, modular construction, the potential for cogeneration, and much lower production of pollutants.

SOFCs are typically constructed using ceramic materials. Therefore, these fuel cells have several advantages over other types of fuel cells, specifically the use of nonprecious metals, solid state construction, and an invariant electrolyte. Ceramic fuel cells, however, place severe demands on the materials used due to their high operating temperature (1000°C) and the dual environments (oxidizing and reducing). Ceramic fabrication processes need to be developed to allow the incorporation of materials into practical stack configurations. At present, the key technical challenges for the development and commercialization of SOFCs are the development of suitable materials (specifically interconnect), improving the performance of the cathode/electrolyte interface and developing low-cost fabrication methods.

The principal components of a SOFC stack are the electrolyte, the anode, the cathode, and the interconnect. Each component serves several functions in the fuel cell and must meet certain requirements. Each component must have the proper stability (i.e.. chemical, phase, morphological, and dimensional) in oxidizing and/or reducing environments, chemical compatibility with other components, and satisfactory conductivity. The components, in addition, must have similar coefficients of thermal expansion. The electrolyte and interconnect must be dense to prevent gas mixing, whereas the anode and cathode must be porous to allow gas transport to the triple phase boundary.

PROJECT DESCRIPTION

The overall approach to this research and development program is to:

1. Modify the current interconnect and cathode materials to minimize or eliminate stability problems, minimizing the number of cations in the composition to reduce potential deleterious interaction with other materials.

2. Electrochemically evaluate (in reproducible and controlled laboratory test) the current state-of-the-art air electrode materials and cathode/electrolyte interfacial properties.

3. Develop methods to improve the air electrode/electrolyte interface through compositional modification. This will require modification of the interface with electronic, ionic, or mixed electronic/ionic conductors.

4. Develop accelerated electrochemical test methods to evaluate the performance of SOFCs under controlled and reproducible conditions.

5. Develop and test materials for use in low-temperature SOFCs. These materials need to be compatible with

the new electrolytes, such as barium cerate, being developed.

RESULTS

Experimental Procedure

$(La_{1-x}Sr_x)CrO_3$ and $(La_{1-x}Ca_x)CrO_3$ powders, where x was varied from 0.1 to 0.4, were synthesized using the glycine-nitrate process (1). All powders synthesized for this study had an A/B cation ratio equal to unity. The powders were precalcined at 650°C for 0.5 hour to decompose any residual organic material and then calcined at 1000°C for 1 hour. The powders were uniaxially pressed into bars (46mm x 16mm x 3mm) at 55 MPa followed by isostatic pressing at 138 MPa. Samples were sintered in air at 1500° to 1650°C for 5 hours ($MoSi_2$ furnace). Additional samples were prepared with the general formula $La_{0.7}Ca_{0.3}(Cr_{0.9}M_{0.1})O_3$ (where M was Ti, Co, Fe, Ni, Zn, Ga, Al, Mn or Cu) to evaluate the effects of isovalent and aliovalent B-site dopants on the dimensional stability. The densities all samples were determined using the Archimedas method.

Dilatometry measurements (Unitherm Model 1161, Anter Laboratories, Pittsburgh, PA) were used to study the dependence of the physical properties of sintered lanthanum chromites on ambient oxygen partial pressure, $P(O_2)$, and temperature. The dilatometry experiments were carried out on sintered bars (30 mm x 3mm x 3mm). Dilatometry experiments as a function of the $P(O_2)$ were carried out using a buffered $CO_2/Ar/H_2$ gas system, enabling expansion measurements to be made over an oxygen partial pressure range from 10^{-5} to 10^{-18} atmospheres at 700, 800, 900, and 1000°C. To ensure complete mixing of the gases, flow rates of 300 sccm were used. Drag forces from the gas flow in the dilatometer were evaluated and corrected for by carrying out similar experiments using an invariant standard (Al_2O_3, 99.9% dense). Calibration curves generated as a function of temperature and $P(O_2)$ were used to normalize the expansion measurements on the chromite specimens. Samples were initially heated at 2°C/min to the soak temperature in air and allowed to equilibrate for approximately 1 hour. After the sample equilibrated the oxygen partial pressure was changed, and measurements were taken until sample expansion was no longer noticeable. This process was repeated until data was collected over the complete $P(O_2)$ range.

The inital oxygen content and Cr^{4+} concentration were determined using a potentiometric titration technique developed by Nadalin and Brozda (2). Sintered samples were ground to a fine powder in a motar and pestle. Approximately 0.15 gm of lanthanum chromite was placed into a teflon pressure vessle with a solution composed of equal parts of vanadium sulfate and sulfuric acid (36N). The pressure vessle was placed into a stainless steel containment vessel and heated at 245°C until the chromite sample dissolve (1 to 6 days). The resultant solution was titrated with a 0.1N ferrous ammonium sulfate solution. The oxygen nonstoichiometry (d) and the Cr^{4+} concentration were then calculated using the electronutrality equation.

Thermogravimetric analysis (TGA, Cahn Microbalance) was carried out at 1000°C in both air and Ar/H_2 gas environments to determine the oxygen loss, upon reduction, and uptake, upon reoxidation. Sintered samples of lanthanum chromite were heated to temperature, allowed to equilibrate, and then reduced. Samples were exposed to the reducing environment for 12 hours after which the TGA atmosphere was changed to air and the sample re-oxidized for up to 12 hours. The relative weight changes were measured and corrected for buoyancy effects.

Stability of Lanthanum Chromite in Reducing Environments

Figure 1 is a plot of the isothermal linear expansion relative to the dimension in air at 700, 800, and 900°C for $La_{0.76}Sr_{0.24}CrO_3$ (denoted LSC-24) as a function of time. The specimens were equilibrated in air, reduced in Ar-4% H_2, and then re-oxidized in air. It was observed that the expansion in reducing environments was rapid at 800° and 900°C (approaching equilibrium in one to two hours) and reversible. When air was introduced the samples contracted very rapidly to within ≈0.05% of their initial length. This figure indicates that LSC expands significantly at temperatures as low as 700°C and that the kinetics of the reaction increase with temperature due to an increasing oxygen exchange rate (3).

Figure 2 shows the effect of decreasing $P(O_2)$ on the linear expansion of LSC-24 as a function of temperature. This sample showed no effect of reducing environment from ambient pressure to 10^{-8} atmosphere at 1000°C. As the $P(O_2)$ was lowered further, the sample steadily expanded, reaching a maximum expansion of ≈0.23% at 10^{-18} atmospheres. Figure 2 also clearly shows that the onset (critical oxygen partial pressure) of expansion moved towards lower oxygen partial pressures with decreasing temperature. Both figures 1 and 2 indicate that the magnitude of expansion decreases with decreasing temperature.

Figure 3 is a plot of isothermal expansion at 1000°C as a function of $P(O_2)$ for $La_{1-x}Sr_xCrO_3$ where x was varied from 0.16 to 0.30. This data indicates that the expansion of these materials increased not only with decreasing $P(O_2)$ but also with increasing Sr content on the A-site. The critical oxygen partial pressure at which expansion was first observed was higher for samples having Sr concentrations greater than 20 mole percent.

Figure 4 shows a comparison between LSC-20 data of Srilomsak et al. (3) and the present measurements for LSC-20 and LSC-24. There are significant differences between the two sets of data. The onset of expansion reported by Srilomsak et al. occurred at 10^{-10} atmospheres, two orders of magnitude higher than equivalent samples examined by the authors. A second notable difference is in the magnitude of the expansions. This 40% difference may be due to the presence of strontium chromate, $SrCrO_4$, as a secondary phase in the sample measured by Srilomsak et al. (4). Observations of single phase Sr and Ca chromate indicate that they expand rapidly on reduction and decomposition.

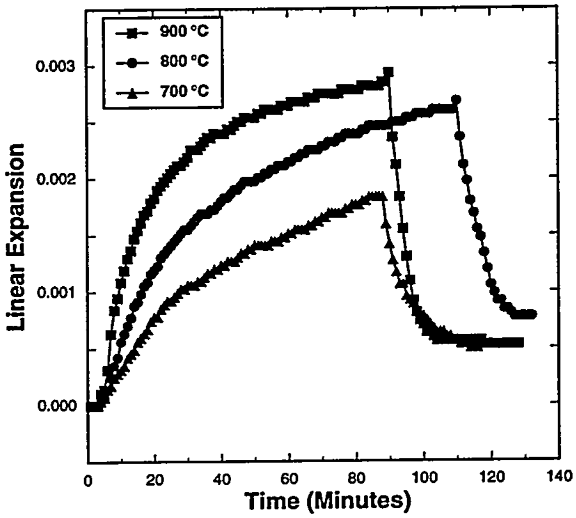

Figure 1. Linear Expansion of LSC-24 in Ar-4%H_2 at 700°, 800° and 900°C

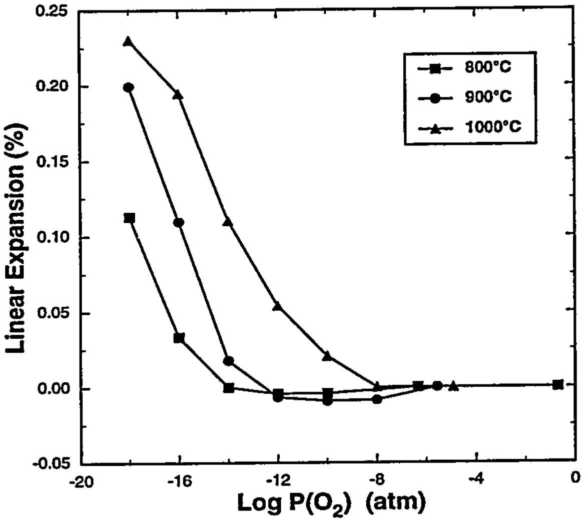

Figure 2. Linear Expansion of LSC-24 asa Function of Temperature and Oxygen Partial Pressure.

The critical partial pressure increases with increasing temperature. Significant amounts of secondary chromate phase may enhance the expansion upon reduction. Another plausible explanation is that the composition of the sample may be closer to LSC-24 than LSC-20 as indicated by the similarity of data in Figure 4.

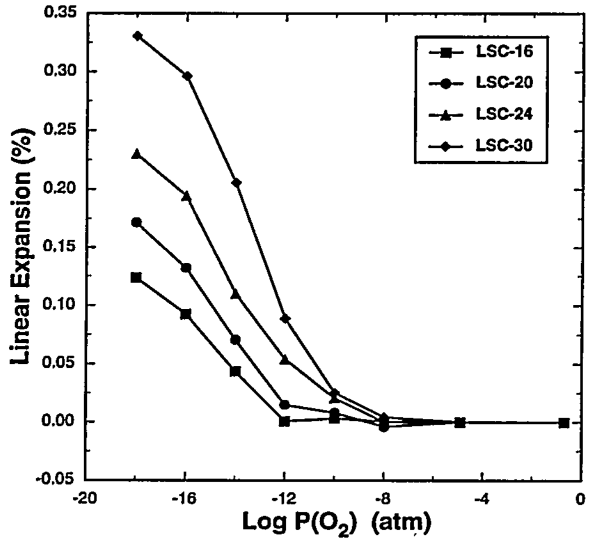

Figure 3. Linear Expansion of $La_{1-x}Sr_xCrO_3$ as a Function of Oxygen Partial Pressure at 1000°C.

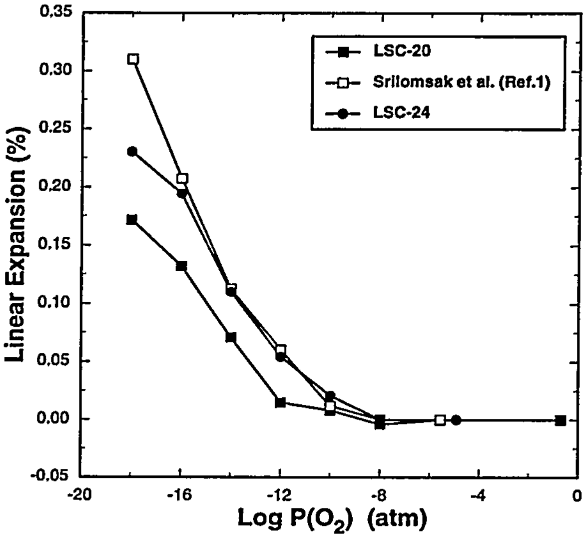

Figure 4. Comparison of LSC-20 LinearExpansion Data.

Specimens with either Sr or Ca show similar expansion behavior in reducing environments. As observed previously in LSC-24 the expansion is reversible and the degree of expansion increases with temperature. However, in LCC-30 the amount of expansion the sample undergoes at 700°C is considerably less than that for comparable Sr materials.

Acceptor doping of lanthanum chromite (substitution of Sr or Ca for La) can be charge compensated either electronically (through the conversion of Cr^{3+} to Cr^{4+}) or ionically (through the creation of oxygen vacancies), as shown by the electroneutrality relation in Equation 1 (Kroger-Vink notation; brackets denote concentration).

$$[Sr'_{La}] = [Cr^{\bullet}_{Cr}] + 2[V^{\bullet\bullet}_{O}] \qquad [1]$$

At high oxygen partial pressures, this compensation is electronic, as verified experimentally in a study by Pederson and Dake using XPS (5) in which the Cr^{4+} concentration was found to be equivalent to the A-site acceptor concentration. At low oxygen partial pressures, charge compensation is dominated by the formation of oxygen vacancies, as shown in Equation 2.

$$La^{x}_{1-x}M'_{x}Cr^{x}_{1-x}Cr^{\bullet}_{x}O^{x}_{3} \rightarrow$$
$$La^{x}_{1-x}M'_{x}Cr^{x}_{1-x+2\delta}Cr^{\bullet}_{x-2\delta}O^{x}_{3-\delta}[V^{\bullet\bullet}_{O}]_{\delta} + \frac{\delta}{2}O_2$$

TGA analysis on these samples by the authors has verified an appreciable decrease in mass due to oxygen evolution during reduction (6). The weight loss (vacancy formation) and gain due to oxygen removal and uptake from the lattice correspond well with the observed reversible linear expansion as well as with electrical conductivity measurements carried out by others (7,8). Based on this information, it would be expected that increasing the acceptor concentration in lanthanum chromites would result in increased expansion upon reduction as the higher initial concentration of Cr^{4+} is reduced to Cr^{3+}, resulting in an increased concentration of oxygen vacancies in the lattice.

The removal of oxygen from the perovskite lattice results in an increase in lattice cell dimensions, and, therefore, an increase in specimen volume as indicated by Srilomsak et al. (4) and Armstrong et al. (6). The observed expansion may result from the increase in cation size accompanying the reduction of Cr^{4+} to Cr^{3+} and/or an electrostatic repulsive forces between adjacent Cr cations separated by oxygen vacancies.

Comparison of the LSC-30, LCC-30 and LSC-20 and LCC-20 expansion data is shown in Figure 5. In both cases the Ca doped lanthanum chromite expands more than the Sr doped lanthanum chromite. Studies are in progress to determine the cause of this difference in behavior. It is possible that, in the same reducing atmosphere, LSCs are more stable toward oxygen loss than the LCCs. Alternatively, the oxygen loss in a given atmosphere may be the same for both materials, but the LSC lattice may expand less than the LCC lattice.

Figure 6 shows data for linear expansion as a function of dopant content from this study and from Schafer and Schmidberger (9) in 10^{-15} atm $P(O_2)$ at 1000°C. In both studies, it is apparent that the relationship between expansion and dopant content was approximately linear over a wide range of composition. The relationship between sample expansion and dopant concentration (denoted by the slope) is 0.013% expansion/mole % dopant for LSC and 0.015% expansion /mole% dopant for LCC materials. It is clear, however, (from the fact that the linear fits shown in Figure 8 have negative y-intercepts) that this linearity cannot hold for very low dopant levels. The data presented in Figure 8 show very good agreement over the entire composition range for the LSC materials. However, there is a large disparity between the expansion measurements for the two sets of LCC data presented in this figure. The data presented by the authors are similar to that of LSC. The data by Schafer and Schmidberger (2) show a significant deviation. While still linear, their data shows nearly a 1:1 correlation between the expansion and the Ca concentration. The data of Schafer more closely match the expansion data at 10^{-18} atmospheres obtained in the present investigation.

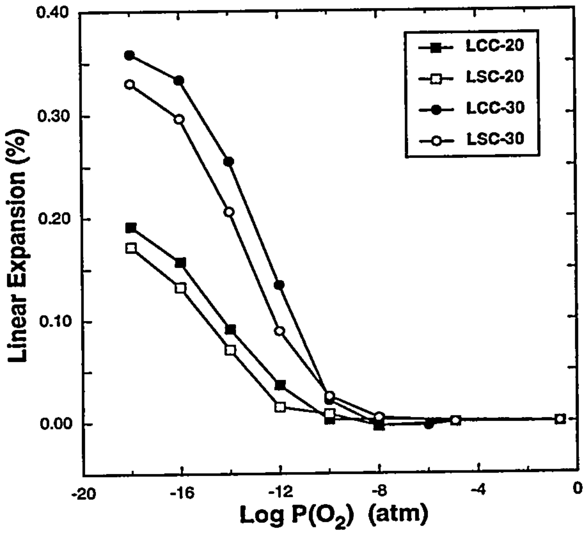

Figure 5. Comparison of Linear Expansion Data of 20 and 30 Mole Percent Doped LSC and LCC at 1000°C.

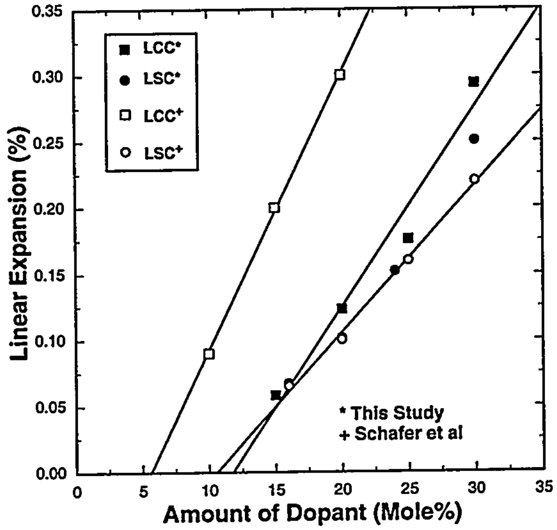

Figure 6. Maximum Linear Expansion at 10^{-15} atm (1000°C) for LSC and LCC compared to Similar Data From Ref. (4).

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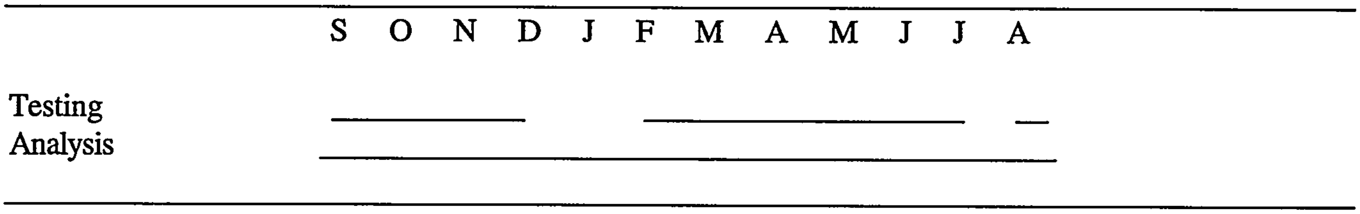
2.4 Pressurized Solid Oxide Fuel Cell Testing

CONTRACT INFORMATION

Contract Number	DE-FC21-91MC28055
Contractor	Westinghouse Electric Corporation 1310 Beulah Road Pittsburgh, PA 15235-5098 (412)256-5313 (telephone) (412)256-2002 (telefax)
Contractor Project Manager	Mr. Emerson R. Ray
Principal Investigators	R. A. Basel and J. F. Pierre
METC Project Manager	Mr. Bruce Harrington
Period of Performance	April 1, 1991 Nov. 30, 1995

Schedule and Milestones

FY1995 Program Schedule



CONTRACT INFORMATION

A cooperative test program between the United States Department of Energy (DOE), Westinghouse, and two customer consortiums is focused on the testing of the tubular solid oxide fuel cell (SOFC) at elevated pressures. The design of a pressurized test facility (PTF) was funded by DOE and Westinghouse. Construction and operation of the PTF, which is located at Ontario Hydro Technologies, Toronto, Canada was funded by a consortium of Canadian utilities and agencies. DOE provides SOFC's and test articles. Operational consultation and data analysis is provided by Westinghouse with financial support from Westinghouse, DOE and a consortium of Japanese electric power companies and NEDO.

The cells and test articles are returned to Westinghouse at the completion of testing.

The Pressurized SOFC Test Program is an integral part of the Cooperative Agreement between Westinghouse and DOE and was put into place to evaluate the effects of pressurization on SOFC performance.

OBJECTIVES

The goals of the SOFC pressurized test program are to obtain cell voltage versus current (VI) performance data as a function of pressure; to evaluate the effects of operating parameters such as temperature, air stoichiometry, and fuel utilization on cell performance, and to demonstrate long term stability of the SOFC materials at elevated pressures.

BACKGROUND INFORMATION

The high temperature Westinghouse tubular SOFC is an electric power generation technology that electrochemically generates electricity at high efficiencies in a clean, environmentally benign process. The technology has progressed to the stage where atmospheric pressure SOFCs have demonstrated operating efficiencies above 50% and the potential for direct utilization of pipeline natural gas (PNG), naphtha, and coal derived fuel gas. In parallel with the technology development work being performed by Westinghouse under the sponsorship of DOE, Westinghouse has completed a number of system studies for DOE and foreign and domestic utilities. These studies indicate that atmospheric pressure SOFC systems have the potential to be economically competitive with and environmentally superior to gas turbines and combined cycles. However, the pressurized technology provides additional performance/ economic advantages which will expand the economically competitive market range and increases the likelihood of investment by utilities in a new technology.

The benefits of pressurization include improved cell performance, (increased power output, efficiency > 60%), smaller equipment sizes, and reduced heat losses and pressure drops. In combination these benefits will yield a lower cost of electricity for SOFC systems. Ongoing system studies indicate that relative to an atmospheric pressure SOFC system, performance should increase by approximately ten percent and the cost-of-electricity may decrease by ten percent for a pressurized SOFC system. These studies are based on predictions of the cell voltage increase that may be realized via pressurizing the SOFC technology.

PROJECT DESCRIPTION

In 1992 Westinghouse and Ontario Hydro Technologies (OHT) entered into a joint program to design, construct, and operate a pressurized SOFC test facility (PTF). The PTF, located at OHT's Kipling Avenue Facility in Toronto, Canada, consists of two independent test stands as shown in Figure 1. Each stand can hold an array of one to four cells with active lengths up to 200 cm. The stands are fully instrumented, have multi-fuel capability, and are capable of operating at pressures up to 15 atm.

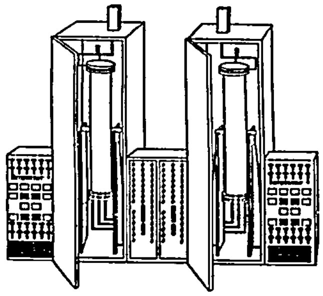


Fig. 1. SOFC Pressurized Test Facility

Figure 2 depicts the pressurized test article (PTA). The SOFCs are located within an alumina muffle tube which in turn is surrounded by a five zone electric heater assembly. The heater and muffle assembly is covered with alumina insulation and alumina sheath. A natural gas reformer inside the muffle tube permits operation on pipeline natural gas. The fully assembled PTA is delivered to OHT and installed into the pressure vessel. Flanges at both the top and bottom of the pressure vessel are fitted with penetrations for fuel, air, and exhaust flows, electrical power, and instrumentation leads.

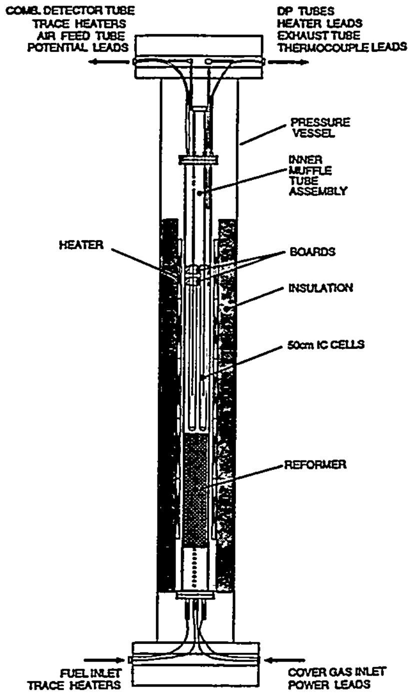


Fig. 2. Pressurized SOFC Test Article

Table 1 delineates the areas of responsibility for Westinghouse and OHT and its funding partners. Title to the SOFCs remains vested with DOE/Westinghouse.

Table 1. Westinghouse/OHT Pressurized SOFC Program

Scope of Supply

- **Westinghouse**
  - Facility Specifications
  - Test Stand Design
  - Test Envelope Design
  - Operating Procedures
  - Training
  - Test Plans
  - Consultation
  - SOFC Test Articles
  - Data Analysis
- **Ontario Hydro**
  - Facility to House Test Stand
  - Test Stand Equipment Design
  - Fabrication and Construction
  - Shakedown
  - Data Acquisition System
  - Control System
  - Operation
  - Consumables

RESULTS

Three PTAs have been tested in the PTF since March 1994. The first two, PTA-400 and PTA-405, served to shakedown the facility. The results discussed herein were obtained using the third test article PTA-443.

PTA-443 contains four 50 cm active length, air electrode supported (AES) cells. These cells are typical of those employed in the present SOFC field units. After an initial test period at Westinghouse, PTA-443 was installed in the PTF in September 1994. The test parameters for the pre-test were replicated at the PTF to verify instrumentation and to ensure that the PTA was not damaged during transport and installation.

Open circuit voltage (OCV) measurements at pressures up to 15 atm were taken and found to be consistent with theoretical predictions (Figures 3 and 4).

In Figure 3, the measured open circuit voltages are slightly less than the theoretical value. Figure 4 shows the open circuit voltage increase in going from 1 atm operating pressure up to 15 atm operating pressure. It can be seen that the measured voltage increases are in good agreement with the theoretical values.

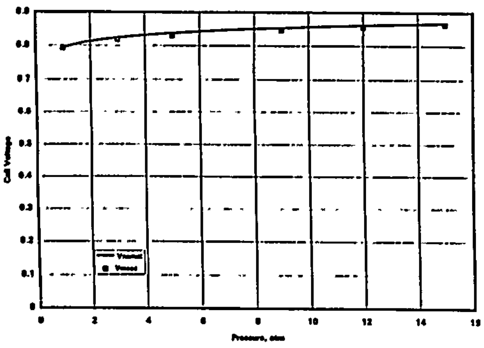


Fig. 3. Open Circuit Voltage for PTA-443

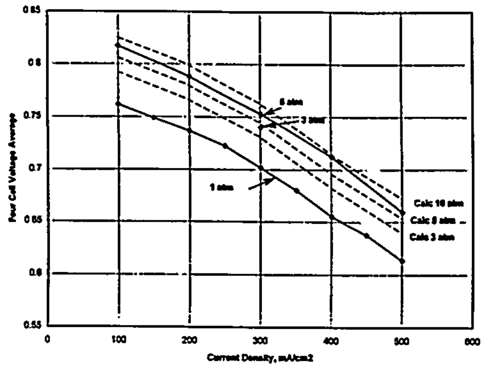


Fig. 5. Four Cell Average Voltage vs Current Density With H_2O Fuel, 85% Fuel Consumption and 6 Air Stoichs

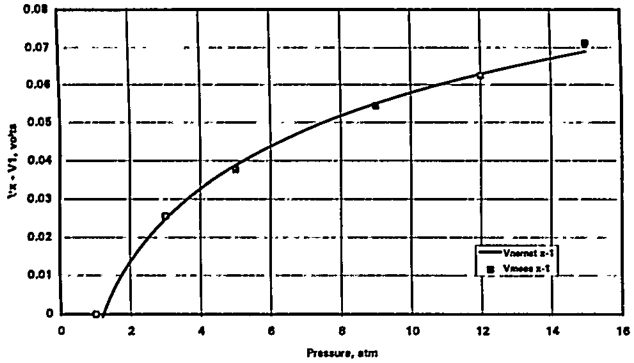


Fig. 4. Comparison of Measure vs Predicted Open Circuit Voltage

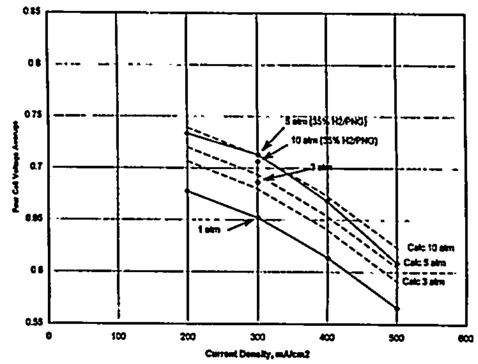


Fig. 6. Four Cell Average Voltage vs Current Density With PNG Fuel, O:C = 4.0 85% Fuel Consumption and 6 Air Stoichs

Voltage versus current density (V-J) curves were obtained at 1 and 5 atm on H2 fuel and PNG fuel as shown in Figures 5 and 6. The calculated values for 3, 5, and 10 atm were obtained by adding the expected increase in the Nernst potential due to increased pressure to the measured voltage at 1 atm. The measured results are in good agreement up to 5 atm. V-J curves at pressures above 5 atm will be obtained in future testing.

The test article was operated for a total of 5000 hours and 5 start/stop thermal cycles. Of the 5000 hours, 1515 hours were spent at 10 atm operating on PNG. The 1515 hours were divided into two portions of 265 hours and 1250 hours due to a scheduled site power outage. Prior to each shutdown the 1 atm V-J curve and the 300 mA/cm2 point at 5 atm were repeated with H2 fuel to assess the condition of the cells. The results are shown in Figures 7 through 10. It can be seen that after 265 hours there was virtually no change in the voltage of all four cells. After 1000 hours one half of the top guard heater zone failed making it impossible to maintain all four cells at a uniform 1000 C, thus making assessment of the cell condition difficult. Figures 7 through 10 show that after 1515 hours at 10 atm cells 3 and 4 show no change in voltage, but cells 1 and 2 show reduced voltage as current increases. The shape of the 7/12/95 V-J curve for cells 1 and 2 is typical of what would be expected if the cell temperatures were lower than for the previous curves. The fact that cells 3 and 4 show the same V-J behavior as before suggests that cells 3 and 4 are still at 1000 C and that the failed heater (each zone consists of two half clam shell cylindrical heaters) is on the cell 1 and 2 side. A post test V-J to assess the condition of the cells will be performed when the test article is returned to Westinghouse.

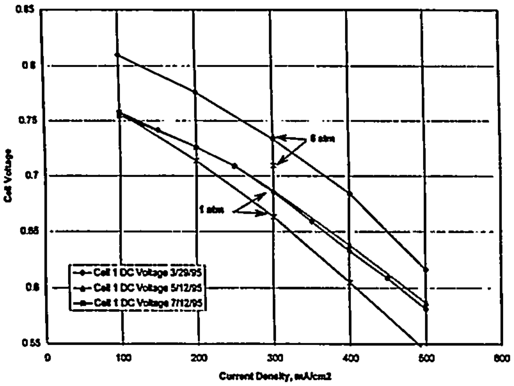


Fig. 7. Cell 1 Voltage vs Current Density With H_2 Fuel, 85% Fuel Consumption And 6 Air Stoichs

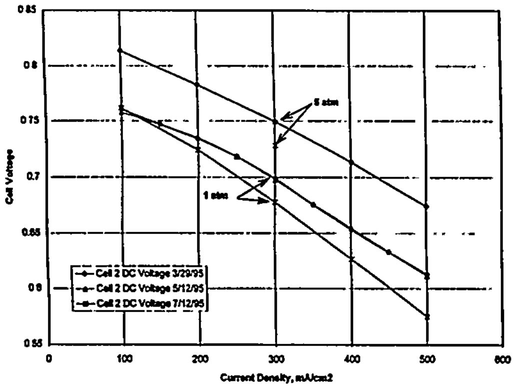


Fig. 8. Cell 2 Voltage vs Current Density With H_2 Fuel, 85% Fuel Consumption And 6 Air Stoichs

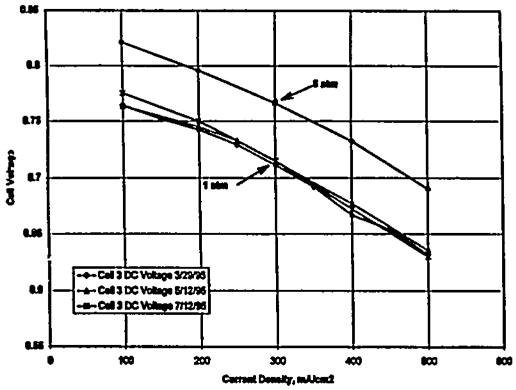


Fig. 9. Cell 3 Voltage vs Current Density With H_2 Fuel, 85% Fuel Consumption And 6 Air Stoichs

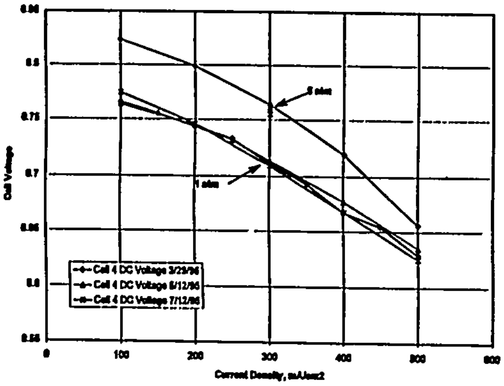


Fig. 10. Cell 4 Voltage vs Current Density With H_2 Fuel, 85% Fuel Consumption And 6 Air Stoichs

Test Article exhaust gas NO_x levels were obtained before the heater failure. Measurements were made using a hand pump/detector tube system. The results are listed in Table 2.

Table 2. Pressurized SOFC Exhaust Gas NO_x Measurements 6 Air Stoichs

	0.80 F.C.	0.75 F.C.
10 atm	1.0 ppm	2 ppm
5 atm	0.5 ppm	--
1 atm	Trace (~0.02 ppm)	--

CONCLUSIONS

Tubular solid oxide fuel cells have successfully operated on reformed natural gas for up to 1500 hours at 10 atm. V-J performance is as expected up to 5 atm. NO_x emissions are 2 ppm or less up to 10 atm. Further test articles are planned.

ACKNOWLEDGMENT

Westinghouse gratefully acknowledges the support of The United States Department of Energy - Morgantown Energy Technical Center for the continued development of the tubular SOFC. Westinghouse also acknowledges the support of the New Energy and Industrial Development Organization; The Hokkaido Electric Power Company; the Tokyo Electric Power Company; Chubu Electric Power Company; Chugoku Electric Power Company; Kyushu Electric Power Company, and the Electric Power Development Company for their financial and programmatic support of the Pressurized SOFC Test Program. The support of Ontario Hydro Technologies, the Canadian Electrical Association, Natural Resources Canada, Gas Technology Canada, Consumers Gas, Union Gas, British Gas, and the Ontario Ministry of Environment and Energy is gratefully acknowledged. A particular acknowledgment and note of gratitude is due to the personnel of Ontario Hydro Technologies for their competent and dedicated efforts in the installation and operation of the Pressurized Test Facility.

Session 3

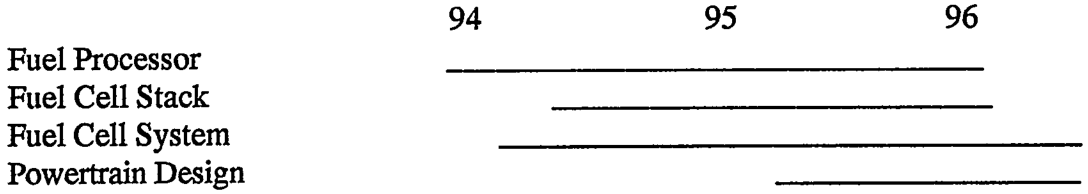
DOE's Transportation Program

3.2 General Motors Automotive Fuel Cell Program

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	October 1994 to June 1995

PROGRAM SCHEDULE (Phase II)



OBJECTIVES

The objectives of the second phase of the GM/DoE fuel cell program is to develop and test a 30 kW fuel cell powerplant. This powerplant will be based on a methanol fuel processor and a proton exchange membrane (PEM) fuel cell stack. In addition, the 10 kW system developed during phase I will be used as a "mule" to test automotive components and other ancillaries needed for transient operation.

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. GM units involved are GM R&D center as prime contractor, Delphi Energy and Engine Mangement Systems, Delphi Harrison, Delphi Packard, and Delco Electronics. Other participants in the program are Los Alamos National Labs, Dow Chemical Co., Ballard Power Systems and DuPont Co. During 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

The GM team is focussed on three major tasks, Fuel Processor, Fuel cell Stack, and Fuel cell System Integration. The fuel processor team is involved in the development of combustors that can operate on both methanol and dilute hydrogen from the anode exhaust, and optimized catalysts for the various fuel processor reactors. Fuel processor reactors under development are the recirculating and plug-flow type reformers, water gas shifter and an isothermal PrOX. Sensors needed for the control and diagnostics of the various reactors are also under development. The fuel cell stack team is involved in the design and testing of various stack components, leading to the design, fabrication and testing of advanced 30 kW PEM stacks for automotive applications. The fuel cell system team is involved in the development of ancillaries required to integrate the fuel processor and the fuel cell stack in to a complete automotive powerplant.

RESULTS

Fuel Processor. Reactors under development are a combustor, plug-flow and stirred reformers, shifter and PrOX. Testing is complete for a combustor that has been adapted for methanol combustion. A 10 kW combustor will also be modified to burn dilute hydrogen from the anode exhaust. A 30 kW combustor is being designed to avoid NOx formation.

Several new catalysts were evaluated for the methanol reforming reaction in a test reactor. One of the catalysts showed almost twice the performance of previously studied catalysts, as shown in Fig. 1. Increasing the steam/methanol ratio from 1 to 1.5 increases the reaction

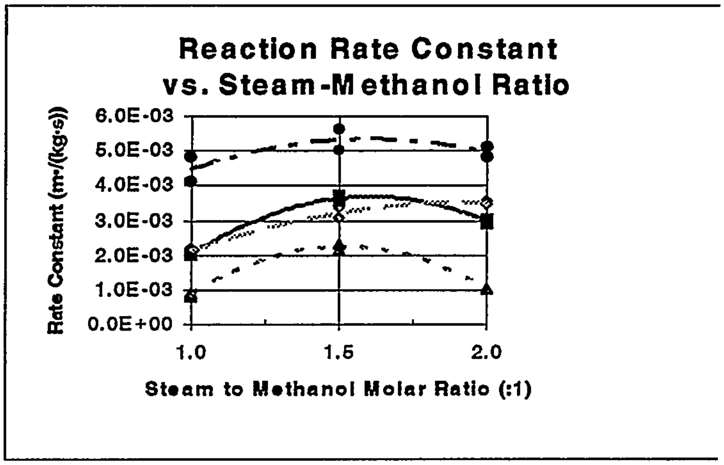


Figure 1. Methanol reforming reaction rate constants for various catalysts

rate, whereas increasing the ratio from 1.5 to 2.0 led to a decrease in rate for most of the catalysts. The effect of pressure, including the partial pressures of hydrogen and CO, has also been studied. Major contaminants that can deactivate the catalysts have been identified. Three types of reformers have been built for testing at the 10 kW level: adiabatic and isothermal plug flow reformers and a combustor-heated stirred reformer.

Fuel Cell Stack. The goal of the stack engineering efforts is the development of an efficient, rugged, low-cost stack suitable for automotive applications. Activities cover development of membranes, membrane/electrode assemblies, bipolar plates, single cell and stack testing, and electrochemical modeling.

Dow and Dupont are working on membrane process research and effecting performance improvements. Dow has synthesized two polymers required for the testing program and has improved the yield during polymer synthesis. DuPont has developed cutting techniques using water-jet processing to eliminate contamination of thin membranes such as Nafion 112. Membrane/electrode fabrication used at GM R&D, LANL and DuPont have followed their earlier patented approaches. The lamination technique used by GM R & D has been improved by suitable optimization of the Nafion and Teflon contents in the catalyst and the gas diffusion layer regions, respectively. The decal transfer method developed at LANL has been improved with major advances in electrode fabrication resulting in >96% decal transfer to a variety of membranes with excellent reproducibility. Single cell testing includes electrodes with varying sizes and platinum loadings. First single cells for the GM GEN1 stack are now undergoing tests. Also, 5 kW Ballard stacks delivered during phase I continue to be tested for reformate operation.

The presence of CO_2 in the fuel mixture not only affects the performance of the anode catalysts but also affects the water balance in the stack. CO_2 causes a large increase in water collected from the anode, probably because the anode gas now has a higher momentum to pick up more water vapor. An increase in stack pressure caused the cathode water to decrease, probably due to a lower volumetric flow rate at higher pressures.

System Integration. Work continues on incorporating more automotive components in the 10 kW system developed during phase I. Examples of such changes are the replacement of a computer-controlled anode exhaust valve with a simple mechanical back-pressure regulator, and the use of various types of temperature and pressure sensors and

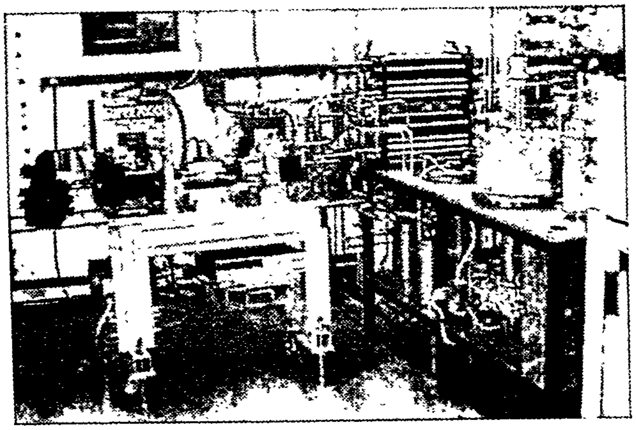


Fig. 2 The 10 kW system including the integrated air compressor/expander

fuel/water injectors. Fig. 2 shows the fully integrated 10 kW system, including fuel cell stacks operating on reformate from the fuel processor and air compression /expansion provided by the Powerex scroll set. Significant progress has been made in specifying the sensors, drivers, actuators ans control functions for the 30 kW system, and hardware design of the Delco Electronics controller for the 30 kW system has been completed.

FUTURE WORK

The 30 kW combustor and reformer will be designed and built by the end of 95, but work on alternative concepts such as the plug-flow reformer will be continued. The Ballard Gen I stack with a 30 kW peak power and an advanced stack with a peak power of 50 kW will be tested next year. Finally, the system team will integrate the fuel processor and the advanced PEM fuel cell stack with necessary support systems and components, and also test the capability of the system to meet required vehicular performance characteristics.

3.3 DOE/FORD Fuel Cell Contract for Automotive Application

CONTRACT INFORMATION

Contract Number	DE-AC02-94CE50389
Contractor	Research Laboratory FORD MOTOR COMPANY P.O. Box 2053, M/D 3439 Dearborn, MI 48121-2053 (313)322-6646 (telephone) (313) 845-4495 (FAX)
Principal Investigator	Djong-Gie Oei
Period of Performance	July 1, 1994 to December 31, 1996

OBJECTIVES

The objectives of the contract are twofold. The first objective is to assess the feasibility of using a direct hydrogen fueled PEM fuel cell engine to power a midsize passenger car through the various drive cycles and test such a propulsion system on a test bed. The second objective is to study the supply infrastructure and safety aspects of hydrogen for future practical implementation of PEM fuel cells.

BACKGROUND INFORMATION

There are several important reasons for developing the direct hydrogen-fueled PEMFC engine. First, hydrogen can be generated from non-petroleum based sources and may ultimately provide a fuel, when used with fuel cells, capable of lower cost per vehicle-mile than gasoline. Using hydrogen as a fuel also leads to a lower dependency on a petroleum based economy. Second and equally important, the direct hydrogen-fueled PEMFC engine is a zero emission vehicle (ZEV) engine. It is also well established that at partial load a fuel cell has a much higher efficiency than a Diesel or internal combustion (IC) engine. And last, but not least, the results of the contract will be an important contribution to the Partnership for a New Generation of Vehicles (PNGV) program.

PROJECT DESCRIPTION

The DOE/FORD Fuel Cell Contract consists of two equally important aspects of the direct hydrogen-fueled PEMFC for automotive applications. First is the development of a PEMFC engine that is suitable for powering a midsize (Taurus) car with a range of 560 km (350 mi). This goal is to be achieved in two phases, with the delivery of a 10 to 12 kW fuel cell stack or system in Phase I, and after careful selection, the delivery of a 50 kW fuel cell system in Phase II. Requirements of power density, platinum-catalyst loading, operational pressure, humidification, and others are specified.

Another important part of the contract is the hydrogen supply infrastructure study. The delivery of a compressed hydrogen gas fuel tank, assessment of hydrogen supply and distribution, and safety aspects studies are included in this effort. A preliminary conceptual propulsion system design will be reported in the first phase of the project.

RESULTS

Power requirements for a pure fuel cell powered vehicle were determined for a small size (ASPIRE), a midsize passenger vehicle (TAURUS) and a van (E-150 ECONOLINE). These requirements were derived from a vehicle simulation program developed by Ford. The weight and size of the fuel cell system were determined using a computer simulation model developed by Directed Technologies Incorporated (DTI) as part of an FC propulsion system packaging study.

Letters of consent from the DOE Chicago Office to start the contract with the fuel cell suppliers were issued at different dates, and therefore, the results obtained depended on their contract start date. The following list shows the fuel cell subcontractors and the date of contract award:

H Power Corp. - December 19, 1994
Energy Partners - January 11, 1995
International Fuel Cells - March 13, 1995
MTI - May 2, 1995
TECOGEN - May 2, 1995

DTI is the coordinator for the hydrogen supply infrastructure and safety related studies. Their contract was awarded on October 11, 1994. The participants to this contract and their contract award dates are:

Praxair - January 24, 1995
Air Products & Chemicals - March 13, 1995
Electrolyser - April 24, 1995
EDO Fiber Science - May 10, 1995
BOC Gases - May 23, 1995
Aero Tec Lab - May 23, 1995

FUTURE WORK

A final conceptual FC propulsion system design will be developed following a favorable review of the preliminary design report by DOE.

At the end of the tenth month of their contract, each fuel cell subcontractor must deliver a nominal 10kW fuel cell stack or system meeting the following minimum requirements:

1) Pt-catalyst loading ≤ 0.25 mg/cm^2,
2) stack power density ≤ 3.7 kg/kW
3) air pressure = 3.04 atm (30 psig)

At the end of Phase II of the contract one or two selected fuel cell supplier for Phase II are to deliver a 50kW fuel cell system. The requirements for this system are:

1) Pt-catalyst loading ≤ 0.25 mg/cm^2
2) system power density ≤ 2.7 kg/kW
3) air pressure ≤ 3.04 atm (30psig)

DTI, coordinator of the hydrogen supply infrastructure and safety investigation, will issue a final report of its findings. An onboard compressed hydrogen gas storage system will be delivered to FORD during this phase of the contract for testing and evaluation.

From the results of experimental verification and testing of the fuel cell, along with the related hydrogen supply infrastructure studies, a report will be issued on the viability of the fuel cell engine for automotive applications. It is also intended that this report will give better indications of the prospects of mass production of fuel cell systems and hence early cost estimates for fuel cell powered vehicles.

3.4 Chrysler Pentastar Direct Hydrogen Fuel Cell Program

CONTRACT INFORMATION

Contract Number	DE-AC01-94CE50390
Contractor	Pentastar Electronics, Inc. P.O. Box 1929 Huntsville, AL 35807-0929 (205) 895-2051 (205) 895-2356
Contractor Project Manager	Daniel H. Tudor
Principal Investigators	Michael Kimble Derrell Deloney
METC Project Manager	Robert Kost
Period of Performance	July 1, 1994 to December 31, 1996

ABSTRACT

The Chrysler Pentastar Electronics, Inc. Direct Hydrogen Fueled PEM Fuel Cell Hybrid Vehicle Program (DPHV) was initiated 1 July, 1994 with the following mission,

> "Design, fabricate, and test a Direct Hydrogen Fueled Proton Exchange Membrane (PEM) Fuel Cell System including onboard hydrogen storage, an efficient lightweight fuel cell, a gas management system, peak power augmentation and a complete system controls that can be economically mass produced and comply with all safety environmental and consumer requirements for vehicle applications for the 21st century."

The Conceptual Design for the entire system based upon the selection of an applicable vehicle and performance requirements that are consistent with the PNGV goals will be discussed.

A Hydrogen Storage system that has been selected, packaged, and partially tested in accordance with perceived Hydrogen Safety and Infrastructure requirements will be discussed in addition to our Fuel Cell approach along with design of the "real" module.

The Gas Management System and the Load Leveling System have been designed and the software programs have been developed and will be discussed along with a complete fuel cell test station that has the capability to test up to a 60 kW fuel cell system.

Future plans call for the integration and testing of a complete 50 kW direct hydrogen PEM fuel cell system prior to the end of 1996. If the system test is promising, there maybe the possibility of putting the system in a "mule" vehicle, funding permitting.

Session 4

ARPA Advanced Fuel Cell Development

4.1 ARPA Advanced Fuel Cell Development

Lawrence H. Dubois
Advanced Research Projects Agency

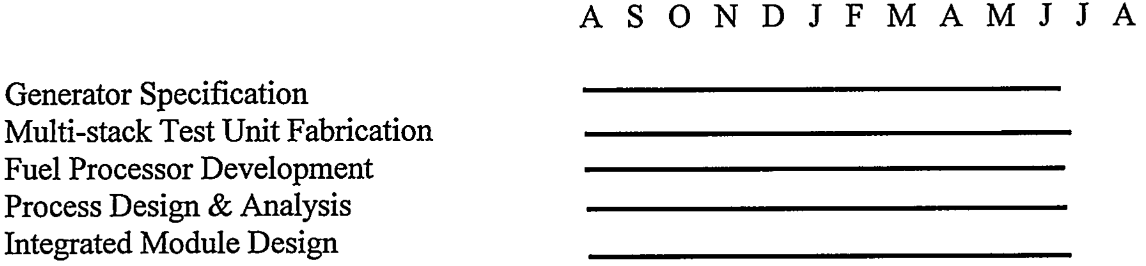
ABSTRACT

Fuel cell technology is currently being developed at the Advanced Research Projects Agency (ARPA) for several Department of Defense applications where its inherent advantages such as environmental compatibility, high efficiency, and low noise and vibration are overwhelmingly important. These applications range from man-portable power systems of only a few watts output (e.g., for microclimate cooling and as direct battery replacements) to multimegawatt fixed base systems. The ultimate goal of the ARPA program is to develop an efficient, low-temperature fuel cell power system that operates directly on a military logistics fuel (e.g., DF-2 or JP-8). The absence of a fuel reformer will reduce the size, weight, cost, and complexity of such a unit as well as increase its reliability. In order to reach this goal, ARPA is taking a two-fold, intermediate time-frame approach to: (1) develop a viable, low-temperature proton exchange membrane (PEM) fuel cell that operates directly on a simple hydrocarbon fuel (e.g., methanol or trimethoxymethane) and (2) demonstrate a thermally integrated fuel processor/fuel cell power system operating on a military logistics fuel. This latter program involves solid oxide (SOFC), molten carbonate (MCFC), and phosphoric acid (PAFC) fuel cell technologies and concentrates on the development of efficient fuel processors, impurity scrubbers, and systems integration. A complementary program to develop high performance, light weight H_2/air PEM and SOFC fuel cell stacks is also underway. Several recent successes of these programs will be highlighted.

4.2 SOFCo Mobile Planar Solid Oxide Generator

CONTRACT INFORMATION

Contract Number	DAAH04-94-C-0012
Contractor	Babcock & Wilcox, a McDermott Company Contract Research Division 2220 Langhorne Road P.O. Box 11435 Lynchburg, VA 24506-1435 (804) 948-4709 (telephone) (804) 948-4700 (telefax)
Other Funding Sources	SOFCo, a Babcock & Wilcox/Ceramatec Company
Contractor Project Manager	L. G. Weatherford
Principal Investigators	A.C. Khandkar, R.M. Privette
U.S. Army Research Office Program Manager	D. Paur
ARPA Program Manager	L. Dubois
Period of Performance	August 1994 through June 1995
Schedule and Milestones	FY95 Program Phase 1 Schedule



OBJECTIVES

This paper presents results from the first phase of a three phase, four-year program with the objective of designing and demonstrating a 10 kW mobile electric power generator operating on logistic fuel. Objectives of the first phase include: the development of a preliminary system design, an assessment of technologies critical to system performance, and the fabrication of three multi-stack test units.

BACKGROUND INFORMATION

This program seeks to incorporate planar solid oxide fuel cells (SOFC) into a compact, integrated generator module operating on military logistic fuel. The integration of fuel processing equipment necessary to reform logistic fuel with SOFC stacks into a compact hardware configuration represents a significant advance in the state of the art in SOFC system technology.

SOFCo, a Babcock & Wilcox/Ceramatec company, plans to design a mobile generator using the patented Ceramatec planar SOFC CPn, staged oxidation design as a basis. The CPn design will be integrated with a proprietary steam reforming catalyst developed by the Institute of Gas Technology (IGT) and the Gas Research Institute (GRI). The integration will be done using the extensive Babcock & Wilcox (B&W) power generation system design experience.

The Ceramatec planar SOFC technology combines state-of-the-art SOFC materials with mature ceramic fabrication processes, allowing independent optimization of materials and processing parameters to achieve reliable, high-performance SOFC stacks. Major technical milestones achieved in planar SOFC technology development at Ceramatec are highlighted in Figure 1.

A key aspect of this program is that military power systems must operate on logistic fuels (JP-8, DF-2). The use of sulfur-laden (0.3% by weight) logistic fuel requires processing to convert the fuel to a hydrogen-rich, low-sulfur gas for use in the fuel cell stacks. While conventional steam reforming, autothermal reforming and partial oxidation processes exist, they have limited application to the present work due to the relatively high sulfur content of the fuel, requirements for mobile operation, and/or undesirable dilution of the product hydrogen gas. SOFCo's preferred reforming approach uses the IGT Sulfur Tolerant Reformer (STR)[1]. The STR does not require the large equipment necessary for hydrodesulfurization, and gas dilution is avoided.

Thermal integration of key processes within an insulated generator enclosure is a key feature of the system. Heat from the fuel cells is used for both fuel reforming and preheating of incoming air. This approach results in high system efficiency, compact system design, and requires no external utilities for operation.

PROJECT DESCRIPTION

A summary of program activities is shown in Figure 2. Phase 1 focuses on: 1) the identification of a target military application and the development of a preliminary system design, 2) an assessment of IGT's STR for reforming DOD logistic fuel, and 3) the fabrication of three test units for conducting multistack tests in Phases 2 and 3 of the program.

Phase 2 will focus on developing a 2-4 kW non-integrated breadboard system employing all key elements of the generator design. Completion of the 10 kW generator design

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[1] A.L. Lee, "Catalytic Steam Reforming of Diesel Fuel for Fuel Cell Hydrogen Production." Final Report to Fort Belvoir, Contract No. DAAK70-86-P-0810 (1986).

and demonstration of a prototype system incorporating the integrated SOFC module concept will be performed in Phase 3.

RESULTS

The U.S. Army Lightweight Multipurpose Shelter (LMS) was chosen as the target application for this project (see Figure 3). The generator fits within a small tunnel, measuring approximately 29" x 25" x 84", near the front of the shelter. The selection was based on the Army's interest in an advanced power supply for the LMS, and the suitable match between the LMS power requirement and the demonstration size for the current program.

In parallel with the selection of the LMS application, candidate process designs were analyzed and conceptual hardware design layouts were completed. A single system was then selected from the candidate designs and refined.

Primary considerations for system designs included complexity and technical risk, overall system efficiency, and hardware size and weight. Two particular systems that were analyzed in depth are shown in Figures 3 and 4. The systems both incorporate steam reforming of the JP-8 fuel, and post-reforming removal of sulfur in a ZnO bed. The primary difference between the schemes is the method for obtaining the steam required for the reforming.

The first system, referred to as "Anode Gas Recycle", recycles a portion of the anode exhaust to supply steam for the reforming reaction. The recycle stream is mixed with the incoming JP-8 fuel prior to entering the reformer. The remaining anode exhaust is mixed with air from the cathode exhaust and burned to provide heat for both fuel reforming and air preheating operations.

The second system, termed "Anode Gas Condensation", incorporates a condenser in the fuel cell anode exhaust stream. The water vapor is first condensed, then separated and later reheated in a steam generator before ultimately being mixed with the incoming vaporized fuel. This system benefits from a smaller water vapor recycle stream and eliminates the need for a high temperature blower. It does, however, require the use of a condenser and small steam generator. Heat is provided to the steam generator by a burning a combination of anode exhaust and incoming JP-8 fuel.

Analyses of both systems were completed using the ASPEN PLUS[2] process simulator. The results of these analyses are shown in Table 1. As can be seen, both systems were projected to achieve 40+% system net efficiency. One significant difference was the steam-to-carbon ratio necessary within the reformer to avoid coking as predicted by an equilibrium analysis.

Since the "Anode Gas Recycle" scheme maintains the composition of the fuel cell anode exhaust in the recycle stream, the carbon remaining in the anode exhaust adds to the overall carbon content in the reformer stream. Thus, due to the "extra" carbon in the recycle stream, additional steam must be added to avoid conditions favorable for coke formation. Since the "Anode Gas Condensation" scheme separates the water vapor from the anode exhaust prior to mixing with the JP-8, the carbon content in the reformer stream is simply the content of the incoming JP-8.

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[2] Aspen Technology, Inc. Cambridge, Massachusetts

The additional steam required for the "Anode Gas Recycle" scheme, combined with the need for a small, high temperature blower were primary factors affecting the selection of the "Anode Gas Condensation" system for future development as shown in Table 2.

Following selection, the "Anode Gas Condensation" configuration was refined to more closely match reformer performance demonstrated during Phase I of this program, and to optimize heat integration between the system components. In particular, the following refinements were made:

- the steam-to-carbon ratio was increased from 1.8 to 2.5,
- a recuperative bayonet reformer design was incorporated,
- the air preheater was modified to provide easier access and better control, and
- the use of a vertical condenser design was evaluated.

Predicted performance of the refined system is shown in Table 3. Net power generation of 10.2 kW is expected at a net system efficiency of 40%. Reformer stream steam-to-carbon ratio is 2.5, and the system air flow is approximately 7 times the stoichiometric flow requirement (a result of stack cooling requirements).

A layout of "Anode Gas Condensation" configuration for the LMS application is shown in Figure 6. The layout is designed to allow easy access to key components through the two ends of the generator. Power conditioning equipment, air intake, and fuel storage are located together at one end of the system layout. Fuel processing equipment is centralized next to the fuel tank with desulfurization equipment located at the opposite end of the layout. This design provides simple access to the zinc oxide beds, fuel tank, and air inlet/exhaust. Not shown in Figure 6 is the insulation that surrounds the generator enclosure.

FUTURE WORK

While the preliminary design fits within the space envelope of the LMS, the weight of the SOFC system is higher than desired. During Phase 2, system design work will focus on reducing system weight and refining component sizing and performance estimates.

Phase 2 work will build on the results of Phase 1 through the breadboard demonstration of a 2-4 kW non-integrated SOFC generator. The breadboard generator will provide valuable experience on fuel processing system scaleup, and fuel cell stack performance operating on logistic fuel reformate. Demonstration of the breadboard system is scheduled for July, 1996.

ACKNOWLEDGEMENT

This work is sponsored by the U.S. Army Research Office. The content of the information does not necessarily reflect the position or the policy of the U.S. Government, and no official endoresement should be inferred.

Figure 1. Ceramatec technical milestones in planar SOFC technology

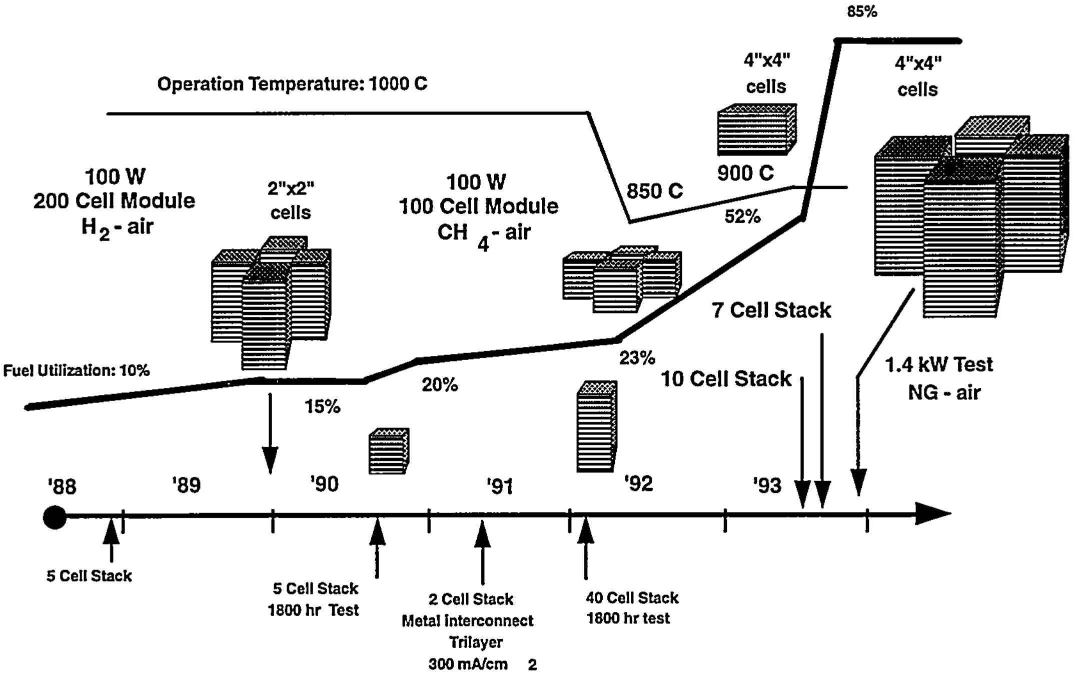
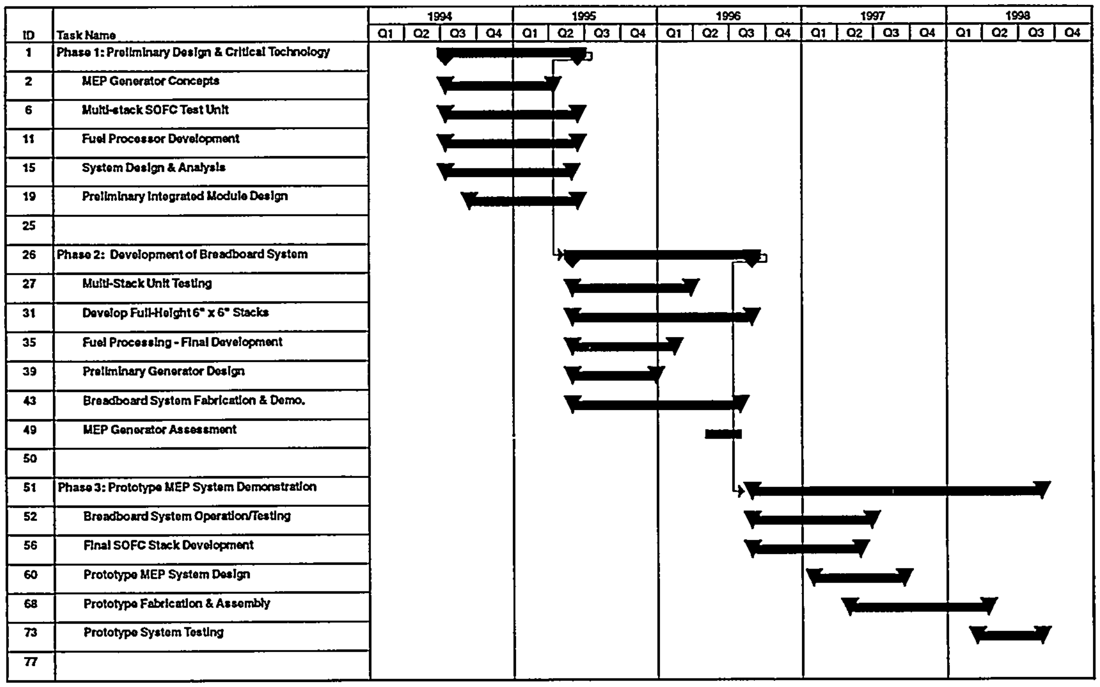


Figure 2. Program schedule



ID	Task Name
1	Phase 1: Preliminary Design & Critical Technology
2	MEP Generator Concepts
6	Multi-stack SOFC Test Unit
11	Fuel Processor Development
15	System Design & Analysis
19	Preliminary Integrated Module Design
25	
26	Phase 2: Development of Breadboard System
27	Multi-Stack Unit Testing
31	Develop Full-Height 6" x 6" Stacks
35	Fuel Processing - Final Development
39	Preliminary Generator Design
43	Breadboard System Fabrication & Demo.
49	MEP Generator Assessment
50	
51	Phase 3: Prototype MEP System Demonstration
52	Breadboard System Operation/Testing
56	Final SOFC Stack Development
60	Prototype MEP System Design
68	Prototype Fabrication & Assembly
73	Prototype System Testing
77	

Figure 3. U.S. Army lightweight multipurpose shelter

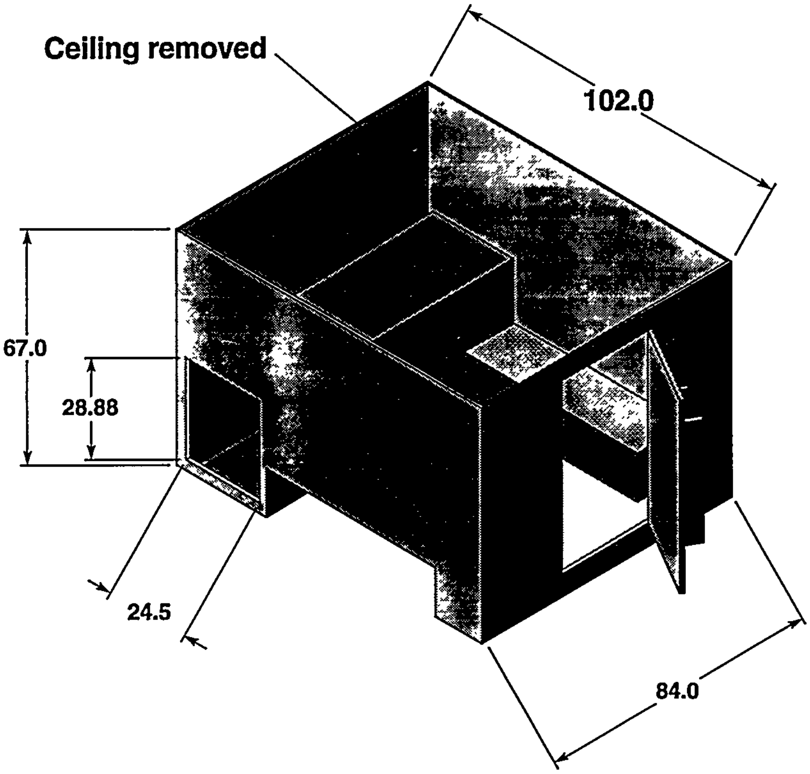


Figure 4. Anode gas recycle process schematic

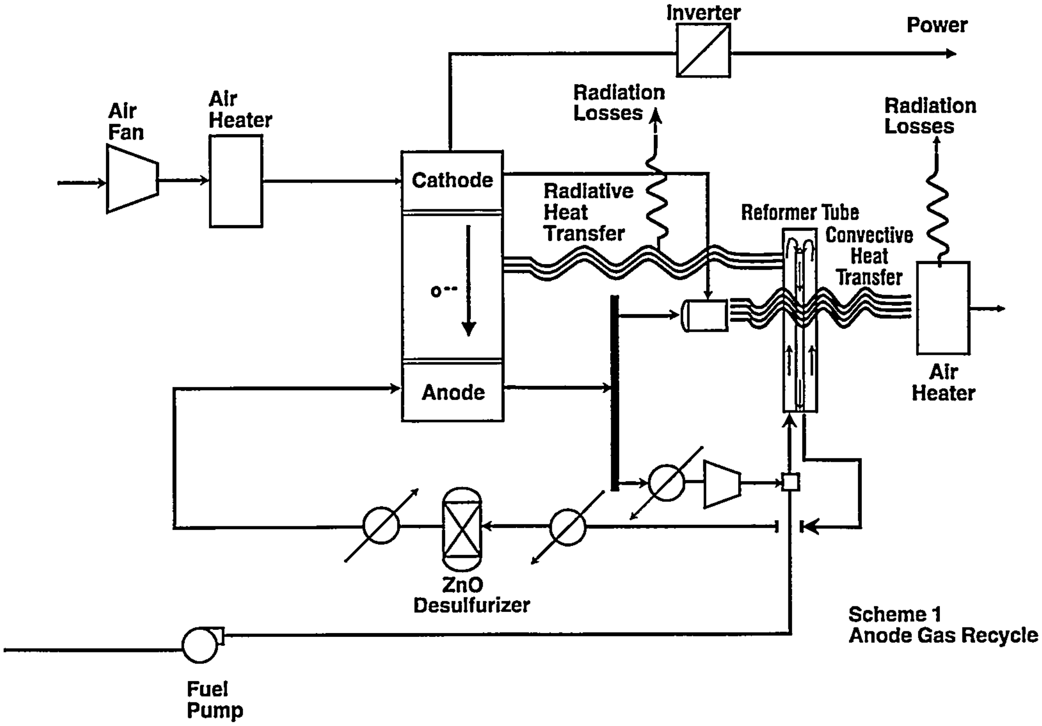


Figure 5. Anode gas condensation process schematic

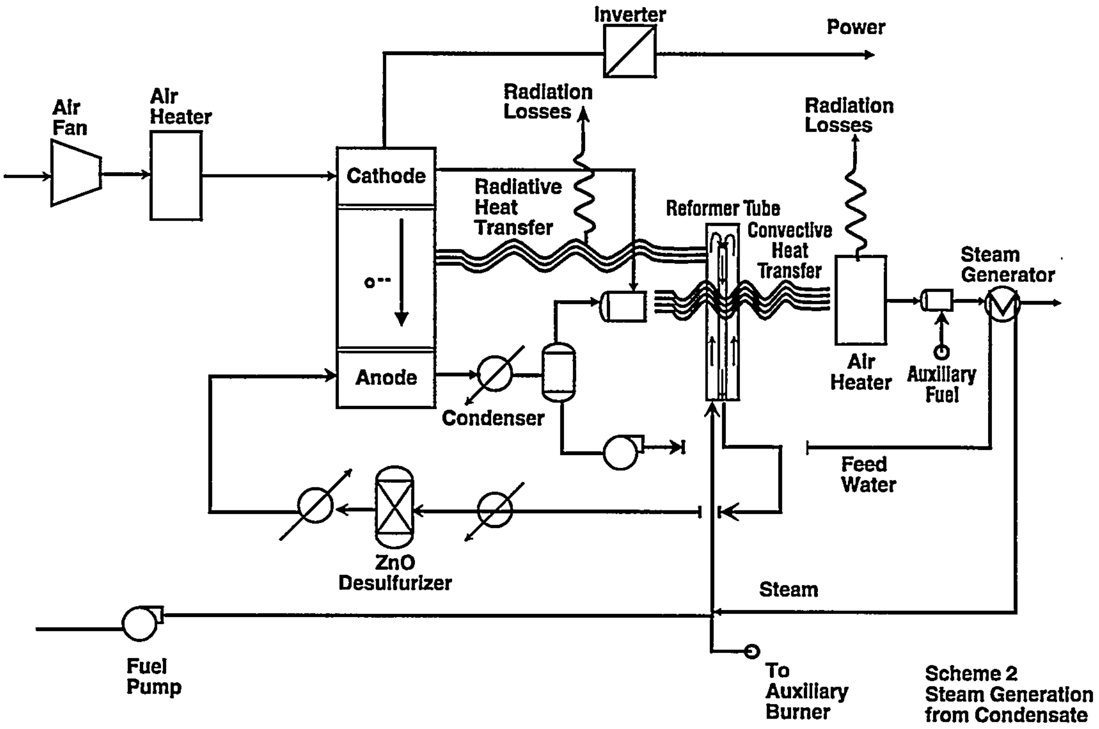


Figure 6. Layout of anode gas condensation system

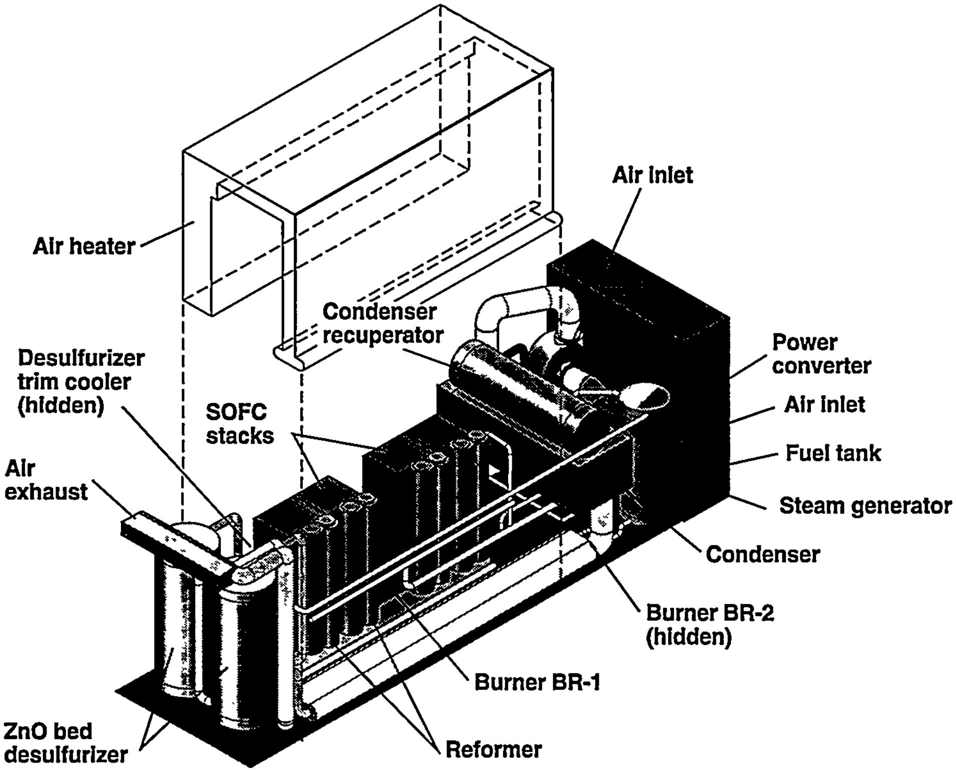


Table 1. Summary of candidate systems' performance

	Scheme 1 *Anode Gas Recycle*	*Scheme 2* *Anode Gas Condensation*
Net system efficiency (%)	45.30	43.80
Fuel flow (lbm / hr)	3.65	3.76
Steam-to-carbon ratio	1.80	1.80
Fuel utilization (%)	82.60	93.30
Air flow (times stoichiometric air)	7.60	7.20
Operating voltage (V_{op})	0.68	0.73
Down-select	Uncertain Catalyst Performance	

Table 2. System selection

Criteria	*Scheme 1*	*Scheme 2*
Size	1	1
Weight	1	2
Efficiency	1	2
Complexity	1	3
Operability	6	3
Risks	10	4
Total Score (Lower is better)	20	15

Table 3. Summary of refined system performance

	Scheme 2 *Anode Gas Condensation*
No. of cells	265
Net system efficiency	40.0
Net power	10.2
Fuel flow (lbm / hr)	4.13
Steam-to-carbon ratio	2.5
Fuel utilization	95.4
Air flow (times stoich.)	6.89
V_{op}	0.71

4.3

Fuel Cell Technology for Prototype Logistic Fuel Cell Mobile Systems

CONTRACT INFORMATION

Contract Number	DAAH04-94-C-0010
Contractor	International Fuel Cells Corporation P.O. Box 739 195 Governor's Highway South Windsor, CT 06074 Telephone: (203) 727-2267; FAX: (203) 727-2319
Other Funding Sources	
Contractor Project Manager	Craig R. Schroll
Principal Investigators	Richard A. Sederquist Jay Garow
ARPA Project Manager	Lawrence H. Dubois, Ph.D.
Period of Performance	June 1, 1994 through August 31, 1997

ABSTRACT

Under the aegis of the Advanced Research Project Agency's family of programs to develop advanced technology for dual use applications, International Fuel Cells Corporation (IFC) is conducting a 39 month program to develop an innovative system concept for DoD Mobile Electric Power (MEP) applications. The concept is to integrate two technologies, the phosphoric acid fuel cell (PAFC) with an auto-thermal reformer (ATR), into an efficient fuel cell power plant of nominally 100-kilowatt rating which operates on logistic fuels (JP-8). The ATR fuel processor is the key to meeting requirements for MEP (including weight, volume, reliability, maintainability, efficiency, and especially operation on logistic fuels); most of the effort is devoted to ATR development. An integrated demonstration test unit culminates the program and displays the benefits of the fuel cell system relative to the standard 100-kilowatt MEP diesel engine generator set. A successful test provides the basis for proceeding toward deployment. This paper describes the results of the first twelve months of activity during which specific program aims have remained firm.

The activity to date has been conducted under two separate tasks. In ***Task 1 - Design of 100-kW Logistic Fuel MEP System***, the requirements for the initial design of the MEP system were interpreted and expanded based on military user input. The requirements thus developed were used to provide a description

of the power system and its subsystems in sufficient detail to identify issues and options. These options were evaluated analytically and a reference system concept was selected. The performance characteristics, operational requirements, and control strategies of the reference system were established. These characteristics provide the basis for the subsystem and component functional requirements. Identification and development of components that satisfy the functional and operational requirements is underway and will allow definition of a preliminary packaging concept now in progress.

In ***Task 2 - Development and Verification of Fuel Processor***, the reforming catalyst developed in earlier IFC diesel-fuel programs was reformulated and verified suitable in laboratory confirmation testing. Definition of the fuel vaporizer and injecter designs was completed and a survey to identify an acceptable start/sustain burner for the ATR was initiated.

These activities are being conducted in concert with the definition of the conceptual design of the ATR. Design was completed to the point of retaining two options for evaluation; down-select will occur in the experimental phase of the program now underway.

Concurrent with ATR development, critical issues in the fuel processing subsystem such as sulfur removal and reformer-generated ammonia scrubbing are being addressed and candidate design concepts for fuel gas cleanup are being defined, for testing in the upcoming year.

4.4 The Direct Methanol Fuel Cell

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ABSTRACT

This presentation describes the approach and progress in the ARPA-sponsored effort to develop a Direct Methanol, Liquid-Feed Fuel Cell (DMLFFC) with a solid Polymer Electrolyte Membrane (PEM) for battery replacement in small portable applications. Using Membrane Electrode Assemblies (MEAs) developed by JPL and Giner, significant voltage was demonstrated at relatively high current densities. The DMLFFC utilizes a 3 percent aqueous solution of methanol that is oxidized directly in the anode (fuel) chamber and oxygen (air) in the cathode chamber to produce water and significant power. The only products are water and CO_2. Presently the catalyst loading is 4 mg/cm^2 of Pt/Ru on the anode and 4 mg/cm^2 of Pt on the cathode. At 95°C, the fuel cell exhibits 0.5 V at 300 mA/cm^2 with 30 psig of O_2, 0.4 V with 30 psig air. However, current densities as high as 600 mA/cm^2 can be sustained. At this performance level, a single 4" x 6" cell can produce 50 to 100 A.

The ARPA effort is aimed at replacing the battery in the BA 5590 military radio. The performance for this application is at a lower temperature and requires operation using low flow rate ambient air. To date the performance in a 2" x 2" cell using the 3 percent aqueous methanol has reached 0.43 V at 150 mA/cm^2 at 60°C with unpressurized air. The goal for this effort is 0.55 V under these conditions. Several areas of investigation are ongoing, e.g., effect of temperature, flow rate, methanol crossover under the various conditions.

From this it appears that the idea of a fuel cell battery for small applications, e.g., radios, cellular phones, and computers based on this technology appears to be feasible. Such a "fuel cell battery" would have several advantages over lithium batteries. These include elimination of costly and complex processes for recharging, replacement, storage, and disposal of the primary and rechargeable lithium batteries in use today. For long-duration applications, the "fuel cell batteries" would be potentially smaller and lighter than these batteries.

The development team is led by JPL and is concentrating on new materials, catalysts, and cell designs. The effort of Giner, Inc., is focused on assembly of hardware and test. The University of Southern California (USC) is developing new polymer electrolytes and evaluating alternate fuels. These efforts will result in (a) improved performance level, particularly efficiency, of the basic cell;
(b) implementation of innovative designs;
(c) establishment of a preliminary design of a DMLFFC version of a BA 5590 battery; and
(d) fabrication and test of a battery to demonstrate that its performance and size/weight are consistent with Department of Defense (DoD) needs.

Poster Session

P1 Sealant Materials for Solid Oxide Fuel Cells

CONTRACT INFORMATION

Contract Number	49638
Contractor	Argonne National Laboratory 9700 South Cass Avenue Argonne, Illinois 60439
Contractor Project Manager	Kevin M. Myles
Principal Investigator	Michael Krumpelt
Co-Investigators	Timothy W. Kueper and John H. Meiser
METC Project Manager	Bruce Harrington
Period of Performance	October 1, 1994 to July 1, 1995

OBJECTIVES

The objective of this work is to complete the development of soft glass-ceramic sealants for the solid oxide fuel cell (SOFC). Among other requirements, the materials must soften at the operation temperature of the fuel cell (600-1000°C) to relieve stresses between stack components, and their thermal expansions must be tailored to match those of the stack materials. Specific objectives included addressing the needs of industrial fuel cell developers, based on their evaluation of samples we supply, as well as working with commercial glass producers to achieve scaled-up production of the materials without changing their properties.

BACKGROUND INFORMATION

Argonne National Laboratory has been developing soft glass-ceramic sealant materials to meet the requirements of planar solid oxide fuel cells. Depending on the design, sealing may be required at the edges of the cells and at the junction of the stack and manifold. A successful sealant for these purposes must meet several demands in addition to relieving stress and matching the thermal expansion of the stack components. The sealant must be chemically compatible with the cell components and the cell environment, while at the same time forming dense, adherent bonds to the cell components. In addition, the material must have low gas permeability and, in manifold sealing applications, it must be electrically insulating.

In early work, many compositions in the five-component system, B_2O_3-SiO_2-

Al_2O_3-SrO-La_2O_3, were studied, and several of these were found to have promising characterisitics. Bonding behavior with SOFC cell components, interaction zones with the cell components, and thermal cyclability were all investigated, and no problems were detected. Thermal expansion and viscosity-temperature behavior were determined, and two compositions in particular were chosen for further development: a high-temperature sealant, Material K, has appropriate viscosity for use in 1000°C stacks and an expansion coefficient of 11.5×10^{-6}/°C, and a low-temperature sealant, Material 43, softens appropriately for use at 800°C and has an expansion coefficient of 9.5×10^{-6}/°C.

The sealing ability of Material K was tested by using it to seal the edges of an electrochemical cell, which consisted of a zirconia wafer and platinum electrodes, to a zirconia tube. When this device was operated as a fuel cell, all EMFs were measured to be greater than 99% of the theoretical values, even after thermal cycles between room temperature and 1000°C.

Other properties of the sealants that were tested in previous work included gas permeability and conductivity as a function of temperature and oxygen partial pressure. Material K had low permeabilities and a predominantly ionic conductivity of 10^{-3} S/cm at 1000°C. These values are suitable for fuel-cell sealing materials.

PROJECT DESCRIPTION

This year, we have continued to study sealant compositions in the same five-component system, and have addressed several more issues and potential problems, some of which were suggested by fuel cell manufacturers working with samples of our materials. These issues included long-term stability under various combinations of conditions. For example, we tested high-humidity reducing atmospheres as well as voltage gradients that were applied by using cell materials as electrodes. Long-term tests in voltage gradients provided information on cation migration between the sealant and interconnect as well as on conductivity changes in the bulk of the sealant and at the interfaces.

A systematic study of mechanical properties (bonding behavior, thermal expansion, and softening) as a function of composition within the five-component system was also performed during this period. Data from these tests were used in a statistical analysis to determine property trends with composition. For the statistics, we used the nominal compositions for each batch, but we also checked for variations from nominal compositions in the actual samples by using ICP-AES (inductively coupled plasma-atomic emission spectroscopy).

We also studied the effect of scaled-up production methods (by commercial glass manufacturers) on the sealant structure and properties. Commercially prepared batches of the sealants differed in composition from those prepared on a laboratory scale. Analytical results (ICP-AES) were used to determine what changes were necessary in the ratios of starting materials to compensate for these variations.

A further element of this project was interaction with industrial fuel cell developers. We have worked with five fuel cell manufacturers and two university research groups to test the performance of the sealants and to study potential problems.

RESULTS

Long-Term Stability Testing

Tests in high humidity (40% H_2O in helium) and in high-humidity hydrogen were done to determine the long-term stability of our materials. In high-humidity reducing atmospheres, we found slight boric acid volatility in the sealant materials, which could be controlled with compositional changes. Volatility was determined by three methods, namely, measurements of weight loss of the sample, weight of material recovered from the furnace exhaust, and void formation within the sample. Weights lost and recovered tended to agree fairly well, whereas void formation often led to a higher porosity fraction than the associated percentage weight loss. This discrepancy was due to plastic deformation of the sample under internal gas pressure.

Volatility reductions were seen in several cases: volatility always decreased in subsequent 1000°C treatments after an initial high-temperature hold. Reduced volatility was also observed for low-silica compositions, with a minimum at 1 mol% SiO_2. Samples high in lanthanum borate were clearly the most stable. These samples showed some weight loss in an initial 1000°C hold in 40% humidity, but had no measurable weight loss during an additional 62 hours under the same conditions.

Stability in Voltage Gradients

In a manifold-sealing application, the sealant will experience voltage gradients during stack operation. Thus, mobile ions (e.g., chromium ions) from stack materials may be driven into the sealant and may affect its conductivity. To simulate these conditions, Material K was bonded between two lanthanum chromite interconnect pellets, with platinum current collectors attached to the chromite and reference platinum wires at the sealant interfaces. This structure was held in air at 1000°C while a 1-V gradient was established across the sealant. The conductivity of the sealant during a 113-h test is shown in Figure 1. The conductivity decrease in the first 20 h was probably due to the formation of an interfacial phase: in post-test analyses using scanning electron microscopy (SEM), it was found that the positive-biased sealant interface was high in strontium and calcium. In addition, some chromium had migrated into the sealant, which may account for the gradual increase in conductivity toward the end of the test. In general, the conductivity values ($< 10^{-3}$ S/cm) are sufficiently insulating for the application.

Thermal Expansion and Softening

Thermal expansion and dilatometric softening data was gathered for a number of sealant compositions that are variations on Material K within the five-component system, B_2O_3-SiO_2-Al_2O_3-SrO-La_2O_3. Materials studied included those low in either silica or borate, and two series in which strontia and lanthana concentrations were varied independently. Data for these nominal compositions were combined with data from previous studies by our group and with data from the literature (for example, on the B_2O_3-Al_2O_3-SrO glass system) to create a database for linear regression analysis.

The thermal expansion data were fit reasonably well by an expression in which the sealant composition was described by four mole fractions: lanthanum borate, alumina, silica, and boron oxide. The intercept value in this case is associated with pure strontia. With an R^2 value of 0.86 and a standard error

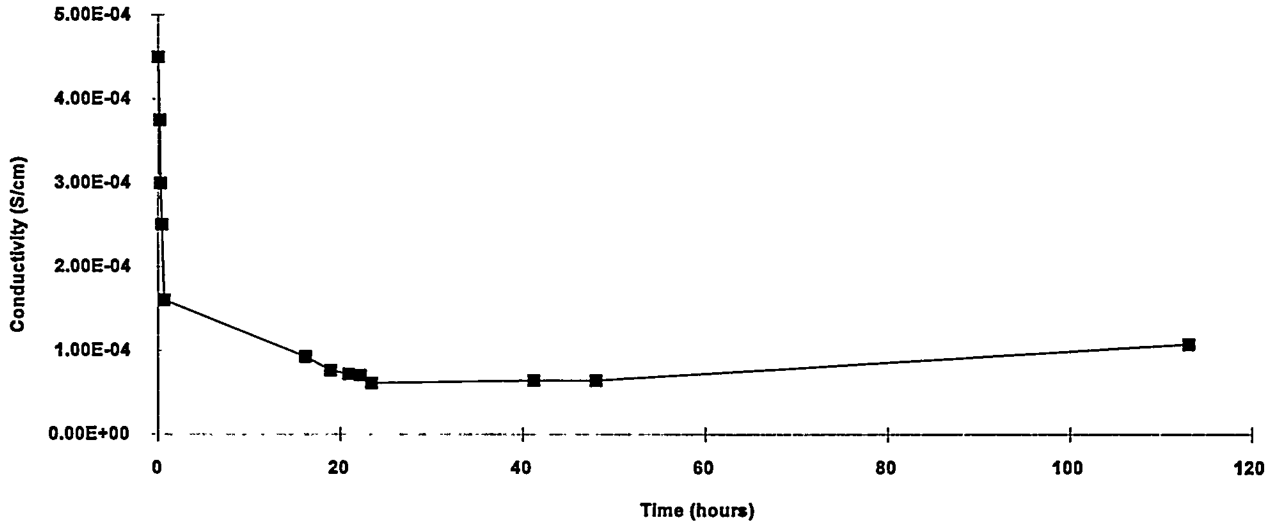

Figure 1. Conductivity of Sealant Material in 1-V Applied Gradient.

of 1.00, the coefficient of thermal expansion, CTE, is given by:

$$\mathrm{CTE} = 18.6 - 3.7[\mathrm{LaBO_3}] - 11.6[\mathrm{Al_2O_3}] - 13.6[\mathrm{SiO_2}] - 16.4[\mathrm{B_2O_3}]$$

This equation allows us to predict with some accuracy the expansion coefficient of a material whose composition falls within the range that we have studied (although this linear expression will probably not be useful for dramatic compositional changes). Linear regression analysis was also used to study the softening behavior of our materials, but no reliable expression was found. In general, materials that were lower in boron oxide, and higher in lanthana and strontia, had higher softening points.

We used ICP-AES analysis to check for variations from nominal compositions in the actual samples. Figure 2 gives the deviations from nominal composition (in percent of the nominal amount of each component) for six different materials. Materials G42, G43, G45 and G47 have low-lanthana, high-silica compositions compared with the two versions of Material K on the left. The general trend is for the low-silica materials to be deficient in strontium and aluminum, while those high in silica are deficient in boron oxide. These trends are probably the result of segregation in the melt, which is likely to occur during small-scale, unstirred melt processing.

Scaled-Up Production Methods

Two commercial glass manufacturers produced scaled-up batches of Material K, and the resulting powders were analyzed for compositional and microstructural changes. The ICP-AES analyses are given in Table 1 for two batches, one from each manufacturer, along with the composition of Argonne-

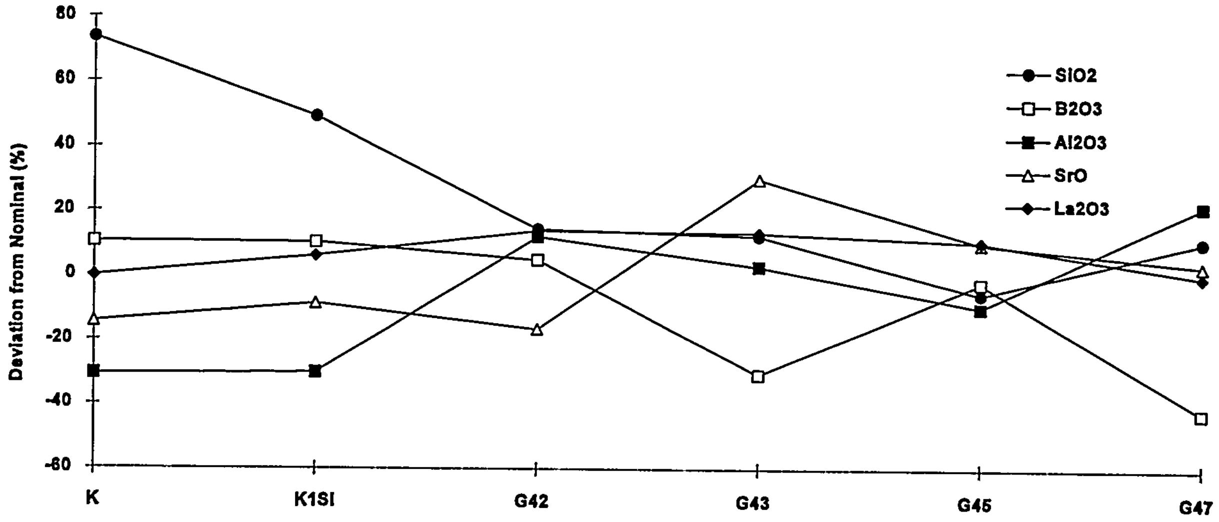

Figure 2. Deviation from Nominal Composition for Six Materials.

produced material. The nominal composition is included for comparison.

The low values for strontia and alumina in the Argonne sample suggest segregation of these oxides during the process. Segregation of strontium-aluminum compounds was confirmed by using X-ray diffraction on an unstirred test melt produced by one manufacturer. Segregation is not expected in full-scale, stirred melt processing.

According to the manufacturer, the large deviation from the nominal strontia amount was due to the raw material: the strontium nitrate was contaminated with sodium. This problem has since been corrected, but is an example of problems that may be encountered during scale-up. The composition of Batch 2 matched the nominal composition fairly well, and with further fine-tuning of the starting composition based on these results, the manufacturer expects to achieve the desired final composition.

FUTURE WORK

Several fuel cell manufacturers have made successful seals with Argonne-produced materials and wish to license the technology for the production of these sealants. Future work will include the analysis of commercial batches now scheduled to be poured, and further modification of the raw material amounts if necessary. Sealants with modified properties for specialized applications may also need to be developed.

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

Table 1. Compositions of Material K Batches Produced by Different Methods

Oxide	Nominal mol%	Argonne mol%	Batch 1 mol%	Batch 2 mol%
SiO_2	4.6	8.11	9.06	4.99
B_2O_3	36.6	40.3	42.6	35.2
Al_2O_3	10	6.92	8.73	7.51
SrO	28.7	24.6	13.6	30.9
La_2O_3	20.2	20.1	22.5	21.4
Na_2O	0	0	3.51	0

P2

Solid Oxide Materials Research Accelerated Electrochemical Testing

CONTRACT INFORMATION

Contract Number (FTPA)	22407
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
Contractor Project Manager	Dr. Timothy R. Armstrong
Principle Investigators	Dr. Chuck Windisch Bruce Arey
METC Project Manager	Bruce Harrington
Period of Performance	October 1, 1994 to June 30, 1995
Schedule and Milestones	

FY95 Program Schedule

	S	O	N	D	J	F	M	A	M	J	J	A
Test Plan		-----	-----	-----								
Testing				-----	-----	-----	-----	-----	-----	-----	-----	-----

OBJECTIVES

The objectives of this work were to develop methods for accelerated testing of cathode materials for solid oxide fuel cells under selected operating conditions. The methods would be used to evaluate the performance of LSM cathode material.

BACKGROUND INFORMATION

Doped lanthanum manganites such as $(La_{1-x}Sr_x)_yMnO_3$ (LSM) have shown promise as cathode materials for high-temperature solid oxide fuel cells. Nevertheless, long-term stability is still a key issue since reactions between fuel cell components, particularly at electrolyte/electrode interfaces, can lead to the formation of resistive layers that result in performance losses. In the case of LSM, a resistive $La_2Zr_2O_7$ (pyrochlore) layer has been reported to form between the LSM cathode and yttrium-stabilized zirconia (YSZ) electrolyte. It was found that 1-2 μm thick layers require more than 100000 h at the SOFC operating temperature of 1000°C, but only a few hours at processing temperatures of 1400°-1500°C (1). Previous studies in our laboratory (2), using reaction couples of LSM and YSZ, have shown that no distinct $La_2Zr_2O_7$ phase forms when the LSM is A-site deficient (where the sum of La and Sr is less than Mn, i.e. $y<1$) after 1 h at temperatures as high at 1500°C nor after extended periods at 1000°C. Rather, for A-site deficient LSM, Mn was found to diffuse into the YSZ.

Diffusion distances were about 20-30 μm after 1 h at 1300-1500°C, and about 35 μm after 1000 h at 1000°C. La, Y and Zr were found not to diffuse across the interface to any significant extent in this study.

While much has been postulated about the effects of the resistive $La_2Zr_2O_7$ layer that clearly forms in A-site enriched manganites, little is known about the practical effects of the Mn diffusion into the YSZ electrolyte for A-site deficient manganites (3,4). Moreover, since most of the studies performed to date focus on thermally induced reactions, either at sintering temperatures or after long thermal treatments at SOFC operating temperatures, additional studies are needed to determine how electrical polarization influences these interactions. To help resolve these and other questions, we have sought to devise *accelerated* electrochemical tests. Similar accelerated testing has been utilized for many years to study corrosion reactions (5).

During the round of development work reported here, LSM/YSZ couples were electrochemically polarized to give some target current density that was significantly higher than that encountered in an operating SOFC. It was hoped that this current density would be high enough to force microstructural and compositional changes that would be encountered in an operating SOFC within a time frame that was manageable for laboratory tests, but not too high to induce changes that would normally not occur. As part of the initial scoping work, it was planned to run experiments at different current densities, as well as different temperatures.

PROJECT DESCRIPTION

Experimental Strategy

Accelerated tests on samples of LSM cathode material sintered to YSZ were performed by subjecting the samples to elevated current densities at various operating temperatures for 1000 h. The material was evaluated during operation by monitoring resistances and by periodically performing electrochemical impedance spectroscopy (EIS). After testing, samples were studied using optical and scanning electron microscopy (SEM), energy dispersive x-ray spectroscopy (EDS), and laser Raman spectroscopy (LRS). Attempts were made to correlate microstructural and compositional changes to resistance changes in the material.

Experimental Procedure

Disks consisting of a layer of LSM with the composition $(La_{0.8}Sr_{0.16})_{0.98}MnO_3$, i.e. A-site deficient, bonded to a layer of YSZ with 8.0 m/o Y_2O_3. The disks were about 1.7 cm in diameter and had an overall thickness of about 0.16 cm. The thickness of the LSM and YSZ layers were about the same, i.e. about 0.08 cm. A set of smaller cylindrical samples, 0.31 cm in diameter, were cut from larger disks for the accelerated electrochemical testing.

The accelerated electrochemical tests were performed using the cell shown in Figure 1. A sample was mounted between two Pt disks, to which were attached the cathode (LSM) and anode lead wires. Both the LSM and YSZ surface of the sample was coated with a thin layer of Pt paste to provide good electrical contact. A Pt mesh was also placed between the Pt disks and the Pt-coated sample to facilitate transport of O_2 to and from the electrodes. The mounted sample was positioned inside an alumina tube furnace and flow of air was maintained across the sample at 0.1 SCFH. Reference electrode lead wires (R1 and R2) were attached to the anode (CE) and cathode (WE) contacts outside of the furnace. With this arrangement, of course, measured cell resistance includes components from the lead wires, contacts electrolyte and both the anode and cathode. The resistance of the lead wires inside of the furnace was measured at temperature (about 3 Ω total) and used to correct all measurements on the samples. The voltage drop at the anode was assumed to be much smaller than at the cathode because the Pt mesh should have significantly reduced mass transport resistance at the anode, which dominates when current flows. In contrast, mass transport should have been more important at the cathode which, while porous, afforded a much denser and more tortuous path for O_2 diffusion. Consequently, in these tests, the corrected cell resistance was assumed to arise primarily from the electrolyte, any contact resistance, and the cathode reactions.

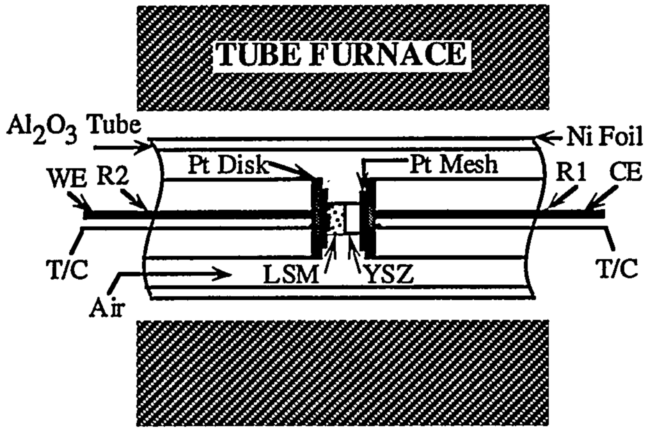

Figure 1. Electrochemical Cell for Accelerated Testing of Cathode Materials

Current and voltage on the cell were controlled with a Solartron (Billerica, MA) 1286 Electrochemical Interface. Electrochemical impedance spectroscopy measurements (EIS) were made with the Solartron 1286 and a Solartron 1250 Frequency Response Analyzer. Data acquisition and control were performed using Scribner Associates (Charlottesville, VA) ZPLOT software running on an IBM compatible computer.

Three 1000-h accelerated electrochemical tests were performed. The target conditions for each test were as follows: (I) 1000°C at 1.0 A/cm^2, (II) 1000°C at 0.5 A/cm^2, and (III) 900°C at 1.0 A/cm^2. Each test was performed by first heating the sample in the cell in air without current for at least 24 h. Current was then applied galvanostatically in increasing steps until the target current density was obtained. This current ramp-up permitted a pseudo-steady state polarization curve to be obtained. The sample was then held at the target current density for at least 24 h, noting any potential drift that occurred during this period. Following this conditioning period, the system was switched to potentiostatic control using the potential noted at the end of the galvanostatic period as the controlling potential. Potentiostatic control was used for the bulk of the accelerated tests so that EIS spectra could be obtained more easily. The cell was run at the set potential for 1000 h while recording current, voltage and temperature, periodically. During each test, the current was observed to vary by as much as 50%, so the target current densities were not maintained throughout. *Nevertheless, the samples are referred to in this report according to their target current density.*

After switching to potentiostatic control, a batch program collected EIS spectra every 24 h for the balance of Tests (II) and (III). (EIS spectra were not collected during Test (I) due to equipment problems.) All spectra were collected under open circuit conditions (i.e. the controlling potential was interrupted briefly so the EIS spectra could be obtained). For this reason, they are mainly an indication of how the material is changing, and not of its actual properties under operating conditions. Spectra were obtained using a ±10 mV excitation voltage over the frequency range 0.1 Hz to 65 kHz.

After testing, the samples were removed from the cell, mounted, cross-sectioned, and polished for optical and SEM microscopic analysis, EDS and LRS. LRS spectra were obtained on the samples using a Spex (Edison, NJ) triple monochrometer spectrometer with microprobe attachment and a Princeton Instruments (Trenton, NJ) CCD detector. The 488.8-nm line of a Spectra Physics (Mountain View, CA) Ar^+ ion laser was used for excitation. Spacial resolution of the microprobe was about 1 µm.

In addition to samples subjected to accelerated electrochemical testing, control samples were also studied. These included a sample that was only heated (no current) at 1000°C for 1000 h and a sample that was neither heated nor electrically polarized after fabrication.

RESULTS

Polarization Scans

As each sample was ramped up to its target operating current density, pseudo-steady state polarization curves were obtained. These curves are shown in Figure 2 for tests at 1000°C and 900°C described above. Data from a 800°C test that is currently being performed is also included.

As shown in Figure 2, as temperature was increased, the instantaneous slopes of the curves also generally increased. In other words, a higher temperature resulted in a lower cell resistance (reciprocal of the slope).

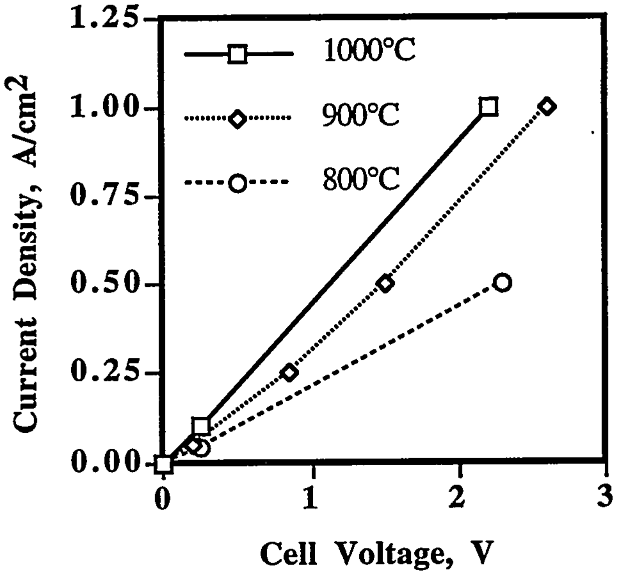

Figure 2. Polarization Curves Obtained Ramping Up to Target Current Density

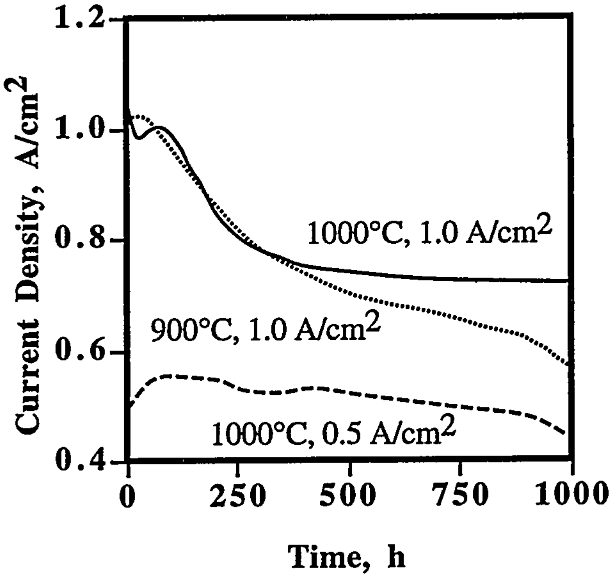

Figure 3. Variation of Current Density with Time for Different Target Conditions

This result cannot be fully explained by the increase in resistance of the YSZ electrolyte with decreasing temperature. Using an equation previously developed for the thermal activation conductivity for YSZ (6), the resistance for the YSZ in the 1000°C and 900°C experiments is calculated to be 0.36 and 0.68 $\Omega \cdot cm^2$, respectively. The slopes of the curves in Figure 2 give actual cell resistances of about 2.2 and 3.8 $\Omega \cdot cm^2$ for 1000°C and 900°C, respectively. Clearly, the electrolyte resistance is only a small part of the cell resistance and other factors such as contact resistance and contributions from electrode processes need to be considered. As described below, the accelerated electrochemical tests were designed to try to determine not only what these resistive components are, but how they change in importance over a 1000 h test.

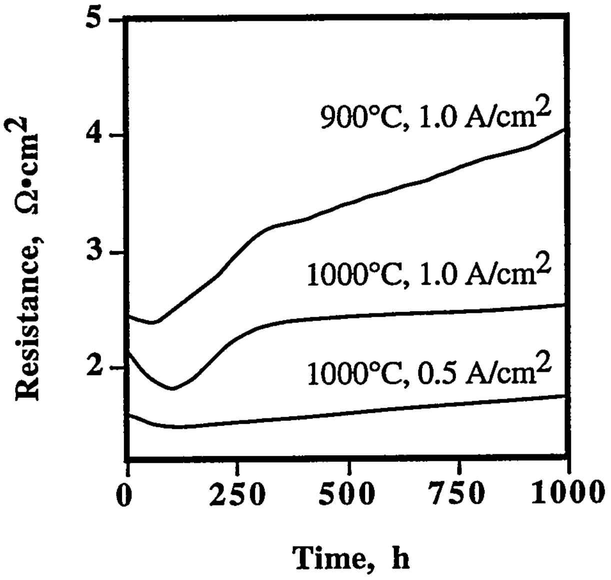

Figure 4. Variation of Cell Resistance with Time

Cell Resistance During Operation

As shown in Figure 3, current density generally decreased during most of the three, potentiostatically controlled, 1000-h tests. The current density and voltage were used to calculate the normalized dc cell resistance in $\Omega \cdot cm^2$ and these are plotted in Figure 4 for each of the three tests. As shown, the cell resistance decreased during the first 100 h and then increased for the balance of the 1000 h.

While the form of variation of resistance with time was similar for the three tests, the magnitude of the changes appeared to depend strongly on current density and temperature. Increasing current density from 0.5 A/cm2 to 1.0 A/cm2 at 1000°C appeared to result in an overall higher resistance, a more dramatic "dip" in resistance during the first 250 h, and a leveling off after about 400 h. Resistance at 0.5 A/cm2 showed no tendency to level off even after 1000 h, although the magnitude of the resistance only reached about 3/4 of the value at 1.0 A/cm2.

Decreasing temperature from 1000°C to 900°C at 1.0 A/cm2 appeared to increase the cell resistance by about 40-50%. This result is similar to the effect of temperature on the polarization scans discussed earlier, and clearly reflects the temperature dependence not only of the electrolyte but of the electrode reactions as well. Unlike the 1000°C test, however, the resistance kept increasing after the first 400 h at 900°C.

EIS Spectra

EIS spectra collected during the 1000-h tests were similar to those shown in Figure 5. In general, they consisted of two or more overlapping loops between 65 kHz and about 10 Hz, and a "nose" at lower frequencies. The overlapping loops were taken as charge transfer reactions and the "nose" was associated with mass transport. These assignments are based on previous work in our laboratory (7) that showed an O_2 concentration dependence for the low-frequency loop consistent with control by the migration of atomic or molecular oxygen, depending on the conditions. Also similar to these previous studies, after interrupting the cell current, repeated EIS spectra showed the low-frequency "nose" grew larger with time.

Since the EIS loops overlapped so much, it was difficult to resolve the contributions from the individual charge transfer processes. Also, the dynamic nature of the low-frequency "nose" precluded detailed analysis of this feature. Rather, the only useful information appeared to be the extrapolation of both "ends" of the spectra. The difference between the high-frequency intercept (R_1) and the resistance from the lead wires (R_w) was taken as the electrolyte resistance (including any contact resistances); while the difference between the low-frequency intercept (R_2) and the high frequency intercept (R_1) was taken as the overall contribution from the electrode processes. While these assignments are not strictly accurate, given the dynamic nature of some of the loops and the ambiguities caused by grain boundary contributions, etc., the approach was thought to give some indication as to how these two components of the impedance varied (relative to each other) with the experimental conditions. Also, since the spectra were collected in the batch mode, the acquisition parameters were exactly the same for each spectrum. Drift in the data subsequent to current interruptions should, therefore, have been the same, or very similar, for each acquisition.

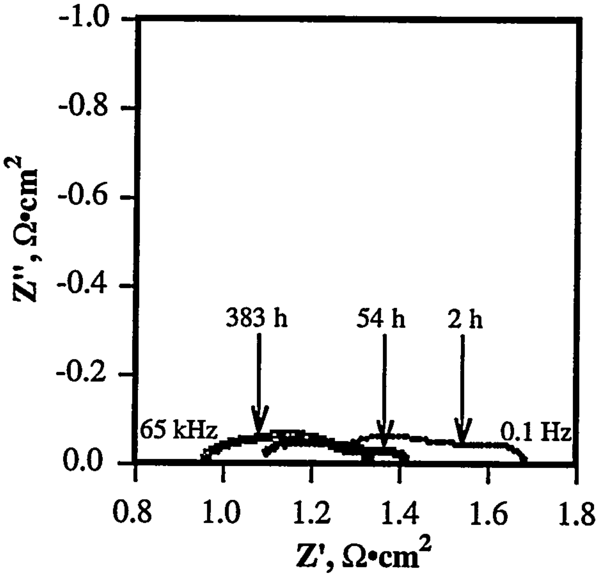

Figure 5. EIS Spectra Collected at Various Times at 1000°C and 1.0 A/cm2

Figures 6 and 7 show how R_2 (the total dc resistance), $R_1 - R_w$, and $R_2 - R_1$ varied with time for Test (II) at 1000°C and 0.5 A/cm2, and Test (III) at 900°C and 1.0 A/cm2. The average value for R_2 over the 1000 h, about 1.5 Ω•cm2 for Test (II) and about 3.2 Ω•cm2 for Test (III), are within the range of cell resistances calculated from the current and voltage in Figure 4. R_2 in Figures 6 and 7 also varies with time similar to the cell resistances in Figure 4. The values are not exactly the same, however; and this was expected, since the EIS data were collected at open circuit while the current and voltage data were measured with the

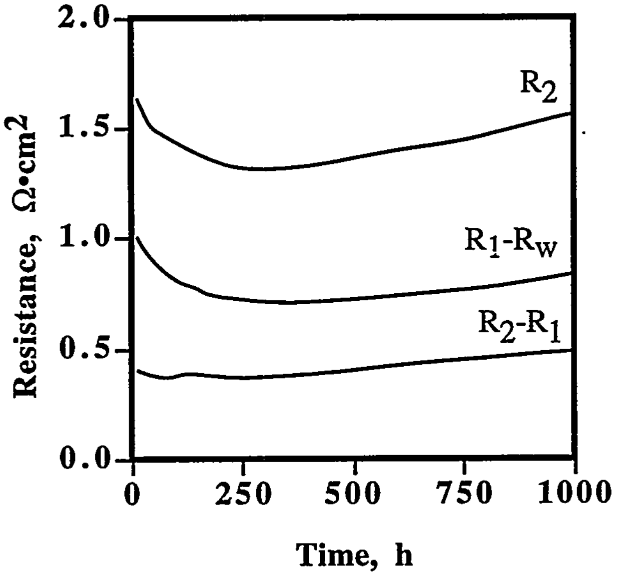

Figure 6. Variation of EIS Components of Cell Resistance with Time for 1000°C and 0.5 A/cm^2

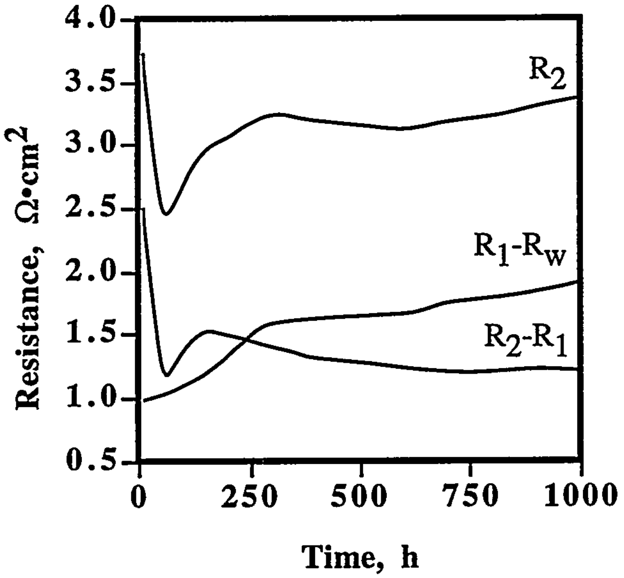

Figure 7. Variation of EIS Components of Cell Resistance with Time for 900°C and 1.0 A/cm^2

cell on. Nevertheless, the trends are similar: cell resistance drops significantly during the first 100 h and then increases slowly for the balance of the test.

The components R_1 - R_w and R_2 - R_1 (Figures 6 and 7) provide insight as to what components are primarily responsible for the changes in cell resistance. At 0.5 A/cm^2 and 1000°C, both R_1 - R_w and R_2 - R_1 contribute to the overall cell resistance in a similar way. The two components decrease initially then increase during the rest of the test. The only difference is that the initial drop appears larger for R_1 - R_w. At 1.0 A/cm^2 and 900°C, R_1 - R_w and R_2 - R_1 behave very differently. R_1 - R_w increases quickly during the first 250 h and then more slowly after that. R_2 - R_1, on the other hand, decreases rapidly at first jumps suddenly, and then decreases again, but more slowly after the first 200 h.

Figures 6 and 7 indicate a complexity of behavior for these electrode materials that will clearly require additional experiments to understand fully. No clear trend is suggested for the two tests that points to a single rate-determining step for the resistance changes. For Test (II), the electrolyte appears to drive the resistance changes within the first 250 h, while both the electrolyte and electrode reactions contribute at longer times. For Test (III), the electrode reactions appear rate-determining within the first 250 h, while the electrolyte dominates at longer times.

Post-Mortem Analysis of Samples

Despite the complexity of the electrochemical data, which further study should clarify, some conclusions can still be made by comparing the electrochemical results with the post-mortem analysis of samples, as discussed below.

Optical microscopy and SEM of the interface region in the sample from Tests (II) revealed a layer, 1-2 µm thick, on the YSZ side (Figure 8). This layer contained significant Zr, but also La and Mn. A control sample heated to 1000°C for 1000 h, but with no current, also showed this layer. The sample from Test (I) and an untreated control sample did not exhibit a layer with comparable thickness. (A thinner, undetectable, layer in these samples cannot be ruled out.) As shown in Figures 9 and 10, EDS detected significant, but varying, amounts of Mn and La in the first few µm of YSZ near the interface for all samples. Samples with the measurable reaction layer gave higher

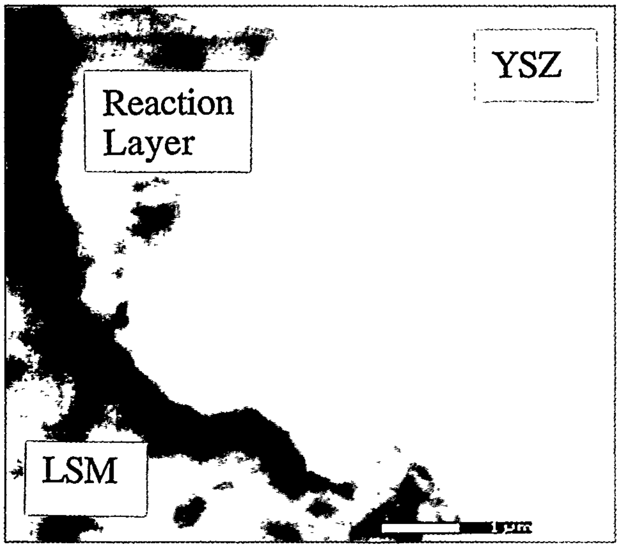

Figure 8. SEM Micrograph of the Reaction Layer in Sample from Test (II)

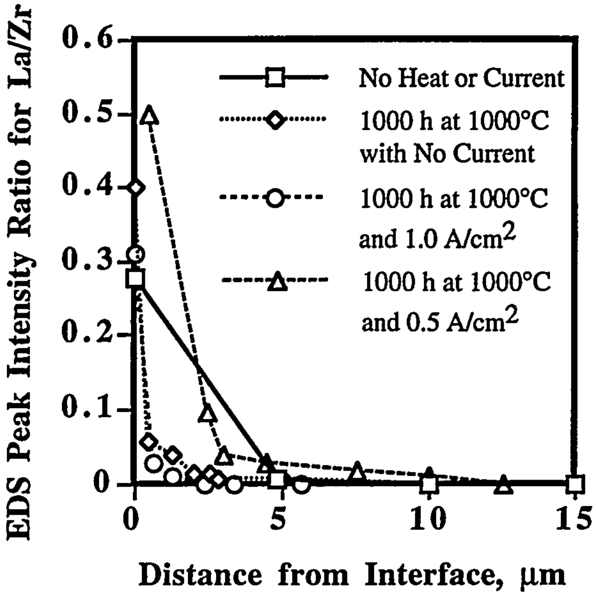

Figure 9. Variation of EDS Peak Ratio for La/Zr as Function of Distance from Interface within YSZ Phase

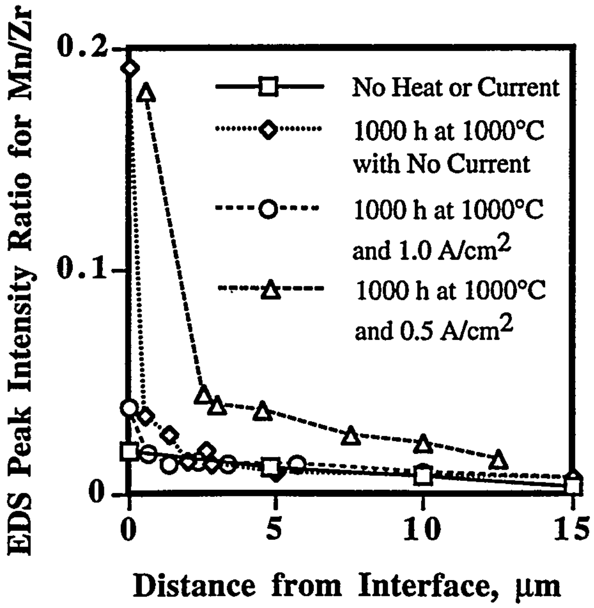

Figure 10. Variation of EDS Peak Ratio for Mn/Zr as Function of Distance from Interface within YSZ Phase

amounts of La and Mn near the interface (within the layer), as expected. The measurements, however, did not indicate the presence of any stoichiometric composition, such as $La_2Zr_2O_7$, which was previously reported for A-site excess compositions (1). The composition varied dramatically and nonstoichiometrically within the interfacial region.

Since the heat-treated control sample contained a reaction layer, it seems unlikely that the current was responsible for its growth. More likely, the layer formed during heat treatment or fabrication. Unfortunately, the specific conditions for its formation are uncertain because of the widely varying results for the few samples tested to date. More importantly, it appears that the layer did not cause an increase in resistance. Test (II) gave the smallest resistances and yet had the thickest layer. The increase in resistances during the latter portion of the tests, shown in Figures 4, 6 and 7, is clearly due to factors other than the growth of this layer.

As shown in Figures 9 and 10, Mn tended to penetrate the YSZ further than La for all of the samples. Significant Mn was measured even at 15 μm away from the interface. This is consistent with previous results for A-site deficient cathode materials (2). It is still not certain from these experiments what role Mn has in the YSZ, particularly in regards to resistance changes. Assuming the Mn is present at grain boundaries, it is possible that it behaves similar to other grain boundary contaminants in zirconia. Elements like Al, Si and Fe are known to migrate to grain boundaries and reduce conductivity by behaving as ion "blockers," especially between 800°C and 1000°C (8). This "aging" process may be induced by Mn as well and be affected by operating conditions such as current density in a way that would explain the results reported here. Other possibilities are: that Ni, which was found as a contaminant in some of the zirconia samples, causes the blocking effect, or that the initial interface morphology differed from sample to sample and these differences had an effect on the resistance changes. Future work will help develop more of a statistical data base for distinguishing the effects of sample differences, as well as in helping to determine the role of Mn at the interface by using molecular spectroscopic techniques such as LRS, as reported below.

In contrast to the above results, the sample from Test (III) showed a more porous reactions layer between the LSM and the YSZ. SEM revealed this layer was essentially a manganese oxide. Sr and La were found as small segregated particles in a crack that had formed at the interface (probably during cool down). This layer may have been responsible for the more drastic increase in cell resistance for Test (III) in Figures 4, as well as the electrolyte resistance in Figure 7. The reaction layer points to a reductive transformation of the manganite when the potential differences are quite large, as they were for Test (III). A test currently being performed at 800°C, where even higher potentials are being used, should help explain this particular result.

Evidence for Mn within the electrolyte was also provided by LRS. The LSM and the YSZ, far from the interface, each give a characteristic spectrum. The spectrum for YSZ, is characterized by a strong peak at about 620 cm^{-1}, while the spectrum for the LSM contains broad features at about 480 and 1000 cm^{-1}. The lower frequency feature for LSM is in the range reported for Mn-O vibrations in various manganese oxides (8). The higher frequency band is more difficult to explain, although it may arise from second order process as observed in La_2CuO_4 and other electrically semiconducting materials (9). In any event, the presence of both the 480 and 1000 cm^{-1} bands are indicative of LSM or some component of this oxide.

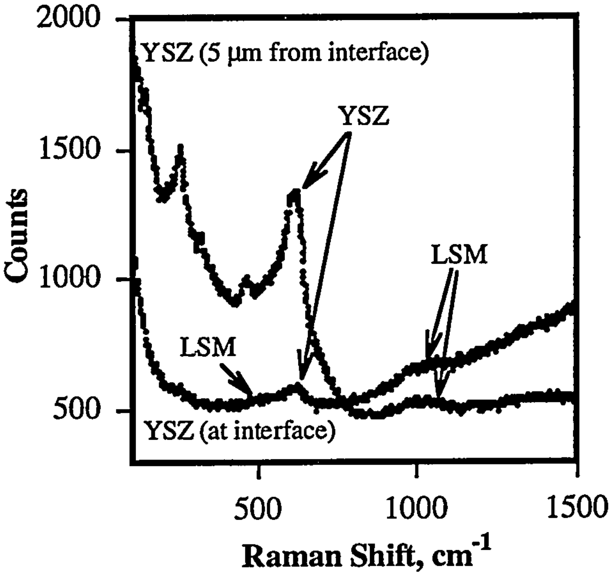

Figure 11. Raman Spectra of YSZ Phase at Different Locations after 1000 h at 1000°C and 0.5 A/cm^2

As shown in Figure 11, the Raman spectra taken on the YSZ layer near the interface for the sample from Test (II) show the presence of the LSM features, in addition to those assigned to YSZ. Although details regarding the chemical environment and bonding are not forthcoming from these preliminary LRS data, the result suggests, at the very least, that Mn-O type structures are present within the YSZ layer in the region near the interface. These spectra are consistent with the EDS data that indicated Mn was present. Additional LRS studies may reveal more information about the chemical bonding of Mn in the YSZ interfacial region and help determine whether (and how) the element affects the electrode properties.

Conclusions

Despite the limited number of experiments performed to date, a few preliminary conclusions can be made regarding the LSM/YSZ couple studied in this work:

- Accelerated electrochemical testing appears feasible using the approach detailed in this report. Studies over a range of current densities and temperatures are needed for a given material, however, to determine those conditions which would be relevant to fuel cell operation.
- EIS measurements have the potential to distinguish contributions from the electrolyte, including any resistive layers, and the electrode reactions. This would make the technique a valuable tool for explaining changes in the performance of cathode materials during extended operation.
- Cell resistance was greater at lower temperatures, partly due to the increase in resistivity of the electrolyte and partly due to other factors.
- Cell resistance appeared to be greater at higher current densities but the reason for this was not clear.
- During 1000-h operation, cell resistance decreases significantly over the first 100 h, and then increases for the remainder of the test.
- The gradual increase in cell resistance after the first 250 h at 1000°C and at either 0.5 or 1.0 A/cm^2 is probably *not* due to the growth of a resistive layer at the interface. A Mn and La-containing layer within the YSZ was observed but could not be correlated with cell resistance or experimental conditions. Alternatively, some other change in the electrolyte, possibly involving the Mn which migrated into the electrolyte during fabrication, may affect the resistance over longer times. Further work is required to resolve this uncertainty.
- A reaction layer composed of mainly Mn oxides formed at the interface for a sample treated at 900°C and 1.0 A/cm^2. It seems likely that the formation of this layer was responsible for the increase in resistance observed in this test.
- LRS was used successfully to measure Mn in the YSZ and has the potential to provide additional information on its chemical environment.

FUTURE WORK

Future work will involve the following:

- Additional 1000 h studies on the LSM/YSZ couple will be performed at other temperatures and current densities. We are currently performing a test at 800°C and 1.0 A/cm^2. Studies at 1100°C and lower current densities are also anticipated.
- Selected experiments will be repeated to obtain better estimates of experimental uncertainties.
- A set of 1000 h tests are planned for a LSM/YSZ couple with more porous LSM. It is hoped that this study will resolve effects from mass transport that are suspected to contribute at the higher current densities.
- Laser Raman studies will be continue to try to determine the role of Mn in the YSZ.
- Samples with LSM that is excess in A-site ions will be tested to see if resistive layers form at the interface under electrochemical polarization. Previous studies indicate a layer forms due to thermal treatment, but little information exists on whether it forms or how it behaves under prolonged electrochemical treatment.
- An additional electrochemical testing rig (Figure 1) is being set up to facilitate the above work. Clearly, the 1000-h tests are time consuming. Only about a half-dozen or so tests can be performed using a single rig within a year; and this assumes that the experiments proceed without interruption from power failures or instrumental problems.

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P3

Processing of $LaCrO_3$ for Solid Oxide Fuel Cell Applications

CONTRACT INFORMATION

Contract Number	DE-FG21-93MC29224
Contractor	University of Missouri - Rolla Ceramic Engineering Department 222 McNutt Hall Rolla, MO 65401 (314) 341-6129
Contractor Project Manager	Wayne Huebner
Principal Investigators	Wayne Huebner Harlan U. Anderson
METC Project Manager	Bruce Harrington
Period of Performance	April 16, 1995 to April 15, 1996

Schedule and Milestones

FY95 Program Schedule

	A	M	J	J	A	S	O	N	D	J	F	M
Influence of B-site Dopants on the Densification Behavior	←	─	─	─	─	─	─	→				
Influence of B-site Dopants on the Conductivity and TEC			←	─	─	─	→					
Effect of Stoichiometry of the Sintering Behavior of $LaCrO_3$		←	─	─	─	─	─	─	→			
Processing of Planar SOFC's with Varying Cathode & Anode Microstructures	←	─	─	─	─	─	─	─	─	─	─	→
Processing of Planar SOFC's with Interfacial Modifications						←	─	─	─	─	─	→

OBJECTIVES

The University of Missouri-Rolla is performing a 5 year research program with two primary objectives: 1) developing $LaCrO_3$-based interconnect powders which densify when in contact with anode and cathode materials for solid oxide fuel cells (SOFC), and 2) developing high performance cathodes, anodes and interfaces for use in planar SOFC's.

With regard to the processing and sintering of $LaCrO_3$, the specific objectives of this research program are to:

- Develop a non-liquid phase sintered $LaCrO_3$-based material sinterable in air.
- Improve and control the properties requisite of $LaCrO_3$ utilizing a B-site acceptor dopant
- Optimize and control the processing conditions associated with $LaCrO_3$.
- Incorporate materials developed in this program into planar cells and measure their performance.

With regard to developing high performance materials for use in planar SOFC's, the specific objectives of this research program over the last year have been to:

- Fabricate single cells with controlled microstructures (i.e. grain size and porosity of electrodes) for operation at 1000°C.
- Gain a better understanding of the mechanisms involved in improving cell performance via electrochemical and impedance techniques.
- Develop processing ↔ microstructure ↔ property relations of electrodes and their corresponding interfacial reactions.

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 the SOFC IV conference held in Yokohama, Japan. At this meeting numerous research groups identified the cost of processing $LaCrO_3$ was prohibitive, as well as its tendency to change dimensions upon reduction. 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]. However, the high mobility of the fluoride fluxes detrimentally affected the other cell components. More recent studies including our own have also shown that while transient liquid phase sintering may allow for lower temperature densification of the interconnect alone, the use of large amounts of A-site dopants (i.e. Ca,Sr) in amounts excess of stoichiometry create problems with second phase formation and limited mechanical stability in reducing atmospheres. Hence in some ways we are back to square one, and in this program we have refocused our attention on densifying acceptor-doped $LaCrO_3$ without the addition of a liquid phase.

Materials research in the area of SOFC's is also driven by the recognition that processing and operating at lower temperatures would circumvent most of the reliability problems which are currently preventing these devices from achieving wide-scale commercialization. These considerations have directed interdisciplinary research thrusts in this field, namely:

- Alternate Materials
  - → higher conductivities at lower temperatures for all four SOFC components
  - → mixed-conducting cathodes
  - → greater phase stability at low pO_2's
- Processing
  - → novel synthesis techniques (for powders and thin films)
  - → controlled microstructure with desired properties
- Reliability Issues
  - → chemical, mechanical, and electrical stability under the temperature/time/atmosphere conditions of cofiring and operation

Hence studies on the optimization of components use of mixed conductors and thin film processing techniques[11-16], as well as studies on structure↔property relationships represent an enabling technology; what remains is to utilize this technology for the fabrication of planar SOFC's. Key to this next stage is the development of a porous anode substrate with the requisite microstructural, electrical and mechanical properties. While many researchers have investigated $Ni:ZrO_2$ cermet anodes[7], many issues related to microstructural evolution and stability, as well as the impact of Ni scale and connectivity on the electrical properties remain unresolved.

PROJECT DESCRIPTION

This report is divided into two primary sections:

#1 $LaCrO_3$ Sintering Studies
#2 SOFC Performance Studies

Results from these studies are presented in the following sections.

$LaCrO_3$ SINTERING STUDIES

Experimental Approach:

Powders of the composition $LaCr_{1-x}M_xO_3$ were prepared by a modified liquid mix process, in which ethylene glycol and citric acid are added to a stoichiometric mixture of carbonates and nitrates of the constituent ions in a 1:1:1 ratio. Starting materials were thermogravimetrically standardized to ± 0.1mg and x-rayed to ensure correct oxide composition. BET measurements indicate resulting powders have a specific surface area of ~5.0 m^2/g. Sintering studies were conducted with 1/2" pellets uniaxially pressed to 3ksi and isostatically pressed to 40ksi. Green densities were taken geometrically, while final densities were measured by the Archimedes technique with kerosene as the medium. Sintering studies were performed on an Orton 1600D dilatometer with 1/4" pellets uniaxially pressed to 0.2ksi and isostatically pressed to 40ksi. Crystallinity and phase purity were determined by x-ray on a Rigaku Miniflex system. Conductivity was determined with a 2-point d.c. apparatus with platinum electrodes over the temperature range of 25-1300°C.

RESULTS

A) Sintering

Recent studies in our laboratory have shown that compositions of $LaCr_{1-x}M_xO_3$ with $x<0.10$ exhibit enhanced sintering in air at temperature up to 1600°C. X-ray and TGA results (Figures 1 and 2) show that the material remains phase pure perovskite throughout the temperature range and that no apparent liquid is formed in the material. Figure 3 shows densification studies performed at 1600°C/6hrs as a function of dopant concentration. Despite problems with the processing resulting in green densities of 48-51% theoretical density, samples were still found to sinter to 90% of theoretical density. The relative densities level off with about 7% dopant additions. Figure 4 shows a polished and thermally etched

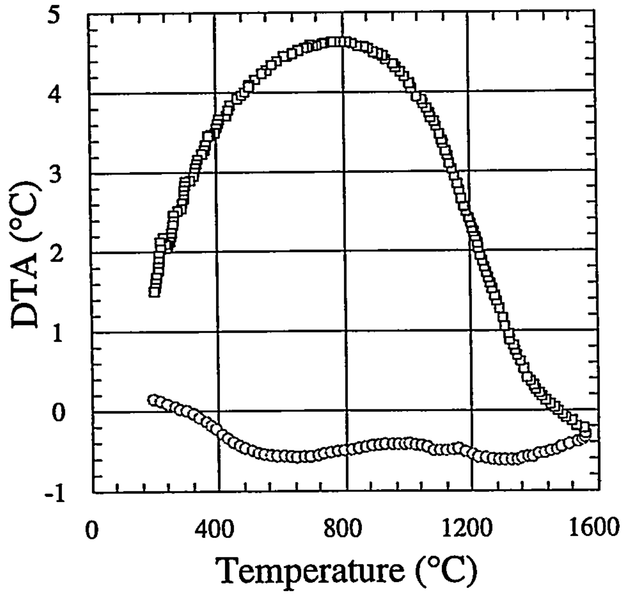

Figure 1: DTA Results on $LaCr_{0.9}M_{0.1}O_3$

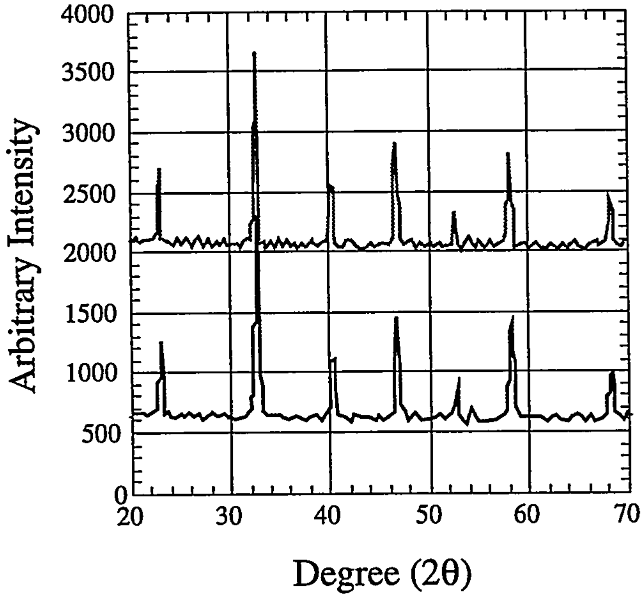

Figure 2: XRD Results on $LaCr_{0.9}M_{0.1}O_3$

specimen of $LaCr_{.90}M_{.10}O_3$ corresponding to a relative density of ~90%. Tests on the effects of stoichiometry were performed for lanthanum contents of 0.98, 0.99, and 1.00. Figure 5 shows

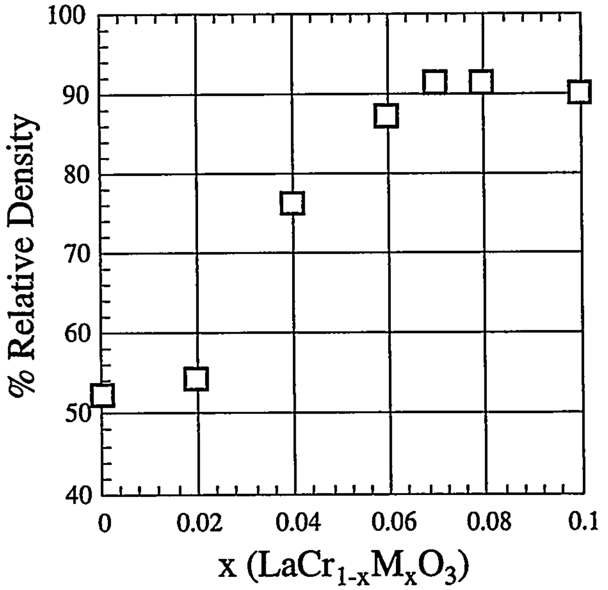

Figure 3: % Relative Density vs. Dopant Concentration for $LaCr_{1-x}M_xO_3$ (1600°C - 6h)

Figure 4: Polished and Thermally Etched Specimen of $LaCr_{.90}M_{.10}O_3$ (1600°C - 6h)

that when the lanthanum content is reduced 1% to a nominal composition of $La_{.99}Cr_{.90}M_{.10}O_3$ the

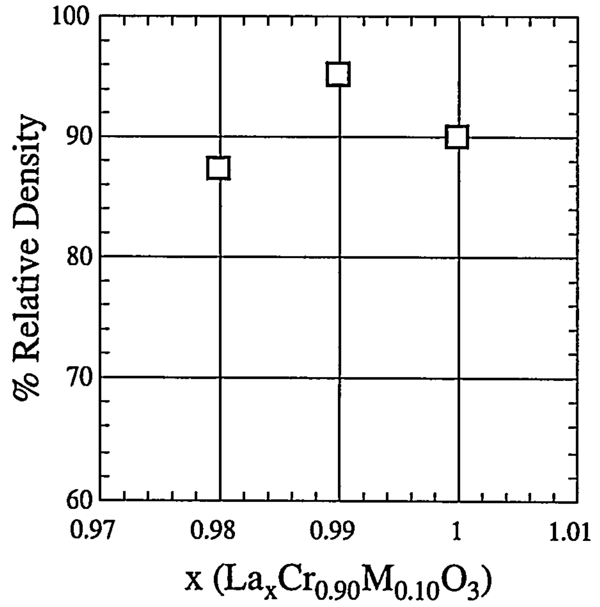

Figure 5: % Relative Density vs. Lanthanum Content for $La_xCr_{0.9}M_{0.1}O_3$ (1600°C - 6h)

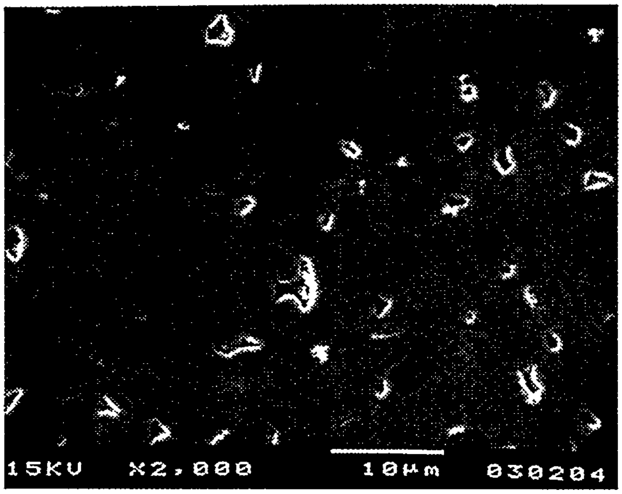

Figure 6: Polished and Thermally Etched Specimen of $La_{.99}Cr_{.90}M_{.10}O_3$ (1600°C - 6h)

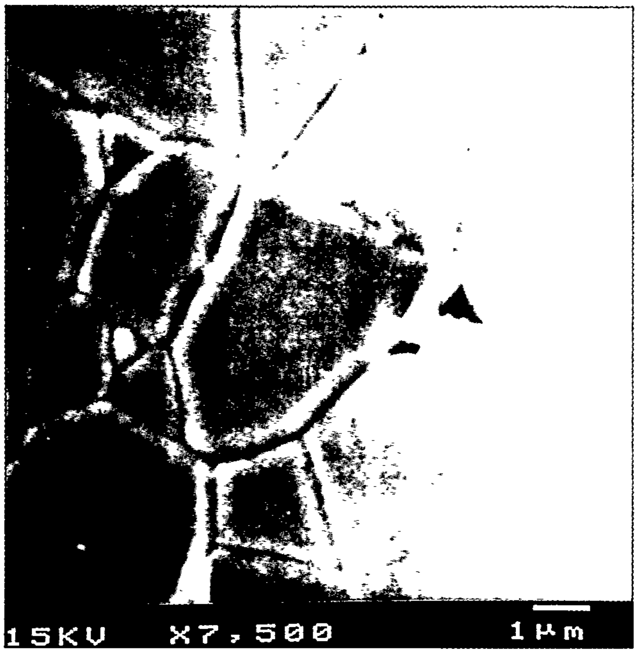

Figure 7: Polished and Thermally Etched Specimen of $La_{.99}Cr_{.90}M_{.10}O_3$ (1600°C - 6h)

relative density is increased to 95%. Figures 6 and 7 show a polished and thermally etched $La_{.99}Cr_{.90}M_{.10}O_3$ specimen . The percent linear shrinkage of $La_{.99}Cr_{.90}M_{.10}O_3$ is shown is figure 8, with the sample exhibiting ~28% shrinkage to a final relative density of ~90% after 6 hours at 1600°C. Sintering starts at ~1100°C reaching a shrinkage of 17% before the hold at 1600°C. Further results have shown that 4 hrs is sufficient to reach the maximum shrinkage. Improvements in processing related issues should allow for even higher sintered densities to be achieved, possibly allowing the lowering of the sintering temperature. Current work is focusing on theses issues.

B) Property Control

Preliminary studies have shown that $La_{.99}Cr_{.90}M_{.10}O_3$ has a conductivity of ~8S/cm at 1000°C, sufficient for fuel cell applications (Figure 9). As seen in figure 10 the plot of Log(σ*T) vs. 1000/T is linear, as is expected under a small-polaron model. Current work is progressing on the evaluation of other properties such as: mechanical and electrical integrity under ambient and hydrated reducing conditions, match of thermal expansion with other cell components, and long term chemical stability with other materials in the fuel cell.

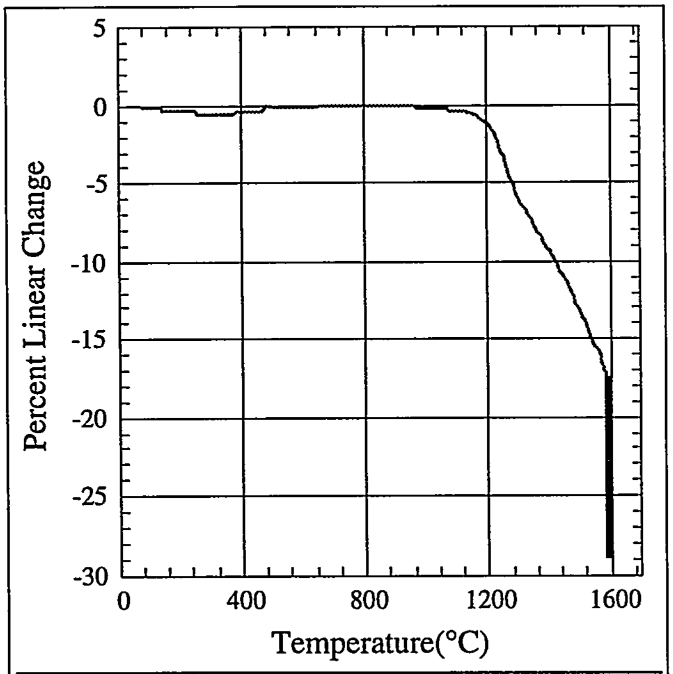

Figure 8: Percent Linear Shrinkage of $La_{.99}Cr_{.90}M_{.10}O_3$

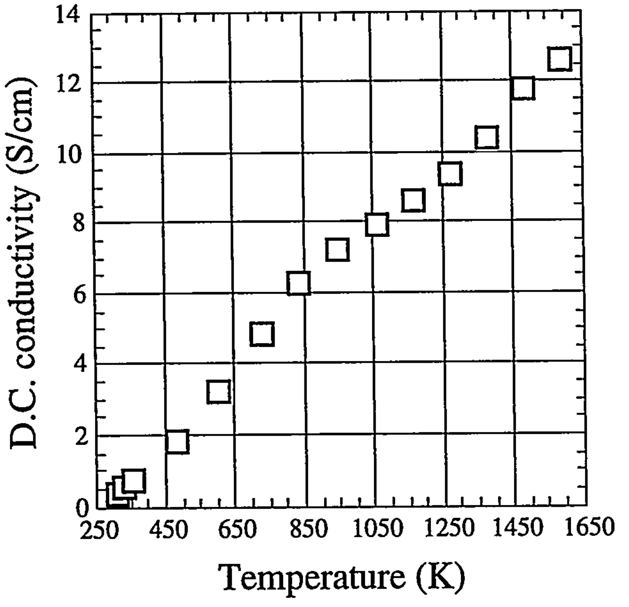

Figure 9: DC Conductivity of $La_{.99}Cr_{.90}M_{.10}O_3$ sintered at 1600°C - 6h

Major Achievement: The development of a non-liquid phase sintered $LaCrO_3$-based interconnect system that utilizes a B-site dopant.

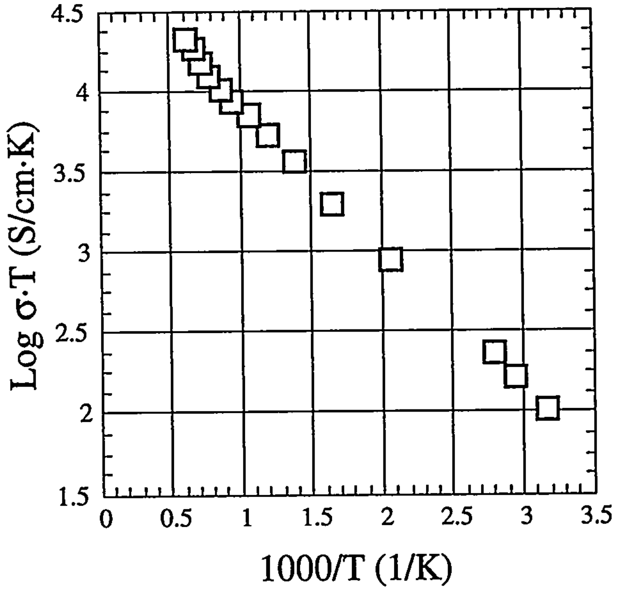

Figure 10: Arrhenius Plot of the DC Conductivity of $La_{.99}Cr_{.90}M_{.10}O_3$ sintered at 1600°C - 6h

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 was completed during Year 2, whereas Phases II and III are presently ongoing.

Experimental Approach:

Sample Preparation

Single cells were fabricated by conventional thick film processing techniques as described in Figure 11. Commercial YSZ powder (Zirconia Sales of America) was mixed with appropriate binders, solvents, plasticizers, and surfactants to prepare a castable slip. The slip was ball milled with ZrO_2 media then cast at a thickness of 10 mils using the doctor blade technique. Circular samples were punched out of the tape and a thin Pt reference probe was screen printed onto the YSZ tape. The samples were laminated and fired flat at 1450°C for 2 hours. Fired thicknesses of the Pt reference electrode and the YSZ electrolyte are 5 and 200µm, respectively (Figures 12 and 13).

Anode powders of NiO, $Ni_{1-x}Mg_xO$, YSZ (Zirconia Sales of America) and Sm doped CeO_2 were used as starting materials. NiO, $Ni_{1-x}Mg_xO$, and Sm doped CeO_2 were prepared by the Pechini liquid mix and glycine -nitrate methods to give powders of varying particle size and surface area. Powder crystallinity, phase and surface area were characterized by X-ray diffraction and BET techniques as a function of calcination temperature. Anode powders were mixed with appropriate binders, solvents, and surfactants and three roll milled to prepare high viscosity pastes with correct rheological behavior for screen printing. Anode pastes were screen printed onto dense YSZ electrolytes and fired at various temperatures (1200 - 1500°C). A typical anode microstructure, NiO - YSZ, fired in air is shown in Figure 14. The anode thickness can be varied depending on the mesh size, emulsion thickness, and wire diameter of the screen.

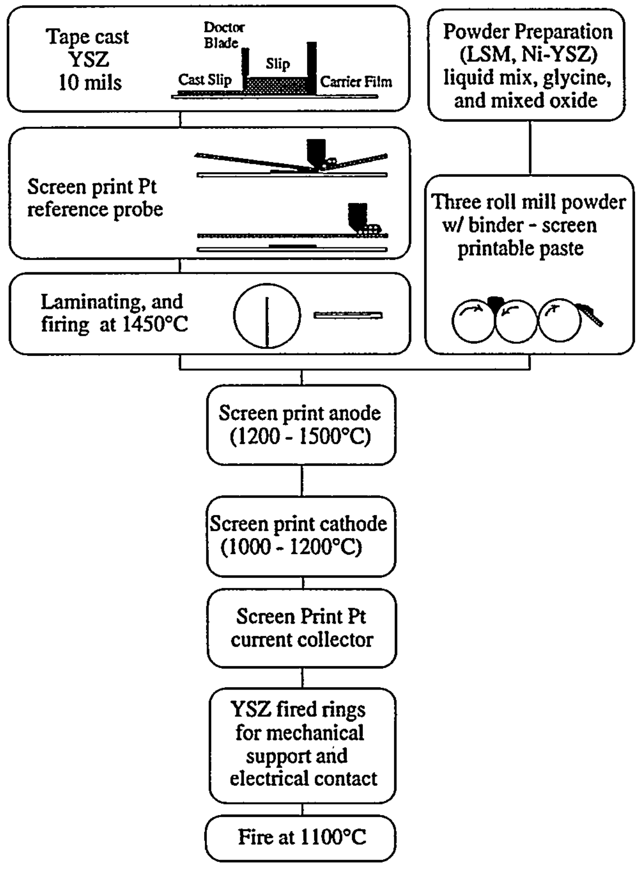

Figure 11. Experimental Overview

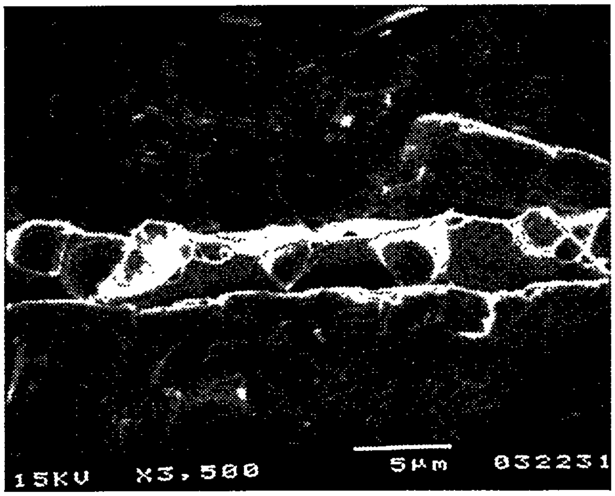

Figure 12: Pt Reference Electrode within the YSZ Electrolyte

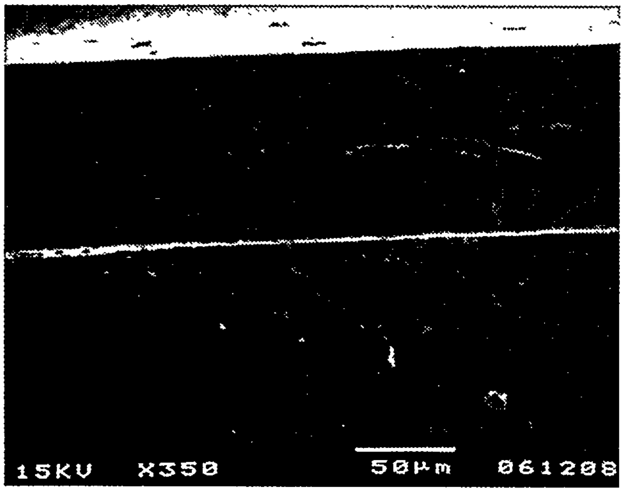

Figure 13: Pt Reference Electrode within the YSZ Electrolyte

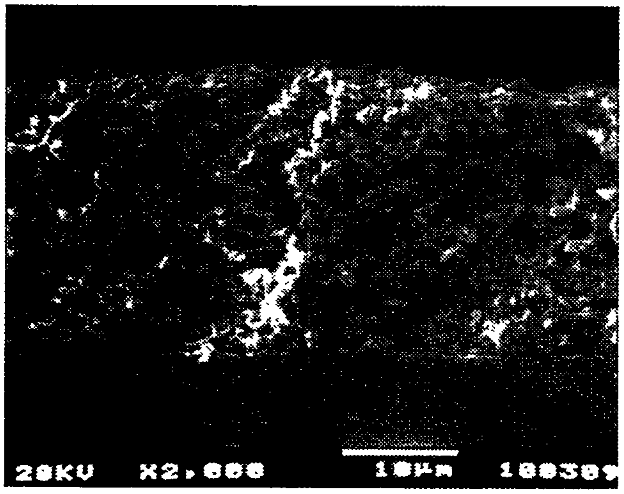

Figure 14: Typical Screen-printed Ni:YSZ Anode

Different cathode powders of composition $La_{0.79}Sr_{0.2}MnO_3$ (LSM-20) were made by various techniques (Pechini liquid mix, glycine - nitrate, and mixed oxide) to give powders of varying particle size and surface area. Powder crystallinity, phase and surface area were characterized by X-ray diffraction and BET techniques as a function of calcination temperature. Cathode powders were prepared into pastes by a similar method as described above, screen printed onto the YSZ electrolyte and fired at various temperatures (1000 - 1200°C). Various cathode microstructures have been developed for the same firing temperature (1100°C/1 hour) with different processing techniques (Figures 15 - 18).

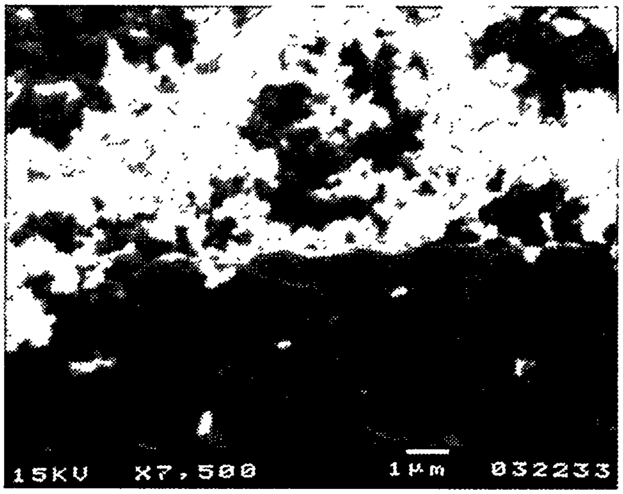

Figure 15: Screen Printed LSM Cathode → Glycine Nitrate Precursor Calcined at 800°C - 1h; fired on at 1100°C - 1h.

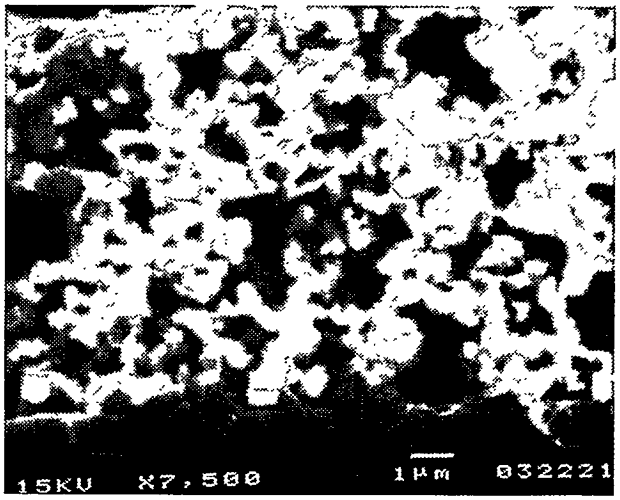

Figure 16: Screen Printed LSM Cathode → Pechini Precursor Calcined at 900°C - 1h; fired on at 1100°C - 1h.

A Pt current collector was printed onto the LSM-20 to mimic a Pt mesh screen. A typical microstructure of the Pt current collector is given in Figure 19.

Dense YSZ rings with a thickness of ~ 400µm were cemented on both sides of the electrolyte for mechanical support and electrical contact.

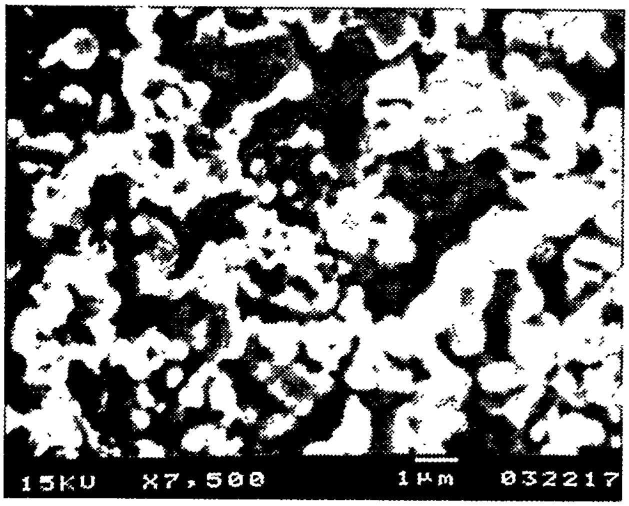

Figure 17: Screen Printed LSM Cathode → Pechini Precursor Calcined at 1100°C - 1h; fired on at 1100°C - 1h.

Figure 18: Screen Printed LSM Cathode → Mixed Oxides Calcined at 1100°C - 1h; fired on at 1100°C - 1h.

Sample Characterization

The system built for measuring cell performance is shown in Figure 20. 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. Partial pressures of O_2 are controlled by O_2 - N_2 mixtures or air on the cathode side and H_2-H_2O, and H_2-N_2-CO_2 mixtures on the anode side. Values obtained from the ZrO_2 sensor are in good

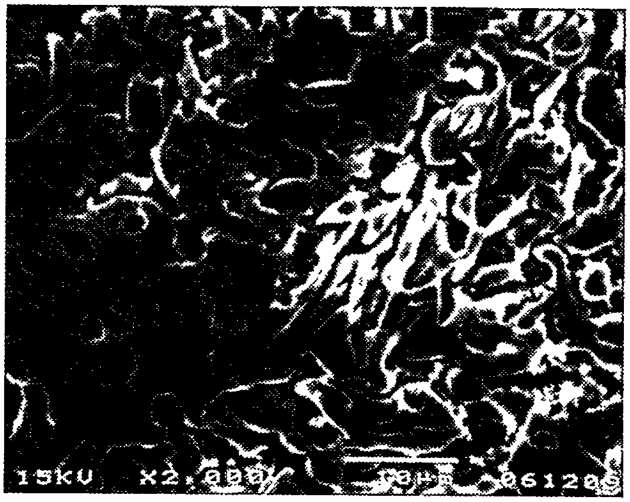

Figure 19: Screen Printed Pt Current Collector.

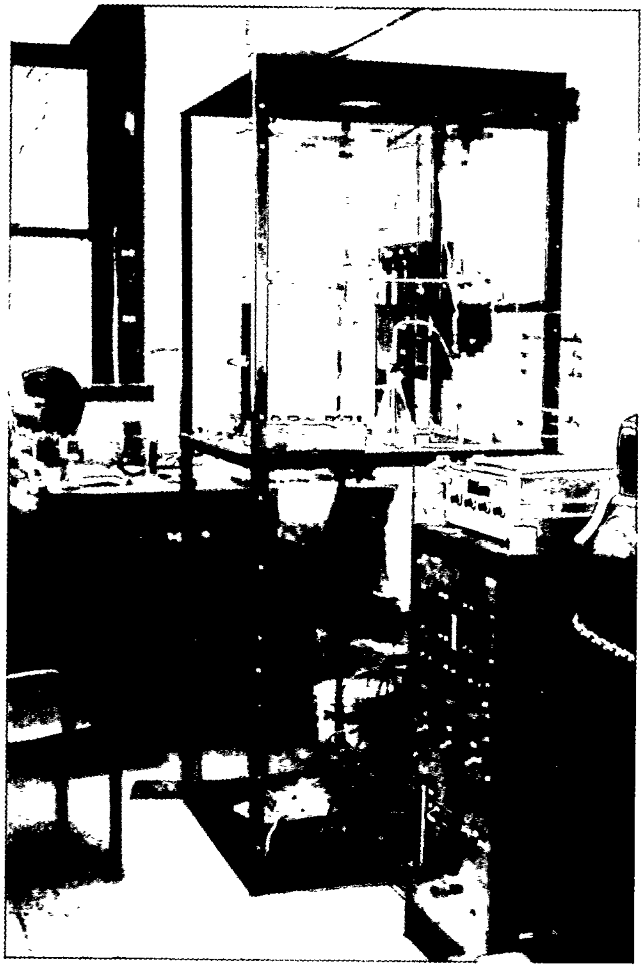

Figure 20: Cell Measurement System

agreement with values derived from thermodynamic data.

Electrochemical measurements are carried out using a three electrode configuration which allows for separation of electrode effects during operation. Pt lead wires are used on the cathode side for electrical connection to both the cathode and reference electrodes while Ni wire is used on the anode side. Electrical characterization, I - V, current density, and interfacial characterization, is performed using a Keithley 197 Microvolt DMM, a Hewlett Packard 4192A Impedance Analyzer, and a computer controlled Solartron Electrochemical Interface (1286) and Impedance/Gain-Phase Analyzer (1260).

RESULTS AND DISCUSSION

Electrochemical data obtained at present has been focused on the overpotential associated with the anode, namely, coarsening of the Ni particles. All cell have an effective electrolyte thickness of 200μm with a Pt reference electrode buried in the center of the electrolyte. The same cathode ($La_{0.79}Sr_{0.2}MnO_3$) has been used for all experiments and was fired on the electrolyte at 1100°C for 1 hour. The only variables to present are the anode composition, Ni content, firing temperature, and gas composition.

Experiments were initially performed on YSZ-NiO anodes containing 43 volume % Ni after reduction with various firing temperatures (1250, 1300, 1400, and 1500°C). Cells were measured with air as the oxidant and a H_2-N_2-CO_2 mixture as the fuel. The theoretical open circuit voltage is 1.00 V. Plots of cell voltage, anode overpotential, and cathode overpotential versus current density measured under load are given in Figure 21. Anode and cathode overpotentials are the voltages measured from the electrodes (cathode or anode) to the Pt reference electrode and do not include any IR losses associated with the anode, cathode, and electrolyte. The IR drop across the electrolyte and anode at the given current density does not have any significant contribution to the total anode overpotential although the IR loss across the cathode may contribute to the total cathode overpotential. Sheet resistance measurements need to be performed on the cathode material to successively separate the cathode overpotential into a IR drop and a nonohmic loss.

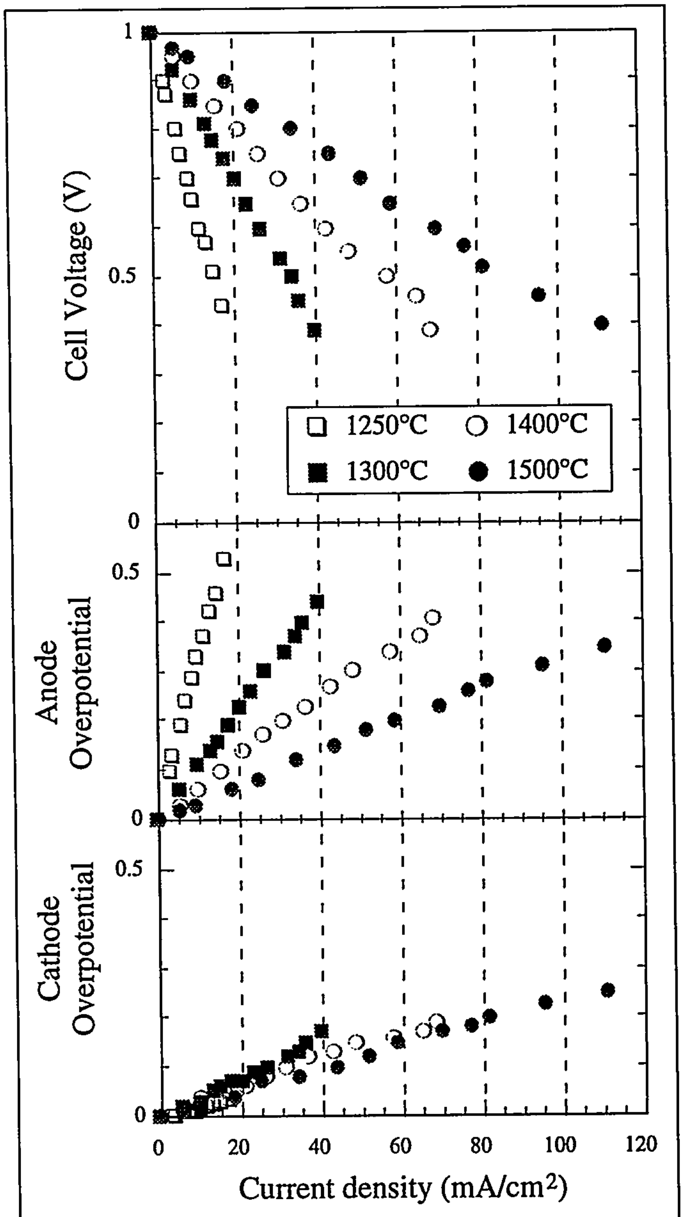

Figure 21: I-V Results on Cells with Anodes Sintered at Different Temperatures

From Figure 21 it can be seen that the cell performance increases as the firing temperature increases. The increase in performance with firing temperature is related to the decrease in the potential drop across the anode. The higher firing temperature may create a more rigid network of YSZ-YSZ contacts thus reducing the tendency of the Ni to sinter. Figure 22 is an SEM micrograph of an anode fired on at 1400°C, reduced and operated under load for 8 hours. The YSZ particle size is approximately 1 µm whereas the Ni particles have coarsened to approximately 5 µm. Before reduction the YSZ and NiO particles were both roughly 1 µm (Figure 14). The cathode overpotentials are essentially the same for a given current density for all samples. Figure 23 shows the cathode microstructure after load for 8 hours; no changes in microstructure are evident.

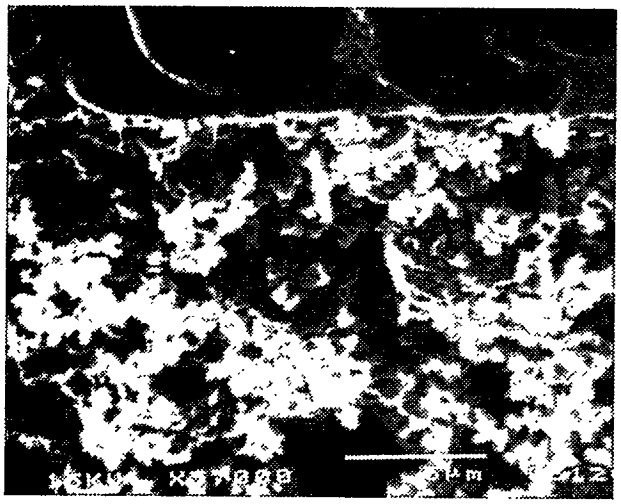

Figure 22: Anode Fired on at 1400°C; Operated as a Cell for 8 h at 1000°C.

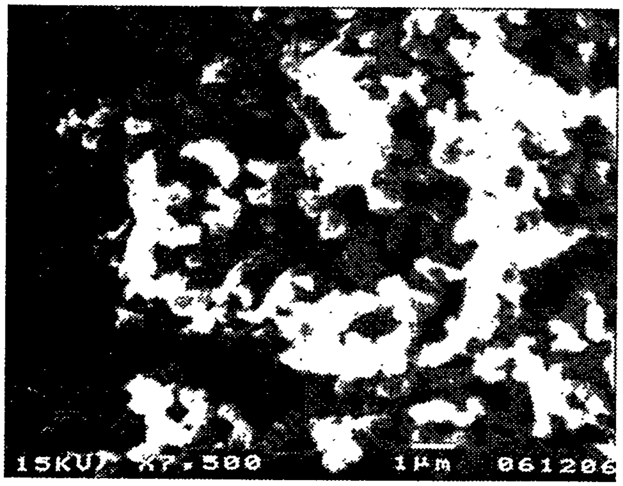

Figure 23: LSM Cathode after Operation as a Cell for 8 h at 1000°C.

The effect of Ni content was investigated with a constant anode and cathode sintering temperature, 1400 and 1100°C, respectively. The same experimental parameters were used as above (i.e. gas composition, electrolyte thickness, and placement of Pt reference electrode). Figure 24 shows the improvement of the cell with a higher loading of NiO, 54 versus 43 volume % Ni after reduction. The anode overpotential is smaller for a higher Ni loading for a given current density

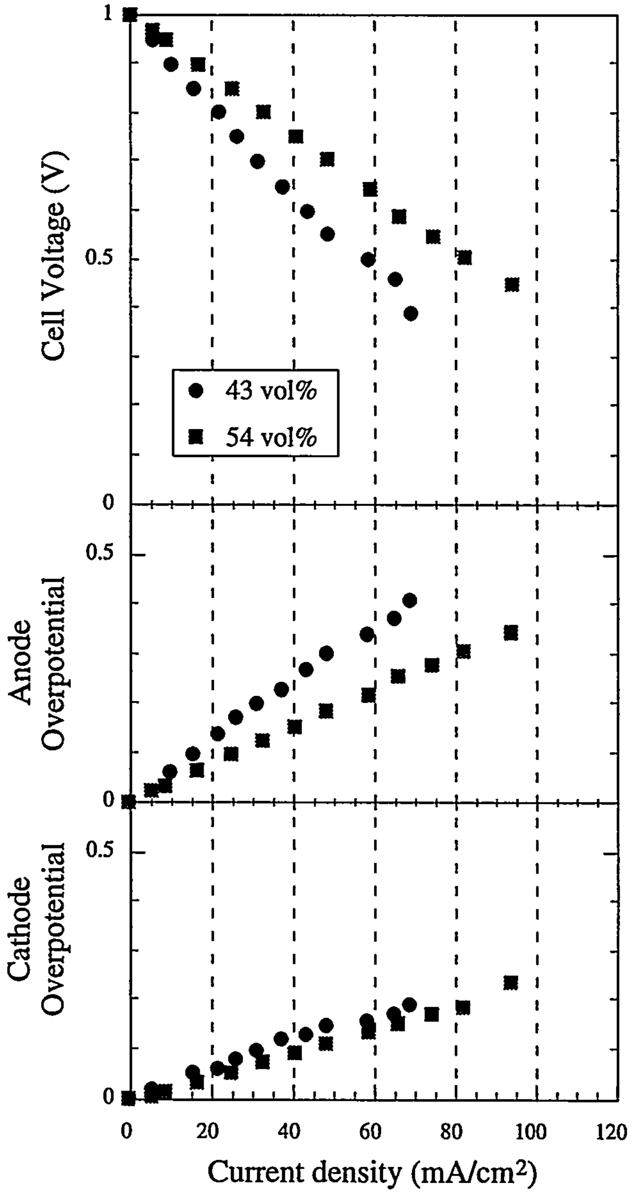

Figure 24: I-V Results on Cells with Anodes Containing Different Amounts of Ni

whereas the cathode overpotentials are essentially the same. A reason for the improvement with a higher Ni loading may be that a higher percentage of contact points (three phase boundaries) are present which increase the electrochemical oxidation reaction rate.

The anode composition was also investigated by replacing YSZ with Sm doped CeO_2, a mixed conductor at low oxygen partial pressures. Figure 25 shows the results of samples containing 54 volume % Ni after reduction with YSZ versus Sm doped CeO_2. Experimental conditions are the same as described above. There is no significant difference between samples but the Sm doped CeO_2 sample does show a slight improvement and a lower overpotential. A possible explanation for the similar results may be that the current density is not high enough to see a significant difference between the two samples. Future investigations will be focus on improving cell performance (i.e. microstructure, anode thickness, etc.) such that the anode composition (YSZ vs. CeO_2) can be better understood.

Anodes composed of 10% MgO doped into NiO to form a solid solution $Ni_{0.9}Mg_{0.1}O$ was investigated as a function of volume % Ni after reduction. Experimental conditions were similar to those given above except H_2 bubbling through H_2O at room temperature was used as fuel. YSZ was used as the other component in the cermet. Figure 26 shows the effect of Ni content for samples with 10% MgO, MgO additions, and gas composition on cell performance. There is a small improvement with increased Ni content for samples containing 10% MgO. The higher Ni loading may increase the number of reaction sites and thus performance as stated earlier. From the data in Figure 26, the improvement in cell performance can be attributed to MgO additions or gas composition, although, the exact parameter(s) controlling performance is not known. Experiments aim at understanding this need to be performed (i.e. YSZ-Ni cermet without MgO utilizing H_2/H_2O as the fuel). Figure 27 is an SEM micrograph of a Ni-MgO-YSZ/YSZ interface reduced at 1000°C for 8 hours. From the micrograph it can be seen that the MgO is successful in reducing the coarsening effect of the Ni in comparison to a Ni-YSZ cermet with no MgO additions (Figure 22).

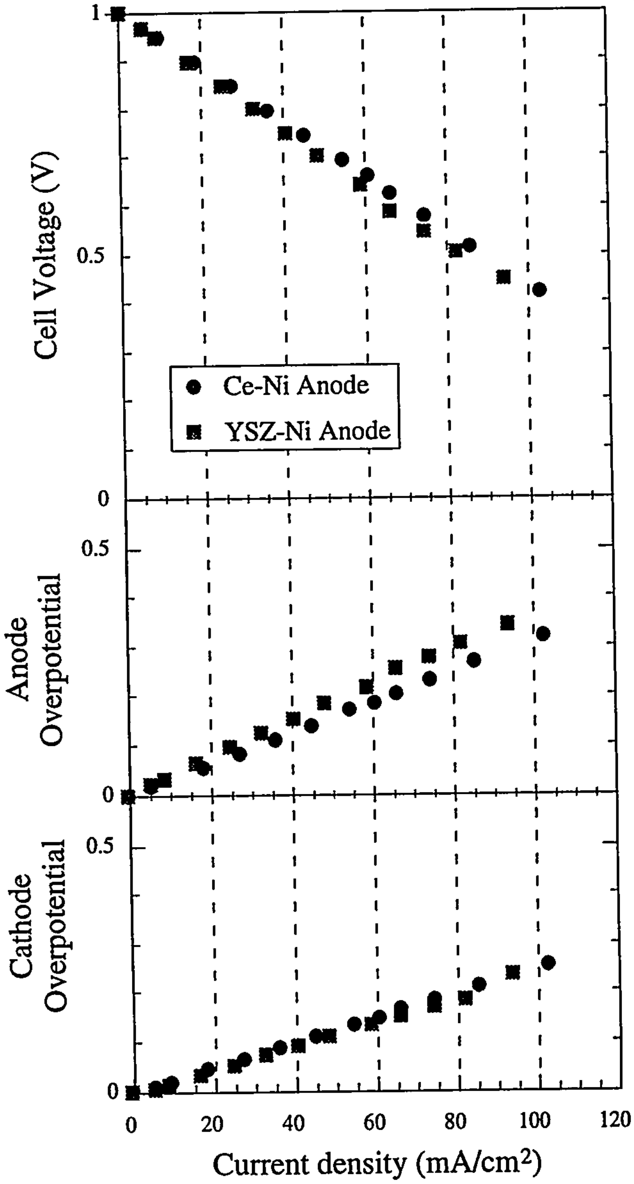

Figure 25: I-V Results on Cells with Anodes Containing Ni: CeO_2 and Ni:YSZ.

Major Achievements During the Past Year

- Developed processing skills necessary to fabricate single cells.

- Incorporated a Pt reference electrode into the electrolyte for separation of electrode effects.

- Developed processing ↔ microstructure ↔ property relations for a number of anodes.

- Developed experimental techniques for measuring cell performance.

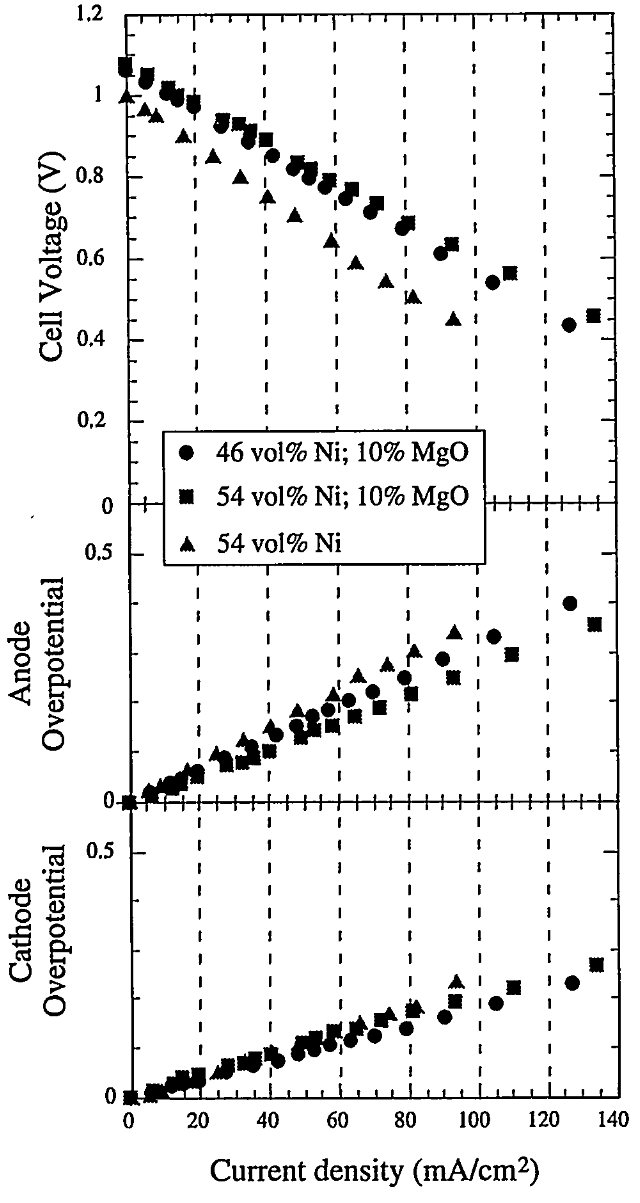

Figure 26: I-V Results on Cells with Anodes with and without MgO Additions to Stabilize the Ni Size

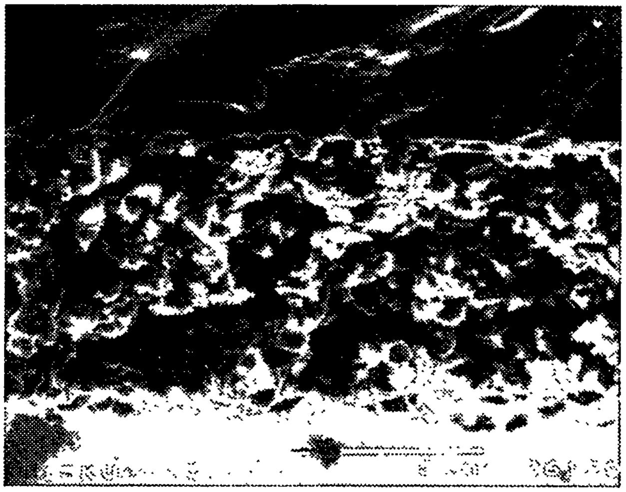

Figure 27: Anode Structure Stabilized with MgO Additions

FUTURE WORK

With respect to the major thrusts of this program:

LACrO3 SINTERING STUDIES

- Continue development of a non-liquid phase sintered $LaCrO_3$-based material sinterable in air.

- Optimize and control the processing conditions associated with $LaCrO_3 \rightarrow$ minimize the particle size and increase the green density.

- Incorporate materials developed in this program into planar cells and measure their performance.

SOFC PERFORMANCE STUDIES

- Fabricate single cells with controlled microstructures of the cathode; incorporate interfacial modifications to improve the catalytic activity.

- Gain a better understanding of the mechanisms involved in improving cell performance via electrochemical and impedance techniques.

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P4 Progress in Carbonate Fuel Cells

CONTRACT INFORMATION

Contract Numbers	49946/49943
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, J. Ernesto Indacochea, Stanley A. Johnson, Michael T. Lanagan, and Sheldon H. D. Lee
METC Project Manager	William C. Smith
Period of Performance	October 1, 1982 to Open

OBJECTIVE

Our objective is to increase both the life and power of the molten carbonate fuel cell (MCFC) by developing improved components and designs. Current activities are as follows:

- Development of lithium ferrate ($LiFeO_2$) and lithium cobaltate ($LiCoO_2$) cathodes for extended MCFC life, particularly in pressurized operation, where the present cathode, NiO, provides insufficient life
- Development of distributed-manifold MCFC designs for increased volumetric power density and decreased temperature gradients (and, therefore, increased life)
- Development of components and designs appropriate for high-power-density operation (>2 kW/m^2 and >100 kW/m^3 in an integrated MCFC system)
- Studies of pitting corrosion of the stainless-steel interconnects and aluminized seals now being employed in the MCFC (alternative components will also be studied)

Each of these activities has the potential to reduce the MCFC system cost significantly. Progress in each activity will be presented during the poster session.

BACKGROUND INFORMATION

Background information was given in four proceedings papers (1-4) that were presented during the previous Contractors Review Meeting. Briefly, we developed a doubly doped $LiFeO_2$ cathode having an acceptable performance; the relative lives of the $LiFeO_2$, $LiCoO_2$, and NiO cathodes are now being measured.

In the studies of distributed-manifold MCFCs, many of the concepts in the original patent (5) were demonstrated. Currently, we are testing 625 cm^2 MCFCs that employ distributed manifolding.

In the high-power-density studies, operation for 1200 h at a specific power >2 kW/m^2 was demonstrated in an MCFC employing a doubly doped $LiFeO_2$ cathode. Current efforts are focussed on design optimization; the goal is a volumetric power density in excess of 100 kW/m^3 for an integrated MCFC system.

The pitting-corrosion studies were initiated in late FY 1994; no data on pitting corrosion were reported at the last Contractors Review Meeting. During this fiscal year, we have observed pitting corrosion on stainless steel and on a nickel-clad stainless steel.

PROJECT DESCRIPTION

In this project, we are investigating ways to increase both the performance and life of the MCFC while reducing its cost.

Studies of cathode materials include basic measurements such as resistivities, Seebeck coefficients, dopant solubilities, and microstructures. MCFC tests to establish cathode performance and life are also conducted.

The distributed-manifold studies include modeling, development of novel methods of dimpling cathodes and anodes (to form gas flow channels), and tests of MCFCs (up to 625 cm^2) to establish their performance and life.

The high-power density studies include modeling, cell design, and measurements of performance and life for MCFCs operated at a current density of 3200 A/m^2 (double the current density usually employed).

The pitting-corrosion studies include potentiostatic and static immersion tests of interconnect and seal materials. The interconnect materials consist of two types (310 and 316) of stainless steel and a nickel-clad stainless steel. The seal material is an aluminized stainless steel. Coupons of these materials are partially immersed in a molten salt (Li_2CO_3-K_2CO_3 or Li_2CO_3-Na_2CO_3) through which either exhaust anode gas or cathode gas is bubbled. The gas stirs the melt and blankets the sample and salt.

Materials from the above studies are generally characterized by scanning electron microscopy and X-ray diffraction.

RESULTS

Cathode Life

Accelerated life tests of MCFCs having $LiFeO_2$ (doubly doped), $LiCoO_2$, and NiO cathodes were conducted for up to 2000 h at high partial pressures of oxygen and carbon dioxide and at a current density of 1600 A/m^2. These tests indicated the potential for long life from both the $LiFeO_2$ and $LiCoO_2$ cathodes. In addition, both cathodes have yielded a voltage of 0.92 V at 1600 A/m^2. This voltage is close to that of the nickel-oxide cathode (0.95 V) under the same test conditions.

Distributed Manifolding

MCFCs (100 cm^2) having distributed manifolding, flat current collectors, and dimpled nickel-chrome anodes and nickel-oxide cathodes (dimpled to form thin gas flow channels) were operated for up to 2000 h with a standard cathode oxidant (air plus 27% CO_2) at one atm. The best had a volumetric power density of 250 kW/m^3 at 1600 A/m^2, which is about twice that of conventional designs under the same test conditions. The volumetric power density of the distributed manifold design under pressurized test conditions is expected to exceed 400 kW/m^3 at 2500 A/m^2 and 2000 W/m^2.

High Power Density

MCFCs having $LiFeO_2$ or $LiCoO_2$ cathodes were operated at a high current density (3200 A/m^2) and at a high specific power (over 2000 W/m^2) for up to 1200 h. These tests demonstrated the high power capabilities of the $LiFeO_2$ and $LiCoO_2$ cathodes.

We also designed a novel MCFC that is expected to have a low cost, a high volumetric power density, and a long life. Due to patent restrictions, details of the new design will not be disclosed during the poster session.

Pitting Corrosion

Pitting corrosion was found on both the nickel-clad anode side and the unclad cathode side of stainless-steel interconnect materials in both the potentiostated and static corrosion tests. On the cathode side of the interconnect, the pitting was found beneath the duplex corrosion layer that usually forms on the cathode side. Pitting-corrosion studies are now being conducted on the aluminized seal material (acceptable samples of it were prepared recently). In addition, other experiments are in progress to aid in understanding the underlying causes of the pitting corrosion.

FUTURE WORK

During FY 1996, the cathode life tests will be extended under a subcontract with M-C Power. The distributed-manifold and high-power-density studies are being merged because they have similar objectives. The pitting-corrosion studies are being expanded, and will address both the interconnect and seal materials in FY 1996. A new activity in FY 1996 will be development of a non-segregating electrolyte.

ACKNOWLEDGMENTS

These studies are being conducted under the auspices of the U.S. Department of Energy, Contract Number W-31-109-Eng-38. This research was sponsored by the DOE Morgantown Energy Technology Center (METC) and by the Electric Power Research Institute (EPRI).

Joseph R. Stapay and Kevin T. Byrne provided technical assistance in the laboratory.

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P5 Carbonate Fuel Cell Matrix Strengthening

CONTRACT INFORMATION

Contract Number	DE-FG05-93ER81512
Contractor	Energy Research Corporation 3 Great Pasture Road Danbury, CT 06813 (203)792-1460 (telephone) (203)825-6150 (telefax)
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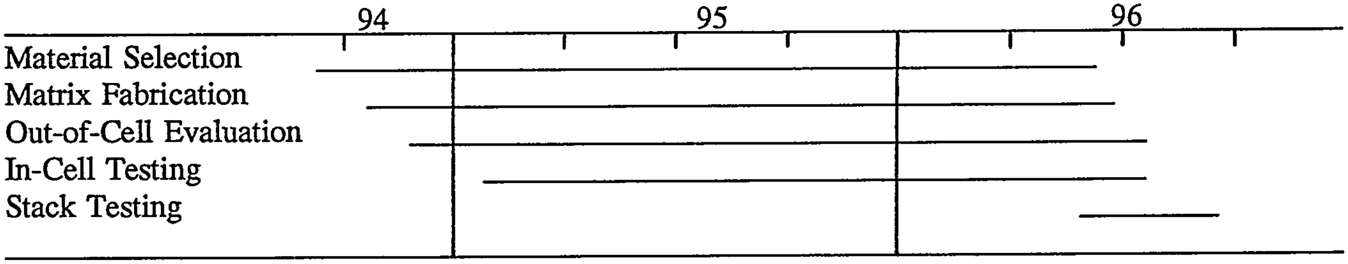
Coinvestigators	Chao M. Huang Richard Johnsen
METC Project Manager	Bruce Harrington
Period of Performance	August 1994 - August 1996 (Phase II)

Schedule and Milestones

Program Schedule



OBJECTIVES

The overall objective of this program is to improve the strength and thermal cycleability of the carbonate fuel cell matrix by reinforcing it with strong as well as stable materials.

BACKGROUND INFORMATION

The present baseline electrolyte matrix is a porous ceramic powder bed impregnated with alkali carbonate electrolyte. The matrix provides both ionic conduction and gas sealing. During fuel cell stack operation, the matrix experiences both mechanical and thermal stresses. Different mechanical characteristics of active and wet seal areas generate stress. Thermal stress is generated by nonuniform temperature distribution and thermal cycling. A carbonate fuel cell generally may experience planned and unplanned thermal cycles between 650°C and room temperature during its 40,000h life. During the cycling, the electrolyte matrix expands and contracts at a different rate from other cell components. Furthermore, the change in electrolyte volume associated with freezing/melting may generate additional thermal stress. Strengthening of the matrix may be beneficial for longer-term stability of the carbonate fuel cell with respect to repeated thermal cycling.

Ceramic particulates and fibers have been used extensively for strengthening ceramic composites (1,2). These types of materials have also been proposed for reinforcing carbonate fuel cell matrix (3). The choice of materials stable in carbonate is rather limited. Strong commercial ceramic fibers (e.g., Al_2O_3) may not have sufficient long-term (40,000 hours) stability in carbonate electrolyte. The γ-$LiAlO_2$ fibers are expected to be stable in carbonate, but currently available ones are nonuniform, defective and weak in structure; their incorporation into matrices has shown no improvement in thermal cycleability in cell testing at ERC. The mechanical property requirements for the ceramic particulates are less stringent, although the incorporation of the particulates may be less effective (1). Nevertheless, the particulates are a lower cost alternative to the fibers. So far, no systematic investigation of the various materials for matrix strengthening has been carried out.

Based on the above discussion, a stable matrix strengthening scheme needs to be defined for long useful life. The effects of the strengthener shape, size and content are yet to be thoroughly investigated. Furthermore, a strengthener stable in a carbonate fuel cell may not have been identified yet. Therefore, there is a need to develop a stable matrix strengthener to optimize the strengthening schemes for further enhancing matrix strength.

PROJECT DESCRIPTION

The major efforts in this SBIR research include: 1) developing and selecting strengtheners and matrix fabrication techniques, 2) out-of-cell evaluation, 3) in-cell testing, and 4) stack testing. The technical approaches are discussed below.

Preliminary development of strengtheners and matrix casting process modifications were carried out in Phase I. The directions to develop stable strengtheners and definition of the processes for implementing the potential strengtheners were identified. A single-cell test was performed to demonstrate the benefit of the proposed strengthening scheme.

In Phase II, manufacturing of the desired strengtheners is being optimized via out-of-cell and in-cell evaluations. Screening of commercial and experimental ceramic fibers and particulates of various compositions, shapes and sizes is first carried out. The experimental strengtheners are being developed either in-house or jointly with fiber developers. The strengtheners thus screened (based on strength and stability considerations) will then be incorporated into the matrix (at different contents) for out-of-cell and in-cell evaluation. The matrix tapecasting process will be adjusted as necessary to uniformly disperse the selected strengtheners.

In the out-of-cell evaluation, the samples are filled with carbonate after binder removal. Metallography, bending and indentation testing are carried out to determine microstructure and mechanical properties. Weibull statistical analysis is conducted to determine matrix reliability. In-cell testing is primarily conducted in $250cm^2$-size single cells. Gas cross-leakage through the matrix is frequently measured to determine the stability of matrix sealing efficiency during thermal cycling. Acoustic emission technique is used to identify test conditions which may result in matrix performance loss. Post-test analysis is performed using chemical methods, SEM/EDAX, and XRD to determine chemical stability of the strengtheners and the mechanisms of matrix strengthening.

Finally, the most promising matrix will be selected for evaluation in a 6-cell 8,000-cm^2 full-area subscale stack test. The optimized strengthening scheme will be recommended for Phase III commercialization.

RESULTS

Strengthener Selection

The considerations for material selection included strengthening potential, stability and cost. In terms of cost, expensive single-crystal whiskers or fibers were not considered. In terms of the fiber mechanical strength, the minimal strength requirement is 700MPa, preferably higher. Porosity and nonuniformity are not acceptable for fibers because they severely degrade fiber strength. High silica content is also not acceptable because silica not only enhances the formation of weak glassy grain boundaries, but also dissolves in carbonate as silicate. Al_2O_3 and ZrO_2 materials may be usable, provided that protective surface oxide layers (lithiated by Li_2CO_3) can form. The morphology and, hence, the protectiveness of the lithiated surface oxide is believed to be strongly affected by the microstructure of the materials. For example, preferred attack of glassy grain boundaries (generally exist in many ceramic fibers) by carbonate with subsequent strength reduction may occur during use. So far, after detailed literature survey, vendor development, and preliminary corrosion testing, several promising materials (including fibers and particulates of Al_2O_3, ZrO_2, $LiAlO_2$, etc.), experimental as well as commercial, were selected for detailed evaluation (see Table 1). Additional strengtheners will be evaluated if they become available in the future.

Table 1. Selection Status of Strengtheners: Two Fibers and Two Particulates Were Selected for Matrix Manufacturing

Strength-ener	Strength	Stability (Short-term)	Status
Fiber			
F1	>1.5 GPa	Good	Selected
F2	>1.5 GPa	Fair	Selected
F3		in Progress	
F4			Under Development
F5	<50 MPa		Not Selected
F6	<50 MPa		Not Selected
F7	>700 MPa	in Progress	
Particulate			
P1	Strong	Good	Selected
P2	Strong	Good	Selected
P3			under Development

Matrix Fabrication

Matrices incorporating the selected strengtheners were then fabricated using processes modified from ERC's baseline. The process variables (viscosity, slurry formulation, doctor blade geometry, etc.) were adjusted so that a uniform dispersion of the strengthener phase while maintaining the strengtheners' aspect ration was successfully achieved. The preferred orientation of the fibers along the casting direction was not significant. All the fabricated green matrices had similar strength, pliability and thickness variation compared to the baseline one, adequate for cell and stack evaluation.

Out-of-Cell Evaluation

Indentation and bending tests were performed on small-size carbonate-filled matrix samples. All the tests were conducted at room temperature where the carbonate was in the solidified state. SEM analysis of the tested samples verified the strengthening mechanism as reported in the literature (1,2). The results of mechanical testing are summarized in Table 2.

Table 2. Results of Mechanical Testing:
Fiber Provides Most Improvements

Strengthener	Fracture Toughness[1] K_{Ic}, MPa$\sqrt{m}$	Strength[2] MPa
None	0.6	10
Particulate P1 (35wt%)	1.5	18
Particulate P2 (35wt%)	1.9	24
Fiber F1 (3wt%)	2.3	27

1: from Indentation Tests
2: from Bending Tests

The fracture toughness from the indentation testing was determined by measuring how far the cracks migrate. The fiber strengthener provided the highest toughening, probably due to fiber debonding and pullout, in addition to crack deflection. Fiber debonding and pullout were observed in the post-test matrix samples. Typical bending test results are shown in Figure 1. Without the strengtheners, the matrix showed brittle failure. With the strengtheners, the tensile strength showed significant improvements, consistent with the indentation results.

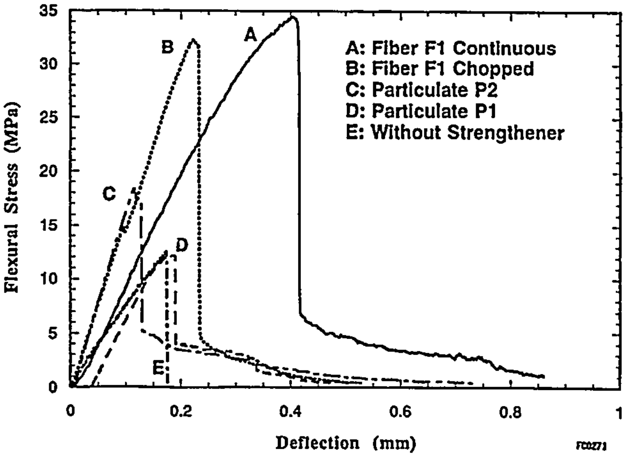


Figure 1. Matrix Bending Test Results:
Fiber Showed the Highest Enhancement in Mechanical Properties

As indicated by the above results, even with only 3% fiber incorporation, the toughness and strength of the matrices have been enhanced significantly. The mechanical tests indicated that the fiber addition provided the highest strengthening effect, followed by particulates.

In terms of the particulate reinforcement, the results of Weibull statistical analysis are shown in Figure 2. The characteristic stresses (63% probability of failure) are 8MPa and 20MPa for small and large particulate matrices, respectively. It is evident that a larger particulate size provides more strengthening. No significant difference in terms of particulate contents (>15%) was observed, consistent with the literature information (1). Similar analysis for the fiber matrices is in progress.

Based on the out-of-cell mechanical evaluation so far, fiber is the best choice for strengthening and larger particulate is preferred. The material stability is being evaluated in long-term corrosion and cell testing.

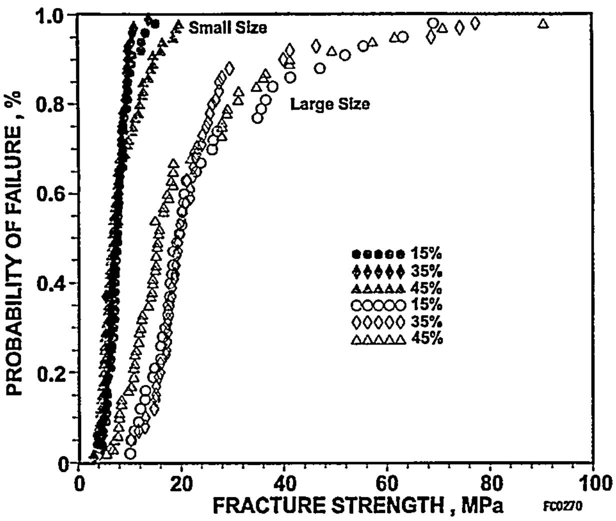


Figure 2. Weibull Analysis of Matrices Strengthened by Particulate P2 of Two Different Sizes:
Larger Particulate Is Desired

In-Cell Evaluation

Matrices incorporating various selected reinforcements were evaluated in 250cm^2 single cells. No difficulties were encountered in implementing these advanced matrices in the baseline design. All the cells were started with ERC's standard start-up procedure. So far, about five fiber cells and five particulate cells incorporating the advanced concept were tested. The primary information recorded is the change in matrix sealing efficiency (i.e., cross-leakage) during thermal cycling. Several of these cells also had acoustic emissions recorded during thermal cycling to provide additional information for diagnosis.

The cell tests so far have shown significant improvements by the strengtheners in maintaining matrix seal efficiency during thermal cycling. The fiber strengthened matrices thermal cycled better than both the particulate ones and the present baseline, as shown in Figure 3. Post-test analysis appears to indicate that one candidate fiber (F1) used here is stable in molten carbonate after 3,000h cell testing (Figure 4). More detailed analysis will be performed in the future (e.g. by TEM) to verify this observation.

Based on the cell thermal cycling results so far, the minimum required strengthener content has also been narrowed down. Higher content is desired for better strengthening. However, the cost of the added strengtheners needs to be considered for selecting the optimal content.

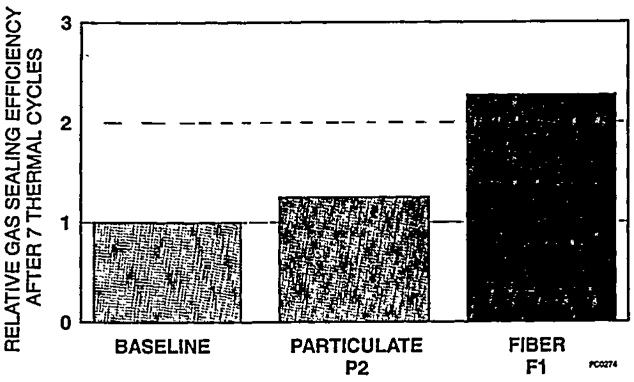


Figure 3. Cell Thermal Cycling Results:
Fiber Matrix Most Effective

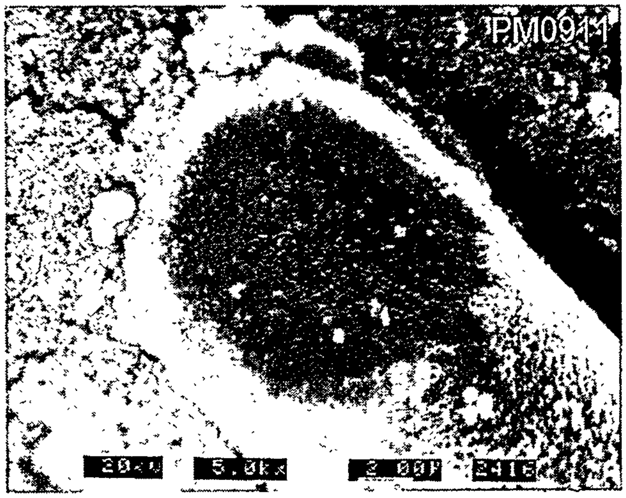


Figure 4. Post-Test Fiber (F1) Matrix (After 3,000h Cell Testing):
Fiber Appears Stable

CONCLUSIONS

- Several promising strengtheners with improved chemical and mechanical stabilities were identified. Fibers provide the highest strengthening effect, followed by particulates.
- Matrix fabrication technique was successfully modified for uniformly incorporating the advanced strengtheners, maintaining the desired aspect ratio.
- Enhanced gas sealing demonstrated using the advanced matrices.

FUTURE WORK

Based on the out-of-cell and in-cell test results available, several promising strengthening schemes were identified for further detailed evaluation. Development effort is in progress with fiber manufacturers to improve fiber quality (strength and stability). Additional candidates obtained recently will also be evaluated. More cell testing to optimize the strengthening scheme will be conducted. The matrix casting method will be simplified and scaled up for large-area manufacturing. Stack evaluation will follow.

ACKNOWLEDGEMENT

ERC would like to acknowledge the support of the U.S. Department of Energy for this SBIR effort.

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P6

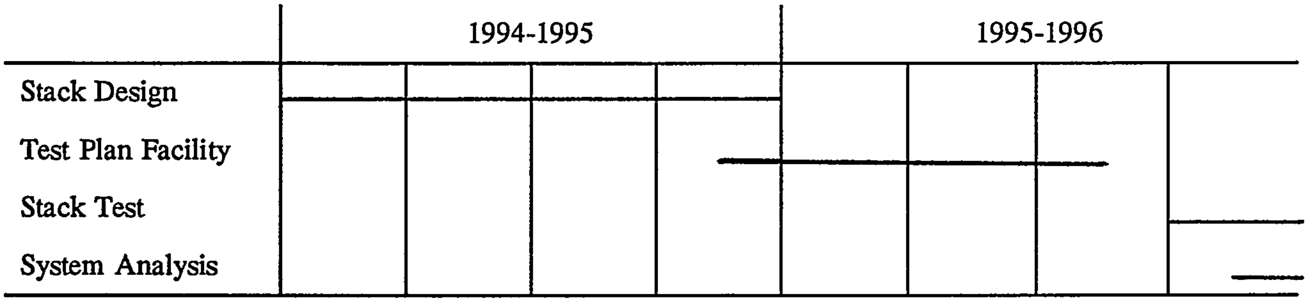
Carbonate Fuel Cell System With Integrated Carbon Dioxide/Thermal Management

CONTRACT INFORMATION

Contract Number	DE-FG05-93-ER81511
Contractor	Energy Research Corporation 3 Great Pasture Road Danbury, CT 06813 (203) 825-6070 (203) 825-6150
Contractor Project Manager/ Principal Investigator	Lawrence Paetsch
Co-investigators	Jianmin Ding, Jennifer Hunt
METC Project Manager	Cliff Carpenter
Contract Period	July 1994 - July 1996

Schedule and Milestones

Program Schedule



OBJECTIVES

The objective of the present work is to define the stack design and system requirements for a commercial-scale carbonate fuel cell with an integrated carbon dioxide management system. Significant simplification and cost reduction of the system is achieved by direct transfer of the fuel exhaust to the oxidant inlet of the fuel cell, thereby eliminating the anode exhaust converter and high temperature piping utilized in conventional system designs.

BACKGROUND

A carbonate fuel cell flow diagram is shown in Figure 1. Carbonaceous fuel at the fuel cell anode provides the source of hydrogen for the anode reaction, while steam and carbon dioxide appear as by-products in the anode exhaust. At the cathode, oxygen and carbon dioxide are consumed. Oxygen is obtained from air, but the carbon dioxide needs to be transferred from the anode exhaust to the cathode inlet. The conventional carbonate fuel cell power plant design collects the anode exhaust from multiple stacks and directs it to a converter where the hydrogen is combusted with excess air. The combustion products, including carbon dioxide, are then directed to the cathode inlet manifolds of the fuel cell. This fuel cell system arrangement is the basis for the direct fuel cell power plant shown schematically in Figure 2.

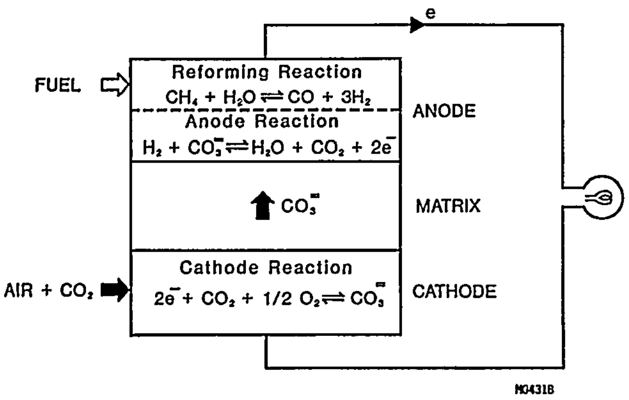


Figure 1. Direct Fuel Cell Diagram: Carbon Dioxide Needs to be Recycled from Anode to Cathode

While this method of recirculating the carbon dioxide is convenient and results in an efficient fuel cell plant, there is a significant cost associated with the anode exhaust converter and high temperature stainless steel piping required for the hot gases. The converter and associated piping can be eliminated and their functions integrated directly into the fuel cell stack itself. This is accomplished by rearranging the stack flow path so that the spent fuel is combusted and the carbon dioxide is transferred *directly* along the face of the stack at the cathode inlet. This concept design is described in U.S. patent 5,422,1950 issued to ERC on June 6, 1995 (1).

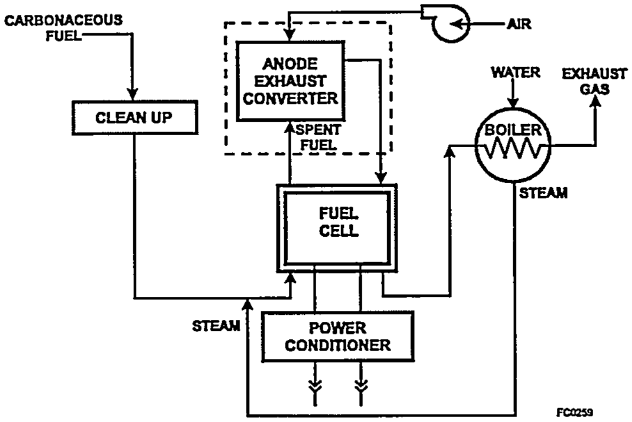


Figure 2. Direct Fuel Cell 2MW Power Plant Schematic: Advanced System Eliminates Equipment Inside Dotted Box

In Phase I of this program, proof-of-concept testing was conducted with laboratory-scale single cells (250 cm^2 cell size). The anode exhaust of the cell was recycled to the cathode which together with the fresh air made up the oxidant stream for fuel cell reaction. Endurance testing was also performed. Stable performance was achieved in a 6,000 hour test (Figure 3). This was the first time anywhere in the world that a direct carbon dioxide transfer without an external anode exhaust converter was demonstrated in a laboratory-scale test. The temperature dependence on fuel conversion was also evaluated, (Figure 4) and the results showed that near complete conversion of the fuel exhaust stream can be expected in the cathode compartment of the fuel cell in commecial size power plants. A short-term test was conducted on a 2kW stack. The stack was able to continue operation with a low cathode inlet temperature, however, it was not tall enough to be thermally representative of a full-height stack. Phase I results are described in detail elsewhere (2).

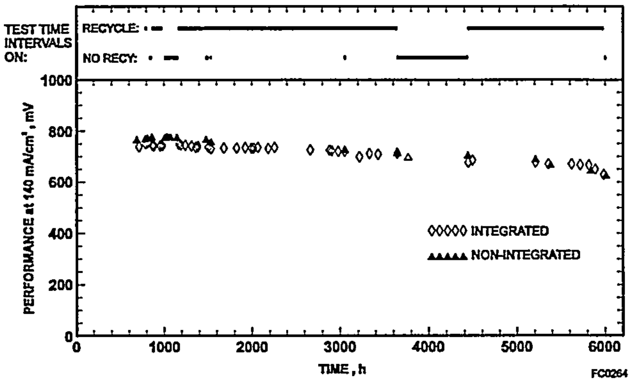


Figure 3. Single Cell Test Results In The Integrated Mode (Fuel Exhaust Recycled To The Cathode):
Stable Performance was Achieved

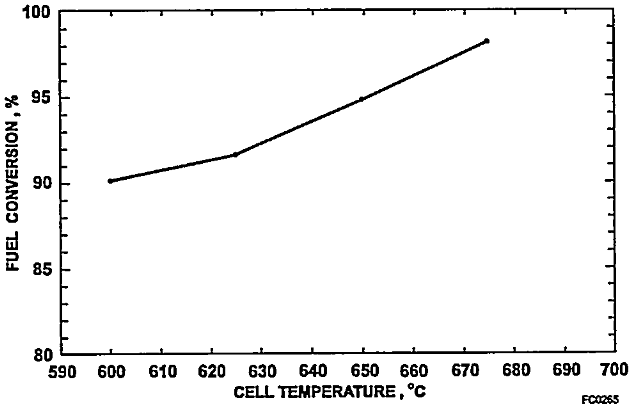


Figure 4. Conversion Of Fuel In The Cathode Gas At Representative Stack Operating Temperatures:
Near-Complete Fuel Conversion is Predicted for Normal Operation

Upon successful completion of Phase I, the Phase II activities were initiated in July 1994 to define the stack design and system requirements for a commercial-scale burnerless carbonate fuel cell stack with an integrated carbon dioxide management system.

PROJECT DESCRIPTION

The major goals of this program are to define the stack design and the system requirements of the integrated design. The approach taken was to maximize the similarities of this stack with ERC's proven baseline stack design and power plant system. Recent accomplishments include a detailed stack design which retains all the essential elements of the baseline stack as well as the power plant system designs. All the auxiliary hardware and external flow patterns remain unchanged, only the internal flow configurations are modified.

RESULTS

Stack Definition

The functions of the anode exhaust converter and piping, which are eliminated, were to:

1. Oxidize the depleted fuel,
2. Recycle the carbon dioxide produced in the anode side of the stack and distribute it to the cathode side for reaction, and
3. Preheat the reactant air during start-up and operation.

The first two functional requirements are integrated *within* the stack design, and the third requirement is satisfied by optimizing the system configuration.

Cross-flow, co-flow and counter-flow arrangements of the fuel and oxidant reactants have long been considered and have been well analyzed for carbonate fuel cell stacks. ERC's baseline design utilizes a cross-flow arrangement, primarily due to its simplicity with respect to gas manifolding arrangement. The functional requirements of 1 and 2 above are most easily satisfied with a counter-flow arrangement since the fuel exhaust must be mixed with air at the cathode inlet. A counter-flow configuration is attractive for this design because it tends to provide for more uniform current density and minimum temperature gradients. The hottest part of the stack, the cathode exhaust, is now on the same face of the stack as the coolest part of the stack, the fuel inlet.

The optimized flow path through the stack elements is shown in Figure 5. Carbonaeous fuel and steam enters the fuel inlet manifold where the fuel is reformed to hydrogen and carbon dioxide. Most of the hydrogen is consumed in the fuel cell reaction and the anode exhaust flows into the stack enclosure. Here, the anode exhaust mixes with air and enters the cathode inlet. The cathode exhaust vents through a manifold to the external piping.

An overall manifold isometric view of the stack is shown in Figure 6. This flow arrangement reduces the number of manifolds by half. Only two manifolds are now required, one for the fuel inlet and one for the cathode exhaust. A cell arrangement showing where the fuel exhaust exits from the anode gas chamber and is mixed with the incoming air before being vented through the oxidant gas chamber is shown in Figure 7. Highlights of this design are:

- Simple, distributed, carbon dioxide transfer.
- Elimination of the fuel exhaust manifold.
- Compact, low risk, stack design.

The design for the 8kW advanced fuel cell stack has been completed. Baseline component designs were selected which are the same as those used in ERC's baseline stack. The stack consists of eighteen 2 ft x 3 ft cells. Experience has shown that the central cells from a stack of this size are thermally representative of taller stacks.

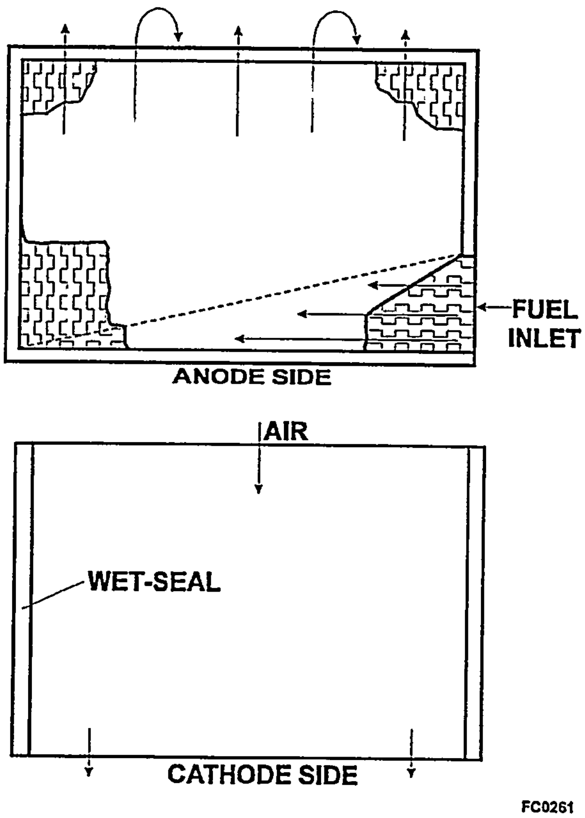


Figure 5. Gas Flow Pattern Through The Various Elements Of The Advanced Carbonate Fuel Cell Stack:
The Path Arrangement Selected for Stack Flow Elements Lends to a Simpler Stack Design

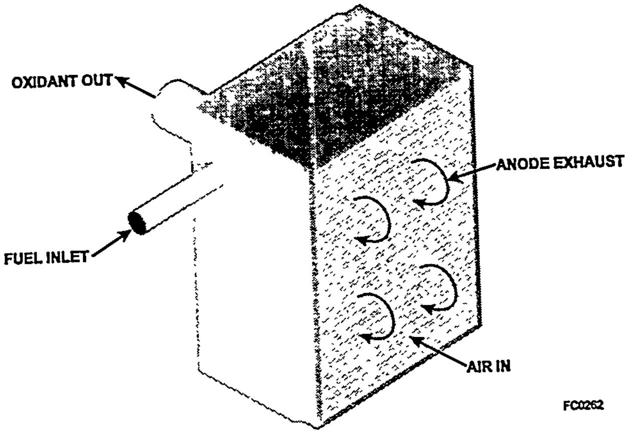


Figure 6. Stack Manifolding With Integrated CO_2 Thermal Management Approach:
Only Two Manifolds are Used

The challenge of this stack design was to obtain a uniform flow profile in a full-size stack while still utilizing baseline cell and stack hardware. The cathode side flow was not altered. Uniform flow distribution was achieved by simple modifications to the baseline anode current collector corrugation.

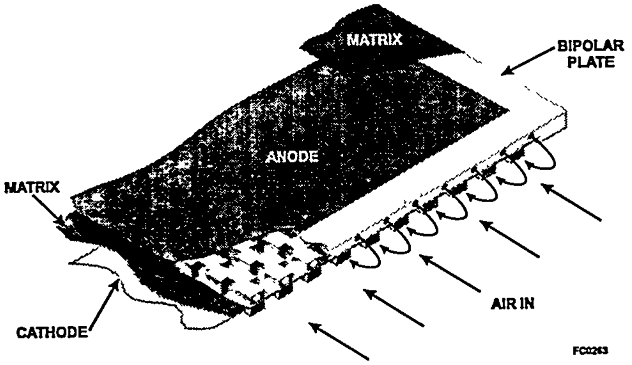


Figure 7. Direct CO_2 Transfer Using Oxidant Inlet Manifold:
A Simple Cell Design was Identified to Provide Direct Transfer of CO_2 to the Cathode Side

A three-dimensional computer model was developed to predict the gas flow distribution in the anode plate. This model is based on the COMMIX-B (3) program and predicts the pressure drop and flow profile based on the type of corrugation used, the flow path and the amount of flow.

The experimental setup shown in Figure 8 was used in conjunction with the computer model. The setup was designed and built to visualize the flow pattern using ammonia sensitive azide paper. Sample ports across the full-size plate allow for local ammonia injections as well as pressure monitoring. Initially, controlled experiments were performed to calibrate the friction factors of flows parallel as well as perpendicular to the corrugation flow channels. Experimental and model-predicted pressure drop data are compared in Figure 9.

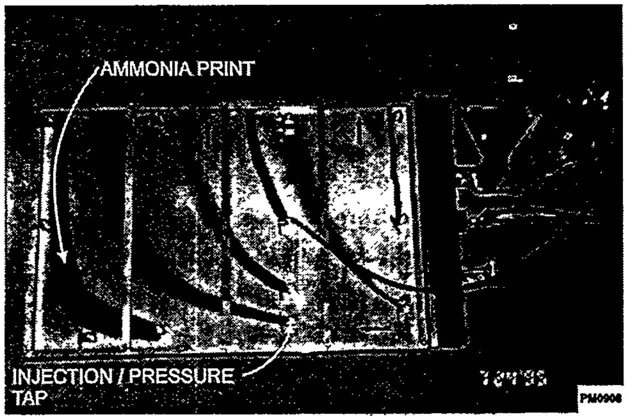


Figure 8. Experimental Setup for Flow Configuration Simulation:
Ammonia and Azide Papers are Used to Visualize Flow Patterns in the Full-Size Corrugated Plate

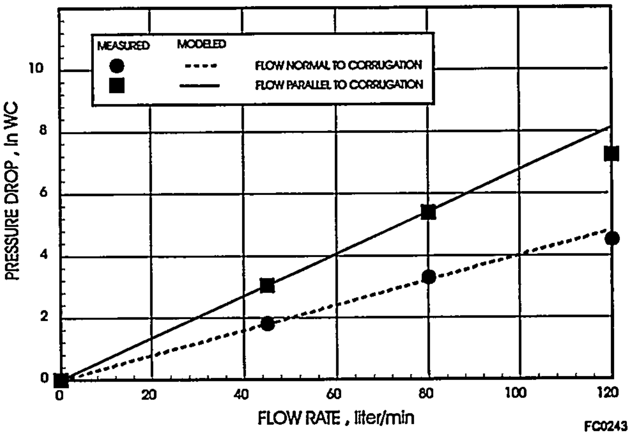


Figure 9. Comparisons of Measured and Computed Pressure Drops in Anode Corrugated Plate:
The Computer Model Closely Predicts the Measured Pressure Drop, Especially at the Full Load Operating Flow Rate of ~45 lpm

Two types of corrugation were modeled and experimentally verified. The first corrugation used was 1994 state-of-the-art anode corrugation.

A second, low-cost corrugation option was also evaluated. This type of corrugation was later chosen as the future standard. Plate design studies, therefore, continued using the low-cost corrugation.

Computer simulations predicted that uniform flow patterns could be obtained with a baffling system. After several baffling studies and extensive computer simulation, an optimized plate design was obtained which incorporates uniform flow and manufacturing simplicity. The experimental and computer modeling results at full load flows are compared in Figures 10a and b. The flow pattern obtained with the ammonia/azide paper in the experimental setup agrees with the model prediction. Both the model and the experimental setup gave a pressure drop of 2.9 inches of water from the fuel inlet. Velocity profiles were computed at these conditions for two distances from the anode exit. The results, shown in Figure 11, predict a ±10% flow velocity variation along the width of the plate. This is well within the design goal.

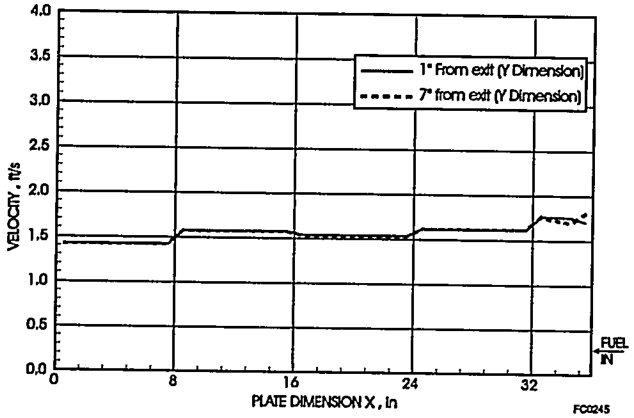


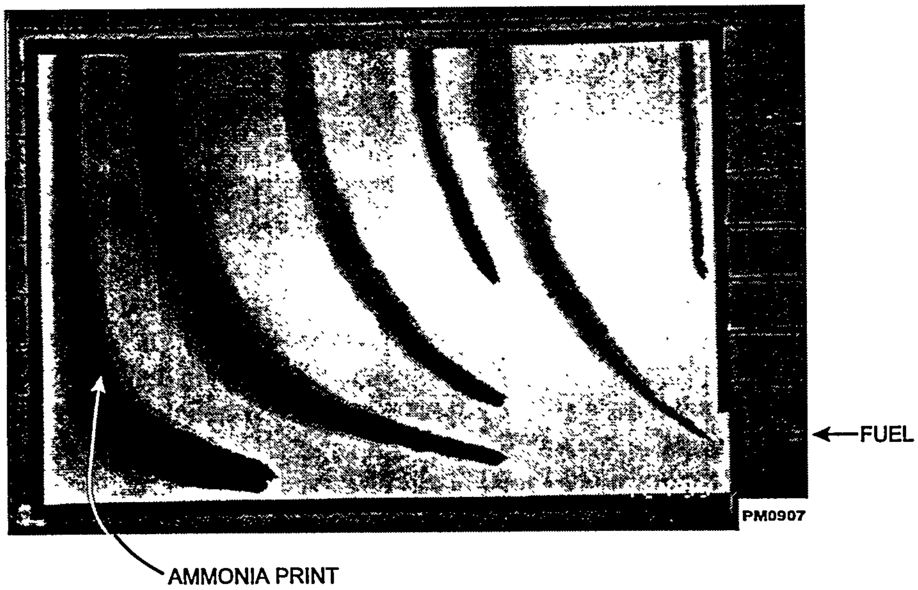
Figure 11. Computed Velocity Profiles with Full Load Flows:
Uniform Velocity Profiles are Projected

Fabrication of this stack will now be initiated. The repeating active components are being provided by ERC's subsidiary Fuel Cell Manufacturing Corporation (FCMC). The auxiliary hardware (compression plates, gas manifolds, etc.) are designed, and the parts are ordered.

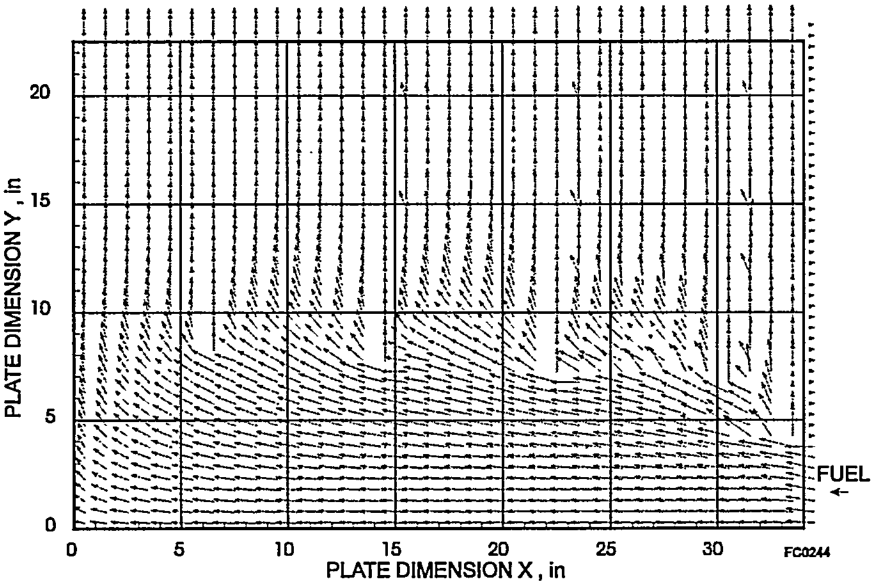
TEST PLAN AND FACILITY

The detailed test plan for the 8kW stack is being finalized. Test conditions will be optimized by varying recycle flow and cathode air inlet temperature as the power output is monitored. Following approximately 500 hours of performance characterization and the establishment of the optimized flow parameter settings, an extended test period is planned. The stack facility available at ERC for baseline short stack tests will be used for this test.

A major effort is focused on defining the operating procedures for a stack with direct and integrated transfer of the fuel exhaust to the cathode inlet. The first step in this process was to review the operating experience obtained from Phase I and ERC's in-house tests. Changes in operating procedures are required because there is no isolation between the fuel and oxidant streams. To accommodate this, the operating strategy was changed for the fuel stream from flow control to differential pressure control. This will protect the anode chambers from air infiltration. After the fuel is exhausted from the anode chambers, it will mix directly with the cathode inlet air along the face of the stack. The flammability of the mixed fuel and oxidant streams was calculated as a function of fuel utilization and temperature, based on the system analysis runs performed in Phase I. These calculations predict where the fuel will combust. At low fuel utilizations, a weak flame is sustainable outside of the stack in the cathode inlet manifold, while at a high fuel utilization combustion will not occur until the mixed gas is preheated within the cathode gas chamber. Therefore, the release of the heat of combustion will be controlled by the operating condition.



a) Experimental Flow Pattern



b) Computer Modeled Flow Pattern: Arrow Length Defines Local Flow Velocity

Figure 10. Experimental and Computer Model Flow Configuration at Full Load Flows: The Flow Pattern Obtained in the Experimental Setup Verified the Computer Model Simulation

First time start-up and conditioning, hot idle, load increase, emergency shutdown, cool down and reheat procedures have all been defined and are currently being incorporated into the test plan.

FUTURE WORK

Near term activities will include:

- Stack Manufacture
- Finalization of Test Plan

ACKNOWLEDGEMENT

The authors wish to acknowledge the support of the DOE/SBIR program.

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1. R. Bernard, "Carbonate Fuel Cell with Direct Recycle of Anode Exhaust to Cathode" U.S. Patent 5,422,195, June 1995.

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3. Argonne National Laboratory, "COMMIX-1B: A Three-Dimensional Transient Single-Phase Computer Program for Thermal Hydraulic Analysis and Multi-component Systems", NUREG/CR-4348, ANL-85-42, September 1985.

P7 High Performance MCFC Using Li/Na Electrolyte

CONTRACT INFORMATION

Contract Number	DE-FC21-95MC30133
Contractor	M-C Power Corporation 8040 South Madison Street Burr Ridge, IL 60521 (708) 986-8040 (Telephone) (708) 986-8153 (FAX)
Other Funding Sources	
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Principal Investigators	Rafael A. Donado Estela T. Ong Chakravarthy I. Sishtla
METC Project Manager	Diane Hooie
Period of Performance	December 19, 1994 - December 18, 1999

ABSTRACT

The substitution of a lithium/sodium carbonate (Li/Na) mixture for the lithium/potassium carbonate (Li/K) electrolyte used in MCFCs holds the promise of higher ionic conductivity, higher exchange current density at both electrodes, lower vapor pressure, and lower cathode dissolution rates. However, when the substitution is made in cells optimized for use with the Li/K electrolyte, the promised increase in performance is not realized. As a consequence the literature contains conflicting data with regard to the performance, compositional stability, and chemical reactivity of the Li/Na electrolyte.

Experiments conducted at the Institute of Gas Technology (IGT) concluded that the source of the problem is the different wetting characteristics of the two electrolytes. Electrode pore structures optimized for use with Li/K do not work well with Li/Na. Using proprietary methods and materials, IGT was able to optimize a set of electrodes for the Li/Na electrolyte. Experiments conducted in bench-scale cells have confirmed the superior performance of the Li/Na electrolyte compared to the Li/K electrolyte. The Li/Na cells exhibited a 5 to 8 percent improvement in overall performance, a substantial decrease in the rate of cathode dissolution, and a decreased decay rate. The longest running cell has logged over 13,000 hours of operation with a decay rate of less than 2 mV/1000 hours.

P8 Technology Base Studies of Long-Term MCFC Performance

CONTRACT INFORMATION

Contract Number	DE-FC21-94MC31078
Contractor	Illinois Institute of Technology 3300 S. Federal- Room 306 Chicago, IL 60616 (312) 567-3035
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Principal Investigator	J. R. Selman
Co-Investigator	M. S. Yazici
METC Project Manager	William C. Smith
Period of Performance	October 27, 1994 to March 27, 1996

OBJECTIVES

Cathode dissolution into the electrolyte matrix and endurance of current collector/separator plate materials are the main life-limiting factors of the state-of-the art MCFC. These components are also major contributors to the total system cost. Therefore, to reduce capital cost, it is necessary to minimize hardware corrosion and increase cell life. This study consists of experimental evaluation of corrosion processes with the objective to further practical;

- understanding of corrosion behavior of alloys and alloy components under cathodic gas conditions. Nickel, iron, cobalt and stainless steels 310 and 316L are analyzed. The experimental study consists of:

1. Observation of open circuit potential (OCP) changes. Surface reactions occuring without net passage of current are proposed based on this measurement.

2.Applying cyclic voltammetry, which provides information about the possible electrode reactions at different stages of polarization.

3. Applying AC impedance to support the result of tasks 1 and 2 at different stages of oxidation, and data analysis by means of equivalent circuits. Open circuit conditions as well as positive and negative polarization are used in the impedance measurements.

4. Surface analysis of the electrodes by SEM-EDX and X-ray diffraction.

To obtain more information from electrochemical measurements a novel approach has been applied in cell design. In this two-electrode approach, one electrode is fully immersed, and the other is wetted by carbonate melt via an alumina tube which has a thin film on it. While camera observation of these two different electrode conditions will identify the effect of carbonate wetting on the state of the surface, electrochemical measurements enable a quantitative comparison between complete submersion and wetting by a film of carbonate.

BACKGROUND

For successful commercialization, it is necessary to raise the power density of the MCFC significantly, and to increase the life time of MCFC stacks. The power density of present MCFC designs under standard conditions is approximately 0.12 W/cm^2, i.e., 0.75V at 165 mA/cm^2, using reformed natural gas as fuel. Cost reduction of the MCFC stack requires raising the power density to 0.75 V at 250-300 mA/cm^2, by drastically reducing the ohmic and polarization losses. Increased power density is made possible by optimal choice of operating conditions, e.g., by pressurized operation. Present information indicates that performance gain is greatest in the 1-3 atm pressure range, mostly due to decrease of cathode polarization. However, pressurization may at the same time accelerate corrosion processes. This is certainly the case for the NiO cathode, but the effect on alloy corrosion is less clear.

Corrosion not only affects the corroding materials but also causes changes in electrolyte composition and physical properties that may give rise, e.g., to electrolyte streaming in the wet seal area and to creepage of electrolyte on corroding surfaces. Most of the electrolyte loss in MCFC operation is due to corrosion, and occurs in the initial stages of operation. There is, therefore, an intimate connection between corrosion of metal and loss of electrolyte and the wetting of metallic and oxidic surfaces by electrolyte.

PROJECT DESCRIPTION

This project aims to improve the long-term power density and endurance of the MCFC by

- developing cathode and anode performance models based on improved agglomerate models. This will identify structural parameters and operating conditions.
- investigating metal oxidation and corrosion of stainless steel(i.e., 310 and 316L).

Electrochemical measurements and surface characterization are the main experimental tools of the project. A fundamental basis for systematic interpretation of alloy corrosion in molten carbonate is developed. The interaction of wetting, migration and corrosion in alloy corrosion is explained in terms of wetted and immersed electrode configuration. This research identifies the driving forces for wetting and corrosion at metal/metal-oxide interfaces in contact with gas atmospheres of varying composition.

In this paper, only experimental part of the project(metal oxidation and corrosion) is covered

RESULTS

Nickel Dissolution: Cyclic Voltammetry

When NiO dissolves in a carbonate melt, the dissolved nickel ions can be electrodeposited on an auxiliary electrode made of inert material during cathodic polarization and stripped off again in a following anodic sweep[Ref. 1&2]. When the potential is scanned through cathodic and anodic limits, the magnitude of the peak currents is related to the scan rate and the concentration of metal ions in the melt. Fig 1 is a typical CV showing electrodeposition and stripping of dissolved nickel on a gold electrode. The anodic scan oxidizes the nickel at the surface of the gold. There is also a shoulder at -350 mV assigned to the lithiation of NiO. The variation of peak current with the square root of scan rate is plotted in Fig 2.

Nickel Dissolution: Stripping Voltammetry

In this part of the experiment, dissolved nickel is deposited on a gold electrode for different holding times at different scan rates. Fig 3 shows current-potential behavior at 50 mV/s anodic scan rate for 5 to 425 seconds holding time. The peak around -350 mV is a feature that is not observed on cyclic voltammograms. It is interesting that this peak becomes stronger in magnitude as the holding time is increased. The dependence of the peak current on the scan rate rather than square root of scan rate suggests that the peak at -350 mV is due to O^{2-} or S^{2-} adsorption on the electrode surface.

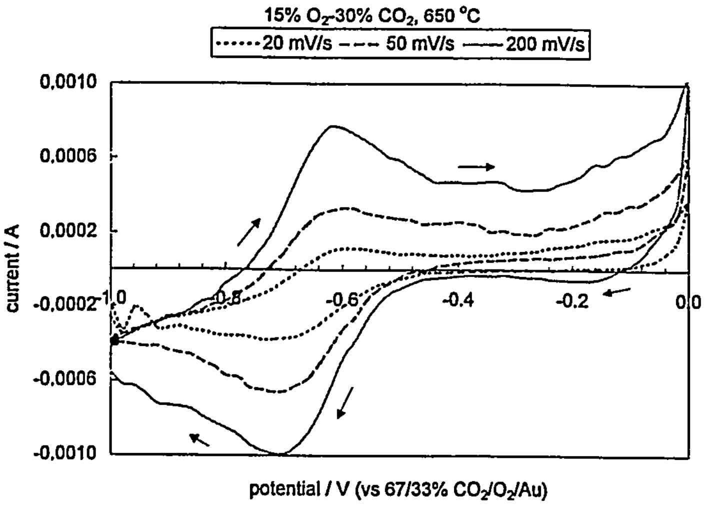

Figure 1. Cyclic voltammograms of nickel deposition and dissolution at an inert electrode.

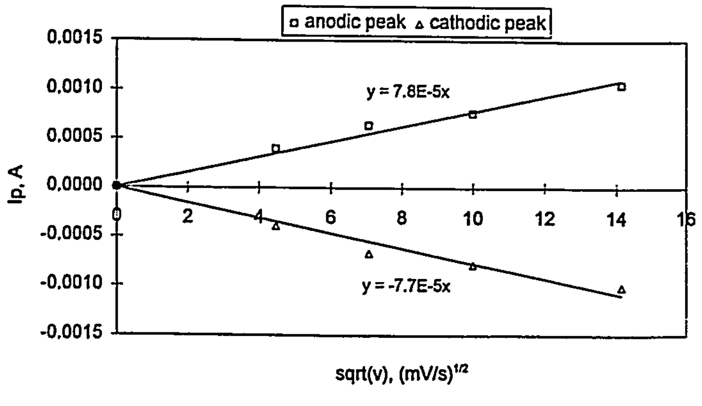

Figure 2. Variation of anodic and cathodic peak current with square root of scan rate

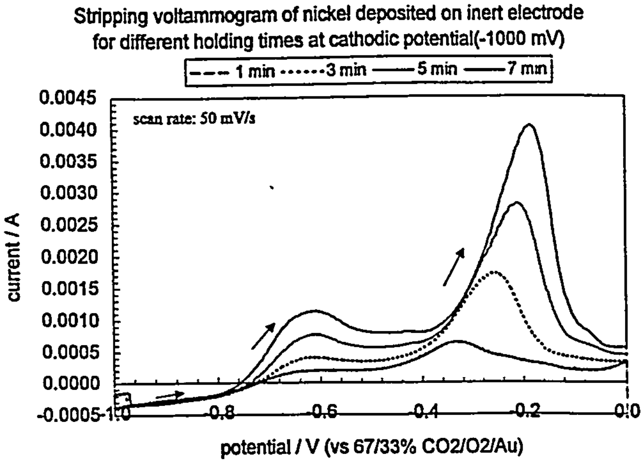

Figure 3. Stripping voltammograms of nickel deposited on an inert electrode for different holding times at cathodic potential(-1000 mV).

Oxidation/Corrosion of Metals and Alloys: Open Circuit Studies

To study oxidation and corrosion, electrochemical measurements are carried out using two electrode designs. One electrode is fully immersed into the melt, the other electrode is mounted on the surface of an alumina tube which provides a substrate for the wetting of the electrode material by a film. Fig 4 is a schematic of the design used. Both electrodes are subjected to the same oxidizing environment and their responses compared.

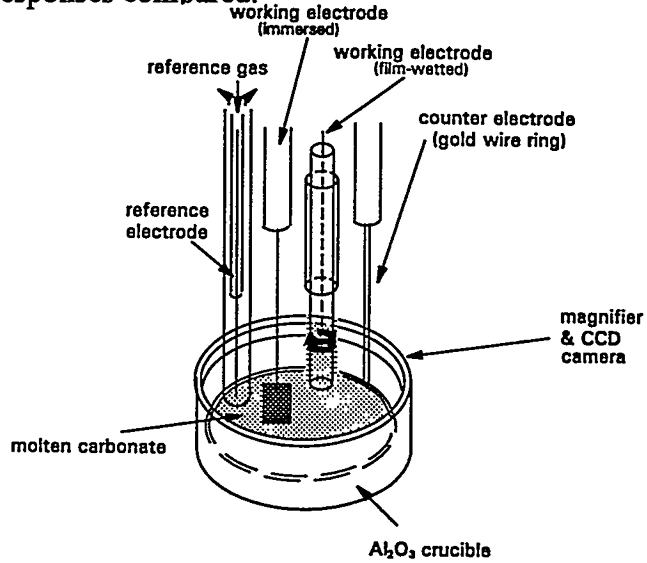

Figure 4. Wetted and Immersed Electrode Configurations in Molten Carbonate.

Fig 5 shows the open circuit potential decay of geometrically similar electrodes, of nickel, iron, cobalt, stainless steel 310 and 316L, under oxidant atmosphere. The response of the nickel electrode to contact with molten carbonate is similar to that observed in previous work[Ref.3] (Fig 5a). When iron metal immersed in molten carbonate initially under CO_2, gas bubbles are observed on the surface although the open circuit potential(OCP) is stable. The OCP change with time(Fig 5b) for both electrodes is similar under oxidant gas. Oxide has a good electronic contact with the base metal, but it is not protective. It takes a long time to recover OCP following polarization measurements.

The potential of immersed cobalt stays at the rest value for almost 200 h.(Fig 5c). This may be explained by convection caused by CO_2 evolution, which makes oxidized and lithiated cobalt lose contact and dissolve into melt. In the case of the wetted electrode, the thin film of electrolyte does permit CO_2 to be released easily from the surface of lithiated CoO. This may explain the OCP shift decay of cobalt/CoO with time.

The OCP of stainless steel Types 310 and 316L under CO_2 is stabilized around -1000 mV(Figs 5d and e). Upon introduction of oxidant gas, the potential of the wetted electrode goes to oxygen reduction potential quickly, i.e., 3 hrs.

The significant differences between the two types of electrode arrangement suggest that half cell studies have to imitate actual MCFC conditions in order to be useful for the improvement of cell materials and for predictions. The film-wetted electrode forms a much more compact and stable passive layer than an electrode immersed in carbonate. During the oxidation process at a film-wetted electrode, the electrolyte intensively moves around the wetted perimeter. The forces that drive this movement at the melt/oxide interface also accelerate corrosion at the oxide/metal interface.

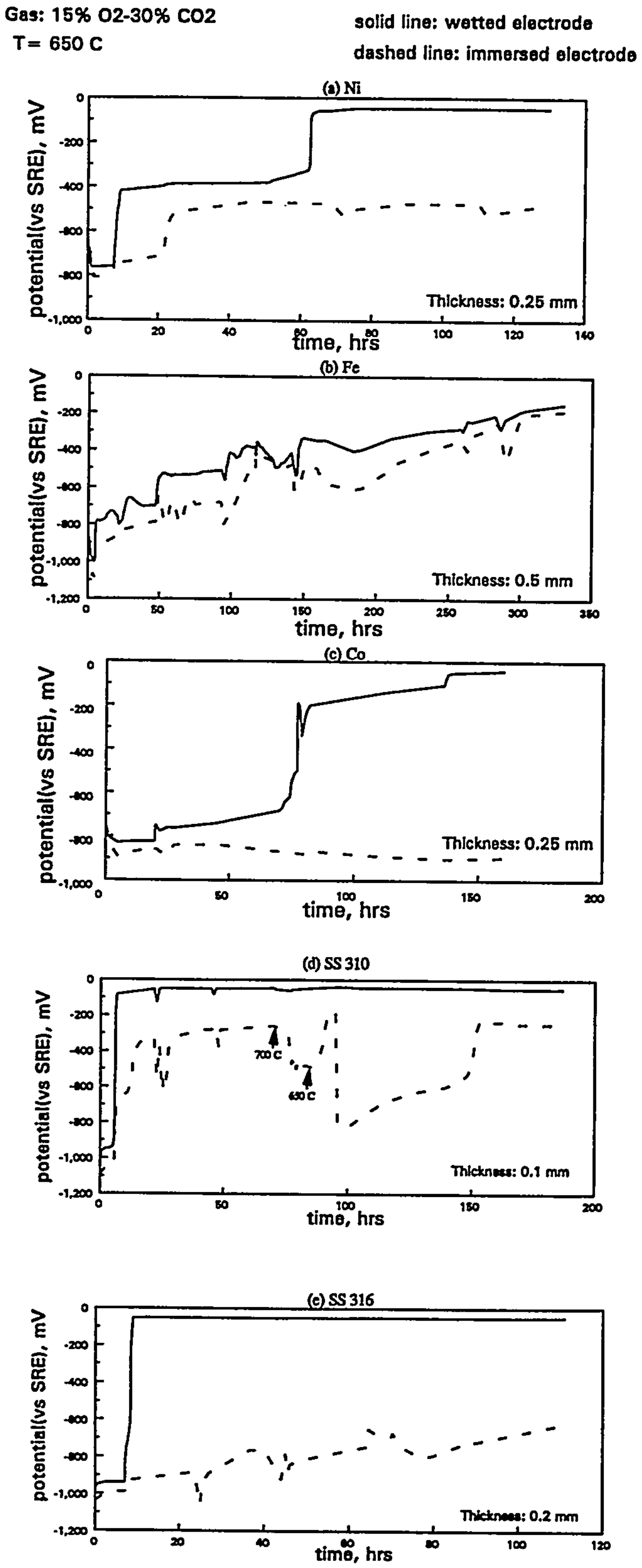

Figure 5. OCP response of nickel, iron, cobalt and stainless steel electrodes under 15%O_2-30% CO_2. Solid line: wetted electrode, dashed line: immersed electrode

Oxidation/Corrosion of Metals and Alloys: Polarization Curves

The polarization response of each metal or alloy is also different. An anodic potential scan causes gas evolution from the surface only at potentials more positive than -600 mV. This leads to the conclusion that the anodic current peak after -600 mV is caused by a process with CO_2 generation. This may be lithiation and/or a higher valent metal oxide formation. Observations for cobalt yield the following conclusions:

1. Very violent surface activity with bubble evolution and splashing starts to take place upon introducing an O_2/CO_2 mixture.
2. For the immersed electrode under CO_2 or 15% O_2-30% CO_2, the potential always oscillates.
3. The electrolyte becomes like slush due to the extreme amount of dissolution into it. Most of the dissolved species deposit on the counter electrode, and on alumina tubes and the surrounding crucible. This behavior is also observed on nickel electrodes, at a slower rate. Iron deposits very little, but after the experiments, it easily dissolved in dilute HCl.

The steady-state polarization of oxide electrodes is shown in Fig 6. The polarization response of stainless steels at slow scan rates is shown in Figs 7 and 8. For Type 310, the initial cathodic potential scan yields only cathodic response. However, Type 316 yields a slight anodic current. This behavior is similar to that observed for nickel, which is related to Ni(III) formation with carbonate decomposition.[Ref.4] This indicates that the two stainless steels have different oxidation characteristics and different protective or non-protective product layers. Since the alloys that we are dealing with here consist of three different elements, sometimes peaks related to the oxidation or reduction of one element may overlap with those for the other two elements, which have the same or approximately the same potentials.

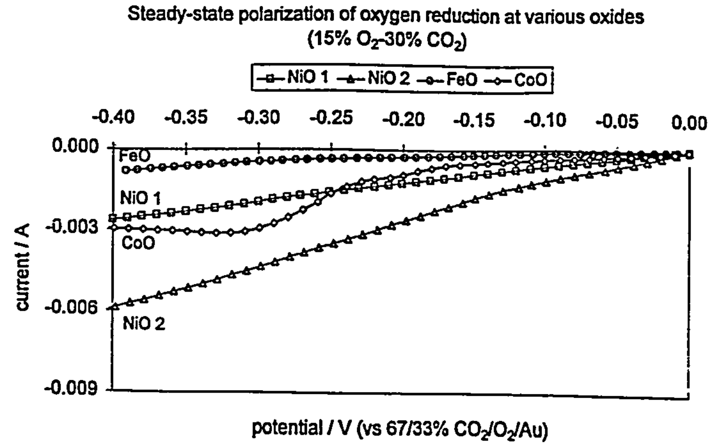

Figure 6. Steady-State Polarization of Wetted Oxide Electrodes.

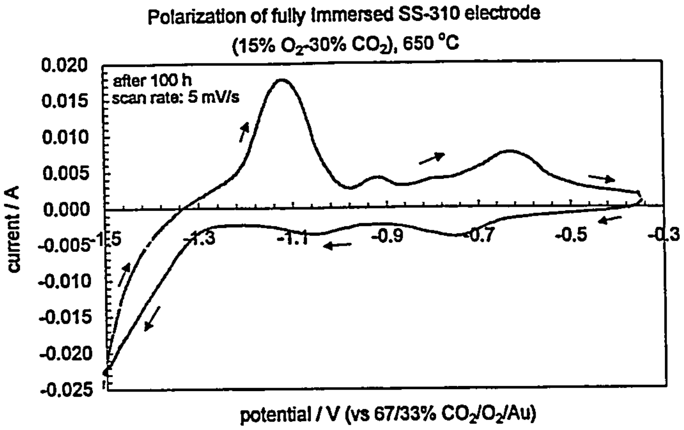

Figure 7. Current-Potential Response of SS310

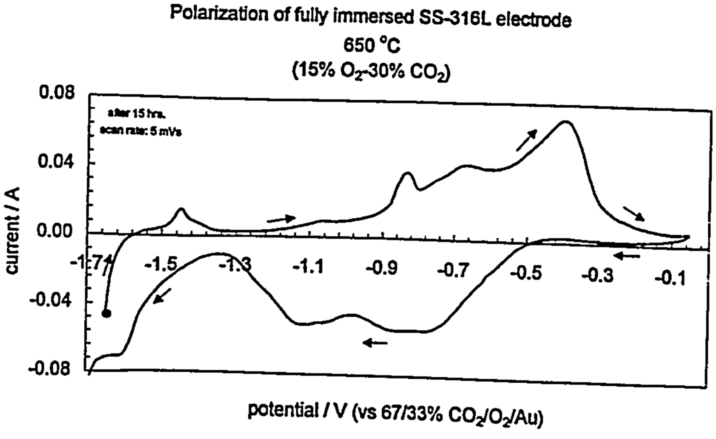

Figure 8. Polarization Curve of SS 316L

P9

Landfill Gas Cleanup for Fuel Cells

CONTRACT INFORMATION

Contract No.	CRADA 95-031
Contractor	Electric Power Research Institute 3412 Hillview Avenue Palo Alto, CA 94304 (415) 855-1057 (415) 855-8501
Other Funding Sources	EPRI TVA ESEERCO NSP NRECA NREL (DOE/EE)
Contractor Project Manager	Douglas J. Herman
Principal Investigator	N/A
METC Project Manager	William Cary Smith
Period of Performance	~ August 1995 to December 1996

ABSTRACT

EPRI is to test the feasibility of using a carbonate fuel cell to generate electricity from landfill gas. Landfills produce a substantial quantity of methane gas, a natural by-product of decaying organic wastes. Landfill gas, however, contains sulfur and halogen compounds, which are known contaminants to fuel cells and their fuel processing equipment. The objective of this project is to clean the landfill gas well enough to be used by the fuel cell without making the process prohibitively expensive. The cleanup system tested in this effort could also be adapted for use with other fuel cells (e.g., solid oxide, phosphoric acid) running on landfill gas.

P10 Engineering a 70-Percent Efficient, Indirect-Fired Fuel-Cell Bottomed Turbine Cycle

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ABSTRACT

We introduce the natural gas, indirect-fired fuel-cell bottomed turbine cycle (NG-IFFC) as a novel power plant system for the distributed power and on-site markets in the 20 to 200 megawatt (MW) size range. The NG-IFFC system is a new METC-patented system. This power-plant system links the ambient pressure, carbonate fuel cell in tandem with a gas turbine, air compressor, combustor, and ceramic heat exchanger. Performance calculations based on Advanced System for Process Engineering (ASPEN) simulations show material and energy balances with expected power output. Early results indicated efficiencies and heat rates for the NG-IFFC are comparable to conventionally bottomed, carbonate fuel-cell steam-bottomed cycles, but with smaller and less expensive components. More recent calculations extended the in-tandem concept to produce near-stoichiometric usage of the oxygen. This is made possible by reforming the anode stream to completion and using all hydrogen fuel in what will need to be a special combustor. The performance increases dramatically to greater than 70 percent.

BACKGROUND

Because of the abundance and relatively low cost of natural gas, gas turbine systems are gaining unprecedented acceptance in the power generation community. There are many advantages to using gas turbine systems. Nominally clean fuels are required to protect the turbine machinery, so the systems tend to also produce low levels of pollutants. When bottomed with a steam turbine, the energy from the exhaust stream inexpensively produces electric or cogeneration power. In addition, gas turbines require low maintenance and provide rapid start up.

There is, however, a limit to their use imposed by material properties — namely, the temperatures needed to achieve high efficiency exceed what materials can reasonably provide. While rapid strides have been made in the last few decades to achieve 50 to 52 percent combined-cycle efficiency, values above 60 percent appear to be close to impossible. Over the next 10 years, the Department of Energy (DOE) Morgantown Energy Technology Center (METC) will co-sponsor research on its Advanced Turbine System (ATS) Program to develop large, utility-scale units that are expected to achieve gas/steam cycle systems at 60-percent efficiency, and smaller scale units with gas/steam turbine systems that should reach 52-percent efficiency. These improvements will be possible if new high-temperature materials are indeed developed as planned. Such thermal systems can achieve higher efficiencies if turbine inlet temperatures are increased by approximately 150 to 200 °C. It will then become ever harder to meet the environmental standards, because nitrogen oxides (NO_X) production tends to increase non-linearly with increases in temperature. A new breakthrough technology is needed to achieve higher efficiencies and maintain low pollutant levels. This is the fuel cell.

The DOE has identified both fuel cells and advanced gas turbines as preferred sources of future electric power. DOE/METC is investigating the possibility of bringing both these two technologies together for the first time. Systems that do this are the subject of this paper.

FUEL CELL TECHNOLOGY

The fuel cell is a device that oxidizes fuel directly into electrical power without the enthalpy of combustion raising the temperature (Appleby and Foulkes 1989). Fuel passes through a semi-permeable membrane in the fuel cell where it electrochemically reacts with the oxidizer (air) compartment or cathode — as in proton-conducting fuel cells. The oxidizer passes through a semi-permeable membrane to the fuel compartment or anode in a molten-carbonate fuel cell (MCFC) or a solid-oxide fuel cell (SOFC).

In MCFCs, the reaction of hydrogen and carbonate ions releases electrons at the anode/electrolyte interface. Water, carbon dioxide, and heat are released by the anode reaction. The electrochemical reaction of oxygen, carbon dioxide, plus two electrons creates carbonate ions at the cathode/electrolyte interface. MCFC stack designs incorporate either internal or external manifolding. Internal and external reforming are being considered in several commercialization concepts. All MCFC concepts employ flat cell components in the cell package (i.e., anode, matrix to hold carbonate, cathode, current collector, and separator plate).

Fuel cells have many advantages that make them the ideal power system of the future, including environmental friendliness because the nitrogen oxide, sulfur oxide, carbon monoxide, and other pollutant exhaust products are insignificant. There is more to this statement than meets the eye. As attempts are made to improve the efficiency of future gas turbines, ever higher temperatures will be necessary to compete with fuel cell efficiency. Thus, these turbines will more likely produce unacceptable pollutant levels or will require expensive catalysis and unacceptable costs. This will not occur with fuel cells, which produce high efficiency through chemical rather than thermal conversion and are controlled by Gibbs' Free Energy rather than high temperature operation. Thus, ultimately, if we are ever to achieve 70-percent power efficiency, we must get away from totally thermal, prime-mover systems.

Fuel cells have other operational benefits. They operate efficiently at small size and even at partial loads. Because of this, fuel cells are ideal for distributed power generation. Fuel cell systems produce power with smaller footprints and hence lower land and power costs. These benefits in turn result in the potential to completely eliminate high voltage lines, reducing health concerns. In turn, a society can be created with fuel cells where the industrial complexes are cleaner and where even residential power services are available.

There is another special advantage to fuel cells, namely low water utilization. This makes fuel cells especially attractive power systems in water-scarce locations. In fact, when natural gas is consumed, it produces relatively pure water, and so could provide a source of water for arid environments.

The many types of fuel cells use different kinds of electrolyte as the principal component by which power is converted, and the types are named after the electrolytes. The MCFC was selected for this analytical evaluation because the temperature required for its operation is nominally about the same as that of the exhaust of a gas turbine. MCFCs operate at about 550 °C (1,050 °F). SOFCs can operate at as much as 1,000 °C (1,800 °F).

Recognizing MCFC operating temperature match with gas-turbine exhaust, we examined the possibility that the clean air exhaust from the turbine could drive the fuel cell. This would all but eliminate a need for yet another heat exchanger to couple the two units.

STATUS OF FUEL CELL COMMERCIALIZATION

Fuel cell technology has evolved from small, curious, laboratory cell tests to fuel cell stack testing and investigations devoted to demonstration testing of complete systems (Energy Research Corporation 1987; Williams 1995; and Williams and George 1990, 1991). Systems testing is the precursor to commercialization. Small units in the 100-kilowatt (kW) range are commercially available. Thus, the fuel cell is no longer a technology of the distant future. Stack life for the MCFC has been extended from 100 hours in the early 1980s to some 5,000 hours for small stacks. Testing is underway to warrant MCFC units for 25,000 hours, and within several years, vendors expect to warrant their units for 40,000-hour operation. The largest MCFC stack now being manufactured and tested is 2-MW size.

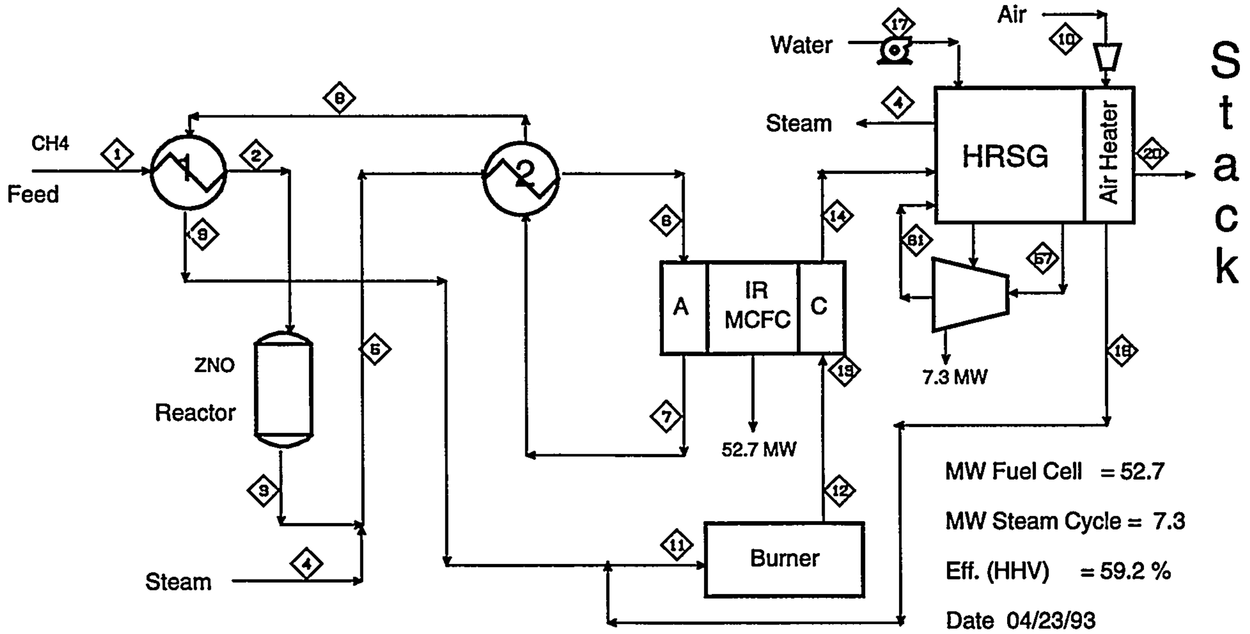
The SOFC uses solid oxides as electrolyte. Tubular and planar SOFC testing is still behind the MCFC in terms of stack size. However, tubular SOFCs have been tested for 40,000 hours. SOFC stacks have achieved better than 0.13 watts per square centimeter (cm^2) and 0.65 volts per cell. For monolithic SOFCs, a type of planar SOFC, better than 1 watt/cm^2 appears feasible. These units could provide eight times the power of present fuel cells in half the stack volume.

FUEL CELL POWER CONVERSION CONFIGURATIONS

While a fuel cell is extremely efficient, not all of the fuel's energy is electrochemically converted to electric power. Fuel conversion always yields moderate to high temperature product streams and other exhaust streams. These energy streams must be thermally converted to electric power.

About 50 percent of the fuel that comes into an MCFC is converted to electric power. The degree of conversion depends on the amount of carbon dioxide in the fuel cell. Carbon dioxide is increased by recycling some of the product stream from the anode, which contains water and carbon dioxide, to the cathode. Recycling increases the conversion from 50 to approximately 60 or 70 percent of the fuel. The excess fuel is then consumed in a secondary combustor. Three possible configurations that compare ways of using the exhaust stream energy are shown in Figures 1 to 3.

The arrangement shown in Figure 1 is one that has been studied in greatest depth: the exhaust energy is used in a steam cycle that acts as a bottomer part of a power system. The combustor raises the temperature using a heat recovery steam generator (HRSG). In Figure 1, a coal gasifier produces fuel for the fuel cell, but natural gas (NG) is equally viable. As an alternative, a gas turbine could be used to



STREAM	1	2	3	4	5	6	7	8	9	10	11	12	13	14	17
FLOW (LBMOLE/HR)	872.95	872.95	872.92	1761.9	2634.9	2634.9	7179.6	7179.4	7179.4	11847	19027	19027	19027	14491	1761.9
TEMPERATURE (F)	60.01	775.00	775.00	740.00	754.40	1100.00	1250.00	1121.03	1030.15	57.00	611.47	1099.84	1100.00	1250.00	60.00
PRESSURE (PSIA)	45.00	30.00	25.20	35.00	25.00	16.00	16.00	15.90	15.70	14.70	15.50	15.50	15.50	15.50	42.00
H (MM BTU/HR)	-28.4	-21.5	-21.5	-173.6	-195.0	-185.0	-824.5	-834.6	-841.6	-15.9	-841.4	-841.4	-841.4	-393.3	-218.5

STREAM	18	20	57	61											
FLOW (LBMOLE/HR)	11847	14491	2697.73	2697.73											
TEMPERATURE (F)	250.00	200.26	1050.00	107.83											
PRESSURE (PSIA)	15.70	14.70	1465.00	1510.00											
H (MM BTU/HR)	0.2	-514.4	-260.4	-331.9											

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Figure 1. Baseline MCFC Steam Turbine System

provide a high temperature, pressurized air stream which then passes into a pressurized fuel cell. This configuration is shown in Figure 2: the fuel cell is a topper for the low-pressure gas turbine. There is little difference in the cost performance trade-off between these two units. One still has the exhaust stream from the fuel cell to deal with. This is probably best utilized by heating the compressor discharge air. Finally, we have the indirect-fired configuration, shown in Figure 3, in which the air used by the fuel cell passes through the heat exchanger, and a combustor raises the temperature of the air to the gas turbine inlet temperature.

Figure 3 illustrates the basic features of a NG-IFCFC power plant. The compressed air for the gas turbine is heated by combustion products from an off-base combustor. While there are many advantages to this configuration, there are also some problems that need to be resolved (Micheli, Williams, and Parsons 1993).

The NG-IFFC configuration in Figure 3 shows only hot air passing through the turbine. This all but eliminates corrosion of the turbine blades, thereby extending turbine life. The hot air source then supplies the fuel cell. The problem of contamination focuses on the heat

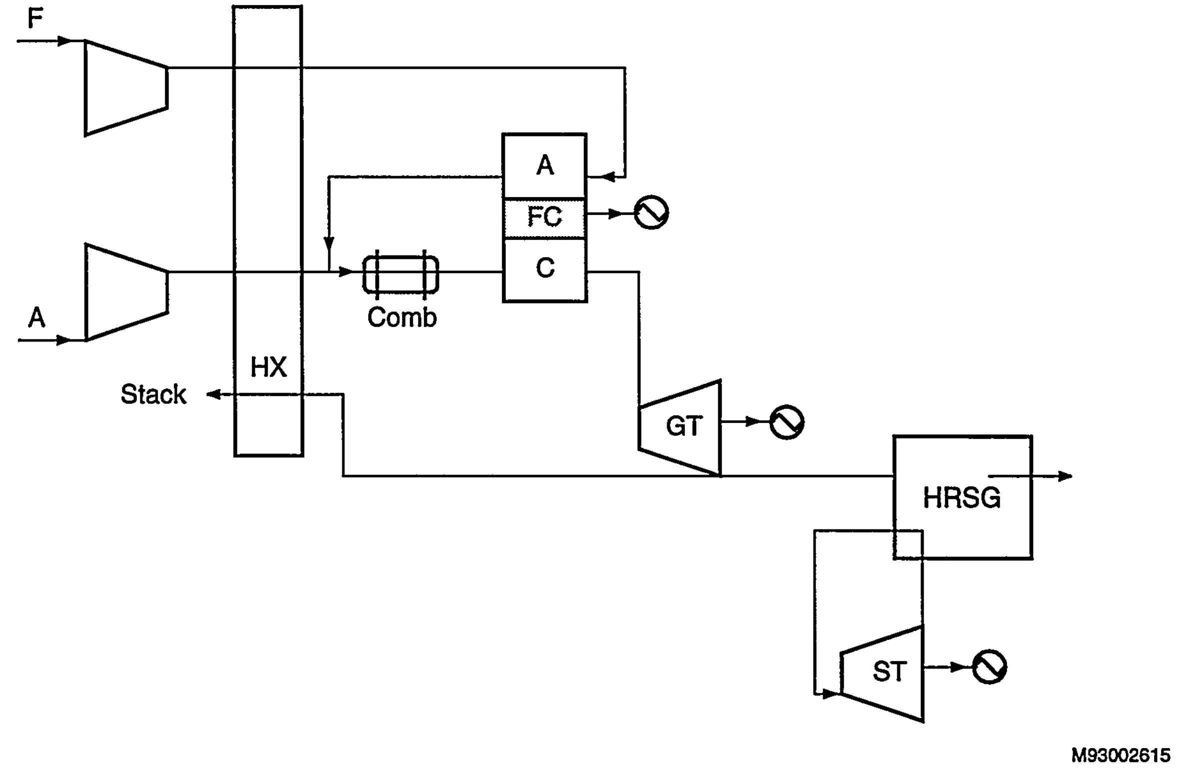


Figure 2. Pressurized Fuel Cell Topper Cycle

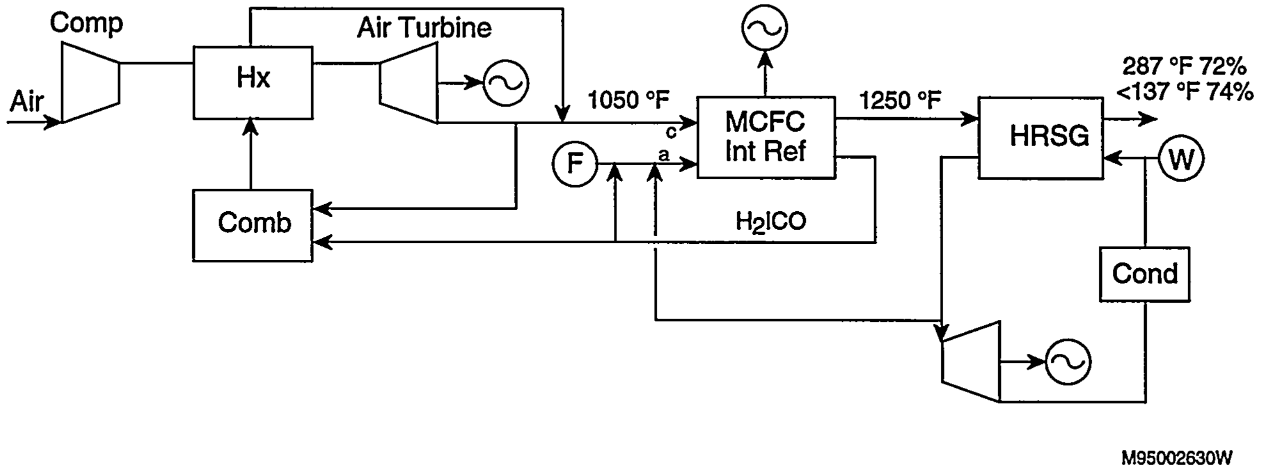


Figure 3. METC ASPEN, ASPENPlus, ERC ChemCAD

exchanger rather than the turbine, which turns out to be a less challenging environment in which to handle both the corrosion and the erosion that commonly damage a turbine.

CYCLE EFFICIENCY CALCULATIONS

Calculations were carried out by DOE using a modified ASPEN code to calculate the conversion of chemical and thermal energy to electric power. The baseline case of the fuel cell plus steam turbine provides a way of comparing the efficiency of using the residual energy. Although higher efficiency is expected to be available by the turn of the century, we conducted the study assuming current steam turbine technology. Hence, the calculations reflect a conservative viewpoint.

Performance calculations using ASPEN simulations present material and energy balances with expected power output. Figure 3 is the schematic used for the ASPEN simulation of the system. The results indicate that efficiencies and heat rates of the NG-IFFC are superior to the conventionally bottomed, carbonate fuel-cell, steam-bottomed cycles. The NG-IFFC also has smaller and less expensive components (Micheli, Williams, and Parsons 1993).

More recent calculations extended the in-tandem concept to produce near-stoichiometric usage of the oxygen. This is made possible by reforming the anode stream to completion and using the all-hydrogen fuel in what will need to be a special combustor. The performance increases dramatically to greater than 70 percent. Figure 4 graphs the capacity and demand of the tandem technology cycle by season, showing the effect of pre-chilling to match capacity and demand requirements. Figures 5 and 6 graph the idealized efficiency of IFFC cycles with the fuel cell operating at 50 and 70 percent of maximum.

Courtesy of Southern California Gas Co.

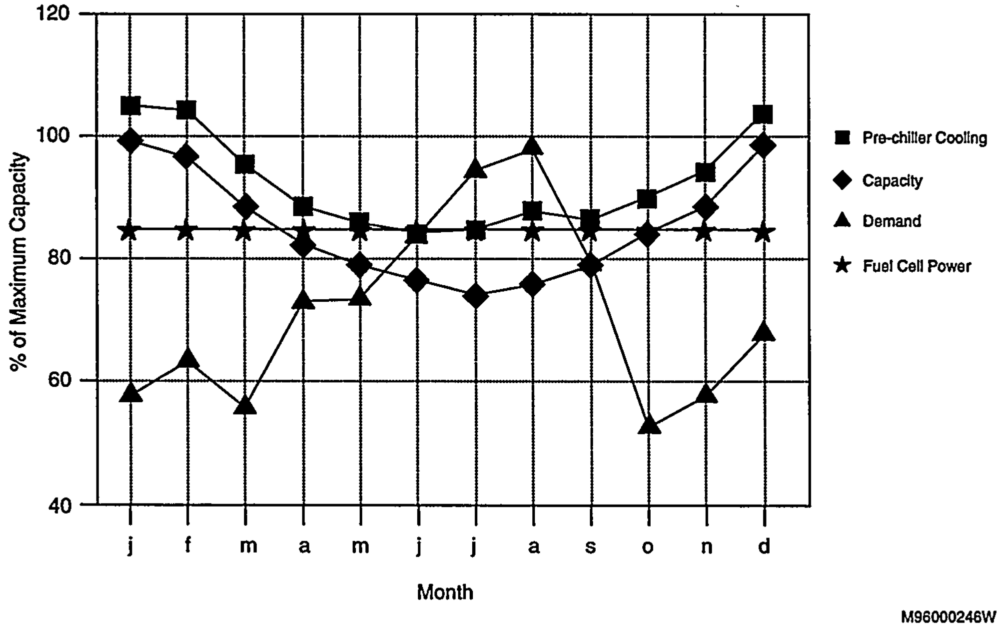
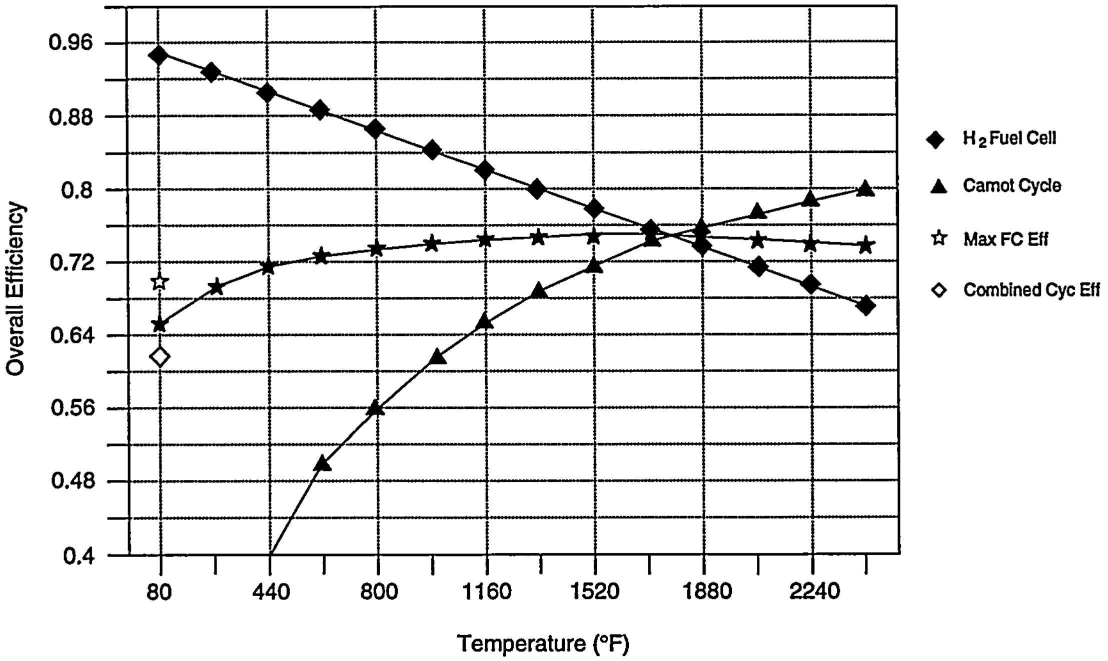


Figure 4. Tandem Technology Cycle, Effect of Pre-Chilling to Match Capacity and Demand Requirements



Fuel Cell @ 70% of maximum
Combined Cycle @ 62% of Carnot

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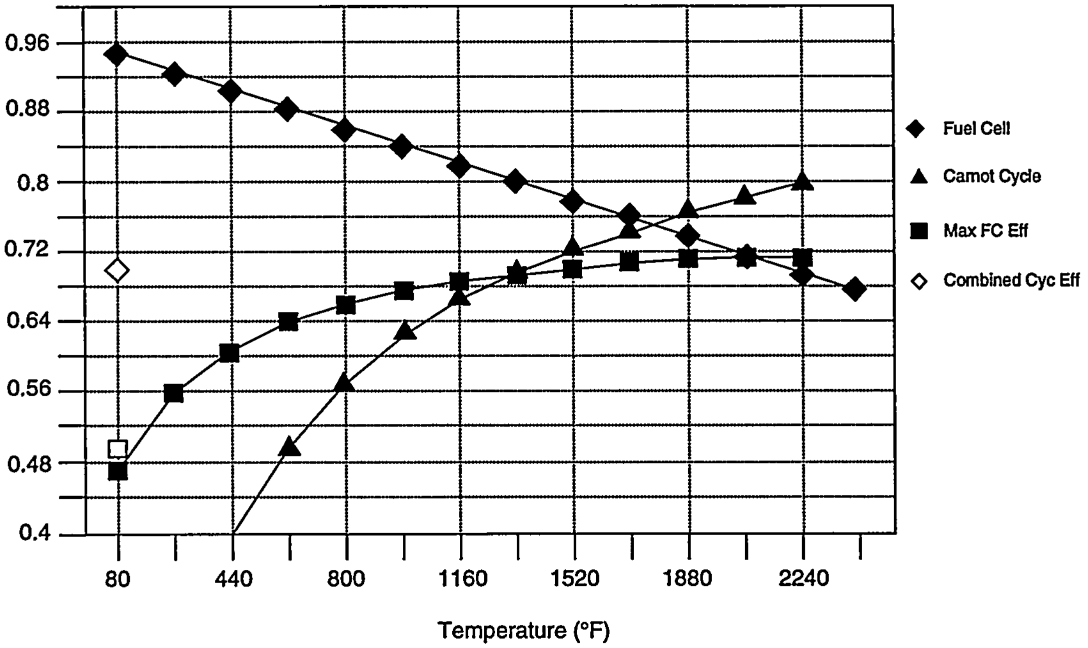
Figure 5. Idealized Efficiency of IF-FCB Cycles, Fuel Cell at 70 Percent of Maximum



Fuel Cell @ 50% of maximum
Combined Cycle @ 70% of Carnot

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Figure 6. Idealized Efficiency of IF-FCB Cycles, Fuel Cell at 50 Percent of Maximum

DISCUSSION

The NG-IFFC system has a 72 to 74 percent efficiency and could have significant use in new specialized (niche) markets, e.g., the distributed power and on-site markets in the 20 to 200 MW size range.

The NG-IFFC has significantly higher cycle efficiency than the gas-turbine combined cycle (GT/CC) alone. A 200-MW utility-size NG-IFFC system will average 72 to 74 percent efficiency compared to 60 percent for a GT/CC system; at the 20-MW industrial-size, the NG-IFFC system averages 68 percent and the GT/CC system averages 52 percent efficiency.

The heat engine and the fuel cell complement each other. They must be linked in tandem: the gas turbine bottomed by an MCFC is an excellent choice, and the gas turbine topper to an SOFC is also good.

The maximum temperature of the gas turbine in the NG-IFFC system reaches about 2,200 °F. Higher temperatures would be wasteful. The system has lower system capital costs with a fuel cell operating at greater than $1,400 per kW, and lower operating costs with a greater than 70 percent efficiency. Table 1 shows the advantages of the fuel cell/heat engine in a tandem approach and the applicable domestic and foreign niche markets. Cost estimates of the NG-IFFC are shown in Table 2.

Table 1. Indirect-Fired, Fuel-Cell Bottomed System

Advantages Fuel Cell and Heat Engine	Applicable Niche Markets (Domestic and Foreign)
System has lower capital costs and life costs than FC + ST.	25 percent higher profit margin.
Constant efficiency over a wide temperature range.	FC/CC overcomes mismatches between annual baseload demand and capacity. - 25 percent smaller power rating.
Low NO_X and SO_X (remove S from fuels).	Sales in non-attainment areas.
	No acid rain implies very low exhaust temperature < 137 °F; provide H_2O for arid climate by condensing water.
Quiet operation.	Siting and permitting simplified; IPP markets and APPA with no overhead power line.
Air turbine operations.	Better RAM, higher profits.

Table 2. Indirect-Fired, Fuel-Cell Bottomed System Cost Estimates

Addition of Combined-Cycle Power Units (GT + ST) Reduces Costs by:	Capital Savings (%)
- Decreasing balance of plant size and cost at higher efficiencies.	5%
- Lowering at-risk cost while FC is still a maturing technology.	10%
- Lowering financing charges since plant is faster on line.	5%
- Lowering capital cost comparing $1000/kW for GT to fuel cell at $2500/kW life replacement costs.	10%
Profit margins significantly higher in niche markets.	**25%**

SUMMARY

We introduce the NG-IFFC as a novel power plant system for the distributed power and on-site markets. The system has a 72 to 74 percent efficiency and could conceivably attain 80-percent cycle efficiency. The system has significant potential use in new specialized (niche) markets, e.g., the distributed power and on-site markets in the 20 to 200 MW size range. The NG-IFFC has significantly higher cycle efficiency than the gas-turbine combined cycle (GT/CC) alone. A 200-MW utility-size NG-IFFC system will average 72 to 74 percent efficiency compared to 60 percent for a GT/CC system; at 20-MW industrial-size, the NG-IFFC system averages 68 percent and the GT/CC system averages 52 percent efficiency.

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P11

Internet Public Information for Fuel Cells

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ABSTRACT

The rapid development and integration of the Internet into the mainstream of professional life provide the fuel cell industry with the opportunity to share new ideas with unprecedented capabilities. The U.S. Department of Energy's Morgantown Energy Technology Center (METC) has undertaken the task to provide a service where current fuel cell descriptions and information are available to customers, manufactures, academia, and the general public.

METC has developed a Fuel Cell Forum where members can exchange ideas and information pertaining to fuel cell technologies using the Internet. Forum membership is encouraged from utilities, industry, universities, and Government. Because of the public nature of the Internet, business sensitive, confidential, or proprietary information should not be placed on this system. The views and opinions of authors expressed in the forum do not necessarily state or reflect those of the U.S. Government or METC.

METC has endeavored to develop a World Wide Web (WWW) location committed to the description and development of the fuel cell. Netscape or compatible software provides access to the METC Homepage. The user then selects Advanced Power Systems, then Fuel Cells. Fuel cell overview and description is followed by a presentation of the fuel cell system characteristics and advantages. Descriptions of major fuel cell projects are provided in the FACTS section. Finally, as a service to METC customers, the homepage provides a calendar and points of contact. Updates to the WWW location are occasionally made revealing current technical advances in fuel cells.

In the continuing effort to further improve public knowledge and perception of fuel cell power generation, METC has created two new modes of communication using the Internet. The FUELCELLS Forum can be accessed using the Internet at the e-mail address:

in:listproc@lists.metc.doe.gov

Under subject Subscribe FUELCELLS <your name>. The WWW METC Fuel Cells Homepage address is accessed using Netscape at

http://www.metc.doe.gov

P12

Fuel Cell Cogeneration

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ABSTRACT

The U.S. Department of Energy's Morgantown Energy Technology Center (METC) sponsors the research and development of engineered systems which utilize domestic fuel supplies while achieving high standards of efficiency, economy, and environmental performance. Fuel cell systems are among the promising electric power generation systems that METC is currently developing. Buildings account for 36 percent of U.S. primary energy consumption. Cogeneration systems for commercial buildings represent an early market opportunity for fuel cells. Seventeen percent of all commercial buildings are office buildings, and large office buildings are projected to be one of the biggest, fastest-growing sectors in the commercial building cogeneration market. The main objective of this study is to explore the early market opportunity for fuel cells in large office buildings and determine the conditions in which they can compete with alternative systems. Some preliminary results and conclusions are presented, although the study is still in progress.

The CNG Tower, a 32-story office building in downtown Pittsburgh, Pennsylvania, served as a specific example for this study. The building presently purchases its electricity, uses natural gas-fired boilers for space heating, and electric chillers for air conditioning. Data from this automated heating, ventilating, and air conditioning (HVAC) system were used to characterize the building's demands for electricity, space heating, and air conditioning on an hourly and/or daily basis throughout an entire year. A cogeneration system, featuring phosphoric acid fuel cells and a lithium bromide absorption chiller, was designed to meet the building's needs. Hot water, produced from fuel cell waste heat, both drives the absorption chiller for air conditioning and provides for space heating. When the fuel cell waste heat is insufficient, a natural gas-fired boiler supplements the hot water supply. The system was studied at two different fuel cell power generation capacities: the building's peak electric demand and an intermediate electric load. At both capacities, the system's mode of operation was to follow the electric load to the extent possible. Future work may consider additional modes of operation, including those which involve selling excess power or storing thermal and electrical energy.

Two economic scenarios were considered for the fuel cell cogeneration system: (1) retrofitting an office building's existing HVAC system and (2) installation into a new office building. In each scenario, the economic performance of the fuel cell cogeneration system was determined as a function of capital cost, fuel cell stack life, and savings in annual energy costs. Savings in annual energy costs were calculated as a function of electricity and natural gas rates.

Fuel cell cogeneration systems can reduce the annual energy consumption of large office buildings which presently purchase their electric power by over 25 percent. Depending on local electricity and gas rates and capital and maintenance costs, fuel cell cogeneration systems may also be economically feasible. In addition, other factors may influence their success in the commercial building energy supply market: environmental regulations, local power transmission costs, the cost of back-up power, and the local utility's need for more power. Furthermore, fuel cells must compete in this market with other cogeneration systems, such as natural gas-fired engines, gas turbines, and low-pressure boilers with back-pressure steam turbines.

Fuel cells enjoy many technical advantages over these competing systems. Air quality permits are more easily obtained due to fuel cells' unsurpassed environmental performance (low NO_x and SO_2). Since the fuel cells themselves have no moving parts, noise and vibrations are low. Fuel cell systems have demonstrated the ability to produce premium quality power at high availability - an attractive combination for computer and other sensitive applications. The load-following capability of fuel cells is excellent and they can be turned down significantly with no reduction in efficiency. Unlike gas turbines, the generation capacity of fuel cells is not derated in warmer seasons. Fuel cells are modular, allowing small, incremental capacity additions as demand grows. Further work is needed to determine the conditions under which these advantages make fuel cells the system of choice for cogeneration in large office buildings, as well as other commercial buildings.

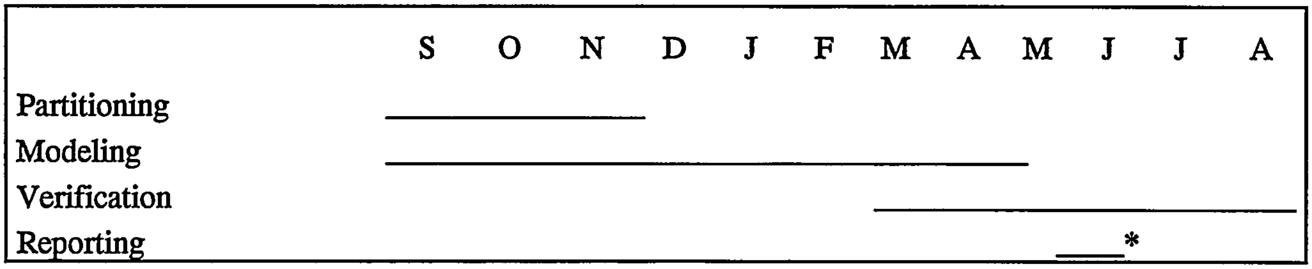
P13 Trace Metal Transformations in Gasification

CONTRACT INFORMATION

Contract Number	DE-AC21-92MC28016
Contractor	Energy & Environmental Research Center University of North Dakota PO Box 9018 Grand Forks, ND 58202-9018 (701) 777-5000
Other Funding Sources	None
Contractor Project Manager	Steven A. Benson
Principal Investigators	Thomas A. Erickson Christopher J. Zygarlicke Cathy A. O'Keefe Karen A. Katrinak Sean E. Allan David J. Hassett William B. Hauserman
METC Project Manager	Dr. Norman T. Holcombe
Period of Performance	August 1, 1992, to December 30, 1995

Schedule and Milestones

FY96 Program Schedule



* Project Review Meetings

OBJECTIVES

The Energy & Environmental Research Center (EERC) is carrying out an investigation that will provide methods to predict the fate of selected trace elements in integrated gasification combined cycle (IGCC) and integrated gasification fuel cell (IGFC) systems to aid in the development of methods to control the emission of trace elements determined to be air toxics. The goal of this project is to identify the effects of critical chemical and physical transformations associated with trace element behavior in IGCC and IGFC systems. The trace elements included in this project are arsenic, chromium, cadmium, mercury, nickel, selenium, and lead. The research seeks to identify and fill, experimentally and/or theoretically, data gaps that currently exist on the fate and composition of trace elements. The specific objectives are to 1) review the existing literature to identify the type and quantity of trace elements from coal gasification systems, 2) perform laboratory-scale experimentation and computer modeling to enable prediction of trace element emissions, and 3) identify methods to control trace element emissions.

BACKGROUND INFORMATION

Trace element emissions pose a potential problem to two emerging coal gasification electric power-generating systems: IGCC and IGFC. The potential problems associated with trace elements are the release of substances that are considered air toxics and the degradation of fuel cell efficiency due to contamination with minor elements. In order to develop effective technologies to control trace element emissions within anticipated regulatory requirements and to ensure the efficient operation of fuel cells, the type and quantity of trace elements emitted from coal gasification-based systems must be determined as a function of system, system conditions, and coal composition. Table 1 shows the potential ranking of trace metals according to their potential impact for IGFC and IGCC (Energy Research Corporation, 1994).

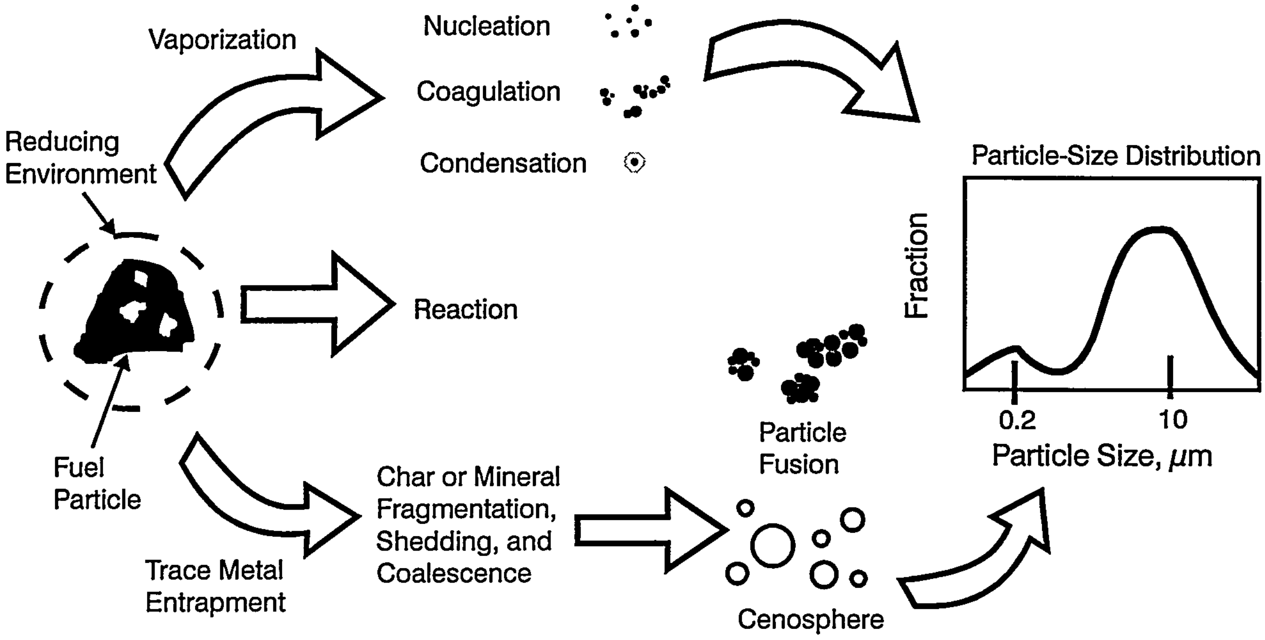
The fate of trace elements in coals used in IGCC and IGFC systems is closely tied to how the trace elements are associated in the coal and the gasification conditions. Trace elements in coals are associated in several forms, including organic associations, such as salts of carboxylic acid groups and organic coordination complexes, and mineral associations, such as sulfides, sulfates, silicates, phosphates, and carbonates. During gasification, these inorganic elements are partitioned into gases, liquids, and solids. The transformation of these trace elements into the various states and phases depends upon the fundamental characteristics of the elements and their association with minerals and coal particles. Minerals that are not directly associated with coal particles experience a different process environment than minerals that are intimately associated with coal particles during gasification.

In order to predict the form of the inorganic species during gasification, it is essential that detailed information on the distribution of major mineral phases and organically associated inorganic elements be determined. This information is critical since the transformations and interactions during utilization impact the partitioning of trace elements. The primary transformations that occur to major and minor trace species during coal conversion are illustrated in Figure 1. Modeling the transformations using thermochemical equilibrium calculations combined with various chemical

Table 1. Possible Ranking of Toxic Trace Elements According to Their Potential Impact on IGFC and IGCC Systems (Energy Research Corporation)

IGFC Trace Species				IGCC Trace Species			
Element	Abundance as Vapors	@ or above	Rank	Element	Abundance as Vapors	@ or above	Rank
As	[Ash_3 – 3ppm]	RT* gaseous	1	As	[AsH_3 – 3 ppm]	RT gaseous	1
Se	[H_2Se – 1 ppm]	RT gaseous	2	Hg	[Hg(v) – 10 ppb]	RT gaseous	2
Pb	[PbSe – 1 ppm]	>600°C	3	Cr	[$CrC\ell_3$ – 4 ppm]	>500°C	3
Mn	[$MnC\ell_2$ – 17 ppm]	>625°C	4	Pb	PbS – 2.6 ppm]	>625°C	4
Be	[$BeC\ell_2$ – 2.5 ppm]	RT gaseous	5	Se	[H_2Se – 1 ppm]	RT gaseous	5
Cd	[Cd(v) – 0.2 ppm]	150°C	6	Cd	[Cd(v) – 0.2 ppm]	>150°C	6
Sb	[$SbC\ell_3$ – 0.1 ppm]	RT gaseous	7	Mn	[$MnC\ell_2$ – 17 ppm]	>625°C	7
Hg	[Hg(v) – 10 ppb]	RT gaseous	8	Ni	[$NiC\ell_2$ – 3 ppm]	>500°C	8
Cr	[$CrC\ell_3$ – 4 ppm]	>550°C	9	Be	[$BeC\ell_2$ – 2.5 ppm]	RT gaseous	9
Ni	[$NiC\ell_2$ – 3 ppm]	>500°C	10	Sb	[$SbC\ell_3$ – 0.1 ppm]	RT gaseous	10

* Run Temperature



EERC CZ10448.CDR

Figure 1. Trace Metal Transformations and Partitioning

and physical constraints to reach equilibrium can be effectively used to estimate the distribution of gas, liquid, and solid components as a function of gasification conditions.

PROJECT DESCRIPTION

The approach of this project is to identify and model important physical and chemical transformation mechanisms of the trace elements during gasification as a function of coal compositions (trace element abundance and association) and gasification conditions. Identification of the reactions and transformations, coupled with accurate modeling of trace element behavior, is providing essential information for the identification of the form of the inorganic species. Gasification conditions—such as reducing and oxidizing atmospheres, gas-phase composition, pressure, and temperature—influence the partitioning of the air toxics between various gases, liquids, and solid inorganic components as a function of location in the gasifier. This information will be used to identify the most effective control technology by predicting the form of the trace element as a function of operational conditions. The model will be used to predict the initial partitioning of the metals in the gasifier and their form downstream of the gasifier in the gas cleanup systems.

This project has four work categories: partitioning experiments and analysis, modeling, identification of control technologies, and model testing and verification. The partitioning experiments and analysis have concentrated on the operation of closely controlled experimental equipment to produce the appropriate samples and the analysis/characterization of those samples by state-of-the-art analytical techniques.

A pressurized drop-tube furnace (PDTF) was utilized for the experiments. Testing has been conducted for three coals under the following conditions: 1000°–1500°C, 50–200 psi, and 0.5–2.0 O/C ratio. The samples produced in the PDTF have been analyzed using inductively coupled plasma spectroscopy, atomic absorption, x-ray fluorescence (XRF), scanning electron microscopy (SEM), and various other techniques.

The modeling work has concentrated on the modification of two existing codes at the EERC: ash transformations (ATRAN) and thermochemical equilibrium analysis of coals and ashes (TEACH). Both codes are being modified to include specific algorithms associated with trace element behavior. The resultant model is a consolidation of the aforementioned codes into a single program capable of predicting the size, composition, and phase of the inorganic species at a given temperature and pressure. An operational shell is being developed to incorporate the two codes.

The identification of control technologies will be incorporated into the project after most of the testing and modeling are complete, at which time multiple scenarios can be addressed using the data and models. The model testing and verification will consist of testing the model and assumptions on the transport reactor scaleup facility located at the EERC.

RESULTS

The levels of the metals found in the coals are summarized in Table 2. The associations of the metals with minerals and organic portions of the coal were described in detail at

Table 2. Abundance of Trace Metals in Coals Tested ($\mu g/g$, dry coal basis)

	Illinois No. 6	Sufco Utah	Pittsburgh No. 8
Arsenic	1.5	0.66	7.80
Cadmium	0.33	0.048	0.10
Chromium	26.0	26.9	18.8
Lead	2.84	0.86	3.87
Mercury	0.067	0.033	0.082
Nickel	15.1	8.35	14.9
Selenium	3.03	1.05	1.41

Table 3. Trace Metal Associations

Element	Associations
As	Pyrite, other sulfides, arsenates
Cd	Sulfides, clays
Cr	Clays, organic compounds
Hg	Pyrite, other sulfides
Ni	Pyrite, clays, organic compounds
Pb	Pyrite, other sulfides, clays
Se	Pyrite, other sulfides, organic compounds, sulfate

the last conference (Benson and others, 1994). Table 3 summarizes the generalized associations found for the metals.

Partitioning of Trace Metals

The partitioning of trace metals was ascertained by determining the fraction in which the different trace metal species were collected. Metals that are not vaporized during gasification remain in the residual ash and are collected in the size fraction greater than 1 μm. Metals that are vaporized but condense in the system are collected in the submicron fraction. Metals such as Se and Hg, which may not condense in the system but remain in a vapor state, will escape the multicyclone system and will be collected in the impinger train. Several runs were made in the PDTF varying pressure, gas temperature, and O/C ratio.

Comparison of the average percentages of each element in the collection devices shows that a substantial portion, 40% or more, of the total amount of the trace elements is recovered in the coarse ash of the first cyclone (Figures 2 and 3). The trace elements have remained with the larger ash particles, have recondensed, or have been captured on the ash. Cr, Ni, (Figure 3), and, surprisingly, Hg (Figure 2) partition with the coarse ash, with an average of more than 65% remaining in the >3-μm fraction of collected sample, and the balance

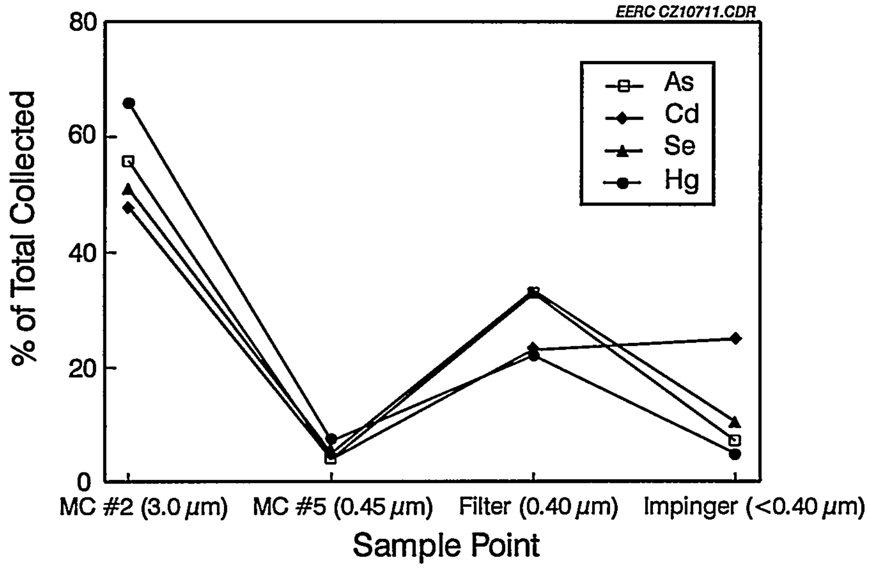


Figure 2. Average Percentages of As, Cd, Se, and Hg Collected in Size Fractions of the Sampling Train

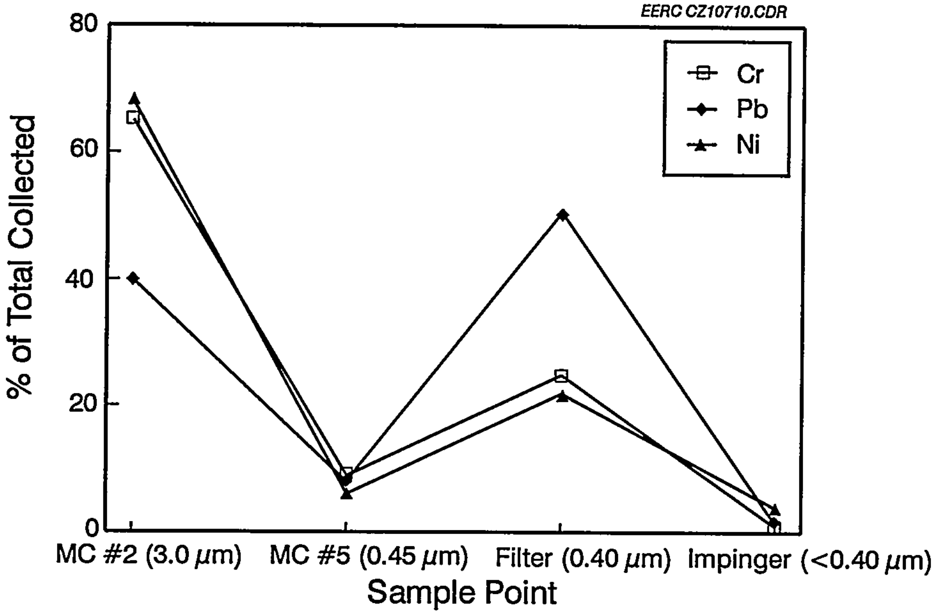


Figure 3. Average Percentages of Ni, Cr, and Pb Collected in Size Fractions of the Sampling Train

appearing in the 1–3-μm fraction or the <0.4-μm filter fraction. Very little Cr and Ni pass through the filter into the impingers (Figure 3), which is probably an indication that they are nonvolatile species, such as refractory oxides. The behavior of Hg is more complex. Although much of the Hg is collected in the cyclones and filter, under certain test conditions up to 30% of the Hg is in a volatile form passing through the filter into the impingers. Pb partitions primarily in the 1–3-μm ash fraction and the <0.4-μm filter, probably as a very fine oxide fume (Figure 3). Generally, little Pb passes through the filter into the impingers. Along with Hg, the trace elements As, Cd, and Se showed appreciable quantities in the impingers. Cd in particular appears to be exceptionally volatile or in the form of a very fine fume, with approximately 25% of that recovered present in the impingers (Figure 2).

In general, it can be summarized that Hg, As, Cd, and Se appear to be the most volatile, simply because a greater portion of the mass of these elements is in what would be considered condensed volatile size fractions. Here we are assuming that the trace metals in these fractions, since they are associated with particulate that is essentially less than 0.45 μm, were in a vapor phase until capture. In contrast, Ni and Cr, along with Pb to a certain degree, concentrate primarily with the larger-sized particulate and are classified more as nonvolatile or vaporized species, with Pb showing consistently more mass in the 1–3-μm range than Ni and Cr. Caution must be taken in making these types of assumptions, because some of these trace metal species may be condensing onto the particulate already trapped in the multicyclones or on the filter. For example, elemental Hg and Hg chloride are known to be captured on activated carbon, coal char, and fly ash. Of special concern in these results is the amount of unburned carbon present in the ash samples. Because the PDTF tests were all run under reducing conditions, unburned carbon was found to varying degrees in nearly every size fraction of product ash collected. It is possible that some of the trace metals have not been released from their original coal host minerals or particles. Several ash grains from the largest cyclone were analyzed using WDS and yielded only Ni and Hg in iron–sulfur-rich particles and Cr in aluminosilicate particles. When this information is compared with Table 2, it could be inferred that trace metal-bearing pyrite and clay mineral aluminosilicates may not have yielded the trace metals contained within them to a vapor or fine aerosol form before they were quenched in the multicyclone. Ideally, the PDTF test runs should have 100% carbon conversion, which would ensure a minimum of carbon carryover and would maximize the release of trace metals into true postflame forms.

The effects of pressure were not clear from the data collected; however, some trends were noted for O/C and temperature effects. Figures 4 and 5 illustrate the effect of O/C ratio on the abundance of As, Cd, Se, and Hg in the assumed vapor phase at 1500° and 1000°C initial gasification temperatures, respectively. The results indicate a general decrease in As, Cd, and Hg with increasing O/C ratio and a slight increase in Se with increasing O/C ratio. A possible explanation for these results is that more oxidized forms of As, Cd, and Hg are less volatile, whereas the oxidized form or species of Se is more volatile. The lower volatility of reduced Se relative to the oxide form has been discussed by Dismukes (1994).

The effect of initial gasification temperature on the abundance of trace metals in the gas phase is depicted in Figure 6. At a constant pressure of 50 psi and an O/C ratio of 2.0, increasing the gas temperature causes the abundance of As, Cd, Se, Hg, and Pb to increase. The effects of carbon burnout, which also tends toward greater values with increasing temperature, may be overshadowing the temperature effect somewhat. Figure 7 illustrates a temperature effect by showing that the concentrations in the vapor phase of two of the more volatile elements, Hg and Se, increase with increasing temperature at constant carbon burnout.

Predicting Trace Metals Behavior

Model Design. A computer model, TraceTran, to predict the evolution of major, minor, and trace elements during coal combustion and gasification has been created (Erickson and Benson, 1993), based on the algorithm shown in Figure 8.

The first task of the model is to determine the associations of the major, minor, and trace elements in the coal prior to utilization. The association of the elements prior to utilization will affect their phase, size, and composition distribution in the residual ash and gas streams. The three primary inorganic associations are water, organic, and mineral. Water-associated constituents are generally in the form of sulfates or chlorides present in the moisture of a coal particle. The organically associated constituents are generally found as the salts of carboxylic acid groups attached to the carbon matrix and as oxygen replacement (such as organic sulfur). The mineral associations are elements that comprise discrete minerals in the coal (such as clays, carbonates, and sulfides).

Because of the complexity of the interaction of inorganics during coal gasification, the mineral associations are further divided into mineral type, trace, minor, and major element content, size, and juxtaposition. Juxtaposition refers to the association of the minerals with the coal matrix and with other minerals. A locked mineral is intimately associated in the coal particle, while a liberated mineral is external to the carbon matrix. The detailed mineral classification is very important because different minerals behave differently. For instance, carbonates will commonly release CO_2, resulting in a greater potential for mineral fragmentation, depending on the system conditions. Clays that contain high levels of moisture may fragment initially because of the release of H_2O from their porous structures. Silicates are much less prone to fragmentation because they lack any of the previously discussed components.

Many of the mineral particles encountered in coal utilization contain trace and minor components. To predict the transformations of the trace and minor elements effectively, their distribution among the minerals is required. Whether a mineral is locked within the coal matrix or external to the coal can also have a large impact on its transformations. Locked minerals will be much more likely to coalesce with other minerals and organically associated constituents than are the liberated minerals. The liberated particles will also experience a slightly different gas environment during coal gasification, since they are not in intimate contact with the highly reducing, exothermic reaction of the carbon matrix.

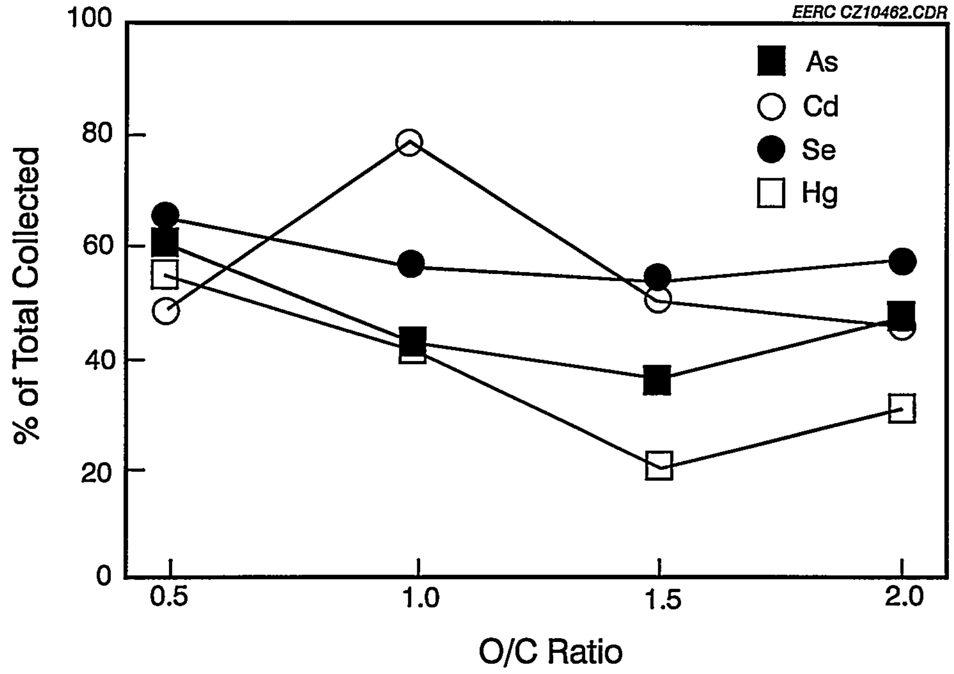


Figure 4. Trace Metal Distribution vs. O/C Ratio for Illinois No. 6

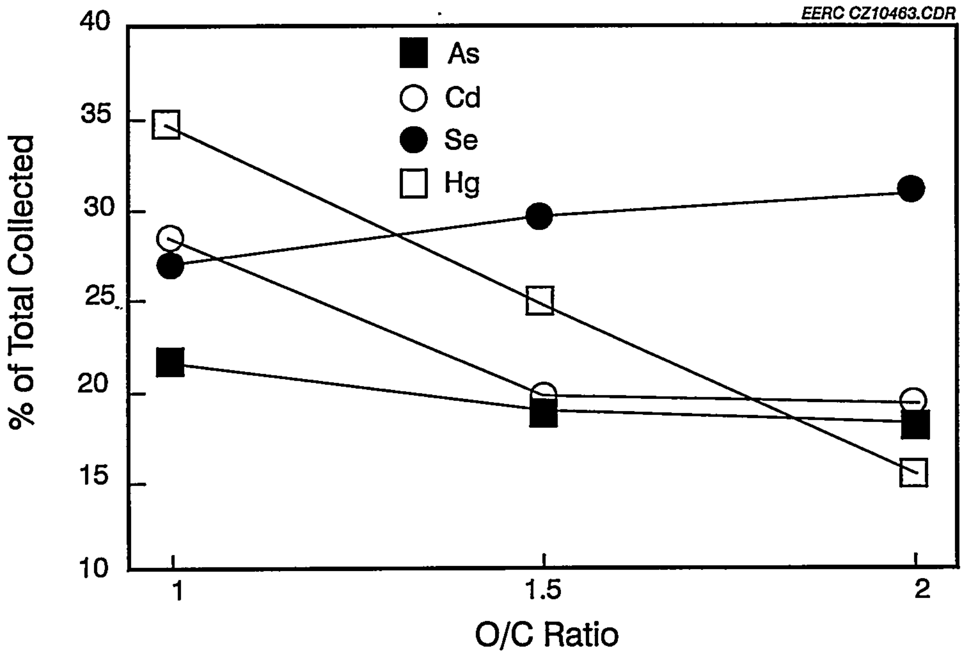


Figure 5. Trace Metal Distribution vs. O/C Ratio at 1500°C

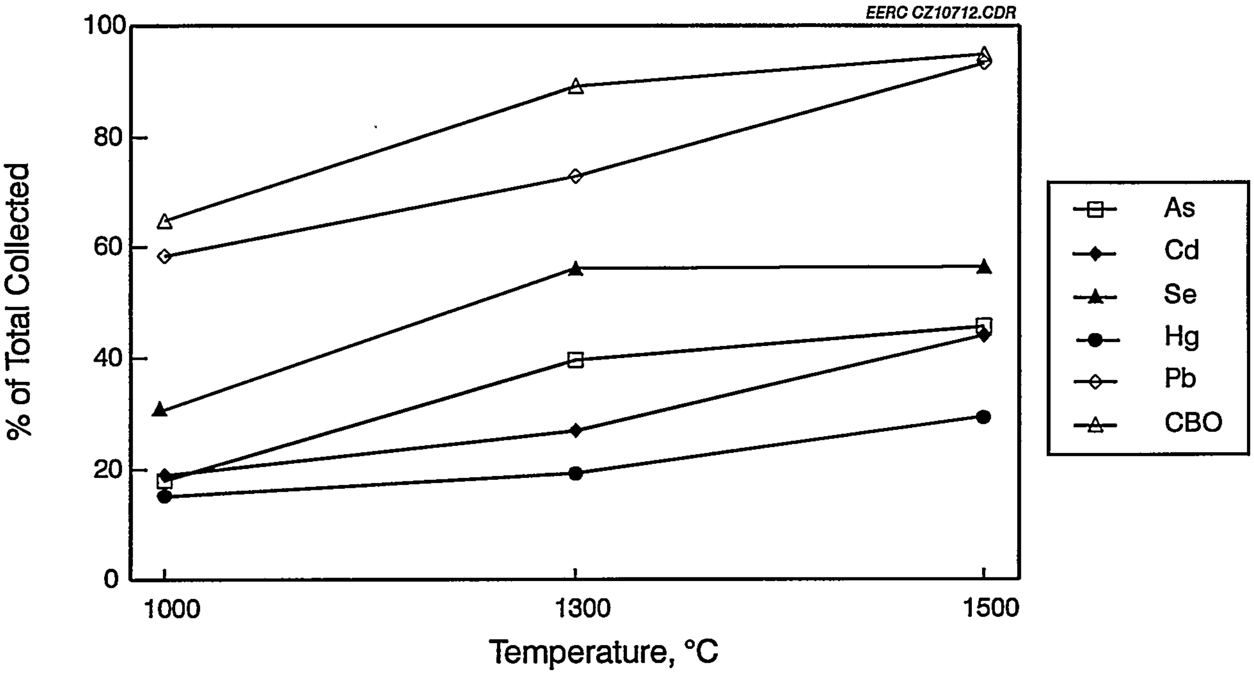


Figure 6. Trace Metal Distribution vs. O/C Ratio at 1000°C

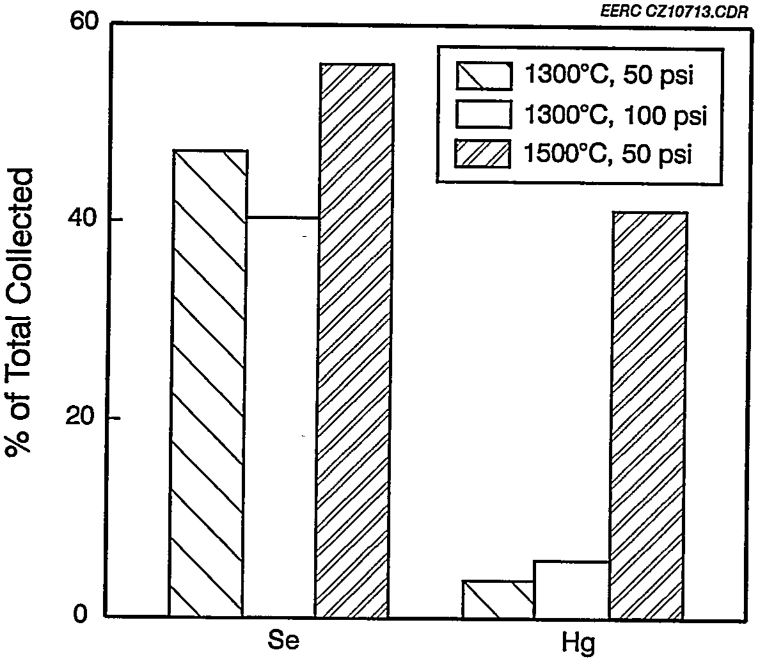


Figure 7. Concentrations in the Vapor Phase of Hg and Se with Increasing Temperature at Constant Pressure and Carbon Burnout

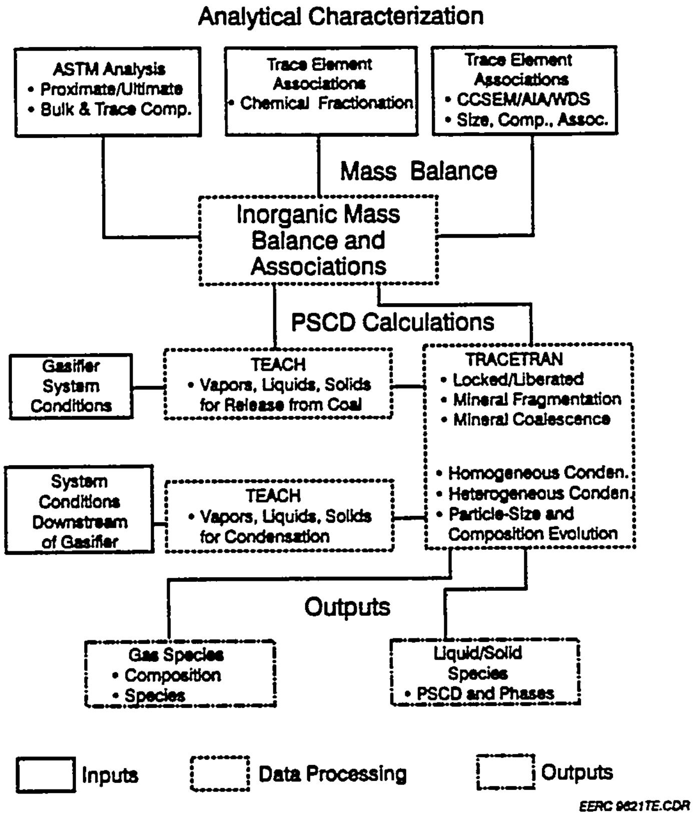


Figure 8. Trace Element Emissions Model Algorithm

Once a mass balance is performed around the coal input data, it is necessary to determine which of the inorganic components will be vaporized during the initial conversion process. These calculations are performed with the use of a thermochemical equilibrium program created at the EERC. This code has been upgraded to include some of the appropriate trace element phases, and the ability to include more phases is being considered through the use of additional thermochemical equilibrium programs. With the exclusion of the vaporized species, the remaining constituents are processed through algorithms for mineral fragmentation, coalescence of both minerals and organically associated species, and the shedding of resulting particles. Examples of data obtained from the thermochemical equilibrium portion of the code are illustrated in Figures 9, 10, and 11.

The fragmentation, coalescence, and shedding algorithms have been developed with the aid of data from full-scale systems and data generated in pilot- and laboratory-scale facilities. These algorithms are designed using data on various frequency distributions for each of the minerals and physical processes. The organically associated species that do not readily volatilize will also undergo coalescence with mineral particles as a function of the surface area of the minerals during coalescence. A portion of the organically associated species also homogeneously coalesces. The liberated minerals undergo fragmentation, but do not appreciably coalesce

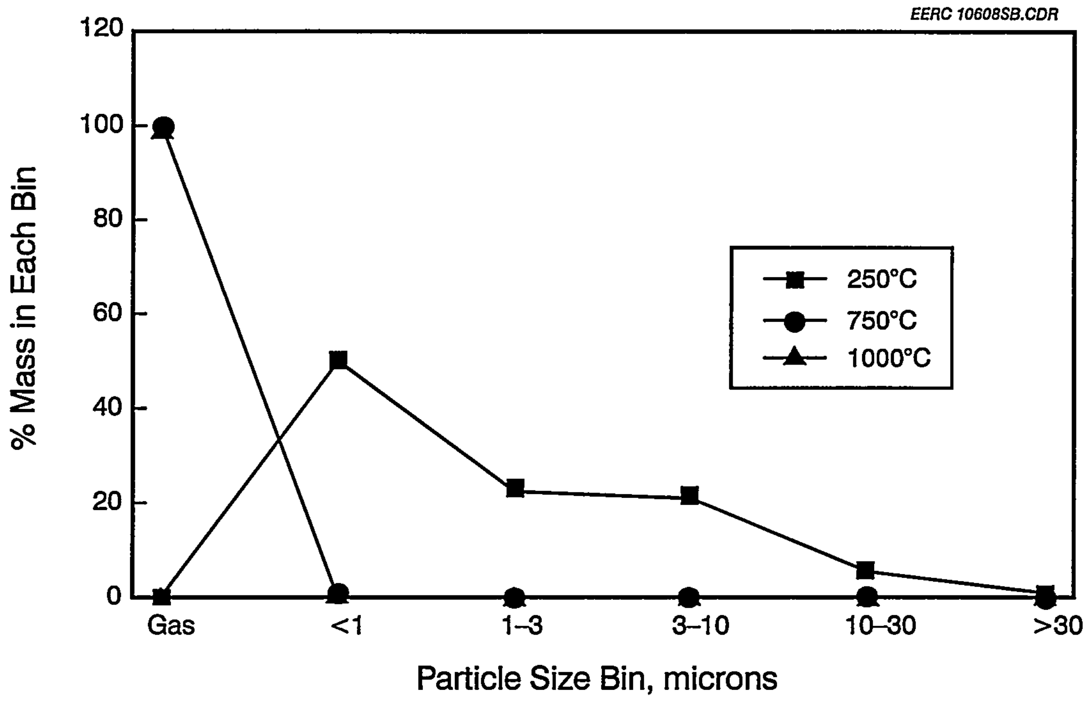


Figure 9. Illinois No. 6: TraceTran Predictions – Arsenic Particle-Size Distribution

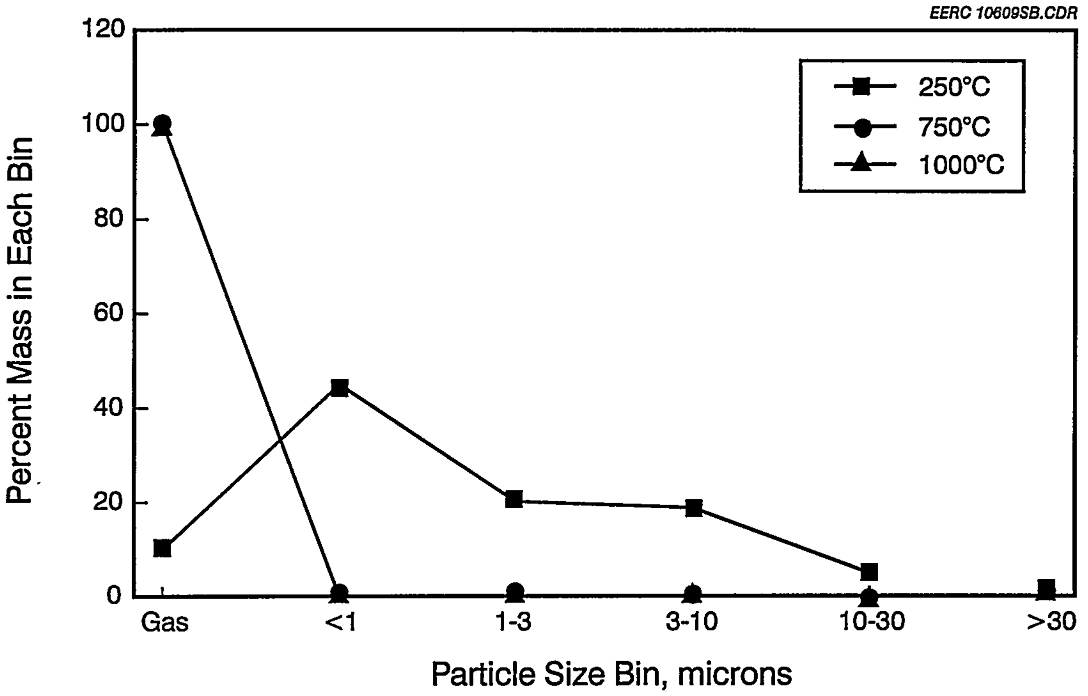


Figure 10. Illinois No. 6: TraceTran Predictions – Cadmium Particle-Size Distribution

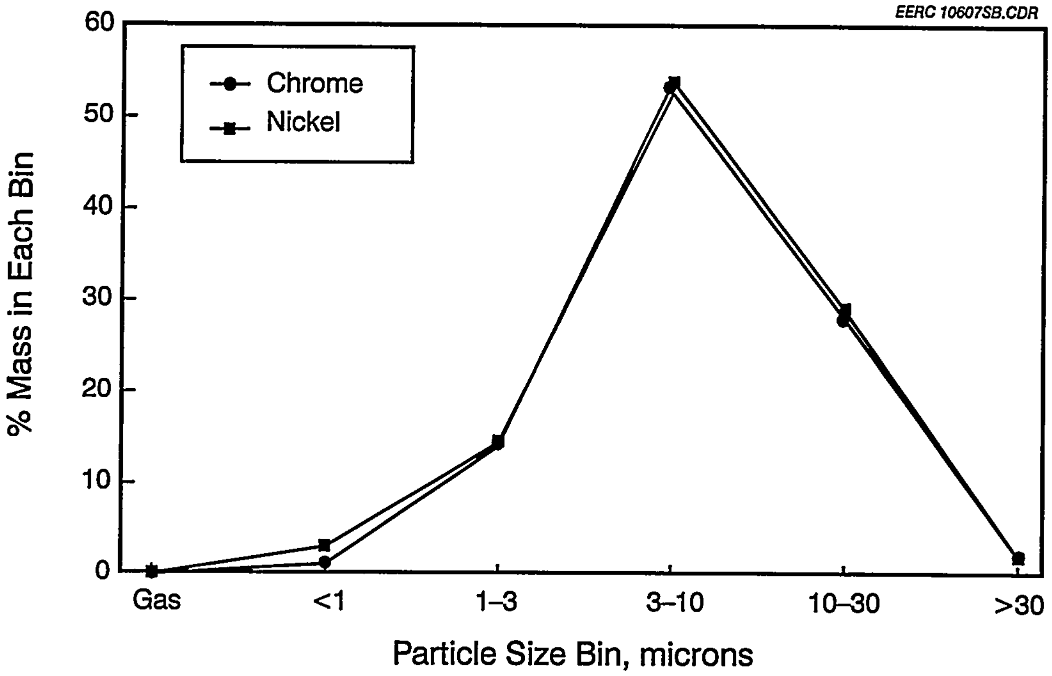


Figure 11. Illinois No. 6 TraceTran Predictions – Particle-Size Distribution, 750°C

with other minerals because of their lack of intimate contact with the coal.

The state of the volatile species at the desired downstream conditions will then be determined, again using the TEACH code. The quantity of species that will condense prior to the given conditions is calculated from the TEACH data. The species will then be both homogeneously and heterogeneously condensed. Heterogeneous condensation is based on the surface area of the existing particles. The resultant particulate and vapor-phase species will be compiled and manipulated into various composition and size distributions at the user's discretion. These distributions can be used to determine effective control technologies for a specific coal or to locate a coal compatible to a specific control technology.

Although the emphasis of this model is to aid in the control of trace element emissions, only minor attention has been given at this time to the engineering mechanisms of the control technologies within a system. Once this model has been fully tested and verified, the next logical step will be to incorporate engineering models that mimic the control technologies. Figure 12 shows an example of the overall model applied to a pressurized fluid-bed combustor for selected elements and the measured element distributions from full-scale sampling. A series of "crude" engineering algorithms were used for bed fluidization, cyclone efficiency, and filter performance. As expected, the model predicts

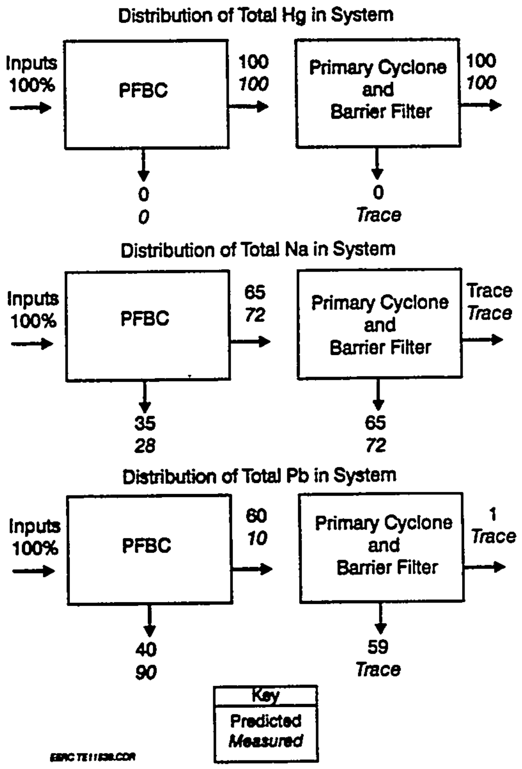


Figure 12. Trace Metal Distributions Predicted and Measured for a Pressurized Fluid-Bed Combustor

well for Hg and Na, which are found largely in the vapor state, but performs only marginally for the mostly particulate lead (primarily because of the lack of robust engineering models).

Similarily, Figure 13 shows the predictions of the seven trace elements in an air-blown entrained flow gasifier at three different locations. In this case, no partitioning was performed to remove some materials in the gasifier slag, ash filter, or gas cleanup device. The calculations were performed assuming all of the inorganics passed throughout the entire system. This is shown as an example of how the different conditions affect the overall vapor phase distributions. In actual application, only the vapor phase and a limited amount of the very fine particulate would pass throughout the system.

FUTURE WORK

Future work will involve the following:

- Completing the integration of the TraceTran computer code.
- Verifying the predictive capability of TraceTran using data obtained from larger-scale systems.

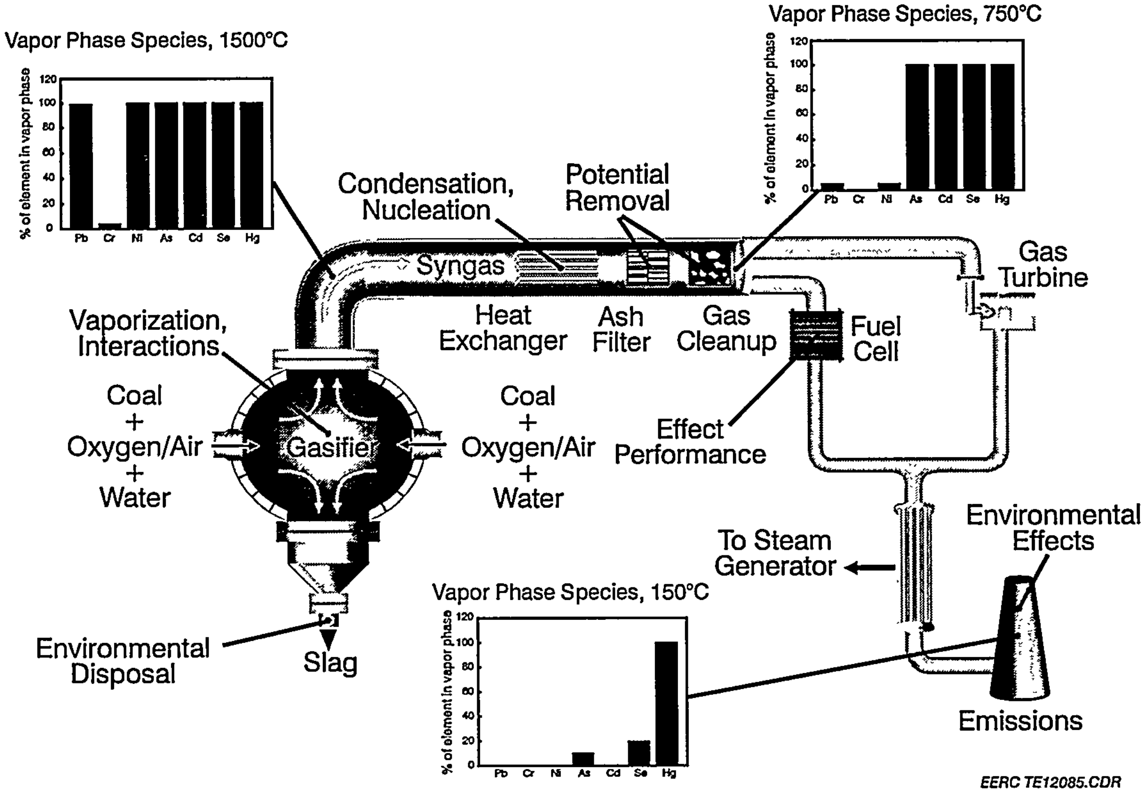


Figure 13. Trace Metal Vapor Phase Distributions in an Air Blown Gasification System at Various Locations. No Partitioning of the Solids Has Been Performed.

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Basic Research Supported by the Office of Basic Energy Sciences U.S. Department of Energy

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Division of Materials Sciences
Office of Basic Energy Sciences
U.S. Department of Energy

ABSTRACT

This presentation will outline the basic research activities of the Office of Basic Energy Sciences (BES) of the U.S. Department of Energy.

The BES mission is to develop understanding and to stimulate innovative thinking needed to fortify the Department's mission. Of particular focus in the presentation are the research programs, amounting to about $10 million, supported by the Materials Sciences Division and the Chemical Sciences Division which are fairly directly related to electrochemical technologies.

Session 5

Molten Carbonate Fuel Cells

5.1 ERC Commercialization Activities

CONTRACT INFORMATION

Cooperative Agreement	DE-FC21-95MC31184
Contractor	Energy Research Corporation 3 Great Pasture Road Danbury, CT 06813 (203) 792-1460 (telephone) (203) 798-2945 (telefax)
Other Funding Sources	ERC, EPRI, NRECA
Contractor Project Manager	Hans C. Maru
METC Project Manager	Tom George
Period of Performance	December 1994 to July 1999

BACKGROUND INFORMATION

The ERC family of companies is anticipating market entry of their first commercial product, a 2.8-MW power plant, in the second quarter of 1999. The present Cooperative Agreement provides for:

- Commercialization planning and organizational development
- Completion of the pre-commercial DFC technology development
- Systems and plant design
- Manufacturing processes' scale-up to full-sized stack components and assemblies
- Upgrades to ERC's test facility for full-sized stack testing
- Sub-scale testing of a DFC Stack and BOP fueled with landfill gas

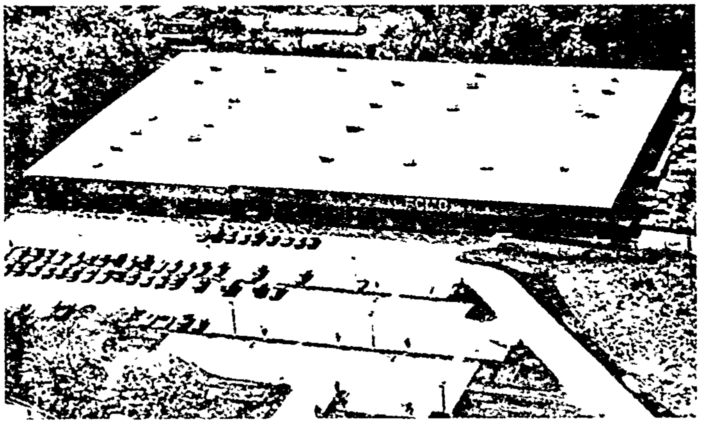
This paper discusses the first item, that of preparing for commercialization.

ERC's formal commercialization program began in 1990 with the selection of the 2-MW Direct Fuel Cell power plant by the American Public Power Association (APPA) for promotion to the over 2000 municipal utilities comprising APPA's segment of the utility sector. Since that beginning, the APPA core group expanded to become the Fuel Cell Commercialization Group (FCCG) which includes representation from all markets - utilities and other power generation equipment buyers.

Through a strong combination of Federal and privately sponsored R&D accompanied by demonstration programs, ERC's family of companies is aggressively making the transition from the laboratory to a manufacturing capability and market entry. Once underway, the planned line of products will reap significant rewards for U.S. enterprise in terms of jobs, revitalization of some primary manufacturing industries (steel, electronics, electrical and control industries, and other equipment suppliers), and other related service-based support organizations. This should also provide a contribution to a more favorable trade balance from the anticipated foreign demand for these systems. Of most importance, however, DFC power plants meet our demands for efficiently produced electricity respectful of our environment in a surging economy.

To serve commercial customers, ERC established two subsidiaries, the Fuel Cell Manufacturing Corporation (FCMC) and the Fuel Cell Engineering Corporation (FCE). Over $12 million of private sector financing has been raised to launch these two entities through an initial capital formation effort in support of ERC's fuel cell activities. Most of these funds have been applied to build and equip an operational manufacturing plant that is supplying DFC stacks and modules for the coming demonstration projects. The FCMC manufacturing facility is shown in *Figure 1.* Both of the new firms are establishing their respective commercial functions through contracts with the DOE and the Santa Clara Demonstration Project (SCDP). As part of the curretn program, a verification of the commercial power plant design is planned.

Figure 1. FCMC's Manufacturing Facility



Since being selected in February 1990 by the Fuel Cell Commercialization Group (FCCG) as the company and technology of choice, FCE/ERC and the FCCG have entered numerous cooperative efforts, all derived from the spirit of the collaborative initiative. ERC/FCE have shared technical data, test experiences, and system design requirements with the group. Each of the buyers/members has executed confidentiality agreements to allow a free transfer of material allowing for a robust interchange to hone the 2-MW power plant to a market-acceptable product.

The FCCG-FCE collaboration is precedent-setting in that a buyer's group is actively participating in the design, demonstration, and commercial introduction phases of a new technology product into a conservative, risk-averse industry. One approach to presenting the dynamics of the commercialization scheme is to describe the separate functions comprising the effort and the ways in which the parties are interacting in each to affect a positive outcome. The FCCG-ERC collaboration is now five years old. The interactive program scopes the functions of information transfer, system planning, design and engineering, early production unit model contract, and organizational cooperation at corporate executive levels. These activities are conducted through the following mechanisms:

- Committee meetings
- Board of Director meetings
- Executive Committee guidance
- Program triggers
- Program reviews

The past two years have seen this structure become increasingly vital to the technical, business, and economic directions of the commercialization effort. ERC has prepared a Commercialization Plan defining the series of technical, business, and financial paths for completion of product development, manufacturing, demonstration and production units, anticipated customers and markets, and the implementation of a corporate organization. This plan is currently being updated.

MARKET CONDITIONS

The utility business is topsy-turvy. Open transmission access, retail wheeling, and EWGs are new terms that translate to deregulation and intensified competition to serve loads. All this comes at a time when electricity demand is growing (as predicted) in response to the ongoing economic recovery. The general response by investor-owned utilities, the segment most impacted by these new situations, is no new large plant orders, further deferrals in older plant retirement, implementing reductions in staff, and cutting r&d expenditures. The net effect is that reserve margins are decreasing while most procurement actions for new generation are overseas.

As these predicted power shortages become reality, u.S. utility and industrial sector expansion could be inhibited. Large new coal- and nuclear-based power plants cannot be constructed inside of 8-10 years from project definition due to the long licensing cycles and environmental barriers. Further, the high investment costs are at risk given the litigious nature of dissident groups who have effectively paralyzed implementation of large central station projects.

Fuel cell power plants can be easily sited due to their inherent quiet, clean-operating features. Because they are modular they can rapidly be erected and place in service. California is setting the precedent in permitting fuel cell by waiving most, if not all, of the combustion limitations applicable to conventional plant site requirements. With these barriers streamlined, new fuel cell-based capacity as well as repowering units can proceed to the construction phase more rapidly. Cost savings will accrue in the shorter overall lead-times from receipt of order to start-up. Fuel cell engineering corporation (fce) projects that our first product, a 2.8-Mw factory-built power plant, can be on-site within 12-16 months after receipt of factory orders. Energy cost savings will accrue due to high efficiency, ease of siting, power quality, cogeneration option, and low environmental intrusion for electricity customers while providing a fuels flexible power generator to utilities. Having said all this, what has changed since the early 1990s that could impact market attractiveness, applications, and procurement of the remaining financial support to complete the development cycle?

NATIONAL POLITICS

In November 1994, the voters elected a Republican controlled Congress for the first time in modern memory. The "Contract with America" called for reductions in (energy) R&D from among the areas where the new Congress targeted dramatic spending reductions along with calls to eliminate the DOE itself. As was the case with the Reagan/Bush Administrations of the 1980s, big ticket programs and high cost demonstrations were said to be the province of the private sector and market-driven forces according to the Republican led legislature. Similarly, environmental improvements gained over the past 15-20 years through stricter emission and waste clean-up standards are being challenged by this same Congress as being too costly and therefore limiting corporate profits and trade competitiveness. Adding further to this situation, fossil energy costs remain low encouraging liberal consumption patterns by all end users.

Juxtaposing on to these movements, the government appears committed to deregulating the electric utility industry. This change may not, of itself, act against utilities accepting better generation investment, the role of transmission and distribution companies aided by the statistics describing a current overcapacity condition. Questions effecting utility investment strategies include loss of their limited monopoly status by allowing open access to all power providers, changes in depreciation allowances for assets including real property and equipment comprising generation facilities, and retail wheeling. These are business tidal waves for the normally conservative, long protected, investor owned systems.

These current issues diminish somewhat the energy conservation-new energy technologies movement of the last decades. The attractiveness of fuel cells for their quiet, high efficiency, low polluting operation remains. However, in preparation for the increased competition among power providers portended by the national policies mentioned, utilities are streamlining their staffs and reducing their characteristically high priority, as well as lowering their funding for R&D. This comes at a particularly crucial period for advanced fuel cell systems where market entry is anticipated by the late 1990s with full scale demonstrations remaining to qualify the commercially configured power plants. It may take a few years for the utility sector to shake out may before the industry resumes a new "business as usual" stance as these impacts become the norm.

Figure 2. Marketing, Distribution and Services Plans

- **MARKETING PLANS**

  MAXIMIZE FCCG PARTICIPATION-CUs ORDERED WITH EPUs
  REGIONAL SALES REPS IN DIVERSIFIED MARKET LOCATIONS
    UPA in 5-State territory
    Others chosen as *quid pro quo* for support, business opportunities
  STRATEGIC ALLIANCES FOR SITE AND SERVICES ACTIVITIES
    FCE-Fluor Daniel
    Timing tuned to program progress-enhances value to FCE/ERC
    Manufacturing & Production Facilities strategies development
  CENTRALIZED SALES-ADVERTISING MANAGEMENT PROGRAM INITIALLY;
  REGIONAL/CUSTOMER SENSITIVE PLAN LATER

- **SUPPLIER & PRODUCT AWARENESS INITIATIVES**

  EXHIBIT-PARTICIPATE IN TRADE MEETINGS
    EEI/APPA/NRECA/IEEE
  PRESENTATIONS-ON AGENDAS AT TARGET MARKET MEETINGS
    NCPA/ESEERCO/EPRI

- **DISTRIBUTION PLAN**

  CONCENTRATE IN TARGETED EARLY MARKETS
    California, Northeast and North Central States
  FORM JOINT VENTURES OR FRANCHISES
    Use FCCG Buyers, other "local knowledge" firms

- **SERVICE PLAN**

  DEVELOP MODULE, SPARE PARTS SUPPLY DEPOTS
  LINK O&M CAPABILITIES WITH DISTRIBUTION FUNCTIONS

MARKETS AND APPLICATIONS

Despite the preceding scenario, in our opinion, the longer range outlook continues to appear most promising for advanced fuel cells and especially the carbonate-based Direct Fuel Cell (DFC) in megawatt class systems. This optimism stems from the nation's sustained strong economic performance, the national will to actually reduce the deficit, and the requirement that older generation systems must be replaced and new capacity must be added for an increasingly electricity dependent U.S. and world.

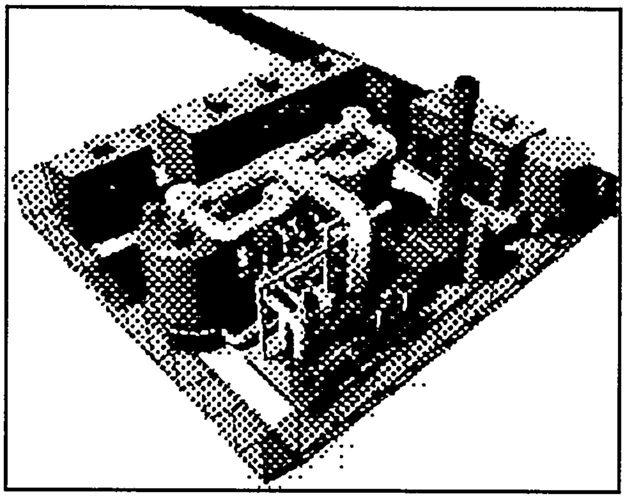
In 1983-84, ERC selected a two-megawatt DFC commercial power plant for our initial commercial product. This size was felt to respond to the widest breadth of potential with a broad diversity of applications. Many of these are shown in *Figure 2.* The application shown are particularly suited for the 2-3 megawatt class system now in primary design. The member-buyers of the Fuel Cell Commercialization Group (FCCG) are working with FCE to finalize a standard contract for their unit purchase, to assist with the plant's performance envelope, and to provide for O&M. FCCG members represent over 20% of the nation's generation capacity and a prime source for early and sustaining unit sales if their initial experience with the first units bears out the potentials of this product and technology.

The 2.8-MW Power Plant

First conceived in 1984, our analysis of the unit's conceptual design indicated power could be produced at an efficiency approaching 60% when fueled by natural gas. The original design concept maximumized fuel utilization, recovery and recycle of unspent hydrogen and product water. Called the "Integrated System" it is believed to be an ultimate carbonate fuel cell system capable of the highest conversion efficiency known by any generator requiring only a natural gas (methane) source for operation (no facility water is needed). This was the commercial baseline until 1991 when a "simplified system" design was conceived. In this configuration, only the most basic power plant functions were retained to truly simplify the system and lower the unit costs. With Fluor-Daniel's assistance, we determined that the unit cost is about $300/kW less while still providing relatively high efficiency at 50%, and preserving the environmentally attractive features of fuel cell plants. The commercial entry unit chosen is the "Simplified System" although there is a high interest to also be able to offer the other system design somewhat later.

The unit, shown in *Figure 3,* requires less than 4500 ft^2 for its installed location and because of its low noise and low pollution operation, can be easily sited in urban or suburban settings, either indoors or adjacent to existing structures. Because of the internal reforming feature, the hydrogen inventory is consumed instantaneously thereby imposing no safety constraints on the unit's installed location.

Figure 3. FCE's 2.8-MW Power Plant



Over the next 12-18 months, FCE expects to have over 30 executed contracts for the first production series, called Early Production Units (EPUs).

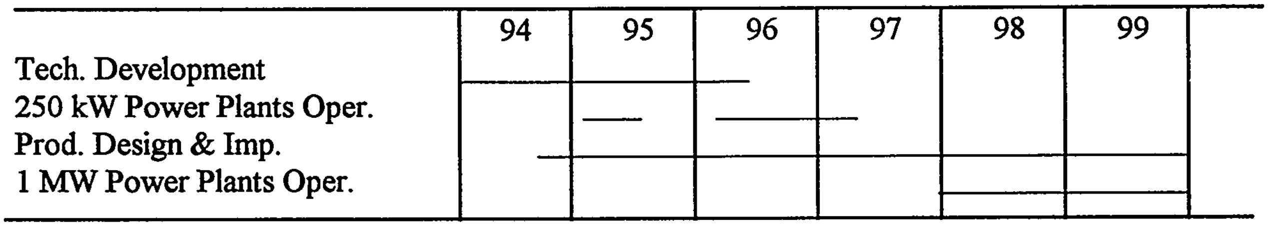
5.2 IMHEX® Fuel Cells Progress Toward Commercialization

CONTRACT INFORMATION

Cooperative Agreement	DE-FC21-95MC30133
Contractor	M-C Power Corporation 8040 S. Madison Street Burr Ridge, Illinois 60521 (708) 986-8040 (708) 986-8153 (Fax)
Other Funding Sources	GRI, EPRI
Contractor Project Manager	Joseph A. Scroppo
Principal Investigators	Rene M. Laurens Vincent J. Petraglia
METC Project Manager	Diane Hooie
Period of Performance	December 19, 1994 to December 18, 1999

Schedule and Milestones

Program Schedule



OBJECTIVE

The overall goal of M-C Power is the development and subsequent commercialization of Molten Carbonate Fuel Cell (MCFC) stacks. More specifically, MCFC's based upon the Internally Manifolded Heat EXchange (IMHEX®) plate design created by the Institute of Technology. In order to achieve the aforementioned goal, M-C Power assembled a formidible team of industry leaders. This group, refered to as the IMHEX® Team, has developed a strategy to move decisively through the stages of Technology Development and Product Design and Improvement through commercialization.

This paper is to review the status of the overall commercialization program and activities. It will also provide an overview of the market entry product. Furthermore, we will evaluate the opportunities and benefits this product brings to a competive power industry.

BACKGROUND INFORMATION

Introduction

Industry Environment. As the U.S. gas industry emerges from the initial uncertainty created from deregulation, the successful participants have developed a greater understanding of market forces. Now offering a myriad of unbundled services the industry allows customers to pick and choose the proper combination of natural gas services at the lowest price. Understanding conditions that enable savvy competitors to prosper is also providing the gas industry with a major advantage as the electric power industry begins its transition toward competition. Competition in the electric market began in the 1980s, but experts anticipate major changes during the next decade. Even today, power marketers are positioning themselves for aggressive competition, even though electricity sales and distribution are still controlled by regulated electric utilities. These new players are approaching individual industrial, commercial, and even residential customers in an effort to define the opportunities being created with the unbundling and repackagine of electric energy services. As competition in the electric power market increases, there will be a downward pressure on the prices charged by the various players.

Participants, which view themselves as integrated suppliers of a full-line of value-added energy services, will become the winners in today's and tomorrow's energy marketplace. Currently a variety of tools help these participants be more competitive and provide lower cost of services to their customers. One technology, that will enable participants to expand their role in the energy services marketplace, is the molten carbonate fuel cell. Distributed power plants based on this technology are uniquely qualified to fill the demand for localized, efficient, and environmentally friendly energy supplies at the lowest possible cost to the consumers. This type of equipment will allow energy service companies to supply the majority of a customer's electric and thermal energy needs from small power plants located at the customer's facilities. These distributed resources offer benefits over and above the commodity electricity and heat produced. Reliability and quality of the electricity, utility grid support, and independence of self-generation are a few examples of these benefits. Over the past decade the regulated electric industry was successful at establishing barriers to cogeneration and self-generation, such as supplemental, backup, and/or standby charges, modified rate structures, and project buy-outs. With full competition in the electric marketplace, some of these services will be available from competitive sources. Therefore, market forces will set the price and distributed generation will compete on a level playing field. For fuel cells to achieve the promise they have always had in the technologist's mind, developers must introduce products that are cost-effective and durable. The IMHEX® Team realizes that its product must compete with commodity priced energy supplies while providing customers added-value services. The commercialization program is focused on developing and verifying the technology that will allow the Team to introduce a cost-effective product with the durability demanded by the marketplace.

Development History

The IMHEX® Team. M-C Power was founded in 1987 with the sole mission of developing and commercializing the molten carbonate fuel cell technology that was initially researched at the Institute of Gas Technology (IGT). In 1989, IGT completed the initial transfer of its sub-scale technology. Since then, M-C Power has concentrated on developing the technology beyond bench-scale models to full-scale stacks that form the building blocks for future commercial power plants. In parallel, a commercialization Team was established with the experience and expertise needed to bring this evolving technology into the marketplace. IGT and M-C Power concentrate on advanced fuel cell technologies and full-scale stack manufacturing, respectively. Bechtel Corporation provides the process and engineering capabilities to design fully-integrated power plants based on the IMHEX® fuel cell stacks. Stewart & Stevenson Services, Inc. brings to the Team not only the ability to cost-effectively package power plant subsystems, but also the marketplace distribution, sales, and service infrastructure demanded by the power industry.

Planning. A series of incremental development and verification steps are focused on the Team's ultimate goal of developing a market-responsive, 1-MW MCFC power plant. Verifications of prototype, 1-MW power plants are scheduled to begin during the late 1990s and initial commercial deliveries to begin before the end of this decade. The commercialization program has been organized into several distinct phases as shown in Figure 1. Each phase achieves critical milestones involved in our quest to commercialize this environmental benign and energy efficient conversion technology.

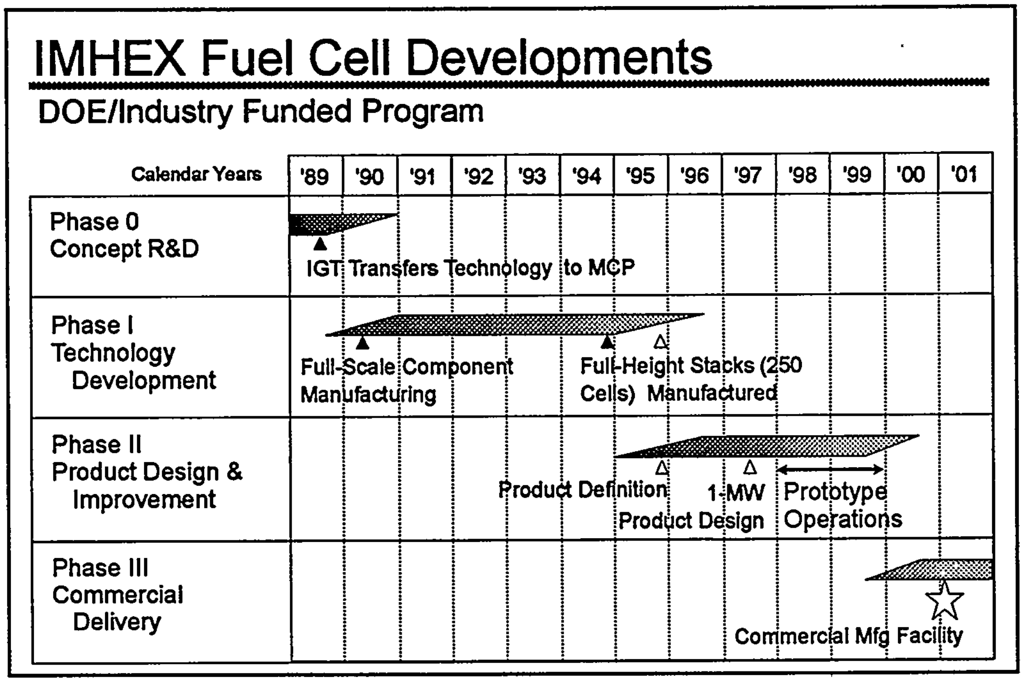


Figure 1 IMHEX® Fuel Cell Commercialization Program Phases

Phase 0 - Conceptual Research. The conceptual research and development work of all the stack components took place in IGT's laboratories during the 1980s. The cell design configuration is based upon the Internally Manifolded Heat EXchanger (IMHEX®) design of the separator plates, which takes on many of the characteristics of plate and frame heat exchanger. Concepts for manufacturing processes and initial component formulations were defined and verified in several $0.1 m^2$ ($1\text{-}ft^2$) stack tests. This technology was subsequently transferred to M-C Power in the form of licensing agreements.

PROJECT DESCRIPTION

Phase I - Technology Development

Scaling-Up of The Technology. During the first phase of the program M-C Power scaled-up the technology to dimensions required for cost-effective stacks. The first step involved scale-up and verification of cells with active areas approaching $1\text{-}m^2$ ($11\text{-}ft^2$). This size enables each cell to generate between 1-kW and 1.5-kW of electricity. After verification testing of short-stacks, the technology was incorporated into full-height stacks consisting of several hundred cells. These full-height stacks were subsequently integrated into developmental power plants that are providing the initial proof-of-concept along with the transition into Phase II developments.

Much of M-C Power's scale-up activities concentrated on the successful implementation of full-size separator plate configuration. This component integrates the functions of cell-to-cell electrical connection, anode/cathode gas separation, formation of process gas flow fields, and soft-rail structures that form wet-seal barriers between internal process gas manifolds and flow fields. The separator plate is made from sheet metal, which has been pressed to form the various features and welded into a final assembly.

Scale-up of the manufacturing processes was a prerequisite to developing commercial-size cells. The world's first manufacturing facility, designed exclusively for fabricating MCFC components, was operating during the early 1990s. These processes are based on tape casting large-area porous components at widths approaching 1.3 meters (52 inches) and component thickness within plus or minus two thousandths of a centimeter. Anodes, cathodes, matrices and electrolyte tapes are all initially manufactured using this tape casting equipment. The metal components (anodes and cathodes) are then sintered in a continuous, dual atmosphere furnace resulting in high quality components with tight control of both bulk porosity and pore size distribution. The individual components are then cut to shape and added to a separator plate prior to final stack assembly.

Beginning in 1992, several short-stacks were fabricated and tested to verify M-C Power's full-scale cell components and continuous manufacturing processes. Verification involved the testing of six stacks containing 20-cells, each producing at least 20-kW of electricity. These tests proved many of the unique characteristics of the IMHEX® cell stack design and laid the foundation for continued developments.

Phase II - Product Design and Improvement

The Product Design and Improvement activities began in 1995 in parallel with the final steps of the Technology Development efforts. The major focus of the Phase II activities is to address cost reduction issues and to establish the commercial readiness of the power plant, stack technology, and marketplace infrastructure. The Team's efforts will initially address these issues at the component level, followed by verification of advanced technologies in prototype power plant hardware.

RESULTS

Unocal Proof-of-Concept

With the successful completion of the initial scale-up verifications, M-C Power and its commercialization Team focused on verification of the IMHEX® technology in full-height stacks integrated into proof-of-concept power plants. The goals of these efforts are to demonstrate the potential of IMHEX® technology in distributed generation and cogeneration applications and to provide the engineering foundation for power plant integration and process optimization. The first proof-of-concept power plant is a 250-kW developmental power plant located at Unocal's Fred L. Hartley Research Center in Brea, California (shown in Figure 2). The full-height stack was fabricated and acceptance tested at M-C Power's facilities in Burr Ridge, IL in October 1994 and shipped to the site in November. Stewart and Stevenson completed the mechanical skid assembly in parallel. Both of these major subsystems were integrated with a conventional natural gas reformer and power conditioner previously installed at the site.

Project Status. In January 1995, the Unocal power plant installation and construction was completed. With this milestone achieved, M-C Power, Bechtel, Stewart & Stevenson, and Unocal personnel began the process and control test phase. The startup team overcame several operational and design issues during this phase. The most significant effort involved the modifications of the reformer so that it would operate near full load conditions.

Significant effort was also expended in debugging the control system and instruments. The process and control testing was completed and on May 4 the stack was integrated into the power plant.

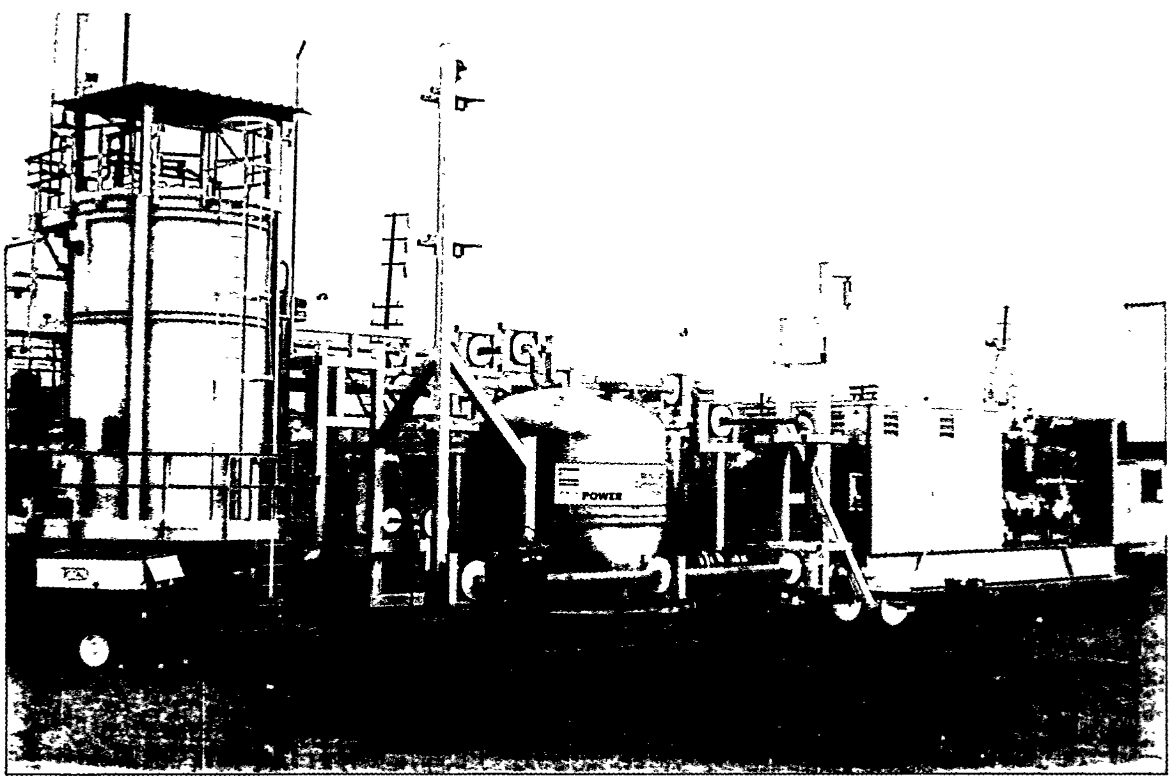


Figure 2 The 250-kW Fuel Cell Power Plant at Unocal's Fred L. Hartley Research Center, Brea, Ca.

During the end of May and early June the startup team conducted two runs in an effort to achieve normal operation. During the first run the stack heaters shorted to ground. This failure slowed the heat-up rate significantly. It was therefore necessary for the startup team to deviate from the normal startup procedure. The stack achieved operating temperature and open-circuit voltages. Several attempts were made to place the stack on load, but its performance was far below design expectations and the nominal performance achieved during the acceptance test at M-C Power. The first run was terminated on May 22, 1995.

A second run was initiated on June 16. Anticipated open circuit voltages were once again achieved, and the internal manifolds in the stack maintained design pressure differentials of over 10 in water column. Although the power plant would maintain low power levels and electricity was exported to the Unocal grid, attempts to sustain significant levels failed. At this point the startup team concluded that the stack was not going to achieve normal operation and decided to initiate a cool down. Preliminary analysis indicates several potential causes, but the primary focus is on the off-design condition used during the initial run when the team deviated from the normal startup procedure. After further assessment these conditions may have potentially damaged the active components, however verification of this requires shipping the stack to M-C Power for further analysis. The team is currently modifying the Unocal project plan to return the stack for diagnostic activities.

Initial Conclusions. Although electric power production never achieved significant levels at Unocal, a great deal of power plant design, fabrication, transportation, siting, operational, and maintenance information was obtained. Operation of the mechanical equipment and the reformer revealed many important areas that must be addressed for success during commercial installations. Several lessons learned are currently being used to modify the control and operational strategies planned for the San Diego Gas and Electric power plant. The IMHEX® stack design was successfully scaled-up ten-fold. The critical stack conditioning phase, which includes the final binder removal, initial electrolyte melting, and cathode oxidation, was accurately implemented in M-C Power's acceptance test facility. During this test the stack produced the target power level of 80 kW (which is the test facility limit). Inverter performance was flawless at low power operation. Operator training was successfully completed, and the Unocal staff achieved practical operational experience during the installation, process and control testing, and heat-up cycles.

San Diego Proof-of-Concept. A second proof-of-concept test is progressing with operation scheduled for early 1996. San Diego Gas and Electric is the host utility for this test. Gas Research Institute is supporting the unit construction and is using this test as a major milestone in their support of the commercialization activity. This power plant is based on a 250-cell/250-kW full-height IMHEX® stack that incorporates an advanced separator plate configuration. The unit also integrates an advanced plate type reformer that allows the full-integration of the power plant processes. Anode exhaust gas from the stack is used to provide the heat for the endothermic reforming reaction. This process change will increase the plant electrical efficiency to approximately 45%. SDG&E has selected the Naval Air Station Miramar, in San Diego, Ca. as the site.

Project Status. M-C Power has fabricated the repeat components and has completed sub-assembly. As a result of the Unocal experience, the assembly of the first SDG&E stack has been delayed until after the analysis of the Unocal stack is complete. Control and startup procedures for the SDG&E power plant are being reevaluated based upon the lessons learned during the Unocal process and control test and initial runs. It is anticipated that the stack assembly delay will not hold up the scheduled operation of the SDG&E power plant in early 1996. Power plant process designs and layouts are complete, and Bechtel has placed orders for the major balance-of-plant components. Stewart & Stevenson has begun mechanical skid fabrication with shipment scheduled for later this year. The advanced plate type reformer has completed factory testing and is being prepared for shipment.

FUTURE WORK

Phase II - Product Design and Improvement

The major barrier to successful fuel cell commercialization has been the higher first cost in comparison to conventional equipment. The MCFC technology is no different, and therefore, the Team's goal is to define and develop advanced stack and power plant technologies that allow the introduction of a cost-effective product even during the low production volumes of the initial few years. M-C Power has defined a range of advanced technologies which have the potential to achieve this goal. These technologies address the two major cost reduction areas of the stack - separator plates and non-repeat hardware. Advanced separator plate designs reduce the number of components needed for the assembly from the fifteen pieces (used in Unocal) to two pieces. Cost reduction of the non-repeat hardware will be achieved through creative engineering efforts which eliminate hardware and integrate functions. In parallel, cost reduction in the repeat components are also being examined. These include faster manufacturing processes and reduced raw material through thinner components and reduced scrap.

Verification tests and power plant efforts are directed toward the construction and operation of a prototype, 1-MW power plant. This unit is intended to reflect all of the design and operational features of the Team's market entry product. The first 1-MW power plant is scheduled for operation during the early part of 1998. Southern California Edison will host this test at their Highgrove Generating Station in Grand Terrace, CA. Product design and definition activities completed in 1994 have projected that the market entry unit can achieve electrical generating efficiencies as high as 60% and overall efficiencies of over 80% with the recovery of by-product heat. Summary characteristics of this unit and an initial layout design are illustrated in Figure 3. The Team estimated the capital cost of this product would be in the \$1500/kW range with production volumes of only 20 to 50 units per year. This price relates to cost of electricity in the 4 to 7¢/kWh range depending on business and application assumptions. Achieving a cost-competitive pricing structure in low production volumes is believed to be critical to marketplace success as the power industry in the U.S. evolves with increasing competition.

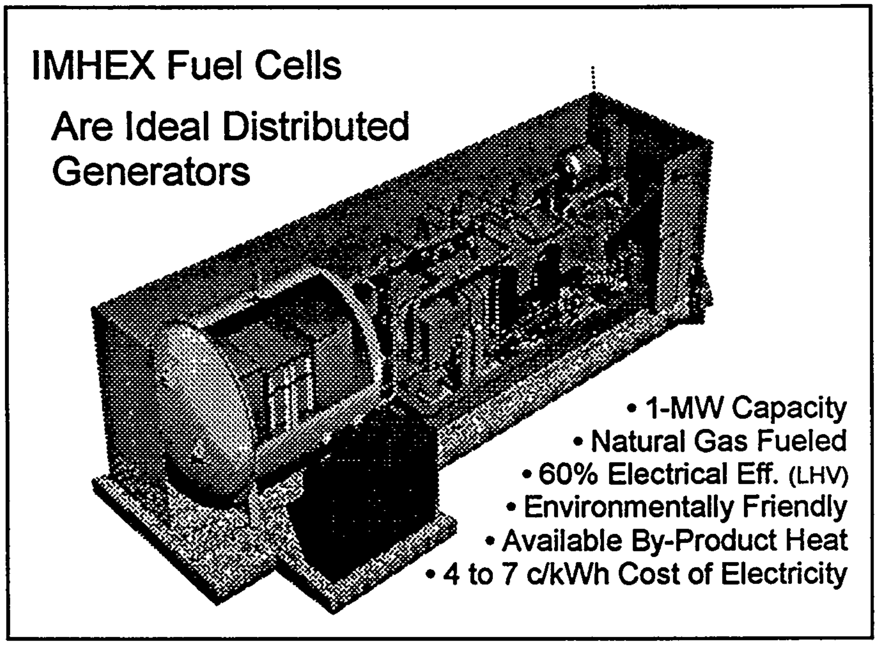


Figure 3 Projected Market Entry, 1-MW IMHEX® Fuel Cell

Phase III - Commercial Delivery

Overview. Commercial Delivery phase of the program is scheduled to begin in 1999. The Team's product introduction strategy begins with gradual penetration of commercial power plants following the successful operation of prototype units during the previous phase. Initially, M-C Power's developmental facility will be used to supply stacks for commercial deliveries, and Stewart & Stevenson will expand their manufacturing and assembly floor space. Delivery schedules are projected to include an initial six units during the first year, followed by ten and twenty the following years. The initial few years of production are limited to help manage the risk of new product introduction and to establish the experience base for product durability and reliability in real world applications. M-C Power will use this time period to establish financing for a semi-automated production facility that is scheduled for operation beginning in 2000/2001. This facility will have the manufacturing floor space for three stack production lines and manufacturing equipment for the first line will be procured. This one line when operated at full-capacity (3-shift operation) is projected to have the capacity of approximately 100 MW of annual production. If market demand meets the Team's expectations, a second and third line of manufacturing equipment will be needed by the 2005/2006 time frame.

Market Opportunities

Industry Situation. The U.S. power generation industry is in the middle of evolutionary changes brought on by deregulation and increased competition, which creates both opportunities for the IMHEX® power plant and uncertainty for the

Team. This evolving marketplace needs advance power generation technologies, such as the molten carbonate fuel cell. This is driven by the industry's need to retain loads through better customer service, decreased capital risks, incremental investments, stronger environmental regulations, and increased generation efficiency to balance the risk of escalating fuel prices.

Assessing and evaluating the market for IMHEX® fuel cell power plants is an important element of the Team's Commercialization Program. Understanding the evolving power generation industry helps ensure that the market entry product developed during the 1990s meets the expectations of our future customers and is responsive to marketplace requirements. Estimated market demand and product acceptance will aid M-C Power and the Team in establishing market entry strategies, manufacturing facilities, and financial requirements.

Segmentation. Market opportunities for stationary power generation can be divided into several categories based on the type of application, ownership, and/or operating strategy. Applications can be categorized by size, location, and function of the facility. The Team believes there is one principle market for its fuel cells during the initial ten years of commercial production. The market is distributed generation (<10 MW capacity). This segment includes electric-only substation applications, cogeneration opportunities within the commercial building and industrial sectors, improved customer service functions in the area of power quality and reliability, pipeline compressor stations, and specialty fuel sources (such as landfills, digesters, and renewable gasification processors). Common to everyone's view is that these small capacity generators must be strategically sited within the electric power distribution network. Selecting sites that provide added-value benefits above the electric capacity and energy they produce, allow distributed generators to compete with the commodity price of retail energy at the customer's site.

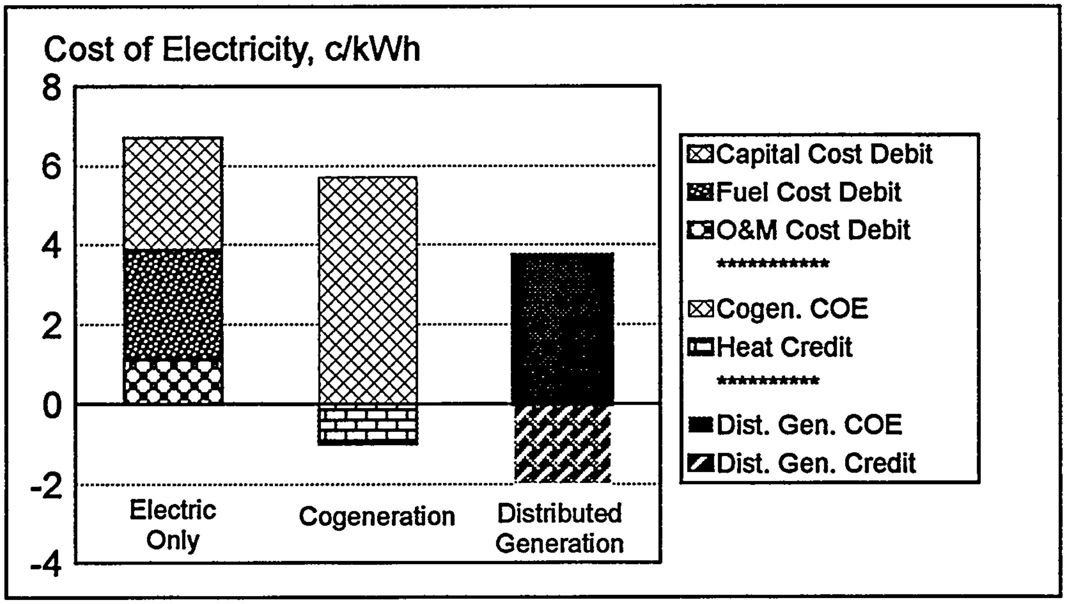


Figure 4 Cost of Electricity in Three Typical Applications

Product Requirements. The product definition activities over the past years have solicited marketplace inputs from the Alliance to Commercialize Carbonate Technology members. Surveys of industry representatives and economic evaluations of the marketplace have shaped the Team's perspective of market-responsive characteristics. First, the power plant must be durable and reliable in its intended applications. Second, it must be cost-effective to install, operate, and maintain. The power plant must produce electricity competitive with the 4 ¢/kWh to 7 ¢/kWh range typical in light industrial and commercial sectors. With the increasing competitive environment within the power industry, these characteristics must be achieved with the initial market entry power plants well before final product maturity.

Economic Analysis. Initial analysis has concluded that the projected IMHEX® power plant's cost of producing electricity is 6.7 ¢/kWh in electric-only applications with a fuel price of $4/MMBtu. This cost of electricity is reduced to 5.7 ¢/kWh in cogeneration applications and to 3.7 ¢/kWh when additional distribution system benefits are included. Figure 4 illustrates these costs and provides a breakdown of energy cost between capital, fuel, and operation and maintenance. Additional economic analyses were conducted to assess the influence of varying capital cost, fuel prices, and operating capacity factors. With a fuel price at $2/MMBtu or with a capital cost approaching $1000/kW, the cost of electricity in typical cogeneration applications drops below the 5¢/kWh range.

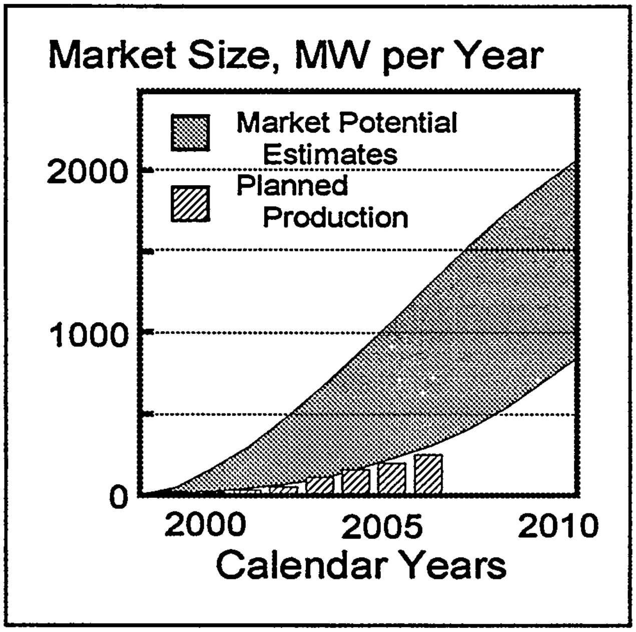


Figure 5 Market Estimates for IMHEX® Fuel Cells Compared to Planned Production

Market Entry. Penetration analysis were completed to estimate the potential rate of market demand during the initial ten years of commercialization. Two cases were evaluated to represent a conservative and an aggressive penetration and are illustrated in Figure 5. In both cases the IMHEX® power plant captured a 4% maximum market share of the total installed U.S. generating capacity. Market capture followed a typical "S" shaped penetration curve beginning in 1999. In the conservative case the IMHEX® product reached its marketplace maturity 40 years after market entry, while under the aggressive scenario maturity was achieved in 20 years. The results suggest that the IMHEX® market share could range from 300 MW/yr to over 1,000 MW/yr by 2005. Demand continues to increase with estimates ranging from 800 MW/yr to over 2,000 MW/yr by 2010. These penetration rates are greater than needed to establish a viable production business for the product. In fact, the Team's production estimates are below the conservative case to help ensure management of the technical and business risk associated with introducing an advanced technology product to the market.

CONCLUSION

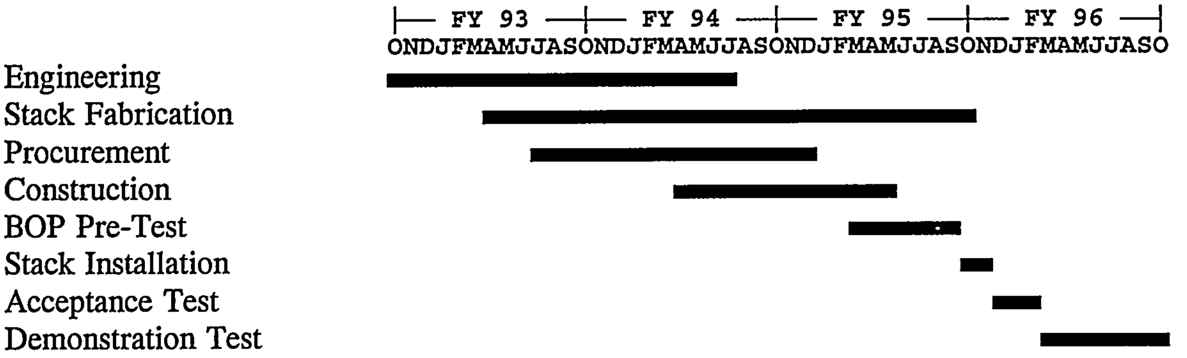
In the U.S. as the gas industry continues its competitive evolution and the electric industry begins its, the concept of supplying energy service will become more central to the strategies of both industries. Few participants in the industry can think of themselves through their previous regulated points of view. Competition will force both to take new perspectives on their business. One important tool in meeting these competitive forces will be efficient, environmentally friendly technologies that can be used to transform fuels, such as natural gas, into the energy products and services their customers need. The fuel cell power plant is one product that holds great promise as competition increases. The IMHEX® Team is dedicated to developing a cost-competitive product before the end of this decade and has focused its ongoing program activities toward this goal. Only through cooperative developments and partnerships with the energy service companies of tomorrow will the Team be successful.

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
Other Funding Sources	Santa Clara Demonstration Project Participants
Contractor Project Manager	Anthony J. Leo
Principal Investigators	Anthony J. Leo (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 direct carbonate fuel cell power plant, such as high efficiency, low emissions, reactive power capability, and high reliability and availability. The project will also provide design input for pre-commercial early production power plants.

BACKGROUND INFORMATION

The project is an outgrowth of the ongoing direct carbonate fuel cell 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 direct 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.

In 1990 ERC formed two subsidiary organizations essential to the commercialization effort: The Fuel Cell Manufacturing Corporation (FCMC) and the Fuel Cell Engineering Corporation (FCE). FCMC will manufacture carbonate stacks and multi-stack modules, initially from its pilot manufacturing facility in Torrington, CT and then from a commercial manufacturing facility. FCE is responsible for power plant design, integration of all subsystems, sales/marketing, and customer services.

FCE began work on the Santa Clara demonstration power plant design in 1990, and submitted a preliminary design to the SCDP participants 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 preparation 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 balance-of-plant (BOP) and the overall plant construction, testing, and operation is being provided by the Santa Clara Demonstration Participants and FCE. 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 (CA), Electric Power Research Institute, National Rural Electric Cooperative Association, Sacramento Municipal Utility District, and Southern California Edison Company. Salt River Project and the Northern California Power Agency (NCPA) are also supplying some project funding through a consortium agreement with the City of Santa Clara. In addition, the California Energy Commission is providing funding to support the balance-of-plant pre-test activities.

PROJECT DESCRIPTION

The 2MW plant is the world's first application of a commercial-scale carbonate fuel cell power plant on an 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, plus space required for an access road around the plant.

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

FCE is providing over-all management of the project. The stack design has been developed by ERC based on technology from ongoing development programs. The stacks are being fabricated by ERC's manufacturing subsidiary, FCMC. Fluor Daniel, Inc. (FDI) has served as the power plant A&E and also provided engineering support to the design of the multi-stack fuel cell modules.

RESULTS

Balance of Plant Design, Procurement, and Construction

The power plant process is ERC's simplified design, which had been developed in earlier system optimization studies. Process design in the demonstration program 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.

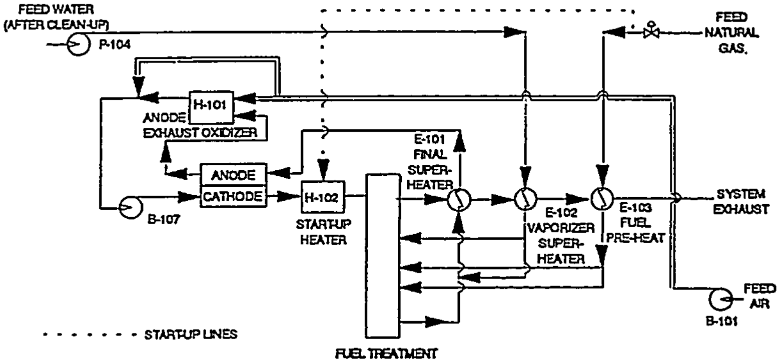


Figure 1. Simplified Process Flow Diagram for 2MW SCDP Power Plant

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 cell anodes consume 75% of the fuel, and the anode exhaust stream is sent to a catalytic oxidizer, H-101, where the residual fuel is 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 cathode exit stream is sent to the heat recovery system, where it provides 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 power conversion unit (PCU), water treatment system, instrument air compressor, and backup power generator. The layout of the plant is shown in Figure 2.

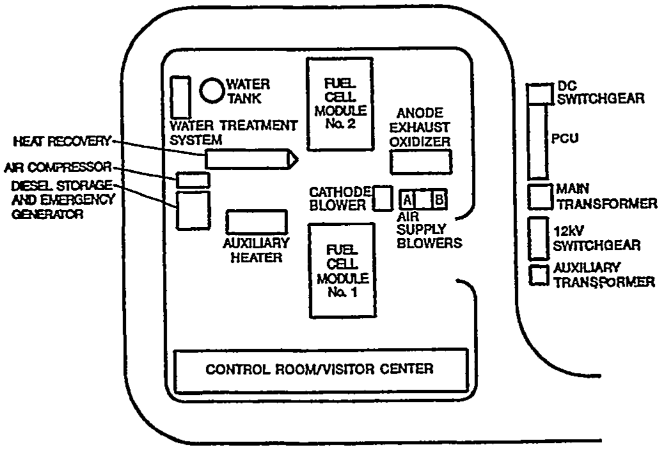


Figure 2. 2MW SCDP Power Plant Layout

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. BOP construction began in April, 1994, and was completed in early June, 1995. Figure 3 shows an overview of the site at the completion of the BOP construction effort. The figure shows piping spool pieces installed where the fuel cell stacks will ultimately be connected. These spool pieces will allow the operational checkout of the full BOP during the pre-test period prior to stack installation.

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 are being shop fabricated and shipped to the FCMC facility in Torrington, CT. There, the fuel cell stacks are being installed in the enclosure and all piping, electrical, and instrumentation work within the enclosure is being completed. All piping connections 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.

A model of one of the 1MW modules is shown in Figure 4. Each 1MW module will consist of two submodules with associated gas distribution piping, instrumentation connections, and electrical connections. Figure 5 shows a rendering of the completed plant with the submodules installed into the 1MW modules.

Fabrication of fuel cell stack components began in April, 1993, with the initiation of cell repeating component production. The stack hardware design was finalized based on a series of full-size stack tests conducted in ERC's technology development program. The first four stacks have been installed into the first submodule enclosure, which is being prepared for shipment to the site in early August.

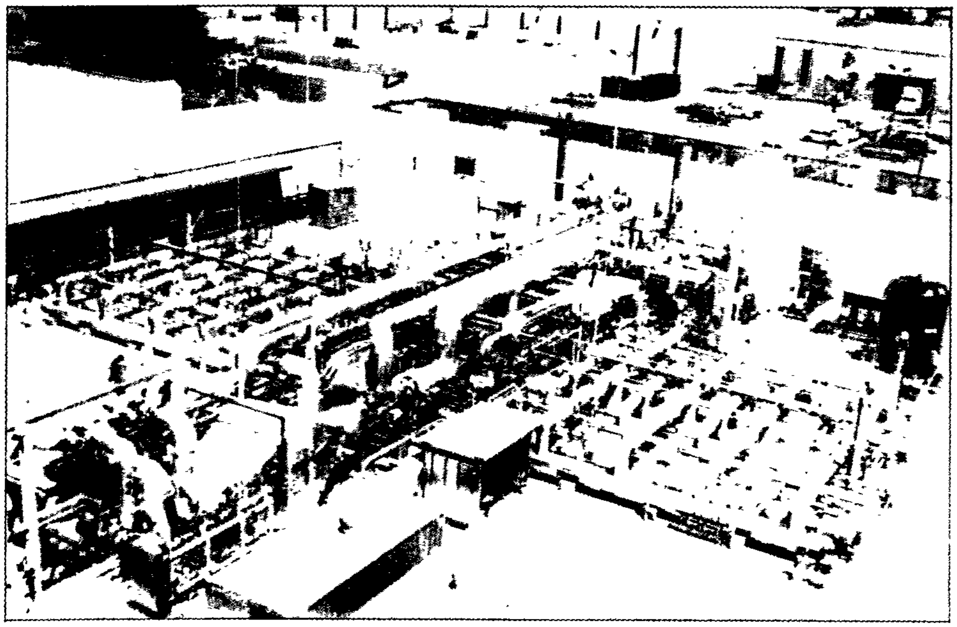


Figure 3. SCDP Power Plant Overview After Completion of BOP Construction

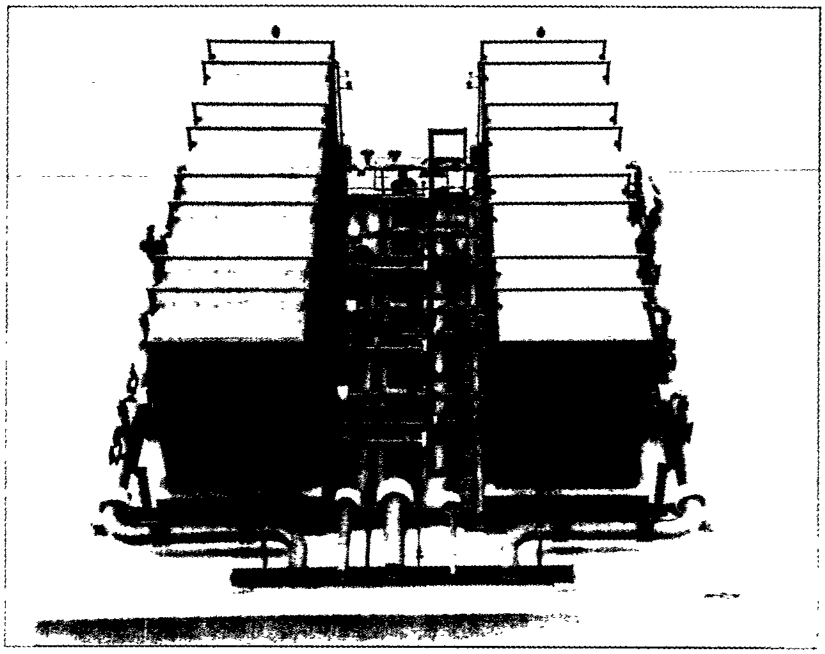


Figure 4. Four-Stack Submodule Model Photograph

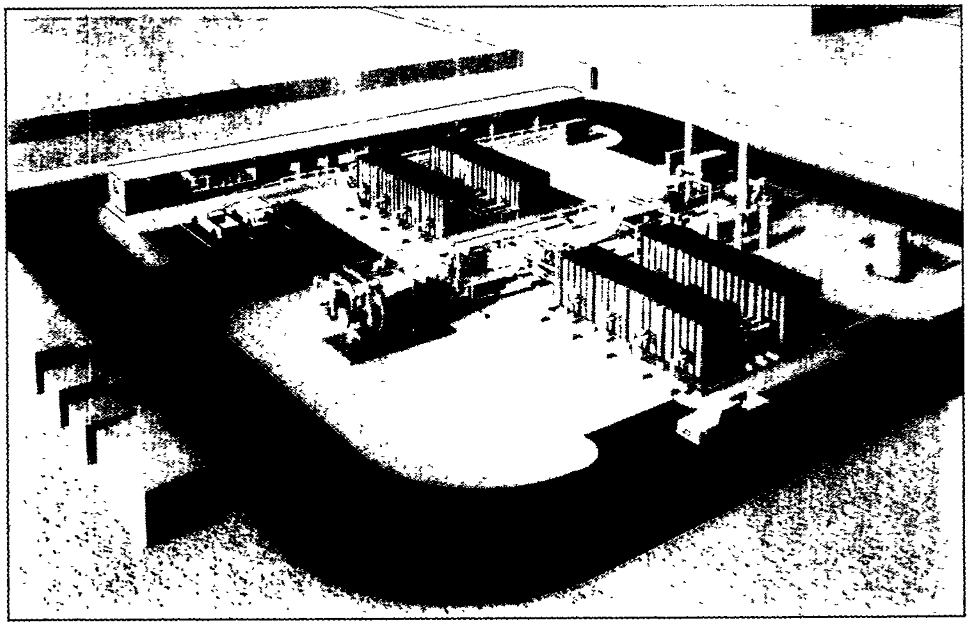


Figure 5. Rendering of Completed Plant with Stacks Installed

An additional eight stacks are in various stages of final assembly, and components for the remaining stacks are being assembled. Completion of all sixteen stacks is scheduled for October, 1995 and shipment of the last four-stack submodule to the site is scheduled for November, 1995.

Plant Operations Status

The full complement of plant operations personnel have been hired and trained, and the balance-of-plant pre-test effort has been initiated. BOP pre-testing is being conducted prior to installation of fuel cell submodules in order to check out the critical BOP subsystems and the BOP as a whole before the system is operated with fuel cell stacks. The BOP pre-testing effort was started in April, 1995. This initial work consisted of testing plant subsystems as they were completed and turned over to FCE. BOP pre-testing will continue through September, 1995. Installation of the fuel cell submodules into the plant will be done in October and November, and be followed by the plant start-up and acceptance testing. The acceptance testing phase is expected to take three months, and will be followed by a sixteen month endurance test phase. Total scheduled operating time for the system is 10,000 hours.

Regulatory and Permit Status

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. The final City occupancy permit is being issued in two phases. The first phase has been issued for the completed BOP construction effort, and the final phase will be issued following the completion of stack installation. Care, custody, and control of the plant has been transferred from the FDI construction team to the FCE operations staff.

FUTURE WORK

As described above, the power plant BOP pretesting effort will continue through September, 1995, when the stack installation effort will be initiated. Following the completion of stack installation, the power plant acceptance testing phase will begin.

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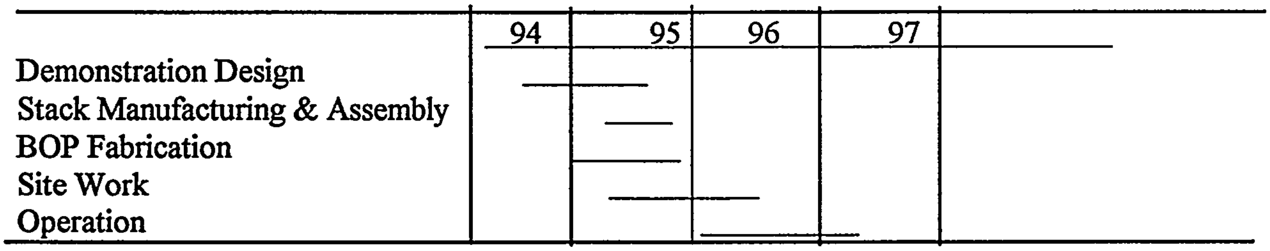
M. Farooque, R. Bernard, P. Patel, A. Skok, C. Yuh, J. Doyon, and H. Maru, Energy Research Corporation, Danbury, CT. 1994. Direct Carbonate Fuel Cell Development. *Fuel Cell Seminar, Program and Abstracts, November/December 1994*, 234.

5.4 Molten Carbonate Fuel Cell Product Development Test at SDG&E

CONTRACT INFORMATION

Cooperative Agreement	DE-FC21-92MC28065
Contractor	M-C Power Corporation 8040 S. Madison Street Burr Ridge, Illinois 60521 (708) 986-8040 (708) 986-8153 (Fax)
Other Funding Sources	ARPA, Bechtel, EPRI, GRI, SDG&E, Stewart & Stevenson
Contractor Project Manager	Joseph A. Scroppo
Principal Investigators	Rene M. Laurens Vincent J. Petraglia
METC Project Manager	Diane Hooie
Period of Performance	September 30, 1992 to March 31, 1997* (pending approval)
Schedule and Milestones	**Program Schedule**



OBJECTIVE

The PDT design objectives will include improved performance at reduced cost compared with the UNOCAL demonstration project. Several specific objectives that differentiate the San Diego Gas & Electric PDT project from the UNOCAL demonstration are the following: packaging designs are more compact in the PDT program; it will also have longer unattended operation and increased reliability. Additionally, the experience gained during the design, construction and start-up of the UNOCAL power plant will be incorporated into the SDG&E design. This power plant is being designed for compatibility with the SDG&E electrical distribution grid.

Design goals for this power plant incorporate the results of product definition work and market research performed over the past few years. Design goals for the PDT demonstration unit include:

(1) High electrical and overall efficiency
(2) Cogeneration acceptability

(3) Environmental acceptability
(4) Size acceptance (size, aesthetics)
(5) Ease of operation (start-up, shutdown, unattended operation)
(6) Reliability, availability, maintainability and safety
(7) Potential for cost competitvness

BACKGROUND INFORMATION

M-C Power Corporation will design fabricate, install, test and evaluate a 250kW Proof-of-Concept MCFC Power Plant. The plant was originally going to be located at Kaiser Permanente's San Diego Medical Center in San Diego, CA. However, after thorough investigation of the costs involved and the advantages and disadvantages that this site had to offer the site was relocated to the Miramar Naval Air Station (NAS) in San Diego, which offered significant advantages, including an anticipated cost reduction for the installation and operation of the power plant. Also, the Navy personnel at the Miramar NAS are very enthusiastic and cooperative about this project, which M-C Power feels will be a great benefit during the construction, installation and operational stages of the project.

M-C Power is managing the overall project, working closely with their sponsors. Bechtel Corporation is responsible for the design, detailed engineering, construction management and start-up of the proposed MCFC PDT power plant. Stewart & Stevenson, Inc. (S&S) is fabricating the balance of plant (BOP) on equipment skids, including all the equipment skids internal piping and valving required. SDG&E is performing some work with its own staff and is using craft subcontractors.

The project, defined as MCFC Product Development Test (PDT), has been developed in accordance with DOE guidelines under the cooperative agreement established between M-C Power and the DOE. DOE is funding the design, manufacturing and fabrication of the 250kW stack for the SDG&E power plant. All costs associated with the necessary BOP are funded by GRI. Funds for testing of the demonstration power plant are being provided by the DOE. The DOD's Advanced Research Projects Agency (ARPA) has funded the site relocation to Miramar and a 20kW stack test. The remaining funded is being supplied by Bechtel Corpoaration, the Electric Power Research Institute (EPRI), Gas Research Institute (GRI), San Diego Gas & Electric (SDG&E) and Stewart & Stevenson (S&S).

The 250kW MCFC stack will be manufactured, assembled and acceptance tested at M-C Power, prior to shipment to the job site at Miramar NAS. The BOP, which includes fuel processing, thermal management/heat recovery, controls and power conditioning systems, will be integrated into a power plant system that will be shop tested at S&S's manufacturing plant in Houston, Texas. The 250kW PDT power plant will be operated for approximately a one (1) year test period. Data gathered during the power plant operation and maintenance will be analyzed by M-C Power, Bechtel and SDG&E personnel. Conclusions and recommendations drawn form this analysis will be used to define future natural gas fueled MCFC power plants that are fully responsive to the needs of potential customers.

PROJECT DESCRIPTION

The MCFC PDT Demonstration is an integral part of the M-C Power Team's comprehensive commercialization program aimed at bringing on-site MCFC power plants to the commercial market by the late 1990s. A key element of the commercialization effort involves the design and field verification of on site power plants. This is a key reason for the 250kW demonstration at the Miramar NAS.

The project has been divided into seven separate tasks which include the following: Management Plan; Demonstration Design; Stack Manufacturing; Balance-of -Plant Fabrication; Site Work; and finally, Power Plant Testing. Each task has been further subdivided with responsiblity for completion of each subtask being delegated to the appropriate team member. A seventh Task "Site Change and Verification of Improvements" was made when the project site was moved to NAS Miramar.

Task 1 Management Planning

Overview. The major focus of Task 1 is the Management of the overall project. This includes the gather information regarding permitting and environmental requirements relating to the project. The project organization is led by M-C Power Corporation as the prime contractor with the Department of Energy and the Gas Research Institute. Bechtel Corp. is responsible for system design along with procurement and construction management. Stewart & Stevenson is responsible for skid assembly and shop testing while SDG&E is responsible for site-related construction and interfacing activities. This team was formed for the specific purpose of combining the professional, technical, and managerial resources required to successfully complete the PDT demonstration project and to move the IMHEX® MCFC toward commercialization.

Management Reporting. M-C Power's Project Manager is responsible for providing overall project direction, including cost control, schedule, and performance. Resources necessary to perform the work are assigned in accordance with the forecasted requirements. Work progress is closely measured relative to materials and labor expenditures. The project schedule is continuously monitored using flow of documents (correspondance, drawings, transmittals, etc.) and personal reviews to detect trends and potential problem areas. Updated schedules provide the reporting mechanism for the execution of action plans to accelerate positive trends, reverse negative trends, and resolve problem areas. Periodic reports from project participants are analyzed for completeness and accuracy. Costs and physical progress to date are verified on a monthly basis. Data for these updates are provided by team participants.

Environmental & Permitting. M-C Power and SDG&E have prepared an environmental assessment which contains the information necessary to allow DOE to prepare documentation to support a request for a categorical exclusion. A categorical exclusion has been recieved.

Task 2 Designing

Overview. The focus of Task 2 is the design of the demonstration power plant unit. This work includes both the stack design and balance-of-plant. The goal is to have an integrated system which can verify prelimanary design concepts for the market entry unit.

Stack Design. The stack design is based on established technical, environmental, and cost goals for the integrated MCFC power plant system. The design takes into consideration manufacturing simplicity, stack cost, reliability, safety, maintenance requirements, and overall performance necessary to achieve MCFC power plant performance goals. A HAZOP analysis has been completed.

Overcoming cost penalties associated with transitioning porduct and process requirements from R&D toward commercialization is being addressed. The stack design has been upgraded using data from applied research, component development studies, pneumatic and structural analyses, and data analysis from the operation of subscale, short stack, and full size stacks.

BOP Design. The goals of the PDT demonstration include producing a compact and highly integrated plant. To help achieve this, the construction concept for the plant is to shop fabricate and minimize field erection. This concept will promote efficient packaging and allow for significant pretesting, while improving quality of construction while saving time and reducing costs.

Stewart & Stevenson is responsible for the detailed engineering of the skids including the design, engineering, and procurement of the piping, instrumentation, controls and structure. All major pieces of equipment have been recieved and are in the final stages of assembly.

Bechtel coordiantes with Stewart & Stevenson for the necessary design interface between the skids and the rest of the plant. This is an iterative process. As the fabricators find out that the skid configuration demands process changes, the process design and piping and instrumentation diagrams are revised accordingly.

Task 3 Manufacturing Development

Stack Manufacturing. The nominal 250-cell stack is the basic building block for all M-C Power IMHEX® power plant applications. We have developed a manufacturing plan under GRI sponsership which is guiding and directing the manufacturing of stack components. Installed and quailified commercial scale tape casting and sintering equipment is in place to manufacture anodes, cathodes, matrices, and electrolyte sheets. Separator plates are being stamped by a company that manufactures formed-plate heat exchangers with plan areas up to 15ft^2. Fabrication variables which affect part quality are being defined and incorporated in a comprehensive QA/QC program.

Testing and Shipping. Stack pre-conditioning is followed by an acceptance test. The acceptance test is used to verify that the stack meets performance, leakage, crossover, and electrical isolation specifications. The stack is operated to verify performance and demonstrate mechanical and electrical integrity.

Following acceptance testing at M-C Power, the stack will be placed into shipping fixtures for transport to the demonstation site. Shock and tilt detection devices are attached to the shipping container to verify that shipping and handling have been completed without exceeding maximum load specifications.

Task 4 Balance-of-Plant

BOP Fabrication. Skid mounted subsystems are being assembled by Stewart & Stevenson based upon Bechtel's design. Stewart & Stevenson has participated in review of Bechtel's design for "constructability, maintence, and cost." Skids are configured in accordance with Bechtel specifications. Stewart & Stevenson has prepared its skid design based on Bechtel's process design. The design allows for routine shipment and easy installation and interconnection at the field site. Stewart & Stevenson will insure that components are integrated into functioning subsystems that will be tested prior to shipment.

Design of the electrical panels and cabinets is being performed by Bechtel and assembled by Stewart & Stevenson. There are three major skids. The inverter and control panels are located on the electrical skid. A second skid contains the reformer, desulfurizer, and gas preheat exchangers. The stack containment vessel, recycle blowers, and turbo compressor make up the third skid. Each skid is being designed to be road transportable.

Task 5 Site

Site Preparation. A site construction plan has been prepared to guide all plant installation activities. SDG&E engineers have worked closely with Bechtel to identify the existing site conditions, define interface requirements, and develop the site construction plan. Ground breaking has taken place and underground trenching is underway. All installation activities will be thoroughly documented through photographs and in writing. The Bechtel site representative will work with SDG&E to insure that all subcontractors perform thier work in accordance with the bid documents, the construction plan, and installation specifications. Prior to turnover of the constructed facility to the Bechtel/SDG&E start-up team, pressurized components will be cleaned and leak tested, rotating equipment will be powered and checked for vibration. Continuity checks will verify component interconnections and all instrumentation and control circuits will be checked against the design specification.

Task 6 Testing Plans

Testing. M-C Power, Bechtel, SDG&E and NAS Miramar are establishing technical objectives and goals for the plant operating period. These goals will guide preparation of test plans and procedures for both steady-state and transient testing. The test plans are being designed to verify plant operating characteristics at power levels from idle to rated and during start-up and shutdown sequences. Each of the components undergoing Proof-of-Concept testing will be included in the test plan which will identify expected process flow, temperature and pressure data, electrical output and fuel consumption, emissions, and other pertinent data. Special instrumentation procedures and emission data requirements will be identified prior to start-up. The plan will include methods for acquistion and analysis of plant operating data and will specify post test dismantling and inspection procedures for the stack, reformer, and other equipment.

Task 7 Site Change and Improvement Verification

This task includes permiting, site investigation, and site design activities related to the relocation of the project to NAS Miramar. It is being funded by ARPA. An objective of this task is to verify the IMHEX® technology (using the PDT components) prior to assembly of the 250 cell Miramar stack. This task has been successfully completed.

RESULTS

Project Status. M-C Power has fabricated the repeat components and has completed sub-assembly. As a result of the Unocal experience, the assembly of the first SDG&E stack has been delayed until after the analysis of the Unocal stack is complete. Control and startup procedures for the SDG&E power plant are being reevaluated based upon the lessons learned during the Unocal process and control test and initial runs. It is anticipated that the stack assembly delay will not hold up the scheduled operation of the SDG&E power plant in early 1996. Site preparation work has begun and will be completed by early fall. Power plant process designs and layouts are complete, and all of the major balance-of-plant components will be received by the end of this month. Stewart & Stevenson has begun mechanical skid fabrication with shipment scheduled for later this year. The advanced plate type reformer has completed factory testing and is being prepared for shipment.

FUTURE WORK

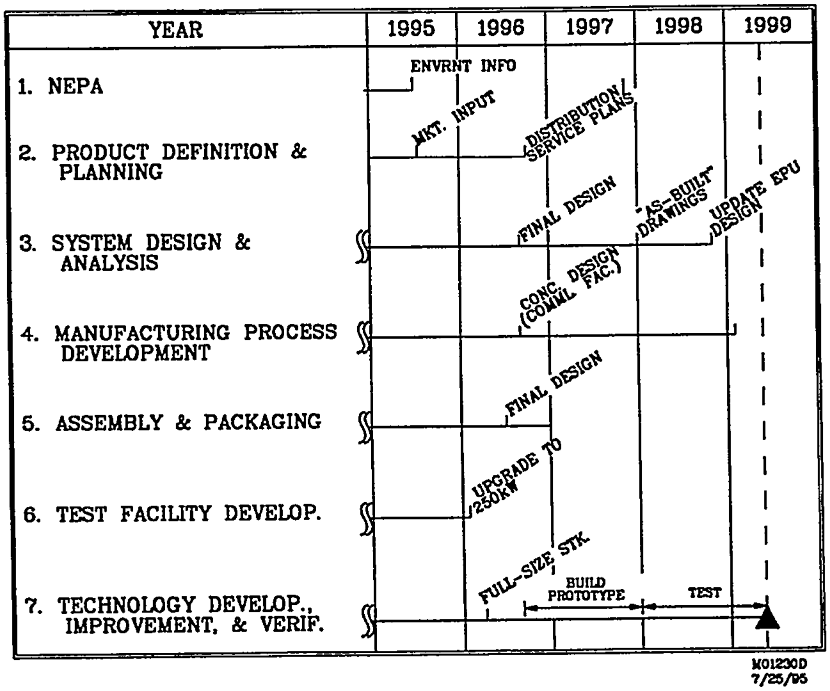
Future work for the Product Development Test includes the completion of site preparation; and expedite the major balance of plant equipment from thier respective vendors. It also includes final assembly of the MCFC stack, followed by stack pre-conditioning, acceptance testing, and shipment to the site. It is anticipated that the stack will be delivered by years end. Stewart & Stevenson will finish the BOP assembly and testing by years end. They anticiapte having the skid installed on site early in 1996. Integration of the reformer, fuel cell stack and mechanical skid will be performed by Bechtel and SDG&E staff with assistance from Stewart & Stevenon and M-C Power staff as well. Upon integration of the system the plant will be started up and tested for approximately one year.

5.5 ERC Product Improvement Activities for Direct Fuel Cell Power Plants

CONTRACT INFORMATION

Contract Number	DE-FC21-95MC31184
Contractor	Energy Research Corporation 3 Great Pasture Road Danbury, Connecticut 06813 (203) 792-1460
Other Funding Sources	DOD/ARPA, Private Sector
Contractor Project Manager	H. C. Maru, Program Director M. Farooque, Associate Program Director
Principal Investigators	C. Bentley, G. Carlson, J. Doyon, D. Glenn, A. Kush, L. Paetsch, P. Patel, C. Yuh
METC Project Manager	Thomas T. George
Period of Performance	December 20, 1994 to March 31, 1999

SCHEDULE AND MILESTONES



OBJECTIVES

This program is designed to advance the carbonate fuel cell technology from the current power plant demonstration status to the commercial design in an approximately five-year period. The specific objectives which will allow attainment of the overall program goal are:

- Define market-responsive power plant requirements and specifications,
- Establish the design for a multifuel, low-cost, modular, market-responsive power plant,
- Resolve power plant manufacturing issues and define the design for the commercial manufacturing facility,
- Define the stack and BOP equipment packaging arrangement and define module designs,
- Acquire capability to support developmental testing of stacks and BOP equipment as required to prepare for commercial design, and
- Resolve stack and BOP equipment technology issues and design, build, and field test a modular commercial prototype power plant to demonstrate readiness for commercial entry.

A seven-task program, dedicated to attaining objective(s) in the areas noted above, was initiated in December 1994. Accomplishments of the first six months are discussed in this paper.

BACKGROUND INFORMATION

The carbonate fuel cell promises highly efficient, cost-effective, environmentally superior power generation from pipeline natural gas, coal-gas fuels, biogas, and other hydrocarbon fuels. ERC has been engaged in the development of this unique technology since the late 1970s, primarily focusing on the development of the Direct Fuel Cell (DFC) technology (1-5) pioneered by ERC in the early 1980s. 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, it contributes to higher overall efficiency for conversion of fuel energy to electricity.

ERC plans to offer commercial DFC power plants in various sizes, initially focusing on the MW-Scale units. The plan is 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 with 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.

ERC has just completed a technology program for product design verification, a predecessor of the current program where the power plant design as well as the technology development were carried out for a full-size demonstration. All the planned goals of this earlier program were met (5,6). As a result of this and prior programs, ERC has built a strong technical base. To date, over 25 short stacks, three 20kW-Class stacks and four 100kW-Class stacks have been tested at ERC, accumulating a total of about 100,000 hours of stack testing experience. In addition, over 50,000 hours of stack testing was conducted at customer sites across USA and Europe. A majority of ERC and customer site tests were conducted at system conditions. In fact, several tests were conducted in integrated systems. A list of these integrated system tests is provided in Table 1. As can be seen, ERC has accumulated both natural gas and coal gas system test experience, totaling ~20,000 hours. The system test experiences have provided the foundation for the full-size demonstration plant. The 1.8MW Santa Clara Demonstration project power plant design was released for procurement in 1994. This power plant test is planned for the fourth quarter of 1995; construction status and test plan are addressed in a separate paper at this conference.

Table 1. Integrated System Test Experience To Date:
Full-Size Demonstration Power Plant Design is Built on ~20,000
Hours of Stack Testing Experience Under System Operating Conditions

Year	No. of Cells	Stack Power, kW	System Test Fuel	Test Site	Test Duration, Hour
1990	54	20	Natural Gas	PG&E, CA	300
1990	18	7	Natural Gas	Elkraft, Denmark	3,600
1992	234	70	Natural Gas	PG&E, CA	1,400
1993	246	120	Natural Gas	ERC, Danbury	250
1993	246	120	Natural Gas	ERC, Danbury	1,800
1993-1994	54	20	Dual Fuel (Natural Gas/ Coal Gas)	Destec, LA	3,900
1994	258	130	Natural Gas	ERC, Danbury	2,000
1994	18	8	Natural Gas	Elkraft, Denmark	6,500
					19,750

In December 1994, ERC launched the final phase of the developmental efforts to advance the power plant design from the demonstration stage to the commercial prototype stage through a concerted, multiyear product improvement effort. Emphasis is being placed on power plant simplification, standardization and packaging, component cost reduction, and performance and life improvement and life prolongation. Significant progress was made in each of the major areas during the reporting period. These accomplishments will be discussed in this paper.

PROJECT DESCRIPTION

To build on the successes of previous programs (discussed in the background section), ERC has embarked on this government/private-sector cooperative program for development and demonstration of a MW-Class power plant. The multipronged program activities have been initiated to attain this overall goal. Figure 1 shows key programmatic events (shaded areas) and their interrelationships. The product definition and specification will be arrived at with input from users. Detailed power plant system and packaging designs will be prepared based on the product definition and the results of stack and BOP development. A 2.8 MW prototype modular power plant representative of an early production unit (EPU) will be constructed and tested. Building of a commercial manufacturing facility for EPU's will take place based on the experience and data generated in the current program.

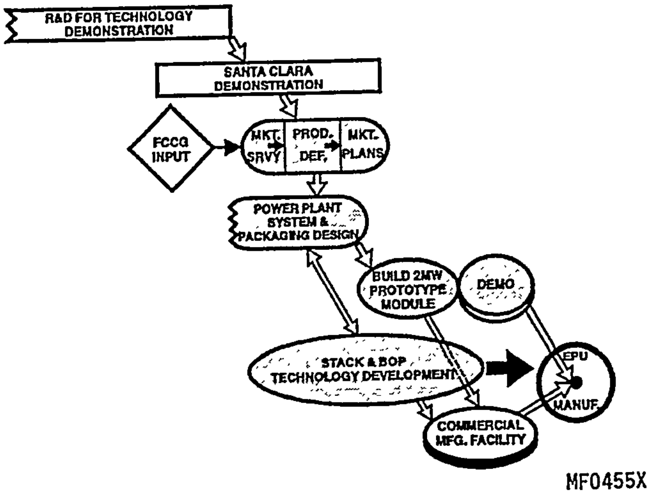


Figure 1. Effort and Interaction of Key Project Events:
The Program Activities will Culminate in a MW-Class Commercial Prototype Demonstration

The project technical team is assembled to supplement all relevant expertise required for product design, improvement, and verifications. The team consists of:

- Energy Research Corporation (ERC), R&D arm focusing on market-entry product improvement, coordinating the effort under all program areas;
- Fuel Cell Engineering (FCE), a subsidiary of ERC, is concentrating on product definition oversight and overall plant construction management and customer service;
- Fuel Cell Manufacturing Corporation (FCMC), another subsidiary of ERC, focusing on manufacturing process development and stack module fabrication;
- Fuel Cell Commercialization Group (FCCG), Washington, D. C. , a group of potential buyers of early production units, collaborating with FCE in product definition, system design, utility system planning and information distribution;
- Fluor Daniel Inc. (FDI), Irvine, CA, assisting FCE in power plant and manufacturer plant design;
- Jacobs Applied Technology (JAT), Orangeburg, SC, assisting in assembly and packaging of fuel cell stack and BOP modules;
- AEG Daimler Benz Industry/Motoren-und Turbinen-Union, Germany, conducting parallel cell and stack technology development focusing on endurance testing and cost reduction.

RESULTS/ACCOMPLISHMENTS

In this program, ERC is developing the detailed design of the commercial entry MW-Class power plant. Initially, ERC is defining the generic power plant specifications and design which will be applicable to a majority of site conditions. The pipeline natural gas composition, water supply quality across the USA, and grid connection requirements have been surveyed; the seismic map of the USA has been reviewed; and federal regulations as applicable to new power plants have been examined. The power plant requirements and the generic power plant design specifications are being developed with these inputs and ongoing interactions with utility users. The baseline natural gas power plant will be for Seismic Zone 2, grid-connected with 15kV breaker-class switchgear. Full range of U.S. pipeline natural gas compositions, except peakshaving gas, will be useable. Other fuels such as peakshaving gas and landfill gas will require additional fuel treatment equipment. The diesel fuel processor design is being developed experimentally under a parallel program. A diesel fuel processor as available from this parallel effort will be interfaced with the generic natural gas power plant so that the ERC power plant can be used for dual (diesel and pipeline natural gas) fuel fixed base DOD applications.

The Santa Clara full-size power plant demonstration unit is currently at the final stage of construction. Based on this power plant design experience, and the requirements established in consultation with the users, the commercial power plant design is being developed. The conceptual design of the plant has been defined.

The plant performance and design criteria are shown in Table 2. The baseline power plant is rated at 2.85 MW on natural gas and has a heat rate of 5900 Btu/kWh (58% efficiency based on lower heating value).

The power plant design has been further simplified from the demonstration design. Several equipment items have been eliminated. As shown in Figure 2, compared to the demonstration power plant, the commercial power plant will not include the emergency diesel generator (and associated fuel storage), auxiliary heater, and the cathode blower. Furthermore, the packaging of the heat recovery unit, which produces the steam and preheats the fuel stream, has been improved. The stack module design has also been optimized. As a result of these design developments and skillful packaging of the major equipment, the power plant hot piping length has been reduced significantly. A computer rendition of the power plant is presented in Figure 3. The power plant will be comprised mainly of two stack, three electrical, and three BOP modules. Modules can be grouped into truck-transportable units and are designed to minimize as well as simplify field connections. Field erection and connection of factory assembled modules will be completed in less than one month time. All the modules are accessible and field maintainable.

Table 2. Commercial Power Plant Design and Performance Criteria:
The Power Plant is Rated at 2.85MW on Natural Gas

Plant Rating, Net AC	2.85 MW
Heat Rate (LHV) at Rated Power	5,900 Btu/kWh (58% LHV)
Footprint	<4500 ft^2
Maximum Emissions at Plant Rating	
SO_x	<Detection Limit
NO_x	<Detection Limit
CO_2	770 lb/MWh

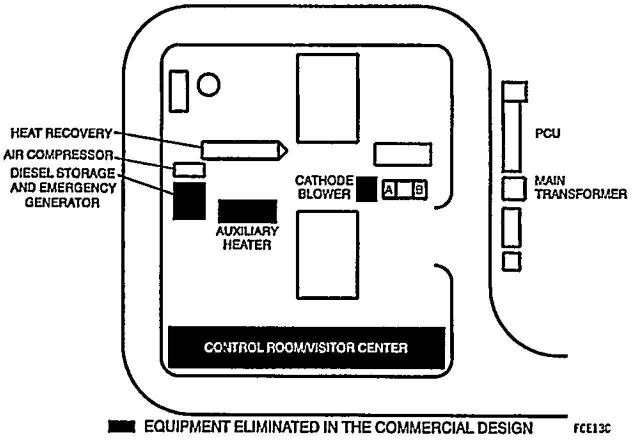


Figure 2. Santa Clara Power Plant Plot Plan: Emergency Diesel Generator, Auxiliary Heater, and Cathode Blower Eliminated in the Commercial Design

The commercial power plant has a footprint of <4500 ft^2, which is approximately one-ninth the size of the Santa Clara demonstration power plant.

The owner's cost, based on this design, is projected to be approximately $1250/kW in 1995 dollars. The cost distribution shown in Figure 4 points out that 65% of the power plant cost is direct material and the stack cost is projected to be only 25% of the total plant cost.

Significant progress has been achieved in optimizing the stack module design. The field demonstration power plant rectangular stack module containing four full-height insulated stacks has been replaced with an internally insulated cylindrical stack module. The cylindrical module will contain four, uninsulated stacks with approximately 50% larger cell area. The cylindrical stack module will also be truck-transportable.

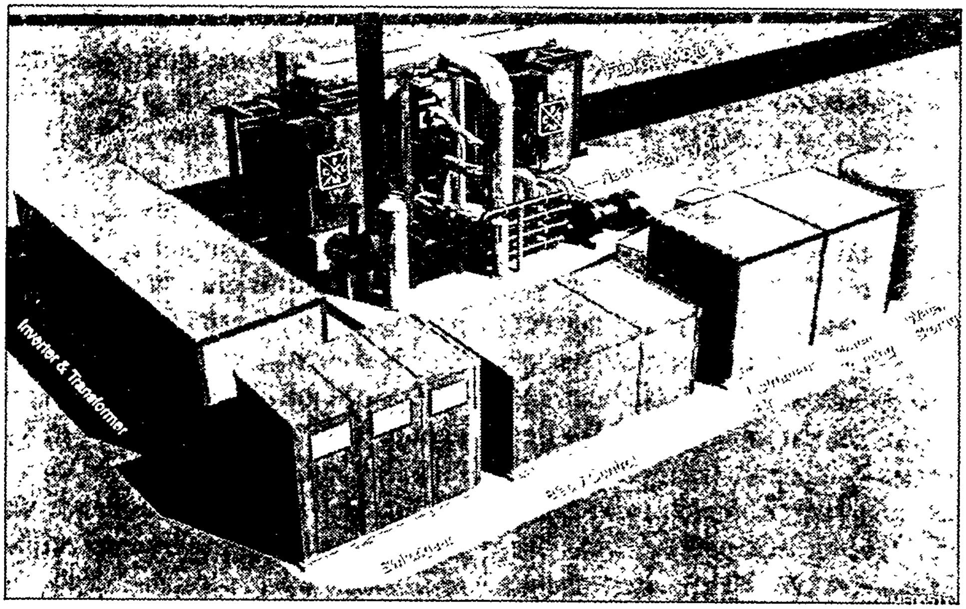


Figure 3. Rendition Of 2.85MW Baseline Natural Gas Power Plant: The Power Plant Modules will be of Standard Size, Factory-packaged, Truck-transported, and Field Connected with Minimal Labor

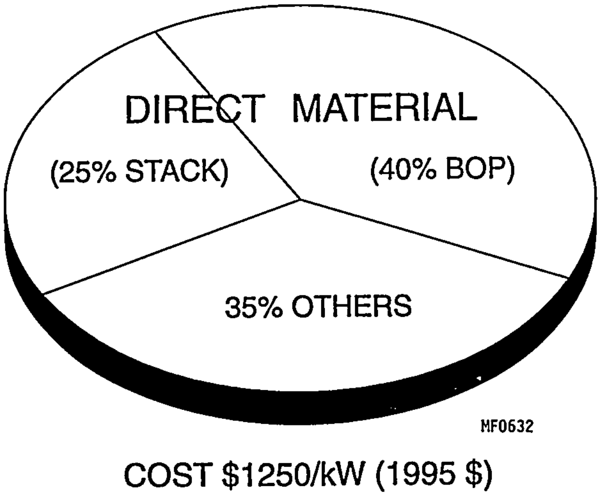


Figure 4. Power Plant Cost Distribution: Stack Cost Projected to be 25% of the Total Plant Cost

This internally insulated cylindrical design provides more efficient packaging of stacks in a module and eliminates a significant portion of hot piping and flexible piping sections. The arrangement has resulted in an increase in cell area packaging density within the module by a factor greater than five. The impact of this is pictorially shown in Figure 5. Two 9 ft wide by 37 ft long demonstration power plant modules and associated piping have been replaced with a 12.5 ft diameter enclosure of similar height. This truck-transportable module will provide 1.4 MW of AC power to the commercial plant output.

The design concept of the cylindrical module involves operation of uninsulated stacks within an internally insulated vessel, maintaining a hot process gas environment within the enclosure and current collection through the hot environment. The container, hot cable, and stack auxiliary hardware design compatible with the concept are being developed. Verification of the initial design of the enclosure and stack hardware has just been initiated. A photograph of an internally insulated carbon steel enclosure which is being used to verify the design concept is shown in Figure 6. This container has been used in conjunction with an 8kW-Class subscale stack to simulate power plant module operation.

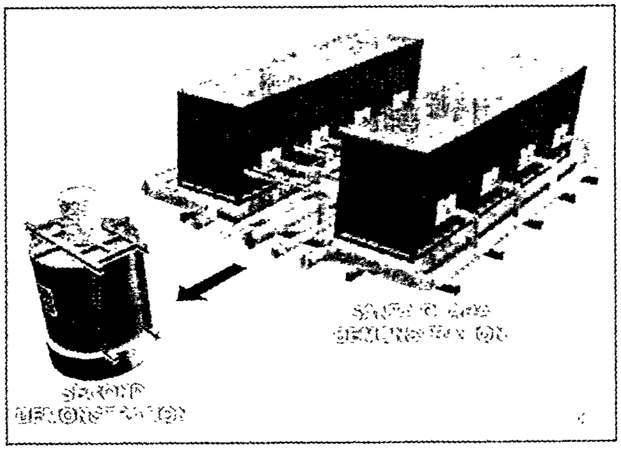


Figure 5. Stack Module: The Commercial Design has Increased Cell Area Packaging Density in a Module by a Factor Greater Than Five

The 8kW Stack AF-8-5 built with the direct internal reforming feature was previously tested as a normal insulated stack in a "cold-vessel" for about 1,700 hours. Following this initial test, the stack hardware was changed to the appropriate "internally-insulated box" compatible design. The stack was installed in the internally insulated cylindrical stack enclosure which incorporated all the key design features of the commercial stack module. The stack was tested using oxidant as the enclosure environment. A photograph of the stack on test is shown in Figure 7. The stack was operated for an additional ~1,000 hours in this mode and was mostly unattended. A comparison of cell performance at different current densities is shown in Figure 8. No change in stack performance was noticed, and no accumulation of combustibles within the enclosure was noted. The enclosure wall temperature was within the design range, thus, validating the design concept. The test was shut down for detailed post-test evaluation and design feedback.

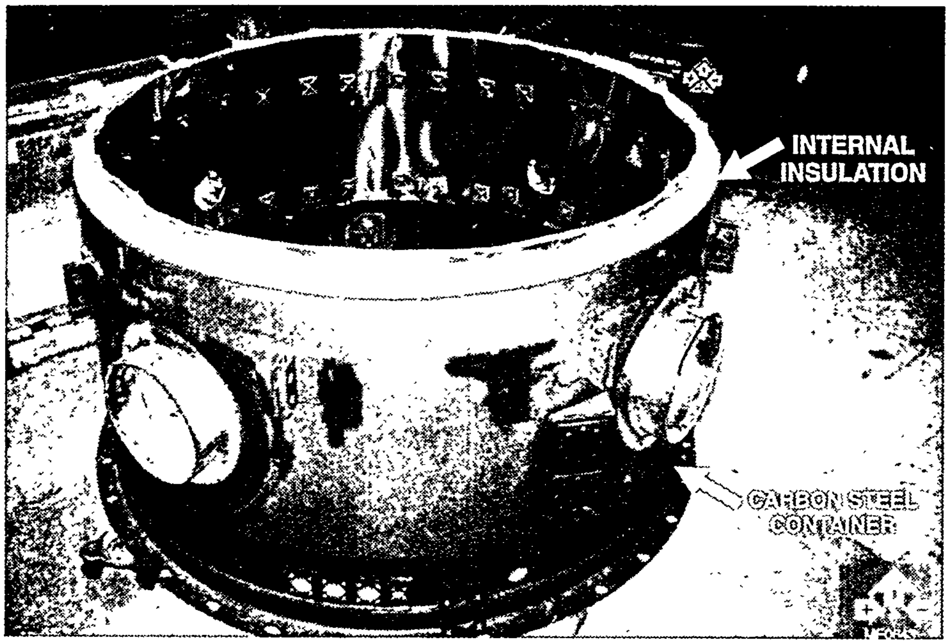


Figure 6. Stack Module Simulator:
This Simulator Provides Verification of Stack Module Design

Fuel cell cost needs to be reduced to attain the commercial cost projections. Parallel activities are underway for design simplification, manufacturing improvement, performance enhancement, and development of low cost material suppliers. An advanced bipolar plate design eliminating an expensive processing step has been developed. A very low cost wet seal corrosion protection process has also been selected. The new design has already been verified in labscale stacks. The design simplification effort has resulted in 40% reduction of cell weight. The manufacturing processes of anode, cathode and matrix have been streamlined. As a result of the manufacturing process improvements and the experience gained from Santa Clara stack manufacture, the yields of cell components have increased while further tightening tolerances. A comparison of yields and corresponding design tolerances for the current design with the baseline stack design is shown in Figure 9. The above mentioned manufacturing simplifications, reduction of material use in active components, and establishment of low-cost material suppliers have led to an overall cell cost reduction by a factor of three, as shown in Figure 10. This comprises a material cost reduction by a factor of 3.5 and labor cost by a factor of 2.4.

Efforts are also underway to reduce and further simplify the stack auxiliary hardware design. As can be seen in Figure 11, over 30% weight reduction of all the key components has been already achieved.

In addition to these manufacturing and cost reduction accomplishments, significant progress has been made in enhancing the performance and extending life. Two improved matrix designs have been identified. A subscale stack built with baseline design has been tested for 10,000 hours,

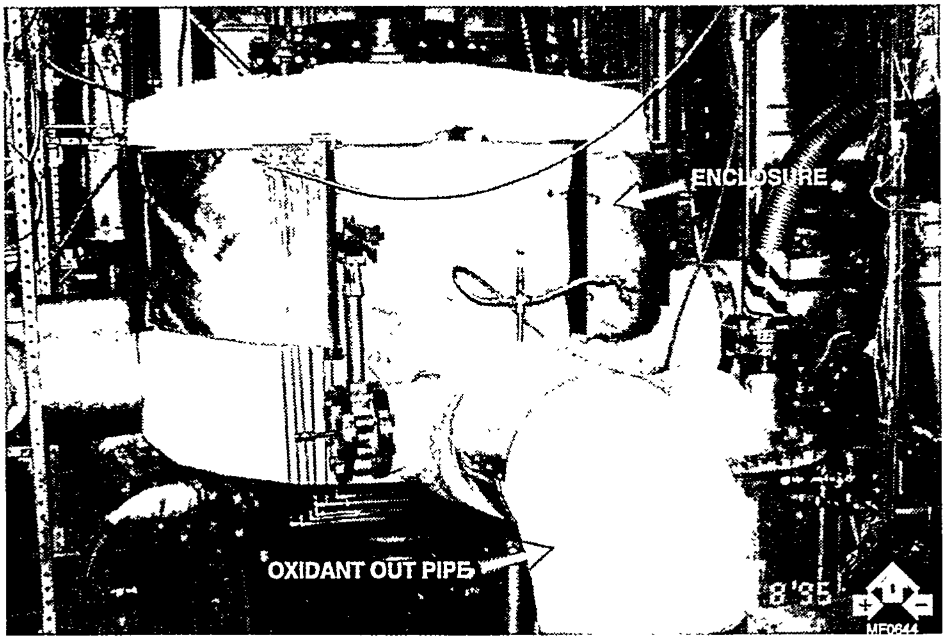


Figure 7. Stack Module Design Being Verified:
Feasibility of Commercial Stack Module Concept Tested with an 8kW Stack

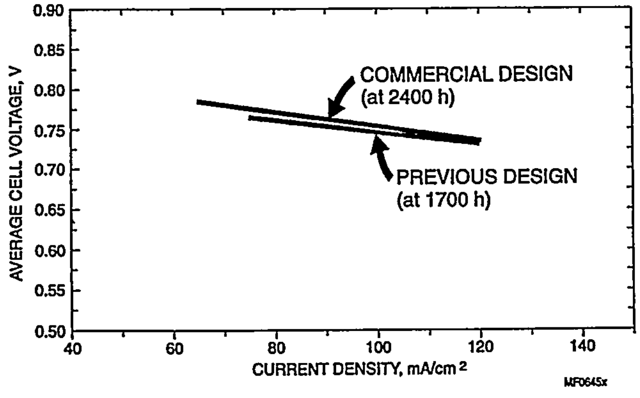


Figure 8. Stack AF-8-5 Performance Comparison (Previous Versus Commercial Design):
No Adverse Effect on Stack Performance

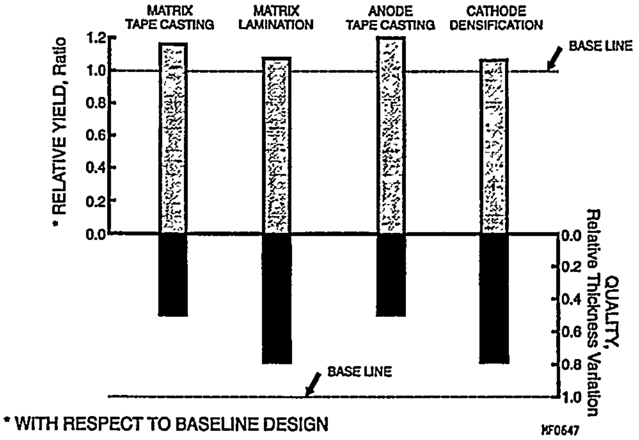


Figure 9. Active Components Manufacturing Improvement:
Yields Have Increased While the Tolerances Have Been Further Tightened

and the test is continuing. The stack lifegraph, as shown in Figure 12, shows stable performance with a decay rate consistent with SCDP demonstration goals. The methane conversion stability shown in Figure 13, indicates that the conversion is expected to remain stable for the 40,000 hour stack life.

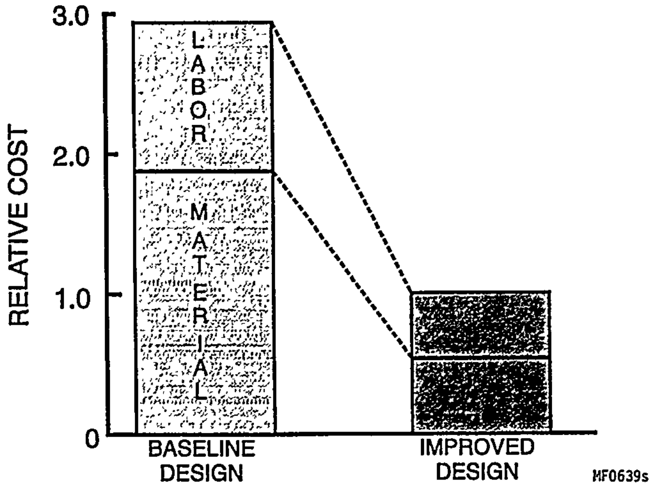


Figure 10. Cost Reduction Of Cell Components:
Cell Components Cost Reduced by a Factor of Three (on Area Basis)

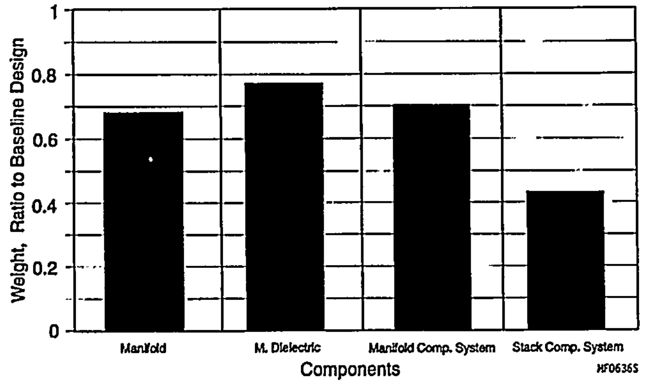


Figure 11. Stack Hardware Design Improved; Weight Reduced:
Over 30% Weight Reduction of all the Key Components has been Achieved

CONCLUSION

Through a concerted product development effort, the direct fuel cell design has been further advanced from the demonstration design. Power plant design simplification and skillful packaging have resulted in a pipeline natural gas fueled, 2.85MW rated truck-transportable, power pant with a footprint <4500 ft^2, and a cost-goal of \$1250/kW (1995\$). The feasibility of an internally insulated cylindrical stack module, which leads to efficient packaging of stacks in a truck-transportable module, has been demonstrated through a 1,000 hour 8kW-size stack test.

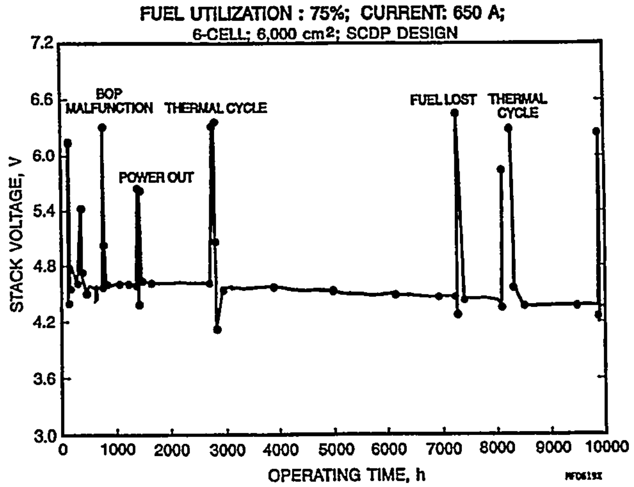


Figure 12. 3kW Stack Endurance:
Stability Goals for Demonstration Met

FUTURE WORK

Building on these accomplishments, the near-term activities of the current program will focus on finalizing product specifications, development of commercial power plant design, verification of cell and stack hardware designs in full area stacks-leading to design of a full-size stack.

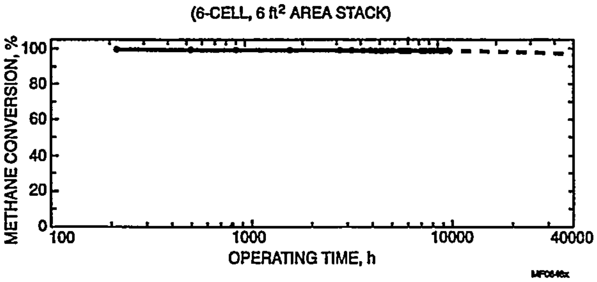


Figure 13. Methane Conversion Lifegraph: Stable Internal Reforming Stack Operation

ACKNOWLEDGEMENT

The support of DOE and DOD/ARPA for the system and technology development and testing is gratefully acknowledged.

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1. P. Patel, et al., "Internal Reforming for Natural Gas Fueled Molten Carbonate Fuel Cells", Final Report for the Period of May 1980 through June 1981 under GRI Contract No. 5080-344-0302.

2. P. Patel, et al., "Internal Reforming Natural Gas Fueled Carbonate Fuel Cell Stack", Final Report for the Period of February 1982 through December 1983, under GRI Contract No. 5081-244-0545.

3. L. Paetsch, et al., "Molten Carbonate Fuel Cell Development", DOE Contract No. DE-AC03-76ET11304, Final Report, April, 1987.

4. M. Farooque, et al., "Development of Internal Reforming Carbonate Fuel Cell Stack Technology", Final Report for the Period of February 1988 through September 1990, under DOE Contract No. DE-AC21-87MC23274.

5. M. Farooque, et al., "Simulated Coal-gas Fueled Carbonate Fuel Cell Power Plant System Verification", Final Report under Contract No. DE-AC21-90MC27168, March, 1995.

6. M. Farooque, et al., "Status of 120kW Direct Fuel Cell Product Improvement and Testing", Proceedings of the Fuel Cell '94 Contractors Review Meeting, DOE/METC-94/1010, August 1994.

5.6 M-C Power's Product Design and Improvement

CONTRACT INFORMATION

Cooperative Agreement	DE-FC21-95MC30133
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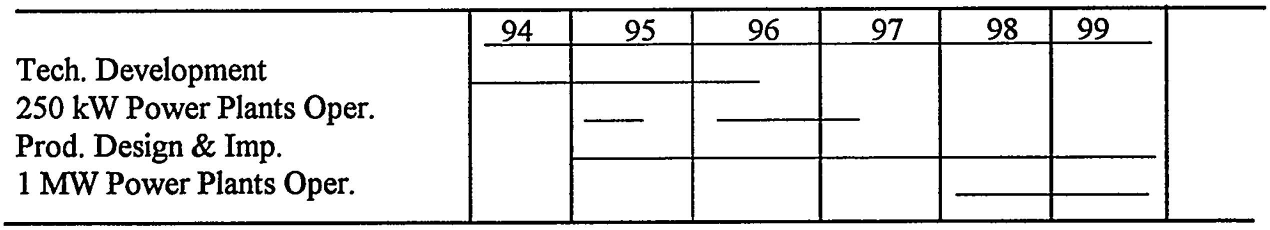
Principal Investigators	Rene M. Laurens Vincent J. Petraglia
METC Project Manager	Diane Hooie
Period of Performance	December 19, 1994 to December 18, 1999

Schedule and Milestones

Program Schedule



OBJECTIVE

The sole mission of M-C Power is the development and subsequent commercialization of molten carbonate fuel cell (MCFC) stacks. These MCFC stacks are based on the Internally Manifolded Heat EXchanger plate design developed by the Institute of Gas Technology. Integration of the MCFC stack into a commercially viable power plant is the mission of the IMHEX® team. The team is composed of leaders in the packaging and design of power generation equipment, including fuel cell technology, and includes Stewart & Stevenson, Bechtel, The Institute of Gas Technology and M-C Power.

In an effort to succeed in their respective missions, M-C Power and the IMHEX® team have developed a commercialization program. At the present time the team is making the transition from Phase I (Technology Development) to Phase II (Product Design & Improvement) of the program. Phase II's objective is a commercially viable (cost effective and technologically reliable) MCFC power plant ready for market by the turn of the century.

BACKGROUND INFORMATION

Introduction

Product Design & Improvement (PDI). The Product Design and Improvement activities began at the start of 1995 in parallel with the final steps of the Technology Development efforts. The major focus of the Phase II activities is to address cost reduction issues and to establish the commercial readiness of the power plant, stack technology, and marketplace infrastructure. The team's efforts will initially address these issues at the component level, followed by verification of advanced technologies in prototype power plant hardware.

The major barrier to successful fuel cell commercialization has been the higher first cost in comparison to conventional equipment. The molten carbonate fuel cell technology is no different, and therefore, the Team's goal must be to define and develop advanced stack and power plant technologies that allow the introduction of a cost-effective product even during the low production volumes of the initial few years.

M-C Power has defined a range of advanced technologies which have the potential to achieve this goal. These technologies address the two major cost reduction areas of the stack - separator plates and non-repeat hardware. Advanced separator plate designs reduce the number of components needed for the assembly from the fifteen pieces (used in Unocal) to two pieces.

Cost reduction of the non-repeat hardware will be achieved through creative engineering efforts which eliminate hardware and integrate functions. In parallel cost reduction in the repeat components are also being examined. These include faster manufacturing processes and reduced raw material through thinner components and reduced scrap.

Verification tests and power plant efforts are directed toward the construction and operation of a prototype, 1-MW power plant. This unit is intended to reflect all of the design and operational features of the Team's market entry product. The first 1-MW power plant is scheduled for operation during the early part of 1998. Southern California Edison will host this test at their Highgrove Generating Station in Grand Terrace, CA.

Market Entry Product

Design Description. Product design and definition activities completed in 1994 have provided the team with a blueprint for a 1MW market entry unit. The unit consists of three highly packaged skids which require minimum field assembly at the customers's site. Overall power plant design and layout is illustrated in figure 1 below.

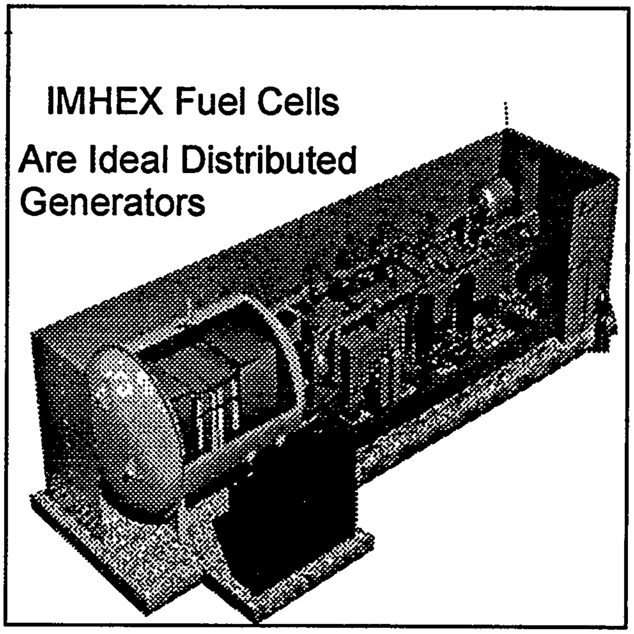


Figure 1. 1MW Market Entry Design

The fuel cell skid includes two 300-cell stacks along with two Ishikawajima-Harima Heavy Industries' plate reformers within a housing vessel.

The reformers convert natural gas into hydrogen and carbon monoxide as the stack fuel feed. Major components of the mechanical skid include; Heat Recovery Steam Generator (HRSG), a turbogenerator, two nitrogen bottles, two desulfurization bottles, six demineralizer bottles, and two boiler feed water pumps. The electrical skid contains a power conditioning unit and the system control unit. The power conditioning unit has an inverter to convert the stack DC power into AC power. The control unit is a simple industrial size PC based system geared toward an unattached operation of the fuel cell unit.

Each skid is sized within the height, length, and width limits for shipping. The unit is designed with maintenance in mind. Equipment requiring frequent servicing or replacement is placed at locations with easy access.

Operating Characteristics. Incorporated within the mechanical skid, the turbogenerator supplies compressed air as the oxidant feed to the stacks and generates additional power by expanding the hot flue gas from the cell reactions. The expanded gas from the turbogenerator flows to the HRSG where waste heat is recovered to perform the following steps: Preheat the compressed air prior to its feeding to the stacks; preheat the desulfurized natural gas; generate steam for the reforming reactions; and finally recover by-product heat for customer use. The HRSG is further equipped with an auxiliary burner, as well as the necessary burner control center to provide startup heat for the fuel cell unit.

A summary of the power plant characteristics is provided in Figure 2. The overall electrical efficiency is very high, 54.4% based upon HHV and 60% based on LHV. The current design is geared toward maximum power production. As a result, the HRSG flue gas is hot enough only for cogeneration of hot water. The overall efficiency including both power and hot water generation is 82% on HHV basis. The fuel cell unit is capable of turning down to approximately 30% load while still maintaining a reasonable electric efficiency close to 35%.

The Team estimates the capital cost of the market entry product will be in the $1500/kW range with production volumes of only 20 to 50 units per year. Achieving a cost-competitive pricing structure in low production volumes is believed to be critical to marketplace success as the power industry in theU.S. evolves with increasing competition.

Market Entry Product

Projected Characteristics - Jan. 1995

Rated Capacity ⇨	1 MW
Heat Rate ⇨	5680 Btu/kWh
Efficiency ⇨	60% LHV
Fuel Source ⇨	Natural Gas
Thermal Output	
250°F ⇨	430 kBtu/h
160°F ⇨	1430 kBtu/h
Foot Print	
Module ⇨	52x19x14ft
Fence ⇨	65x25ft
Emissions ⇨	<1 ppm NO_X

Figure 2. Market Entry Power Plant Characteristics

PROJECT DESCRIPTION

Product Development and Improvement

Planning. The PDI project has been structured into six tasks directed at successfully developing a commercially viable MCFC power plant. These tasks are: Product Definition and Market Assessment; System Design and Analysis;

Manufacturing Process Development; Packaging and Assembly Verification; Test Facility Design and Development; and Technology Development Improvement and Verification. The interrelationships between the program tasks are illustrated in Figure 3.

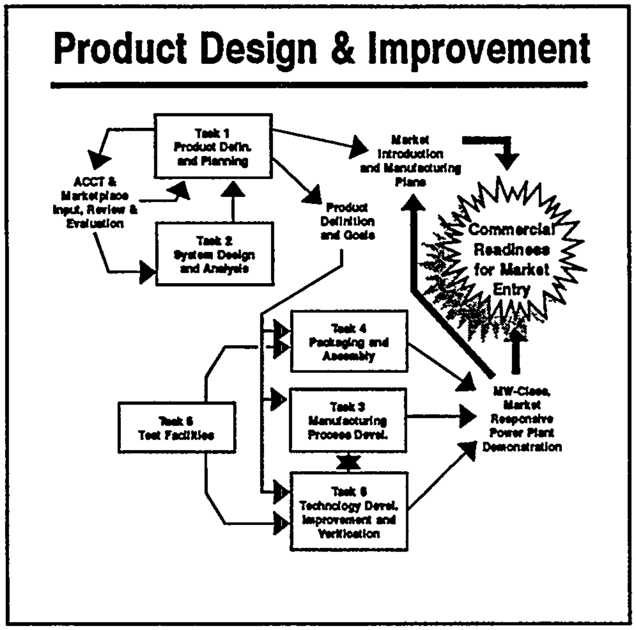


Figure 3. Product Design and Improvement Project Flow

Task 1. The product definition and market assessment involves the definition of a market driven power plant and the planning required to successfully introduce the power plant into the marketplace. Development of a market-driven product is enhanced by bringing the end users into early planning, product definition, and design phases. Preliminary marketplace inputs have emphasized the importance of reliability, durability, and cost. Although the initial introduction may require an elevated pricing structure in comparison to conventional technologies, reliability and durability of the power plant cannot be compromised.

Market surveys are being developed to identify the marketplace requirements for a MW-class MCFC power plant for distributed power generation, cogeneration, and compressor station applications. The surveys will address such issues as capacity, footprint, operating characteristics, interface requirements, and product trade-offs such as cost vs. efficiency. The results of the market surveys, in conjunction with applicable codes & regulations and design optimization from Task 2, will be analyzed to define preferred market entry power plant options.

The preferred power plant characteristics will be used to establish performance, life, and cost goals for the stack, mechanical skid equipment, and the power plant system. An ongoing technical progress evaluation are being maintained to measure development status compared to goals.

Task 2. System Design and Analysis will culminate in design and costing of a market-entry MW-class power plant. A preliminary baseline system has been selected against which trade-off studies will be performed. This baseline system expands upon design experience from Phase I (Technology Development) activities of the overall commercialization program.

Bechtel Corporation has developed design documents for the baseline system which include: a process flow diagram of the baseline design, complete with process flow characteristics (compositions, pressures, and temperatures); a description of the process; and a list of major equipment. Trade-off studies are being performed to determine the optimum operating parameters and process configuration for the market entry unit. Variables are independently changed to identify their impact on plant performance, reliability, and cost. Efficiency improvements will be weighed against increases in plant complexity, cost, size, reliability, and operating risk.

A design and cost estimate of the optimized plant has been generated. The cost estimates currently includes capital cost. Operating cost estimates and economic sensitivity analysis programs are being developed. The performance (design, off-design, and reliability, availability, maintainability, and safety) will be evaluated and dynamic simulations will be run to analyze the system behavior during normal and off-design conditions.

Task 3. The primary focus of the Manufacturing Process Development is to reduce the cost of the IMHEX® stack. This effort will build on the component and stack manufacturing capabilities developed at M-C Power since 1989, the year in which we established our first manufacturing facility dedicated to developing full area MCFC . This facility has been producing full area components and test stacks since 1991. In 1994 the largest MCFC stack was manufactured and conditioned in the facility and subsequently shipped to Unocal's Research Center in Brea, California. Currently, M-C Power is preparing to assemble the first of two 250-kW stacks to be tested at the Miramar Naval Air Station in San Diego, California. Progress in cost reduction from the initial 250kW stack (Unocal) to through the manufacturing of the two San Diego stacks is illustrated in Figure 4.

The optimization and automation of active cell component manufacturing process, consolidation of in-house separator plate manufacturing, and upgrade of QA/QC and analytical laboratory capabilities will enhance cell component manufacturing for the prototype power plant. The identification, qualification, and implementation of advanced component formulations and manufacturing processes will further reduce cell costs for market entry.

The manufacturing processes developed will be used to prepare a conceptual design for M-C Power's market entry stack manufacturing facility. The conceptual design will include site and facility layouts, staffing requirements, inventory and throughput analysis, and manufacturing cost analysis.

Stack's Major Cost Elements

Progress Being Demonstrated Today

- Separator Plate Assembly
    - 47% Reduction: Unocal to SDG&E-1
    - 74% Reduction: Unocal to SDG&E-2
- Non-Repeat Parts
    - 15% Reduction: Unocal to SDG&E-1
- Repeat Parts
    - 50% Reduction: Unocal to SDG&E-1
    - 73% Reduction: Unocal to SDG&E-2
- Performance Improvements
    - 120 W/ft2 Achieved in 20kW Stack/Lab Cells
- Life Improvements
    - 4mV/1000hr Achieved in 1ft2 Stack/Lab Cells

Figure 4. Reductions in Stack Costs

Task 4. Packaging and assembly techniques used for the IMHEX® power plant will affect cost, footprint, weight and heat loss from the plant. Stewart & Stevenson will build on their experience with packaging of the Unocal and San Diego Gas & Electric demonstration units, along with their proven expertise with packaged power generation systems, to develop optimized packaging and assembly techniques.

Definition, verification, and implementation of packaging concepts and assembly methodologies will reduce the size and cost of the fuel cell power module, mechanical skid, and electrical module through efficient integration and assembly. Packaging concepts will be confirmed by scale models. The packaging concepts will be translated into three-dimensional computer models for verification of the assembly methodology and will be

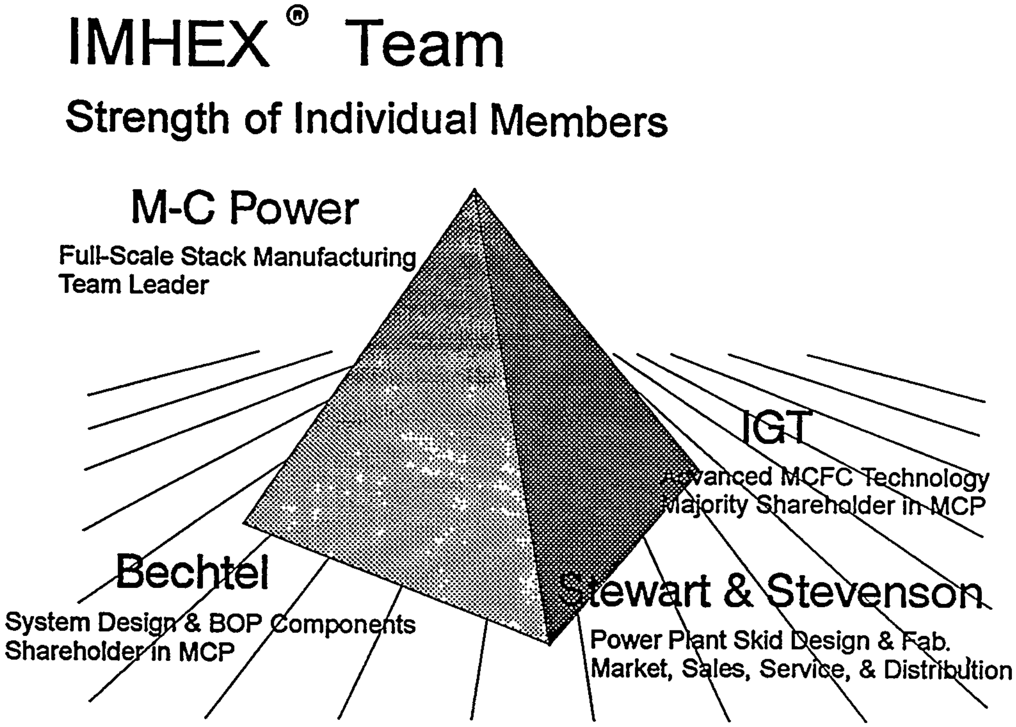


Figure 5. IMHEX® Team

Tasks 1,2, and 4 emphasize not only the fuel cell stack but the balance-of-plant components as well. Task 1 is critical for defining a market driven product and development of business plans designed to guide the team through commercialization. Task 2 is essential for development of a quality power plant. The trade off studies currently being conducted will provide confidence that the plant design will be responsive to the needs of the power generation industry. An essential element also because lessons learned from the fabrication and shipping of proof-of-concept power plants will be used to drive BOP cost down. This will be accomplished through the development of effect assembly techniques as well as analysis of logistical issues.

The IMHEX® Team is focused on accomplishing the tasks that lay ahead. Successful commercialization of the IMHEX® MCFC power plant requires that the team move decisively forward. The particular strengths of individual members of the team, summarized in Figure 5, alone could not accomplish the goals laid out, in the time frame required. The best way to insure success is in the form of our current alliance.

Appendices

Agenda

FUEL CELLS '95
REVIEW MEETING
August 9-10, 1995

WEDNESDAY, AUGUST 9, 1995

7:30 a.m.	REGISTRATION/BREAKFAST	
8:30 a.m.		*Welcome to METC* Mark C. Williams Morgantown Energy Technology Center

SESSION 1 -- OPENING SESSION

Session Chair: Mark C. Williams

8:35 a.m.	1.1	*DOE/METC Perspective* Rita A. Bajura Morgantown Energy Technology Center
8:50 a.m.	1.2	*An EPRI Perspective* — 1995 John B. O'Sullivan Electric Power Research Institute
9:10 a.m.	1.3	*Fuel Cells '95 GRI Perspective* James Kimball Gas Research Institute
9:30 a.m.	1.4	*DOE Perspective on Fuel Cells in Transportation* Robert Kost U.S. Department of Energy, Office of Transportation Technologies and Renewable Energy
9:50 a.m.	1.5	*DOD ARPA Perspective* Lawrence H. Dubois Advanced Research Projects Agency
10:10 a.m.	BREAK	

SESSION 2 -- SOLID OXIDE FUEL CELLS

Session Chair: Bruce C. Harrington

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	*Advanced Materials for Solid Oxide Fuel Cells* Timothy R. Armstrong Pacific Northwest Laboratory
12:00 p.m.	2.4	*Pressurized Solid Oxide Fuel Cell Testing* Joseph F. Pierre Westinghouse Electric Corporation
12:30 p.m.	LUNCH	

SESSION 3 -- DOE'S TRANSPORTATION PROGRAM

Session Chair: Robert Kost

1:30 p.m.	3.1	*Fuel Cell Transportation Overview* Robert Kost U.S. Department of Energy
1:45 p.m.	3.2	*General Motors Automotive Fuel Cell Program* Swathy Swathirajan General Motors Corporation
2:00 p.m.	3.3	*DOE/FORD Fuel Cell Contract for Automotive Application* Djong-Gie Oei Ford Motor Company
2:15 p.m.	3.4	*Chrysler Pentastar Direct Hydrogen Fuel Cell Program* Daniel H. Tudor Chrysler Corporation
2:30 p.m.	BREAK	

SESSION 4 -- ARPA ADVANCED FUEL CELL DEVELOPMENT

Session Chair: Lawrence Dubois

3:00 p.m.	4.1	*ARPA Advanced Fuel Cell Development* Lawrence Dubois Advanced Research Projects Agency
3:15 p.m.	4.2	*SOFCo Mobile Planar Solid Oxide Generator* Robert M. Privette Babock & Wilcox
3:30 p.m.	4.3	*Fuel Cell Technology for Prototype Logistic Fuel Cell Mobile Systems* Craig R. Schroll International Fuel Cells
3:45 p.m.	4.4	*The Direct Methanol Fuel Cell* Gerald Halpert Jet Propulsion Laboratory
4:00 p.m.	**POSTER SESSION**	
		Session Chair: William C. Smith
	P1	*Sealant Materials for Solid Oxide Fuel Cells* Timothy W. Kueper Argonne National Laboratory
	P2	*Solid Oxide Materials Research* Timothy R. Armstrong Pacific Northwest Laboratory
	P3	*Processing of $LaCrO_3$ for Solid Oxide Fuel Cell Applications* Wayne Huebner University of Missouri - Rolla
	P4	*Progress in Carbonate Fuel Cells* Michael F. Roche Argonne National Laboratory
	P5	*Carbonate Fuel Cell Matrix Strengthening* Chao-Yi Yuh Energy Research Corporation

P6 *Carbonate Fuel Cell System with Integrated Carbon Dioxide/Thermal Management*
Lawrence Paetsch
Energy Research Corporation

P7 *High Performance MCFC Using Li/Na Electrolyte*
L. G. Marianowski
Institute of Gas Technology

P8 *Technology Base Studies of Long-Term MCFC Performance*
J. Robert Selman
Illinois Institute of Technology

P9 *Landfill Gas Cleanup for Fuel Cells*
Douglas J. Herman
Electric Power Research Institute

P10 *High Efficiency MCFC Bottom Cycle*
Paul L. Micheli
Morgantown Energy Technology Center

P11 *Internet Public Information for Fuel Cells*
Frederick A. Sudhoff
Morgantown Energy Technology Center

P12 *Fuel Cell Cogeneration*
John G. Wimer
Morgantown Energy Technology Center

P13 *Trace Metal Behavior During Gasification*
Thomas A. Erickson
Energy & Environmental Research Center
University of North Dakota

P14 *Research and Basic Science in Electrochemical Technology*
Dick Kelley
U. S. Department of Energy

5:30 p.m. ADJOURN

THURSDAY, AUGUST 10, 1995

7:30 a.m. REGISTRATION/BREAKFAST

9:15 a.m. *Welcome Back to METC*
Edward L. Parsons
Morgantown Energy Technology Center

SESSION 5 -- MOLTEN CARBONATE FUEL CELLS

Session Chair: Diane Hooie

9:30 a.m. 5.1 *ERC Commercialization Activities*
Donald R. Glenn
Energy Research Corporation

10:00 a.m. 5.2 *IMHEX® Fuel Cells Progress Toward Commercialization*
Richard Woods
M-C Power Corporation

10:30 a.m. BREAK

11:00 a.m. 5.3 *ERC MCFC Product Development Test*
Anthony J. Leo
Fuel Cell Engineering Corporation

11:20 a.m. 5.4 *Molten Carbonate Fuel Cell Product Development Test at SDG&E*
Joseph A. Scroppo
M-C Power Corporation

11:40 a.m. 5.5 *ERC Product Improvement Activities for Direct Fuel Cell Power Plants*
Mohammad Farooque
Energy Research Corporation

12:00 p.m. 5.6 *M-C Power's Product Design and Improvement*
Lee Camara
M-C Power Corporation

12:30 p.m. LUNCH

1:30 p.m. PANEL: *Key Issues in Commercialization*
Mark C. Williams, (Moderator)
Morgantown Energy Technology Center

Elias Camera
M-C Power Corporation

William Lueckel
International Fuel Cells

Hans C. Maru
Energy Research Corporation

Emerson Ray
Westinghouse Electric Corporation

2:30 p.m. *Closing Remarks*
Edward L. Parsons
Morgantown Energy Technology Center

2:45 p.m. METC SITE TOUR

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