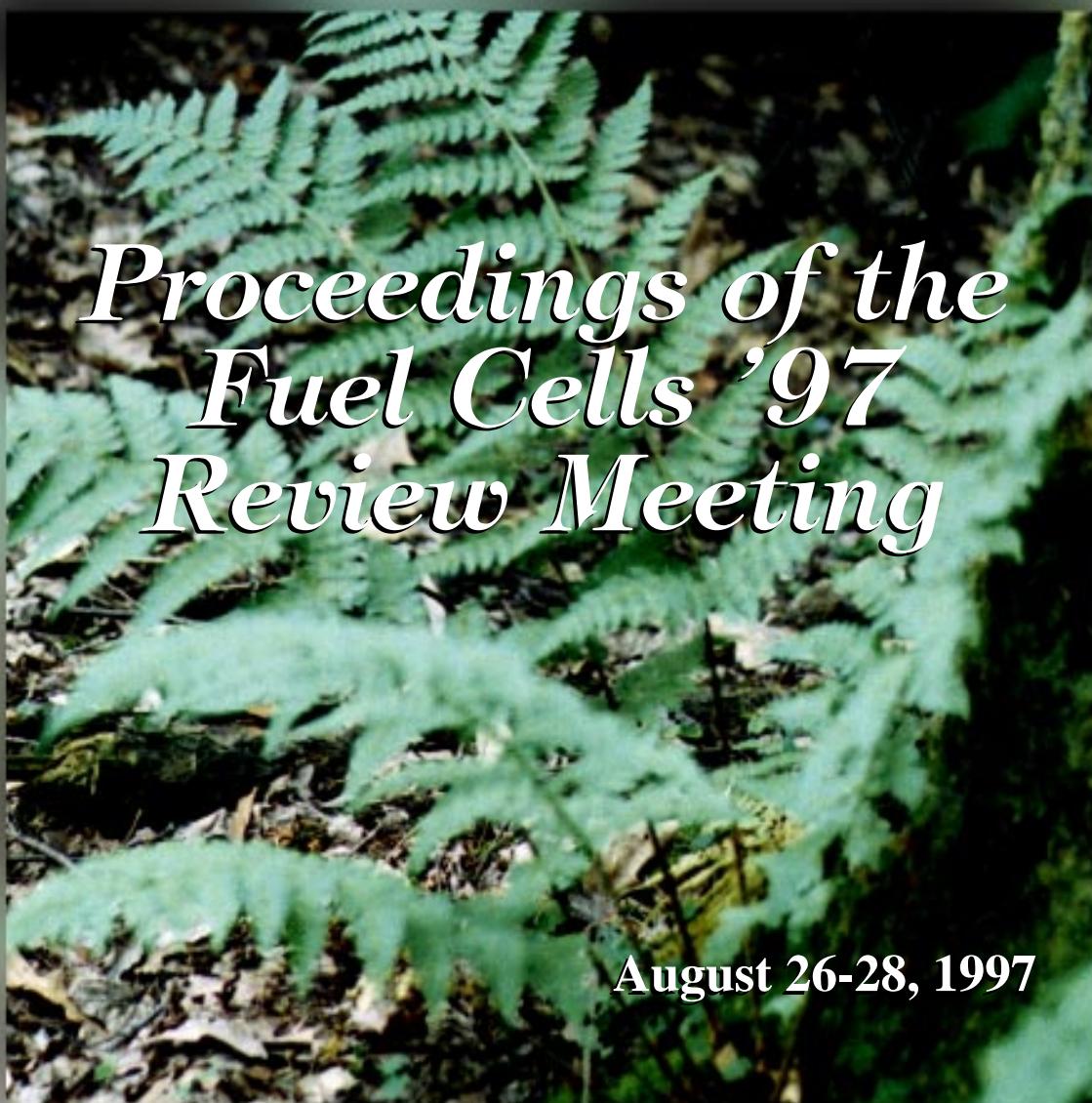


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# Contents

This primary contents page has been set up with each section (or session) title linked to the corresponding section/session. To use this feature, move your cursor to the section/session you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that session.

**Disclaimer**

**Foreword**

**Conference Participants**

**Papers and Presentations**

Session 1: Overview

Session 2: PAFC Program

Session 3: MCFC Program

Session 4: SOFC Program

Session 5: AR&TD Solid Oxide Fuel Cells I

Session 6: AR&TD Solid Oxide Fuel Cells II

Session 7: Other Fuel Cell Developments

Session 8: Advanced Research and Technology Development Workshop

**Fuel Cell Fact Sheets**

To return to the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

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To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## **Foreword**

The Federal Energy Technology Center (FETC) sponsored the Fuel Cells '97 Review Meeting on August 26-28, 1997, in Morgantown, West Virginia. The purpose of the meeting was to provide an annual forum for the exchange of ideas and discussion of results and plans related to the research on fuel cell power systems. The total of almost 250 conference participants included engineers and scientists representing utilities, academia, and government from the U.S. and eleven other countries: Canada, China, India, Iran, Italy, Japan, Korea, Netherlands, Russia, Taiwan, and the United Kingdom.

On first day, the conference covered the perspectives of sponsors and end users, and the progress reports of fuel-cell developers. Papers covered phosphoric, carbonate, and solid oxide fuel cells for stationary power applications. On the second day, the conference covered advanced research in solid oxide and other fuel cell developments. On the third day, the conference sponsored a workshop on advanced research and technology development. A panel presentation was given on fuel cell opportunities. Breakout sessions with group discussions followed this with fuel cell developers, gas turbine vendors, and consultants.

Papers and presentations in this document were produced from electronic files provided by the authors. They have been neither refereed nor extensively edited. The meeting Proceedings was provided to participants on CD. The Proceedings is available as paper copy or microfiche from the Office of Scientific and Technical Information, or as microfiche or blowback from microfiche from the National Technical Information Service. The Proceedings is also available worldwide on the FETC Homepage over the Internet (URL: <http://www.fetc.doe.gov>).

### **Conference Technical Coordinator**

William Cary Smith, for Diane T. Hooie

### **Workshop Coordinator**

Douglas F. Gyorke

## **Acknowledgment**

As Conference and Workshop Coordinators, we could not have operated without the complete support of many people. We especially want to acknowledge Betty Robey, Carolyn Moore, Pam Stanley, and Beckie Pestun for their conference services support; Mary Micciche and Marlene Williams for audiovisual and photography support; the entire Graphics staff and Duplicating staff; and Kate Lessing for assistance in compiling the Proceedings.

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 1 — Overview

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 1.1** *FETC Perspective on the DOE Stationary Power Fuel Cell Program:* Rita A. Bajura — Federal Energy Technology Center
- 1.2** *EPRI Perspective, Fuel Cells in Evolving Energy Markets:* Daniel M. Rastler — Electric Power Research Institute
- 1.3** *GRI Fuel Cell Perspective:* Kevin Krist — Gas Research Institute
- 1.4** *DOE/EE Transportation Perspective:* Steven G. Chalk — U.S. Department of Energy; and James F. Miller — Argonne National Laboratory

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 2 — PAFC Program

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 2.1** *Fuel Cells: DOD-Unique Applications:* Ronald Patun and Heather Moyer — Concurrent Technologies Corporation
- 2.2** *DOD Fuel Cell Demonstration Program:* Michael J. Binder, William R. Taylor, and Franklin H. Holcomb — U.S. Army Construction Engineering Research Laboratories
- 2.3** *ONSI PC25<sup>TM</sup> Fuel Cell Power Plant Experience/Status/Progress:* Joseph M. King, Jr. — International Fuel Cells Corporation

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 3 — MCFC Program

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 3.1** *ERC Direct Carbonate Fuel Cell Program Overview:* H. Maru, M. Farooque, G. Carlson, P. Patel, and C. Yuh — Energy Research Corporation; C. Bentley — Fuel Cell Manufacturing Corporation; and D. Glenn and A. Kush — Fuel Cell Engineering Corporation
- 3.2** *Santa Clara Direct Carbonate Fuel Cell Demonstration:* Anthony J. Leo and Andrew J. Skok — Fuel Cell Engineering Corporation; and Thomas P. O'Shea — Santa Clara Demonstration Project
- 3.3** *M-C Power Commercialization Program Overview:* Elias H. Camara — M-C Power Corporation
- 3.4** *NAS Miramar Molten Carbonate Fuel Cell Demonstration Status:* Joseph A. Scroppi and René M. Laurens — M-C Power Corporation; and R.A. Figueroa — San Diego Gas & Electric

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 4 — SOFC Program

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 4.1** *Westinghouse Program Overview:* R. George — Westinghouse Science & Technology Center
- 4.2** *Westinghouse SOFC Field Unit Status:* S. Veyo — Westinghouse Science & Technology Center
- 4.3** *SOFC System Analysis:* Tan-Ping Chen — Bechtel Corporation; John D. Wright — TDA Research, Inc.; and Kevin Krist — Gas Research Institute

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 5 — AR&TD Solid Oxide Fuel Cells I

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 5.1** *Thin-Film Fuel Cells:* Steven J. Visco, Craig Jacobson, and Lutgard C. De Jonghe — Lawrence Berkeley National Laboratory
- 5.2** *Advanced Materials for Solid Oxide Fuel Cells: Mechanical Properties of Lanthanum Chromite:* Timothy R. Armstrong, Suresh Baskaran, and Steven W. Pulik — Pacific Northwest National Laboratory
- 5.3** *Micro-Engineered Cathode Interface Studies:* Rajiv Doshi and Michael Krumpelt — Argonne National Laboratory
- 5.4** *Composite Ceria Electrolytes:* Scott L. Swartz and William J. Dawson — NexTech Materials, Ltd.
- 5.5** *Lanthanum Gallate as a New SOFC Electrolyte:* John B. Goodenough and Kequin Huang — University of Texas at Austin

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 6 — AR&TD Solid Oxide Fuel Cells II

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 6.1** *Solid Oxide Fuel Cell Performance Studies: Anode Development:* Wayne Huebner and Harlan U. Anderson — University of Missouri-Rolla
- 6.2** *Planar SOFC Integrated System Technology Development:* S. Elangovan, J. Hartvigsen, A. Khandkar, R. Privette, K. Kneidel, M.A. Perna, and M. Tharp — SOFCo
- 6.3** *Tape-Calendered SOFC Stack Development:* Nguyen Q. Minh, Brandon Chung, and Kurt Montgomery — AlliedSignal Aerospace Equipment Systems
- 6.4** *Solid Oxide Fuel Cells (SOFCs) for the Direct Oxidation of Methane:* Raymond J. Gorte, John M. Vohs, and Wayne L. Worrell — University of Pennsylvania
- 6.5** *Low Temperature, High Performance, Planar Solid Oxide Fuel Cells and Stacks:* Anil V. Virkar, Jai-Woh Kim, and Karun Mehta — University of Utah; and Kuan-Zong Fung — Materials and Systems Research, Inc.
- 6.6** *Application of Ceria Layers to Increase Low-Temperature SOFC Power Density:* Scott Barnett, Erica Perry, and David Kaufman — Northwestern University

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 7 — Other Fuel Cell Developments

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 7.1** *SOFC Stack Performance Goals:* John D. Wright — TDA Research Inc.
- 7.2** *Evaluation of Small-Particle Plasma Spray for SOFC Electrolyte Deposition:* Daniel E. Boss, Thomas Bernecki, David Kaufman, and Scott Barnett — Northwestern University
- 7.3** *Non-Segregating Electrolytes for Molten Carbonate Fuel Cells:* T.D. Kaun, I.D. Bloom, and M. Krumpelt — Argonne National Laboratory
- 7.5** *200 kW B Anaerobic Digester System:* Yan Kishinevsky — New York Power Authority
- 7.6** *An Integrated Fuel Processor for PEM Fuel Cells:* David J. Edlund and William A. Pledger — Northwest Power Systems, LLC
- 7.7** *Researches and Development of a Fuel Cell With Protonic Conductivity:* Yuri N. Kleschev — Russian Federal Nuclear Center; Valery P. Gorelov — Institute of High-temperature Electrochemistry; and Vladislav V. Sagalovich — Scientific-Technological Enterprise “STIN”

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Session 8 — Advanced Research and Technology Development Workshop

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

- 8.1** *Results of FETC Pre-Workshop Survey:* Douglas F. Gyorke — Federal Energy Technology Center
- 8.2** *Panel Discussion: Fuel Cell Opportunities*
  - 8.2a** *EPRI Assessment of Fuel Cell R&D Needs:* Daniel M. Rastler — Electric Power Research Institute
  - 8.2b** *GRI Basic Solid Oxide Fuel Cell Research:* Kevin Krist — Gas Research Institute
  - 8.2c** *The DOE Fuel-Cell AR&TD Program:* Mark C. Williams — Federal Energy Technology Center
  - 8.2d** *Fuel Cell Opportunities in the Division of Materials Sciences, Office of Basic Energy Sciences, U.S. Department of Energy:* Richard D. Kelley — U.S. Department of Energy
  - 8.2e** *DARPA Advanced Energy Technologies:* Robert Nowak, U.S. Department of Defense
  - 8.2f** *U.S. Department of Agriculture Small Business Innovation Research Program:* Charles F. Cleland and Ruth Lange — U.S. Department of Agriculture
- 8.3** *Identification of Breakout Topics, and Initiation of Breakout Group Discussions*

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

## Fuel Cell Fact Sheets

This session contents page has been set up with each presentation title linked to the corresponding presentation. To use this feature, move your cursor to the presentation you wish to view. (Your cursor arrow will change to a pointing finger.) Click your left mouse button to jump to the beginning of that presentation.

*Climate Change Fuel Cell Program Fact Sheet:* Mark Williams and Diane Hooie — Federal Energy Technology Center

*Developing the Second-Generation Fuel Cell, The Energy Research Project Fact Sheet:* Bernard Baker — Energy Research Corporation; and Mark Williams — Federal Energy Technology Center

*Developing the Second-Generation Fuel Cell, The M-C Power Project Fact Sheet:* Elias Camara — M-C Power Corporation; and Mark Williams — Federal Energy Technology Center

*Developing the Solid Oxide Fuel Cell Fact Sheet:* Stephen Veyo — Westinghouse Electric Corporation; and Mark Williams — Federal Energy Technology Center

*Solid Oxide Fuel Cell Project Fact Sheet:* Stephen Veyo — Westinghouse Electric Corporation; and Mark Williams — Federal Energy Technology Center

*Phosphoric Acid Fuel Cell Commercialization Fact Sheet:* Frederick L. Whitaker — ONSI Corporation; and Mark C. Williams — Federal Energy Technology Center

To return to the primary *Contents* page or the *Cover*, click the **Go Back** button (the double left-pointing arrow) from the tool bar at the top of the screen, or choose **Go Back** from the view menu.

# FETC Perspective on the DOE Stationary Power Fuel Cell Program

**Rita A. Bajura**  
Director, Federal Energy Technology Center  
U.S. Department of Energy

Good Morning, welcome to the *Fuel Cells '97 Review Meeting*.

Let me start with a few words about the Federal Energy Technology Center — the FETC. As I am sure you know, last December, DOE merged the Morgantown and Pittsburgh Energy Technology Centers into the FETC. This merger was the result of a nationwide trend, in the words of President Clinton, for a *smaller, humbler Government*.

We have responded. Our staff is down 15 percent over the past two years through attrition. We operate the FETC as if our two sites were co-located — thanks goodness for E-mail. A common management team serves both sites. Our programs typically span both sites. No more lead centers.

So you will start to see some fuel cell work in Pittsburgh office of the FETC. I'd like to introduce the FETC management team deal with fuel cells:

- Ralph Carabetta, my Deputy.
- Joe Strakey, heads the Office of Power Systems Product Management.
- Mark Williams is the Fuel Cell Product Managers in that office.
- Ken Markel heads the Office of Project Management.
- Chuck Zeh is Director of the Gas Power System Division.

Fuel cells! DOE/FE is responsible for the fuel cell program for stationary power. Our sister agency, DOE/Energy Efficiency & Renewable Energy, is responsible for fuel cells for transportation. This morning, I want to talk about:

- The status of DOE/FE's fuel cell program.
- Five trends and their impact on the future of the program

## Status of Fossil Energy's Fuel Cell Program

Let me begin with the first *real* fuel cell success story — PAFCs!

From 1976 to 1992, the DOE and its predecessors invested \$290 million to develop PAFCs. The Gas Research Institute, EPRI, and gas and electric utilities also invested in the development effort. ONSI Corporation, a subsidiary of International Fuel Cells (IFC), emerged

as the leading PAFC technology vendor. Currently, ONSI is actively marketing PAFC systems. More than 100 of their 200-kW systems are operating in countries around the world. Reliability has been excellent. Some have operated for more than 40,000 hours and are still going strong! Reducing the capital cost is the major issue for PAFC systems — as it is for *all* fuel cell systems. The ONSI unit sells for \$3,000/kW; the goal is to reduce the cost to \$1500/kW.

In 1996, FETC awarded grants for 42 of the 200-kW units. The Department of Defense provided this funding as part of the Administration's climate-change program. Here, the goal was to reduce CO<sub>2</sub> emissions by accelerating the commercialization of PAFCs. This year, DOD's Picatinny Arsenal is managing the buy-down program. DOD intents to award grants for 50 to 70 units before the end of this calendar year.

DOE/FE is now out of the PAFC business. We focus on advanced fuel cells — MCFCs and SOFCs. These operate at higher temperatures than PAFC systems, have higher fuel-to-electric efficiencies, potentially lower capital costs, and can be integrated with coal gasifiers. DOE/FE's total FY97 funding for the MCFC and SOFC programs is \$50 million. And we leverage this with an additional 40-percent cost-sharing from the private sector.

We are funding two major developers of MCFCs:

- Energy Research Corporation of Danbury, CT, and
- M-C Power Corporation of Burr Ridge, IL.

In FY97, we funded each of these developers at the \$15 to 16 million level. Both of the developers conducted a major demonstration in 1996-97.

- ERC demonstrated a 2-MW unit at Santa Clara, California. This was the world's largest MCFC demonstration. It was also the largest, *fuel cell* power plant to operate in the U.S.
- M-C Power demonstrated a 250-kW demonstration at the Miramar Naval Air Station, near San Diego, California.

You will hear more about these two demonstration projects during this meeting. *It is no secret* that the original objectives for the two demonstrations were not *all* achieved. Fortunately, the problems encountered were not inherent to the technology. Rather, they were a normal part of the technology maturation process. Both MC-Power and ERC plan an additional demonstration project as part of the DOE program. They are still assessing sites.

FETC also funds SOFC technology. We currently support one major SOFC developer — Westinghouse — who, we feel, is the world leader in the SOFC technology. We are funding Westinghouse at the \$12 million/year level in FY97.

Westinghouse made significant progress in reducing manufacturing costs of their tubular cells. They eliminated two electrochemical vapor deposition steps. Currently, Westinghouse is preparing for a **100-kW demonstration test** in the Netherlands, in January 1998. This will be the largest SOFC system tested to date. They are also planning (1) a 250-kW test at Southern

California Edison, (2) a 1-MW test at Ontario hydro, and (3) a 3-MW, demo at Fort Meade, MD sponsored by the EPA. This will be a coupled turbine/fuel cell system.

## **Five Trends and Impact on the Future of the Fuel Cell Program**

I want to talk about five policy issues and then give some of my thoughts on how these issues might impact the fuel cell program.

### **Issue 1: Global Warming — *Greenhouse Gas Emissions*.**

Let me put this into context: CO<sub>2</sub> concentration levels in the atmosphere have increased by 25 percent over the past 100 years. That is a fact. We are still debating — does this influence our climate? If it does, what will change? Will the changes result in a better or worse climate? What is the rate of change?

2,500 experts from more than 80 countries studied climate change as part of the United Nation's Intergovernmental Panel on Climate Change (IPCC). There is some disagreement over the final wording but their IPCC report stated:

*The balance of evidence suggests a discernible human influence on global climate.*

Where do we go from here? Developing countries do not want to limit their CO<sub>2</sub> *until* their standard of living catches up with developed countries.

But many developed countries want binding limits on CO<sub>2</sub> emissions. Binding limits are not that onerous for some developed countries.

- England — when they privatized their coal industry, they switched from high-cost, thin-seam coal to lower-cost North Sea gas;
- France — which is heavily nuclear;
- Germany — whose baseline in 1990 included the terribly inefficient East German plants that are already being upgraded;
- Japan — a compact nation with small cars and short commutes, and also high energy-efficiency because of long-standing, high energy prices.

Thus, the U.S. is under considerable pressure to commit to binding limits. The Administration has stated they want binding limits that are “flexible, cost-effective, realistic, achievable, and ultimately global in scope.” They are currently negotiating binding limits with 130 other nations. The Administration plans to sign a greenhouse gas reduction treaty during a climate change meeting in Kyoto, Japan this December.

Meanwhile:

- Econometric models by Argonne and others show that binding limits would significantly reduce output and employment in six industries: Aluminum, Cement, Chemical, Steel, Petroleum Refining, and Paper and Pulp.
- Many, but not all, industry groups complain that binding limits will hurt the U.S. economy. They want more public debate on climate change. They want to see the specific details on the plan that the Administration will sign at Kyoto.
- The U.S. Senate voted 95 to 0 that they wanted **both** developing and developed nations to be covered in any climate change treaty negotiated by the Administration.

The concern over greenhouse gases is a threat for the fuel cell program. **Any technology that uses a carbon-based fuel is suspect!** And natural gas is a carbon based fuel.

But climate change concern is also an opportunity! In July, President Clinton announced a new initiative: **A Climate Change Technology Strategy**. He asked DOE to help plan this initiative, an initiative to seek low-cost technologies to address climate change. As we speak, DOE's laboratories — including FETC — are planning this initiative. The initiative will:

- Define roadmaps for addressing global warming.
- Identify technology targets.
- Scope out the needed R&D.
- Form industry-university-laboratory partnerships to carry out the R&D Program.

The scope of the initiative includes:

- Clean power generation — everything is on the table, including nuclear.
- Energy efficiency in the transportation, building, and manufacturing sectors.
- Long-range solutions for beyond 2025.

The planning schedule for this initiative is aggressive. DOE's goal is to deliver the completed plans for the initiative to the White House by October 1. We plan to start work on the Climate Change Initiative in FY98! Initially, the program could involve **only repackaging** of our current R&D activity. But depending on the level of Congressional and public interest, this **could evolve** into a major, new program!

I see this initiative as an opportunity for DOE and industry and academia to partner (1) to reduce the cost of fuel cells, and (2) to develop the ultra high-efficiency fuel-cell systems. Hybrid systems that couple gas turbines and fuel cells can slash CO<sub>2</sub> emissions in half compared with current, baseline systems. We can reduce CO<sub>2</sub> emissions even more by coupling high-efficiency power generation systems (1) with more efficient, end-use technologies; and (2) more efficient motors and buildings, the kind of technology DOE's Office of Energy Efficiency is developing.

## Issue 2: Deregulation and convergence

Deregulation of the energy industry, both domestically and internationally, is forcing all segments of the energy industry to reduce. In response to this pressure to reduce costs, gas, electric, and oil companies are merging to become “energy companies.”

Energy companies will build “energy plexes” — flexible plants that

- Consume gas or coal or waste material, and
- Produce a slate of products: electricity, liquid fuels, chemicals.

You are seeing this concept in Italy where Texaco is building three IGCC plants that will use a waste — refinery bottoms — to produce electricity and other chemicals.

DOE calls this concept Vision 21 — the plant for the 21st century. Vision 21 has a flexible design. It is tailored to meet the economics of a specific site. Vision 21 plants will have fuel-to-electric efficiencies higher than 60 percent. To achieve this efficiency level, the plant design includes a turbine/fuel cell combination.

In addition, Vision 21 plants will have no net CO<sub>2</sub> emissions. **No net CO<sub>2</sub> emissions!** In Vision 21, we are exploring CO<sub>2</sub> removal and sequestration. At a minimum, we need to develop information on the real cost and performance implications of removing and sequestering CO<sub>2</sub>. This is a long-term effort. Possible sequestration techniques include:

- Reforestation projects.
- Deep ocean disposal in clathrate hydrates.
- Geologic disposal in depleted gas and oil reservoirs.
- Disposal in deep, un-minable coal seams.

I need to insert a touch of realism in this discussion on deregulation. Fuel cells have serious competition from competing technologies! Gas-fired reciprocating engines in the 50-kW to 5 MW size cost \$200 to \$350/kW. Lower-heating-value efficiencies are 35 percent. Thanks to work by GRI and others, NO<sub>x</sub> levels are not that bad.

The message is that fuel cell costs must come down before fuel cells will be competitive in anything other than niche markets.

## Issue 3: New R&D approaches ★ A new model for structuring R&D programs

Over the past few years, Congress and the Administration have had diametrically opposed views of R&D: **Congress** held that “basic research is good, and applied research is bad — it’s corporate welfare.” The **Administration** championed “technology partnerships with industry” as the cornerstone of economic development.

To solve this impasse, Congressional leaders are attempting to reach a consensus on a new R&D model — a model that would blur the border between basic and applied research. The

emerging model is based on partnerships among government, universities, and industry. The view is partnerships leverage the Government and the private-sector's investment in R&D. Industry involvement ensures the relevance of the R&D. The model calls for Consortia to:

- Focus on a defined problem — for example, develop a 60 mpg car.
- Cut across an industry — for example, all fuel cell manufacturers.

Industry produces a roadmap of technology needs and it may share ideas. This model suggests, but doesn't demand, that the research be pre-competitive. There is a whole range of potential opportunities for fuel cell consortia in pre-competitive R&D:

- Advanced manufacturing processes
- Advanced materials

I recognize that all three major developers in the DOE/FE program have an advisory group or a commercialization consortia that support their respective programs.

- ERC has the Fuel Cell Commercialization Group (FCCG).
- MC-Power has the Alliance to Commercialize Carbonate Technology (ACCT).
- Westinghouse has a 100-member fuel-cell advisory group.

Currently, EPRI is forming a new focused advisory group for Westinghouse. But the new R&D model I described may become a requirement. I invite your comments on how we can make it work if it *does* become a requirement.

## **Issue 4 is Accountability**

The Government Performance and Results Act, or GPRA, passed in 1993. In it, Congress demanded that all Government programs contribute to measurable, desirable outcomes. The outcomes must benefit the public — and be something that the private sector cannot or will not do on its own. R&D programs are not exempt: they must produce measurable outcomes, for example, tons per year of pollutant reduced. These measures are used as part of the budget formulation process. The message for this audience is that, from a budget perspective, it is critically important that we hold to the schedule for fuel cell demonstrations and commercialization.

Starting in the FY99 budget cycle, DOE's budget requests to Congress will be based on amount of public good achieved. In this case, the public good is the potential for future emission reduction.

## **Issue 5 is budget pressure**

The requirement to balance the budget is producing tremendous pressure on Congress to reduce discretionary funding, like R&D programs. As most of you know, the House FY98 appropriations bill told DOE to "down select" from three fuel-cell vendors to two. The Senate language was silent on "down select," and it contained enough funding that we should

be able to continue with all three vendors. The issue will be resolved by the conference committee after Labor Day. We are looking at various scenarios, in the event the House version prevails, none of them pleasant. I hope we will not be forced into a down selection, particularly since a down selection is likely to drive the technology abroad, with the U.S. losing much of its investment.

I think it is important to have multiple vendors in the fuel cell program, and maintain the competition to reduce the cost of the commercial fuel cell product.

I conclude with a request: that *you enter into the public debate on global warming*. Energy use has a major impact on the U.S. economy! Energy is a \$560 billion per year business in the U.S. It accounts for 8 percent of our Gross Domestic Product.

Fossil fuels supply 85 percent of the energy we consume in the United States. And they ***will continue*** to be the dominant energy source in the U.S. for the foreseeable future.

DOE projects that fossil fuels will supply 88 percent by 2015. In the U.S., the use of fossil fuels has made our energy cheap — this has enhanced our quality of life — I like my dishwasher and I like my Chevy Blazer.

And it's fossil fuels that have made our economy competitive in the world market. This is our economic imperative — an abundant supply of low-cost energy. But we also have an environmental imperative — I want my children and grandchildren to live in a world that is non despoiled.

The U.S. needs to reconcile our economic and environmental imperatives. It's up to us in the scientific and technical community to weigh in on how best this might be done — or we will be told how it will be done.

Thank you.

# EPRI Perspective

## Fuel Cells in Evolving Energy Markets

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### Perspectives on 1996 Technology Progress

During the past year, fuel cell technology for stationary power applications continued to advance in both commercial applications; first of a kind field demonstrations; and laboratory scale breakthroughs. PAFC systems are carving out a real niche as premium power and grid independent power systems for commercial sector markets. Field units continue to illustrate fuel cell systems can have superior reliability and availability characteristics. While costs are still about a factor of two higher than what more main-stream markets will bear - PAFC applications & adoption process clearly point the way in demonstrating how fuel cells will play out in the evolving energy services markets.

Multi-million dollar carbonate fuel cell field tests were completed the past year. A 2 MW system developed by ERC successfully operated for 4090 hours sending power to the Santa Clara electric grid. A 250 kW pilot system supplied by M-C Power also completed demonstration tests with over 1500 MWhs supplied to the Miramar Naval Air Station. Both these demonstrations are enormous steps forward in developing an understanding of how carbonate systems need to be designed and how carbonate stacks need to be configured both electrically and process wise. While these demonstrations provided valuable information for the developers, they fell short, however, in providing the necessary "technology comfort" for many of EPRI's electric and gas utility clients. More specifically, the demonstrations did not confirm if large carbonate stacks can be effectively cycled and provide load following. They also did not answer important buyer questions on durability; performance, life and information regarding costs and impacts of long term operation on maintenance and operations. As both carbonate fuel cell demonstrations fell short of their intended operating test plans, concerns need to be raised regarding the robustness of the technology that was put in the field; the relative timing of these field tests; the need for continued RD&D "in-house"; and eventual commercialization plans. As will be discussed later, fuel cell technology is trying to be introduced into an rapidly changing electric power industry, where it will be very difficult for utilities to be the early adopters and initial buyers of potentially high risk technology. New and creative commercialization approaches will be needed.

Solid oxide fuel cell systems are advancing both in size and in integrated systems coupled with small gas turbines. Westinghouse has fabricated and is testing a 100 kW system; ZTEK has fabricated and is testing a 25 kW planar system. Both are planning to enter the market with ultra high ( 60% ) SOFC-gas turbine systems. Innovations in the laboratory have yielded breakthrough power densities. EPRI sponsored work at the University of Utah developed a planar solid oxide fuel cell with a power density 5-6 times higher than the current state of the art.

Historically, PEM fuel cells did not fit into any EPRI advanced power generation systems. However, both the industry and the technology has changed and we believe there are several strategic fits for PEM fuel cells. Polymer electrolyte membrane fuel cells continue to make technological advances in both increased power density; weight reduction; and cost reduction. As such, we are starting to look very closely at PEM systems for stationary power markets and applications. Their operating and power characteristics may make PEM systems potentially very attractive in new energy services. The past year has also shown that fuel processing systems, once a critical enabling technology for PEMFCs, are also advancing. Examples of this are the work at ADL and Northwest Power. Advances in PEM technology are also leading us to re-examine the role of PEM electrolyzers and regenerative PEM systems in energy market applications. If critical technical issues can be resolved this may open up complete new markets for PEM fuel cells in stationary power and transportation markets.

## **Role for Fuel Cells in Evolving Electric Utility Industry**

Electric industry restructuring is radically evolving to a new paradigm where customers will be able to purchase electricity as a commodity. New energy service providers will emerge to “package” and offer a wide variety of energy service products to customers. Customers will have a landmark opportunity to choose their own energy providers. These structural changes, deregulation and the resulting competitive environment will significantly impact commercialization strategies and markets for fuel cell technology.

While electric and gas utilities have historically pioneered the introduction of new technology, the associated risks of using “emerging technologies” were moderated by monopoly status and allowance for rate-based costs to be passed on to customers. New regulatory changes favor enhanced competition, lowest-cost providers, and differentiated services that deliver value to customers beyond traditional kilowatts. Therefore, utilities will not be able to provide the “market pull” support in the way previously envisioned for commercialization of fuel cells. Competing technologies continue to place pressure on the performance and cost requirements for fuel cell products. Compact aeroderivative turbines and proven heavy-frame (industrial) gas turbines are commercially available in 1 to 100 MW sizes and can be deployed on a one-year schedule; several micro-turbine generator systems are also entering the market for on-site distributed power generation applications. Their initial low cost ( \$ < 500/kW) and attractive operating characteristics are receiving the attention of many of our utility clients.

We do not believe fuel cells will be competitive with bulk power generation resources for quite some time, if ever. That is, it will be very difficult for fuel cell systems to compete in a deregulated bulk power commodity market. Fuel cells will most likely find applications in a variety of distributed generation/energy services applications.

The most likely adopters of fuel cells will be non-regulated Energy Service Companies (ESCOs) offering packaged bulk power and valued added services to customers. ESCOs will favor fuel cell products in the 10 kW to 5,000 kW sizes. GENCOs and Mini-Gencos may adopt larger fuel cell products (e.g. 1 to 20 MW SOFC-CT and MCFC systems) if they can compete with bulk power or offer other quantifiable values to end use customers. Fuel cell produced

power will be used to augment bulk power most-likely through a premium service or cogeneration driven customer need. Near term applications could be in industrial and commercial cogeneration sites where electric and gas rates are high; in certain commercial sectors where high reliability and premium service will be valued high enough to off-set the higher cost of fuel cell power. Early adopter markets are most likely to be in the commercial sector which could include hotels; data processing centers; grocery stores; health care and educational facilities. Eventual mass markets could be served by either PEM or SOFC systems designed for loads around 2 to 10 kW. However, significant technical and cost reduction challenges exist for fuel cells in this market.

The electric utility industry is entering into a competitive era in which the business environment requires investments in new technology to provide an immediate as well as long term return. Electric utilities seek to deploy new technologies that respond to customer needs and enhance their ability to compete on a domestic and global basis. Several of our member utility clients have recently made significant investments in fuel cell technology ventures.

### **Reinventing EPRI's Fuel Cell & Distributed Generation Program**

Industry restructuring, the convergence of electric, gas and telecommunications are requiring EPRI to re-invent the collaborative R&D business model upon which the Institute was founded 25 years ago. During the past three years we have unbundled our R&D portfolio which now enables utilities to choose among 100 research targets and to customize their participation in EPRI R&D. We have also recently established two taxable subsidiaries epriCSG and epriGEN to enhance the competitive value of R&D investments by our members and to provide increased flexibility with respect to intellectual property protection, publication and access of information.

The new corporate vision for 1998 is to "Provide science and technology based solutions of indispensable value to our global energy customers". We are in the process of changing our organization from a structure designed to serve an integrated utility to one that will provide solutions to numerous individual participants in the electric and energy enterprise including: power marketers; generating companies; transmission and distribution companies; system operators; gas and oil companies; energy service and retail service companies; and telecommunications companies.

Fuel Cell R&D at EPRI has undergone dramatic changes in program emphasis and direction. All future long term fuel cell R&D and basic science & technology will continue to be funded by our strategic R&D program. While the development and commercialization of new fuel cell products for retail and small distributed power markets will be very much market driven and conducted in our new epriGEN taxable subsidiary target. Our fuel cell program has changed the past few years from a "technology push" R&D strategy to more of a market and "customer driven" R&D focused program.

Our program areas for 1998 include:

- Evaluate ONSI 200 kW PAFC systems in distributed generation applications

- Facilitate the commercialization and early introduction of MCFC systems
- Provide intelligence and assessments on vendor fuel cell programs
- Sponsor development of SOFC based systems for serving retail markets
- Participate in development, evaluation, and demonstration of SOFC-CT systems
- Evaluate & develop PEM based fuel cell systems for retail markets
- Catalyze the formation of initiatives, joint ventures involving fuel cell technology

Our primary goals are to provide intelligence and fuel cell technology solutions to our clients to help them grow new and profitable business strategies based on advanced distributed resource technologies. By participating in bench scale testing, evaluation, development and demonstration we will provide validated data to support member interests in fuel cell technology investments. EPRI is in a unique position to assist both energy companies and fuel cell developers in defining/developing marketable products and in implementing their commercialization business plans within the evolving restructured electric power industry.

# GRI Fuel Cell Perspective

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## Outline

- A. Power Generation Perspective
- B. Basic Research on Solid Oxide Fuel Cells

## GRI Power Generation Strategic Elements

1. Use of Natural Gas in (1) competitive dispatch — central station; and (2) emissions control — central station.
2. Customer Generation — onsite.
3. Distributed Generation — utility controlled.

## GRI's Changing Imperatives

1. The gas industry is rapidly evolving from full-cost recovery to market pricing.
2. There is an increasing perception that share-holder dollars, rather than rate-payer dollars, are at risk.
3. Share-holder dollars carry a **much higher** discount (investment hurdle) rate.
4. The R&D portfolio must equilibrate to a higher discount rate **and** reduced resources.

## GRI Response

1. Re-examine project time frames, risk factors, and expected benefits.
2. Develop a lower-risk, nearer-term R&D portfolio.
3. No 1997 applied R&D fuel-cell budget or activities, although basic SOFC research continues.

## Where Do Fuel Cells Fit?

- **Onsite Generation**

**Premium Power** - applications needing sine-waves, not volts and amps; energy costs defer to other cost factors.

**Configuration** - base-load fuel cell on a dedicated circuit with grid as backup. *Fuel cell reliability must be unquestioned.*

- **Distributed Generation**

**Definition:** modular generation under the dispatch control of electric utilities.

**GRI focus** on characterizing distributed generation benefits, and defining required technology features.

**Application pull**, not technology push.

**Gas turbine and reciprocating engine** technologies continue to progress.

**Vast network** of unknown costs. Price transparency will uncover pockets of very high cost service.

**Performance-based** rate-making will motivate wire companies to a least-cost solution.

**Is modular generation cheaper** than substations and wires? Probably **yes** in enough cases to support multiple vendors.

## GRI View of Fuel Cell Status

1. The continuing challenge for the PAFC is to find high-value, mission-critical power niches that can easily afford the high first cost.
2. Recent MCFC test results point out how much research is still needed, while the MCFC slowly progresses toward product goals.
3. Basic research focuses on the SOFC because of its advantages for stationary applications, but is also monitoring the recent PEM progress.

## **SOFC Advantages in Stationary Applications**

- Natural gas fuel is processed directly within the stack — by internal reforming or possibly by direct oxidation.
- One compact system with effective heat transfer from the stack to reforming and air preheating steps.
- Long life because of the all-solid-state construction.
- Very high efficiency: SOFC only — 55 percent HHV; SOFC and Gas Turbine Cycles — 65 to 70 percent HHV.
- High quality heat for direct use, or gas turbine cycles.

## **Current GRI SOFC Research**

1. Reduced-temperature, planar systems (AlliedSignal, Univ. of Utah, Univ. of Pennsylvania)
  - Anode-supported, thin-film, scaled-up cells
  - Mixed-conducting electrode cells,  $\text{CH}_4$  oxidation
  - Inexpensive fabrication
  - Small stacks, metallic interconnects
2. Tubular systems (Westinghouse)
  - Fabrication methods
  - Cell design
3. Technical evaluation (TDA Research, Bechtel)
  - Manufacturing cost, system issues

## Planar Manufacturing Cost Assumptions

200 MW/yr, 24 hr/day operation, 95% availability	
Fixed capital investment:	3.9 times major equipment
Working capital:	20% of fixed capital
Raw materials:	Vendor projections
Labor:	200 employees at \$35,000/year
Maintenance:	4.2% of fixed capital
Depreciation:	10% of fixed capital
Property taxes and insurance:	2.6% of fixed capital
Overhead & administration:	8% of labor
Distribution, marketing, and R&D:	15% of total pretax expense
Profit:	Adjusted for ROR of 20%
Income taxes:	50% of profit

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ROR = (depreciation + profit - income tax)(fixed + working capital)

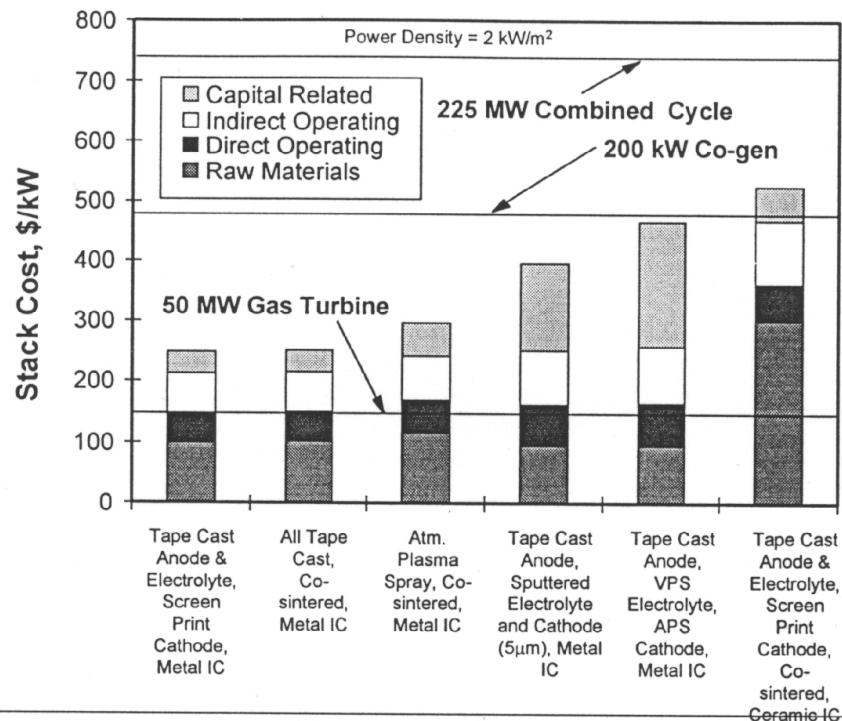
Annual cost = raw materials + operating + capital-related

Stack capital cost = (annual cost)(annual production)

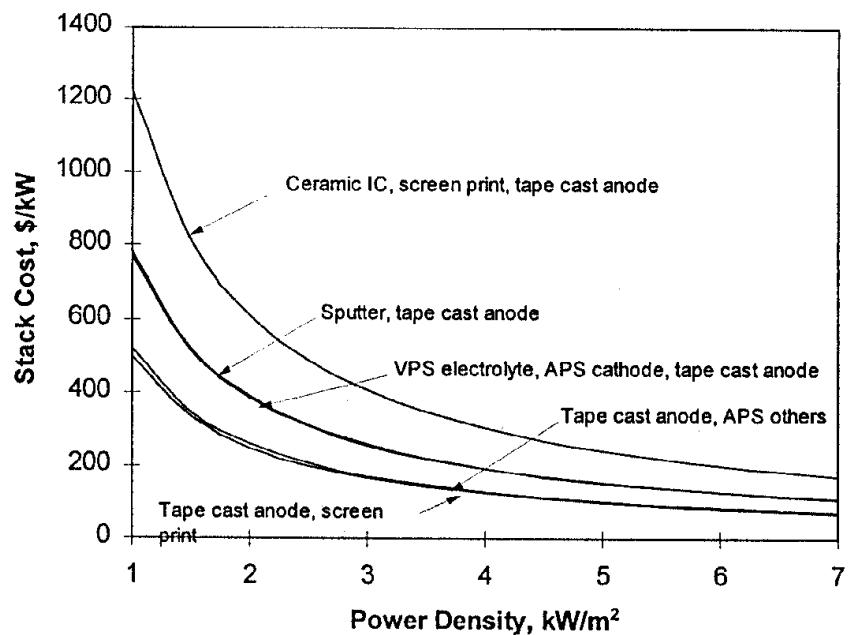
## Conclusions

1. Rapid changes in the energy industry have produced a nearer-term focus in GRI's power generation program.
2. Applied R&D is supporting improvements to microturbines, engines, and industrial turbines.
3. Basic research in reduced-temperature, planar SOFCs:
  - Technically challenging, longer-term option
  - Potential for more than \$700/kW capital cost
  - Higher power density, small size, good manufacturability
4. Basic research in tubular SOFCs:
  - Possibly ready for commercialization in about 3 years
  - Seal-less design, tolerance to thermal stress, recent technical progress, operability in high-efficiency, pressurized SOFC/turbine cycles

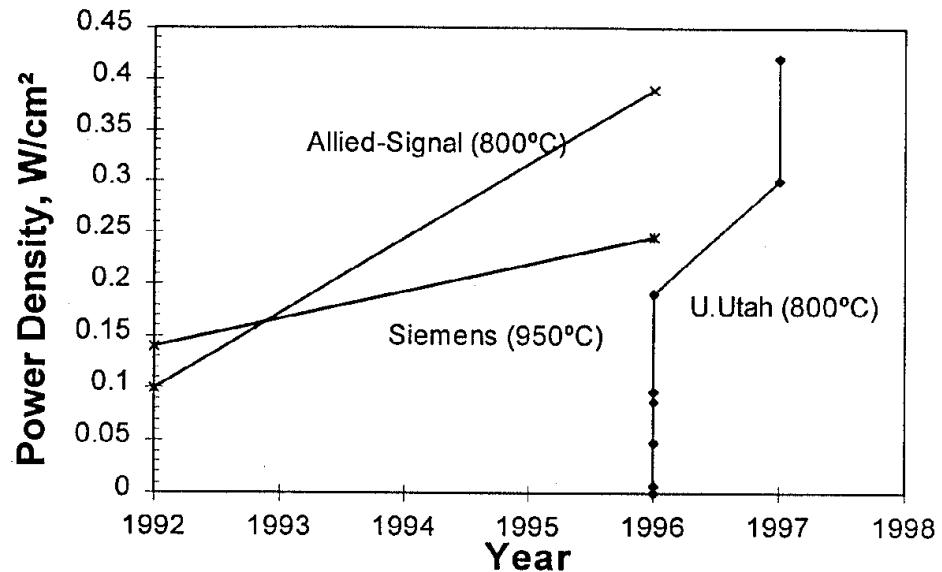
# SOFC STACK MANUFACTURING COSTS



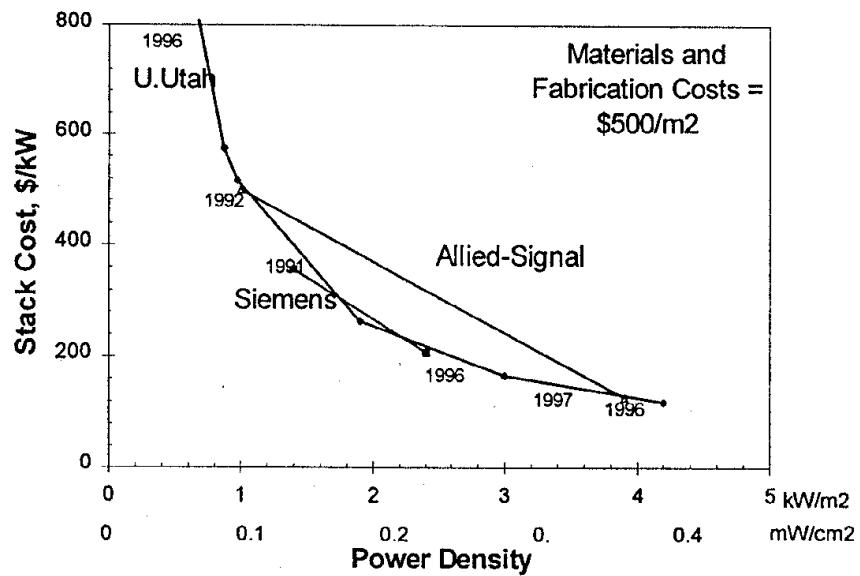
## EFFECT OF POWER DENSITY ON SOFC STACK COST



## COMPARISON OF PLANAR GEOMETRY STACK POWER DENSITIES

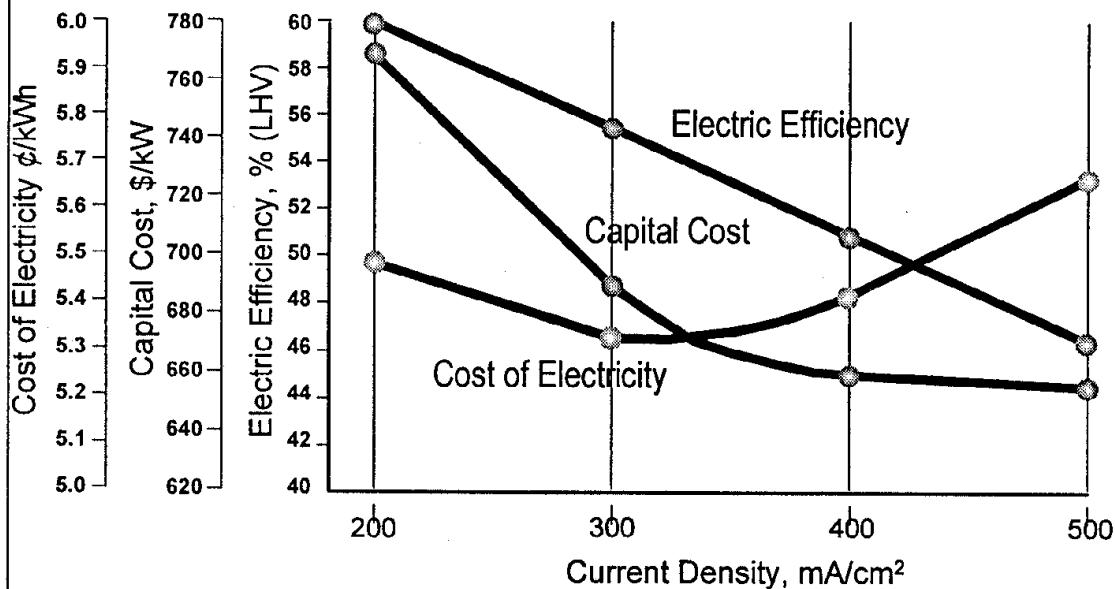


## THE COST OF SOFC STACKS IS DECREASING AS POWER DENSITY IS IMPROVED



## PLANAR SYSTEM COST AND EFFICIENCY

(800°C, 1 Atm, 85% Fuel Utilization)



# DOE/EE Transportation Perspective

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## Introduction

Fuel cells represent a developing technology which could potentially replace the internal combustion engine in all areas of the transportation sector. They operate with significantly higher fuel efficiency, greatly reduced emissions, and the capability of running on a variety of fuels (such as hydrogen, ethanol, methanol, and natural gas). The widespread introduction and use of fuel cell vehicles could have a major impact on reducing petroleum consumption and on improving air quality in urban areas. This paper provides an update on the status of the U.S. Department of Energy (DOE) program directed at the development of fuel cell propulsion systems for transportation applications.

## Program Drivers

This program is responsive to requirements of the U.S. Energy Policy Act of 1992 (EPACT) which authorizes the development of fuel cell vehicles. It also represents the key fuel cell work being done under the Partnership for a New Generation of Vehicles (PNGV) -- a U.S. government/industry research and development initiative involving representatives from seven Federal agencies and the three major domestic automakers (Chrysler, Ford, General Motors) that began in 1993 to strengthen U.S. competitiveness in the automotive industry. DOE's program specifically addresses the PNGV goal of developing a vehicle to achieve up to three times the fuel efficiency of today's comparable vehicle.

Besides the legislative drivers for this program, there is keen international competition in the race to develop PEM power systems for automobiles -- extensive efforts are underway in North America, Europe and Japan. Toyota demonstrated a fuel cell powered vehicle in late 1996. Daimler-Benz has built a second-generation six-passenger van powered by a 50-kW PEM fuel cell and has announced plans to demonstrate a small A-class car powered by fuel cells at the October 1997 Tokyo Auto Show. Daimler-Benz and Ballard Power Systems of Canada have announced a joint venture in which they will invest over \$200 million during the next three years in the development and commercialization of fuel cell engines.

## Program Goal

The goal of the DOE Fuel Cells in Transportation Program is to develop highly efficient, low or zero emission automotive fuel cell propulsion systems. Specific objectives include: By the year 2000, validate fuel cell propulsion systems that are (a) 2-3 times more energy efficient than today's comparable vehicles; (b) more than 100 times cleaner than Federal EPA Tier II emissions standards; and (c) capable of operating on hydrogen, methanol, ethanol, natural gas, and gasoline. In addition, by the year 2004, our objective is to validate fuel cell propulsion systems that meet customer expectations in terms of cost (competitive with conventional vehicles) and performance (equivalent range, safety, and reliability as conventional vehicles).

## **Technical Accomplishments**

Recent technical accomplishments of the program will be presented. Some highlights include:

- Platinum Catalyst Reduction -- Catalyst loadings on fuel cell electrodes were reduced to 0.25 mg/cm<sup>2</sup> with performance validated in 10-kW size stacks.
- Advanced Bipolar Plates -- Low-cost, lightweight metallic plates were developed by Mechanical Technology Inc, Allied Signal, and Delphi Automotive Systems and validated in fuel cell stacks of up to 10-kW size. As a result, stack power densities of greater than 400 W/kg were attained.
- Automotive-Size Stacks -- A full-size 50-kW PEM fuel cell stack was built and tested.
- Fuel-Flexible Reformer -- A 50-kW partial oxidation reformer was developed and demonstrated by Arthur D. Little, with measured efficiencies of 78% and 84% for gasoline and ethanol, respectively.
- Carbon Monoxide Clean-Up System - Los Alamos National Laboratory developed a preferential oxidation (PrOx) system to convert low levels of carbon monoxide to carbon dioxide in reformed fuels. The PrOx system reduces the CO concentration from greater than 1% to less than 0.01%, a level that does not degrade fuel cell performance.
- Low-Temperature Reforming Catalyst for Gasoline -- Argonne National Laboratory developed a novel, fuel-flexible partial-oxidation (POX) catalyst for reforming gasoline and alternative fuels. The product gas contained 60% hydrogen for gasoline and ethanol, 65% for methanol, and 72% for natural gas.
- High-Efficiency Air Compressors and Expanders -- Scroll, variable-displacement, and turbo compressors were built and tested. Air compressor efficiencies of 86%, and expander efficiencies of 90%, were measured with minimal loss in efficiency over a broad range of flow conditions.
- Fuel Cell Vehicle Design -- Fully configured fuel cell vehicle designs were developed by Ford, General Motors, and Chrysler Pentastar.
- Membranes for Direct Methanol Fuel Cells -- Improved membranes were developed by a team from International Fuel Cells and the Universities of Connecticut, Iowa, and Southern Mississippi. A Nafion/poly (propylene) dendrimer membrane reduced methanol crossover by 67% while adding only 8 mV to the cell voltage loss compared with Nafion 117 at 0.1 A/cm<sup>2</sup>.
- Modeling and Simulation -- Detailed systems models were developed at Argonne National Laboratory to permit PNGV to conduct systems analysis and vehicle engineering studies for fuel cell vehicles.
- Manufacturing Assessments -- Studies by divisions of General Motors, Ford and Chrysler concluded fuel cells could be manufactured in quantity at a price comparable to today's internal combustion engine.

## **Future R&D Activities**

DOE conducted a competitive procurement in 1997 for focused R&D on transportation fuel cells. DOE's future fuel cell R&D activities, to be completed during the next 2-3 years under \$50 million of new cost-shared initiatives, will be disclosed and discussed.

# Fuel Cells: DOD-Unique Applications

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## Abstract

The National Defense Center for Environmental Excellence (NDCEE), operated by Concurrent Technologies Corporation (CTC), has been tasked to support the Environmental Security Technology Certification Program (ESTCP) in investigating the use of fuel cells in Department of Defense (DOD) applications. This study emphasizes stationary power applications, as apposed to transportation and portable power applications. In addition, this analysis does not preclude any type of fuel cell technology. The applications identified in this task may be met by phosphoric acid, molten carbonate, polymer electrolyte membrane, or solid oxide fuel cell technology.

The information presented is based on the second report in a series of three documents written in support of the ESTCP task.

The first report, issued June 2, 1997, provides a detailed overview of fuel cell technology, fuel cell manufacturers, and key support organizations. In addition, the report summarizes current DOD fuel cell applications and installations.

The second report, which is the focus of this presentation, was issued on June 20, 1997. This report identifies fuel cell applications not currently pursued by the DOD, including premium power, DC power, and hydrogen source applications. To date, most fuel cells installed or to be installed under the fiscal year 1993/1994 DOD fuel cell programs handle non-critical electrical loads, with the primary focus on effective utilization of the recovered thermal energy. However, many other, more critical applications exist that could benefit from fuel cell technology.

The third report is scheduled to be issued in October 1997. This report will provide DOD decision makers with the information necessary to determine if they have a potential fuel cell application, and analyze the economics of installing a fuel cell at their facility. The guidebook will also provide information on siting considerations, environmental issues, and effect of deregulation of the electric power industry on fuel cell implementation.

Fuel cells can provide "premium power" to those critical or semicritical loads that require higher quality and/or reliability than that normally provided by the electric utility grid. Depending on their configuration, fuel cells can provide continuous power while serving as a backup or

uninterrupted power supply. Ten high-impact, premium power applications have been identified for the DOD, as follows

- Medical Treatment Facilities
- High-Security Facilities
- Communications and Data Centers
- Advanced Manufacturing Processes
- Electronics Manufacturing Processes
- Air Traffic Control Facilities
- Radar Sites
- Shipboard Service
- Research and Testing Facilities
- Remote Sites and Field Operations

Similar types of applications exist in the commercial industry and non-DOD government sectors. Examples of premium power applications include the trans-Alaska pipeline, U.S. embassies, and American Indian reservations.

In addition to these premium power applications, several potential DOD applications have been identified for the direct current (DC) power generated by fuel cells. These applications include many electrotechnologies, such as electroplating, electrocoating, and other industrial processes found in many Army, Navy, Air Force, and Marine maintenance and repair facilities.

Finally, potential hydrogen sources that can be used to power a fuel cell have been identified. The DOD can take advantage of this fuel cell applications in facilities that have sewage treatment plants or other means of generating a hydrogen-rich gas, such as heat treatment processes.

Meeting the high standards and critical power requirements of the DOD, fuel cells offer premium power in high-impact applications. In addition, these fuel cell applications meet similar needs found in the government (non-DOD), commercial, and industrial sectors. Successful implementation of fuel cells in these applications will, in turn, facilitate the commercialization and further development of this technology and reduce DOD energy costs, thereby reducing life-cycle costs.

## **National Defense Center for Environmental Excellence**

### **Vision:**

To be a National Resource for the Development, Application, and Dissemination of Advanced Environmental Technologies to the DOD, Other Government Agencies, and Industry.

### **Mission:**

- Assess and Prioritize the Nature and Seriousness of DOD Environmental Problems, and Identify Potential Solutions that Support the DOD Environmental Strategy.
- Transition Environmentally Acceptable Materials and Processes to Defense Industrial Activities and Private Industry.
- Provide Training Which Supports the Use of New Environmentally Acceptable Technologies.
- Perform Applied R&D, Where Appropriate, to Accelerate the Transition of New Technologies.

### **Environmental Security Technology Certification Program (ESTCP) Validation Task**

POP: October 1, 1996 - November 1, 1997

Task 1: Current Uses (June 1997)

Task 2: DOD-Unique Applications (June 1997)

Task 3: DOD Guidebook (October 1997)

## **Objectives**

- Look beyond current Congressionally funded work:
  - Most of the 30 fuel cells, installed or to be installed under the FY 93/94 program, are for non-critical electric and heating applications.
  - Emphasize unique, premium power applications.
- Focus on stationary applications (not transportation or portable power).
- Consider all fuel cell types (PAFC, MCFC, SOFC, PEMFC).

## **DOD Direction**

DOD's goal is to purchase utilities and services, including electricity, from public or private sources, especially for those sites located within the U.S.

The findings from this report will be made available to energy service providers for the purpose of evaluating this opportunity.

## **Methodology**

Literature Search: Web sites, reports, conference proceedings

NDCEE contacts

Survey

## **Expected Benefits of Premium Power**

Reliability (Backup power and UPS capability)

Quality

Security

Cost Avoidance (data loss; product loss; health and safety)

## **High-Impact Premium Power Applications**

Medical Treatment Facilities  
High-Security Facilities  
Communications and Data Centers  
Advanced Manufacturing Processes  
Electronics Manufacturing Processes  
Air Traffic Control Facilities  
Radar Sites  
Shipboard Services  
Research and Testing Facilities  
Remote Sites and Field Operations

### **Medical Treatment Facilities (MTF)**

- Fuel cells could provide premium power for critical and life safety operations, such as surgical services, emergency room, cardiology, intensive care, X-ray, labor and delivery, emergency lighting, and fire detection.
- 130 DOD hospitals and medical centers and 500 clinics worldwide.
- 41 MTFs could benefit from fuel cell technology.
- 163 veterans administration medical centers meet the same criteria for installation of a fuel cell as DOD MTFs.

### **High-Security Facilities**

Examples of secure facilities include: (1) National Simulation Center, F. Leavenworth, KS; (2) Ft. Leavenworth Prison (new facility); (3) National Gold Reserve, Ft. Knox, KY; (4) U.S. Mint, West Point, NY; and (5) Nuclear weapons storage, Elsworth AFB, SD. Devices such as electric magnetic card swipe entryways, and surveillance cameras require reliable power.

### **Communications/Data Centers**

Mission critical data centers support human life critical applications, such as early warning systems, combat information centers, emergency evacuation and support operations, and manned space mission satellite-operations centers.

High-demand data centers support critical applications, such as logistic processing centers, financial processing centers, and telecommunication processing centers.

## **Advanced Manufacturing Processes**

Quality controlled processes having extremely tight tolerances include CNC processing, CAD/CAM, and robotics.

A power outage may result in: (1) increased scrap material, (2) loss of production (throughput), and/or (3) possible equipment damage.

20 DOD sites were identified that have advanced manufacturing processes, including Army depots, Navy aviation depots, air logistics centers, and ship repair yards.

## **Electronics Manufacturing Processes**

Power quality in electronics manufacturing has a significant effect on the quality and cost of DOD electronic devices.

Power problems can cause large financial losses because of lost product and deferred production, for example, semiconductor manufacturing, PCB assembly, and system-level assembly.

## **Air Traffic Control Facilities**

1383 airports worldwide handle military air traffic.

These systems require clean, reliable power for safe operation of communications, computer systems, and runway lights.

## **Radar Sites**

National security is dependent on the reliable operation of worldwide radar sites. NORAD tracks over 8,000 objects in space using information from radar sites and satellites.

Fuel cells could supply primary or backup electrical power at these, often remote, locations.

## **Shipboard Service**

The U.S. Coast Guard and Navy have analyzed the potential use of fuel cells on board smaller combatant ships. Their findings note that fuel cells could:

- Increase survivability by distributing electrical power from multiple sources within the ship.
- Improve fuel efficiency by 12 to 20 percent.
- Lower noise and thermal signatures.
- Reduce environmental impact.
- Match future electrical (DC) power requirements for Naval weapon systems.

## **Remote Sites/Field Operations**

Deployed military units, particularly those that are permanent or stationary, include headquarters units, maintenance units, support units, and field hospitals. DOD facilities are located in Arctic and Antarctic regions. DOD facilities are also located on remote islands.

## **Other (non-DOD) Premium Power Applications**

- **Trans-Alaska Pipeline**, including pumping stations, remote electronic communications stations, and the proposed pipeline expansion.
- **U.S. Embassies:** 253 embassy-related facilities worldwide.
- **American Indian Reservations:** 314 federally recognized reservations; often located in remote areas.

## **DC Power Applications**

Very large amounts of DC power are used in electrochemical processes, such as anodizing, electrocoating, electroplating, electrolytic etching, and electrostatic spray painting.

49 DOD sites require DC power for industrial operations.

## **Hydrogen Source Applications**

Potential hydrogen sources include: industrial processes, such as heat treatment; sewage treatment plants; and other non-DOD sources, such as landfills and agricultural waste.

17 DOD sites have sewage treatment plants that could be sued to power a fuel cell.

## **Summary**

Fuel cell technology offers several environmental and operational advantages over conventional fossil fuel power generation methods.

In support of an ESTCP task to investigate the use of fuel cells in DOD applications, CTC has identified several premium power, DC power, and hydrogen source applications for fuel cells.

## **Recommendations**

Based on the positive results presented, additional work is indicated to properly evaluate the DOD applications identified. DOD sites with high-impact applications need to be identified. Information on fuel cell technology needs to be conveyed to the sites. And the potential application and economics of fuel cell technology need to be evaluated, using the DOD Guidebook (to be available October 1997).

# **DOD Fuel Cell Demonstration Program**

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## **INTRODUCTION**

The supply of reliable, cost-effective electric power with minimal environmental impact is a constant concern of Department of Defense (DOD) installation energy personnel. Electricity purchased from the local utility is expensive and represents only about 30% of the original energy input at the generating station due to generation and distribution inefficiencies. Because of master metering and large air conditioning loads, the demand portion of the installation's electric bill can be in excess of 50% of the total bill.

While the electric utilities in the United States have a very good record of reliability, there is significant potential for improving the security of electrical power supplied by using on-site power generation. On-site, dispersed power generation can reduce power outages due to weather, terrorist activities, or lack of utility generating capacity. In addition, as increased emphasis is placed on global warming, acid rain, and air pollution in general, the development of clean, highly efficient power producing technologies is not only desirable, but mandatory. Since the majority of central heat plants on U. S. military installations are nearing the end of their useful life, there is an opportunity to replace outdated existing equipment with modern technologies.

## **THE TECHNOLOGY**

Fuel cells are electrochemical power generators with the potential for attaining very high electrical energy conversion efficiencies while operating quietly with minimal polluting emissions. In addition, by-product thermal energy generated in the fuel cell is available for use for cogeneration of hot water or steam, bringing the overall potential conversion efficiency (electrical plus thermal) to approximately 85%. Air emissions from fuel cells are so low that several Air Quality Management Districts in the United States, including several in California which has the nation's strictest limits on air pollutants, have exempted them from requiring a permit to operate.

Phosphoric Acid Fuel Cells (PAFCs) are in the initial stages of commercialization. While PAFCs are not economically competitive with other more conventional energy production technologies at the present time, current cost projections predict that PAFC systems will become economically competitive within the next few years as market demand increases.

## THE DOD FUEL CELL DEMONSTRATION PROJECT

The FY 1993 Defense Appropriations Act provided \$6.0M worth of equipment procurement funds per Service for the implementation of "non-developmental item natural gas fuel cells currently in production in the United States ... for power generation at military installations ... " with the recommendation that "... some of the cells be installed at locations in need of enhanced air quality ...". The purposes of this demonstration project are to stimulate growth in the fuel cell industry, which will lower costs through economies of scale and competition, and to determine the role fuel cells should play in DOD long-term energy supply strategy. The three Services, acting through the Defense Utilities Energy Coordinating Council (DUECC), requested that the U. S. Army Construction Engineering Research Laboratories (USACERL), a U. S. Army Corps of Engineers research laboratory affiliated with the University of Illinois at Urbana-Champaign, coordinate this fuel cell demonstration program for all three Services.

The FY 1994 Defense Appropriations Act provided \$6.25M worth of equipment procurement funds per Service "to continue procurement of nondevelopmental item (NDI) 200 kW phosphoric acid natural gas fuel cells currently in production in the United States".

Specific tasks associated with USACERL's coordination role in this program include the following: (1) Procurement of turnkey Phosphoric Acid Fuel Cell Power Plant packages; (2) Evaluation of potential DOD site installation candidates in order to identify the specific sites where the PAFCs will be installed; (3) Monitoring of the electrical generation efficiency, degree of thermal utilization, air emission characteristics, and overall system reliability of the PAFCs to determine the economic and environmental benefits of owning and operating these systems; (4) Development of application guidelines based on the results of this project for the implementation of PAFC technology at DOD facilities; and (5) Documentation of all aspects of the entire DOD Fuel Cell Demonstration Project.

A solicitation was prepared for the purchase of turnkey Phosphoric Acid Fuel Cell Power Plant packages, to include purchase, site engineering, installation and startup, operation and maintenance training, and a five year warranty, maintenance and repair period. Following a negotiation period, ONSI Corporation was awarded a contract for the purchase of these turnkey PAFC systems. The terms of this contract involve cost-sharing on the part of ONSI Corporation and calls for partnering with the local utility serving the selected posts. A total of 12 200-kW PAFCs were purchased with the FY 1993 Appropriations and have been, or will be, installed at DoD installations, with specific installation sites being identified through contract modifications.

## SITE SELECTION CRITERIA

Initial candidate sites were identified by Army, Air Force, and Navy/Marine Corps Headquarters through solicitation of their respective Major Commands/Major Claimants. As awareness of the program grows, individual installations are requesting to become a part of this program. Initial screening of candidate sites is performed through an economic analysis based on total electricity and natural gas usage and average unit costs as provided by the Defense Energy Information System (DEIS). This economic analysis considers the electrical savings available

through operation of a fuel cell power plant. the associated natural gas costs to operate the system, and the natural gas savings obtainable through recovery of the by-product thermal energy.

Installations which appear to be good potential candidates as a result of this initial screening are then asked to submit copies of their actual past utility bills for a twelve month period in order that the economic analysis can be refined through the use of actual monthly energy consumption and utility rate schedule data. In addition, each candidate installation is asked to provide information regarding the degree of air quality attainment for the region in which they are located, as well as a description of the intended application for the recovered by-product thermal energy and an estimate of the amount of this recovered thermal energy which they could use. At the same time, potential opportunities for financial leveraging through cost sharing and/or rebates by the local utilities providing service to these candidate sites are investigated. Efforts are also made to insure equal distribution of fuel cell installation sites among the three Services, and to provide as wide a geographical and climatic distribution as feasibility allows.

Site visits are then made to those installations which still appear to be good potential candidate sites at the end of this evaluation process. These site visits allow for refinement of the estimate of by-product thermal energy usage, an analysis of the logistical factors surrounding potential fuel cell installation (e.g. distance from gas line, lengths of pipe and wiring runs, availability of sufficient land space for siting, etc.), and the development of a conceptual design package. The successful candidate sites are then identified to the ONSI Corporation to be selected installation sites through individual contract modifications. A kickoff meeting is held on site shortly after each contract modification to initiate the design and installation process.

## CURRENT STATUS

From the FY 1993 appropriation, ten ONSI model B PAFCs have been installed and are operational at Natick Research, Development and Engineering Center, MA. Newport Naval Education Training Center, RI, the 934th Tactical Air Group, Minneapolis, MN, Kirtland Air Force Base, NM, Twenty-Nine Palms Marine Corps Base, CA, Nellis Air Force Base, NV, Camp Pendleton, CA, Ft Eustis, VA, U. S. Military Academy, West Point, NY, and Picatinny Arsenal, NJ. A PAFC is also slated for installation at the Galley at the U. S. Naval Academy, Annapolis, MD during 1997 as part of an overall building renovation project. (Another PAFC was installed by others at Vandenberg AFB.)

From the FY1994 appropriation, five PAFC are operational (as of July 1997). The five operational plants are at the following sites: 911th Airlift Wing, Pittsburgh, PA; Naval Air Station (NAS), Jacksonville, FL; NAS Fallon, NV; Fort Richardson, AK; Edwards AFB, CA. Twelve additional sites are scheduled to be installed and operational by the end of September 1997. The twelve additional sites are: Laughlin AFB, TX; Watervliet Arsenal, NY; Fort Huachuca, AZ; Westover ARB, MA; Little Rock AFB, AR; Stennis Space Center, MS; Davis-Monthan AFB, AZ; Fort Bliss, TX; Pine Bluff Arsenal, AR; Port Hueneme, CA; Barksdale AFB, LA; and NDCEE, PA. Of the seventeen total PAFCs to be installed from the FY 1994 appropriation, thirteen will be ONSI model C power plants and four will be the older model B power plants.

As of July 1997 with 15 PAFCs installed operational, the fleet has logged more than 136,000 hours of operation (mostly by model B power plants). The first units came on line during February 1995. As of June 1997, the total fleet unadjusted availability was 74%. Several factors have contributed to this 74% unadjusted availability which is somewhat lower than the manufacturers stated (> 90%) availability for the worldwide fleet. In early 1996 after one unit experienced water chemistry problems related to local conditions, some units were intentionally shut down while the problem was being investigated. Another site intentionally shuts down their PAFC during winter months due to high gas prices. Adjusting for these intentional shutdowns, the fleet has an adjusted availability of 85%. This availability could be adjusted further by following the common industry practice of adjusting for down time which is not attributed to power plant itself. For the 136,000 fleet hours, estimated (cumulative) dollars savings (based on the actual Mwh generated) are more than \$900,000. Also, estimated (cumulative) pollution abated (compared to a typical mix of conventional electric utility generation) is approximately 45 tons of NOx and 90 tons of SOx.

Monitoring of PAFC performance continues while new plants are added to the fleet. It is anticipated that continued monitoring (contingent on available funding) will eventually permit a more detailed analysis of PAFC long term performance, identifying causes for outages, and development of application guidance for DoD facilities.

# **ONSI PC25™ Fuel Cell Power Plant Experience/ Status/Progress**

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## **Abstract**

Manufacturing of the PC25 fuel cell power plant began in 1991 and operation in on-site applications began early in 1992. Since that time, nearly 150 power plants have been delivered to customers in 13 countries throughout North America, Asia, and Europe, and the fleet has accumulated 1.5 million hours of operations.

Concurrent with the manufacturing and operating activity, the equity partners in ONSI have invested substantial resources in developing further improvements to the product and the supporting business capabilities. The most visible result of these activities is a new power-plant model, referred to as the PC25 C, which is one third smaller and lighter than the first power-plant model delivered to the market. Improvements to this model are in various stages of development, and introduction of these improvements will begin later in 1997. Sponsors of these improvements include Toshiba and United Technologies Corporation, who are owners of International Fuel Cells Corporation of which ONSI is a subsidiary, and a subsidiary of Ansaldo, who along with Toshiba and International fuel Cells, is an equity partner in ONSI.

The PC25 experience includes operation on natural gas, propane, light naphtha, and waste gas from waste-water treatment and landfill facilities, and hydrogen. The electrical configurations include both 60 and 50 Hz as well as grid-connected with automatic transfer to grid-independent operation in the event of a grid outage; other power plants have been operated as grid-independent units with transfer of the load to the grid in the event of a fuel cell outage. The product heat from the fuel cell has been used for domestic hot water, space heating, air conditioner reheat, and to drive adsorption air conditioners. A wide range of operating environments has been encountered, including temperatures from minus 40 to 128 Fahrenheit; altitudes from sea level to one mile above sea level; and shock and vibration from truck transport, transportation and rigging accidents, and earthquakes.

Durability and reliability statistics are encouraging. Individual power plants have operated for total periods in excess of 35,000 hours (over 4 years), confirming the long periods between overhaul, which has long been a projection for fuel cells. A few dozen continuous runs have exceeded six months and several have exceeded one year. The power plant incorporates diagnostics and data recording, which permit rapid restoration to operation after a forced outage. Remote control and diagnostics have been used to avoid shutdowns and to facilitate maintenance actions.

The PC25 is designed and fabricated in accordance with requirements developed by the American Gas Association Laboratories. These requirements are currently in the process of being upgraded to an ANSI standard. Other standards activities with the IEEE, ASME, are addressing interconnection, installation, and applications issues.

Improvements incorporated in the PC25 C model include a higher power density cell stack; improved cell stack manufacturing processes; an inverter utilizing IGBTs to reduce size and weight; a simplified and smaller fuel processor; compact heat exchangers; and an improved control system and improved packaging, including hinged panels for improved maintenance access. Further improvements under development are associated with these same components; these improvements will be inserted into production over the next few years. The improvements will also include broader operating capabilities associated with assured power applications.

# ERC Direct Carbonate Fuel Cell Program Overview

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## 1.0 INTRODUCTION

The carbonate fuel cell promises highly efficient, cost-effective, environmentally superior power generation from pipeline natural gas, coal gas, biogas, and other gaseous and liquid fuels. ERC has been engaged in the development of this unique technology, focusing on the development of the Direct Fuel Cell (DFC) technology[1,6]. 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 reforming reactor and associated heat exchange equipment. This approach provides upgrading of waste heat to chemical energy; thereby, it contributes to higher overall efficiency for conversion of fuel energy to electricity with low levels of environmental emissions. Among the internal reforming options, ERC has selected the Indirect Internal Reforming (IIR) - Direct Internal Reforming (DIR) combination as its baseline design. In the IIR-DIR stack, a reforming unit (RU) is placed in between a group of fuel cells. The hydrocarbon fuel is first fed into the RU where it is reformed partially to hydrogen and carbon monoxide fuel using heat produced by the fuel cell electrochemical reactions. The reformed gases are then fed to the DIR chamber, where the residual fuel is reformed simultaneously with the electrochemical fuel cell reactions.

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 by the end of the decade. The power plant design will include a diesel fuel processing option to allow dual fuel fixed DOD base applications. These power plants, which can be shop-fabricated and sited near

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Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center and the Department of Defense (DARPA) under contract DE-FC21-95MC31184 with Energy Research Corporation, 3 Great Pasture Road, Danbury, CT 06813; telefax; 203-825-6150.

the user, are ideally suited for distributed generation, industrial cogeneration, and uninterrupted power for military bases. 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 or other fuels in the early part of the 21st century.

ERC operated a 1.8 MW plant in 1996-97, the largest fuel cell power plant that ever operated in North America, at a utility site. This first-of-a-kind power plant has demonstrated high efficiency, low emissions, reactive power, and unattended operation capabilities. These accomplishments are addressed in a separate paper at this conference. Built on the experience of this full-size power plant field test, ERC launched the Product Design Improvement (PDI) program sponsored by government and the private-sector cost-share. The PDI efforts are focused on technology and system optimization for cost reduction, commercial design development, and prototype system demonstration.

## **2.0 OBJECTIVES AND APPROACH**

This program is designed to advance the carbonate fuel cell technology from the current full-size field test to the commercial design by the turn of this century. The specific objectives selected to attain the overall program goal are:

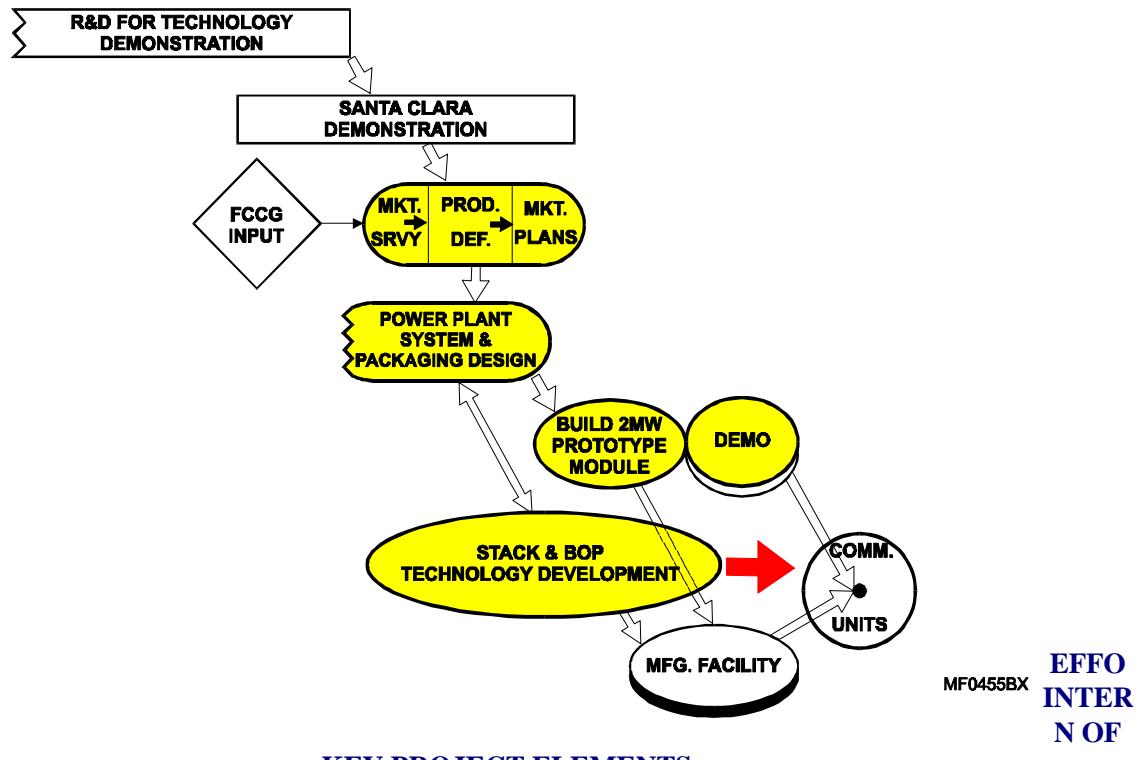
- Define power plant requirements and specifications,
- Establish the design for a multifuel, low-cost, modular, market-responsive power plant,
- Resolve power plant manufacturing issues and define the design for the commercial-scale manufacturing facility,
- Define the stack and balance-of-plant (BOP)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

A seven-task program, dedicated to attaining the objectives in the areas noted above, was initiated in December 1994. Program accomplishments of the past year is discussed in this paper. The previous year's progress was reported earlier[5-6].

## **3.0 PROJECT DESCRIPTION**

ERC is currently in the third year of the multiyear program for development and demonstration of a MW-class power plant supported by DOE/FETC with additional funding from DOD/DARPA and the ERC Team. Figure 1 shows key program elements (shaded area) and their interrelationships. The product definition and specification have been derived with input from potential users, including the Fuel Cell Commercialization Group (FCCG). The baseline power plant

final design has been completed. Detailed power plant system and packaging designs are being developed using stack and BOP development results. A MW-scale prototype modular power plant representative of the commercial design is planned to be constructed and tested. Based on the experience and data generated in the current program, ERC also plans to acquire manufacturing capability for market-entry products through expansion of the existing Torrington production facility.



A project team is in place to supplement all relevant expertise required for product design, improvement, verifications, and marketing.

#### 4.0 RESULTS/ACCOMPLISHMENTS

In the past year, the ERC team has made steady progress in the areas of system design, enhancing manufacturing capabilities, resolving technology issues, and preparing for system verification. Major accomplishments in each of these areas are discussed below:

##### Power Plant Design

The ERC team has completed the Final Design engineering effort of the baseline DFC power plant in collaboration with Fluor-Daniel, Inc. This design effort has already factored in the Santa Clara

Demonstration Project (SCDP) operating experience. The highlights of the SCDP operating experience input include improvements to the anode exhaust oxidizer, inverter electrical configuration, and plant layout for heat loss and maintenance considerations. The cost impact of the SCDP experience driven improvements/adjustments has also been evaluated and audited by an independent consultant and found to be within 1% of an earlier prediction[6]. The baseline plant is highly modularized featuring high efficiency, quiet operation, negligible emissions, and a small footprint that allows it to be sited in virtually any location. The plant has a rectangular footprint with a plot area of <420 square meter. The height of the plant will not exceed 7.6 meter. The DFC plant is designed for natural gas fuel, but modifications will allow use of other fuels such as landfill gas, and military logistic fuels. The plant will provide unattended operation with remote dispatching capability. The power plant specifications and engineering drawings available from the effort are sufficient to initiate the standard power plant procurement and construction activities.

The packaging approach for BOP and the stack module has been defined. The BOP equipment will be packaged into truck-transportable skids, complete with pre-installed piping, valves, insulation, instrumentation and electrical wiring. The truck-transportable fuel cell stack modules, each containing four fuel cell stacks and gas distribution to and from those stacks were, designed in conjunction with Jacobs Applied Technology, Inc. Progress in defining the stack module design is discussed later in this report.

## **Manufacturing Capabilities**

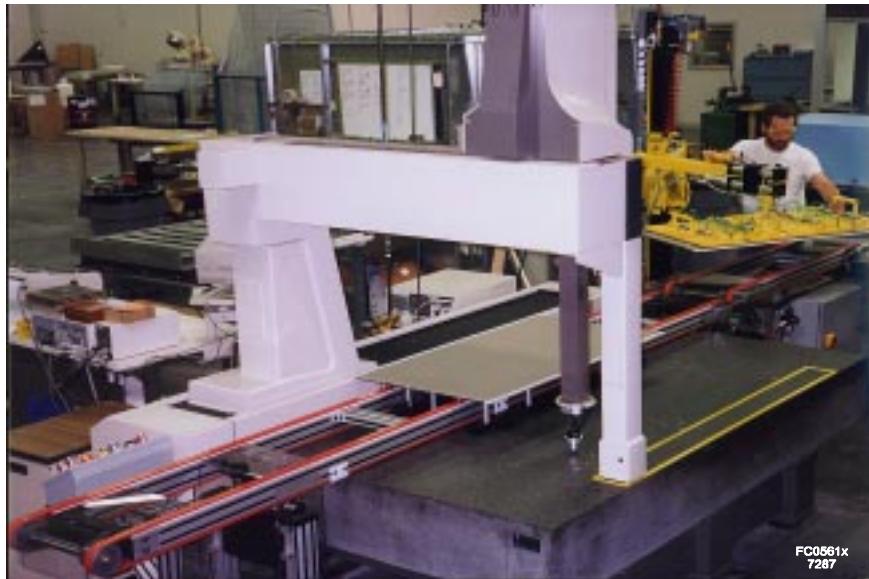
The stack manufacturing has been significantly enhanced by expanding the floor space, acquiring new tooling, and automating the quality check procedure for the cell packages. About 1300 meter square additional manufacturing floor space has been added to the existing manufacturing plant space of 5000 meter square. The components manufacturing processes have been streamlined to improve throughput, yield, and quality. FCMC has also prepared a semiautomatic full-height stack assembly facility. This facility will be used for assembly of the 300 plus cell stacks, the building block of ERC's DFC power plant. As the commercial components manufacture has been defined, the plant layout for the 24 MW/yr production capacity have been identified. Facilitization in accordance with this plan has been initiated. The present equipment capacity is approximately 17 MW/yr.

In parallel with manufacturing capacity enhancement, the components manufacture processes were improved and streamlined. The bipolar plate design has been refined to reduce thermomechanical stress and cost as well as improve mass manufacturability. Following successful qualification of the innovative design in the full area cell stack, the bipolar plate high rate manufacture tooling was designed, fabricated, and qualified in trial runs at the vendor's site. Next, the tooling will be commissioned at the Torrington, CT, manufacturing site. An automatic cathode manufacturing line has been commissioned to manufacture a fully quality-checked cathode every six minute starting with nickel powder. A photograph of the automatic cathode manufacturing line is shown in Figure 2. The matrix manufacture rate and yield have also been increased using a lower cost casting substrate.

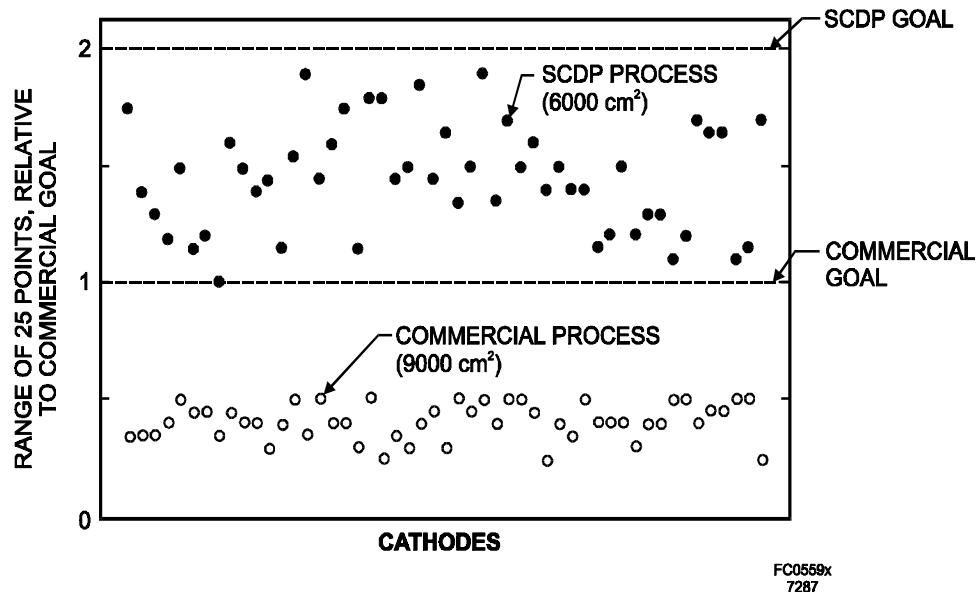


**Figure 2. AUTOMATIC CATHODE MANUFACTURING LINE:**  
Can Produce One Cathode Every Six Minutes

The cell components are quality checked by measuring thickness at twenty-five locations with a computer controlled thickness measuring machine. A photograph of the setup is shown in Figure 3. The range of the twenty-five measurements, i.e. the difference between the maximum and minimum values, is used as the individual components acceptance criteria.

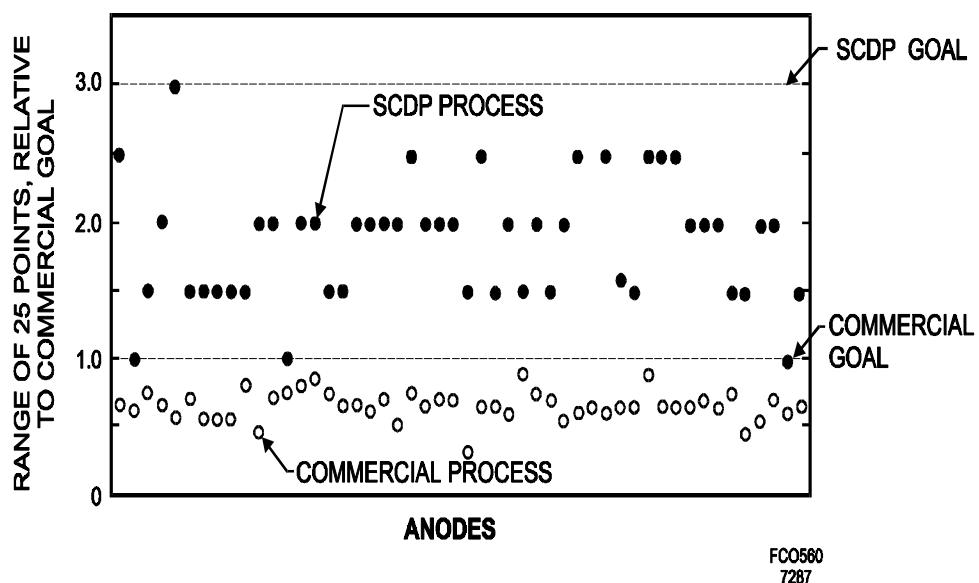


**Figure 3. COMPUTER CONTROLLED THICKNESS MEASUREMENT EQUIPMENT:**  
Thickness is Measured at Twenty-Five Points for Quality Check

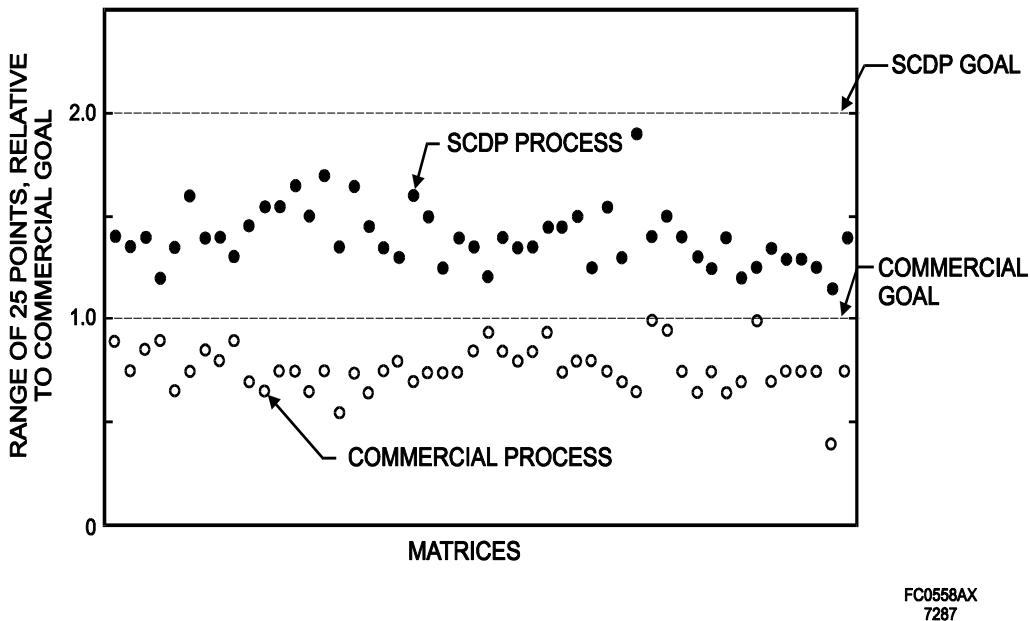


**Figure 4. COMPARISON OF THICKNESS RANGE OF COMMERCIAL PROCESS CATHODES WITH SCDP PROCESS CATHODES:**  
Thickness Variation Reduced by a Factor of Three

Together with the area scaleup, manufacturing rate, yield, and manufacturing tolerance of the cell components have been enhanced significantly. A comparison of the new generation  $9000\text{ cm}^2$  cathode quality measurement data (of fifty pieces of cathodes) with the previous generation  $600\text{ cm}^2$  cathodes is provided in Figures 4. The cathode thickness variation has been reduced by a factor of three and the piece-to-piece reproducibility, as shown by the scatter of the individual cathode measurements, has also been improved significantly. Similar quality improvements have been achieved for the anodes (Figure 5) and the matrices (Figure 6).



**Figure 5. COMPARISON OF THICKNESS RANGE OF FIFTY COMMERCIAL PROCESS ANODES WITH SCDP PROCESS ANODES:**

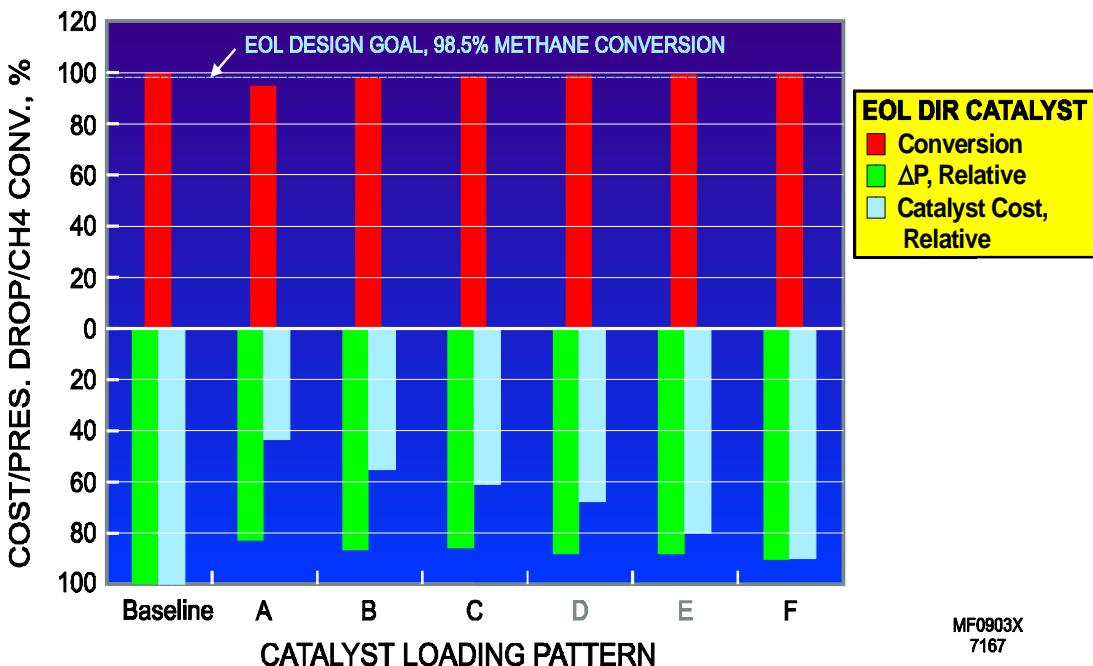


Thickness Variation Reduced by a Factor of Three **Figure 6. COMPARISON OF THICKNESS RANGE OF FIFTY COMMERCIAL PROCESS MATRICES WITH SCDP PROCESS MATRICES:**  
Thickness Variation Reduced by a Factor of Two

### Technology Improvement

A three-dimensional transient/steady-state computer model, describing fluid flow, heat and mass transfer, and chemical and electrochemical reaction processes has been developed based on the COMMIX computer code[8] for guiding the direct carbonate fuel cell stack design optimization. The computer model predicts three-dimensional distribution of gas flows, temperature and gas components as well as cell current density profile, cell potential, and pressure drops. The prediction is derived from numerical solutions of the conservation equations of mass, momentum and energy coupled with reaction kinetics and cell performance model. The models in the computer code have been validated by extensive laboratory data. Reasonable agreements between computed and fuel cell results, such as flow variations, temperature distributions, cell potentials, exhaust gas compositions, and methane conversions were obtained. Details of the model and modeling results with experimental DFC stack data were presented in a recent paper[9].

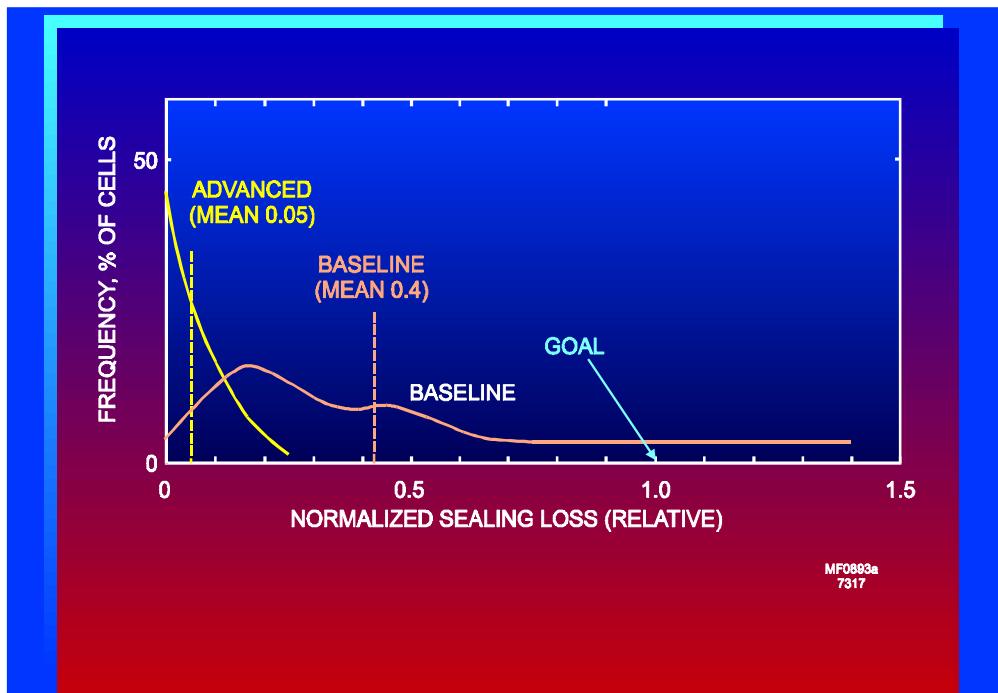
The model is being used at ERC as a cost effective tool for: 1) the DIR and reforming unit design optimizations, 2) evaluating stack performance (temperature profile, cell voltage, and pressure drops), and 3) investigating transient response to load changes. An example of the model utilization for optimization of the DIR design is discussed next. Six different catalyst loading designs were evaluated using the model. The model predicted end-of-life (EOL) methane conversion, anode chamber pressure drop, and the catalyst cost are compared in Figure 7. The Design E which promises 10% pressure drop reduction, 20% catalyst cost reduction over the baseline design and at the same time assures greater than 98.5% methane conversion at EOL (end-of-life) has been selected for stack testing.



**Figure 7. DIR CATALYST LOADING PATTERN OPTIMIZED USING MODEL:**  
Design E Selected for Stack Evaluation

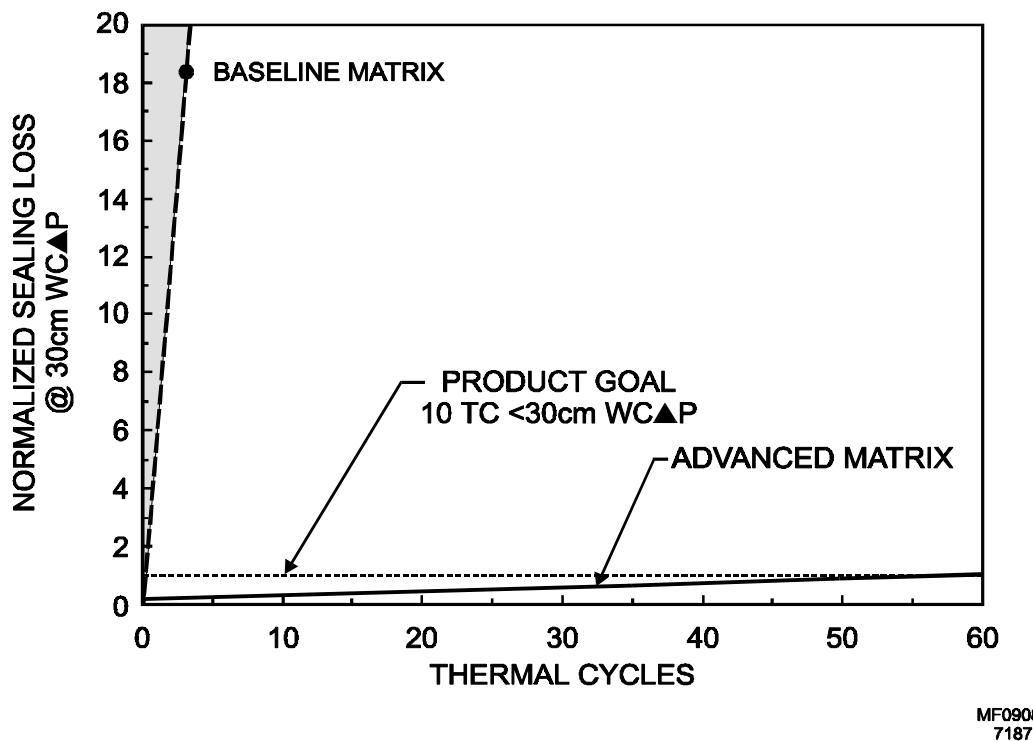
A high performance rugged matrix has been developed in a DOE/SBIR program and incorporated in the baseline DFC stack design. This matrix has resulted in excellent gas sealing capability and performance reproducibility in the DFC cells. A comparison of beginning-of-life gas sealing performance of laboratory cells ( $250\text{ cm}^2$ ) are compared in Figure 8 for two types of matrices, the advanced and the baseline designs. The frequency of cells, percent of the cells, is plotted against the gas sealing performance normalized with respect to the product goal. The laboratory cell data (built in 1996 to 1997 time period) show that for the cells built with the high performance matrix: 1) 100% of the cells met the design goal, 2) the mean gas sealing efficiency is approximately a factor of ten better than the design goal, and 3) cell-to-cell reproducibility is excellent. This improved design has also significantly enhanced thermo-mechanical ruggedness of the carbonate fuel cell.

The DFC power plant targeted for the baseload power generation application is expected to undergo five to ten thermal cycles over its life for planned and unplanned maintenance operations. A laboratory cell built with the advanced design has already been thermal cycled sixty times (the test is continuing) using an accelerated scheme for gas sealing loss. The cell maintained the design goal for gas sealing (as shown in Figure 9). The gas sealing goal set here is very tight,  $<0.5\%$  fuel loss at 30 cm of water column differential pressure which is equivalent to  $<0.2\%$  fuel cell performance loss. The cell electrochemical performance has also been unaffected by the repeated thermal cycles to room temperature (Figure 10).

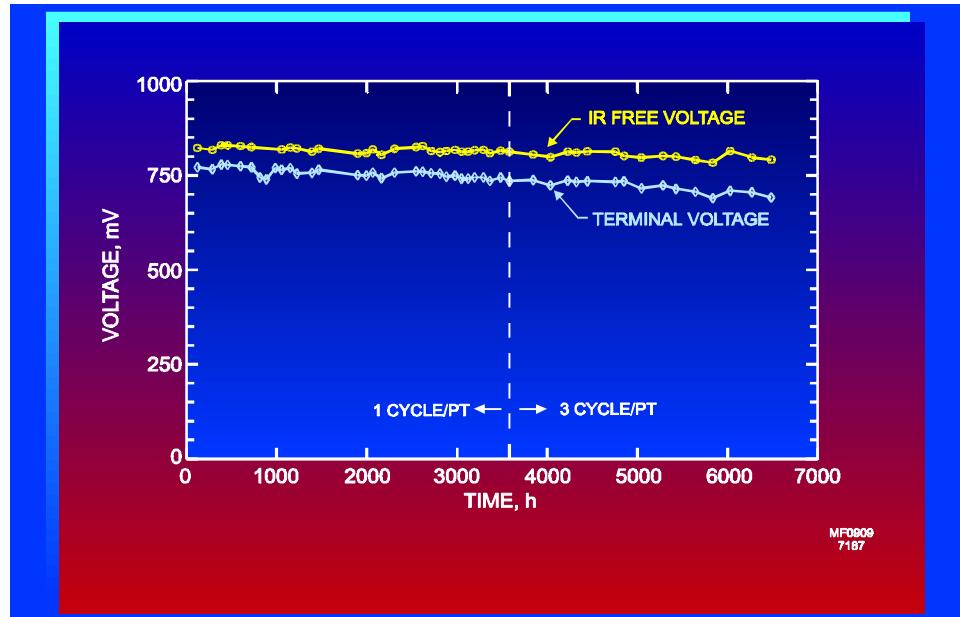


**Figure 8. COMPARISON OF GAS SEALING EFFECTIVENESS  
OF TWO TYPES OF MATRICES**

A Factor of Ten Improvement Achieved with the Advanced Matrices

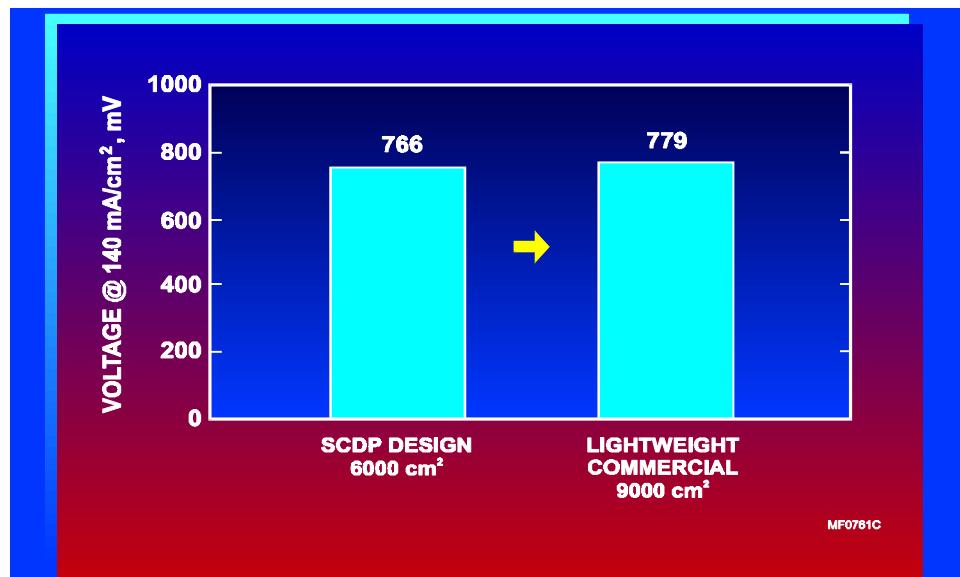


**Figure 9. EFFECT OF THERMAL CYCLING ON GAS SEALING:**  
Gas Sealing Goal Met Even with Sixty Thermal Cycles

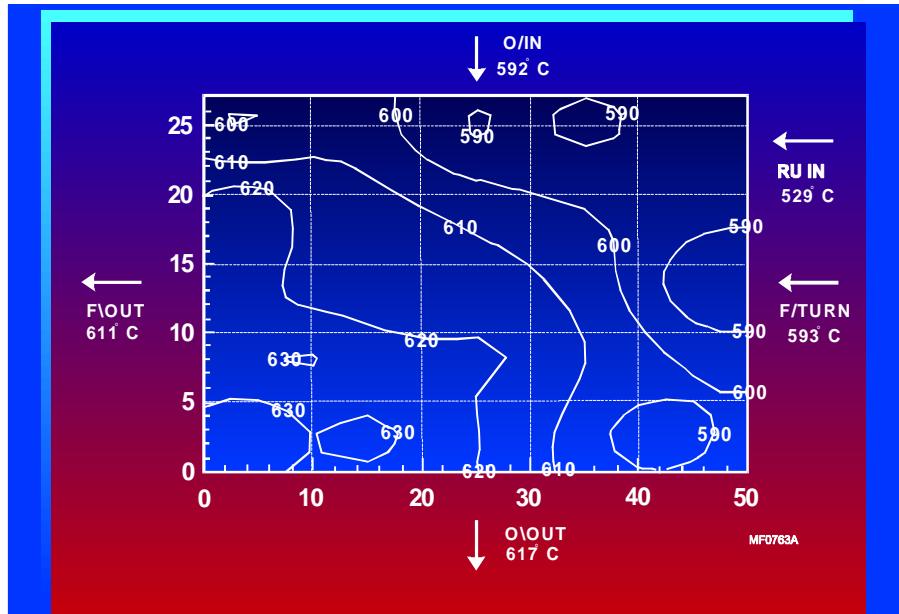


**Figure 10. EFFECT OF THERMAL CELL VOLTAGE:**  
Performance Unaffected by Thermal Cycles

The robustness of the commercial DFC design has been enhanced significantly by incorporating a thermo-mechanically compliant cell design and the advanced rugged matrix discussed earlier. The 10 kW-Class subscale stack (10-cell, 9000 cm<sup>2</sup>) test using this design has been initiated. The performance of this improved design stack is compared in Figure 11 with the Santa Clara generation 6000 cm<sup>2</sup> area stack. This lightweight, robust stack provided about 2% performance enhancement. The stack was operated at 160 mA/cm<sup>2</sup> producing ~1 kW per cell which is more than two times the cell power corresponding to the SCDP's maximum power operation point. The cell temperature distribution corresponding to the 160 mA/cm<sup>2</sup> operating case is shown in Figure 12. As the data show, the cell was operated with a temperature gradient of only 40°C even with a cold fuel inlet temperature of~520°C. This improved thermal distribution has resulted from the optimization of the IIR as well as the DIR designs.

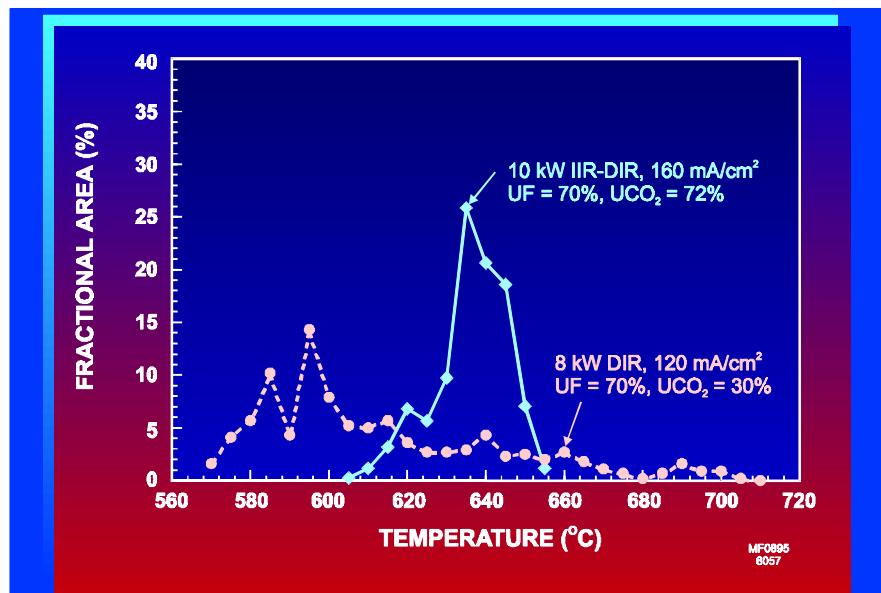


**Figure 11. PERFORMANCE STATUS:**  
~2% Performance Enhancement Achieved in Lightweight Full-Area Stacks



**Figure 12. TEMPERATURE PROFILE AT 160 mA/cm<sup>2</sup> (9000 cm<sup>2</sup>, 10-CELL DFC STACK):**  
Excellent Temperature Achieved at Rated Load

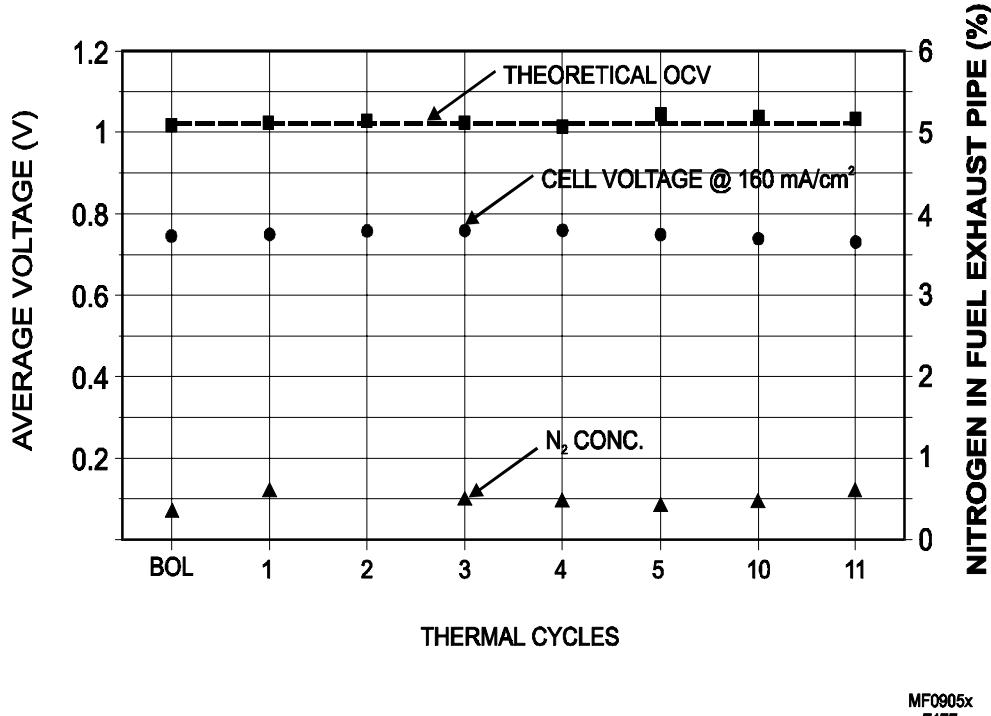
The IIR-DIR feature used in the ERC design lends to unique temperature control capabilities as compared to a DIR-only stack. This feature is illustrated in Figure 13, where the fraction of the cell area is plotted against the local temperature for two different internal reforming design types, the DIR and the IIR-DIR. These results point out that the IIR-DIR stack operates with a narrow temperature range. Therefore, for a given maximum cell temperature, the IIR-DIR design will provide the highest average cell temperature, hence, a better electrochemical performance. The baseline ERC IIR-DIR design can be optimized further by using the comprehensive DFC model discussed previously to fine tune the temperature profile.



**Figure 13. IIR-DIR AND DIR STACK TEMPERATURE DISTRIBUTIONS:**  
The IIR-DIR Provides Much Further Improved Thermal Uniformity

The 10 kW stack was thermally cycled eleven times without showing effects on gas sealing efficiency, electrochemical performance and cell internal resistance (Figure 14).

**Figure 14. EFFECT OF THERMAL CYCLING ON STACK**

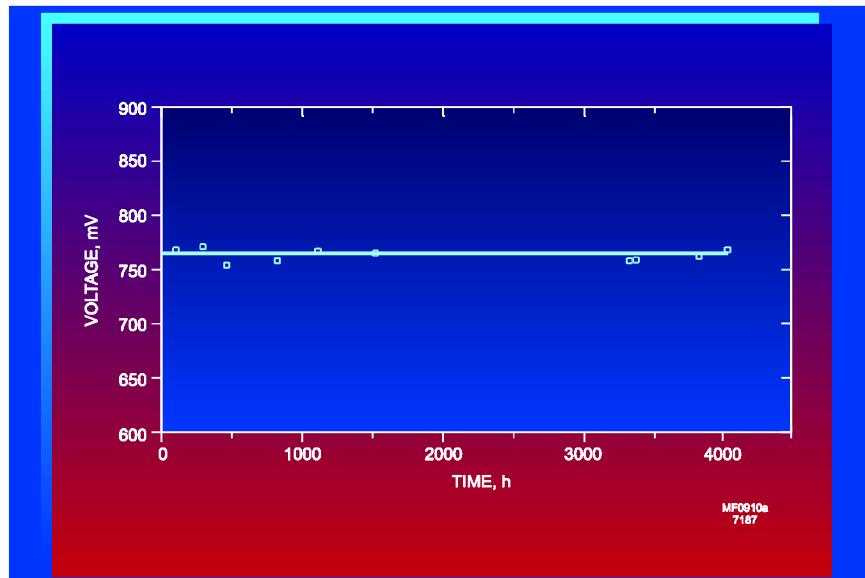


**PERFORMANCE PARAMETERS:**  
Stack Robust to Thermal Cycling

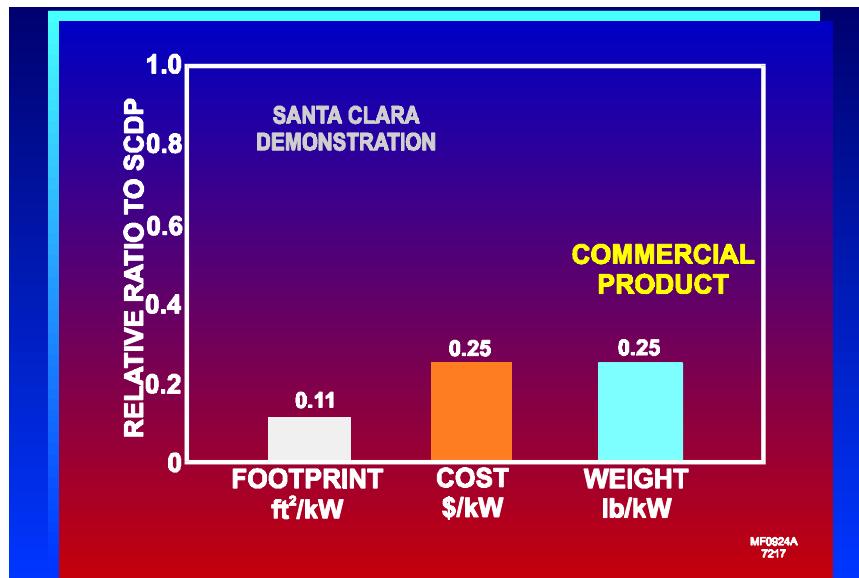
The baseline matrix uses a high surface area submicron gamma-LiAlO<sub>2</sub> powder as the matrix support material. Recently, the alpha-LiAlO<sub>2</sub> has been shown to be most stable in the carbonate environment. Use of this material is, therefore, expected to enhance matrix electrolyte retention in the carbonate fuel cell. The advanced matrix has been successfully fabricated using the alpha phase material. Two single-cell tests conducted with this material for greater than 4000 h each have demonstrated significant performance stability improvement. In fact, no decay in cell terminal voltage has been noticed in the initial data (Figure 15). The current focus is scale up of this design and implementation in full-area stacks.

Compared to the individually insulated stacks in a rectangular box used in the SCDP plant, the stack enclosure in the commercial product is an internally insulated circular box. The packaging, cost, and weight benefits of the new design are shown in Figure 16. On an equal output power basis, the stack enclosure footprint, weight, and cost have been reduced by factors nine, four, and four, respectively. The stack enclosure conceptual design was developed in 1994-95 time period. The important considerations for the design are: 1) high performance insulation, 2) electrical penetrations and conduction through hot environment, 3) internal gas distributors, 4) baseplate, and 5) packaging of four stacks and the gas distribution system within the truck transportable envelope. These designs have been evolved in 1996-97 time period through iterative stack tests. A multifunctional end plate which has integrated end plate and the stack compression plate functions has been made available (a

photograph of the plate is shown in Figure 17) from this effort. This design has resulted in elimination of end heaters, cathode side pipe dielectrics, and a 40% reduction of the expansion joints. To date six subscale tests of the stack module design have been completed. A full-height enclosure stack has also been fabricated for a test to be conducted towards the end of 1997. Based on the single stack enclosure experience, the four- stack enclosure design is being finalized. Once the design is finalized, a full-size simulator will be constructed for cold and hot verification tests. The four-stack module is truck-transportable. A “goose-neck” type transporter design is being finalized in collaboration with a transporter vendor.



**Figure 15. STABILITY OF ADVANCED CELL DESIGN:**  
No Decay Observed



**Figure 16. COMPACT STACK MODULE DESIGN:**  
Footprint Lowered by Factor of Nine



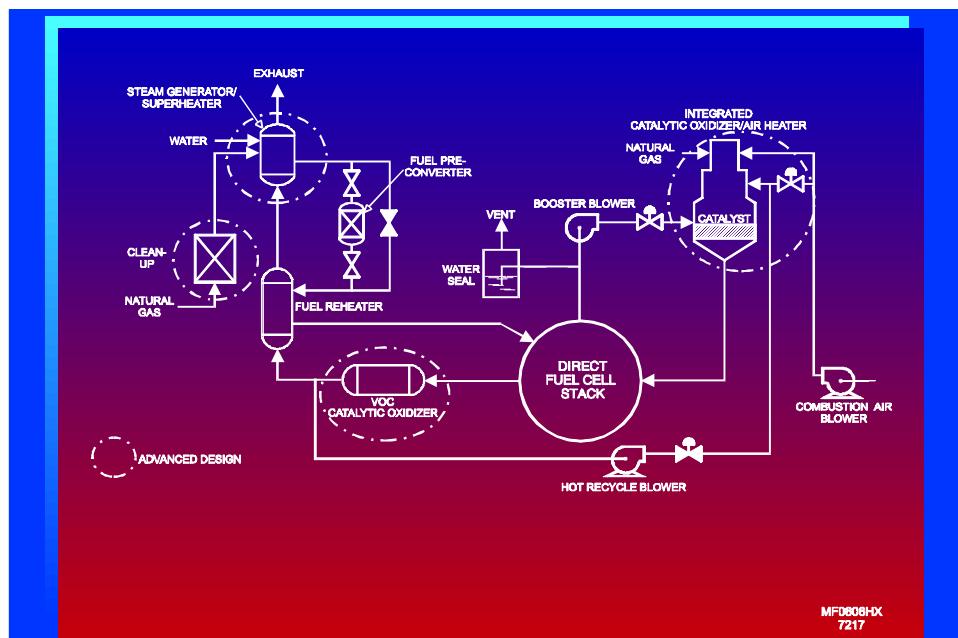
**Figure 17. MULTIFUNCTIONAL END PLATE DEVELOPED:**  
Eliminated Parts, Reduced Cost, and Allowed Compact Packaging

### System Design Verification

A 400 kW subscale power plant test facility has been constructed at ERC for evaluation of the major BOP equipment, full-size stack module, and power plant control and operational parameters. A process flow diagram of the power plant is shown in Figure 18. The power plant test facility includes a unique anode exhaust oxidizer design developed by ERC. The basic design has been validated by computer simulation in collaboration with the DOE/FETC in-house R&D engineers under a CRADA. This test facility simulates ERC's baseline power plant operations including automatic startup, shut-down, load change and steady state as well as provides conditioning of the full-size stack. The power plant construction and assembly have been completed (a photograph is shown in Figure 19). It is currently undergoing PAC (process and control test) and is expected to be ready for full operation by the fourth quarter of 1997.

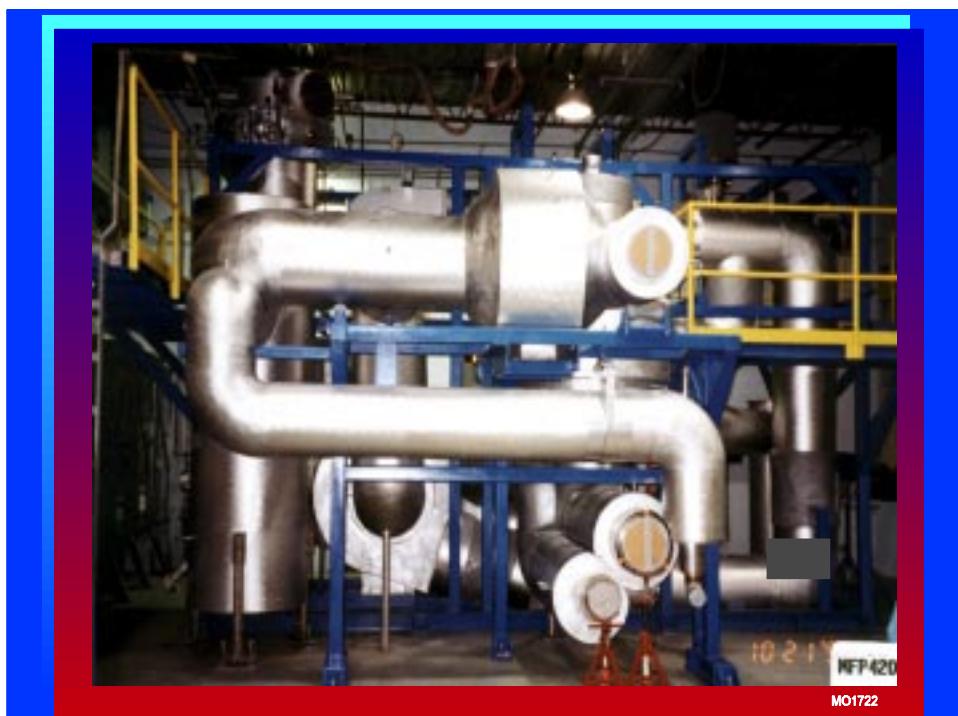
### Summary

The major accomplishments of the past year include the following: 1) completed baseline power plant final design and constructed 400 kW power plant test facility, 2) established commercial manufacturing processes for cell (repeat) components, 3) verified robustness of commercial cell design at subscale, and 4) verified stack module design in subscale stack tests; scaleup is in progress. The future activities will focus on testing full-size stacks and BOP equipment in the power plant test facility and construction of the stack conditioning facility.



**Figure 18. ERC 400 kW-CLASS POWER PLANT PROCESS FLOW DIAGRAM:**

Will be Used as Test Bed for Advanced Subsystems



**Figure 19. 400 kW POWER PLANT MECHANICAL CONSTRUCTION AND ASSEMBLY COMPLETED:**

This Power Plant test Facility will be Ready in the Latter Part of 1997

## ACKNOWLEDGMENT

The support of DOE and DARPA for the system and technology development and testing is gratefully acknowledged. The insight guidance and support of the FETC contracting officer's representative, Mr. Tom George, and DARPA representative Dr. Robert Rosenfeld are also appreciated.

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# Santa Clara Direct Carbonate Fuel Cell Demonstration

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## Introduction

Fuel Cell Engineering Corporation (FCE) has been involved in a DOE Cooperative Agreement Program (private-sector cost-shared) aimed at the demonstration of direct carbonate fuel cell (DFC) technology at full scale. FCE is a wholly owned subsidiary of Energy Research Corporation (ERC), which has been pursuing the development of the DFC for commercialization near the end of this decade. The project involves the design, construction, and testing of a 2MW carbonate fuel cell demonstration power plant in the city of Santa Clara, California. Construction of the plant - including the installation of the fuel cell stack modules - was complete in March 1996. Testing was begun in April 1996 and the test program was completed in this reporting period, in March 1997.

## Objectives and Approach

Potential users of the commercial DFC power plant under development at ERC will require that the technology be demonstrated at or near the full scale of the commercial products. The objective of the Santa Clara Demonstration Project (SCDP) was to provide the first such demonstration of the technology. The approach ERC has taken in the commercialization of the DFC is described in detail elsewhere [1]. An aggressive core technology development program is in place which is focused by ongoing contact with customers and vendors to optimize the design of the commercial power plant. ERC has selected a 2.4 MW power plant unit for initial market entry. Two ERC subsidiaries are supporting the commercialization effort: The Fuel Cell Manufacturing Corporation (FCMC) and the Fuel Cell Engineering Corporation (FCE).

FCMC manufactures DFC stacks and multi-stack modules, currently from its manufacturing facility in Torrington, CT. FCE is responsible for power plant design, integration of all subsystems, sales/marketing, and client services. The commercial product specifications have been developed by working closely with the Fuel Cell Commercialization Group (FCCG). FCCG members include municipal utilities, rural electric co-ops, and investor-owned utilities who have expressed interest in being the initial purchasers of the first commercial DFC power plants. The utility participants in the SCDP have been drawn from the membership of FCCG.

FCE was the prime contractor for the design, construction, and testing of the SCDP Plant, and FCMC manufactured the multi-stack submodules used in the DC power section of the plant. Fluor Daniel Inc. (FDI) served as the architect-engineer for the design and construction of the plant, and also provided support to the design of the multi-stack submodules. FDI is also assisting the ERC companies in commercial power plant design.

## Project Description

The project involved the design, construction, and testing of megawatt-scale DFC demonstration power plant in the city of Santa Clara, California. The rated output of the nominally 2MW SCDP plant was 1.8 MW. The plant is located at 1255 Space Park Drive in Santa Clara. The site is owned by the City's Electric Department and is adjacent to a 115/60kV switching station on the City electrical system. A photograph of the power plant is shown in Figure 1.

The natural gas fueled power plant consists of the fuel cell power section (16 electrochemical fuel cell stacks, configured into four 4-stack submodules) and the balance of plant (BOP) equipment. The BOP is comprised of the process, mechanical, and electrical equipment which provides the required gas flows to the stacks and converts the stacks' DC power to AC power at the required grid voltage. The design of the power plant is based on ERC's proprietary DFC "Simplified Design," which is also the basis for ERC's initial commercial offering. The Simplified Design includes provision for startup, fuel cleanup, recirculation of carbon dioxide to the cathode side and exhaust of spent gases through a Heat Recovery Unit (HRU) which provides the required fuel pre-heat and steam generation. ERC has also investigated other process design options which provide higher efficiency operation, but at the expense of increased system complexity and higher capital cost



**Figure 1**  
**Santa Clara Demonstration Project Power Plant**

The design and fabrication of the fuel cell stack modules was done with the support of the U.S. Department of Energy through FETC, under Cooperative Agreement DE-FC21-92MC29237. The period of performance for the Cooperative Agreement is October 1, 1992 through September 30, 1997.

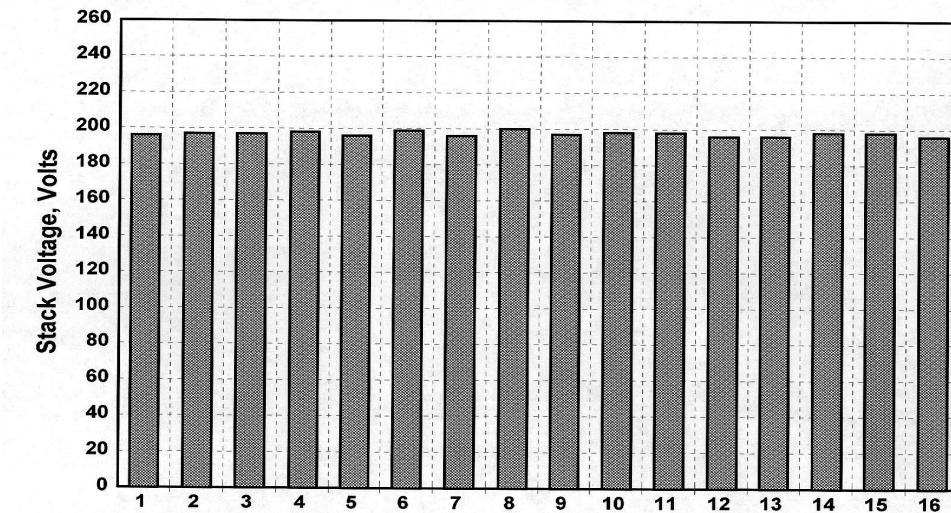
The design and procurement of the balance of plant and the construction and testing of the complete system was supported by the Santa Clara Demonstration Participants. The participants in the SCDP are as follows: City of Santa Clara, City of Los Angeles Department of Water and Power, City of Vernon, 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) also supplied some project funding through a consortium agreement with the City of Santa Clara. In addition, the California Energy Commission has provided funding to the City of Santa Clara to partially support the balance of plant pre-test activities. ERC also contributed to the project, at about the level of an SCDP share.

## Results

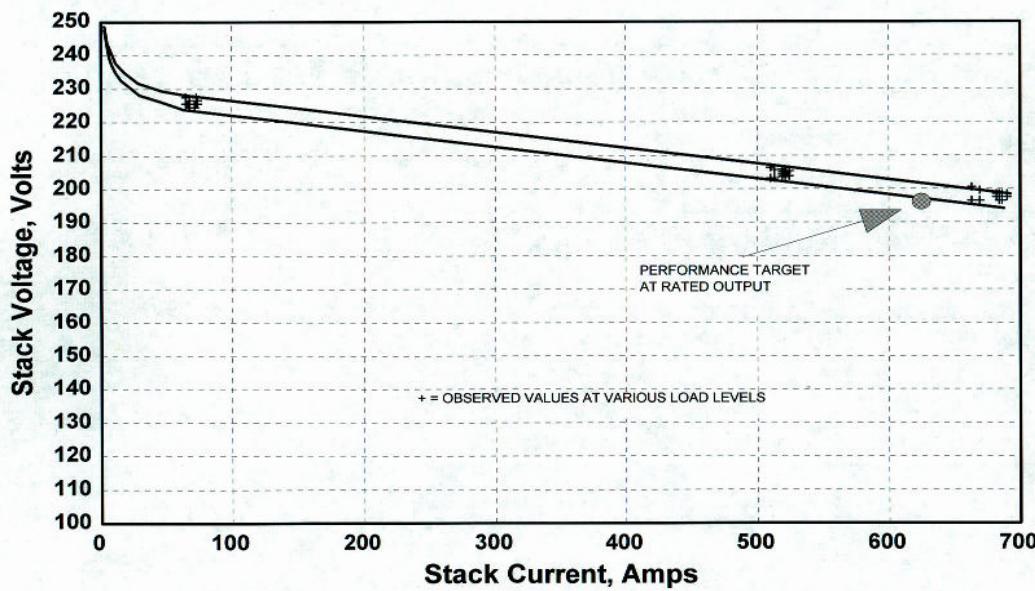
As reported last year, the plant startup was begun in April 1996. After the initial heatup, power generation operations began with a period of gradual power level increases during Power Conversion Unit tuning operations. The plant output was increased beyond the 1.8 MW rated capacity to 1.93 MW, during which time the fuel cell stacks exhibited extremely uniform performance (+/- 1% variation in stack voltage) at levels in excess of the plant design projections. The voltage uniformity at 1.93 MW net AC is shown in Figure 2, and the voltage performance of the stacks over the load range is shown in Figure 3. In the first run of operation at rated power, the plant was operated in a conservative mode, since the stability of the PCU system had not yet been confirmed and the response of the system to grid disturbances had not yet been demonstrated. This involved running the two auxiliary natural gas burners in the plant (one in the anode exhaust line and one in the HRU) in order to enhance system stability in the event of a plant trip. These burners would normally be off, and would be started in the event of a trip to standby, so keeping them running would provide a “soft landing” plant trip.

With the auxiliary burners firing, the system efficiency did not meet the target 49% (LHV) level, but an efficiency of 44% (LHV) was achieved, a record for a simple cycle natural gas power plant at this size level. The performance of the stacks and the system operating parameters indicated that once the auxiliary firing was curtailed, the 49% target could easily be met.

During operation at rated power, transients were observed in some of the stack voltages and the system was shut down for inspection. The cause was determined to be parasitic electrical circuits, caused by a breach of the electrical isolation between the fuel cell stacks and the gas distribution piping. The source of these circuits was traced to a glue used to install the thermal insulation around the feed and exit process lines to the fuel cell stacks. At elevated temperatures glue became electrically conductive, diminishing the effectiveness of dielectrics used to isolate fuel and air metal pipes from the fuel cells. Since differential potentials can reach 1000 VDC in the power plant, it became possible for stray parasitic currents to flow through the tainted dielectrics.



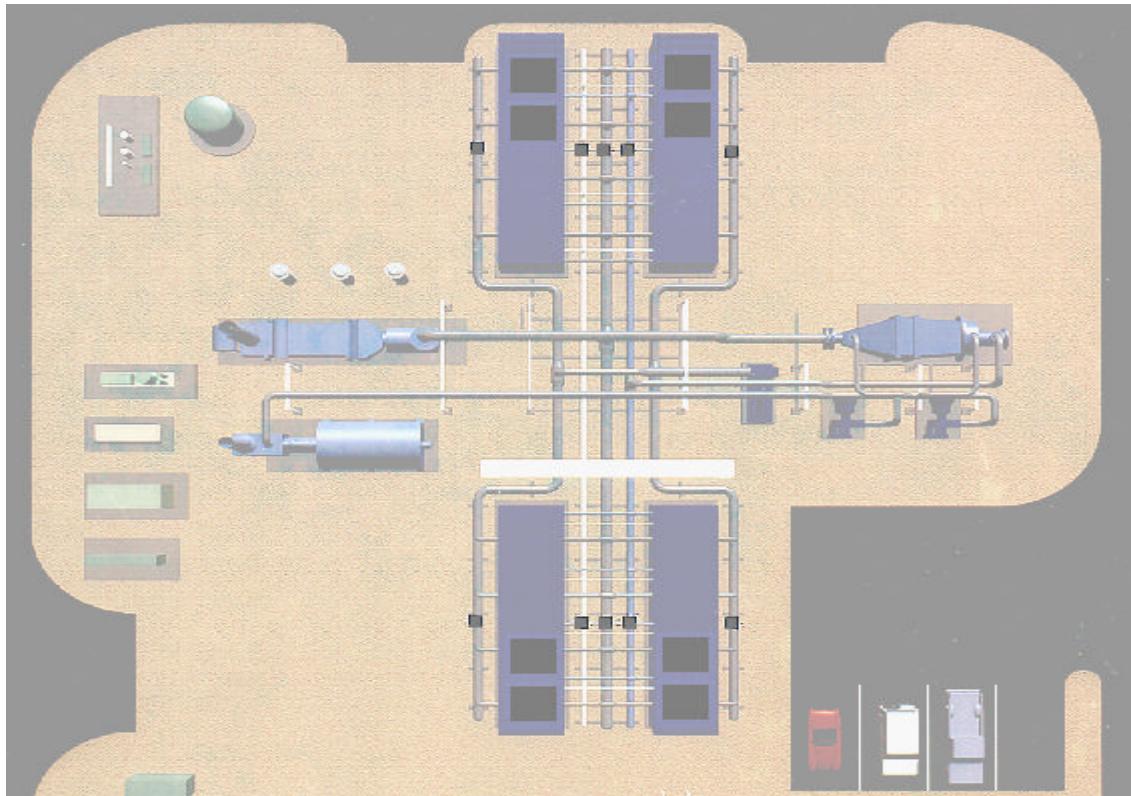
**Figure 2**  
**Voltage Performance of 16 SCDP Stacks on Load at 1.93 MW net AC Power**  
**The Stack Voltages Were Uniform to Within  $\pm 1\%$**



**Figure 3**  
**Voltage Performance of SCDP Stacks Over Full Range of Power Output**  
**The Stacks Performed Slightly Above the Target Design Basis Level**

Power plant operations were resumed once the compromised dielectric components were repaired or replaced. During this second operating period, power output was ramped to 1.2 MW gross DC (1.0 MW net AC) before the performance of some of the stacks precluded further ramping. It was observed that all of the stacks operating below design levels shared a common position in the electrical circuit. It appears that during the pipe dielectric shorting which forced the first plant shutdown, the voltages of the shorted pipe and stack hardware in some portions of the circuit were driven to levels which triggered secondary electrical damage at the stacks. This left residual damage which became progressively worse during the second operational run. The eight stacks in the unaffected portion of the circuit did not incur this problem, and all eight were observed to operate well.

As a result, operations were continued using the unaffected eight stacks in a reconfigured 1MW operating mode. Isolation of the desired stacks was done by installing blinds midway down the run of the flow headers, as shown in Figure 4. This eliminated the eight stacks at the four ends of the piping system, preserving a symmetrical flow configuration which is important in maintaining a uniform pressure profile throughout the stacks. The mechanical reconfiguration was done in a 10 hour operation, during which time the stacks were not delivered any gas flows.

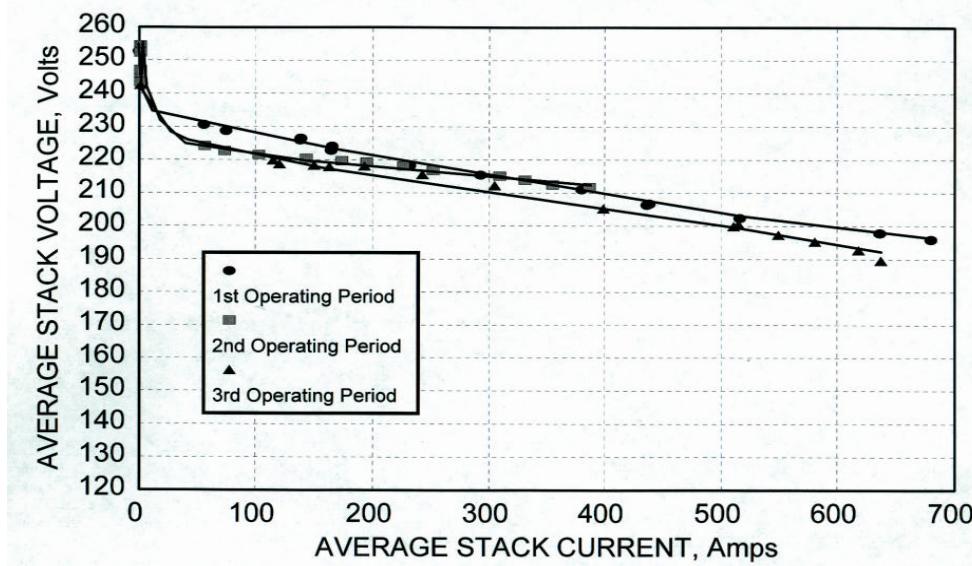


**Figure 4**  
**Stack Isolation Approach Used For Plant Reconfiguration**  
**The Isolation Preserved a Symmetrical Flow Configuration to the Eight Operating Stacks**

Following the mechanical reconfiguration, the plant was left in hot standby awaiting the delivery and installation of an additional transformer between the inverter and the grid, which was needed to allow the lower voltage output of the reconfigured power plant to interface with the City's electrical system. Once the transformer was installed the third operating period was begun. The planned curtailment of auxiliary fuel firing could not be done in the third operating period, since the heat output from the eight stacks was not sufficient to support the HRU pre-heat and steam generation functions, without the use of auxiliary firing.

The initial power ramp in the third operating period brought the plant to a power level of 950 kW gross DC, 95% of the Gross DC target level for the 8-stack system. Figure 5 compares the average performance of the operating eight stacks during this load ramp to the performance of the eight stacks during the load ramps in the first and second operating periods. It can be seen that the performance of the stacks in the load ramp in the second period was very similar to the ramp at the beginning of the first operating period.

The load curve for the third period is similar in terms of polarization slope, but the average stack voltage was slightly less than that seen in the previous two operating periods. The slightly lower voltage was due to the lower operating temperatures of the stacks. As the 8-stack process gases were circulated through the 16-stack piping system, the normal plant heat losses resulted in twice the cooling effect due to the reduced mass flow. As a result, the inlet temperatures to the stacks were significantly cooler than in earlier operating periods and the exit temperatures were slightly cooler than in previous runs. The lower voltage of the stacks was consistent with model predictions of the temperature effect. The temperature differential across the stacks - the difference between the coolest inlet and the warmest outlet process gas - was 30 to 40 percent higher than in earlier power generation periods at comparable power output levels.



**Figure 5**  
**Comparison of Initial Power Ramps in Three Operating Periods**  
**Performance Variations Were Limited To Effects Of Temperature And Gas Composition**

While operating at the 95% DC power level, the performance of two of the stacks began to decline. The possibility that the wide thermal gradients were causing localized contact problems was considered, and the plant was put on standby for about 150 hours to allow the stacks to thermally equilibrate on standby. On resumption of power generation, one of the two weak stacks recovered performance and the other stabilized, indicating that the thermal gradients had played a role in their performance loss. In this and subsequent operations the thermal gradient across the stack was minimized by running with a higher than normal steam content in the fuel to the stacks. This increased the heat capacity of the fuel inlet, lessening the heat loss impact on the system.

The stacks used in the SCDP plant represent a 1993 design, and incorporate cell hardware which is less tolerant of extreme thermal gradients than the hardware currently being used at ERC. The advanced hardware which is currently being tested in full area short-stacks is designed to tolerate thermal gradients significantly beyond those seen during the off-design operating periods in the SCDP plant. Testing to date on the new hardware has verified this capability, which was a major objective of the new design.

Once the process adjustment was made to limit the thermal gradient across the stacks, the balance of the third operating period was characterized by stable operation at 50% to 75% power for extended time periods. There were three short BOP-related shutdowns in this period. One was caused by a short circuit in a power cord in the control room and two were caused by the buildup of debris in the HRU. The HRU includes a catalyst block for oxidation of VOCs during the initial power plant heatup (this initial conditioning of the fuel cell stacks was done in the field at Santa Clara but is expected to be a factory operation in commercial stack production). This catalyst block began to collect deposits related to the use of unclean pipeline natural gas in the startup burner firing into the system exhaust. Changes in the commercial BOP configuration and stack designs will preclude this from recurring. These three outages were the *only* BOP related outages in the entire test program since the initial startup.

The few outages that did occur in the third operating period provided the opportunity to evaluate one of the SCDP's key performance criteria, ramp rate. The criteria to be demonstrated was a ramp rate of standby to full power in 30 minutes, a rate of 3.3% per minute. Seven automatically controlled power ramps were performed in the third operating period, the fastest of which was at a rate of 4.8% power per minute. The stable operations observed in the third operating period also provided the opportunity to evaluate other design criteria, such as noise level, power quality, and emissions. The testing of the reconfigured power plant was continued through March 3, 1997, when the testing operations were concluded. The plant was operated in grid-connected mode for a total of 4000 hours. Following the completion of the test program, the fuel cell stack submodules were shipped back to FCFC's Torrington, CT facility for post-test analysis, which is now ongoing.

In addition to meeting and exceeding its rated power criteria and ramping criteria, the power plant operations met many other key project objectives. Specific project criteria which were successfully demonstrated included rated output, peak operation, voltage harmonic power quality, low NO<sub>x</sub> and SO<sub>x</sub> emissions, and operation within noise limits. A summary of the power plant performance against key project criteria is shown in Table 1. The SCDP balance of plant proved to be exceptionally reliable. The system rode through minor grid disturbances and responded to major grid problems

exactly as intended. The overall availability of the balance of plant during the test program was 99%, and the entire test program (including the BOP pre-test) was carried out with an excellent safety record, with no lost-time accidents. These would be excellent results for any plant, but are particularly impressive in this first-of-a kind demonstration plant.

**Table 1**  
**SCDP Power Plant Performance vs Key Project Criteria**  
**Almost All of the Key Project Criteria Were Met in the Demonstration Program**

Power Output	1.8 MW Rated Power Target Exceeded with Power Output up to 1.93 MW.
Heat Rate	Stack Performance Level Necessary for Target Heat Rate Achieved.  < 7000 Btu/kWh (48.7% Efficiency) Target Not Achieved due to Conservative Approach To Operating the Plant in the Early Phase Of Testing, and Process Impacts in Reconfigured Plant. Minimum Heat Rate of 7820 Btu/kWh (43.6% Efficiency) Achieved.
Power Quality	Voltage Harmonics Less Than Half of IEEE 519 5% Distortion Level. Overall Current Harmonics Below IEEE 519 Level, With Four Individual Harmonics Above Level. Correctable With Additional PCU Tuning.
Ramp Rate	3.3% Power per Minute Target Rate Exceeded  Maximum Ramp Rate Tested was 4.8% per Minute
Emissions	SOx Emissions Level Undetectable  NOx Emissions Level Undetectable Upstream of Startup Burner, only 2ppm at System Exhaust Downstream of Burner
Noise	Met SCDP and City of Santa Clara Requirements  <60 dB(A) 100 feet from Equipment and <70 dB(A) at Property Line

Beyond its success at demonstrating many of the key project criteria, the test program was also invaluable as a learning process for the DFC development team. The project represents the first time that global power plant type issues (e.g. heat loss, system control, grid interface, multi-stack operation, etc.) have been dealt with for carbonate fuel cells in the field and at the megawatt scale. The advantages of the simplified BOP design utilized in ERC's DFC concept were clearly demonstrated by such results as the excellent BOP reliability. The limitations of the design (e.g. the impact of heat loss on the stack temperature gradient in early operations of the 8-stack system) were successfully faced and resolved (e.g. by increasing steam content to minimize heat loss impacts). In terms of both capabilities demonstrated and lessons learned, the test program has been an important advancement toward the commercialization of the Direct Fuel Cell. The accomplishments of this project were recognized through an APPA Energy Innovator Award and an EPRI Technical Achievement Award. A summary of the overall program accomplishments is shown in Table 2.

**Table 2**  
**Key SCDP Accomplishments**

<b>Permitting/Construction</b>	<p>Ease of Permitting for Direct Fuel Cell (DFC) Powerplant Demonstrated</p> <p>Demonstrated DFC Construction Approach</p> <p>BOP Configured with Vendor Supplied Skid Mounted Modules and Shop Fabricated Piping Spools</p> <p>DC Power Block with Multi-Stack Submodules Truck Shipped Across U.S.</p> <p>Engineering, Procurement and Construction Completed Within 0.1% of 1993 Budget Forecast.</p>
<b>Power Plant Start-Up</b>	<p>Start-Up of Multiple-Stack System in the Field Demonstrated</p> <p>Automatic Control of Start-Up Parameters Demonstrated</p>
<b>Stack Operation</b>	Uniform Performance of DFC Stacks Above Target Performance Level Achieved
<b>Power Output</b>	<p>Power Output at and Above 1.8 MW Rated Power Level, up to 1.93 MW Net AC</p> <p>1710 MWh Delivered to City of Santa Clara Grid</p> <p>Largest Fuel Cell Powerplant Operated in Western Hemisphere</p> <p>Largest DFC Powerplant Operated in the World</p>
<b>Power Quality</b>	<p>Voltage Harmonics Less Than Half IEEE 519 5% Distortion Limit</p> <p>Overall Current Harmonics Below IEEE 519 Level, with Four Individual Harmonics Above Level. Correctable with PCU Retuning .</p>
<b>BOP Operation</b>	<p>Automatic Control of BOP Equipment and Power Level Demonstrated</p> <p>99% BOP Availability</p> <p>Plant Staffed by Locally Hired Power Plant Operators</p> <p>No Nuisance Trips. Plant rode through minor grid disturbances and responded to major grid disturbances exactly as intended</p>
<b>DC Power Module</b>	<p>Fuel Cell Stacks Exhibited Uniform Performance, in Excess of Design Projections</p> <p>Design and Electrical Configuration Issues Continue to be Addressed in Ongoing Development Program, along with Thermal Cycle and Durability Issues</p>
<b>Learning</b>	<p>Operation of Multiple Stacks in Common Flow System and Electrical Circuit Provided Insight into Multiple Stack Power Plant Design Issues</p> <p>Experience Gained in DFC Power Plant Operation</p> <p>Dynamic Response of Stacks and BOP Equipment During Transients and Load Ramps Provided Insight Into Design of Commercial Power Plant.</p>

## **Future Activities**

As noted above, the fuel cell stacks have been returned to FCMC's facility in Torrington, Connecticut, where a process of post test analysis is now ongoing. ERC, FCMC, and FCE personnel are participating in the post test program, which will be complete later this year, and reported on at the next conference.

## **Acknowledgments**

The guidance and support of the FETC Contracting Officer's Representative, Mr. Thomas J. George, has been greatly appreciated.

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## **M-C Power Commercialization Program Overview**

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### **Introduction:**

Since its incorporation in 1987, M-C Power has focused on development and integration of molten carbonate fuel cell technology by combining individual cells into cost-effective stacks, which are then integrated into a power plant system utilizing commercially available balance of plant equipment. Demonstration testing of cogeneration power plant concepts that generate electric power and steam in practical applications at the desired capacity level are an integral part of our commercialization strategy.

We have successfully operated increasingly larger fuel cell stacks while improving power output and cell life. Development activities include the improvement of component performance, endurance tests, and cost reduction methods. This work represents an important milestone in M-C Power's commercialization program and has culminated in the design, construction, and testing of a fully integrated 250-kW molten carbonate fuel cell (MCFC) power plant at the Naval Air Station (NAS) Miramar in San Diego, California.

The purpose of the Product Development Test (PDT) at the NAS Miramar was to demonstrate the internally manifolded heat exchanger (IMHEX<sup>®</sup>) MCFC technology in a thermally integrated power plant. This project has provided the data required to finalize the commercial design for M-C Power's market entry product. The findings from the PDT project are currently being incorporated into our Product Design and Improvement (PDI) project. This paper addresses the importance of the PDI program in commercializing MCFC technology by the year 2001 and M-C Power's approach to achieving that goal.

### **Objective:**

The objective of the PDI project is to establish commercial readiness of M-C Power's MCFC system for distributed generation and other applications. To date, work performed under this project has focused on quantifying the market potential for MCFC technology, recognizing and addressing the requirements of the customer base, and resolving cost reduction and technical issues affecting the marketability, performance, and reliability of MCFC power plant systems. M-C Power's ongoing commercialization program is enhanced by recent development activities and market studies.

Research sponsored by the U.S. Department of Energy and administered by the Federal Energy Technology Center, under contract DE-FC21-95MC30133 with M-C Power Corporation, 8040 S. Madison Street- Burr Ridge, IL 60521; (630) 986-8040. Dr. Diane T. Hooie is the Contracting Officer's Representative.

When M-C Power was founded, its initial mission was to develop, produce, and commercialize a high efficiency, environmentally benign energy conversion device based on a molten carbonate fuel cell utilizing the IMHEX® design. Today, our corporate goal reflects the many accomplishments we've made since our founding. We plan to enter the market with nominal 250-kW and 1-MW flexible and modular power plant products. We based this decision upon several factors which include market studies completed in the past year; input from members of the Alliance to Commercialize Carbonate Technology (ACCT), which includes their views of the future in a deregulated marketplace; analysis and recommendations of the commercialization team members; and the results of the demonstration test at the NAS Miramar. A great deal of the technology development work has been completed. The remaining barriers have been identified and a strategy has been implemented. We have production capabilities established—and we've set our sights on accomplishing the full scale commercialization of this technology.

## **Approach**

The approach for developing this technology was defined early on when M-C Power established a commercialization team in order to succeed at meeting its mission. The commercialization team is comprised of major players in the fields of energy research, power plant design, packaging, distribution, and maintenance. It is led by M-C Power and is balanced out by Stewart & Stevenson Services, Bechtel Corporation, and the Institute of Gas Technology. This team provides the expertise needed to develop and commercialize a cost-effective power plant based on M-C Power's molten carbonate fuel cell MCFC stack technology.

Although M-C Power is managing the commercialization and development of the MCFC technology, the contributions of its supporting team members and their significance can't be overlooked. The Institute of Gas Technology is responsible for component development to increase cell performance and endurance. M-C Power then takes this technology and scales it up to commercial size utilizing its proven manufacturing processes. Bechtel Corporation provides the process design for the overall plant and the detailed design of the systems needed to support the fuel cell stacks and form a completely integrated operational power plant. Stewart & Stevenson Services receives the specified equipment, packages it on transportable skids that are completed with piping, instrumentation, and wiring. The skids are then factory tested to verify performance and operational characteristics.

Verification tests and power plant efforts within the Product Design and Improvement project are directed toward the construction and operation of a prototype power plant system. The system is intended to reflect all of the design and operational features of the market entry product. The commercialization team has developed a strategic approach to implementation of the project. This approach capitalizes on past development projects and enables the team to accelerate its efforts to

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satisfy our commercialization objective through the successful design, manufacture, field test, and evaluation of the prototype power plant.

## **Project Description**

The commercialization team has chartered the best course of action in order to achieve the highest probability of success. The performance of market assessments and product definition activities was the first order of business under this project. They are discussed in further detail in the results portion of this paper. They have led to a framework from which the system design and analysis tasks could proceed in a timely fashion. Bechtel had previously selected a baseline system which they have been using to perform trade-off studies. The results of these studies are discussed in the following section.

The other major areas covered under the PDI program include the optimization of the manufacturing processes developed in an earlier program and verification of advanced component concepts. Optimization and automation of active cell component manufacturing processes, consolidation of separator plate manufacturing, and the upgrading of our QA/QC capabilities are all serving to enhance our ability to produce quality stacks for prototype power demonstrations.

Through the identification, qualification, and implementation of advanced component formulations and manufacturing processes, we are reducing the cost of producing the cells that comprise the fuel cell stack. We have been developing, verifying, and improving upon the critical components and subsystems required in a market responsive product. Efforts are focusing on the cell package, stack module, and balance-of-plant components.

Future testing will be carried out using existing facilities. Modification will be made to these facilities as deemed appropriate by the commercialization team and its sponsors. Tests are being performed to verify advanced engineering designs and component technologies. Balance of plant components have been identified and will be qualified through strategic vendor alliances. These verification efforts will be focusing on the turbogenerator, recycle blower, and power conditioning units. All subsystems and BOP components will be accepted only after rigorous factory endurance verification. Fuel processing is addressing a variety of alternate fuels.

## **Results**

The results of the PDI program to date have contributed to a better understanding of the markets for MCFC technology. Those results are discussed further in the application section of this paper.

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Market analysis completed over the past year has led to a detailed product definition and realignment of our program strategy to reflect the dynamics of a changing market.

Bechtel has been charged with the task of developing simple and cost effective concepts for nominal 250-kW and 1-MW class power plants. The decision to further explore the opportunities of a power plant with the capacity of 250-kW has been purely market driven. The members of ACCT and other industry players have expressed their interest in a power plant of this size, and we have responded. From early on they have conveyed the importance of reliability, durability and cost. We have been working to address those issues from the onset of our development program.

The path to allowable installed power plant cost (stack and balance of plant) has been established. Stack costs are being reduced by elimination and integration of processing steps, design simplification, and the use of low cost raw materials. Many of these cost reduction approaches are currently being tested at the 100 cm<sup>2</sup> single cell scale and in 1000 cm<sup>2</sup> stacks. The cost of the balance of plant is being reduced by compaction, component elimination, and system simplification.

## **Applications**

More recently, the markets for this technology have been driven by the issues surrounding deregulation and competition in the electric industry. Some of the issues driving the market for fuel cell technology include retaining loads through better customer service, environmental regulations, power quality, reliability, constrained capital for utility transmission and distribution (T&D) investments, and distributed vs. centralized power generation. Our market analysis gives us a clearer understanding of these issues and their impact on the PDI program.

The target market within the United States can be segmented into four main areas. They are commercial applications, light industrial applications, distributed power, and several special niche applications. The commercial segment includes small to medium sized hospitals, hotels schools, and shopping malls. For these applications our technology would serve as a co-generation unit; supplying heat and electricity for cooling and illumination. The light industrial segment includes, but is not limited to, the chemical, paper, metal, food, and plastics industries. In this application, the MCFC would serve as a co-generator of electrical power and high quality steam to serve the respective industrial processes.

The customers for the distributed power segment would include the traditional utilities and their unregulated subsidiaries. The main driver behind this application is the avoidance or deferral of T&D costs. The MCFC would alleviate energy losses within the system, supply reactive power support, and defer the construction of costlier system upgrades.

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The special niche segment includes applications such as computer centers that require premium power quality and high reliability. Also within this segment are producers of opportunity fuels such as landfills, waste water treatment plants, and refineries. In these applications the MCFC will help them to lower current power costs by using available waste streams as fuel.

## **Future Activities**

M-C Power and the commercialization team will continue with the tasks outlined under the PDI program. The most important element in the near term is to integrate and verify that the balance of plant components can perform reliably. Concurrently, we will expand upon our accomplishments in the area of cell performance and cost reduction.

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# **NAS Miramar Molten Carbonate Fuel Cell Demonstration Status**

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## **Introduction**

There is a need to demonstrate Molten Carbonate Fuel Cell (MCFC) power plant systems in order to establish a cost and performance track record for the emerging MCFC technology. It is intended that the MCFC demonstration power plant would provide a configuration which supports the demonstration of a M-C Power internally manifolded (IMHEX®) 250 kW MCFC stack. The demonstration provides an ideal vehicle for developing utility and other end-user support for the technology which would provide the confidence necessary to encourage the purchase of MCFC power plants. The demonstration incorporates full-scale and full-height stack and balance of plant (BOP) system testing. Testing goals are consistent with M-C Power's commercialization plan and with product definition requirements identified through extensive market research. The target capacity range for commercial plants is from 250 kW to 1 MW and higher which makes the power plant size well suited for a wide range of on-site and distributed power applications that are expected to grow in importance over the coming decade.

## **Objective**

The objective of this Cooperative Agreement is to support the Product Development Test (PDT) of an M-C Power full-area, full-height 250 kW MCFC stack in order to accelerate commercialization of the technology. Another objective is to demonstrate the M-C Power team's capability to design, manufacture, assemble and test MCFC power plants which are close to a commercial configuration. By definition, the 250 kW NAS Miramar molten carbonate fuel cell demonstration is an integrated power plant which is consistent with M-C Power's commercial development plan.

## **Approach**

The project has been developed in accordance with DOE guidelines under the Cooperative Agreement established between M-C Power and the DOE. DOE funded the design, manufacturing and fabrication of the 250 kW stack which consisted of 250 cells with an active area of 10,800 cm<sup>2</sup> per cell. All costs associated with the necessary BOP were funded by the Gas

Research Institute (GRI). Funds for testing of the demonstration power plant were provided under tailored collaboration with the Electric Power Research Institute (EPRI). The DOD's Advanced Research Projects Agency (ARPA) has funded the site relocation to Miramar and a 20 kW stack test. Additional funding was supplied by Bechtel Corporation, Pennsylvania Power & Light, San Diego Gas & Electric (SDG&E), Southern California Gas (SoCalGas), Stewart & Stevenson Services, Inc. (S&S) and Sydkraft AB of Sweden.

M-C Power managed the project. Bechtel was responsible for the design, detailed engineering, construction management and plant start-up. S&S fabricated the balance of plant (BOP) in their shop on a skid and tested the performance prior to shipping to the site. SDG&E performed site construction work with its own staff which was supplemented by craft labor under subcontracts with them. The project was executed over the period September 30, 1992 through March 31, 1997. M-C Power operated the plant with its own funding and utility support 2 1/2 months beyond the March 31, 1997 Cooperative Agreement end date.

## **Project Description**

The project includes the design, procurement, construction, start-up and operation of the world's first 250 kW cogeneration MCFC power plant. The demonstration plant is designed to produce 250 kW net of electricity as well as 340 - 640 pounds per hour of 100 psig steam for use in NAS Miramar's district heating system. It is located on the NAS Miramar military base which is about 15 miles northeast of downtown San Diego. The plant, occupying a 40 x 80 square foot area, has three main components: a fuel processor (reformer), a BOP skid and the fuel cell stack. Auxiliary equipment includes a nitrogen storage tank and vaporizer, and a boiler feed water make-up system which are separate from the BOP skid. In the market entry plant, this auxiliary equipment will be either eliminated or included within the BOP skid. The electrical equipment and control system are located in a separate building. The electrical and control systems are being reduced to cabinet size in the market entry design.

## **Results**

### Construction Schedule

Site preparation and civil construction were completed in November 1995 which was six months after the construction start date. The stack was shipped to the site in August of 1996. The plant was mechanically complete the following month and electrical acceptance took place on October 15, 1996 as shown in Figure 1 (NAS Miramar Operating Schedule). Process and Control (PAC) testing started in September 1996 and was completed in December.

### Fuel Cell Stack

Between April and July 1996 the final assembly, conditioning and acceptance testing of the stack took place. The stack was conditioned in the Acceptance Test Facility (ATF) and tested at 400 to 425 amps load. Conditioning and testing lasted a total of 593 operating hours. Binder removal was conducted for approximately 247 hours. Electrolyte melting was completed in 162

hours. Throughout the electrolyte melting, the gas manifold pressures remained positive indicating excellent sealing. Cathode oxidation was completed in 39 hours.

### Acceptance Testing

During acceptance testing the stack generated power for 210 hours. The maximum load current obtained during this testing was 546 amps with the maximum power output of 107 kW. The stack operated at a steady state load of approximately 400 amps for 164 hours with the output power of 90 kW. During operation the stack met the stated acceptance criteria: > 207.5 VDC at 40 mA/cm<sup>2</sup>. No apparent cell package voltage decay was observed during operation. Final stack cooling after acceptance testing was completed in 160 hours. Ambient temperature crossover testing before and after stack conditioning met the acceptance criteria of less than 0.4 slpm/cell gas leakage at a stack clamping force of 40 psi and differential pressure of 10" WC.

### Plant Performance

The demonstration project first produced power on January 24, 1997. In addition to an impressive list of technology successes and the verification of key operating systems, the power plant produced 158 megawatt hours of electricity and 296,000 pounds of steam. This was the first time any molten carbonate fuel cell power plant has been used for cogeneration, providing both electricity and high quality steam in a utility grid application. The quality of electricity and steam produced was excellent. Transitions between being on and off-line were smooth. These transitions were completely unnoticeable to power and steam users in the local NAS Miramar systems.

Figure 2 (NAS Miramar 250-kW Stack - Stack DC Cumulative Power) presents the electricity production history of the demonstration plant. The slow initial power production during the first 900 hours corresponds to the time when instrumentation electrical interference issues were being resolved. At this time the inverter was taken off line. Between 900 hours and 1900 hours of operation took place utilizing a load device. At 1250, 1450 and 1650 hours the stack exhibited off load operation due to malfunction of rotating equipment. At approximately 1900 hours through 2300 hours the inverter was operational when rotating equipment again malfunctioned.

Figure 3 (NAS Miramar Stack Performance - OCV) shows excellent cell to cell uniformity. The average voltage is shown in the left bar. There is excellent correlation between the OCV's in the demonstration stack and with small-height scale tests in the laboratory even with the scale up factor of 12.5 times. Bench scale tests were run in parallel for diagnostic purposes.

Figure 4 (NAS Miramar Stack Performance - 1140 ADC, 206 kW) shows excellent voltage distribution. This uniformity is at 66% of design load and at actual power plant conditions (low CO<sub>2</sub>/O<sub>2</sub>) concentration. The upward trend is due to temperature gradient from the bottom to the top of the stack. Voltage uniformity also indicates an above average gas distribution demonstrating that hydrodynamic design is under control.

Figure 5 (250 kW Miramar Stack Actual versus Predicted Performance) is a voltage comparison between the electrochemical model predictions and actual voltage performance. The difference between predictions and actual performance achieved is less than 8%. Even though results are extremely good, further enhancements of the model to predict low oxidant conditions are under development.

There was superior stack voltage distribution throughout the stack and once again confirmed no evidence of electrolyte movement. All internal seals held over a wide range of test pressures which verified that they would hold under adverse operating conditions. The plant easily moved between standby and on-load operations without the use of supplemental CO<sub>2</sub>.

The DOE Cooperative Agreement ended on March 31, 1997, but M-C Power chose to operate the plant for an additional 2 1/2 months. With limited additional funds, the decision was made to shut down the plant. The stack was removed and transported to M-C Power's manufacturing and testing facilities where it will be analyzed.

The demonstration project met its test objective by:

- Demonstrating major equipment/system performance in an actual end-user application using commercial operating parameters and fuel conditions.
- Proving that the technology can operate in a distributed generation mode.
- Confirmed that the plant can start up and operate with standard electric utility operating personnel.
- Proved that there is no electrolyte migration in the stack.

There are some enhancements to the BOP and stack which will move the technology closer to commercialization. On the BOP side these improvements include better performance from the hot gas blower and turbocharger. New and improved equipment are ready for installation and testing. In future operations, stack performance will be improved by a new plate design which will incorporate 310 SS instead of 316 SS for better corrosion resistance.

## Application

Plant startup and operating experience has amply demonstrated that M-C Power's MCFC stack can be integrated effectively into a power plant configuration. The stack design has shown impressive stability and resiliency under adverse operating conditions as has the new reformer. Design and operating experience is already being used to further improve the reliability and operability of the next generation of power plants which are currently being designed. Plant operating experience has revealed that BOP equipment specifications should be relaxed to provide improved power plant operating flexibility. Major equipment and systems performance has shown that simpler and cheaper designs are possible for the market entry power plants. An elaborate instrumentation and control system at NAS Miramar has provided data which will further

contribute to simple and reliable commercial designs. The demonstration has also proven that commercial cost targets are achievable.

## **Future Activities**

The NAS Miramar Demonstration Plant is currently in a standby mode of operation. Plans are being developed to use the power plant as a test facility for MCFC stacks which incorporate advanced components. These components, already proven in the laboratory, will be included in the early market entry power plants. The Miramar plant will also be used to test improved BOP equipment and control systems which meet commercial operating specifications and cost goals.

## **Acknowledgments**

In addition to the funding organizations and project participants mentioned above, the author would like to acknowledge IHI for their technical assistance and NAS Miramar for their use of the demonstration site and continuing support of the demonstration. Dr. Diane Traub Hooie and Dr. Mark Williams of the U.S. Department of Energy's Federal Energy Technology Center were most helpful in providing guidance and independent progress assessment throughout the Cooperative Agreement. A special thanks to Mr. Al Figueroa, San Diego Gas & Electric Project Manager, Narasimha Kudlu, Bechtel Project Manager and Tom Robertson S&S Project Manager. Together they worked as a very efficient team. Their work resulted in a plant that is well designed with high quality construction and able to operate in accordance within specifications.

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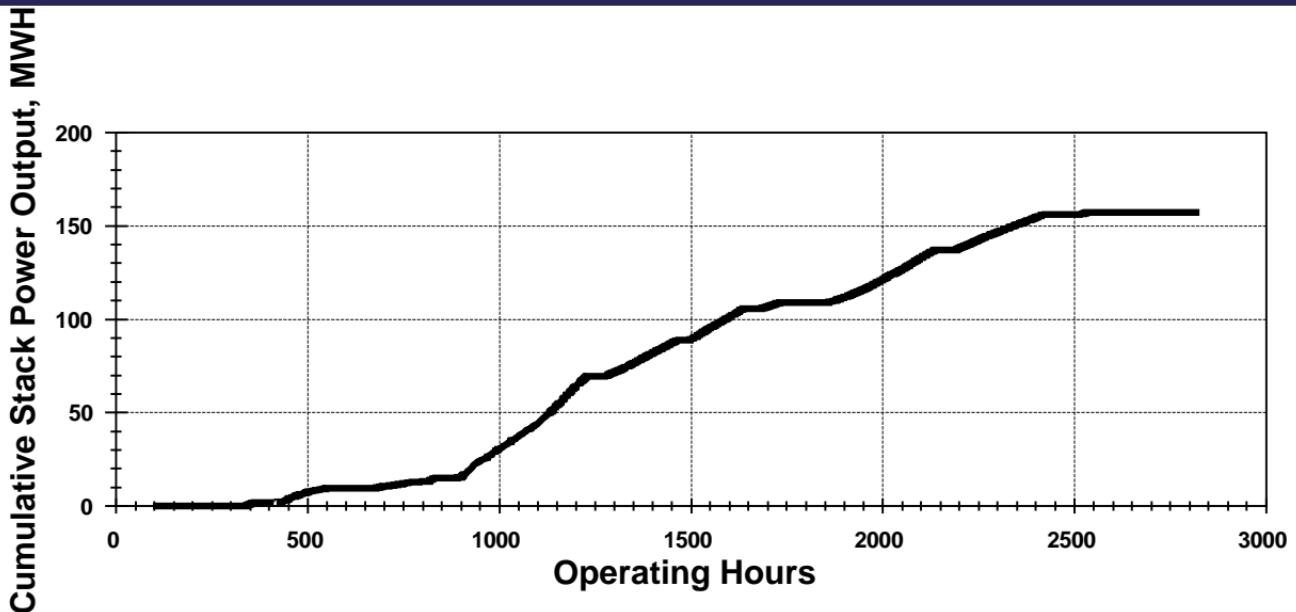
The Molten Carbonate Fuel Cell (MCFC) Product Development Test project is a Cooperative Agreement (DE-FC21-92MC28065) between the U.S. Department of Energy and M-C Power Corporation, 8040 S. Madison Street, Burr Ridge, IL 60521; phone (630) 986-8040; fax (630) 986-8153. The project was administered by the Federal Energy Technology Center (FETC). Dr. Diane Traub Hooie is the FETC Contracting Officer's Representative (COR). The project was executed over the period September 30, 1992 through March 31, 1997. Subcontractors included San Diego Gas & Electric and Bechtel Corporation. The Institute of Gas Technology (IGT) and Stewart & Stevenson Services, Inc. (S&S) performed work under a Gas Research Institute (GRI) subcontract which was cost shared into the project. Under the Cooperative Agreement with DOE. The U.S. Advanced Research Projects Agency (ARPA) provided financial assistance.

**Figure 1**

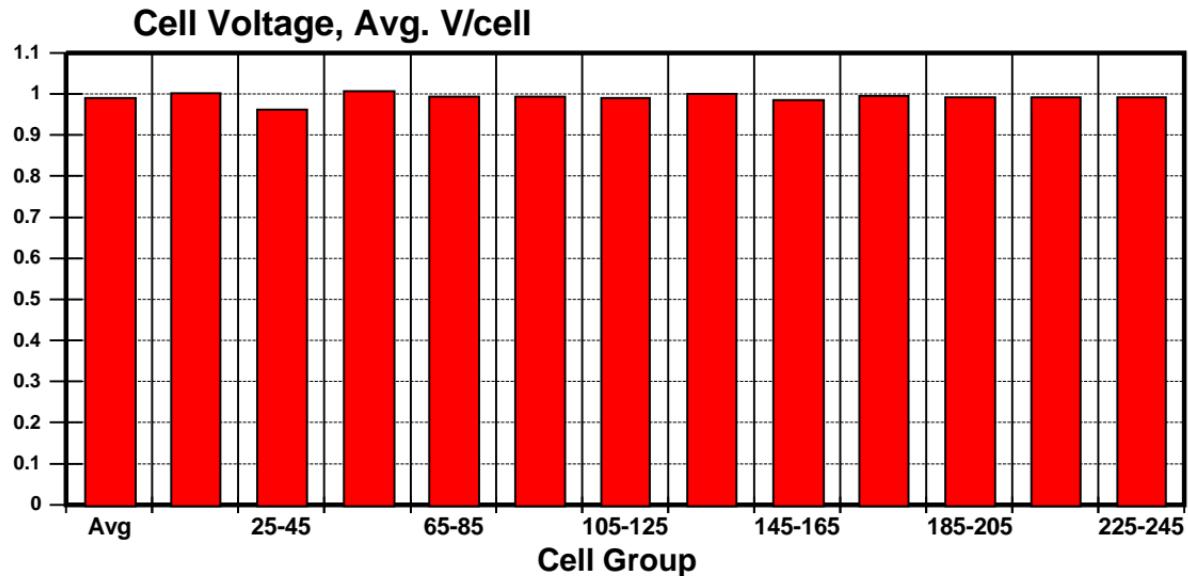
# NAS Miramar Operating Schedule

# Fig. 2 NAS Miramar 250-kW Stack

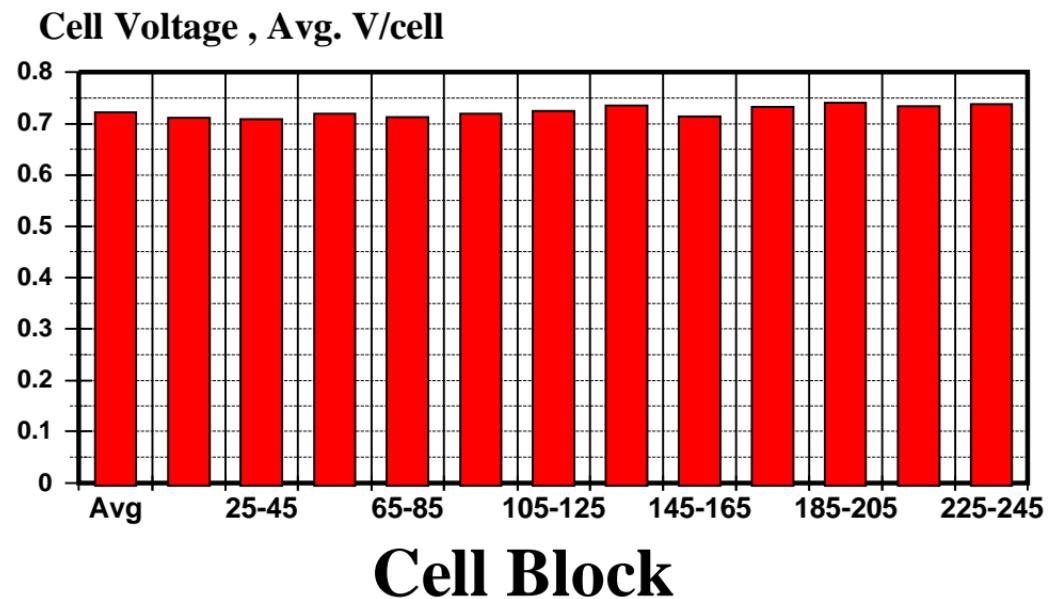
Stack DC Cumulative Power



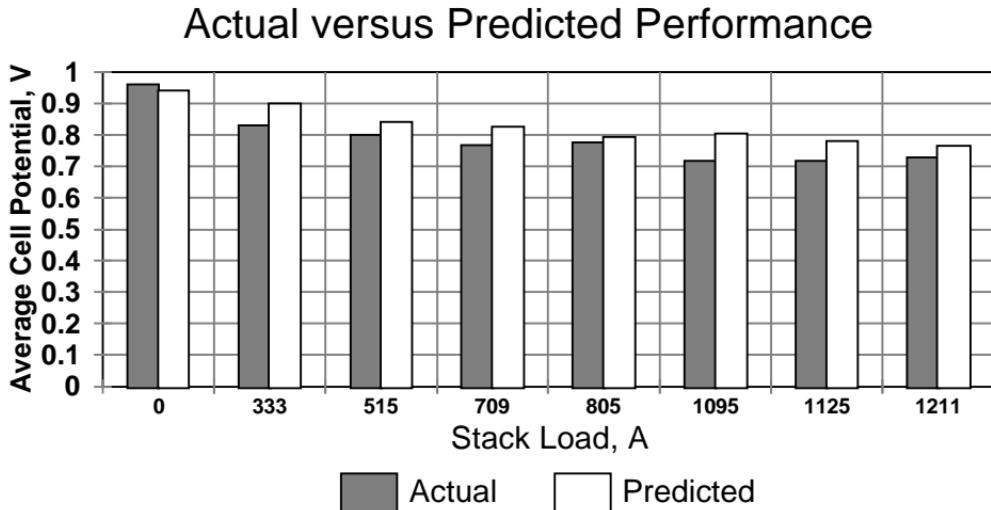
# Fig. 3 NAS Miramar Stack Performance - OCV



# Fig. 4 NAS Miramar Stack Performance-- 1140 ADC, 206 kW



# Fig. 5 250 kW Miramar Stack



# Westinghouse Program Overview

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## Introduction

The Westinghouse Solid Oxide Fuel Cell Development Program continues to make steady progress towards commercialization. The vision of a cost-competitive, ultra-high efficiency, environmentally friendly SOFC power generation product line for distributed power and cogeneration applications is nearing a reality. This vision is centered around Pressurized SOFC/Gas Turbine (PSOFC/GT) combined cycle power generation systems in the hundreds of kW<sub>e</sub>'s to tens of MW<sub>e</sub>'s power range, which are capable of producing electricity at efficiencies between 62% and 72% depending on size, configuration and equipment selection. This paper summarizes 1) our important technical accomplishments over the past few years, 2) our final phase development program covering the time-frame 1997 to 2001, and 3) our vision of a commercial enterprise structure including partners.

## Objectives

The objective of the Westinghouse SOFC Development Program is to complete the development of atmospheric SOFC power systems and PSOFC/GT power systems for cogeneration and distributed power applications, and initiate commercial manufacture of such systems by 2001.

## Approach

The Westinghouse SOFC Development Program is multi-faceted and consists of the following major tasks: cell cost reduction, module scale-up and cost reduction, system (balance-of-plant) scale-up and cost reduction, field unit program, and commercial manufacturing facility design and construction. This final phase development program will cover the time span 1997-2002. In order to secure non-Federal funds for cost share obligations and for construction of the commercial manufacturing facility, and to selectively augment the existing skills and capabilities in such areas as ceramic manufacturing, automation, gas turbine technology, market access and distribution channels, etc., Westinghouse has been negotiating with a number of other corporations concerning the formation of a Joint Venture for the purpose of commercializing SOFC.

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Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-FC21-91MC28055 with Westinghouse Electric Corporation, Science and Technology Center, 1310 Beulah Road, Pittsburgh, PA 15235-5098; telefax: 412-256-2012.

## Project Description

The cell development tasks are focused on developing lower cost materials and ceramic manufacturing processes for our commercial size (2.2 cm O.D., 150 cm active length) air electrode supported cell. The nominal material composition of each cell component has been fixed, and the evaluation of lower grade (i.e., lower purity) raw materials is in progress. Over the last several years, Westinghouse has made significant progress in developing lower cost manufacturing processes; plasma spray of the interconnection has replaced the more expensive electrochemical vapor deposition (EVD) process in production and fuel electrode slurry dipping/sintering intended to replace fuel electrode slurry spray/EVD has been successfully developed in the laboratory. Implementation in production is expected by mid-1998.

The module development tasks are focused on scale-up, pressurization, and cost reduction. A cell stack, consisting of 1152, commercial size, air electrode supported cells with integral stack reformers and having a maximum power output at 1 atmosphere pressure of 170 kWe DC, has been fully designed and built for the EDB/ELSAM Field Unit described below. The above stack design will form the basis of the stack designs in our field unit program described below, and in our commercial modules.

The field unit program consists of 3 field unit tests: 1) the EDB/ELSAM (Dutch/Danish utilities) 100 kWe, atmospheric cogeneration system scheduled for start of site testing in December 1997, 2) the SCE 250 kWe, PSOFC/GT combined cycle system scheduled for start of site testing in January 1999, and 3) the MWe-Class PSOFC/GT system at EPA's Fort Meade, MD laboratory scheduled for start of site testing in 2000. The first field unit will demonstrate the performance of the commercial size cell in large quantities and the new stack components including the stack reformers. The second field unit is a first-of-a-kind test demonstrating the coupling of a PSOFC module and a gas turbine at 3-4 atmospheres pressure. Finally, the third field unit is a demonstration of a multi-module commercial prototype system. Both PSOFC/GT field units are expected to have an electrical efficiency in the 60-65% range. In addition, a task to upgrade the MWe-class system to a 70% efficient system is included in our field unit program.

Assuming the success of the cost reduction tasks and field unit program, a decision to build the first commercial production line will be made around mid-1999. Construction is estimated to take about 2 years. Thus commissioning of the commercial manufacturing facility is expected by mid-2001.

## Accomplishments

The SOFC Power Generation organization is proud of and encouraged by its technical accomplishments over the past few years. The major accomplishments include:

- Over 13,000 hours (1½ years) of power operation for a 25 kWe SOFC power generation system with approximately 0.1% voltage degradation per 1000 hours of operation,
- Nearly 7½ years of power operation for two cells with a voltage degradation rate of about 0.5% per 1000 hours,
- Successful development of our air electrode supported cell, and successful scale-up from a 50 cm active length cell (65 watts per cell at 1 atmosphere) to a 150 cm active length, larger diameter cell (210 watts per cell at 1 atmosphere),
- Construction, commissioning, and operation of a 4 MWe per year Pilot Manufacturing Facility,
- Completion of the manufacturing of the 100 kWe (150 kWe max.) cogeneration power system for EDB/ELSAM (a Dutch and Danish Utility Consortium), and
- Successful pressurized cell testing up to 15 atmospheres in collaboration with Ontario Hydro achieving a SOFC cell power output record of 280 watts.

## **Benefits**

This program is expected to culminate in a MWe-class product offering having an electrical efficiency of at least 63% on natural gas, an NO<sub>x</sub> emission of  $\leq 2$  ppmV, and a total installed cost of \$1500/kWe or less.

## **Acknowledgments**

Westinghouse acknowledges the guidance and assistance of Mr. William C. Smith, Project Manager, Gas Power Systems Division, US-DOE-FETC, in the course of this Cooperative Agreement spanning the period from December 1, 1990 through November 30, 1997.

# Westinghouse SOFC Field Unit Status

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## Introduction

Fuel cells are without question the most non-polluting of all fuel consuming power generation technologies. Intrinsically, the potential for NOx generation is virtually non-existent because fuel oxidation occurs electrochemically rather than by combustion, and in the case of the solid oxide fuel cell (SOFC), isolated from contact with atmospheric nitrogen. In general, sulfur must be removed from the fuel prior to its oxidation in a fuel cell, thus there is no potential for the emission of SO<sub>2</sub>, the precursor for acid rain. In addition, across the range of power generation capacity levels, a fuel cell power system can generate electricity more efficiently than any other fossil fuel consuming electric power generator. Since power generation at the highest practical efficiency serves to minimize the emission of carbon dioxide while conserving fuel, fuel cells represent the best way to ameliorate concern over the “green house effect” and dwindling or politically insecure sources of fuel. Among fuel cell types, only the SOFC has the recognized potential to achieve power generation efficiencies in excess of 70% using a hybrid cycle that is both simple and dry, the SOFC/Gas Turbine. Further SOFC development is needed, however, in order to achieve commercially competitive cell and stack cost and to demonstrate SOFC power systems at commercially viable capacity levels. In the following will be summarized the status of the Westinghouse SOFC field unit program and its contribution to an improved prospect for SOFC commercialization.

## Objective

The objectives of the Westinghouse experimental SOFC field unit program are: the development of a viable SOFC electrical power generation system that meets customer needs; the first-hand demonstration to customers of the beneficial attributes of the SOFC; the exposure of deficiencies through experience in order to guide continued development; and the garnering of real world feedback and data concerning not only cell and stack parameters, but also transportation, installation, permitting and licensing, start-up and shut-down, system alarming, fault detection, fault response, and operator interaction.

## Approach

Westinghouse has practiced the deployment with customers of fully integrated, automatically controlled, packaged solid oxide fuel cell power generation systems for over ten years. These 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 field units has been funded with customer and Westinghouse moneys. DOE has participated in these programs by providing the cells and in part, the stacks or generator modules. Field units have been operated at customer expense and in most cases solely by the customer, with data shared with Westinghouse and DOE. Cells and generator modules are returned to Westinghouse at the completion of the customer test program.

## Project Description

A simplified process flow diagram for an atmospheric pressure SOFC cogeneration system is shown in Figure 1. A motor driven blower draws ambient air through a filter and compresses it to a pressure of the order of 1000 mm water column. The process air then flows through an exhaust gas heated recuperator where its temperature is increased to approximately 500°C. From the recuperator, the air flows through an air heater and then into the SOFC generator module. The air heater in field units to date is electrically powered and is used only during start-up and during periods of low power operation when needed to maintain the SOFC at set point temperature, nominally 1000°C. Natural gas at a pressure of nominally three atmospheres gauge is desulfurized and routed to the SOFC generator module through a flow control device. Within the SOFC generator module, the natural gas is reformed to hydrogen and carbon monoxide and subsequently electrochemically oxidized generating dc power. Typically, the SOFC operates at 85% electrochemical fuel utilization. In the SOFC, the electrochemical oxidation of the fuel occurs with complete isolation from nitrogen, therefore with no potential for NOx formation. Depleted fuel is burned completely in a combustion zone within the generator module where it preheats incoming air. Exhaust gas exits from the SOFC generator module with in-stack radiantly heated reformers at a temperature of approximately 850°C. In earlier 25 kW class field units with a convectively heated reformer, the exhaust gas exited at a temperature of approximately 700°C. The exhaust gas is routed through a recuperator, which may consist of a high temperature section, a heat recovery steam generator, and a low temperature section, followed by an exhaust gas heated water heater.

The Joint Gas Utilities (JGU), a consortium of the Tokyo Gas Company and the Osaka Gas Company, sponsored in 1992 a 25 kW SOFC cogeneration system that utilized two SOFC generator modules in a single packaged enclosure. Each generator module used 576 cells, 16 mm diameter by 500 mm active length, of the now obsolete porous zirconia support tube (PST) design. A description of this unit and its performance can be found in the literature. As this unit did not perform satisfactorily, it was modified and repaired. The JGU cogeneration system was modified to accept a single generator module utilizing 576 air electrode supported (AES) cells of 16 mm diameter by 500 mm active length in place of the previously used pair of PST type modules. This unit was originally scheduled to be shipped to Osaka, Japan for application testing, but the great Hanshin earthquake which struck the Kobe area in January 1995 disrupted the operations of Osaka Gas to such an extent that this proved infeasible. An unequivocally successful extended factory test was initiated on March 22, 1995 and concluded on February 10, 1997.

The world's first 100 kWe class Solid Oxide Fuel Cell (SOFC) power generation system is being supplied by Westinghouse and is sponsored by EDB/ELSAM, a consortium of Dutch and Danish utilities. This natural gas fueled unit will be installed near Arnhem, The Netherlands, at an auxiliary district heating plant [Hulp Warmte Centrale] at the Rivierweg in Westervoort, a site provided by NUON, one of the Dutch participants and will supply ac power to the utility grid and hot water to the district heating system serving the Duiven/Westervoort area.

The 100 kW SOFC generator module utilizes tubular Air Electrode Supported (AES) SOFCs of nominally 22 mm diameter by 1500 mm active length. The generator module or stack is of seal-less design and employs 1152 tubular SOFCs oriented vertically and arranged in twelve bundle rows. Each bundle row consists of four bundles, with each bundle a rectangular cell array having three cells in parallel and eight cells in series. The bundle rows are connected in electrical series yielding a serpentine current path. A cross section of the 100 kW SOFC stack is shown in Figure 2. The thermal-hydraulic design for the 100 kW stack differs from previous Westinghouse practice in that the natural gas reformers are integral with the insulation barriers between bundle rows, with heat supplied by thermal radiation directly from the SOFCs. (In the 25 kW SOFC units, the reformers are hydraulically integrated with the cell stack, but heated by exhaust gas.) As in prior Westinghouse practice, spent anode gas is recirculated and mixed with fresh fuel (desulfurized natural gas) using an ejector with pressurized natural gas as the primary fluid. See Figure 3. The outer canister of the 100 kW generator module is cooled with process air to limit dissipation to the ambient and to limit canister temperature.

The 100 kW SOFC power generation system is composed of five discrete skid mounted assemblies or "skids". The generator skid supports the SOFC stack and the electrically powered process air heater used for startup. The Thermal Management Skid (TMS) supports the recuperators, the air movers (blowers), air and exhaust piping and air control valves, and the Electrical Distribution System (EDS), a shallow set of enclosures which houses all electrical distribution and electronic hardware including the control computer. The Fuel Supply System (FSS) skid supports the fuel and purge gas control valves, the desulfurizers, and the small steam generator used during startup along with a small water supply tank. These three skids are arranged in a rectilinear package as shown in Figure 4 measuring 8.42 m long by 2.75 m wide with a maximum height of 3.58 m. The power conditioner and the hot water heater are also skid mounted, but supplied by EDB/ELSAM.

Westinghouse tubular AES-SOFCs are being tested not only at Westinghouse, but also at the Kansai Electric Power Company in Japan and Ontario Hydro in Canada. Kansai Electric purchased from Westinghouse a fully automated atmospheric pressure tubular cell test stand designed and built by Westinghouse and capable of exercising at steady or cyclic conditions either a single cell or a "short stack" consisting of two or four tubular cells with cell active lengths up to 1500 mm. To date, they have tested two test articles with four 500 mm active length cells and are presently testing a single 1500 mm length cell.

The cell test facility at Ontario Hydro consists of two test stands that can each exercise test articles similar to those at Kansai Electric, but at elevated pressure up to fifteen atmospheres.

The cell test facility built by Ontario Hydro was designed in collaboration with Westinghouse. To date, OH has tested seven test articles.

Ontario Hydro is presently completing the construction of a pressurized bundle test facility designed in collaboration with Westinghouse. The 10 kW test article will contain two bundles of twenty four cells each and is expected to begin operation in the second half of 1997.

Negotiations are underway for the renewal of the cooperative agreement between DOE and Westinghouse for tubular SOFC development. In addition to a focus upon cell and stack manufacturing cost reduction, the proposed renewal program will design, develop, build, and test pressurized SOFC Gas Turbine (PSOFC/GT) hybrid cycle power systems at customer sites. A simplified schematic for a PSOFC/GT hybrid cycle power system can be seen in Figure 5. Simplistically, the expansion turbine has replaced the blower drive motor, and the compressor has replaced the blower. No electrical power is therefore consumed to move process air, a major element of parasitic power consumption for atmospheric pressure systems, and additional electric power can be generated from the turbine shaft. The first field unit proposed for the renewal program is a 250 kW system which will utilize a stack of the same design as used for the EDB/ELSAM 100 kW unit, but operating at approximately 3.5 atmospheres pressure in conjunction with a micro turbine generator (MTG) of nominally 50 kW capacity. The expected electrical generating efficiency of the 250 kW PSOFC/MTG power system is 60% (ac/LHV). The second field unit, a one MW system, will utilize four SOFC stacks and a 250 kW turbine generator to yield a system with over 60% electrical generation efficiency. The third proposed field unit will upgrade the second field unit by adding four additional SOFC stacks and upgrading the turbo-machinery in order to achieve 70% efficiency. Startup of the 250 kW PSOFC/GT is planned for early 1999 while startup of the one MW PSOFC/GT system is expected in the year 2000.

## Results and Accomplishments

A summary of the characteristics and results for Westinghouse SOFC field units to date is given in Table 1. Factory test of the JGU 25 kW AES-SOFC Cogeneration system was initiated on March 22, 1995 at Westinghouse's Pre-Pilot Manufacturing Facility (PPMF) in Monroeville, PA. The unit was shut down after 1200 hours of operation to repair a fuel leak from a reformer and to replace deactivated reformer catalyst. The unit was subsequently relocated to the Westinghouse Pilot Manufacturing Facility at the Westinghouse Science and Technology Center upon decommissioning of the PPMF. Operation of the JGU 25 kW AES-SOFC Cogeneration System was terminated on February 10, 1997 in order to permit contract mandated inspection of the cells and stack internals prior to the end of the Japanese fiscal year on March 31. The JGU 25 kW AES-SOFC Cogeneration System generated power for 13,194 hours (282 MW-hrs). In a maximum power test, the unit generated 25 kW at 84% fuel utilization at 1855 operating hours and 24.9 kW at 79% fuel utilization just prior to shut down at 13,191 operating hours. A plot of kW, Voltage and Amperes vs time can be found in Figure 6. Nominal conditions during operation were 170 Amperes, (306 mA/cmsq), 79% fuel utilization, 1020°C set point temperature. The degradation in terminal voltage over the test period at these set point conditions was 1.86%. Thermal control of the generator to set point is governed by the centroidal

temperature of the hottest quadrant. During operation, the centroidal temperatures of the quadrants drifted apart. After adjusting the observed quadrant voltage to a fixed temperature, the coldest quadrant experienced no voltage degradation. During the course of operation, the unit endured a total of ten thermal cycles between operating temperature (1000°C) and ambient temperature and thirteen instances of sulfur break-through from the desulfurizer. Sulfur poisoning was reversible since complete voltage recovery was observed after replacement of the desulfurizer reagent. The longest period of continuous operation without outage of any kind was 6500 hours. Upon disassembly of the stack, visual inspection using a borescope found no cracked cells. The physical appearance of cells and bundles was virtually indistinguishable from newly-manufactured samples. The primary reason for the observed degradation in voltage was the development of porosity in the interconnection resulting in gas leakage between cathode and anode. Interconnection porosity development after 13,000 hours was an increasing function of increasing temperature. At 1000°C, interconnection porosity development was insignificant.

The EDB/ELSAM 100 kW SOFC field unit is presently undergoing a process and control test of the balance of plant (BOP). The SOFC stack is in the final stages of assembly. The power conditioner has satisfactorily completed factory acceptance testing in Europe. The Heat Export System (hot water heater) is in the vendor solicitation phase. Factory acceptance tests are scheduled to begin in September\_97 with site acceptance tests expected in December\_97. Analytical estimates of performance show that maximum system efficiency will occur at thermal balance, that operating condition where no ancillary energy is required to maintain the SOFC stack at operating temperature. The maximum efficiency of the 100 kW SOFC power generation system is estimated as 47% (net ac/LHV) at 100 kW ac net output. Overall fuel effectiveness at this point will exceed 70%. System maximum power is estimated to be approximately 160 kW-ac with a fuel effectiveness approaching 80%. These estimates of performance are shown in Figure 7. The EDB/ELSAM 100 kW SOFC is expected to operate for two years.

At Kansai Electric Power Company facilities, a short stack of four 16 mm diameter by 500 mm active length AES-SOFCs endured 100 thermal cycles between operating temperature (1000°C) and ambient temperature and generating power for over 10,100 hours at 1000°C, 450 mA/cmsq and 85% fuel utilization. Presently under test is a 22 mm diameter by 1500 mm active length cell which has achieved 1500 hours of operation without evident degradation.

At Ontario Hydro, a 22 mm diameter by 1500 mm active length AES-SOFC is presently under test and has passed 3700 hours of operation at elevated pressure (nominally 5 atmospheres) with no evident degradation after two thermal cycles to ambient conditions.

The SOFC at modest elevated pressure (4 atmospheres) can easily yield a 50% conversion of natural gas fuel energy into electricity. Noting that all of the fuel energy not converted to electricity by the SOFC is contained in the exhaust gas at 850°C, a modest recuperated GT-generator efficiency of 20% will yield a system efficiency of 60%. More elaborate GT cycles that employ an intercooled, recuperated, reheat GT cycle can then be expected to approach and exceed a 70% efficiency level.

## **Benefits**

Westinghouse tubular AES-SOFCs have demonstrated superior voltage stability (approximately 0.1 per cent per thousand hours) in tests exceeding 13,000 hours of power generation at high fuel utilization. Westinghouse tubular AES-SOFCs have demonstrated superior thermal toughness by enduring without deleterious effect 100 thermal cycles from power generation at 1000°C to ambient conditions. Atmospheric pressure tubular SOFC systems can approach a simple cycle power generation efficiency of 50% (net ac/LHV). Pressurized SOFC/simple cycle Gas Turbine hybrid cycle power systems can exceed 60% efficiency at approximately the 250 kW capacity level while GT cycles with intercooling and reheat can exceed 70% electrical generation efficiency at the integer MW capacity level.

## **Acknowledgments**

Westinghouse acknowledges the guidance and assistance of Mr. William C. Smith, SOFC Project Manager, Gas Power Systems Division, Office of Project Management, Federal Energy Technology Center, Morgantown, WV, in the course of this Cooperative Agreement (DE-FC21-91MC28055) spanning the period from Dec. 1, 1990 through Nov. 30, 1997.

Table 1 — Westinghouse SOFC Field Units

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 <sub>2</sub> +CO	0.5
1987	Osaka Gas	3	1	TK-PST	360	144	3012	H <sub>2</sub> +CO	6.1
1987	Osaka Gas	3	1	TK-PST	360	144	3683	H <sub>2</sub> +CO	7.4
1987	Tokyo Gas	3	1	TK-PST	360	144	4882	H <sub>2</sub> +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	36.0
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	99.1
1995	SCE-2	27	1	AES	500	576	5582	PNG/DF-2/JP-8	118.2
1995	JGU-2	25	1	AES	500	576	13,194	PNG	282.1
<i>Future Work</i>									
1997	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

24 March 97

Figure 1 — Process flow schematic for an SOFC Cogeneration System

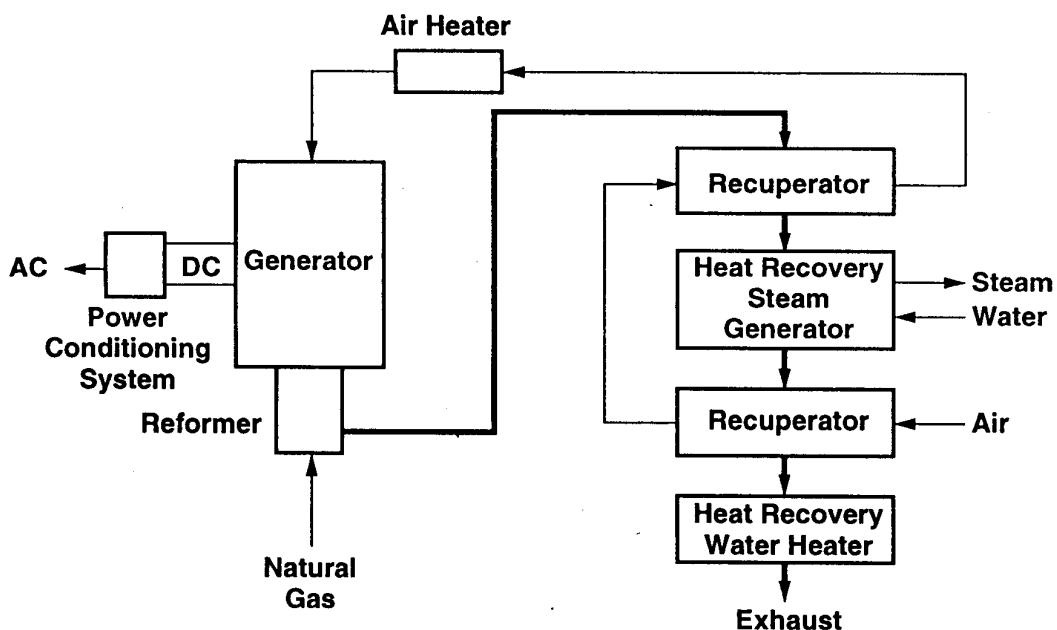


Figure 2 — 100 kW SOFC Stack Cross Section

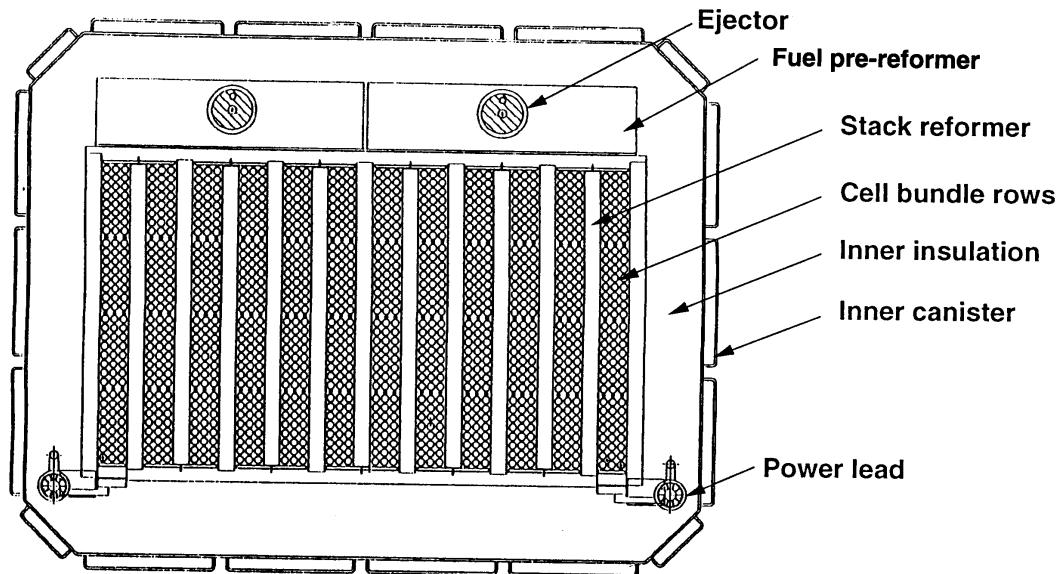
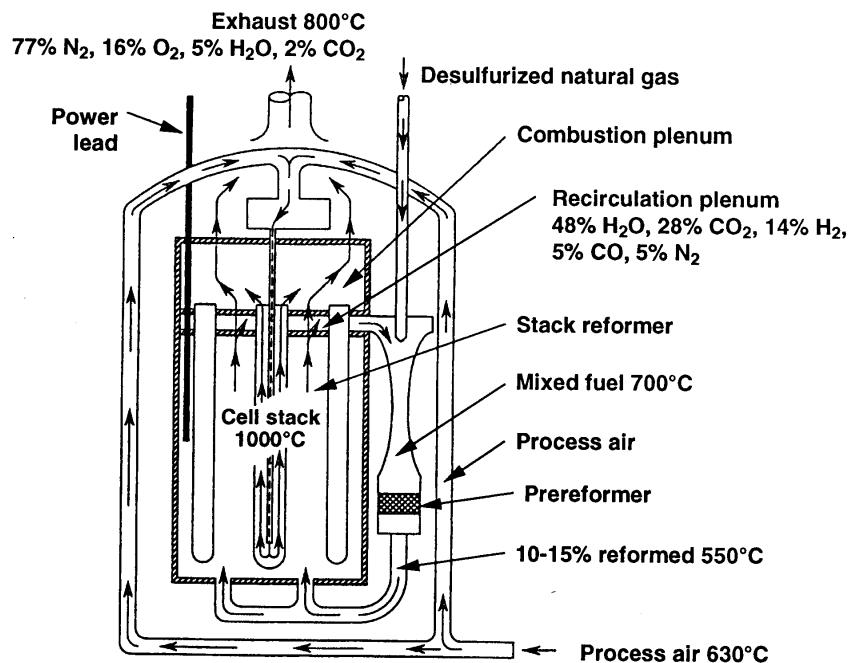
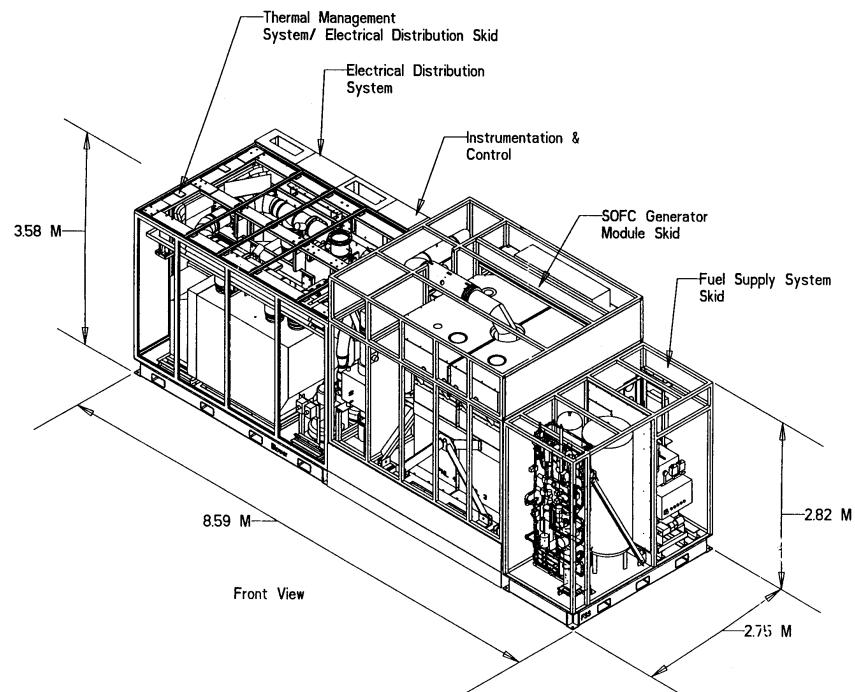


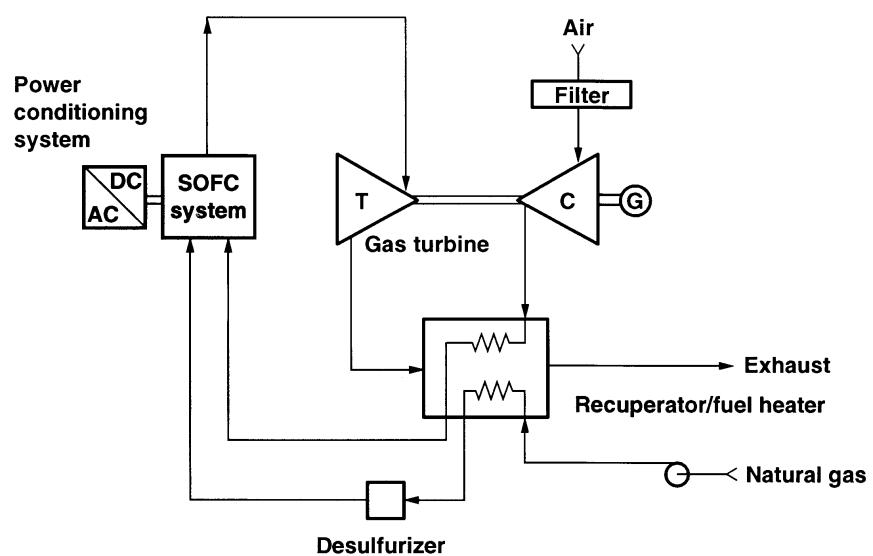
Figure 3 — 100 kW SOFC Stack Gas Flows



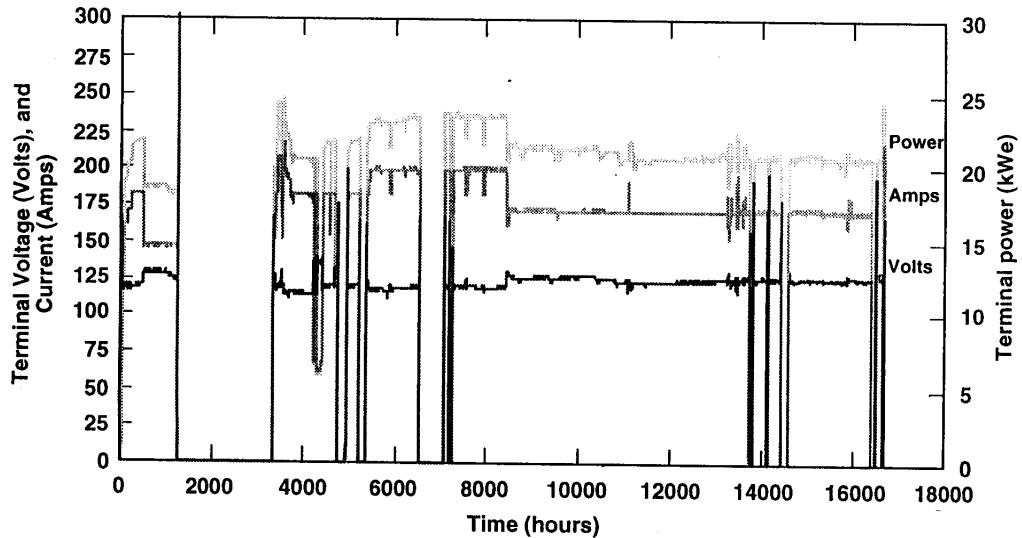
**Figure 4 — 100 kW SOFC Field Unit (interior view)**



**Figure 5 — Single Shaft PSOFC/GT**



**Figure 6 — JGU 25 kW AES-SOFC Power System Performance**



**Figure 7 — EDB/ELSAM 100 kW SOFC Performance Estimate**

<b>Rated Power (net)</b>	<b>100 kWe AC</b>
<b>Efficiency @ Rating (AC/LHV)</b>	<b>47%</b>
<b>Maximum Power (net)</b>	<b>160 kWe AC</b>
<b>Thermal Recovery @ Rating (Hot Water)</b>	<b>52 kWth</b>
<b>Thermal Recovery @ Maximum (Hot Water)</b>	<b>125 kWth</b>
<b>Maximum Fuel Effectiveness</b>	<b>80%</b>

## SOFC SYSTEM ANALYSIS

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A preliminary design and cost estimate of a 500 kW SOFC commercial unit was conducted to assess its economic potential for distributed power generation. The stacks used were of planar design based on the thin electrolyte technology. The study results indicate that the optimum operating temperature for the thin electrolyte is 800C, the product can be produced at \$700-800/kW with 55-60% overall electric efficiency (LHV), and the cost of electricity (5-6 cents/kWh based on \$4/MMBtu retail natural gas price and 25% annual capital recovery) is sufficiently low to capture the distributed generation market.

This study is funded by the Gas Research Institute (GRI). Bechtel Corporation is the prime contractor, responsible for the overall system design and cost estimate. TDA Research provided the stack performance and cost estimate.

## INTRODUCTION

The planar SOFC has the potential to be more efficient and lower cost than the tubular design because the cells used have shorter current path and are simpler to manufacture. However, it is difficult to find suitable low cost materials for the sealant and interconnect at the 1000C cell operating temperature. To overcome this technical barrier, Gas Research Institute (GRI) has been funding research for developing thin electrolyte cells to reduce the operating temperature. TDA Research, in a recent GRI funded stack cost study (1, 2), showed the thin electrolyte stack could be manufactured at a very low cost of \$230/kW due to the use of metallic instead of ceramic interconnects. However, the stack cost typically represents only 20-40% of the total system cost. To determine its commercial viability for distributed power generation, GRI has engaged Bechtel to conduct a cost analysis of the entire system. In this study, TDA Research assisted Bechtel in estimating the stack performance and cost.

In this system analysis, a system simulation model was built and tradeoffs were performed to select the optimum operating parameters and system configuration. The tradeoffs were geared to address the issues such as:

- Will the increased cathode polarization resistance at the reduced operating temperature significantly penalize the overall system efficiency? How much can the increased Nernst potential at the reduced temperature help improve the system efficiency? Will the reduced temperature also reduce the supporting facility cost, such as the air preheater? What would be the optimum operating temperature when all the factors are considered?
- Is it beneficial to operate the cell at higher current density?
- Is it beneficial to operate the cell at higher fuel utilization?
- Is there any advantage to use pressurized operation?
- What is the best integration scheme between the stacks and supporting facilities?

This paper summarizes the preliminary results of this study.

## STUDY CASES

Twenty five study cases divided into seven groups were analyzed as shown in Table I.

Groups 1-5 are ambient pressure operation cases. Groups 6 and 7 are pressurized operation cases. A comparison of them establishes the relative advantages between the ambient pressure operation and pressurized operation.

Groups 1-4 search for the optimum current density at the stack operating temperatures of 700, 800, 900, and 1000C, respectively. Three or four different current densities were analyzed in each of these groups. All the cases are based on a fuel utilization of 85%. The optimum case in each temperature group is then selected for comparison to establish the effects of operating temperature.

Group 5 searches for the optimum fuel utilization. Three different fuel utilization levels were analyzed under the condition of 800C stack operating temperature and 300 mA/cm<sup>2</sup> current density. Case 5B is actually a duplicate of Case 2B.

Group 6 searches for the optimum operating pressure under the condition of 800C stack operating temperatures, 300 mA/cm<sup>2</sup> current density, and 85% fuel utilization. Four different pressure levels were analyzed. Group 7 also searches for the optimum operating pressure. The current density and fuel utilization used are the same as Group 6 but the stack temperature is increased to 1000C to determine the benefits of a hotter gas for a more efficient operation of the downstream turbogenerator. Five different operating pressures were analyzed in Group 7.

All the cases were designed for a minimum excess air level of 30% to ensure there is adequate oxygen concentration available in the cathode. This minimum excess air requirement has forced the high stack temperature cases in Groups 3 and 4 to operate at high current densities. For example, the minimum current density that a 1000C stack can operate is 600 mA/cm<sup>2</sup>. Below this current density level, the stack is too efficient and the waste heat generated is not sufficient to heat up a large amount of air to the stack operating temperature.

## SYSTEM CONFIGURATIONS

### Ambient Pressure Operation

All the atmospheric pressure cases (Groups 1-5) are based on the system configuration shown in Figure I.

The natural gas feed is desulfurized and fed to the ejector as the motive gas to induce an anode gas recycle. The anode gas recycle provides an internal steam supply for the pre-reformer by using the cell reaction product water in the anode exhaust gas. In the pre-reformer, the natural gas is partially reformed to ensure there is hydrogen available for the cell reactions at the cell entrance to generate sufficient heat for the highly endothermic internal reforming reaction. An anode preheater is included as an extended part of the pre-reformer to heat the pre-reformed gas to the anode inlet temperature. Heat required for the pre-reforming and anode preheating is provided by a waste heat recovery from the fuel cell stack flue gas.

Multiple stacks (only one shown in Figure I) made of small size (10 cm diameter) cells are used. The stack heat is removed by a direct heat dissipation to the air preheater coils (only one shown in Figure I) placed in between the stacks. The small cell size was chosen to facilitate this type of heat removal. It prevents the cells from developing a large temperature gradient between the center and edge. A blower supplies the air feed to the air preheater coils. The preheated air is further heated to the cathode inlet temperature by a direct combustion in a “pre-burner” with the spent fuel in the anode exhaust gas. In other SOFC system designs, the anode exhaust is usually burned off with the cathode exhaust gas in an “after-burner”.

An effective heat integration between the stack heat removal and air preheating has been a major system design challenge for the SOFC. A standard heat integration scheme employed by many SOFC developers uses the cathode gas for the heat removal and preheat the air feed by heat exchange with the cathode exhaust gas. As the temperature rise of the cathode gas in the stacks is limited (usually less than 100C), the required flow is very large. Typically, a stoichiometric air ratio of 4-5 is necessary for the heat removal. This large air flow significantly increases the air preheater size. The large size, in conjunction with the high air discharge temperature required, significantly increases the air preheater cost. This has been one major reason that the SOFC system cost is high. The large air flow also increases the system pressure drop. The combined effect of large flow and high pressure drop increases the

air blower size and the auxiliary power consumption. As a result, the system efficiency is reduced.

The present design does not depend on the cathode gas for the stack heat removal. The air flow required is substantially smaller. Thus, the air preheater is much smaller and the auxiliary power consumption is reduced. Also, a much hotter stack flue gas is available for downstream generation of steam, hot water, or additional power because the cathode exhaust gas is no longer used to preheat the air feed.

The “pre-burner” used in the present design reduces the duty requirement and air discharge temperature of the air preheater. As a result, the air preheater can be even smaller and constructed of a lower cost material. The “pre-burner”, however, decreases the oxygen concentration in the cathode feed by one to two percentage points. This was found to generate no substantial efficiency penalty in the present study.

The anode exhaust gas from the stacks is split into two streams: one to the ejector and other one to the “pre-burner”. The cathode exhaust gas, after heat recovery for the pre-reformer/anode preheater, is discharged to the atmosphere. As indicated previously, plenty of high temperature heat is available in this stream for further generation of steam, hot water, or power, if desired. All the high temperature system components are housed in a vessel to minimize high temperature pipe penetration through the vessel. The DC power produced from the stacks is converted to AC power in the inverter. Not shown in Figure I but included in the cost estimate are a startup boiler, a nitrogen system, and a control system.

### Pressurized Operation

All the pressurized operation cases (Groups 6 and 7) are based on the system configuration shown in Figure II. It is essentially the same as that for the atmospheric pressure cases except a turbogenerator is included to produce additional power and to supply the compressed air feed by expansion of the stack flue gas.

The fuel cell/turbine integration described above is only one of many schemes commonly postulated. One other possible scheme is to have the gas turbine placed upstream of the fuel cell unit. The turbine exhaust becomes the cathode feed with the fuel cell flue gas to preheat the air feed to the turbine combustor. In this case, the fuel cell stacks can be operated at atmospheric pressure. Another possible scheme is to generate steam or hot air in the air preheater coils for expansion in a steam or gas turbine. These schemes will be investigated in the future in this study.

## SYSTEM SIMULATION MODEL

The system simulation model used in this study performs an overall heat and material balance to determine the process stream flows and conditions, sizes the major equipment, estimates the capital and maintenance costs, and analyzes the cost of electricity. In the heat and material balance, the stack performance was estimated based on use of the following cell components:

- Anode: Ni/Zr cermet, 100 micron thick
- Electrolyte: YSZ, 5 micron thick
- Cathode: Sr-doped La Manganite, 100 micron thick
- Interconnect: stainless steel for the 700C and 800C operating temperatures, high alloy metal for the 900C operating temperature, and La Chromite for the 1000C operating temperature; all materials are 1000 micron thick

The stack costs previously estimated by TDA Research (2) based on 200 MW/yr production were \$43/ft<sup>2</sup>, \$70/ft<sup>2</sup>, and \$98/ ft<sup>2</sup> cell area for the 700/800, 900, and 1000C operating temperatures, respectively. The ionic resistance of electrolyte used are 0.048, 0.017, 0.007, and 0.003 Ohm-cm<sup>2</sup> at the 700, 800, 900, and 1000C operating temperatures, respectively. The corresponding total area specific resistances, which also include ohmic and polarization resistances of electrodes and ohmic and contact resistances of the interconnect, are 1.01, 0.68, 0.401, and 0.284 Ohm-cm<sup>2</sup>, respectively. The ionic resistances are seen to be a very small fraction of the total cell resistances due to the use of the thin electrolyte. The compressor and expander of the turbogenerator were assumed to have 76% and 86% polytropic efficiencies, respectively. The inverter was assumed to have 95% efficiency.

## STUDY RESULTS

A summary of the system performance for all the cases, including feed requirements, a breakdown of the cell voltage drops, cell area required, amounts of power generated and consumed, and electric and cogeneration efficiencies, is shown in Table II.

A cost summary of all the cases is shown in Table III. The O&M cost component of the cost of electricity consists of maintenance cost, stack replacement cost, and catalyst consumption. As the fuel cell unit was designed for unattended operation, there is no operating labor cost. The annual maintenance cost, including both materials and labor, was assumed to be 1% of the capital cost. The stack replacement cost was based on a 5-year stack life with a salvage value equal to 1/3 of the original stack cost. The cost of electricity was calculated based on \$4/MM Btu natural gas price and 25% annual capital recovery (or 4 year payback) which are the typical values anticipated in the United States for the distributed power generation. Results of the specific tradeoff analysis are discussed below.

### Optimum Current Density (Groups 1-4)

At higher current density, the cell voltage drops, the stacks are less efficient, and the system electric efficiency is reduced. The larger amount of heat generated from the stacks increases the stoichiometric air ratio and the air preheater size. On the other hand, the power density is increased and the total cell area required is reduced. The optimum current densities at 700, 800, 900, and 1000C operating temperatures are 200, 300, 500, and 600 mA/cm<sup>2</sup>, respectively.

### Optimum Stack Operating Temperature

As the operating temperature increases, the stacks become more efficient but also more expensive. A comparison of the optimum current density cases from Groups 1-4 (Cases 1A, 2B, 3B, and 4A) in Figure III shows that the optimum stack temperature is 800C. This optimum temperature is a result of the tradeoff between the efficiency and stack cost. It should be noted that, due to the use of the “pre-burner”, the air preheater temperatures, even in the 1000C stack operating temperature cases, never exceed 660C. As a result, none of the study cases needs to use high alloy metals or ceramic materials for the air preheater. The optimum operating temperature will decrease if the cell resistance is further reduced in the future.

### Optimum Fuel Utilization (Group 5 Cases)

The stacks are more efficient as the fuel utilization increases, even after taking into account the cell voltage reduction due to the lower fuel concentration in the anode. The more efficient stacks release less heat and, thus, the stoichiometric air ratio and the air preheater size are reduced. The total cell area, on the other hand, increases due to the lower power density. Overall, the total capital cost is not sensitive to the fuel utilization level. The higher efficiency is the main reason that the higher fuel utilization case is more economical. However, there is an upper limit for the practical fuel utilization level. Beyond that, certain areas of the cells could be deprived of fuel if a mal-distribution of gases develops due to the stack design imperfection, stack aging, or other reasons. Only the stack developers based on their actual operating experience can determine whether the upper limit is 85%, 90% or some other values.

### Optimum Operating Pressure (Groups 6 and 7)

As the operating pressure increases, the turbogenerator has to compress the air to a higher pressure and this results in a hotter air feed to the fuel cell unit. The hotter air is less effective in removing the stack heat. Thus, the stoichiometric air ratio and air preheater size are increased. Due to the larger air flow and higher working pressure, a larger size turbogenerator is also required. The stacks, on the other hand, are more efficient because of the higher reactant partial pressure available. The expander gas of the turbogenerator for the cases

studied is in a temperature region that the turbogenerator produces less power as the pressure increases. To compensate for this lower power production, the stacks have to produce more power. Therefore, the cell area required does not necessarily decrease when the stack efficiency increases with the operating pressure. Overall, the capital cost increases as the operating pressure increases.

The stacks become more efficient and turbogenerator becomes less efficient as the operating pressure increases. Due to these two opposing effects, the overall electric efficiency slightly increases and then decreases as the operating pressure increases. As the efficiency variation is very small, the cost of electricity reflects the change of capital cost with the operating pressure.

A comparison between Groups 6 and 7 indicates that the higher stack operating temperature, even though offers a higher electric efficiency, has no net economical advantage for pressurized operation. The major reason is that stack cost is substantially higher at 1000C than at 800C.

A comparison of the best pressurized case (Case 6A) with the best atmospheric pressure case (Case 2B) under the same current density and fuel utilization indicates the pressurization offers no major economical advantage. Given the same cost of electricity, the atmospheric pressure operation is preferred because the lack of high temperature rotating equipment can make the unit more reliable, less noisy, and safer to operate. Also, the atmospheric operation is less likely to need feed gas compression if the natural gas supply pressure is not sufficiently high.

## CONCLUSIONS

This study shows that a properly designed planar thin electrolyte SOFC unit can be produced at \$700-800/kW with 55-60% efficiency (LHV). The cost of electricity based on the retail natural gas price and capital recovery rate anticipated for the distributed power generation is around 5 cents/kW. In comparison, the retail electricity cost in the United States, ranges from 6 to 12 cents/kW. Thus, the SOFC can be commercially viable for capturing the distributed power generation market.

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**Table I Study Cases**

<b>Group 1</b>	<b>Ambient Pressure</b> <b>85% Fuel Utilization</b> <b>700 C Stack Temperature</b>	<b>Case 1A: 200 mA/cm<sup>2</sup> current density</b> <b>Case 1B: 300 mA/cm<sup>2</sup> current density</b> <b>Case 1C: 400 mA/cm<sup>2</sup> current density</b>
<b>Group 2</b>	<b>Ambient Pressure</b> <b>85% Fuel Utilization</b> <b>800 C Stack Temperature</b>	<b>Case 2A: 200 mA/cm<sup>2</sup> current density</b> <b>Case 2B: 300 mA/cm<sup>2</sup> current density</b> <b>Case 2C: 400 mA/cm<sup>2</sup> current density</b> <b>Case 2D: 500 mA/cm<sup>2</sup> current density</b>
<b>Group 3</b>	<b>Ambient Pressure</b> <b>85% Fuel Utilization</b> <b>900 C Stack Temperature</b>	<b>Case 3A: 400 mA/cm<sup>2</sup> current density</b> <b>Case 3B: 500 mA/cm<sup>2</sup> current density</b> <b>Case 3C: 600 mA/cm<sup>2</sup> current density</b>
<b>Group 4</b>	<b>Ambient Pressure</b> <b>85% Fuel Utilization</b> <b>1000 C Stack Temperature</b>	<b>Case 4A: 600 mA/cm<sup>2</sup> current density</b> <b>Case 4B: 700 mA/cm<sup>2</sup> current density</b> <b>Case 4C: 800 mA/cm<sup>2</sup> current density</b>
<b>Group 5</b>	<b>Ambient Pressure</b> <b>300 mA/cm<sup>2</sup> current density</b> <b>800 C Stack Temperature</b>	<b>Case 5A: 80% fuel utilization</b> <b>Case 5B (2B) : 85% fuel utilization</b> <b>Case 5C: 90% fuel utilization</b>
<b>Group 6</b>	<b>Pressurized</b> <b>85% Fuel Utilization</b> <b>300 mA/cm<sup>2</sup> Current Density</b> <b>800 C Stack Temperature</b>	<b>Case 6A: 3 atm operating pressure</b> <b>Case 6B: 4 atm operating pressure</b> <b>Case 6C: 5 atm operating pressure</b> <b>Case 6D: 6 atm operating pressure</b>
<b>Group 7</b>	<b>Pressurized</b> <b>85% Fuel Utilization</b> <b>300 mA/cm<sup>2</sup> Current Density</b> <b>1000 C Stack Temperature</b>	<b>Case 7A: 5 atm operating pressure</b> <b>Case 7B: 6 atm operating pressure</b> <b>Case 7C: 7 atm operating pressure</b> <b>Case 7D: 8 atm operating pressure</b> <b>Case 7E: 9 atm operating pressure</b>

**Table II Summary of System Performance**

Study Case	1A	1B	1C	2A	2B	2C	2D	3A	3B	3C	4A	4B	4C
	1	1	1	1	1	1	1	1	1	1	1	1	1
Operating Pressure, atm	700	700	700	800	800	800	800	900	900	900	1000	1000	1000
Stack Operating Temp., C	85	85	85	85	85	85	85	85	85	85	85	85	85
Fuel Utilization, %	200	300	400	200	300	400	500	400	500	600	600	700	800
Current Density, mA/cm <sup>2</sup>													
Natural Gas Feed (HHV), MMBtu/h	3.32	3.64	4.57	3.15	3.41	3.72	4.09	3.50	3.71	3.94	3.76	3.93	4.11
Stoichiometric Air Ratio	1.66	1.99	2.32	1.32	1.49	1.65	1.82	1.35	1.44	1.54	1.31	1.37	1.43
Cell Voltage, Volt	0.741	0.642	0.542	0.779	0.721	0.662	0.602	0.702	0.664	0.625	0.653	0.626	0.598
Stack Power Density, kW/m <sup>2</sup>	1.48	1.92	2.17	1.56	2.16	2.65	3.01	2.81	3.32	3.75	3.92	4.38	4.79
Total Cell Area Required, ft <sup>2</sup>	3,867	2,975	2,653	3,657	2,639	2,159	1,901	2,030	1,720	1,524	1,456	1,303	1,193
Power from Inverter, kW	503.9	505.5	507.6	503.0	503.6	504.4	505.3	503.4	503.8	504.3	503.5	503.8	504.2
Power from Turbogenerator, kW	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Power Consumed for Blower, kW	3.9	5.5	7.6	3.0	3.6	4.4	5.3	3.4	3.8	4.3	3.5	3.8	4.2
Net Power Export, kW	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0
Electric Efficiency, % (LHV)	57.0	49.2	41.4	60.1	55.5	50.9	46.3	54.1	51.1	48.1	50.3	48.2	46.1
Cogeneration Potential, % (LHV)	35.2	42.6	50.0	32.7	37.2	41.6	46.1	39.0	41.9	44.9	43.1	45.2	47.3
Cogeneration Efficiency, % (LHV)	92.2	91.8	91.4	92.8	92.7	92.5	92.4	93.1	93.0	93.0	93.4	93.4	93.4

**Table II Summary of System Performance (continued)**

Study Case	5A	5B	5C	6A	6B	6C	6D	7A	7B	7C	7D	7E
	1	1	1	3	4	5	6	5	6	7	8	9
Operating Pressure, atm	800	800	800	800	800	800	800	1000	1000	1000	1000	1000
Stack Operating Temp., C	80	85	90	85	85	85	85	85	85	85	85	85
Fuel Utilization, %	300	300	300	300	300	300	300	300	300	300	300	300
Current Density, mA/cm <sup>2</sup>												
Natural Gas Feed (HHV), MMBtu/h	3.56	3.41	3.26	3.02	2.97	2.95	2.95	2.82	2.80	2.79	2.78	2.78
Stoichiometric Air Ratio	1.56	1.49	1.42	1.71	1.82	1.94	2.07	1.34	1.39	1.45	1.50	1.56
Cell Voltage, Volt	0.733	0.721	0.711	0.735	0.739	0.742	0.745	0.758	0.762	0.765	0.767	0.770
Stack Power Density, kW/m <sup>2</sup>	2.20	2.16	2.13	2.20	2.22	2.23	2.24	2.27	2.28	2.29	2.30	2.31
Total Cell Area Required, ft <sup>2</sup>	2,595	2,639	2,673	2,337	2,301	2,286	2,285	2,184	2,166	2,155	2,150	2,149
Power from Inverter, kW	504.0	503.6	503.3	454.5	450.1	449.4	451.0	438.4	436.8	436.4	436.9	438.1
Power from Turbogenerator, kW	0.0	0.0	0.0	45.5	49.9	50.6	49.0	61.6	63.2	63.6	63.1	61.9
Power Consumed for Blower, kW	4.0	3.6	3.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Net Power Export, kW	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0	500.0
Electric Efficiency, % (LHV)	53.1	55.5	58.0	62.7	63.7	64.1	64.1	67.1	67.6	68.0	68.1	68.2
Cogeneration Potential, % (LHV)	39.5	37.2	34.7	29.4	28.1	27.4	27.1	25.8	25.1	24.6	24.3	24.1
Cogeneration Efficiency, % (LHV)	92.6	92.7	92.7	92.1	91.8	91.5	91.2	92.9	92.7	92.6	92.4	92.3

**Table III Cost Summary, 1996 Pricing**

Study Case	1A	1B	1C	2A	2B	2C	2D	3A	3B	3C	4A	4B	4C
	1	1	1	1	1	1	1	1	1	1	1	1	1
Operating Pressure, atm	700	700	700	800	800	800	800	900	900	900	1000	1000	1000
Stack Operating Temp., C	85	85	85	85	85	85	85	85	85	85	85	85	85
Fuel Utilization, %	200	300	400	200	300	400	500	400	500	600	600	700	800
Current Density, mA/cm <sup>2</sup>													
Capital Cost, \$/kW													
Stacks	331	254	227	313	226	185	162	285	241	214	284	254	233
Air Preheater/Vessel	93	114	149	76	81	92	107	74	80	88	74	80	86
Air Blower/Turbogenerator	2	2	3	2	2	2	2	2	2	2	2	2	2
Inverter & Control System	200	200	200	200	200	200	200	200	200	200	200	200	200
Desulfurizer, Ejector, Prereformer	25	28	32	24	25	26	28	25	26	27	26	27	27
Other Support Facilities	155	155	155	155	155	155	155	155	155	155	155	155	155
Total	806	754	766	769	689	660	656	741	705	687	741	718	703
Cost of Electricity, Cent/kWh (a)													
Capital Recovery (25% annualy)	2.42	2.27	2.30	2.31	2.07	1.98	1.97	2.22	2.12	2.06	2.23	2.16	2.11
Natural Gas (\$4/MMBtu)	2.66	3.07	3.66	2.52	2.73	2.98	3.27	2.80	2.97	3.15	3.01	3.14	3.29
O&M	0.69	0.57	0.55	0.66	0.51	0.45	0.42	0.62	0.55	0.50	0.62	0.57	0.54
Total	5.77	5.91	6.50	5.49	5.31	5.41	5.66	5.64	5.63	5.72	5.85	5.87	5.94

**Table III Cost Summary, 1996 Pricing  
(continued)**

Study Case	5A	5B	5C	6A	6B	6C	6D	7A	7B	7C	7D	7E
	1	1	1	3	4	5	6	5	6	7	8	9
Operating Pressure, atm	800	800	800	800	800	800	800	1000	1000	1000	1000	1000
Stack Operating Temp., C	80	85	90	85	85	85	85	85	85	85	85	85
Fuel Utilization, %	300	300	300	300	300	300	300	300	300	300	300	300
Current Density, mA/cm <sup>2</sup>												
Capital Cost, \$/kW												
Stacks	222	226	228	200	197	195	195	426	423	420	419	419
Air Preheater/Vessel	83	81	78	78	81	84	87	62	63	65	66	68
Air Blower/Turbogenerator	2	2	2	64	85	105	125	78	90	101	112	124
Inverter & Control System	200	200	200	200	200	200	200	200	200	200	200	200
Desulfurizer, Ejector, Prereformer	25	25	25	21	21	21	21	20	20	20	19	19
Other Support Facilities	155	155	155	201	201	201	201	201	201	201	201	201
Total	688	689	688	764	784	806	829	988	997	1007	1019	1032
Cost of Electricity, Cent/kWh (a)												
Capital Recovery (25% annualy)	2.07	2.07	2.07	2.30	2.36	2.42	2.49	2.97	2.99	3.03	3.06	3.10
Natural Gas (\$4/MMBtu)	2.85	2.73	2.61	2.42	2.38	2.36	2.36	2.26	2.24	2.23	2.22	2.22
O&M	0.51	0.51	0.51	0.47	0.47	0.47	0.47	0.86	0.85	0.85	0.85	0.85
Total	5.42	5.31	5.19	5.18	5.20	5.25	5.32	6.08	6.09	6.11	6.14	6.18

(a) Based on 95% On-Stream Factor

Figure I System Configuration (Ambient Pressure Operation)

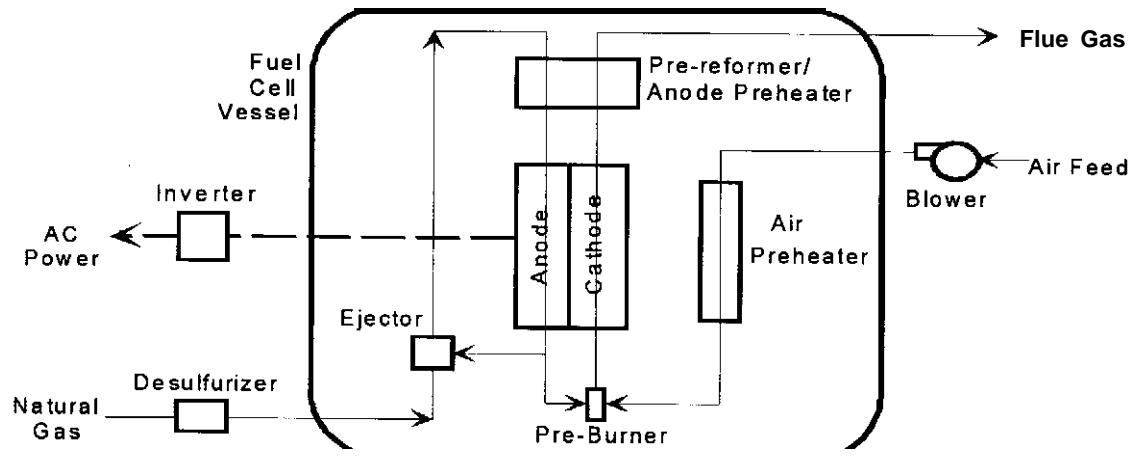
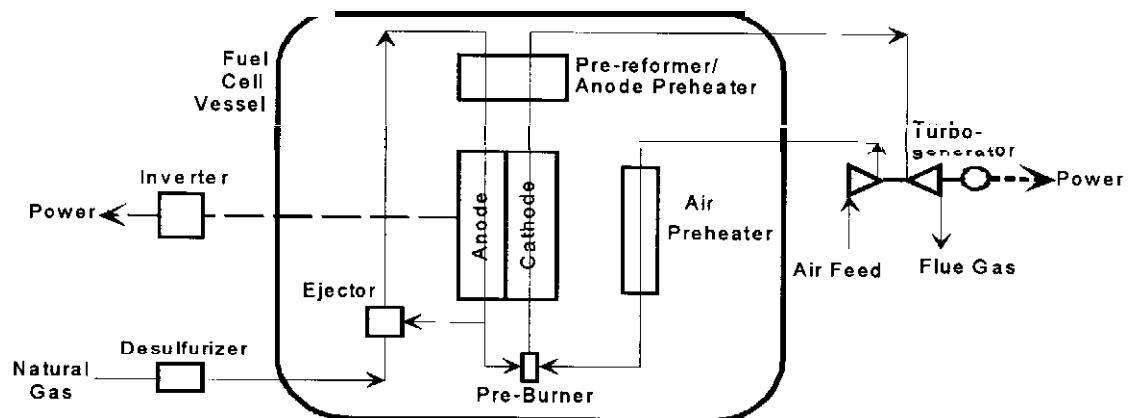
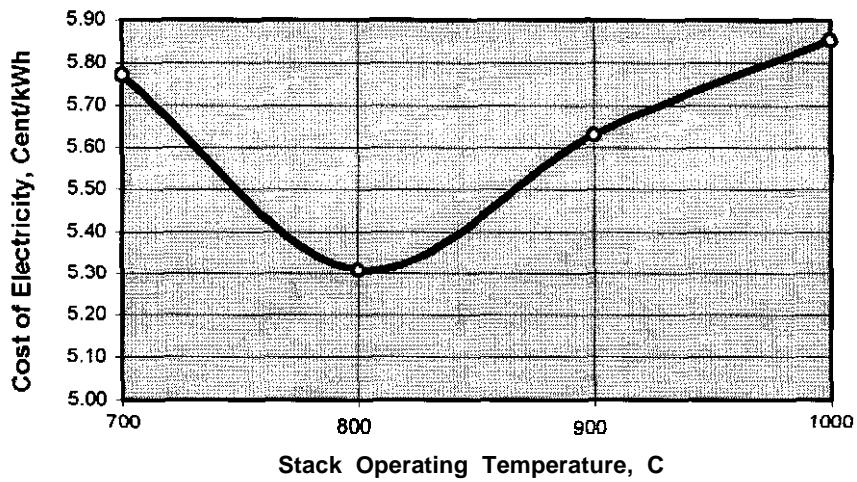


Figure II System Configuration (Pressurized Operation)



**Figure III**  
**Effect of Stack Operating Temperature on Electricity Cost**



## THIN-FILM FUEL CELLS

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### ABSTRACT

Bilayers consisting of highly porous substrates and dense thin-films of zirconia, ceria, and perovskite mixed conductors ( $\text{SrZrO}_3$ ,  $\text{SrCeO}_3$ , and  $\text{LaSrCoFeO}_3$ ) have been successfully fabricated using colloidal deposition techniques. The films are well bonded to the substrate, gas tight, and exhibit little interfacial resistance. Performance of solid oxide fuel cells (SOFCs) based on YSZ electrolytes of less than 10  $\mu\text{m}$  have achieved power densities close to 2 watts/cm<sup>2</sup> at 800°C. Ceria based SOFCs exhibited excellent electrochemical performance over the temperature range of 600 to 800°C with peak power in excess of 650 mW/cm<sup>2</sup> at 750°C and close to 300 mW/cm<sup>2</sup> at 600°C. Bilayers of thin  $\text{SrCeO}_3$  and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_3$  (LSCF) suitable for gas separation or membrane reactors have been fabricated as well as fuel cells based on proton conducting  $\text{SrZrO}_3$  films have also been fabricated and tested. The approach is generic and allows deposition of a variety of thin ceramic films on porous substrates. The methodology is scaleable and inexpensive, and should allow substantial cost-savings in the manufacture and operation of electrochemical devices in the intermediate temperature range of 500-800°C.

### INTRODUCTION

Ionic and mixed-conducting ceramic devices such as solid-oxide fuel cells, gas separation membranes, and membrane reactors require a dense electrolyte and highly porous electrodes. The motivation to fabricate thin film ceramic electrolytes derives from the benefits associated with lowering of ohmic losses across ionic and mixed ionic-electronic conducting materials as membrane thickness is reduced. When films are very thin (5-15  $\mu\text{m}$ ) the resistance of the electrolyte at intermediate temperatures is almost negligible, for example a ~10  $\mu\text{m}$  thick YSZ electrolyte has been shown to have an  $iR$  drop of 0.025  $\Omega\text{cm}^2$  at 800°C<sup>1</sup>. This allows the electrochemical device to operate at lower temperatures and higher thermodynamic efficiency where less expensive materials may be used in device construction. The technical challenge involves depositing pinhole and crack free dense layers of electrolyte 5 to 40  $\mu\text{m}$  in thickness on substrates of high porosity. The film must be well bonded to the substrate without excessive infiltration into the electrode porosity and there must be minimal interface polarization.

Several approaches to thin film fabrication have been reported including physical vapor deposition techniques<sup>2,3</sup>, tape calendaring<sup>4</sup>, sol-gel deposition<sup>5</sup>, sputtering<sup>6</sup> and in our group colloidal deposition. Many of these approaches have allowed the fabrication of high quality films, however, the high cost of capital equipment and/or operating costs for several of these

approaches presents a considerable barrier to their commercialization. In our laboratory we have focused on the use of wet chemical techniques such as colloidal deposition wherein a fine powder is dispersed in solution, deposited onto a green substrate, and fired such that the film fully densifies and the substrate remains highly porous.

Colloidal deposition of dense electrolyte layers on porous substrates requires that the materials are chemically compatible at the processing temperature and there must be adequate thermal expansion match between the layers. It is critical to develop a thorough understanding of the sintering behavior of both film and substrate materials. Once compatible materials have been selected, fabricating dense films of 5-40  $\mu\text{m}$  is achieved by careful control of the sintering profile (shrinkage vs. temperature) and the magnitude of the shrinkage of the materials. This is accomplished by systematically modifying the sintering profiles of film and substrate through control of particle size and morphology of green substrates. Poor understanding of these parameters often leads to electrolyte films of low density (pinholes) or cracked films composed of islands of high-density film. Even in cases where the shrinkage of film and substrate are sufficiently close to generate dense electrolyte films, residual stresses can lead to highly distorted films with significant curling. Importantly, the electrode substrate must be processed to yield continuous porosity and a high surface area microstructure, without compromising the strength of the bilayer.

At Lawrence Berkeley National Laboratory (LBNL) we have perfected the technique of colloidal deposition whereby high quality films of a wide variety of ionic and mixed ionic-electronic conductors can be deposited onto porous electrode substrates. Sintering profiles of films and substrates are matched to the extent that bilayers can be free sintered to a high degree of flatness with no compressive load (or with minimal load). Using these techniques, we have fabricated bilayers having dense electrolyte films of many electrolytes including yttria-stabilized zirconia (YSZ)<sup>1,7,8,9</sup>, ceria<sup>10</sup>, strontium cerate<sup>10</sup>, and LSCF. Colloidal deposition is a flexible process in that a wide variety of materials can be deposited as thin films with no (or minimal) alterations to fabrication equipment. Further, only small amounts of material are needed for bilayer fabrication making this approach suitable for novel or expensive conductors. Thin-film solid oxide fuel cells (SOFCs) fabricated using these techniques have demonstrated exceptional performance at reduced temperatures relative to conventional thick-film SOFCs.

## EXPERIMENTAL

*Porous Substrate:* Yttria-stabilized zirconia (YSZ) powders were obtained from Tosoh Corporation and nickel oxide powders (NiO) from J.T. Baker Inc. Ceria powders,  $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.8}$  (CGO), were supplied by NexTech Materials, 720 Lakeview Plaza Blvd., Worthington, OH, 43085. The substrate mixture containing approximately 45 w/o electrolyte (YSZ or CGO), 45 w/o NiO and a 10 w/o pore former (cornstarch) was attritor milled in acetone. The finely ground mixture was then dried, re-ground using a mortar and pestle, and sieved to <100  $\mu\text{m}$ . Samples were weighed out and pressed in a 3.81 cm diameter steel die. The green disks were then heated and held at the decomposition temperature of the pore former for 1 hour to ensure complete burnout of the cornstarch. The green disks were then partially fired to a green body strong enough to be handled for electrolyte deposition. Porous ceria or YSZ substrates were made in a similar manner, without NiO additions.

Thin-film Electrolyte: Electrolyte powders were obtained commercially (YSZ from Tosoh Corporation, CGO from NexTech Materials) or prepared by the glycine nitrate combustion process<sup>11</sup>. Electrolyte powders were dispersed in isopropanol with an ultrasonic probe and applied to the anode substrate. To avoid cracking of the thin film or warping of the substrate due to the 2D confinement of the film during sintering, the shrinkage of the substrate was carefully matched to that of the electrolyte. The electrolyte/electrode bilayer was fired at 1400-1500°C to fully densify the electrolyte film.

Cathode: Cathode powders of  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.98}\text{Ni}_{0.02}\text{O}_3$  (LSCN) and  $\text{La}_{0.85}\text{Sr}_{0.15}\text{MnO}_3$  (LSM) powders were prepared by the glycine nitrate process using a glycine to nitrate ratio of 0.5. These were mixed with the electrolyte material in approximately 50/50 weight percent ratio and applied to a 1 cm<sup>2</sup> masked off area of the bilayer. The structure was then fired at 1000-1250°C.

Current Collectors: Platinum paste (Heraeus OS3) was applied to both anode and cathode and fired to 950°C for 30 min. Platinum screens were then applied to the electrodes and bonded using Pt paste; Pt wires were spot-welded to these screens.

Cell Test Rig: The thin-film cell was sealed to the end of an alumina tube with Aremco cement. A type K thermocouple was fixed at the surface of the cathode current collector to obtain accurate fuel cell temperatures as a function of furnace temperature and current density during fuel cell operation. Hydrogen gas was saturated with water vapor at room temperature. Fuel cell temperature was monitored and controlled remotely through software written at LBNL.

Electrical Measurements: A high current galvanostat/potentiostat (PAR 371) was controlled by the use of an external voltage signal generated by software written in our lab using LabVIEW by National Instruments. This software allows current to be incremented monotonically for defined time increments as determined by the user. The program also automates current interrupt measurements across a cell with or without reference electrodes. Typically, currents were increased in 50 mA increments and the experiment was terminated when the cell potential dropped below 0.1 volt. The current interrupt method was used to separate the ohmic losses across the cell from other polarization losses. The interrupt current was typically greater than 750 mA/cm<sup>2</sup> and a minimum of two current interrupts were performed at each temperature.

## RESULTS AND DISCUSSION

Thin Ceramic Films: Thin films of oxygen conductors (YSZ), mixed conductors (CGO, LSCF), and proton conductors ( $\text{SrZrO}_3$ ) were successfully fabricated on porous substrates. A SEM micrograph of the fracture surface of a YSZ thin-film on a porous Ni-YSZ substrate is shown in Figure 1. The sample is at slight tilt so the top surface of the YSZ film can be seen along with the edge of the film and a portion of the porous substrate. Since the electrolyte is cofired onto the electrode, intimate bonding of electrode and electrolyte occurs leading to very low polarization of the bilayer under current load<sup>1,10</sup>. In Figure 2 the fracture surface of a CGO thin film on a porous Ni-Ceria substrate is shown. Thin films of  $\text{SrZr}_{0.95}\text{Y}_{0.05}\text{O}_3$  and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$  were also successfully fabricated and the SEM micrographs of fracture surfaces of these films are shown in Figures 3 and 4.

Cell Performance: The performance of cells (porous substrate/electrolyte/ cathode) were evaluated under current control using flowing  $H_2 + 3$  volume percent  $H_2O$  at the anode and static air at the cathode. Figure 5 shows the open circuit voltage of three different electrolyte systems. Figure 6 shows the current-voltage ( $i$ - $V$ ) curves for single cells of YSZ tested at 800°C and of CGO at 750°C. The performance of a thin-film Ni-YSZ/YSZ/LSM-YSZ fuel cell over the intermediate temperature range of 650-800°C is shown in Figure 7. As can be seen from the power density curves, the cell achieves a maximum power output of close to 2 watts/cm<sup>2</sup> at 800 °C. The performance of thin-film cells having LSM-YSZ cathodes is a strong function of temperature, due to polarization of the air electrode at temperatures below 800 °C. Mechanical integrity of YSZ thin films has been demonstrated by fuel cell tests lasting 1000's of hours at 800°C. Thin-film ceria based fuel cells also demonstrate excellent performance. In figure 8 the observed power density for a Ni-CGO/CGO/LSCN-CGO thin-film fuel cell is shown as a function of operating temperature. As can be seen from figure 8, thin-film ceria based cells achieve peak power densities of over 650 mW/cm<sup>2</sup> at 750 °C and close to 300 mW/cm<sup>2</sup> at 600 °C.

Figure 9 shows the fracture surface of a  $SrZr_{0.95}Y_{0.05}O_3$  thin film fuel cell after testing. Though the open circuit voltages (OCVs) were high the initial test performance of these proton conductors has been poor and the large resistive loss is attributed to a reaction layer between the electrolyte and cathode.

Voltage Losses: Due to the constraints of thin-film fuel cell geometry, only one reference electrode can be used in electrochemical testing (Figure 10). The use of a reference electrode allows separation of voltage losses into contributions from the anode and electrolyte/cathode components of the cell. For thin film cells tested in our laboratory at 800°C the ohmic resistance across the entire thin-film cell measured by current interrupt is typically in the range of 0.05 to 0.20  $\Omega cm^2$  dependent on the electrolyte. The cell resistance across a fuel cell is shown in Figure 11. The exceptionally low values for the area specific resistance (ASR) observed for thin-film fuel cells also highlights the impact of resistive interfaces in SOFCs due to interface instability and/or poor electrode processing. For state-of-the-art planar SOFCs where the YSZ electrolyte is about 200  $\mu m$  in thickness, values of the ASR are typically in the range of 0.8 to 1  $\Omega cm^2$  at 950°C. Remarkably, at operating temperatures 150°C lower than for thick-film cells, thin-film fuel cells exhibit almost an order of magnitude improvement in ASR. Notably, deterioration in the ASR for thick-film cells of 0.05 to 0.2  $\Omega cm^2$  would result in degradation of performance by 6 to 20%. However, this increase in ASR for a thin-film cell would effectively double the ASR and half the thin-film cell performance. In other words, as the electrolyte thickness in a SOFC is reduced to zero, ohmic losses across the electrolyte vanish, and interface/contact resistance and electrode polarization limit cell performance. Clearly, to attain and maintain high levels of performance in thin-film SOFCs, critical attention must be paid to electrode microstructure and processing in order to preclude formation of resistive interfaces.

Improving Device Performance: At 800°C YSZ thin film cells demonstrated excellent performance, achieving power densities of 800 mW/cm<sup>2</sup> at 0.8 volts and exhibiting maximum power densities of close to 2000 mW/cm<sup>2</sup>. However, as can be seen in Figure 12, the ohmic drop (as measured by current interrupt) across the cell increased rapidly below 800°C compared with

ceria electrolyte based cells. Improvement of the low temperature performance of YSZ based cells could be accomplished by replacing the LSM electrode with a cathode having better low temperature oxygen reduction kinetics such as LSC. In this case, a reaction barrier such as a 1  $\mu\text{m}$  layer of ceria might be necessary between the YSZ and LSC to prevent the formation of a resistive interface through chemical reaction. As can be seen from Figure 12, thin film ceria based cells have very low resistive losses. Still, in order to attain power densities greater than 650  $\text{mW/cm}^2$  (at 750°C) it might be necessary to reduce voltage losses due to electronic conductivity in the ceria electrolyte. This could be accomplished by either choosing a dopant composition that reduces the electronic conductivity of the ceria electrolyte, or by depositing a very thin ( $<1\ \mu\text{m}$ ) layer of YSZ to block electronic transport.

Table 1 - Summary of Systems Investigated

THIN FILM	SUBSTRATE	CATHODE
YSZ*	YSZ-NiO*	LSM-YSZ* LSCN-CGO
YSZ	$\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_2\text{-NiO}$	LSCN-CGO
$\text{Ce}_{0.9}\text{Y}_{0.1}\text{O}_2$ $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_2^*$	$\text{Ce}_{0.9}\text{Y}_{0.1}\text{O}_2\text{-NiO}$ $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_2\text{-NiO}^*$ $\text{Ce}_{0.9}\text{Y}_{0.1}\text{O}_2\text{-NiO}$	LSM-CYO LSCN-CGO* LSCF-CGO
$\text{Ce}_{0.85}\text{Sm}_{0.15}\text{O}_2^*$	$\text{Ce}_{0.85}\text{Sm}_{0.15}\text{O}_2\text{-NiO}^*$ $\text{Ce}_{0.85}\text{Sm}_{0.15}\text{O}_2\text{-NiO}^*$	LSCN-CGO* LSM-CSO*
$\text{SrCe}_{0.9}\text{Y}_{0.1}\text{O}_3$ $\text{SrZr}_{0.95}\text{Y}_{0.05}\text{O}_3$	$\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_2\text{-NiO}$ $\text{Ce}_{0.85}\text{Sm}_{0.15}\text{O}_2\text{-NiO}$ YSZ-NiO	LSCN-SrCeYO LSM-SrZrYO
$\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_3$	$\text{Ce}_{0.85}\text{Sm}_{0.15}\text{O}_2$	
$\text{LaSrGa}_3\text{O}_7$ (note: reacted with Ni)	YSZ-NiO with ceria barrier layer	

\*Denotes systems that exceeded 500  $\text{mW/cm}^2$

## CONCLUSIONS

The broad application of bi-layer fabrication by colloidal techniques has been demonstrated by making and testing several ionic and mixed ionic-electronic ceramic thin film devices. Systems investigated are summarized in Table 1. Thin film SOFCs fabricated using these techniques demonstrate excellent performance; YSZ based cells have achieved maximum power densities of close to 2000  $\text{mW/cm}^2$  at 800°C and CGO based cells have achieved power densities above 650  $\text{mW/cm}^2$  at 750°C. Current interrupt techniques indicate that the majority of the efficiency loss is due to ohmic losses associated with the cathode or cathode/electrolyte interface resistance. The goal of improving fuel cell performance at lowered temperatures by reducing the electrolyte thickness has been realized. Electrochemical characterization of the thin film fuel cells has indicated that ohmic drop across the electrolyte layer is almost negligible. Clearly, limitations to performance have been shifted from the electrolyte to interfacial and charge transfer resistance as well as mass transfer polarization at high current densities. Improved performance of thin film SOFCs, particularly at further reduced temperatures, will be

accomplished through systematic studies of alternative cathode materials and electrode microstructures.

## ACKNOWLEDGMENTS

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## FIGURES

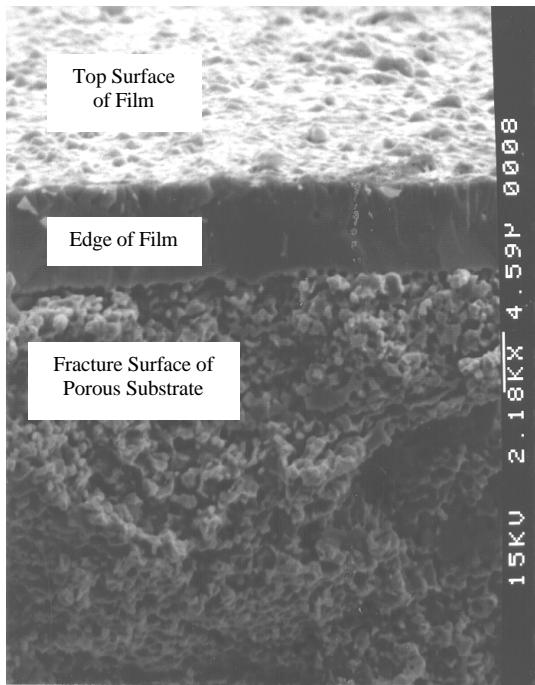


Figure 1. 7 $\mu$ m YSZ on Ni-YSZ Substrate

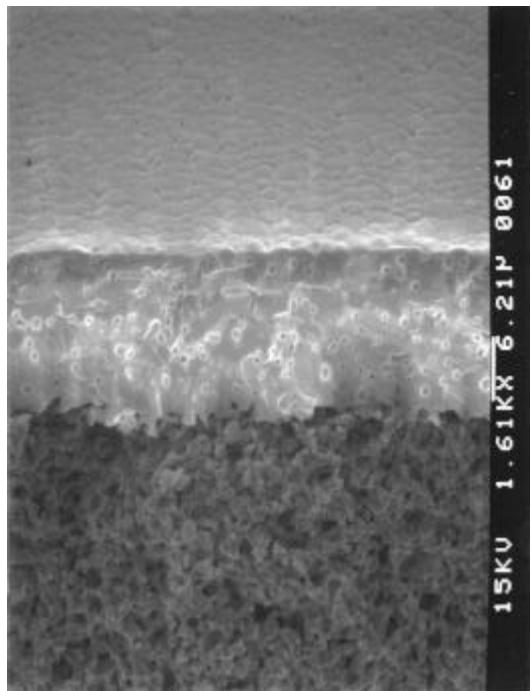


Figure 2. 15 $\mu$ m CGO on Ni-Ceria Substrate

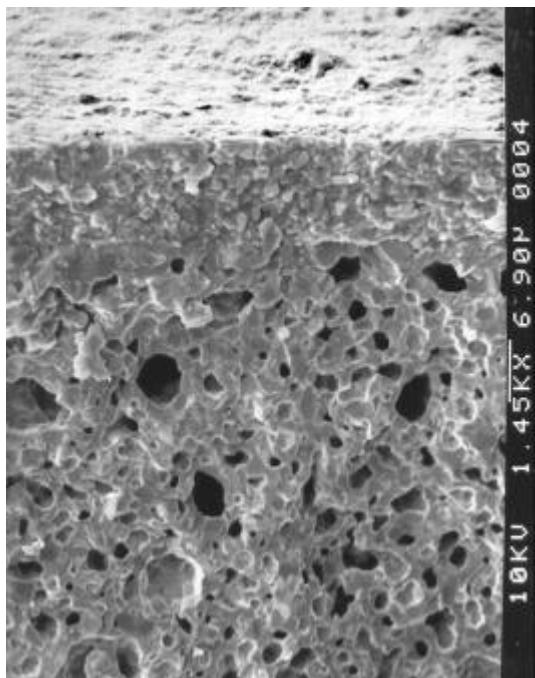


Figure 3. 10  $\mu$ m  $\text{SrZr}_{0.95}\text{Y}_{0.05}\text{O}_3$  on NiO-ceria.

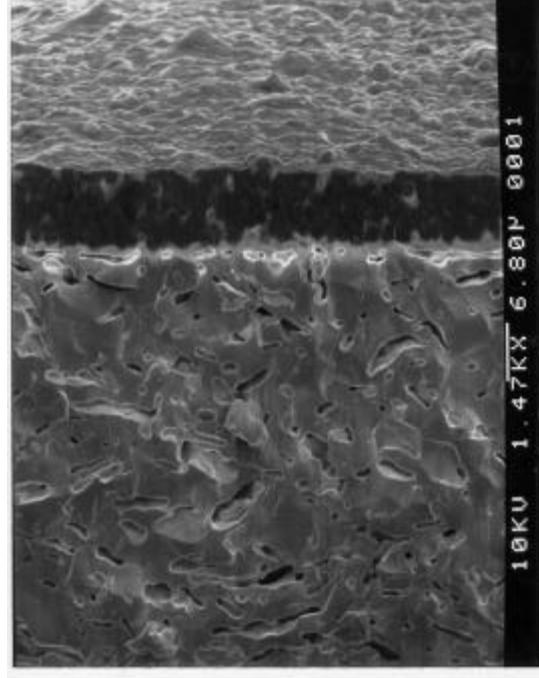


Figure 4. 8  $\mu$ m  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_3$  on ceria.

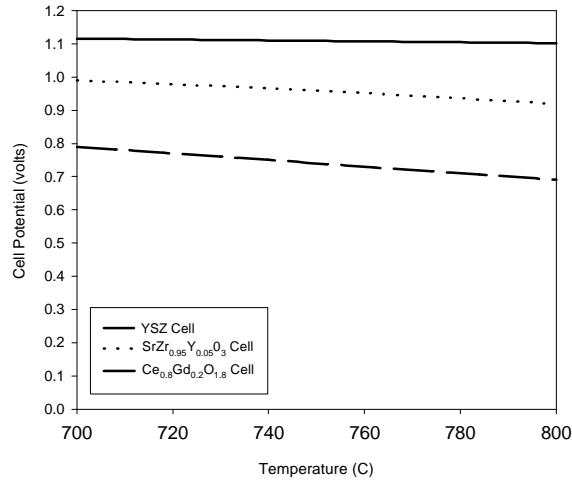


Figure 5. Open circuit potentials of three electrolyte cells in air/  $H_2$  + 3 v/o  $H_2O$ .

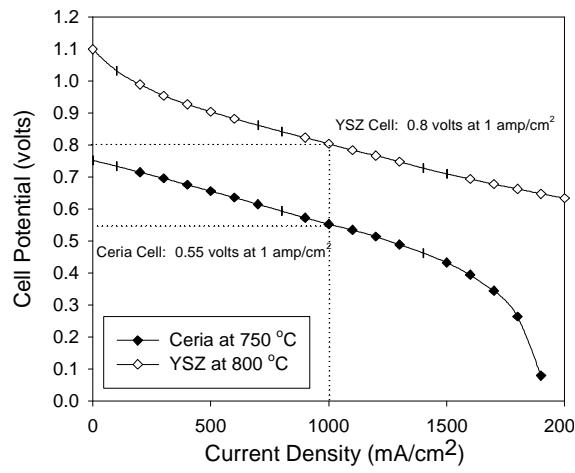


Figure 6.  $i$ - $V$  characteristics of YSZ and CGO cells (air/cathode/electrolyte/anode/  $H_2$  + 3 v/o  $H_2O$ ).

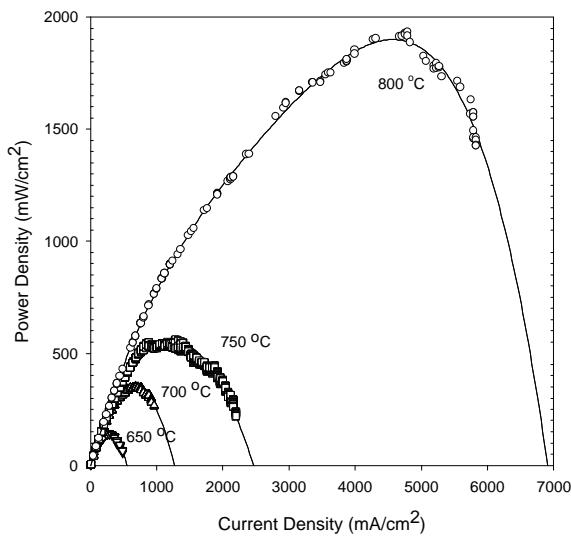


Figure 7. Performance of thin-film Ni-YSZ/YSZ/LSM-YSZ fuel cell as a function of cell temperature.

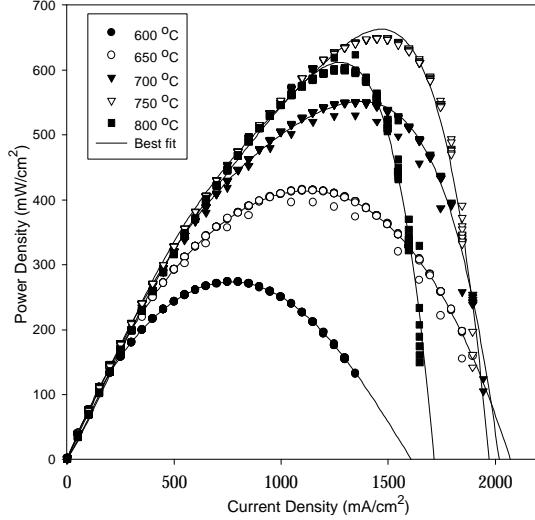


Figure 8. Performance of thin-film ceria cell as a function of temperature.

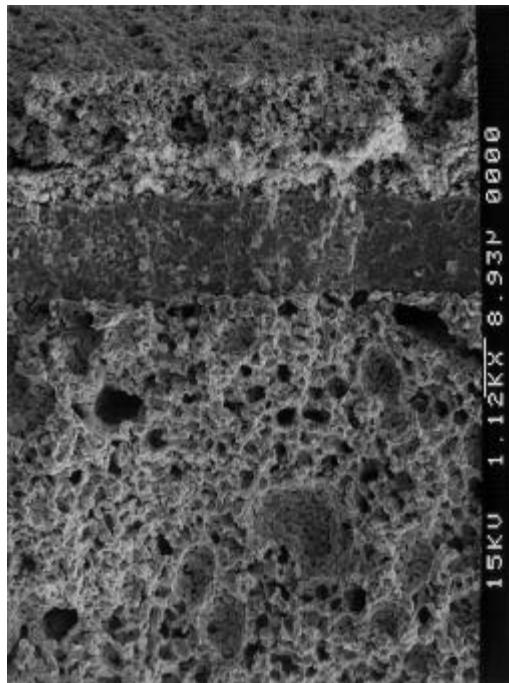


Figure 9.  $\text{SrZr}_{0.95}\text{Y}_{0.05}\text{O}_3$  fuel cell with LSCN cathode after testing.

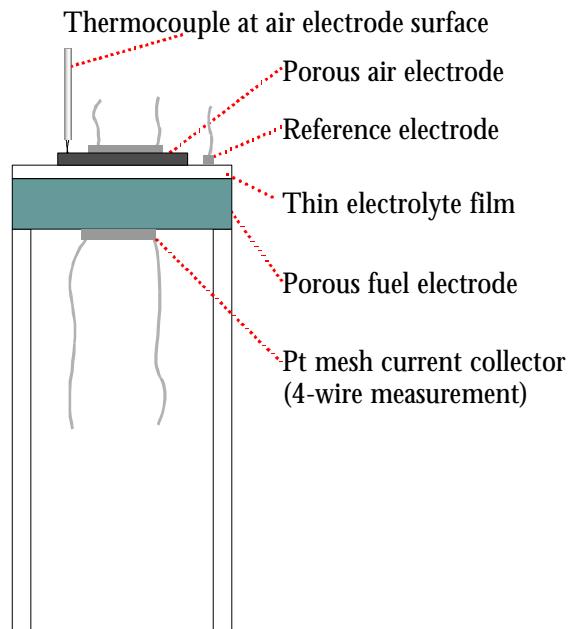


Figure 10. Location of reference electrode in cell geometry

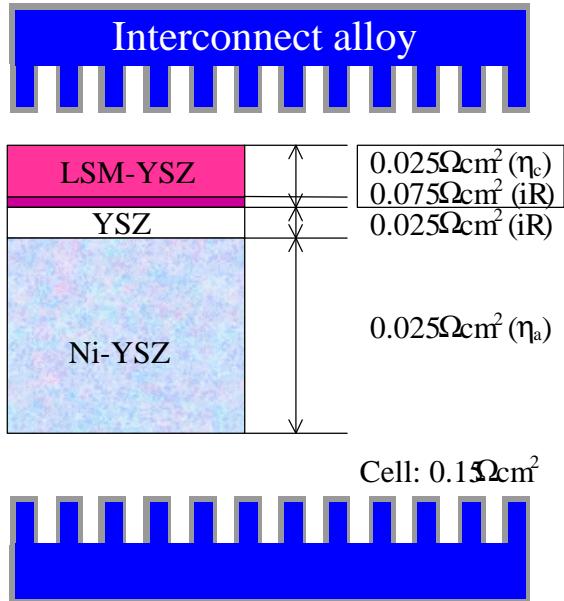


Figure 11. Importance of low resistance interconnect for thin-film SOFC

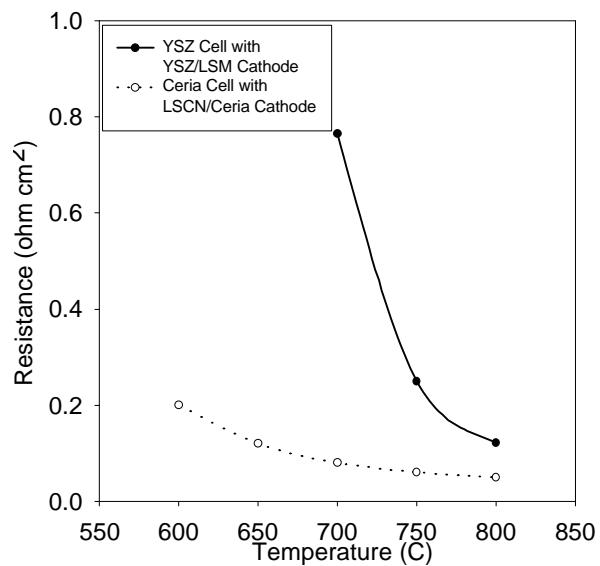


Figure 12. Total ohmic drop ( $iR$ ) of YSZ cell with LSM/YSZ cathode and of CGO cell with LSCN/CGO cathode.

# Advanced Materials for Solid Oxide Fuel Cells: Mechanical Properties of Lanthanum Chromite

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## Introduction

Solid oxide fuel cells (SOFCs) with high power density and efficiency are currently being developed for a variety of mobile and stationary power applications. SOFCs are solid-state devices consisting of two porous electrodes (anode and cathode) separated by an ionically conducting electrolyte, typically stabilized zirconia. The electrode materials commonly used are nickel/zirconia cermet for the anode and doped lanthanum manganite for the cathode. The final component is the interconnect which connects the individual cells in series.

The interconnect, which provides the electrical pathway from the anode of one cell to the cathode of the adjacent cell, must be chemically and physically stable in both reducing and oxidizing environments. The interconnect material must be compatible with all other cell components, must be electrically conducting, and have negligible ionic conductivity. In the planar SOFC design, the interconnect is the load bearing component that supports the other cell components, and must therefore also have adequate strength at high temperature. Only acceptor substituted lanthanum chromites meet these severe environmental, thermal, and structural requirements.

The fundamental material properties of lanthanum chromite under oxidizing conditions have been measured and reported extensively [1,2]. However, there has been only limited characterization of the mechanical properties of these materials [3-10]. The flexural strength has been characterized, both at room temperature and high temperature under oxidizing conditions, using 3 and 4 point sample geometries. In general, room temperature strength increased with increasing acceptor content; the increase was attributed to an increase in sintered density [3]. The strength of both strontium and calcium substituted chromites was found to decrease with increasing temperature [4,5].

Initial investigations into the dependence of the strength on reducing atmospheres have shown mixed results. Milliken *et al.* [6] reported a 50% increase in bend strength of  $\text{La}_{0.83}\text{Sr}_{0.16}\text{CrO}_3$  samples after exposure to an oxygen partial pressure ( $\text{P}(\text{O}_2)$ ) of  $\sim 2 \cdot 10^{-18}$  atm at  $1000^\circ\text{C}$  relative to unexposed samples, whereas Montross *et al* [7] did not observe any improvement in strength for similar compositions in a reducing ( $\text{H}_2$ ) atmosphere. In addition, Montross reported that the

bend strength at 1000°C of  $\text{La}_{0.85}\text{Sr}_{0.15}\text{CrO}_3$  and  $\text{La}_{0.70}\text{Sr}_{0.30}\text{CrO}_3$  in hydrogen was significantly less than that in air, while  $\text{La}_{0.80}\text{Sr}_{0.20}\text{CrO}_3$  was relatively unaffected. Preliminary studies by Paulik and Arsmtrong [11,12] have shown significant strength losses after annealing in low oxygen environments, indicating the need for additional characterization and microstructural control in the lanthanum chromites to ensure adequate long-term performance in fuel cell environments .

## Objective

It was the objective of this study to provide an assessment of acceptor (both calcium and strontium) substituted lanthanum chromite in terms of their mechanical behavior and performance in application environments. Strength, toughness and elastic properties were characterized as a function of acceptor concentration and temperature, over the range of oxygen partial pressures.

## Experimental Procedure

$\text{La}_{1-x}\text{Ca}_x\text{CrO}_3$  and  $\text{La}_{1-x}\text{Sr}_x\text{CrO}_3$  powders (with acceptor concentrations varied from 0.15 to 0.3) used in this study were synthesized using the glycine-nitrate combustion process (Praxair Specialty Ceramics, Seattle, WA), and then calcined in air at 1000°C for 1 hr. Due to the high surface area and low packing density of the as-synthesized powder, a processing sequence involving pressing, grinding of the pressed compact, sieving the ground powder through a 150- $\mu\text{m}$  size screen, followed by a final consolidation (isostatic pressing at 276 MPa), was required to obtain high green densities between 65 and 68% of theoretical. The green billets, • 34 x 34 x 64 mm in size, prepared in this manner were sintered in air between 1600 and 1690°C for 2 to 6 hr, and then cooled slowly at 2°C/min to obtain dense crack-free samples. Selected billets were machined into 3 x 4 x 45 mm size bars for flexural tests. Additionally, one billet of each composition was sliced and polished into 10 mm thick square samples, with parallel top and bottom surfaces, for modulus and indentation measurements. Sample densities were measured using the Archimedes method with ethyl alcohol.

Four point bend strengths were measured (Instron model 1125) with a crosshead speed of 0.5 mm/min using a fully articulated fixture with a 20 mm inner and a 40 mm outer span. Flexural strengths were measured in air at 25, 600, 800 and 1000°C. A minimum of ten samples corresponding to each annealing treatment was tested. Samples were heated at approximately 20 to 25°C/min and allowed to equilibrate for 15 min prior to testing.

Selected flexure samples were annealed at 1000°C in a reducing environment for 2 hours, and then tested at room temperature. Samples were first heated to 1000°C in air in a silica tube furnace and allowed to equilibrate. The gas environment was then controlled with a buffered  $\text{CO}_2/\text{Ar}/\text{H}_2$  system metered using mass flow controllers. The use of this system allowed the oxygen partial pressure to be accurately controlled from  $10^{-5}$  to  $10^{-18}$  atm. During cooling, constant  $\text{P}(\text{O}_2)$  was maintained to approximately 700°C.

Fracture toughness was evaluated by the indentation crack-measurement technique [3] for all the  $\text{La}_{1-x}\text{Ca}_x\text{CrO}_3$  and  $\text{La}_{1-x}\text{Sr}_x\text{CrO}_3$  compositions as a function of the annealing treatment. A 5-kg

indentation load (Zwick of America, Inc., E. Windsor, CT) with a 30-s residence time was used. Five indentations and the associated radial cracks, corresponding to each experimental condition were measured and averaged to determine the hardness and fracture toughness. Elastic moduli required for the toughness calculations were obtained on polished samples prior to indentation by the sonic pulse technique [14].

The initial oxygen content of sintered samples was determined using an potentiometric (oxidation-reduction) titration technique described elsewhere [15]. Samples were then annealed at various oxygen partial pressures for 2 hours. After annealing, samples were reoxidized in a thermogravimetric analyzer (TGA, Cahn Microbalance). The relative weight changes were measured and corrected for buoyancy effects, and the oxygen content of the annealed samples was determined using the initial oxygen content and the electroneutrality equation.

Scanning electron microscopy (SEM) was used to examine polished sections and fracture surfaces of samples. The grain size was measured using standard stereological techniques described by Underwood [16]. As-sintered and reduced samples were ground to <100 mesh size powder using a mortar and pestle and analyzed by x-ray diffraction [Phillips XRG3100]. X-ray spectra were collected from 15 to 75° 2θ at 0.04° increments for 2 s using Cu-K<sub>α</sub> radiation.

## Results

### *Microstructure*

For convenience, calcium substituted lanthanum chromites will be designated by LCC, followed by numerals referring to the amount of dopant added. For example,  $(La_{0.8}Ca_{0.2})CrO_3$  is denoted as LCC-20. The average sintered densities of both LSC and LCC samples are listed in Table 1. The calcium doped lanthanum chromite (LCC) samples demonstrated increasing density with increasing acceptor content similar to previously reported data by Chick et al. [17,18] for strontium additions to lanthanum chromite. The addition of calcium to LaCrO<sub>3</sub> allows the formation of a transient liquid phase, CaCrO<sub>4</sub>, above 1150°C, which enhances densification [19,20], resulting in sintered microstructures with grain sizes ranging from 6 to 9 μm. For the strontium substituted lanthanum chromite (LSC) samples, densities and average grain sizes were slightly less than similarly doped LCCs, except for the composition with only 16% A-site substitution, which developed a bimodal grain size distribution.

SEM examination of polished and thermally etched (1500°C) sections revealed small quantities of a grain boundary phase in the LSC-20, LSC-24, LCC-20, LCC-25 and LCC-30 samples. It is useful to point out that the thermal etching treatment can cause some exsolution of dopant-rich phases. However, it is likely that even the materials slow-cooled from the sintering temperature are not single phase, since some second phase inclusions are observed in fracture surfaces of samples tested in air at much lower temperatures (<<1000°C). Energy dispersive x-ray spectroscopy (EDS) revealed calcium-rich phases in fractured surfaces of as-sintered LCC samples, and strontium/lanthanum-rich phases in fractured surfaces of reduced ( $PO_2 = 10^{-16}$  atm., 100°C) LSC samples. From the polished (thermally etched) cross-sections, it was apparent that LSC samples had more grain boundary phase than similarly doped LCC samples, especially with the higher acceptor contents. Minor phases, when present in the sintered samples, were below

detectable limits by X-ray diffraction. At room temperature, the diffraction patterns revealed only the orthorhombic phase in as-sintered LCCs, and the rhombohedral phase in the LSCs.

### *Flexural Strength Behavior*

#### Compositional Effects on Strength

The room temperature bend strengths of selected LCC and LSC compositions as a function of acceptor content are listed in Table I. The room temperature strength of the LCC samples increased with increasing calcium content. The density of LCC-15 was only •87% of theoretical, so a significant strength improvement was realized by increasing the dopant level to 20% and attaining high sintered density (•95%). Beyond the 25% Ca-dopant level, density and strength did not change significantly. The overall observations on density variation with composition shows good agreement with other studies [3]. With the Sr-doped compositions, strength of the LSC-16 material was limited by the exaggerated grain growth that occurred as a result of a nonuniform distribution of  $\text{SrCrO}_4$  that forms during sintering. Improved strengths are obtained with higher amounts of dopants (LSC-25 and -30), which promotes liquid-phase sintering and prevents discontinuous grain growth.

Table 1. Density, Grain Size and Flexural Strength of Acceptor-Substituted Lanthanum Chromites

Composition	% Theoretical Density	Grain Size ( $\mu\text{m}$ )	Strength (MPa, 25°C)
LCC-15	87.3	$7.0 \pm 1.2$	$61.0 \pm 11.5$
LCC-20	94.9	$6.2 \pm 1.1$	$96.0 \pm 14.4$
LCC-25	96.4	$9.0 \pm 1.3$	$122.7 \pm 26.0$
LCC-30	97.2	$6.6 \pm 0.8$	$107.4 \pm 5.9$
LSC-16	93.1	Bimodal: $2.9 \pm 0.4$ $46.7 \pm 17.2$	$48.6 \pm 10.8$
LSC-20	91.6	$3.7 \pm 0.8$	$75.7 \pm 8.3$
LSC-24	91.2	$4.1 \pm 0.7$	$65.6 \pm 9.6$

#### Temperature Effects on Strength

Bend strengths for the LCC (-20, -25 and -30) as a function of temperature are shown in Figure 1. All LCC compositions showed a decrease in strength at 600°C relative to room temperature. This is in good agreement with the strength variations recently observed by Montross et al [10] for an LCC-20 material. According to Montross [10] the loss in strength at low temperatures was attributed to a progressive decrease in “transformation-toughening” contributions with increasing temperature, since the strength loss with temperature as well as an observed grinding-induced phase transformation phenomenon were similar to the behavior of transformation-

toughened zirconia. This interpretation of the strength behavior appears to need further corroboration, since transformation-toughening does not appear to play any role in the chromites. The orthorhombic chromite phase at room temperature is the equilibrium phase, unlike transformation-toughened zirconia where the high temperature tetragonal phase has to be retained metastably at room temperature for high toughness and strength. In the samples used in this study, no grinding-induced transformation was noted - the phase composition of as-fired, ground and polished surfaces of samples were identical.

A more likely explanation for the strength loss in LCCs with increasing temperature is a decrease in toughness or introduction of strength-controlling flaws due to the phase transition at  $\bullet 300^{\circ}\text{C}$ . Upon heating from room temperature, the LCC compositions undergo a phase transition from orthorhombic to rhombohedral symmetry, similar to undoped  $\text{LaCrO}_3$  [21,22]. The phase transformation is detectable by dilatometry, as shown in Figure 2, where dilatometric traces for 20% acceptor-substituted chromites are compared to undoped  $\text{LaCrO}_3$ . The measured strength at  $225^{\circ}\text{C}$  is identical to the room-temperature strength, but decreases significantly above  $300^{\circ}\text{C}$ . The significant decrease across a narrow temperature range at such low temperatures for such an inherently refractory oxide (the lanthanum chromites were considered for high temperature fiber reinforcement applications [23]) is most likely due to transformation-weakening, where the phase transition at  $250^{\circ}\text{--}300^{\circ}\text{C}$  results in a high temperature rhombohedral phase with lower toughness or defects from the dimensional changes. With further increase in temperature, from  $500^{\circ}\text{C}$  up to  $1000^{\circ}\text{C}$ , no significant change in strength were observed for the LCC-20 and -25 compositions, but the strength of the LCC-30 continued to decrease steadily with temperature. With the highest dopant level (LCC-30), residual calcium-rich second phases can lead to lower strengths with increasing temperature, especially since the test temperatures ( $800\text{--}1000^{\circ}\text{C}$ ) are a significant fraction ( $>>0.5T_m$ ) of the melting points of calcium chromate phases [17-19] that form in this system. It is well established that transient low-melting calcium chromate phases form in the Ca-doped chromites [17,18, 20], and a complex series of reactions control the final phase composition in sintered chromites [19, 20]. A-site dopant levels at 20-25% are barely adequate for sintering, resulting in almost complete redissolution of the calcium-rich phases into the grains during high temperature treatment [19], but additional dopant amounts can lead to excess calcium chromate

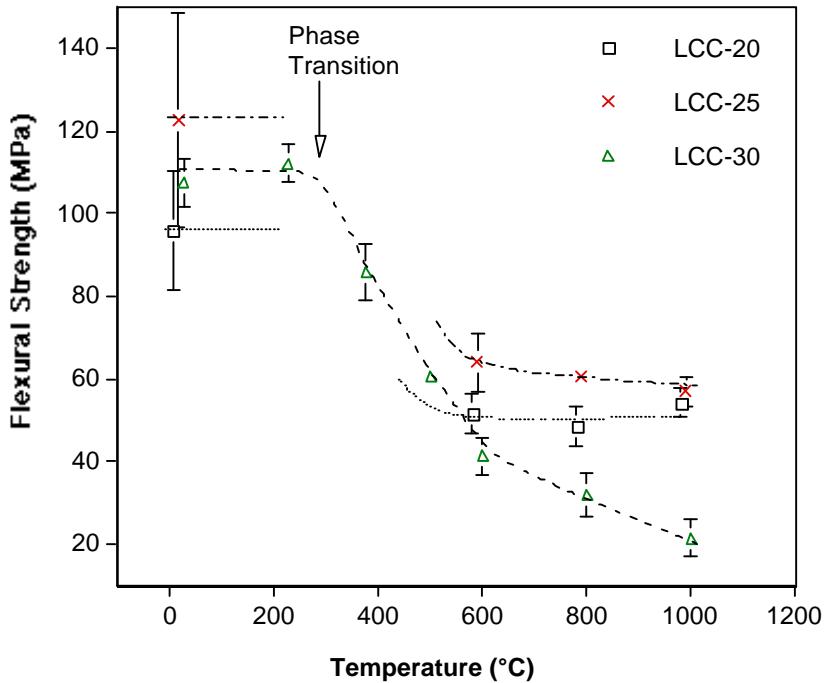


Figure 1. Flexural strength as a function of temperature for Ca-substituted lanthanum chromites

phases. Inspection of the fracture surfaces by SEM confirmed that all the LCC samples tested in air failed by intergranular cracking. But the LCC-30 samples tested at 1000°C showed significant non-linear deflection behavior with distinct curvature evident in the broken bars. This is indicative of fracture being preceded by deformation due to low-melting residual second phases at this dopant level.

Bend strengths for the LSCs (-16, -20 and -24) as a function of temperature are shown in Figure 2. The average four-point bend strengths are in range 50-80 MPa which are essentially comparable to the 600 °C strengths reported by Sammes and Ratnaraj [8] for LSC-20, after taking into account their use of 3-point flexure geometry and a small loading span, which gives higher strengths than the four-point geometry used in this study. The strength of the LSC samples remained unchanged over the entire temperature range from room temperature to 1000°C. Unlike the LCCs, the rhombohedral phase in the LSCs are stable with increasing temperature, resulting in a constant strength in the low temperature regime, with no inflection in the thermal expansion. At higher temperatures, 600°C-1000°C, constant strength is maintained, primarily because of low amounts of residual Sr-rich second phases at the 16-24% dopant level, and possibly because second phases do not have a significant effect on fracture when material densities are only 91-94% of theoretical. This strength retention with temperature for the LSCs is in contrast with the results of Sammes and Ratnaraj [8] who report a decrease in strength by •30-40% from 600°C to 1000°C in samples with various compositions (LSC-10,-20 and -30) and densities (•88%, •95% and 97% of theoretical). Intergranular phases play a major role in controlling strength, and discrepancies in the measured strengths values can arise from the differences in the amounts and composition of these second phases in the test samples at temperature.

### Environmental Effects on Strength

Figures 3 and 4 are plots of the room temperature strength as a function of  $P(O_2)$  for the annealed LCC and LSC samples, respectively. The results indicate that both acceptor content and type affect

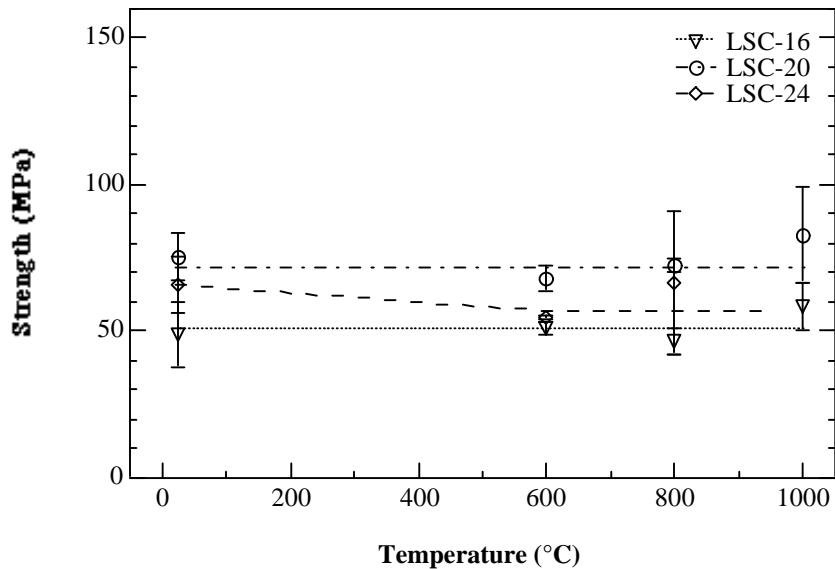


Figure 2. Flexural strength as a function of temperature for Sr-substituted lanthanum chromites

the mechanical performance of the material under reducing conditions. With the LCC materials, significant losses in strength were observed after annealing below a critical oxygen partial pressure. The reduction in strength was most significant with the highest dopant level (LCC-30). Furthermore, the loss in strength occurred at higher oxygen pressures with the higher dopant levels. These environmental effects on the strength behavior are related to structural changes in the chromite lattice (grains), which also results in significant changes in fracture morphology. SEM micrographs in Figure 5 illustrate a transition in fracture mode, from intergranular to transgranular fracture, that accompanies this decrease in strength observed upon annealing at progressively lower oxygen pressures. It is apparent that the structural changes caused by annealing at very low oxygen partial pressures lowers the cohesive strength of the chromite lattice.

The environmental effects on strength behavior of LSCs (Figure 4) are qualitatively similar to that of the LCCs, except much lower oxygen partial pressures are required to significantly affect the retained strength. A slight increase in strength is observed with decreasing oxygen pressure in the annealing treatment, before strengths begin to decrease after very low oxygen heat-treatments. Again, the materials with higher dopant levels begin to show strength losses after annealing at higher oxygen pressures. The fracture surfaces illustrated in Figure 6 also indicate that structural changes in the lattice upon annealing at low oxygen environments results in lower grain strengths, i.e., cohesive strength in the chromite lattice is reduced, leading to transgranular

fracture. The theoretical cohesive strength of the lattice is controlled by the fracture energy and the elastic modulus of the material [24], both of which can also undergo changes with annealing at low oxygen pressures.

### Fracture Toughness and Elastic Modulus

The variation in indentation fracture toughness with annealing treatment for LSC (LSC-24) and an LCC (LCC-25) samples are shown in Figure 7. Prior to heat-treatment, the fracture toughness of the as-sintered LSC sample was  $\bullet 1.1 \text{ MPa}\cdot\text{m}$ , and the LCC material  $\bullet 2.1 \text{ MPa}\cdot\text{m}$ . Similar to the flexural strength, the average toughness decreased significantly below a critical oxygen pressure. The indentation cracks also showed, in general, more intergranular fracture after high oxygen pressure treatments, and evidence of transgranular fracture after heat treatments at very low

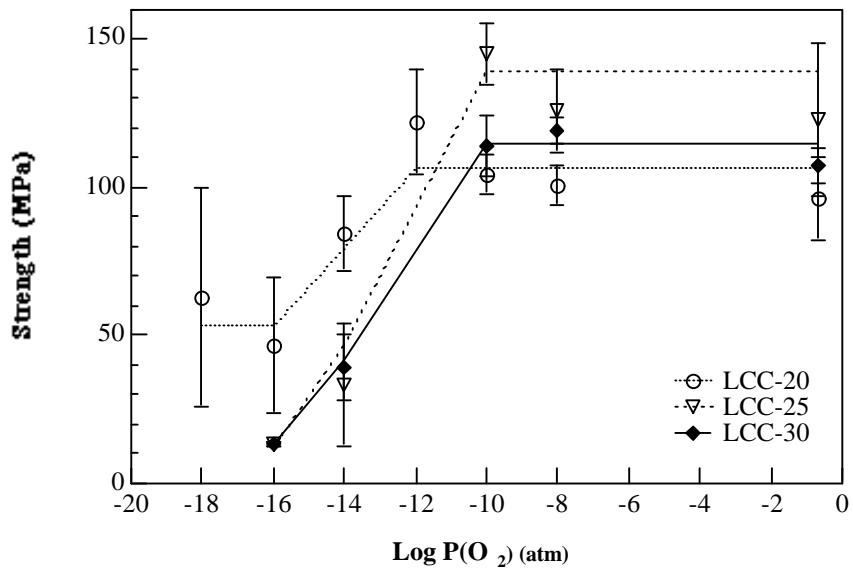


Figure 3. Room-temperature strength of annealed Ca-substituted lanthanum chromites as a function of the oxygen partial pressure used in the 1000°C heat-treatment

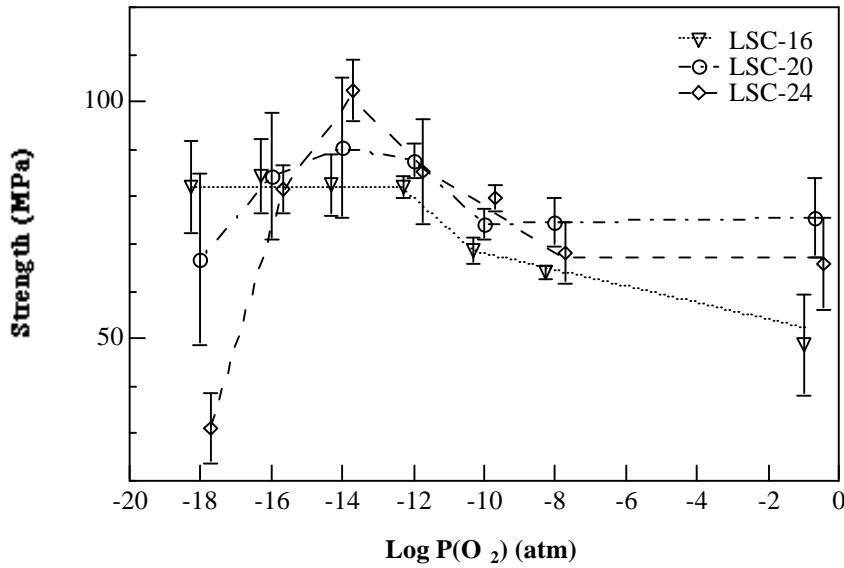


Figure 4. Room-temperature strength of annealed Sr-substituted lanthanum chromites as a function of the oxygen partial pressure used in the 1000°C heat-treatment

oxygen pressures, indicating that structural changes in the lattice also lowers the fracture surface energy of the perovskite grains. This behavior showing decreased fracture toughness after heat treatments at low oxygen pressures is similar to the fracture behavior observed in  $\text{Y}_2\text{O}_3$  [25] where  $K_{\text{Ic}}$  was found to decrease by  $\sim 30\%$  after annealing at 1700°C for 2h in a vacuum of  $\sim 13.3$  kPa. For the reduced yttria material, the lower toughness was correlated to a high concentration of oxygen vacancies, and reduced ionic character of the Y-O bond which lowers the bond strength.

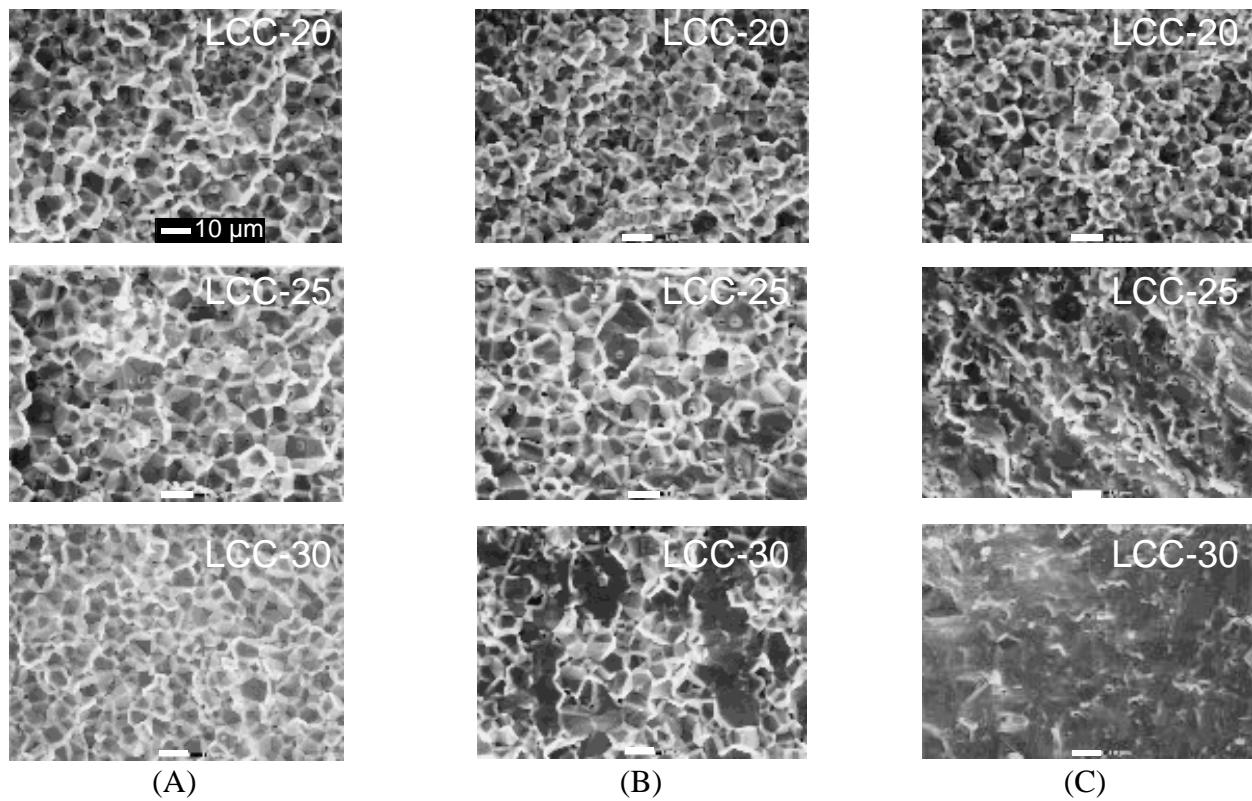


Figure 5. SEM micrographs of fracture surfaces of annealed Ca-substituted lanthanum chromite bar specimens. Samples annealed in (A) air, (B)  $10^{-10}$  atm.  $P(O_2)$  and (C)  $10^{-16}$  atm.  $P(O_2)$ . Note the transition from intergranular to transgranular fracture with decreasing oxygen partial pressures in the annealing treatment.

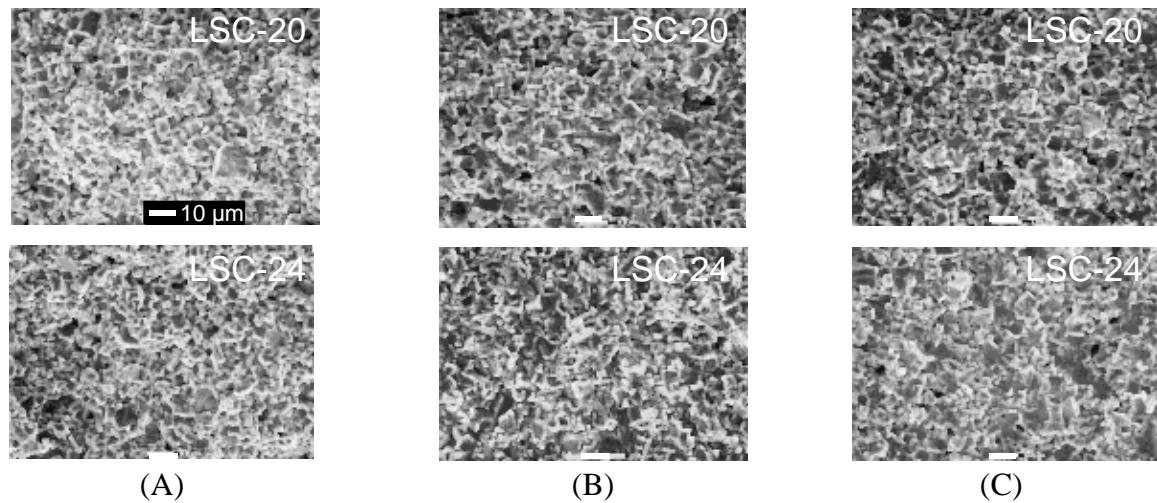


Figure 6. SEM micrographs of fracture surfaces of annealed Sr-substituted lanthanum chromite bar specimens. Samples annealed in (A) air, (B)  $10^{-14}$  atm.  $P(O_2)$  and

(C)  $10^{-18}$  atm.  $P(O_2)$ . Note the transition from intergranular to transgranular fracture with decreasing oxygen partial pressures in the annealing treatment.

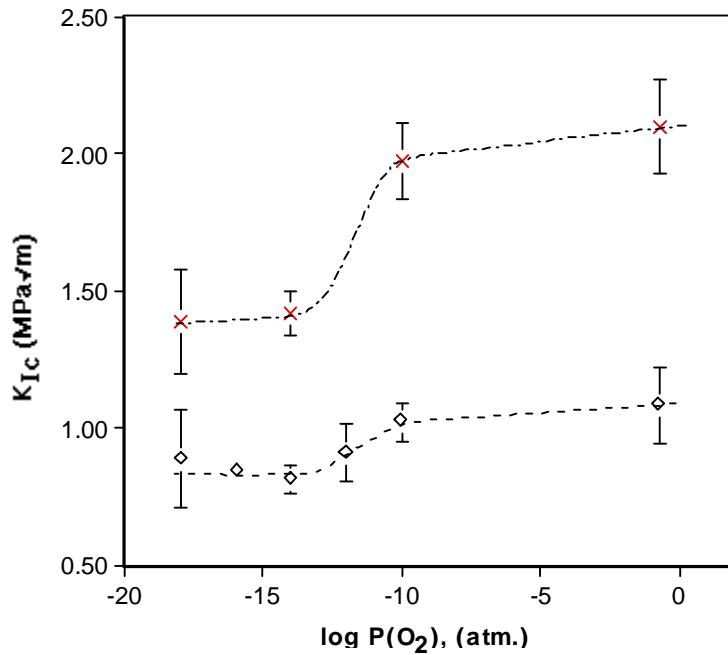


Figure 7. Indentation fracture toughness of annealed LSC-24 and LCC-25 samples as a function of oxygen partial pressures used in the annealing treatment.

## Discussion

The variations in strength, toughness and fracture morphology with these annealing treatments arise from structural changes in the chromite lattice, which also affect the thermal expansion behavior and dimensional stability of these materials [15,26]. It has been noted that doped lanthanum chromite samples exhibited increasing expansion below a critical oxygen pressure, with the onset of expansion occurring at a higher  $P(O_2)$  for compositions with higher acceptor content [26]. Similarly, the magnitude of the expansion after a specific annealing treatment was greater in samples with higher acceptor contents.

In reducing environments at high temperatures, oxygen is lost from the chromite lattice, resulting in the formation of oxygen vacancies, and reduction of  $Cr^{4+}$  to  $Cr^{3+}$  to provide charge compensation. For LCC-30, the mole fraction of  $Cr^{4+}$  is reduced from 0.26 to 0.06 [26]. Armstrong et al [26] have argued that the lattice expansion ( $\sim 1.3\%$ ) upon reduction is primarily because of the change in ionic radius of Cr ( $0.55\text{\AA}$  to  $0.62\text{\AA}$ ) associated with the change in the Cr oxidation state (4+ to 3+). The increased ionic size from the annealing treatment can also potentially decrease the strength of the chemical binding, or the cohesive strength of the lattice. Accurate prediction of fundamental strengths of these oxides awaits rigorous molecular dynamics calculations, but to a rough approximation, the strength of chemical binding in simple oxide structures can be assessed from the cationic field strengths computed at the oxygen sites of

the lattice [27]. A simple calculation of the coulombic force can be performed for the Cr-O bond. The field strength is proportional to  $Z/R^2$  where  $Z$  is the cationic charge, and  $R$  is the interatomic distance,  $r_{\text{Cr}} + r_{\text{O}}$ . The field strength in  $\text{e}/\text{\AA}^2$  is 2.10 with Cr in the 4+ state, and 1.47 with Cr in the reduced 3+ state, using ionic radii values for a coordination number of 6 [28]. The significant difference in the estimated field strength between the different Cr oxidation states for this simple case suggests that the cohesive strength of the more complex chromite lattice could also change significantly when  $\text{Cr}^{4+}$  is reduced to  $\text{Cr}^{3+}$  during annealing.

The decrease in strength and fracture toughness upon annealing in reducing atmospheres could also depend on the distribution of oxygen vacancies in the lattice. Strength and fracture toughness as a function of the oxygen stoichiometry (number of oxygen ions per formula unit) are plotted in Figures 8 and 9. The data for both unannealed and annealed samples are included in the plots. The data in each plot fall into two groups, with the lower values corresponding to the highly reduced samples. Since the reduced samples show predominantly transgranular fracture, it appears that oxygen vacancies in small (overall) concentrations can affect the crack propagation characteristics through the lattice. The modulus is not changed significantly by the annealing treatment, thus the ratio of fracture toughness to modulus is lowered by the formation of oxygen vacancies. For other materials where this ratio is low, the fracture behavior is believed to be controlled by the easy cleavage fracture along specific crystallographic planes of the lattice [29]. Information concerning oxygen vacancy distribution could not be obtained by X-ray diffraction, as no differences in relative peak intensities were observed in X-ray spectra corresponding to annealed and unannealed samples of the same composition. Further characterization by electron microscopy and scattering techniques is required to determine whether preferred fracture paths through the grains could arise from ordering of oxygen vacancies in these chromites.

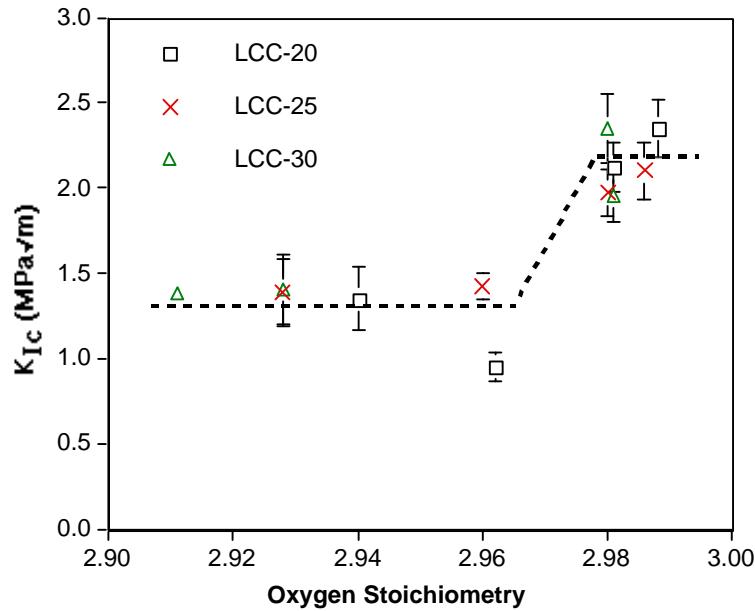


Figure 8. Indentation fracture toughness of Ca-substituted lanthanum chromite as a function of oxygen stoichiometry ( $y$  in  $\text{La}_{1-x}\text{Ca}_x\text{CrO}_y$ )

## Conclusions

Strength behavior of acceptor-substituted lanthanum chromites depends on acceptor type and amount. The flexural strength of Ca-substituted chromites decreases with temperature, even at low temperatures, while Sr-doped chromites show no change in strength over a wide temperature range. Annealing acceptor-substituted chromites at low oxygen partial pressures results in significant decreases in strength, as a result of structural changes to the lattice that accompany the reduction of  $\text{Cr}^{4+}$  to  $\text{Cr}^{3+}$  and formation of oxygen vacancies. In reduced chromites, cracks preferentially propagate through grains. A lower fracture surface energy is inferred from the decreased fracture toughness measured after the annealing treatment at low oxygen partial pressures.

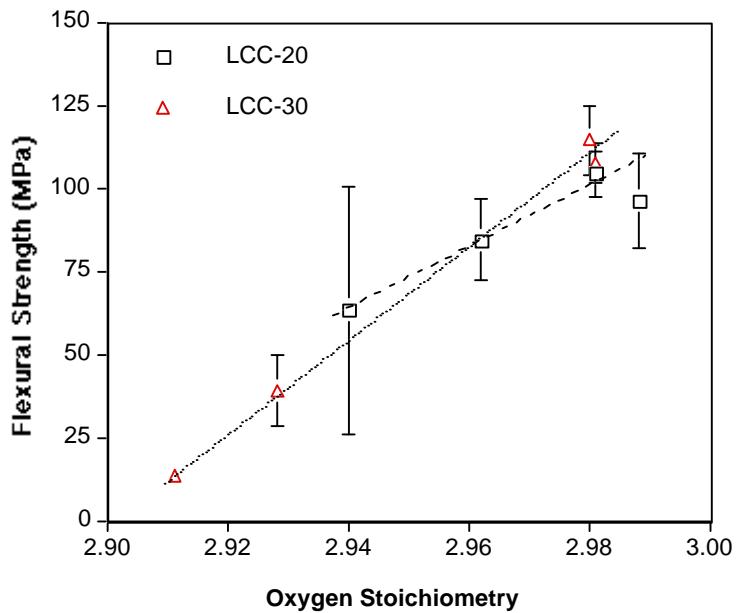


Figure 9. Room-temperature strength of Ca-substituted lanthanum chromite as a function of oxygen stoichiometry ( $y$  in  $\text{La}_{1-x}\text{Ca}_x\text{CrO}_y$ ). The highly reduced samples with poor mechanical integrity are shown separated from the samples with higher strengths by the thick vertical line.

## Acknowledgments

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# **MICRO-ENGINEERED CATHODE INTERFACE STUDIES**

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## **INTRODUCTION**

The aim of this work is to increase the performance of the cathode in solid oxide fuel cells (SOFCs) operating at 1000°C by decreasing the polarization resistance from  $0.2 \Omega\text{-cm}^2$  at  $300 \text{ mA/cm}^2$ . Decreased polarization resistance will allow operation at higher current densities. This work is in support of the Westinghouse tubular SOFC technology using YSZ electrolyte and strontium doped lanthanum manganite (LSM) cathode.

## **THE PROBLEM**

As a result of work performed last year at Argonne National Laboratory and information derived from the literature, the limitations at the cathode/electrolyte interface can be classified into two main areas. First, the ionic conductivity of the LSM cathode material is low which limits the reaction zone to an area very close to the interface, while the rest of the cathode thickness acts essentially as current collector with channels for gas access. Second, the electronic conductivity in YSZ is very low which limits the reaction zone to areas that are the boundaries between LSM and YSZ rather than the YSZ surface away from LSM at the interface.

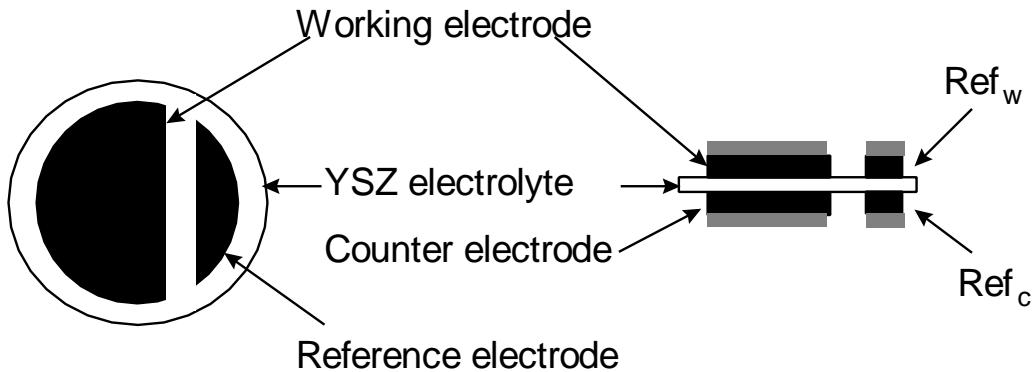
## **APPROACH**

Possible solutions to this problem being pursued are: 1) introducing an ionic conducting YSZ phase in LSM to form a porous two-phase mixture of LSM and YSZ; 2) applying a thin interlayer between the electrolyte and the cathode where the interlayer has high ionic and electronic conductivity and high catalytic activity for reduction of  $\text{O}_2$ ; 3) increasing the ionic conductivity in the LSM by suitable doping; and 4) increasing the electronic conductivity in the electrolyte by doping or by depositing an appropriate mixed conducting layer on the YSZ before applying the cathode.

## **PROCEDURE**

We obtained YSZ electrolyte discs of  $200 \mu\text{m}$  from Marketech International. The appropriate microstructure of the electrode to be studied was then created on top of the YSZ membrane so as to

form the working electrode. A reference electrode was also made the same way on the same side of the disk as the working electrode but separated from the working electrode by about 5 mm. Similarly, a counter electrode and another reference electrode were deposited on the other side of the YSZ disc. The experimental arrangement is shown in Fig. 1.

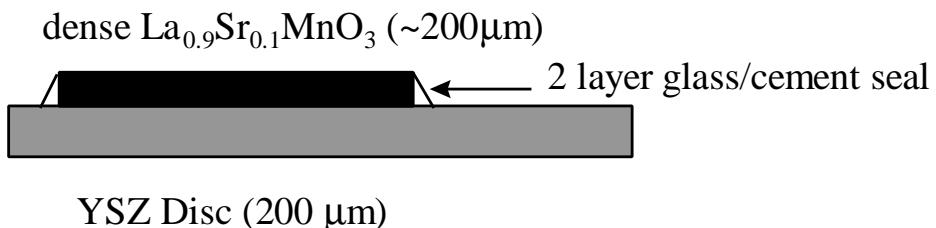


**Fig. 1. Electrode setup for polarization tests.**

Polarization resistance of each electrode was measured against the corresponding reference electrode using impedance spectroscopy. Overpotential measurements were performed as a function of current density using a galvanostat, and the potential of the working and counter electrodes were measured against each other and against the corresponding reference electrodes.

## RESULTS

The effect of low ionic conductivity in LSM on the cathode polarization resistance ( $R_c$ ) is evident from Fig. 2, which shows a dense LSM disc bonded to a YSZ disc with a thin layer of LSM applied between the two. These edges were sealed as shown with a glass-plus cement seal so that gas access to any LSM/YSZ interface is blocked off. In this way, all the oxygen from the air is forced to diffuse through the dense LSM. The  $R_c$  from impedance spectroscopy in such a case was high,  $11 \text{ k}\Omega\text{-cm}^2$ . A cathode with high oxide ion conductivity should decrease the polarization resistance.

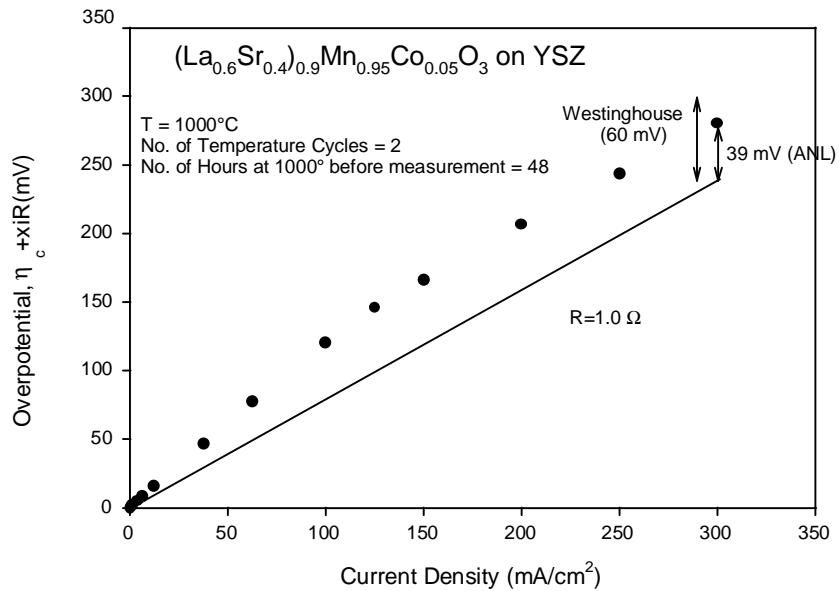


**Fig. 2. Configuration of dense LSM on YSZ with sealed edges.**

In another experiment a YSZ disc was ion implanted with manganese ions to 0.1  $\mu\text{m}$  below the surface. Upon heating up to 900°C, the polarization resistance was found to be low, 0.38  $\Omega\text{-cm}^2$ . Over time, this value increased as manganese ions diffused away from the surface of the YSZ into the bulk. However, it showed that increasing the ionic conductivity of YSZ at the surface helps to spread out the reaction zone and decreases polarization.

Based on the above two observations, compositions were devised to increase the ionic conductivity of the cathode by doping an LSM composition with Co and by increasing the substitution of La by Sr. New materials like the cobalt-ferrite composition were also tried. The aim here is to either replace the LSM cathode or use the new materials as a thin interlayer between LSM and YSZ.

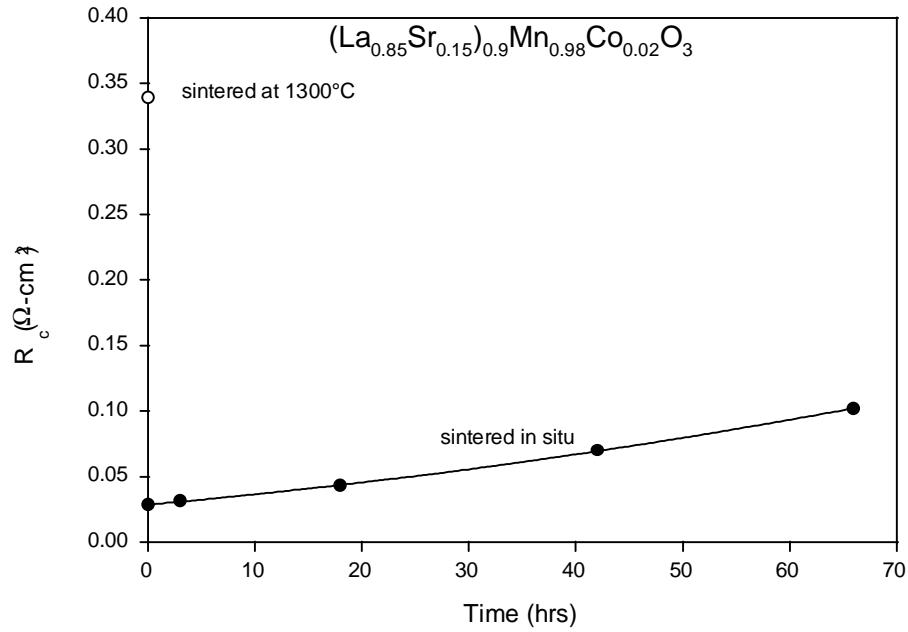
A cobalt-doped LSM composition,  $(\text{La}_{0.6}\text{Sr}_{0.4})_{0.9}\text{Mn}_{0.95}\text{Co}_{0.05}\text{O}_3$  (designated LSCM-2) sintered at 1300°C and tested last month showed good performance ( $R_c$  around 0.1  $\Omega\text{-cm}^2$ ) as a cathode on YSZ with no load. This was presumably due to increased ionic conductivity from the cobalt doping. Further testing at different loads (current densities) showed good performance as well. Figure 3 is a plot of overpotential versus current density after holding the sample at a 1000°C for a cumulative 48 hours with two temperature cycles where it was cooled to room temperature and heated back up to 1000°C. The overpotential at 300 mA/cm<sup>2</sup> was 39 mV, compared with a typical 60 mV overpotential for Westinghouse's cathode at the same current density.



**Fig. 3. Overpotential of LSCM-2 decreases with increasing I.**

Another cobalt-doped composition,  $(\text{La}_{0.85}\text{Sr}_{0.15})_{0.9}\text{Mn}_{0.98}\text{Co}_{0.02}\text{O}_3$  (LSCM-1), was tested both with sintering at 1300°C and sintered in situ at the operating temperature of 1000°C, (see Fig. 4). The sintered composition exhibited a higher  $R_c$  of about 0.34  $\Omega\text{-cm}^2$ , while the in situ sintered sample had an initial  $R_c$  of 0.03  $\Omega\text{-cm}^2$  without any load. This compares very well to a value of 0.2  $\Omega\text{-cm}^2$

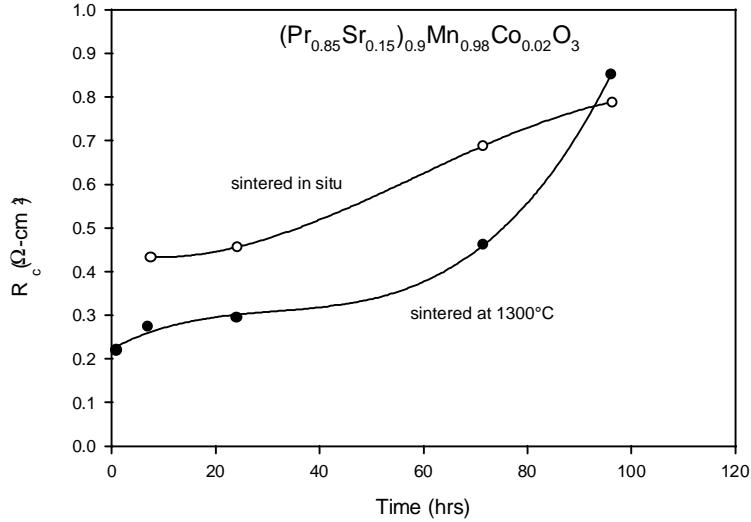
for Westinghouse's cathode at 300 mA/cm<sup>2</sup>. The in situ composition started sintering with time at 1000°C, which caused an increase of the  $R_c$  from an initial value of 0.03 Ω-cm<sup>2</sup> to 0.1 Ω-cm<sup>2</sup> after 67 hr. Sintering this composition caused a decrease in the actual area of contact between YSZ and LSCM while increasing the area of contact between LSCM particles. This indicated that cobalt-doped LSM (LSCM) shows promise as an interlayer if it is made with a large contact area between LSCM and YSZ that does not decrease with time under operating conditions.



**Fig. 4. LSCM-1 shows good performance initially when sintered in situ.**

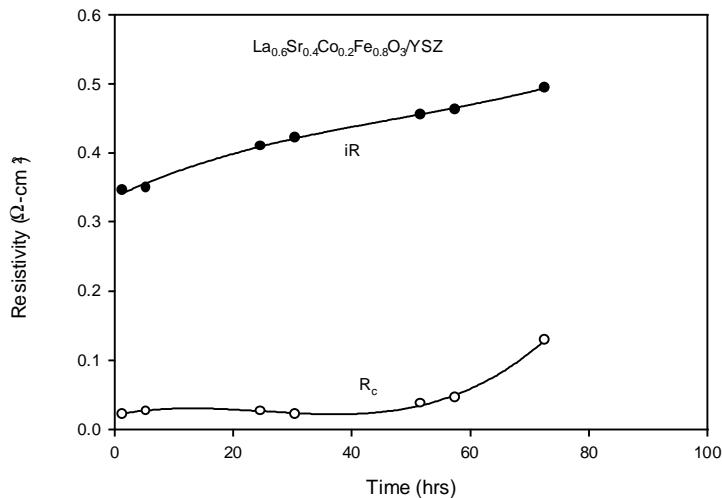
It has been reported in the literature that manganites of praseodymium ( $\text{PrMnO}_3$ ) showed higher performance than  $\text{LaMnO}_3$ . To utilize this fact, we synthesized praseodymium counterparts of LSCM-1 and LSCM-2, by the Pechini method,  $(\text{Pr}_{0.85}\text{Sr}_{0.15})_{0.9}\text{Mn}_{0.98}\text{Co}_{0.02}\text{O}_3$  (PSCM-1) and  $(\text{Pr}_{0.6}\text{Sr}_{0.4})_{0.9}\text{Mn}_{0.95}\text{Co}_{0.05}\text{O}_3$  (PSCM-2). It is expected that the cobalt-doping in PSM also increases ionic conductivity.

Figure 5 gives the performance of PSCM-1 for one electrode sintered at 1300°C and another in situ. In this case the 1300°C sintered composition exhibited better performance than the one sintered in situ, and both cathodes showed a deterioration in performance with time at 1000°C. The PSCM-2 also showed the same trend, but with a much steeper increase in  $R_c$  with time. Since both compositions show the same trend, the deterioration is probably due to change in cathode composition with time, possibly by Co or Mn diffusion from PSCM to YSZ.



**Fig. 5.**  $R_c$  for PSCM-1 sharply increases with time after 40 hrs.

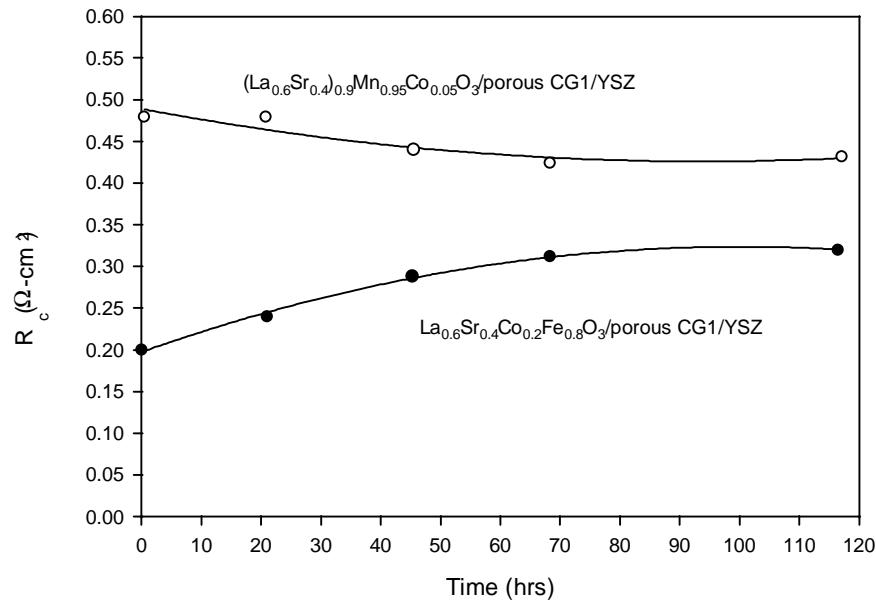
High ionic and electronic conductivity is well known in  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$  (LSCF) and similar cobaltite or cobalt-ferrite compositions. We previously reported data on some LSCF compositions where the LSCF composition was slightly deficient on the A-site,  $(\text{La}_{0.6}\text{Sr}_{0.4})_{0.99}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$ . While this yielded very low  $R_c$  values of  $0.02 \Omega\text{-cm}^2$  or even less initially, the formation of an insulating  $\text{CoFe}_2\text{O}_4$  caused the  $R_c$  to increase to almost  $10 \Omega\text{-cm}^2$  in less than three days. This time we report results on stoichiometric LSCF with the composition  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$ . The polarization resistance,  $R_c$  for this LSCF still increases with time, (see Fig. 6), but at a slower rate than before. The ohmic resistance component through the electrolyte plus cathode, if any, also rises. This indicates that the increase in  $R_c$  is accompanied by an increase in resistance. The rate of increase in resistance indicates the formation of an insulating phase. Both  $\text{La}_2\text{Zr}_2\text{O}_7$  and  $\text{SrZrO}_3$  have been reported to form when LSCF electrode is used on a YSZ electrolyte at  $1000^\circ\text{C}$ .



**Fig. 6.** Both the  $R_c$  and the resistance part of iR drop increase with time for LSCF.

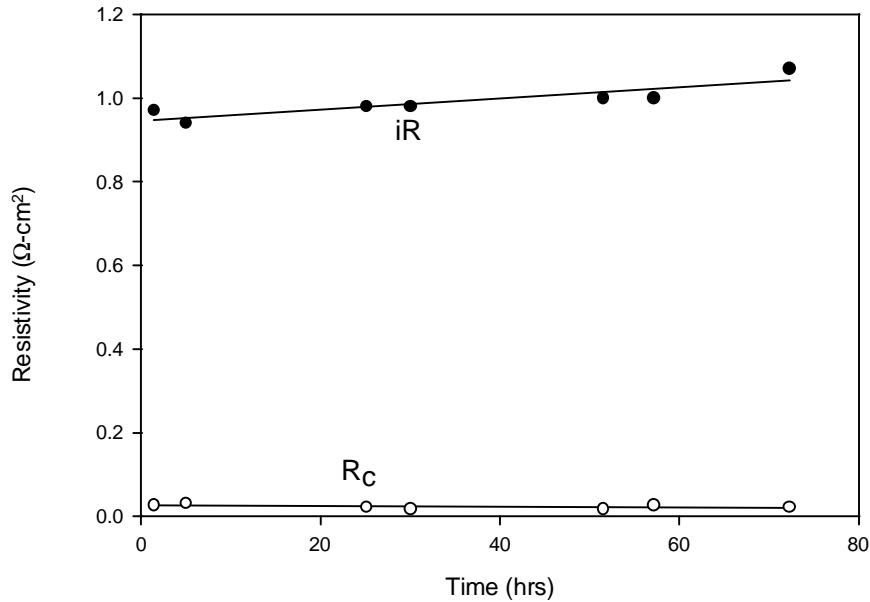
In order to decrease the interaction between YSZ and LSCF, an attempt was made to deposit a thin (< 1  $\mu\text{m}$ ) dense layer of  $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{2-\delta}$  on YSZ, followed by the deposition of LSCF on top of the ceria layer. The ceria layer made by us turned out to be porous. Nevertheless, two such electrodes were tested with the following configurations: stoichiometric LSCF/porous CG1/YSZ and LSCM-2/porous CG1/YSZ.

Results of cathode tests of both configurations are shown in Fig. 7. The LSCF cathode showed a higher initial  $R_c$  (0.2  $\Omega\text{-cm}^2$ ) than the previously tested LSCF/YSZ configurations and also showed an increase in  $R_c$  to 0.33  $\Omega\text{-cm}^2$  over 115 hr. However, the plot shows that the  $R_c$  value stabilized and, after 115 hr, is less than the LSCF/YSZ polarization resistance of 1  $\Omega\text{-cm}^2$  after 63 hr. Therefore, a ceria interlayer did prevent significant reaction between LSCF and YSZ. Similarly, for the LSCM-2 cathode with ceria interlayer, the value of  $R_c$  was initially 0.47  $\Omega\text{-cm}^2$ , which actually decreased and stabilized at 0.44  $\Omega\text{-cm}^2$  after 115 hr. The reason for a decrease in  $R_c$  for LSCM-2 versus an increase for LSCF is not known at this time; however, it may be linked to differences in thermal expansion coefficient between the cathode and the interlayer.



**Fig. 7. Stable but higher polarization resistances obtained with an interlayer of porous ceria.**

In order to improve the overall  $R_c$  value for LSCF/CG1/YSZ cathodes, a thin 0.5  $\mu\text{m}$  dense CG1 film was deposited by Prof. Harlan Anderson at the University of Missouri-Rolla on a YSZ disc supplied by us. We then applied the LSCF cathode and tested the cell as a function of time. The results of this test (Fig. 8) show a low initial  $R_c$  of 0.024  $\Omega\text{-cm}^2$  which then decreases slightly and monotonously to less than 0.018  $\Omega\text{-cm}^2$  over 72 hrs. This electrode again is a combination of steps 3 and 4 as outlined earlier.



**Fig. 8. Very low and stable  $R_c$  obtained with a thin dense interlayer of ceria.**

We are also currently working on a composition that can be used directly as a cathode with no interlayers. The goal here is to increase the ionic conductivity without decreasing the electronic conductivity of LSM or its catalytic activity for oxygen reduction. In addition, the thermal expansion coefficient will be matched to the YSZ electrolyte and it will not be reactive with YSZ. Initial results show some promise with about a 112 mV polarization at 300 mA/cm<sup>2</sup>. Further work is in progress to understand and improve this composition.

## BENEFITS

Increased cathode performance or decreased cathode polarization resistance will help to operate at higher power density or with higher efficiency.

## FUTURE WORK

- 1) Development of new compositions that can be used as cathodes without interlayers. These materials will be analyzed for performance over time and as a function of compositional changes. The aim here is to increase ionic conductivity substantially compared to LSM.
- 2) Continued development of Co or other transition metal-doped LSM. We will select a promising material and develop a cathode with an interlayer microstructure of a two-phase mixture of YSZ and doped LSM. It will be analyzed in terms of performance over time and its thermal expansion coefficient will be measured.

- 3) Analysis of previously tested cathode/electrolyte and cathode/interlayer/electrolyte samples. These samples will be analyzed for microstructural characteristics including porosity, interface bonding, and interdiffusion of cations across the interface.

## **ACKNOWLEDGEMENT**

Research sponsored by the Department of Energy through the Morgantown Energy Technology Center under contract no. W-31-109-ENG-38 with FETC Project Manager, Richard Johnson; Contractor Project Manager, Kevin M. Myles; Principle Investigator, Michael Krumpelt. This contract covers work done in FY1997.

## Composite Ceria Electrolytes

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*Over the past two years, NexTech Materials has been developing ceramic materials technology for applications in solid oxide fuel cells and other electrochemical systems. This work has been performed under a collaborative project funded by Ohio's Edison Materials Technology Center (EMTEC), and under three SBIR and STTR projects funded by the U.S. Department of Energy. The focus of NexTech's EMTEC project is the development of hydrothermal synthesis methods for producing nano-scale and crystalline powders of useful ceramic electrolytes. In a recently completed SBIR project, NexTech developed a new family of composite ceria-based ceramic electrolyte compositions providing a superior combination of electrical and mechanical properties. In a recent STTR project, NexTech collaborated with Lawrence Berkeley National Laboratory to apply colloidal deposition techniques to the fabrication of thin-film ceria fuel cells with excellent low-temperature SOFC performance. In an ongoing SBIR project, NexTech is collaborating with Westinghouse on the development of a low-cost process for depositing membrane films of yttria-stabilized zirconia onto lanthanum manganite air-electrode tubes. This paper describes the materials technologies being developed in these four projects.*

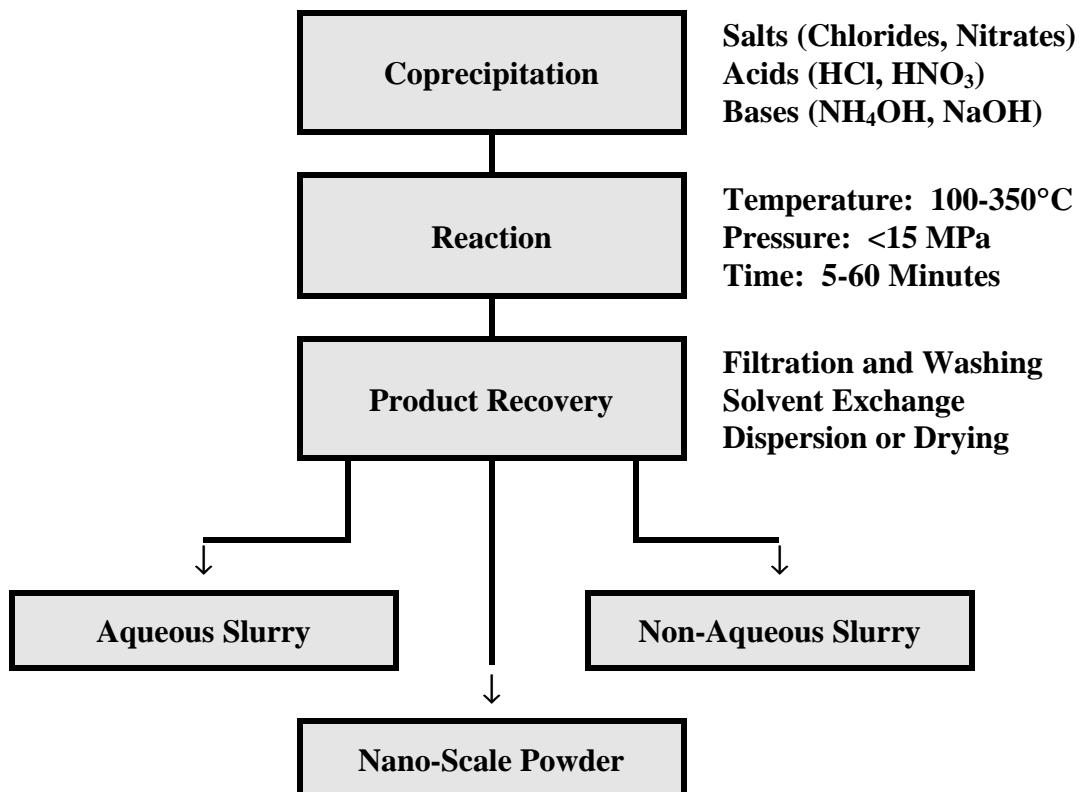
### NexTech's EMTEC Project

NexTech's EMTEC project was initiated in October of 1995 and runs through June of 1999. The objective of this project is to develop innovative ceramic powder synthesis and fabrication methods for the manufacture of oxygen-conducting ceramic membranes. Targeted applications include solid oxide fuel cells, oxygen generation systems, and ceramic membrane reactors. Team members are listed below:

- NexTech Materials
- University of Cincinnati
- Argonne National Laboratory
- Lawrence Berkeley National Laboratory
- Pacific Northwest National Laboratory
- Sandia National Laboratories
- Institute of Gas Technology
- Eltron Research
- Orton Ceramic Foundation
- Superconductive Components, Inc.
- DAI Ceramics
- Westinghouse
- BP Chemicals
- Litton Life Support
- SOFCo
- Babcock & Wilcox
- Motorola
- Coulter Corporation
- Consolidated Natural Gas
- Columbia Gas
- Plastronic, Inc.

NexTech is utilizing hydrothermal synthesis methods (see Figure 1) to produce zirconia and ceria-based ceramic electrolytes. NexTech's hydrothermal process is based on a coprecipitation step (i.e., neutralization of aqueous acid solutions), followed by a mild hydrothermal treatment (i.e., at temperatures less than 300°C and pressures less than 15 MPa). The process results in the formation of an aqueous suspension of nano-scale (5-15 nm) crystallites with the cubic fluorite structure typical of zirconia and ceria-based electrolyte materials. A TEM micrograph of a hydrothermally produced powder of a  $(\text{Ce}_{0.80}\text{Gd}_{0.20})\text{O}_{1.90}$  powder produced by this process is shown in Figure 2. As-produced zirconia and ceria-based electrolyte powders have large surface areas ( $>100 \text{ m}^2/\text{gram}$ ), consistent with their nano-scale crystal size. An advantage of the hydrothermal process is the flexibility of product forms, such as dispersed aqueous suspensions, non-aqueous suspensions, or dried nano-scale powder. The surface area can be controlled over a wide range, either by modifying initial hydrothermal synthesis conditions, or by calcining dried powders at modest temperatures (see Table 1).

In this project, NexTech is conducting process development and materials characterization, evaluating sintering performance and electrical properties, and providing samples for evaluation by participating team members and other collaborators. NexTech has shown that hydrothermally derived zirconia and ceria-based powders can be sintered to high density at relatively low sintering temperatures. For example, an SEM micrograph of a 98% dense gadolinium-doped ceria ceramic, sintered at 1250°C, is shown in Figure 3.



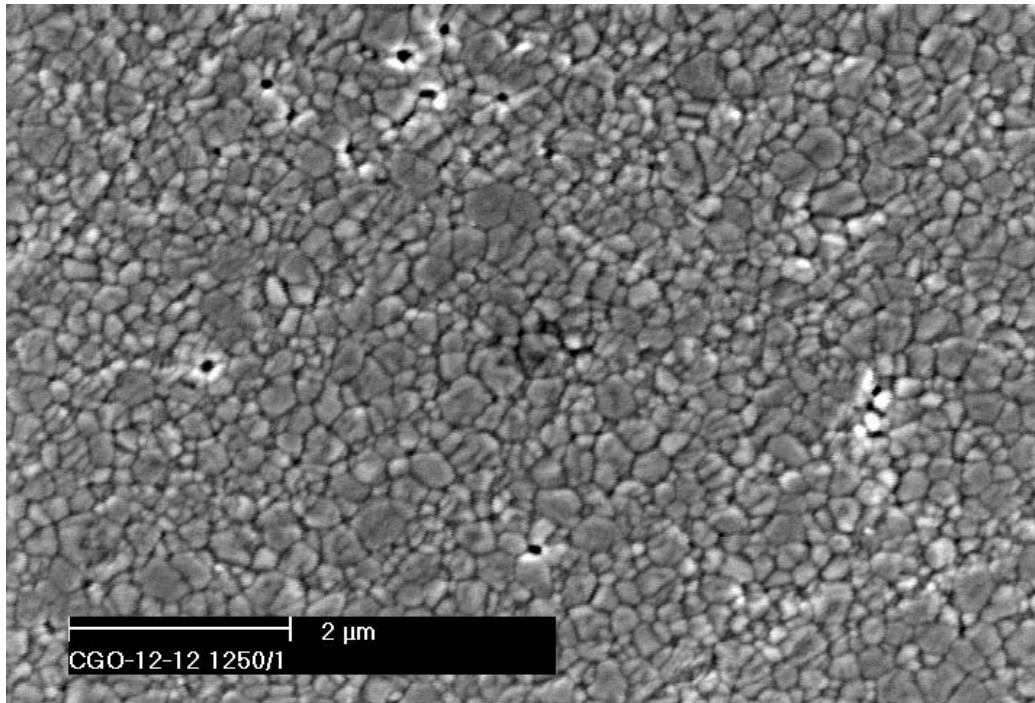
**Figure 1. NexTech's Hydrothermal Powder Synthesis Process.**



**Figure 2.** TEM micrograph of a hydrothermally derived Gd-doped ceria powder.

**Table 1.** Surface Area of Zirconia and Ceria Powders

Calcination	BET Surface Area (m <sup>2</sup> /gram)		
	(Zr <sub>0.852</sub> Y <sub>0.148</sub> )O <sub>1.926</sub>	(Ce <sub>0.80</sub> Gd <sub>0.20</sub> )O <sub>1.90</sub>	(Ce <sub>0.85</sub> Sm <sub>0.15</sub> )O <sub>1.925</sub>
as-produced	154	126	127
600°C, 4 hours	108	---	79
700°C, 4 hours	91	33	45
800°C, 4 hours	65	18	25
900°C, 4 hours	40	9.5	12
1000°C, 4 hours	14	4.4	5.0



**Figure 3. SEM micrograph of a hydrothermally derived Gd-doped ceria ceramic, sintered at 1250° C.**

NexTech has completed a comprehensive set of two-lead electrical conductivity measurements on hydrothermally derived ceria-based ceramics. Two-lead measurements cannot differentiate between the contributions of electrolyte and electrode, and interfacial resistances. However, the two-lead method does allow for relative comparisons regarding effects of material composition, synthesis methods, and sintering conditions. For example, with platinum electrodes, a profound effect of sintering temperature on two-lead conductivity was observed (see Figure 4). The conductivity decreased as the sintering temperature was increased above 1300°C; this trend was common to all of the hydrothermal ceria ceramics evaluated. These results suggest either highly conductive grain boundaries in the fine-grained ceria ceramics sintered at low temperatures, and/or dopant segregation at grain boundaries in coarser-grained ceria ceramics sintered at higher temperatures. Additional microstructural characterization, in combination with ac impedance spectroscopy, may be needed to clarify specific causes.

The two-lead method also provides a means to compare the electrochemical performance of different electrode materials, as shown for Gd-doped ceria ceramics in Figure 5. Compared to platinum and silver electrodes, the conductivity was almost doubled by using cermet electrodes comprised of  $(\text{La},\text{Sr})(\text{Co},\text{Fe})\text{O}_3$  and silver phases. Since identical ceramic samples were used, the improvement can only be related to superior electrochemical performance of the cermet LSCF/Ag electrodes. However, additional work is needed to determine whether composition or morphology of the electrode was responsible for the improvement.

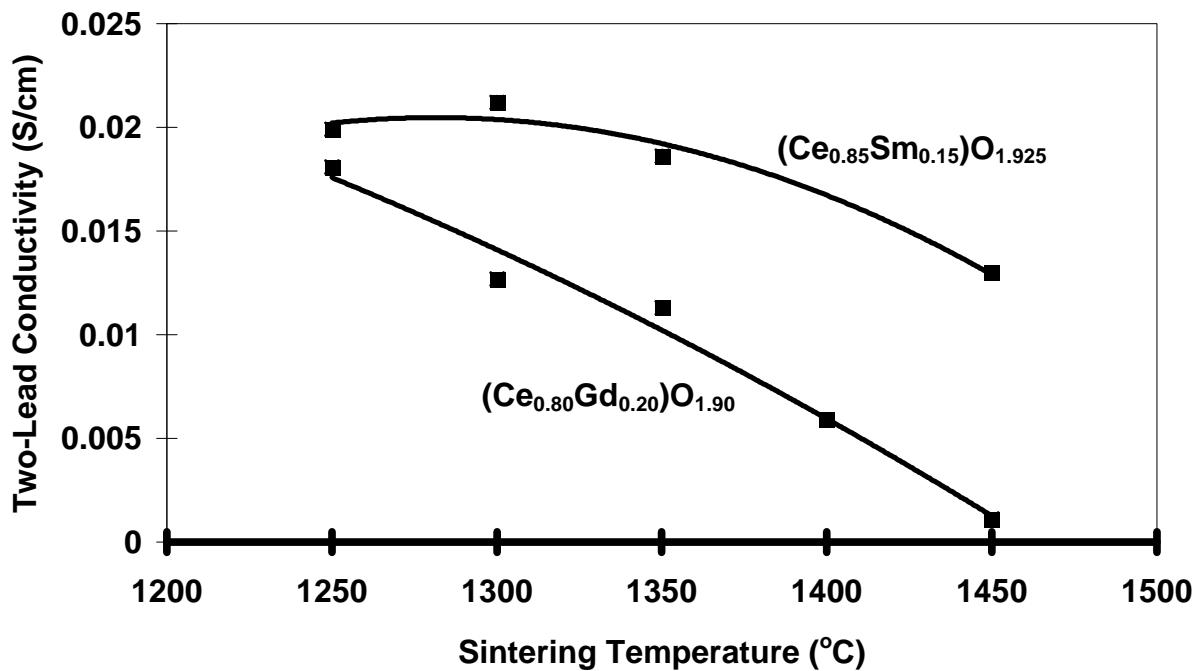


Figure 4. Two-lead conductivity versus sintering temperature for hydrothermally derived ceria-based ceramics (platinum electrodes).

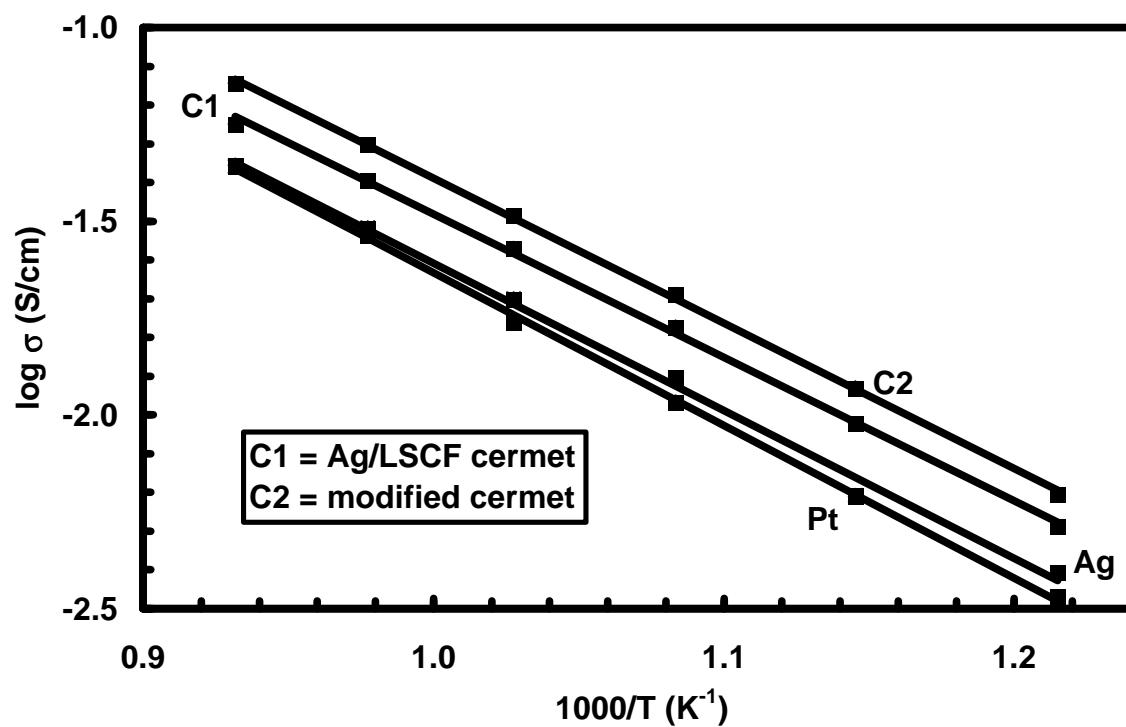


Figure 5. Arrhenius plots of two-lead conductivity versus reciprocal temperature for hydrothermal  $(\text{Ce}_{0.80}\text{Gd}_{0.20})\text{O}_{1.90}$  ceramics with different electrode materials.

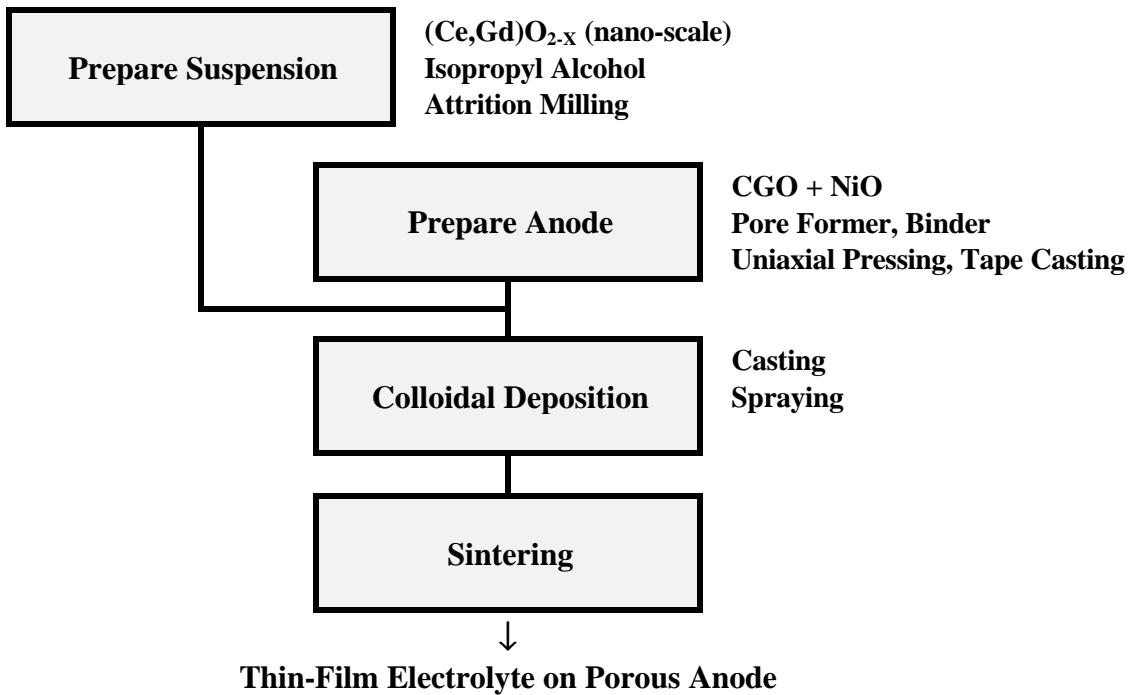
## **Thin-Film Ceria Fuel Cells**

In this STTR project, NexTech Materials and Lawrence Berkeley National Laboratory (LBNL) collaborated on the fabrication of thin-film ceria fuel cells and evaluation of their low-temperature SOFC performance. Contract information is provided below:

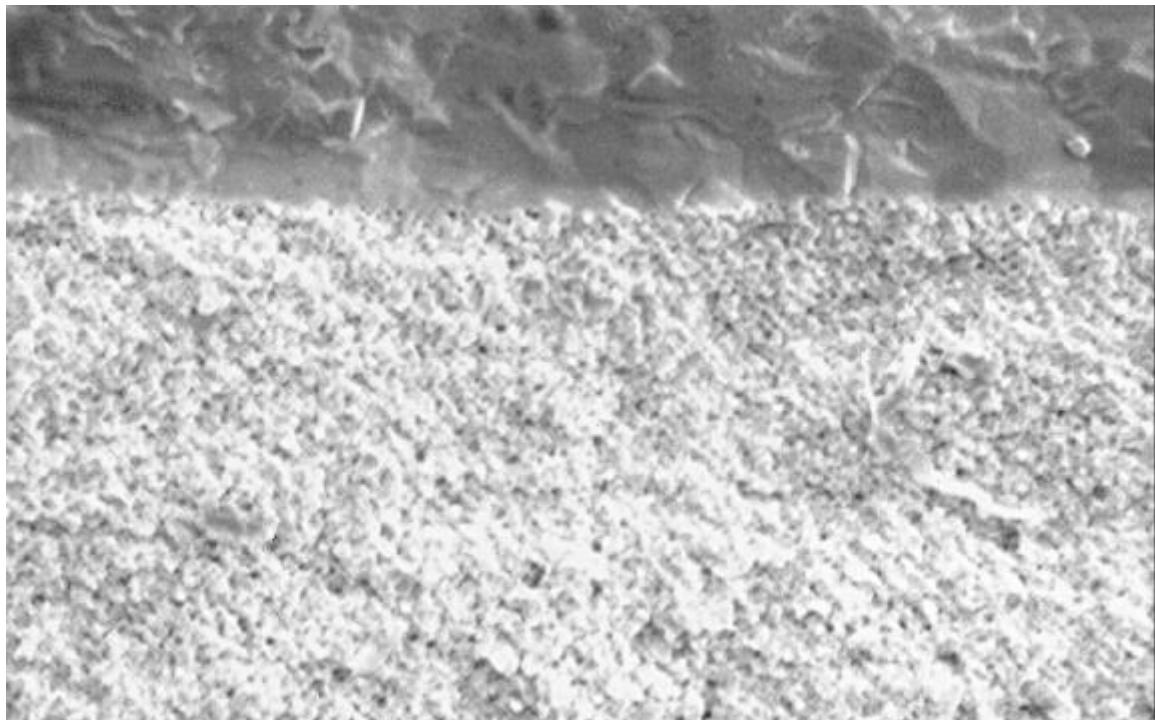
- **Title:** *Thin-Film Ceria Fuel Cells with Low Operating Temperature*
- **Sponsor:** U.S. Department of Energy, Office of Transportation Technologies
- **Contract Number:** DE-FG02-96ER86051
- **Period of Performance:** 6/22/96 - 3/21/97
- **Contracting Officer:** Dr. JoAnn Millikin (202-586-2480)
- **Subcontractor:** Lawrence Berkeley National Laboratory
- **Principal Investigator:** Dr. Scott L. Swartz (614-842-6606)

Several developers have shown that deposition of thin-film yttria-stabilized zirconia (YSZ) membranes on porous electrode (anode or cathode) supports is a feasible approach to reduce the operating temperature of solid oxide fuel cells. However, even with thin-film YSZ membranes, the SOFC operating temperatures cannot be reduced to below 600°C, without substantial degradation in performance and efficiency. The use of higher-conductivity electrolyte materials, fabricated in thin-film configurations, is the most feasible approach for reducing SOFC operating temperatures to below 600°C. It is well known that ceria-based ceramic electrolytes have higher conductivity than YSZ, especially at temperatures below 600°C. A common problem with ceria-based electrolytes has been the onset of electronic conductivity at high temperatures, which reduces efficiency of SOFC operation. However, at temperatures below about 600°C, electronic conductivity of ceria-based electrolytes is negligible, so that SOFC performance is not degraded. Thus, this STTR project was undertaken to evaluate ceria thin films as an approach for reducing SOFC operating temperatures to 600°C and below. Successful development of the technology demonstrated in this project will open up opportunities for SOFCs to compete as lower cost and higher performance options to PEM-based fuel cells for future transportation applications.

The approach pursued in this project was based on the synergistic combination of NexTech's hydrothermal synthesis process for preparing nano-scale ceria suspensions (as discussed above and shown in Figure 1) and LBNL's colloidal deposition method for fabricating thin-film fuel cells. The colloidal deposition process, as shown in Figure 6, is an inexpensive approach for fabricating bilayer electrolyte elements comprised of an electrolyte film on a porous electrode substrate. A colloidal suspension of the desired electrolyte material is first prepared, and then cast onto a highly porous electrode plate. A green electrolyte film is formed as the solvent evaporates from the suspension, and then the bilayer is sintered so that the electrolyte film densifies fully and the electrode substrate densifies partially. If the relative shrinkages and green densities of the two layers are properly controlled, then the process results in a bilayer element comprised of a dense electrolyte film (10-15  $\mu\text{m}$  thick) on a porous and flat electrode substrate (200-300  $\mu\text{m}$  thick). This process was successfully applied to the fabrication of a  $(\text{Ce}_{0.80}\text{Gd}_{0.20})\text{O}_{1.90}$  (CGO) electrolyte film on a porous NiO/CGO anode substrate. An SEM micrograph of a cross-section of a sintered bilayer is presented in Figure 7, which clearly shows the pore-free nature of the CGO film.

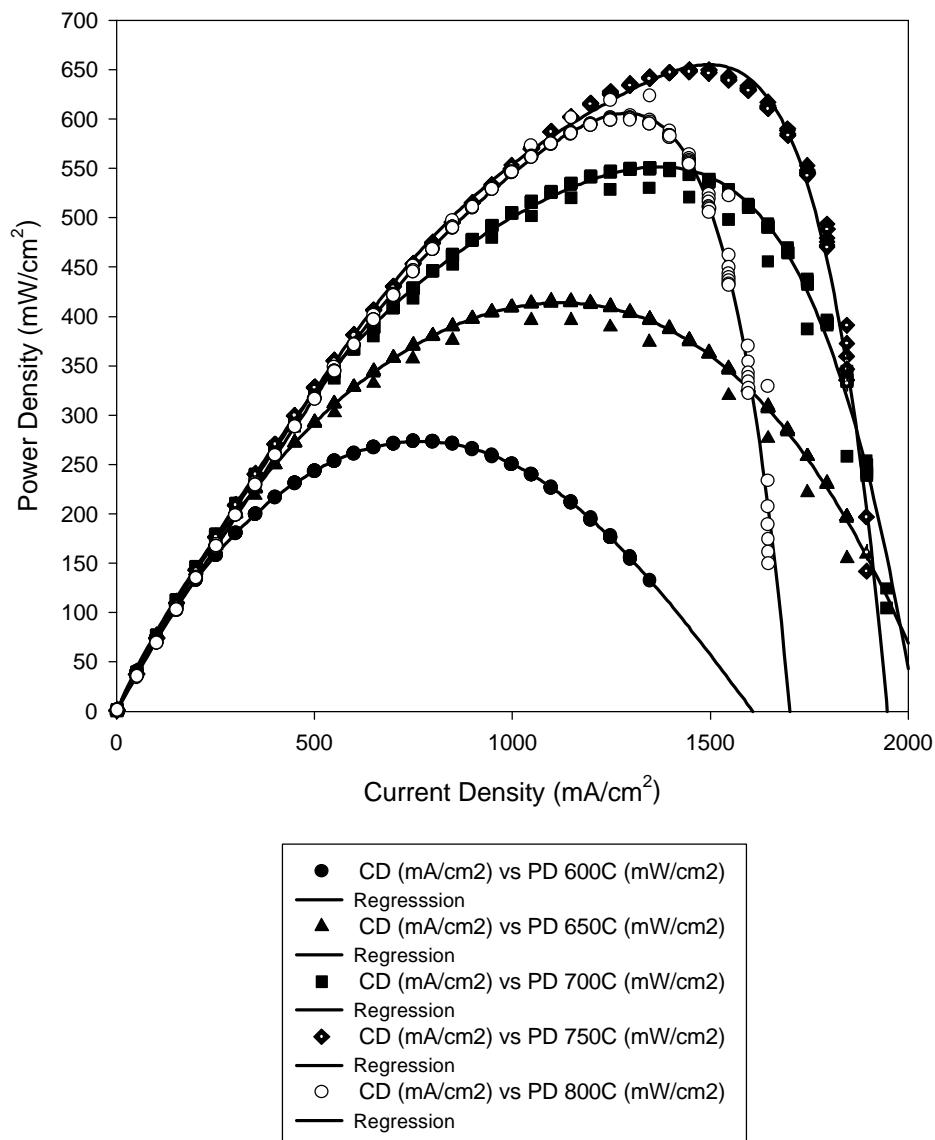


**Figure 6.** LBNL's colloidal deposition process for fabricating thin-film fuel cells.



**Figure 7.** SEM micrograph of a cross-section of a bilayer element comprising a porous NiO/CGO anode with a dense CGO electrolyte film.

After successful fabrication of sintered bilayers, fuel cell elements were completed by the deposition of a (La,Sr)CoO<sub>3</sub>-based cathode. Fuel cell testing was conducted using H<sub>2</sub> and air as fuel and oxidant, and the power density curves obtained are presented in Figure 8. To our knowledge, the power densities achieved (~650 mW/cm<sup>2</sup> at 750°C) in the thin-film ceria cell represent the *highest ever reported* for a ceria-based solid oxide fuel cell. The power density achieved at 600°C (~270 mW/cm<sup>2</sup>) also represents one of the highest values reported for *any type of fuel cell* operating at this temperature. These results are especially promising, given the limited amount of optimization work directed at the anode microstructure and the cathode material.



**Figure 8. Power density versus current density curves at different temperatures for a colloidally deposited thin-film ceria fuel cell.**

## Composite Ceria Electrolytes

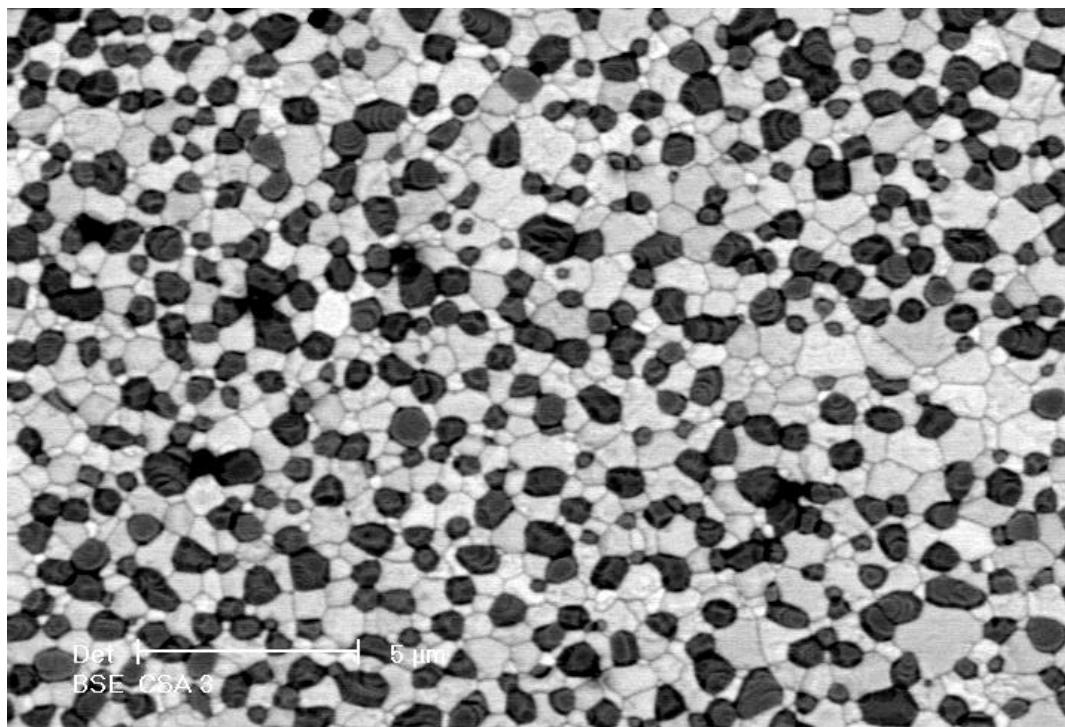
In this recently completed SBIR project, NexTech Materials demonstrated a novel composite materials approach for improving the performance of ceria-based ceramic electrolytes. The objective of this project was to improve the oxygen ion transference number of ceria-based electrolytes, through the controlled addition of a nano-dispersed insulating second phase. Contract information is provided below:

- **Title:** *Composite Ceria Electrolytes for Solid Oxide Fuel Cells*
- **Sponsor:** U.S. Department of Energy, Federal Energy Technology Center
- **Contract Number:** DE-FG02-96ER82236
- **Period of Performance:** 8/13/96 - 3/6/97
- **Contracting Officer:** William Cary Smith (304-285-4260)
- **Principal Investigator:** Dr. Scott L. Swartz (614-842-6606)

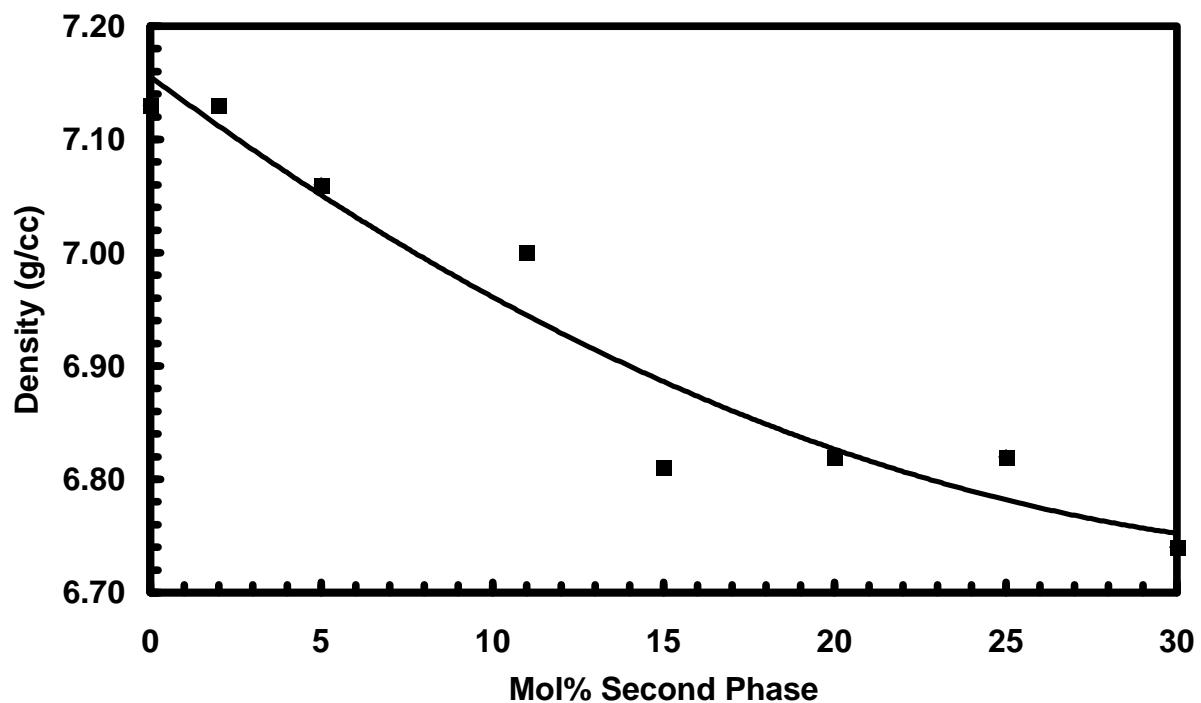
Based on their relatively high oxygen ion conductivities, doped cerium oxide ceramic electrolytes are potential electrolyte materials for intermediate-temperature solid oxide fuel cell applications. However, the utilization of single-phase ceria electrolytes in SOFCs has been limited by electronic conductivity (which results in power loss) and by poor mechanical properties (which leads to failure of planar electrolyte elements). NexTech's approach to solving these problems involved the controlled addition of an insulating second phase to a nano-scale ceria powder, followed by the preparation of a composite ceria ceramic electrolyte. Previous theoretical and experimental work at Arizona State University suggested that the insulating second phase reduces electronic conductivity by trapping electronic charge carriers within space charge regions at the interface between the ceria and insulating grains. This program was conducted to evaluate this approach for improving the oxygen ion transport properties of ceria-based electrolytes.

NexTech's hydrothermal synthesis process was used to prepare nano-scale ceria-based powder, which when sintered, provided the desired two-phase microstructure, as shown in Figure 9. Sintered densities for all compositions were in excess of 95% theoretical, although densities exceeding 98% theoretical were obtained when the second phase content was less than 15 mol%, as shown in Figure 10. Two-lead conductivity measurements determined that the second phase addition reduced the conductivity, but this reduction was fairly small for second phase contents of less than 20 mol%, as shown in Figure 11. Galvanic cell measurements were inconclusive, but indicated an apparent improvement in oxygen ion transference number at  $pO_2 \sim 10^{-22}$  atm. Additional measurements are needed to quantitatively determine the effect of the second phase on oxygen ion transport in these composite ceria ceramics. Perhaps, the most beneficial effect of the second phase addition was on the mechanical properties, as shown in Table 2. Compared to an unmodified ceria ceramic, the second phase addition (11 mol%) led to a significant improvement in hardness and a two-fold increase of fracture toughness, as shown below:

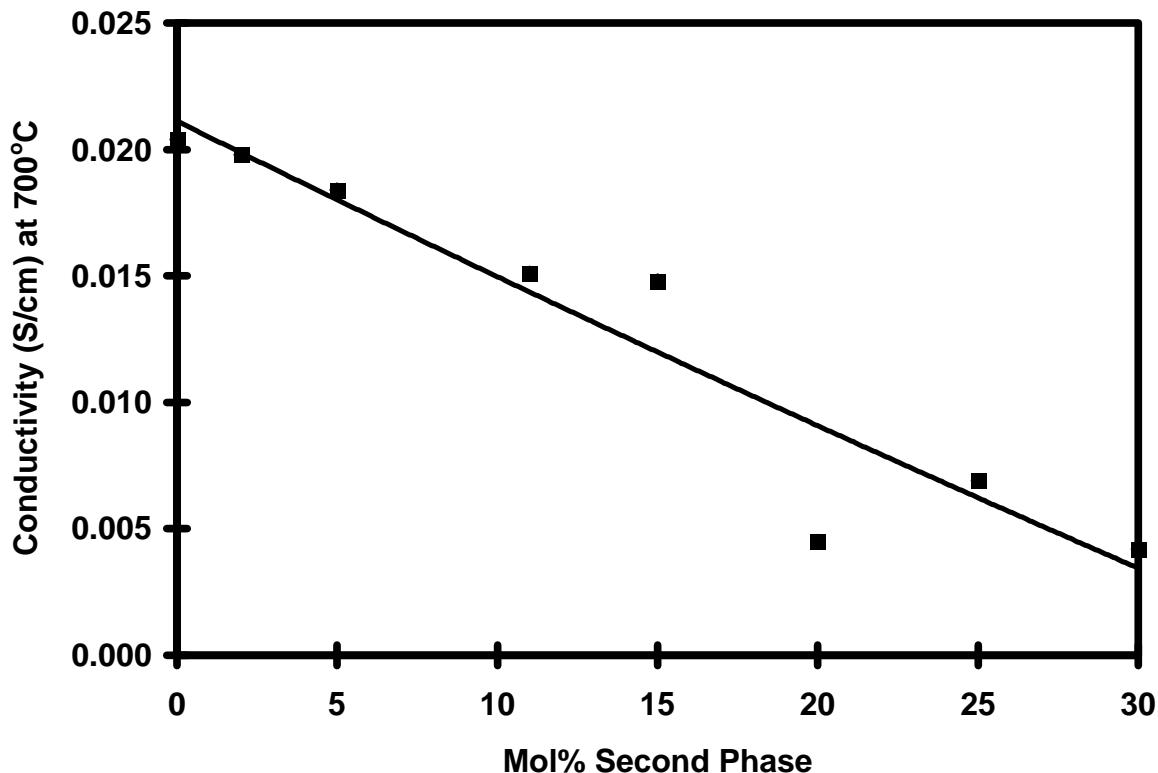
- **Fracture Toughness ( $K_{IC}$ ):**  $0.9 \rightarrow 1.8 \text{ MPa}\cdot\text{m}^{-0.5}$
- **Vickers Hardness ( $H_V$ ):**  $780 \rightarrow 940 \text{ kg/m}^2$



**Figure 9.** Backscatter-mode SEM micrograph of composite ceria ceramic.



**Figure 10.** Effect of second phase content on the sintered density of composite ceria ceramic electrolytes.



**Figure 11. Effect of second phase content on the two-lead conductivity of composite ceria ceramic electrolytes (platinum electrodes).**

### Nano-Scale YSZ Electrolytes

NexTech Materials recently initiated an SBIR program, in collaboration with Westinghouse, to develop a low-coat electrolyte deposition process for the Westinghouse tubular SOFC, as a replacement for electrochemical vapor deposition. The approach is based on NexTech's hydrothermal synthesis process for making nano-scale YSZ powders. Electrolyte films will be deposited onto lanthanum strontium manganite (LSM) cathode tubes from colloidal YSZ suspensions, and then sintered to high density at low temperatures. With the low sintering temperatures possible with hydrothermal YSZ powders, adverse reactions between the YSZ electrolyte and the LSM cathode will be avoided. Contract information is provided below.

- **Title:** *Tubular SOFC with Deposited Nano-Scale YSZ Electrolyte*
- **Sponsor:** U.S. Department of Energy, Federal Energy Technology Center
- **Contract Number:** DE-FG02-97ER82443
- **Period of Performance:** 9/3/97 - 3/17/98
- **Contracting Officer:** William Cary Smith (304-285-4260)
- **Collaborating Partner:** Westinghouse
- **Principal Investigator:** Dr. Scott L. Swartz (614-842-6606)

# Lanthanum Gallate as a New SOFC Electrolyte

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## Abstract

The single-phase, cubic-perovskite region of the  $\text{LaO}_{1.5}\text{-SrO-GaO}_{1.5}\text{-MgO}$  phase diagram has been determined from room-temperature and high-temperature x-ray diffraction. Two impurity phases were identified,  $\text{LaSrGaO}_4$  and  $\text{LaSrGa}_3\text{O}_7$ . An essentially pure oxide-ion conductivity of the oxygen-deficient perovskite phase was formed over the wide range  $10^{-22} \leq P_{\text{O}_2} \leq 1$  atm of oxygen partial pressures. The highest values of  $\sigma_o$  were found for  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.83}\text{Mg}_{0.17}\text{O}_{2.815}$  with  $\sigma_o = 0.17$  S/cm and 0.08 S/cm at 800°C and 700°C, respectively. The oxide-ion conductivities remained stable over a week-long test. The Arrhenius plot of  $\sigma_o$  is curved, dividing into two straight-line regions with different activation energies above and below a critical temperature  $T^* = 600^\circ\text{C}$ . This behavior is well-described by a model in which all the oxygen vacancies are mobile at temperature  $T > T^*$ , but they progressively condense into clusters of ordered vacancies with decreasing temperature  $T < T^*$ .

SEM microstructures of samples  $\text{La}_{1.8}\text{Sr}_{0.2}\text{Ga}_{1-y}\text{Mg}_y\text{O}_{2.9-0.5y}$ ,  $0.05 \leq y \leq 0.30$ , were obtained after thermal etching at 1350°C for 2 h. Smaller grain sizes were found in the two-phase regions. The impurity  $\text{LaSrGa}_3\text{O}_7$  appearing for  $0.05 \leq y \leq 0.10$  is seen distributed over the grain boundaries. The impurity  $\text{LaSrGaO}_4$ , existing in samples with  $0.25 \leq y \leq 0.30$ , melts at 1400°C and is in a molten state at the sintering temperature 1470°C; it therefore acts as a flux and appears to drain away from the grain boundaries to form globular second-phase regions at grain-boundary intersections. TEM micrographs show well-bonded grain-boundary interfaces where x-ray diffraction shows a  $\text{LaSrGaO}_4$  second phase.

AC-impedance spectroscopy was used to investigate the grain-boundary contribution to the DC resistance and the influence of impurities on this contribution. The impurity  $\text{LaSrGa}_3\text{O}_7$  in samples  $0.05 \leq y \leq 0.10$  contributed to a perfect or a depressed grain-boundary semicircle in the complex-impedance plane. The depressed semicircle represents a constant-phase element (CPE) indicative of an oxide-ion insulator, *viz*, the  $\text{LaSrGa}_3\text{O}_7$  impurity, at the grain boundary. This grain-boundary semicircle vanishes if a hydrogen-containing atmosphere is applied, which indicates good proton conduction across the impurity phase at the grain boundaries. In contrast, the impurity  $\text{LaSrGaO}_4$  seen in samples with  $0.25 \leq y \leq 0.30$  gives no grain-boundary contribution to the impedance; in fact, the grain-boundary semicircle vanishes in these samples.

The performance of the optimized electrolyte  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.83}\text{Mg}_{0.17}\text{O}_{2.815}$  (LSGM) in a SOFC was tested on single cells having a 500-μm-thick electrolyte membrane and a

$\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-0}$  cathode. It was found that poor anode performance was due to a reactivity of the NiO in the composite  $\text{NiO}\cdot\text{CeO}_2$  or  $\text{NiO}\cdot\text{LSGM}$  anodes with LSGM to form  $\text{LaNiO}_3$  at the anode-electrolyte interface. To prevent this reaction, a thin Sm-doped  $\text{CeO}_2$  layer was introduced between the electrolyte and the anode. Comparison of  $\text{Sm}:\text{CeO}_2/\text{Sm}:\text{CeO}_2 + \text{Ni}$  and  $\text{Sm}:\text{CeO}_2 + \text{Ni}$  anodes showed that introduction of the  $\text{Sm}:\text{CeO}_2$  interlayer gave an exchange-current density four times larger. The peak power density of the interlayered thick-electrolyte cell was 100 mW higher than a cell without the interlayer. The improvement was shown to be due to a reduction of the anode overpotential. Comparison of the peak power density in this study with that of a previous study, also with a 500- $\mu\text{m}$ -thick electrolyte, shows a factor of two improvements, *i.e.*, from 270 mW/cm<sup>2</sup> to 550 mW/cm<sup>2</sup> at 800°C. This excellent cell performance indicates that an LSGM-based SOFC operating in the temperature range  $600^\circ\text{C} < T_{\text{op}} < 800^\circ\text{C}$  is a realistic goal.

Electric Power Research Institute (EPRI)

Contract Number: W08062-08

Wate Bakker, COR

Two year total contract

# Lanthanum Gallate as a New SOFC Electrolyte

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## Executive Summary

With the identification of Sr- and Mg- doped  $\text{LaGaO}_3$  as a superior oxide-ion electrolyte, the task of this grant was to optimize the composition and to evaluate its suitability for use in a solid oxide fuel cell (SOFC).

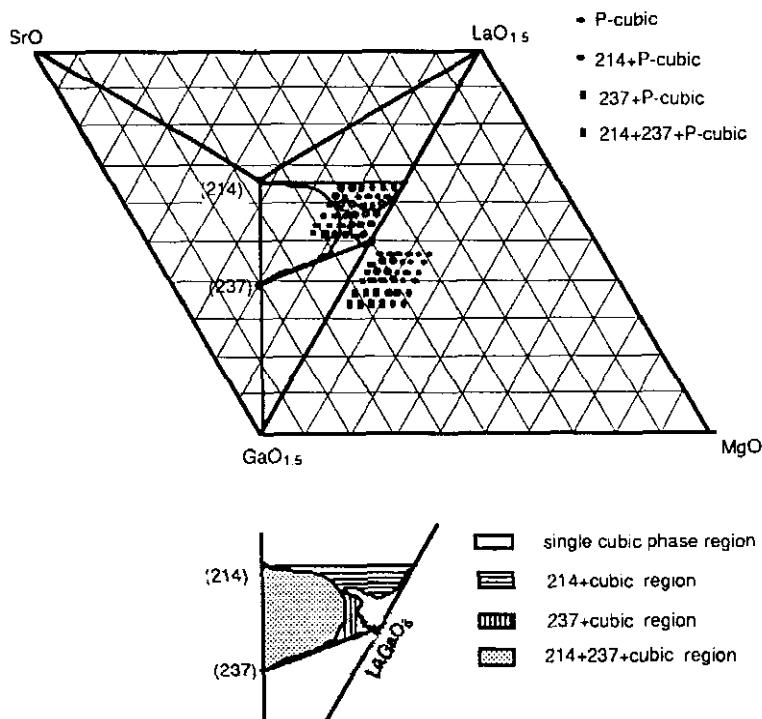


Fig. 1 Phase diagram of  $\text{LaO}_{1.5}-\text{SrO}-\text{GaO}_{1.5}-\text{MgO}$  system

A quaternary phase diagram for  $\text{LaO}_{1.5}\text{-SrO}\text{-GaO}_{1.5}\text{-MgO}$  was determined, Fig. 1; a single perovskite phase was found to be restricted to a narrow range of compositions  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$ .  $\text{LaSrGa}_3\text{O}_7$  and  $\text{LaSrGaO}_4$  were identified as impurity phases condensing out at the grain boundaries or the pores; excess MgO precipitated in the grains. The system was also found to tolerate vacancies in the  $\text{La}_{1-x}\text{Sr}_x$ -cation subarray, thereby allowing a greater Mg substitution. Calcining of commercial "Mg carbonate" at 1000 °C for more than 3 hours was needed to obtain a reliable MgO starting material.

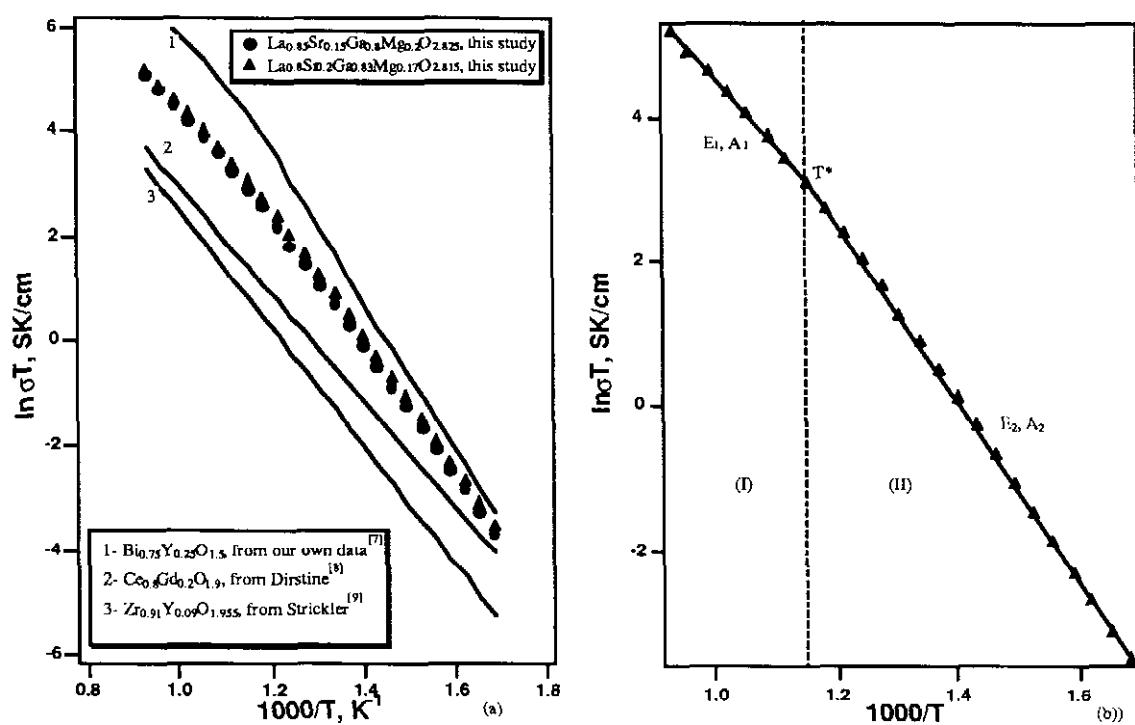


Fig. 2 (a) Arrhenius plots of conductivity for two typical compositions compared with other well-known oxide-ion conductor; (b) Separation into regions (I) and (II) at  $T^*$

Electrical measurements showed only oxide-ion conduction, without degradation in life tests, over the wide range of oxygen partial pressures existing in fuel cell operation ( $1 \leq P_{\text{O}_2} \leq 10^{-22}$  atm). Arrhenius plots, Fig. 2, of the oxide-ion conductivity  $\sigma_0$  versus reciprocal absolute temperature  $T$ ,  $\ln(\sigma_0 T)$  vs  $1/T$ ,

distinguished two temperature regions: for  $T \geq T^* \approx 600$  °C,  $A = A_1$  and  $E_a = E_1$ ; for  $T < T^*$ ,  $A = A_2$  and  $E_a = E_2$  in the expression

$$\sigma_0 T = A \exp(-E_a/kT)$$

This behavior was interpreted with a model in which the oxygen vacancies are disordered above  $T^*$ , which makes  $E_1 = \Delta H_m$ , the motional enthalpy, whereas below  $T^*$  the vacancies become progressively trapped out at a vacancy-ordered condensate by a trapping energy  $\Delta H_t$ , which makes  $E_2 = \Delta H_m + \Delta H_t$  and  $A_2 = A_1 \exp(\Delta H_t/kT^*)$ . At 800 °C, the region of highest oxide-ion conductivity,  $\sigma_0 > 0.14$  S/cm, falls in the compositional ranges  $0.125 \leq x \leq 0.25$  and  $0.125 \leq y \leq 0.25$ . The maximum oxide-ion conductivities found at 800 °C, Fig. 3, 700 °C, and 600 °C were, respectively, 0.17, 0.08 and 0.03 S/cm for  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.83}\text{Mg}_{0.17}\text{O}_{2.825}$ .

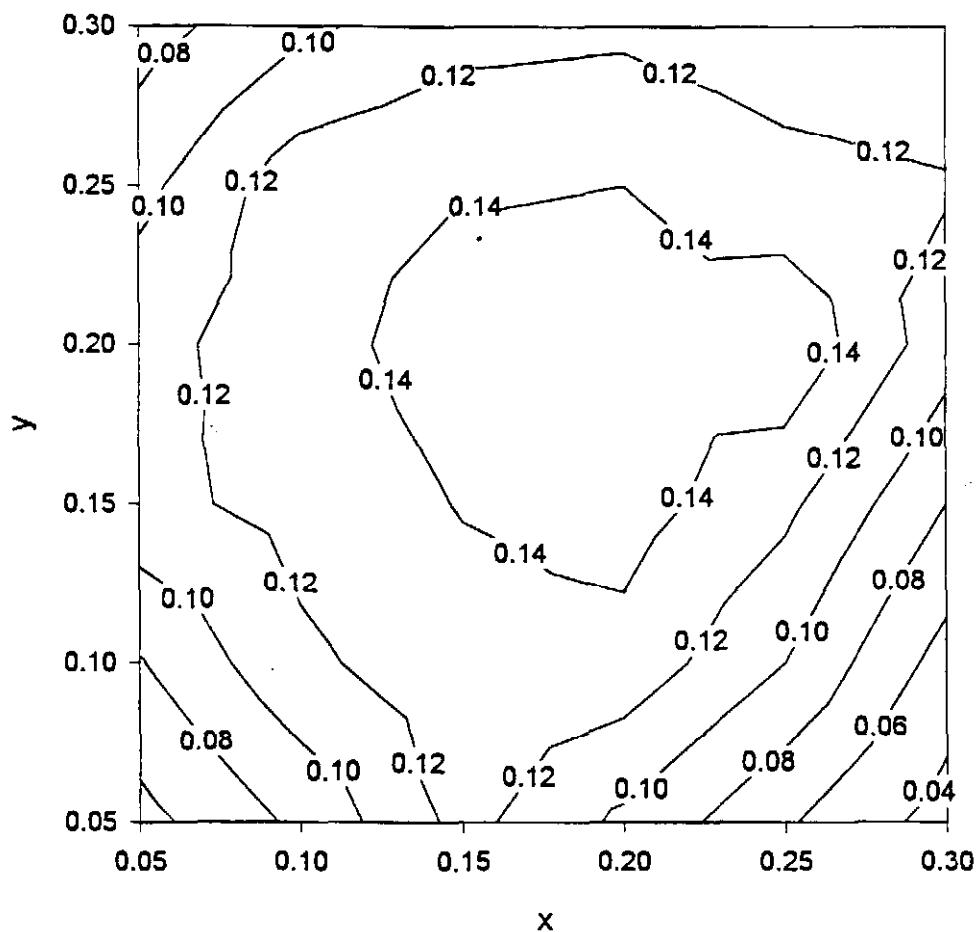


Fig. 3 Isoconductivity diagram of  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$  at 800 °C

SEM microstructures of samples with  $x = 0.20$  showed precipitation of  $\text{LaSrGa}_3\text{O}_7$  over the grain boundaries for  $y \leq 0.10$  and  $\text{LaSrGaO}_4$  particles pinning the grain boundary for  $y \geq 0.25$ ; in addition, intragranular  $\text{MgO}$  precipitates were found for  $y = 0.30$ . AC-impedance spectroscopy showed an important grain-boundary contribution to the oxide-ion impedance for  $y \leq 0.10$ , but no grain-boundary contribution for larger values of  $y$ . The  $\text{LaSrGaO}_4$  phase is liquid at the sintering temperature and promotes strong intergranular bonding.

Three soft-chemical routes for the synthesis of high-purity, ultra-homogeneous  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$  powders were compared; the sol-gel process, co-precipitation, and hydrothermal reaction. Powders produced by these routes reduce the sintering temperature required for materials processing. The powders obtained were characterized by x-ray diffraction, thermal analysis, transmission electron microscopy, and fourier-transformed infrared spectroscopy in order to elucidate the reaction mechanisms operating in each synthesis. The electrolyte could be prepared as a single phase by both the sol-gel and the Pechini co-precipitation methods, but not by hydrothermal treatment.

Chemical compatibility between the electrodes and the electrolyte were investigated by monitoring with EDS the diffusion of cations across the interface between them. Conventional cathodes were studied: porous  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  and films of  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ . The former did not react with the electrolyte; the latter interdiffused at higher temperature, but not appreciably at an operating temperature of 800 °C. An electrode that conducts both oxide ions and electrons allows use of an electrode film; an electrode that conducts only electrons must be made as a thick, porous layer.  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  has a thermal expansion compatible with the electrolyte, but it only conducts electrons.  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$  conducts both electrons and oxide ions, but it has an anomalously large thermal expansion. Nevertheless, we used  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$  cathodes in our fuel-cell tests because of its excellent mixed electronic and oxide-ion conduction.

Conventional anodes are formed from composites of NiO with either the electrolyte or  $\text{Ce}_{1-x}\text{Sm}_x\text{O}_{2-0.5x}$ . The reducing atmosphere at an anode reduces the NiO to elemental Ni deposited on the walls of a porous structure. However, we found that interdiffusion of Ni and Ga across the electrode-electrolyte interface introduces at the interface metallic  $\text{LaNiO}_3$ , which blocks oxide-ion conduction. To circumvent this problem, we introduced  $\text{Ce}_{1-x}\text{Sm}_x\text{O}_{2-0.5x}$ , which conducts both oxide ions and electrons in a reducing atmosphere, as a thin buffer layer between the electrolyte and a  $\text{Ce}_{1-x}\text{Sm}_x\text{O}_{2-0.5x} + \text{NiO}$  anode.

Single fuel cells were constructed for testing. Introduction of the buffer layer at the anode reduced significantly the anode overpotential and extended the life of the cell, Table 1. With a 500- $\mu\text{m}$ -thick electrolyte, maximum power densities of 550 mW/cm<sup>2</sup> at 1.1A/cm<sup>2</sup> were obtained, Fig. 4. A life test showed a stable power output for at least a period of 1500 hours under a loading current 250 mA/cm<sup>2</sup>. Reducing the thickness of the electrolyte would greatly improve performance.

**Table 1 Resistances ( $\Omega$ ) at a frequency of 5Hz at 800 °C in the atmosphere of wet hydrogen**

time (h)	anodes	LSGM+NiO	Sm-CeO <sub>2</sub> /LSGM+NiO	CeO <sub>2</sub> +NiO	Sm-CeO <sub>2</sub> / Ca-CeO <sub>2</sub> +NiO
0		648	146	354	78
5		691	124	273	74
10		622	124	284	74
15		611	124	295	74
20		625	126	306	75

In conclusion, we have demonstrated that the  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$  electrolyte promises to provide a SOFC operating at 800 °C with a thick-film (*ca.* 100  $\mu\text{m}$ ) electrolyte, at 600 °C with a thin-film (*ca.* 10  $\mu\text{m}$ ) electrolyte.

The fuel-cell tests reported were done in collaboration with Dr. Chris Milliken, Dr. Ashok Khankar and Dr. S. Elangovan of Ceramatec, Inc.. We are grateful for their valuable cooperation.

Sponsor: Electric Power Research Institute (EPRI)  
Contract Number: WO 8062-08  
Dr. Wate Bakker, COR  
Two year total contract

**Keywords:** Solid electrolyte, oxide-ion; perovskite, lanthanum gallate; solid oxide fuel cell; phase diagram,  $\text{LaO}_{1.5}\text{-SrO}\text{-GaO}_{1.5}\text{-MgO}$ ; ionic conduction; buffer layer.

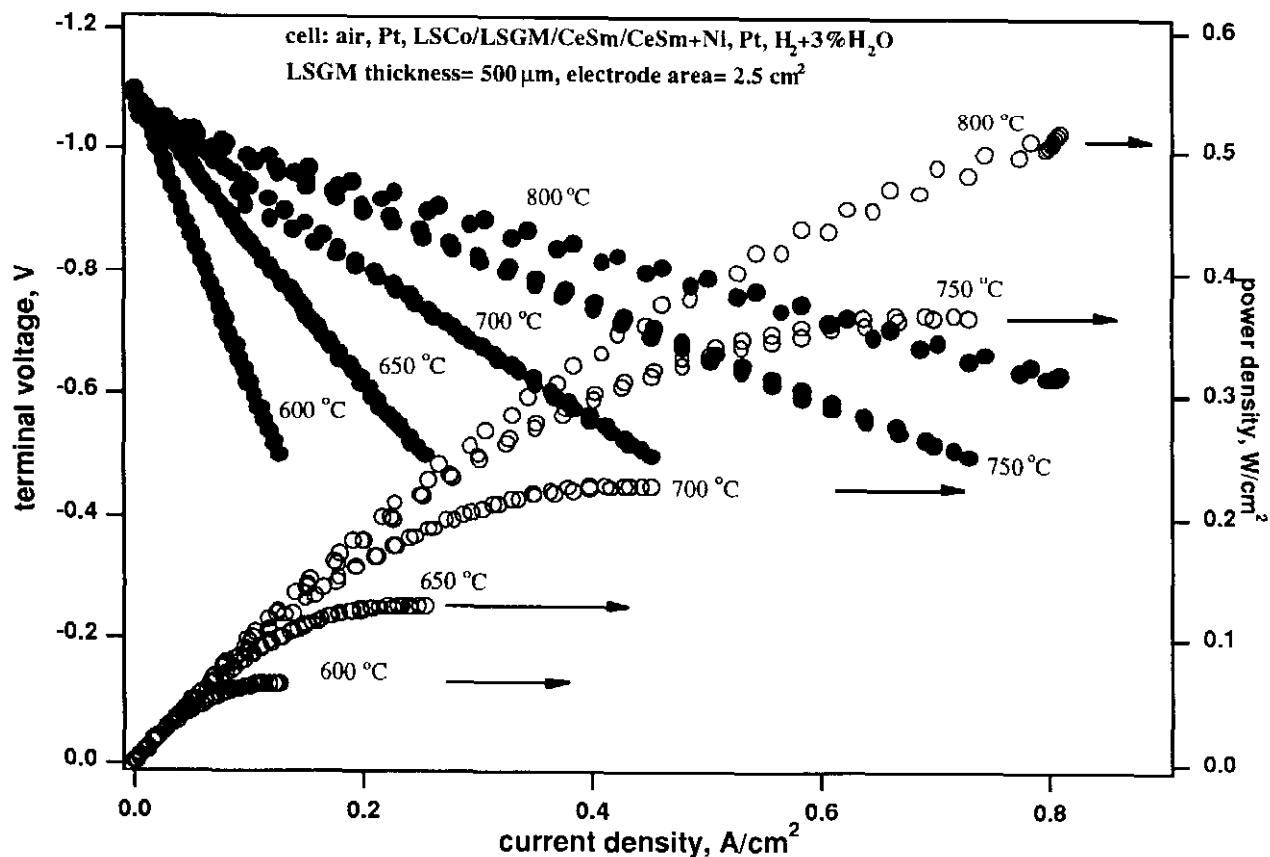


Fig. 4 Single fuel cell performance based on LSGM electrolyte with LSCo as cathode and  $\text{Sm-CeO}_2/\text{Sm-CeO}_2\text{+Ni}$  bilayer anode at different temperatures

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# LANTHANUM GALLATE AS A NEW SOFC ELECTROLYTE

John B. Goodenough & Keqin Huang

EPRI Sponsor: Dr. Wate T. Bakker

# OUTLINE

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- Optimal compositions in  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$  system
- Condensate of short-range order  $T < T^* \approx 600^\circ\text{C}$
- Microstructures and ac impedance spectroscopy
- Introduction of anode layer

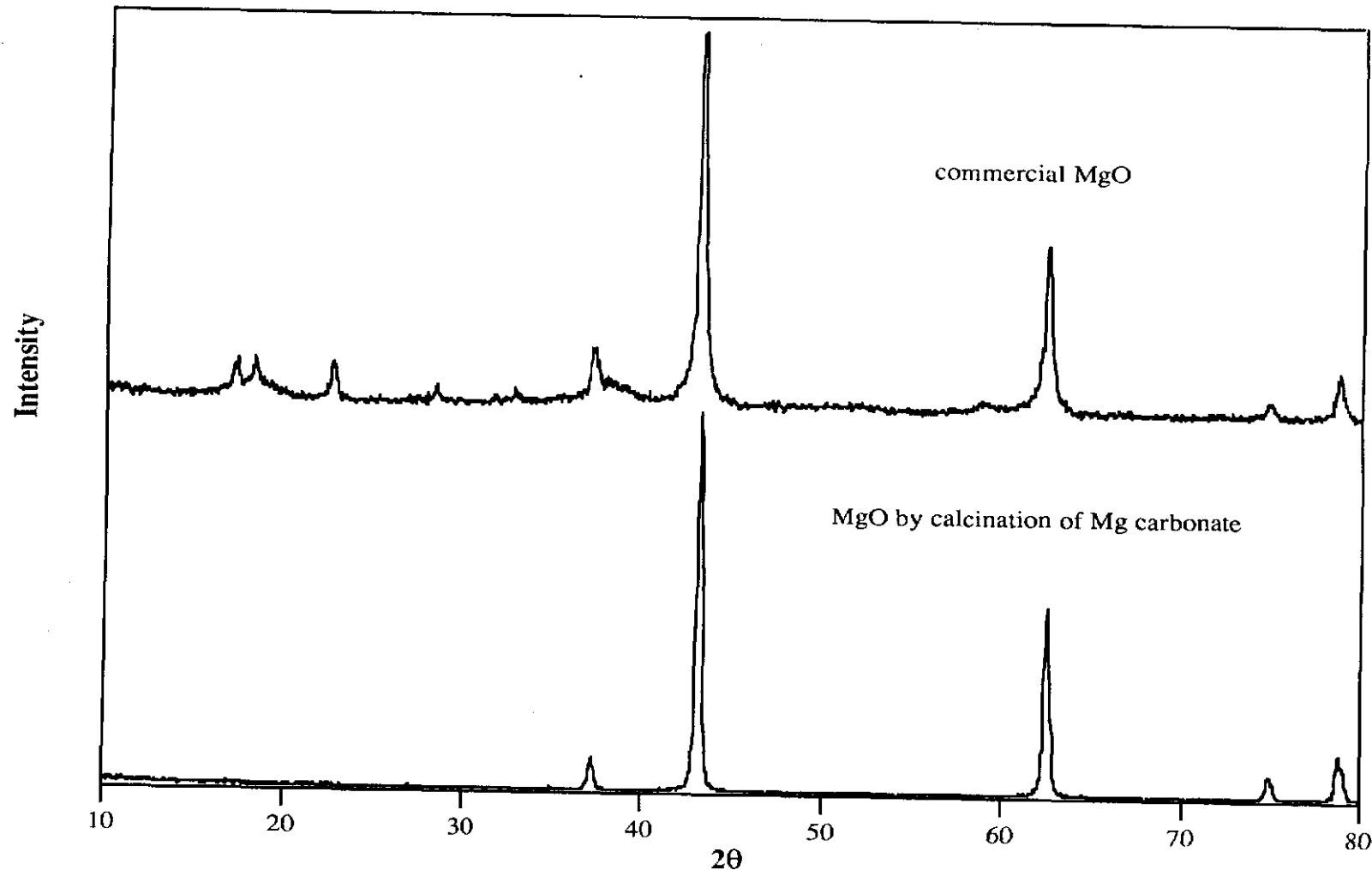
# SAMPLE PREPARATION

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- Intimate mixing of  $\text{La}_2\text{O}_3$ ,  $\text{SrCO}_3$ ,  $\text{Ga}_2\text{O}_3$  and  $\text{MgO}^*$
- Pressed pellets fired overnight at 1250 °C
- Partially sintered pellets reground and ball-milled
- 6-mm-diameter x 5-7 mm pellets fired 24 h at 1470 °C

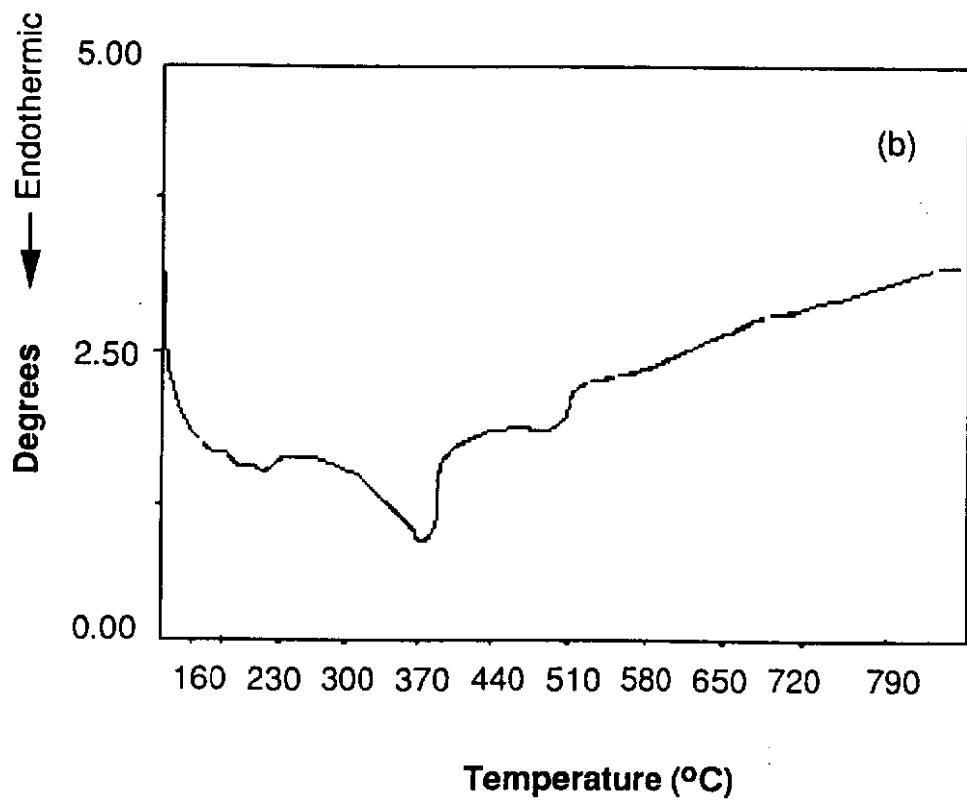
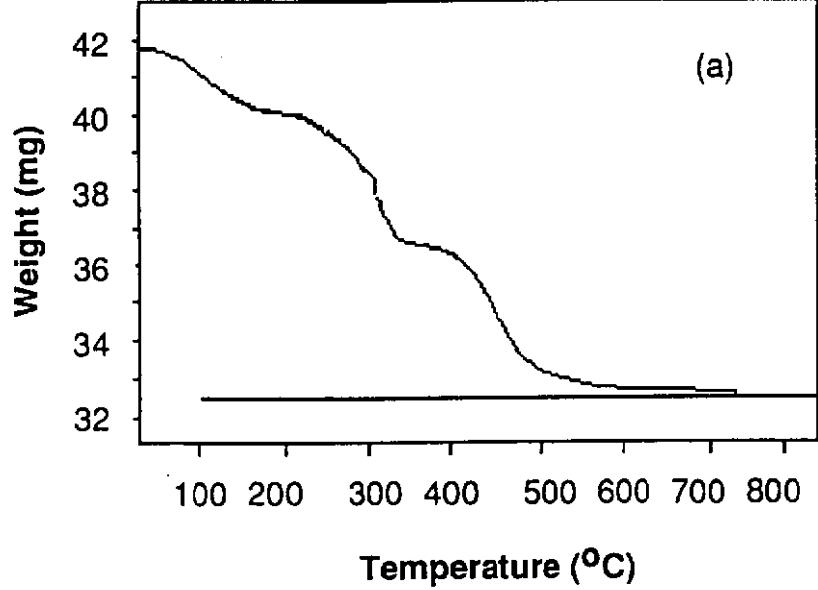
*\* after calcining Mg carbonate for 4 h at 1000 °C*

# XRD Patterns of Dried and Undried Commercial MgO

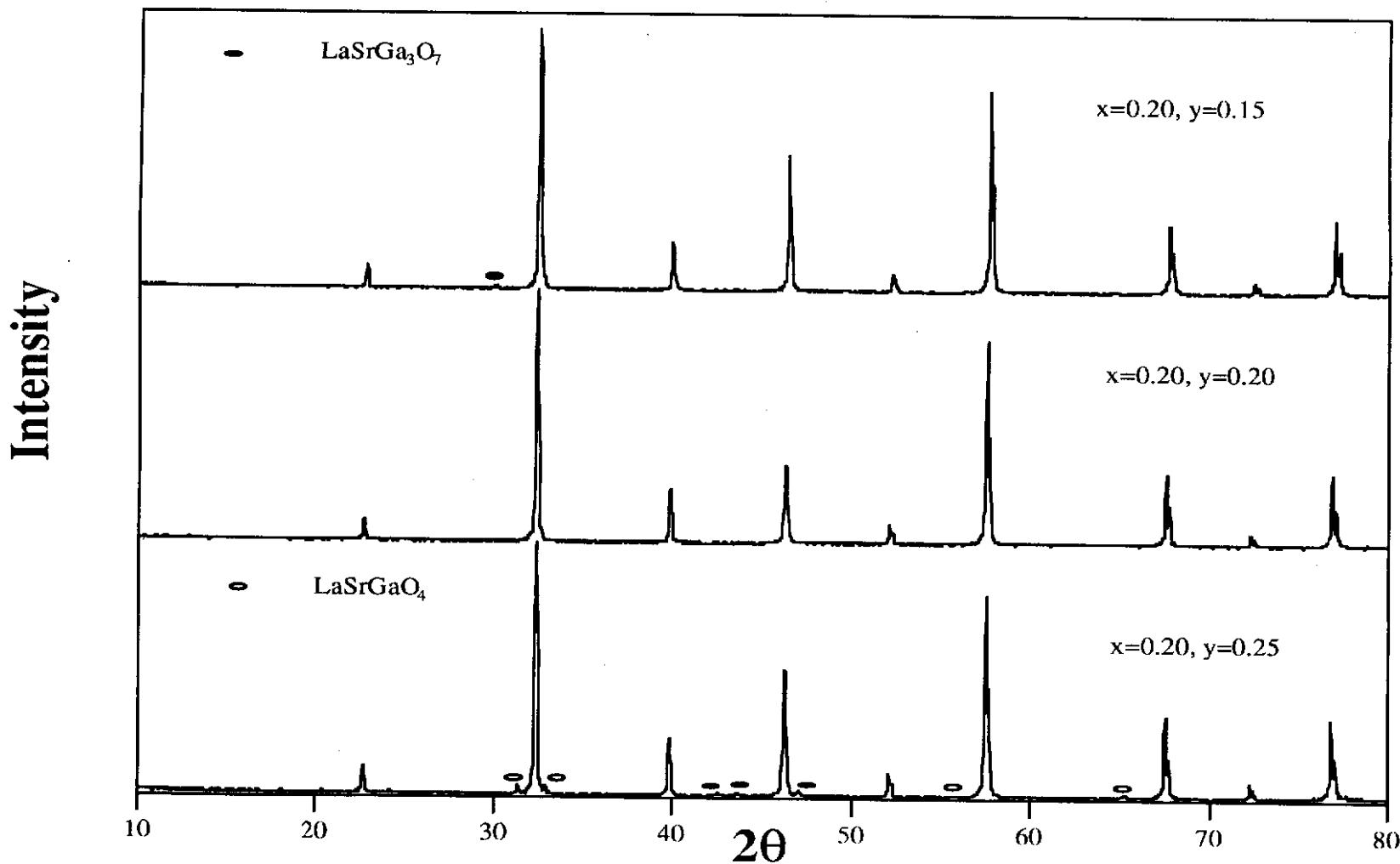


# TGA/DTA Curves of Commercial MgO

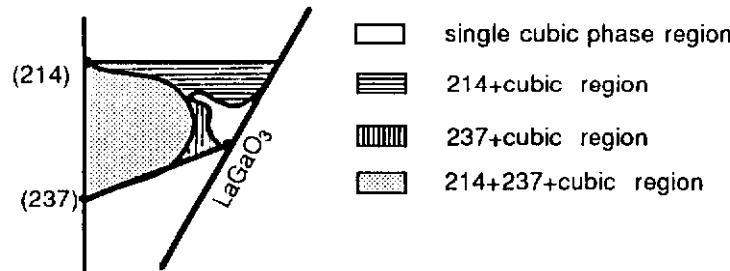
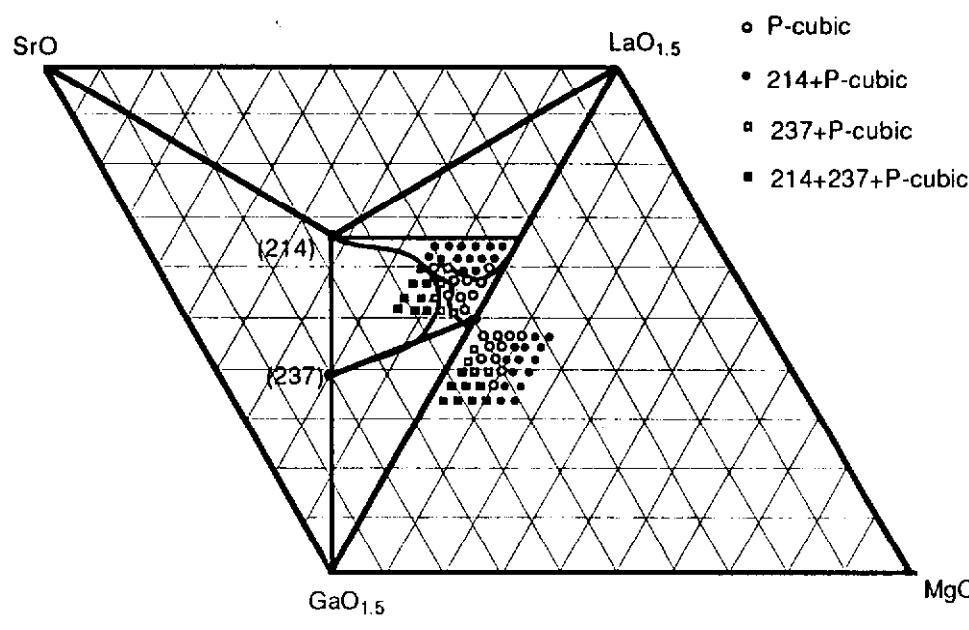
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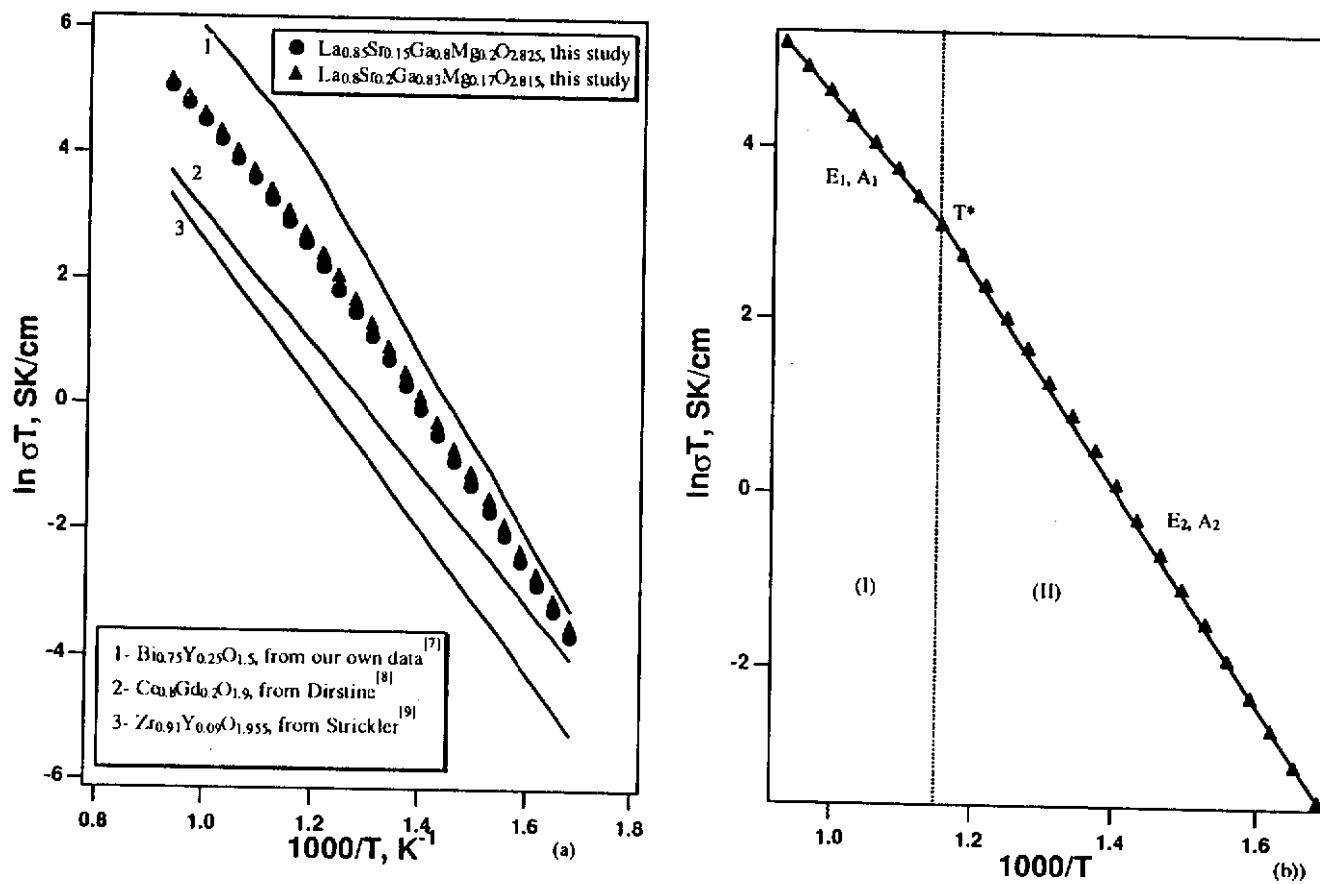
# XRD Patterns of $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(0.2+y)}$



# Phase Diagram of $\text{LaO}_{1.5}\text{-SrO-GaO}_{1.5}\text{-MgO}$



# Conductivity vs Temperature



# IONIC CONDUCTIVITY

---

$$\mu_O = qD/kT = (qD_O/kT)\exp(-\Delta G_m/kT)$$

$$\sigma_O = cNq\mu_O = (A/T)\exp(-E_a/kT)$$

Assume:

$T \geq T^*$ , vacancies disorder

$\therefore E_a = E_1 = \Delta H_m$ ,  $c = c^*$ ,  $A = A_1$

$T < T^*$ , vacancies progressively trapped  
in condensate

$$\ln(c/c^*) = -(\Delta H_t/kT)[1 - (T/T^*)]$$

$\therefore E_a = E_2 = \Delta H_m + \Delta H_t$ ,  $A_2 = A_1 \exp(\Delta H_t/kT^*)$

$$\ln(A_2/A_1) = \Delta H_t/kT^* = (E_2 - E_1)/kT^*$$

# PROOF

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Let:

$\mu_V^*(\text{cond})$  = chemical potential of condensate

$\mu_V^\circ(\text{sol})$  = chemical potential of pure solid solution

$$\Delta G_t(T) = \mu_V^\circ(\text{sol}) - \mu_V^*(\text{cond})$$

At equilibrium:

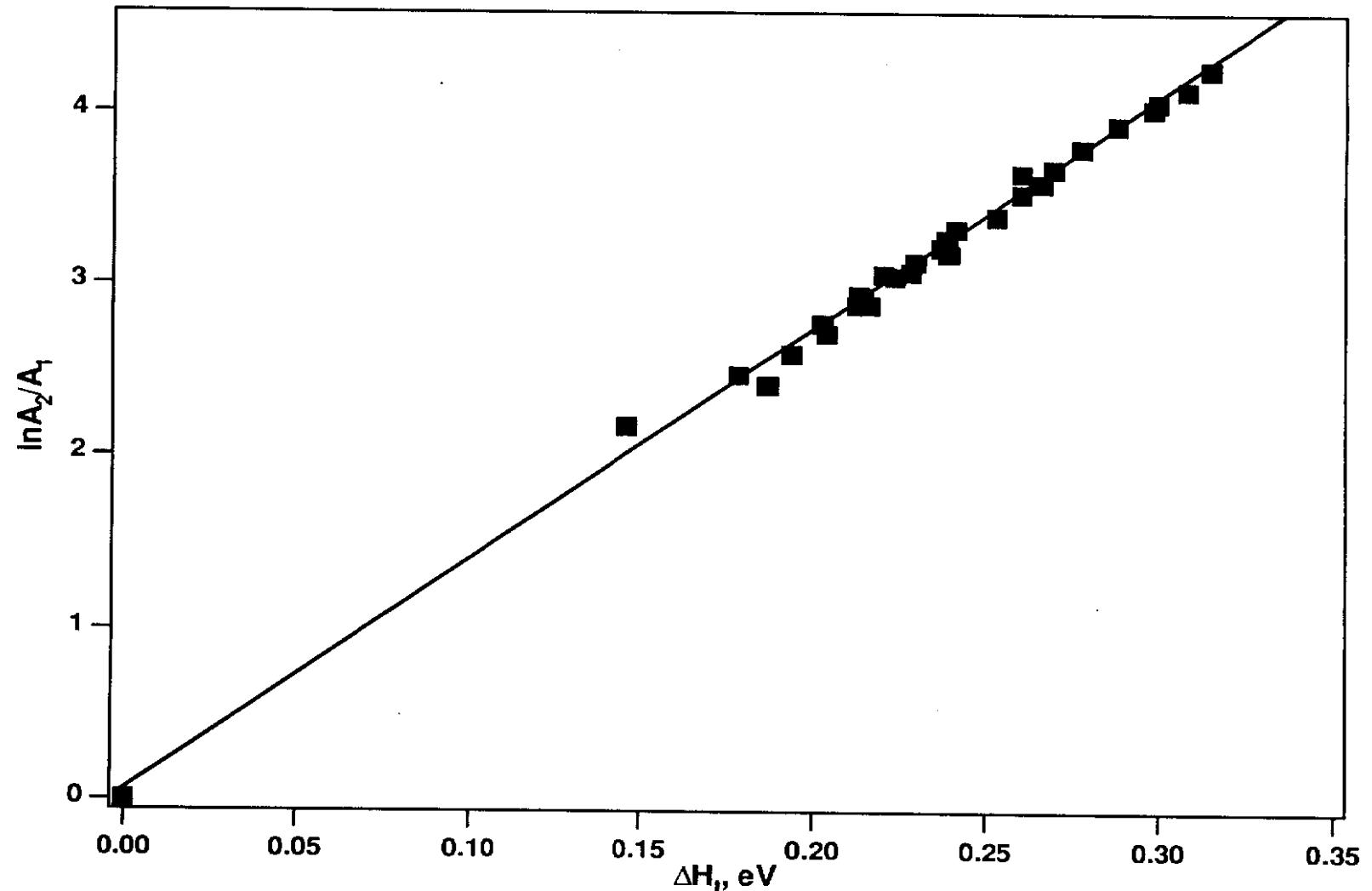
$$\mu_V^*(\text{cond}) = \mu_V^*(\text{sol}) = \mu_V^\circ(\text{sol}) + kT \ln(c/c^*)$$

Since  $\Delta G_t(T^*) = 0$

$$\begin{aligned} \ln(c/c^*) &= -[\Delta G_t(T)/kT - \Delta G_t(T^*)/kT^*] \\ &= -\Delta H_t(1-T/T^*)/kT \end{aligned}$$

Note:  $T^* = 599 \pm 49$  °C for all samples

# $\ln A_2/A_1$ vs $\Delta H_t$



# INFLUENCE OF DOPING METHOD

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If method of doping, *i. e.*, Sr for La *vs* Mg for Ga, to obtain a given  $c^*$  effects  $\Delta H_t$ , but not  $\Delta H_m$  or  $T^*$ , then:

$\ln(\sigma_0 T)$  vs  $(1/T)$  is  $\left\{ \begin{array}{l} \text{common for } T \geq T^* \\ \text{diverges for } T < T^* \end{array} \right.$

# MEYER-NELDER RULE

## OBEYED $T < T^*$

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For some semiconductors with  $\sigma_0 T = A \exp(-E_a/kT)$

$$\ln A = aE_a + b$$

is found empirically. In our case with  $T \leq T^*$ ,

$$\sigma_0 T = B \exp(-E_2/kT)$$

$$\sigma_0 T^* = B \exp(-E_2/kT^*) = A \exp(-\Delta H_m/kT^*)$$

∴

$$\ln B = aE_2 + b$$

with  $E_2 = \Delta H_m + \Delta H_t$ ,  $a = 1/kT^*$ ,  $b = \ln A - (\Delta H_m/kT^*)$

# MOBILITY & DIFFUSIVITY

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For  $T \geq T^*$

$$\sigma_O = c^* N q \mu_O = 2 e c^* \mu_O / a^3 = (A/T) \exp(-\Delta H_m / kT)$$

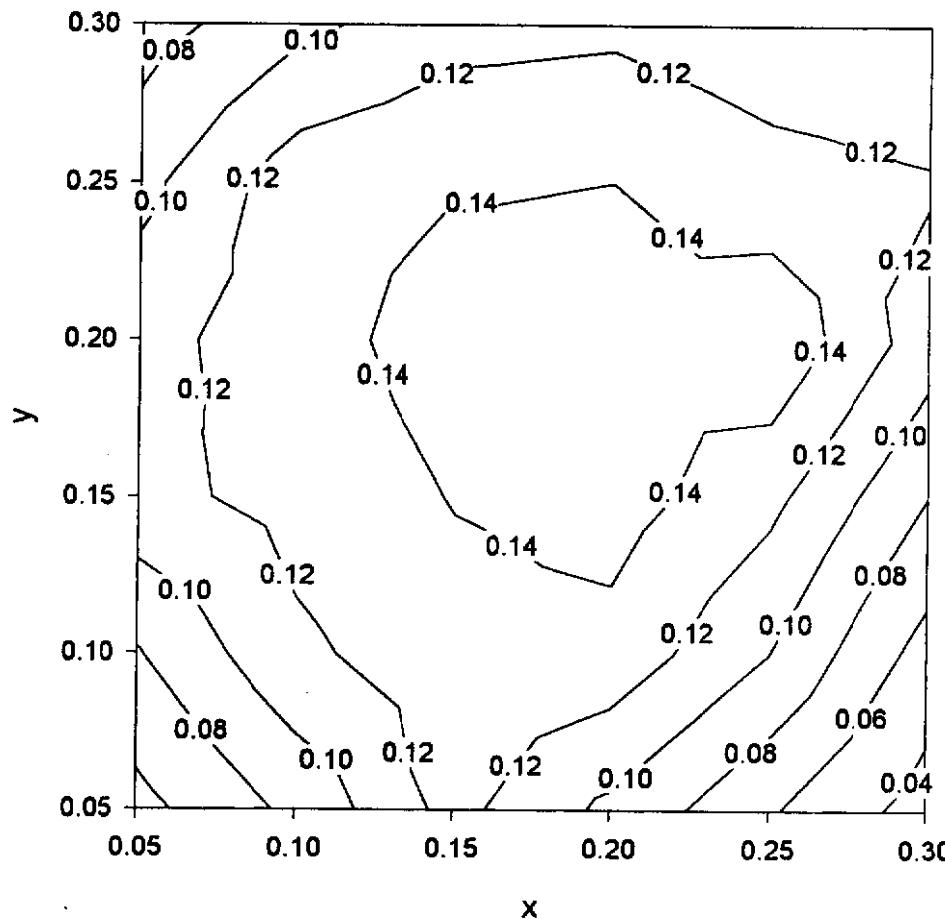
$$\therefore \mu_O \exp(\Delta H_m / kT) = A a^3 / 2 e c^* T = e D_O / kT$$

We measure  $\Delta H_m$ ,  $A$ , lattice constant  $a$ ,  $c^*$  to obtain:

<b>Sample</b>		<b><math>c^*</math></b>	<b><math>\mu_O, \times 10^{-5} \text{cm}^2/\text{Vs}</math></b>			<b><math>D_O, \times 10^{-6} \text{cm}^2/\text{s}</math></b>		
$x$	$y$		600	700	800 $^{\circ}\text{C}$	600	700	800 $^{\circ}\text{C}$
0.15	0.15	0.05	8.72	26.3	53.9	3.29	11.0	24.9
0.15	0.20	0.06	7.16	22.9	49.2	2.70	9.60	22.8
0.20	0.15	0.06	8.12	23.8	48.7	3.06	9.98	22.6

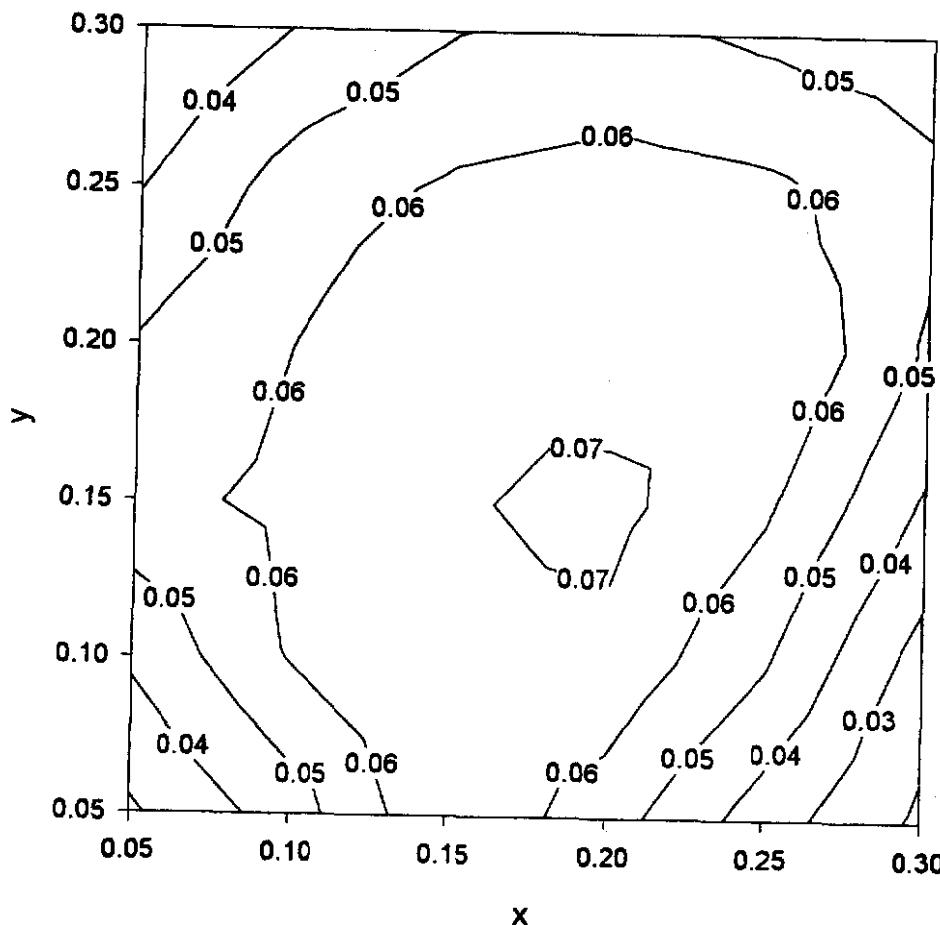
# Isoconductivity Contours at 800 °C (S/cm)

for  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$



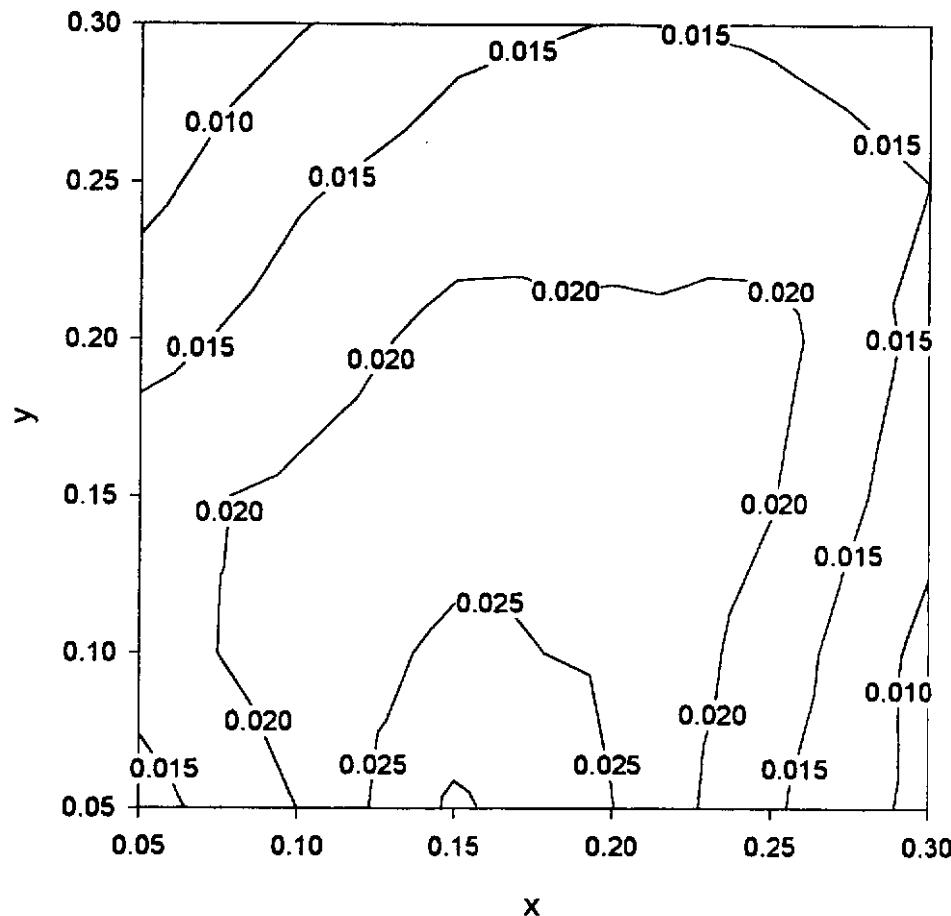
# Isoconductivity Contours at 700 °C (S/cm)

for  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$

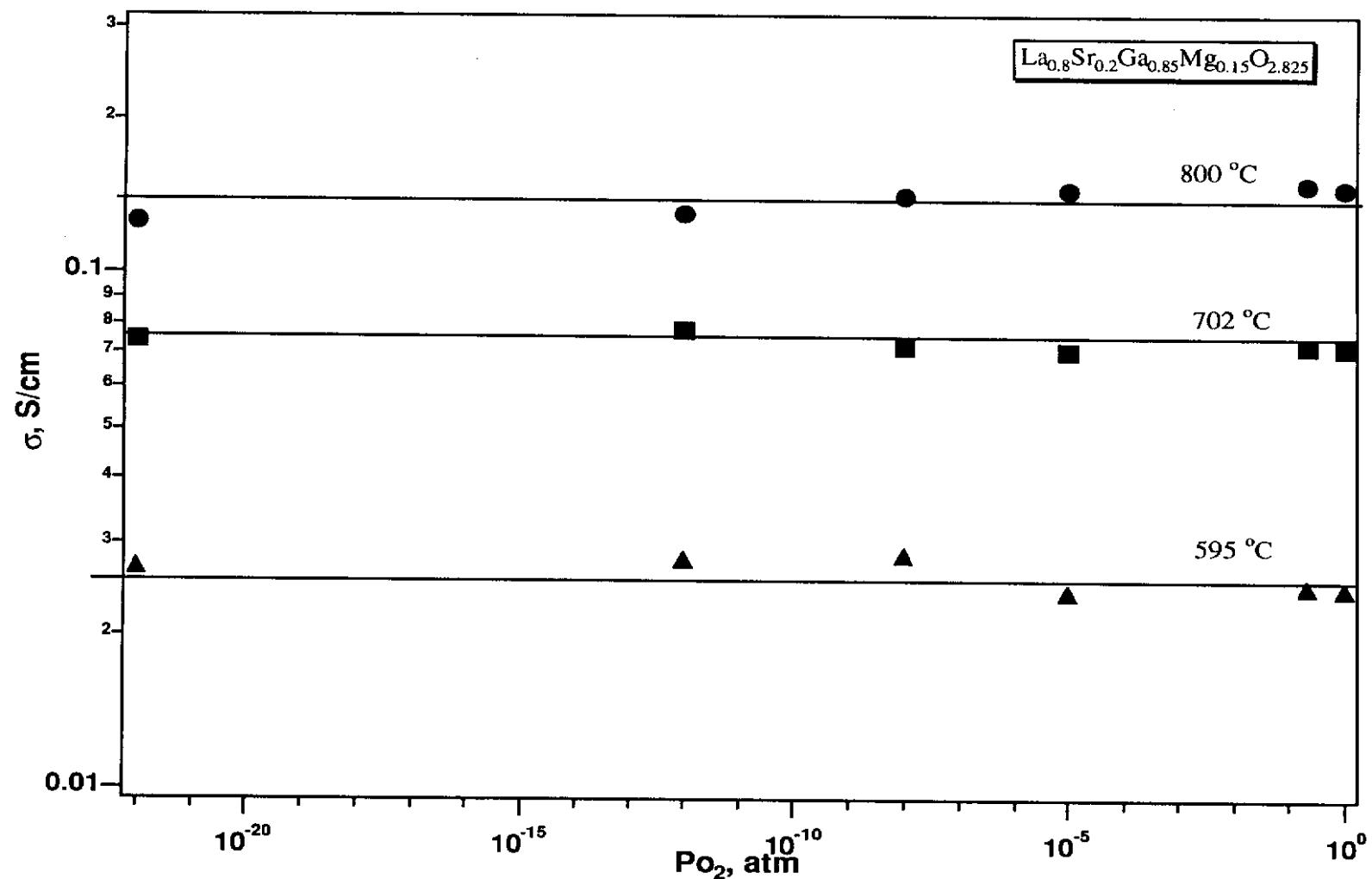


# Isoconductivity Contours at 600 °C (S/cm)

for  $\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$

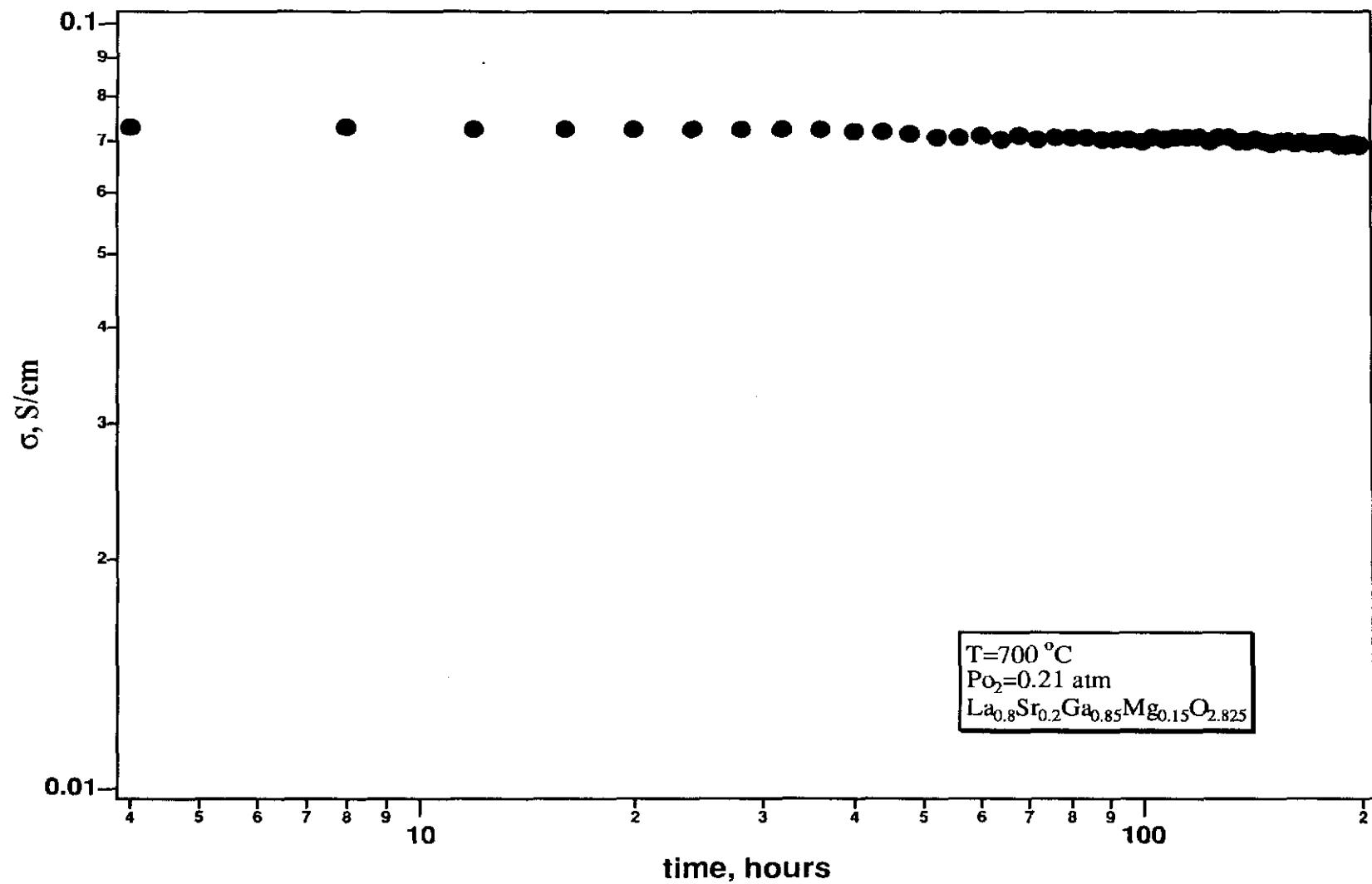


# Conductivity *vs* $P_{O_2}$



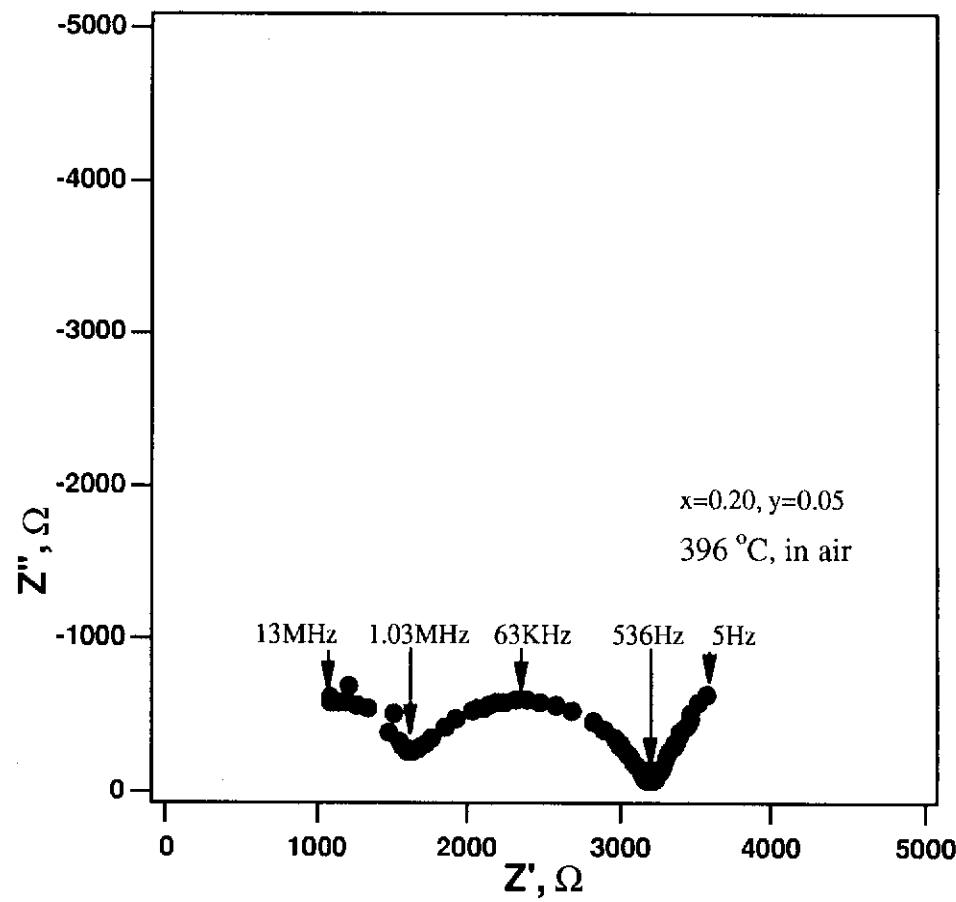
# Conductivity *vs* time

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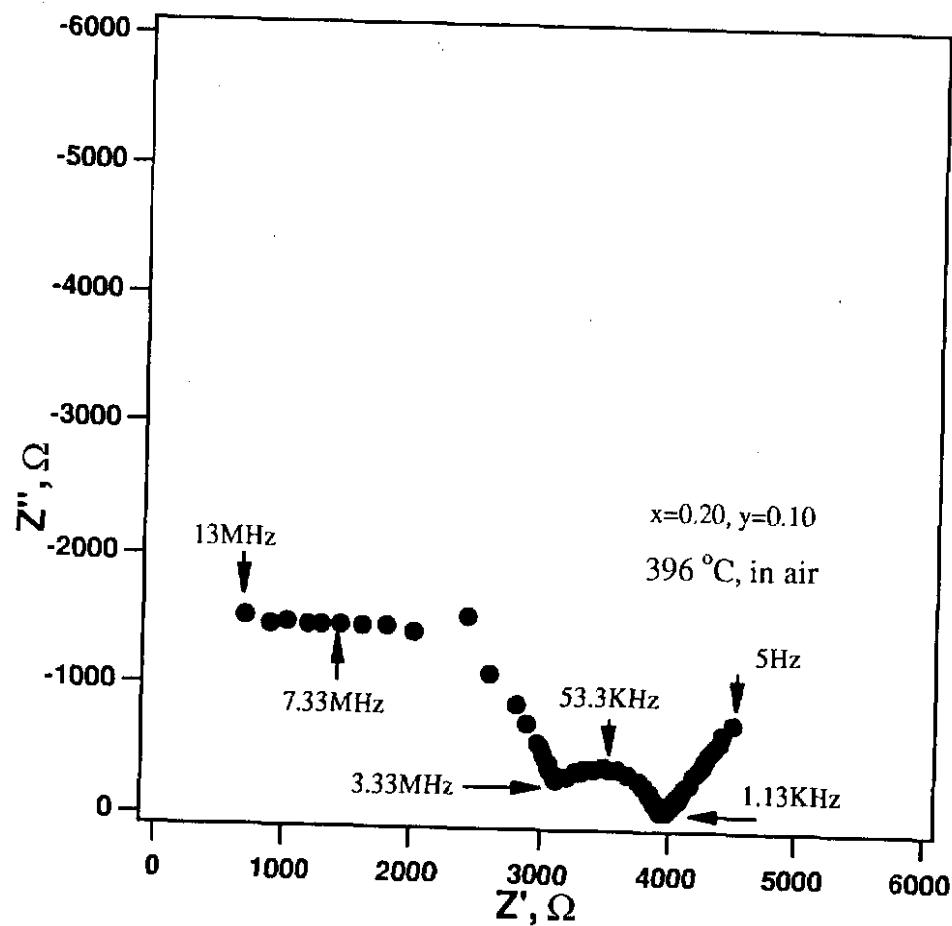


# Impedance Spectrum

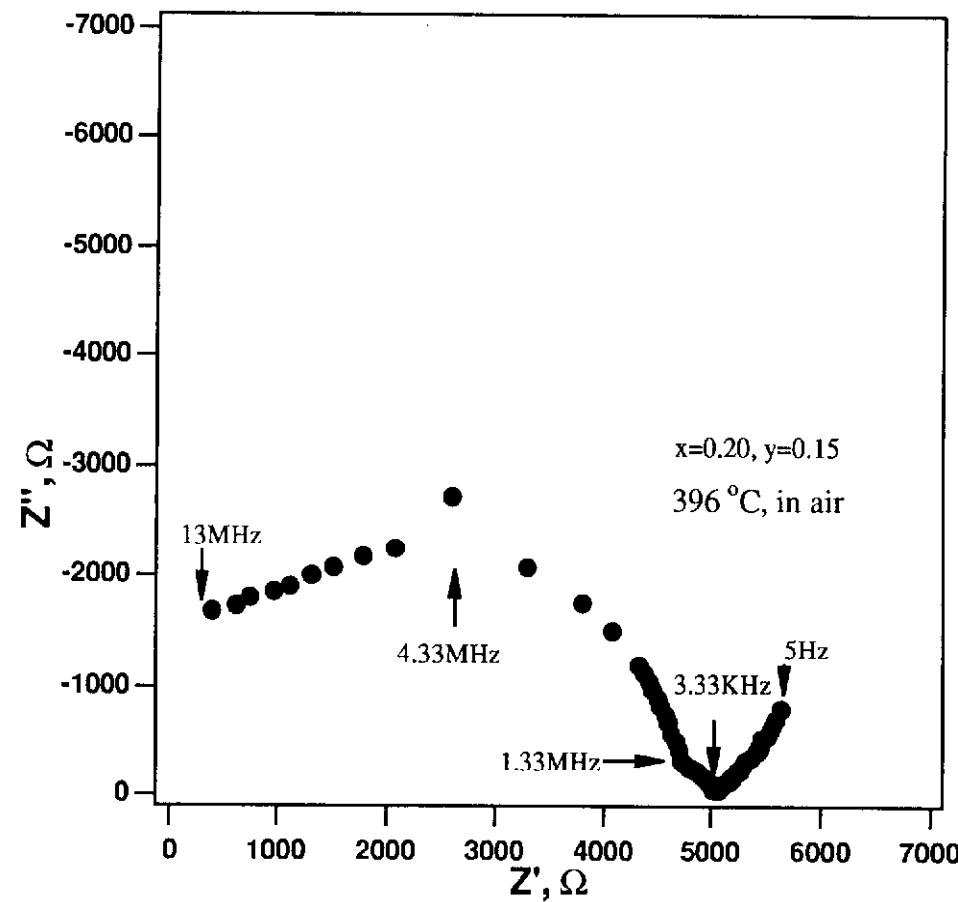
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# Impedance Spectrum

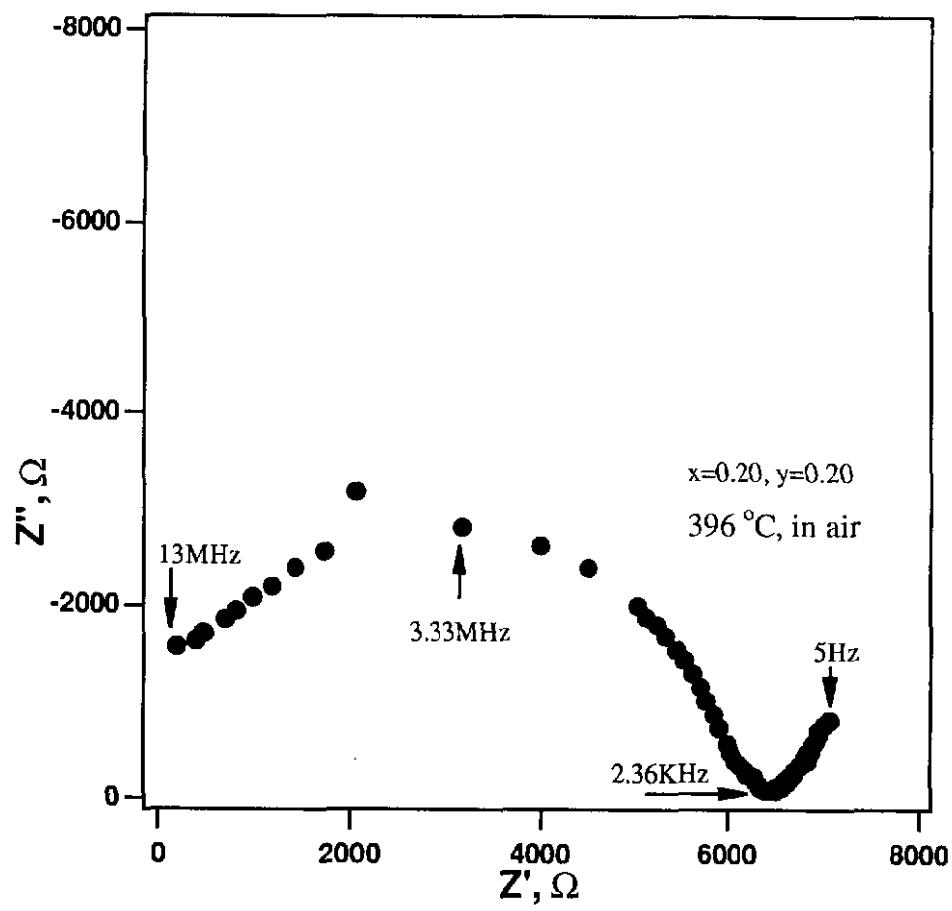


# Impedance Spectrum



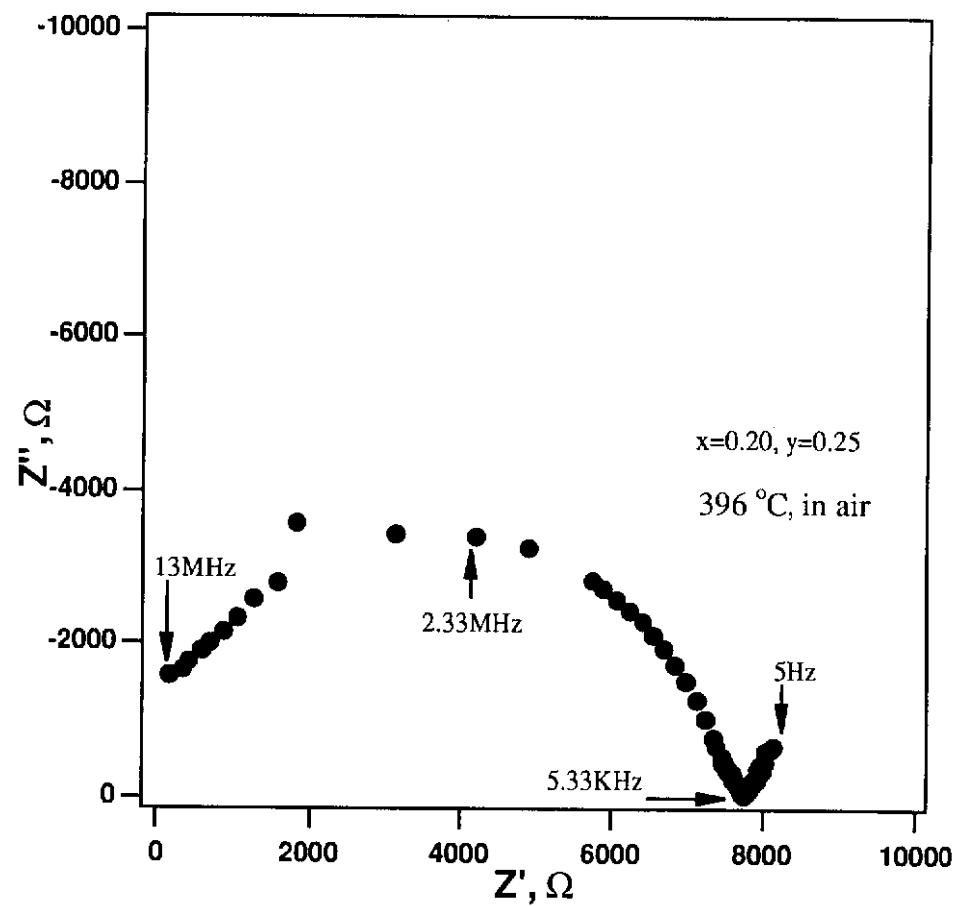
# Impedance Spectrum

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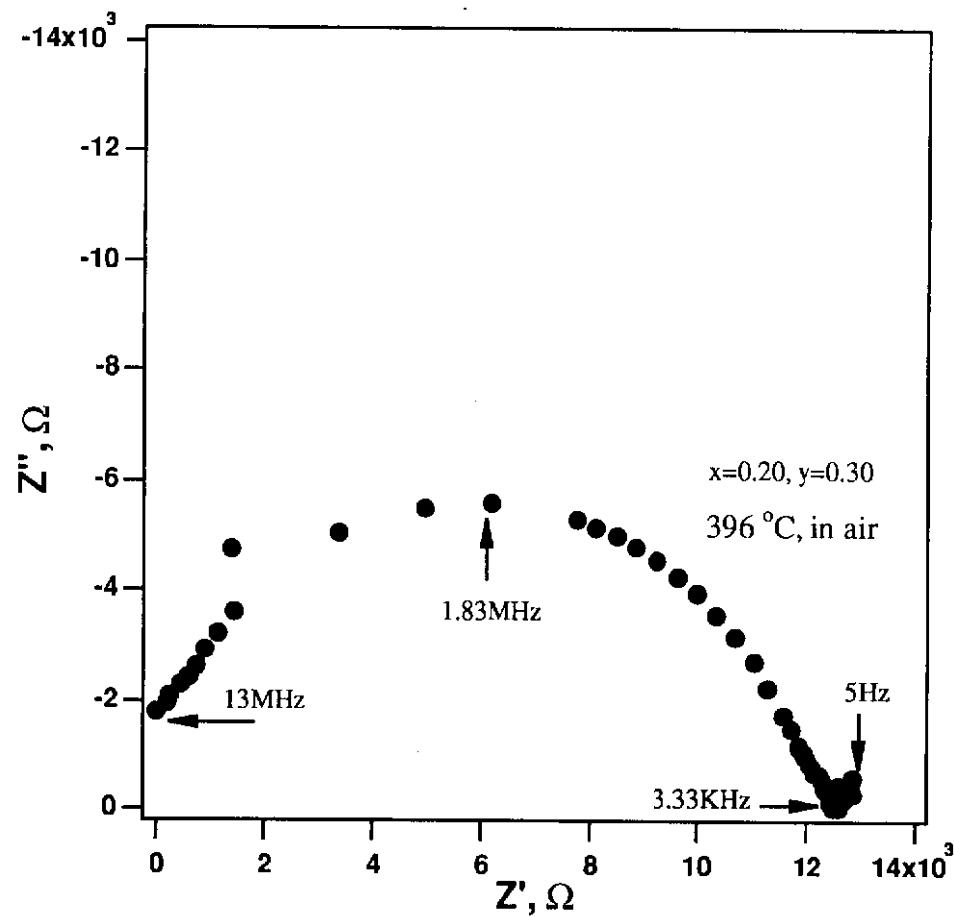


# Impedance Spectrum

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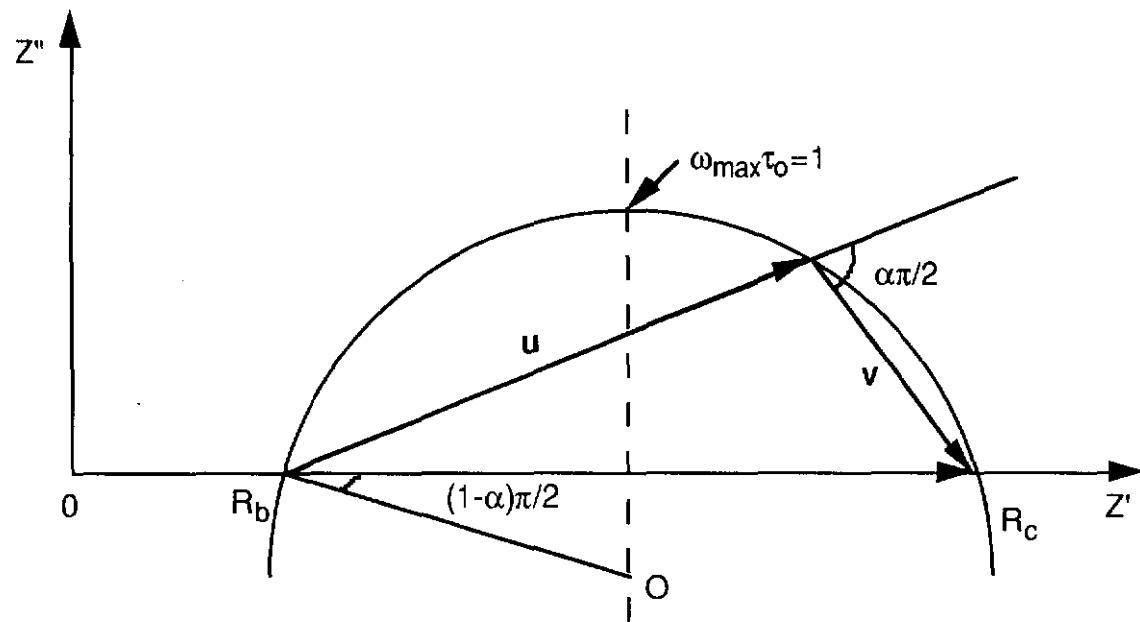


# Impedance Spectrum



# A Graphic Presentaion of CPE

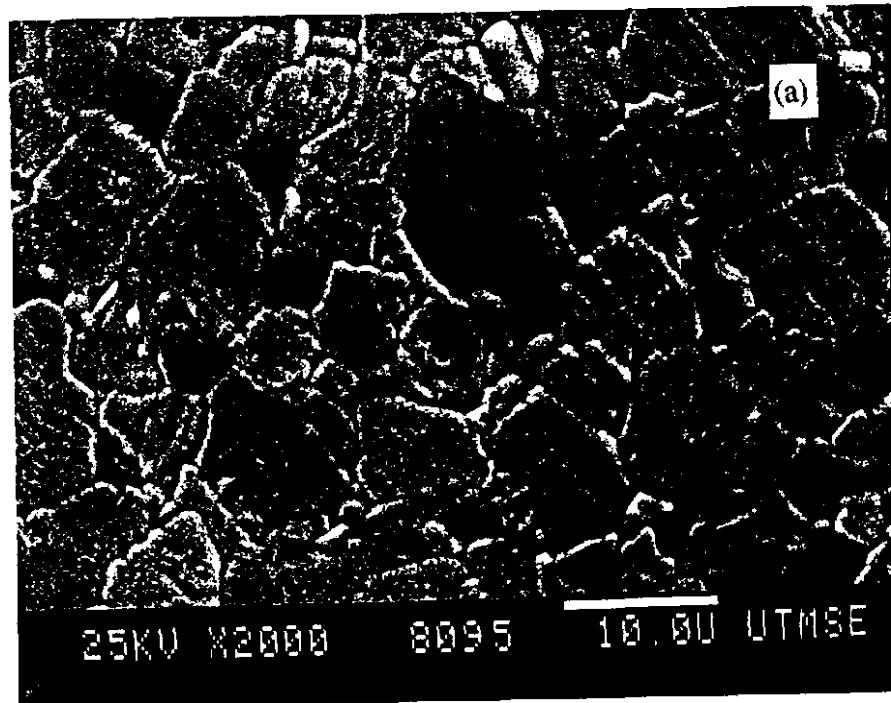
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# SEM Microstructures

## $x=0.2, y=0.05$

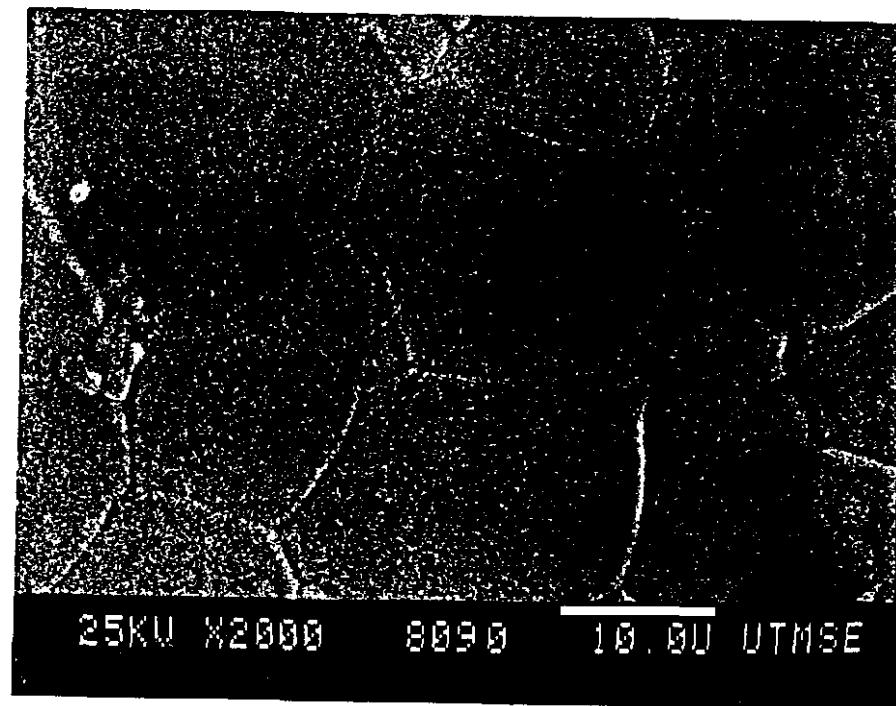
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thermal etch at 1350 °C for 1 h

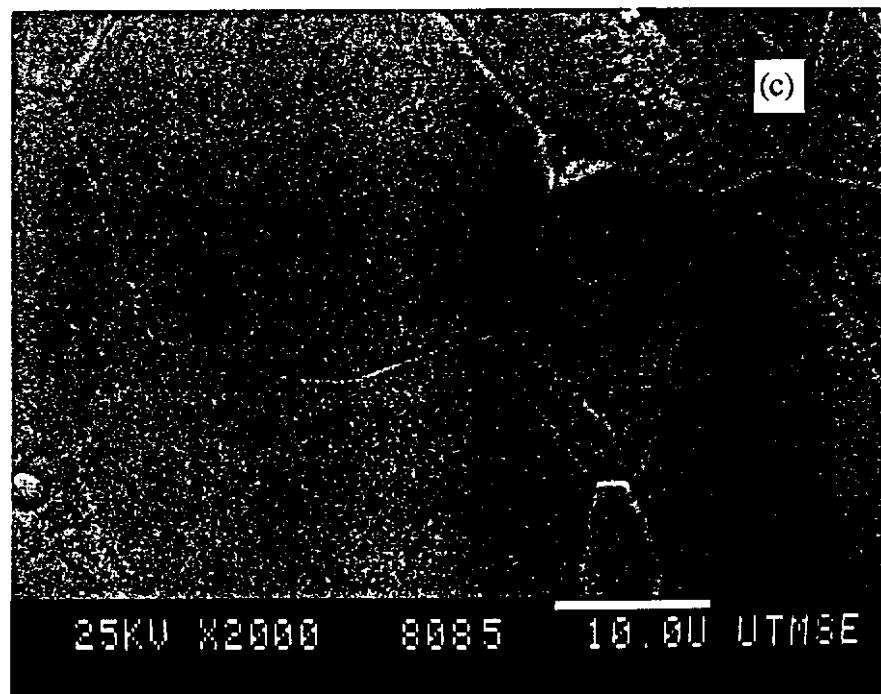
# SEM Microstructures

$x=0.2, y=0.1$



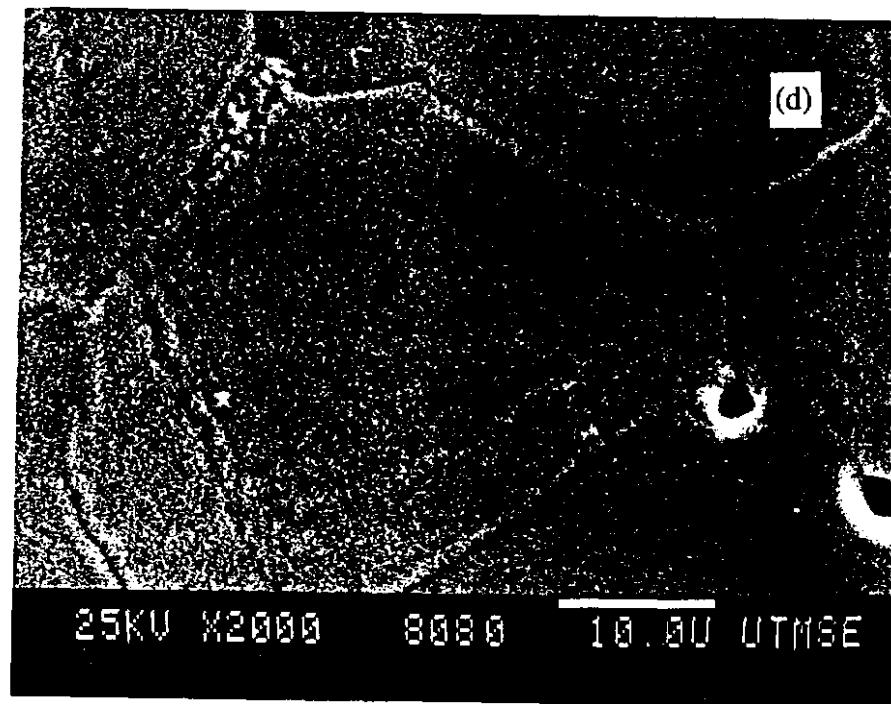
# SEM Microstructures

$x=0.2, y=0.15$



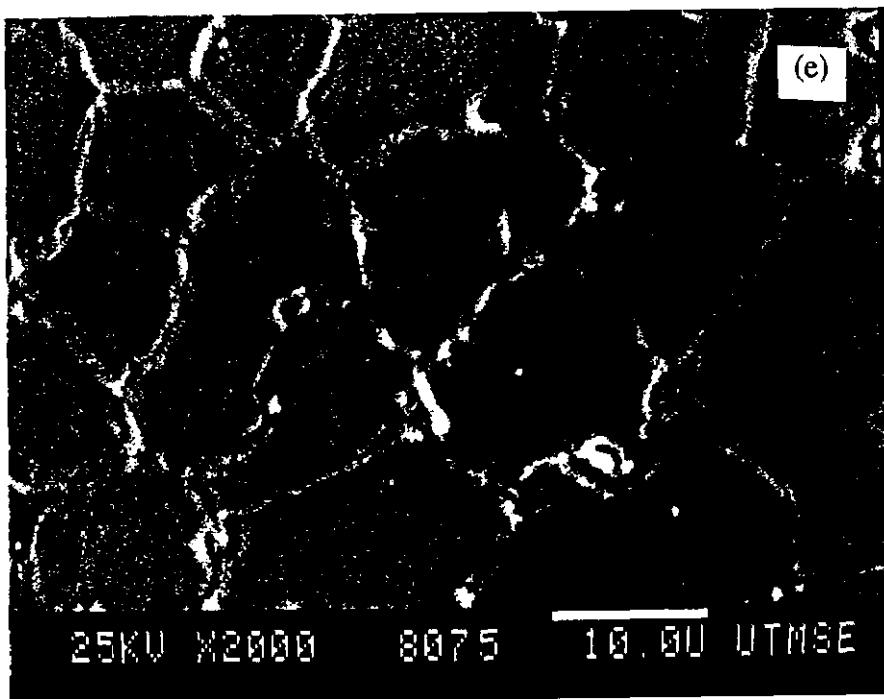
# SEM Microstructures

## $x=0.2, y=0.2$



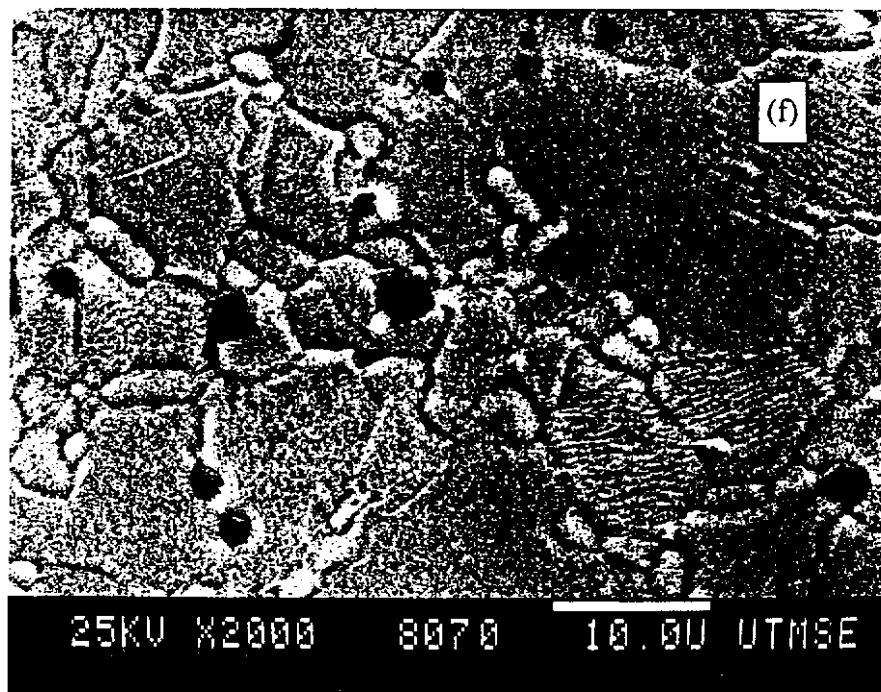
# SEM Microstructures

$x=0.2, y=0.25$



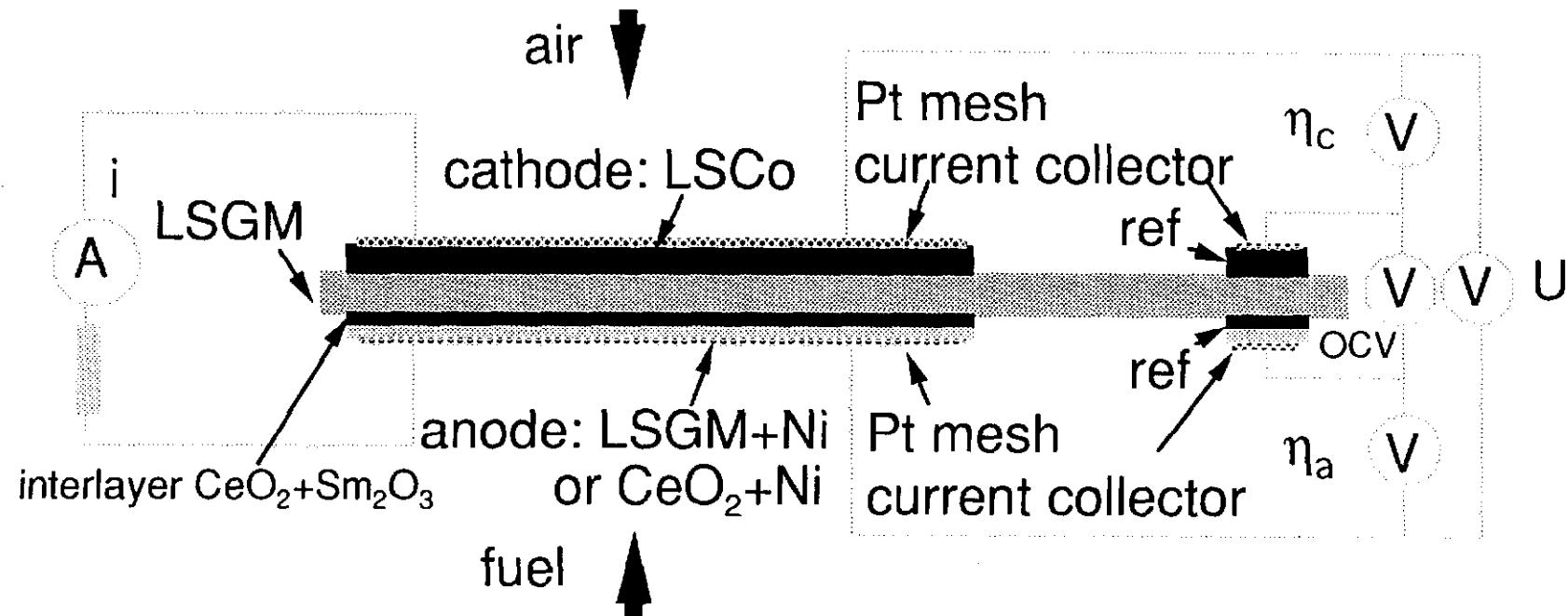
# SEM Microstructures

$x=0.2, y=0.3$

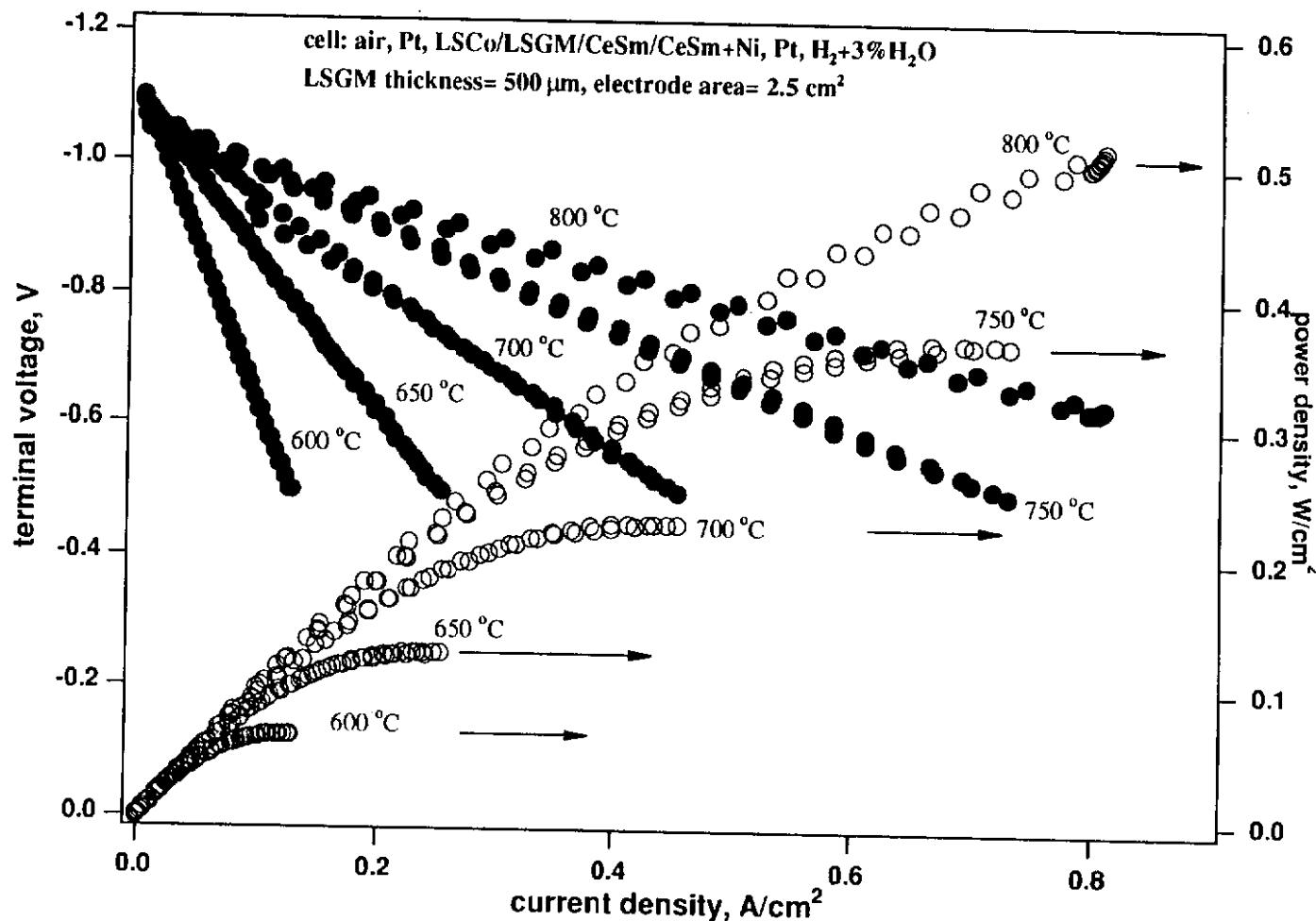


# Single Fuel Cell Configuration

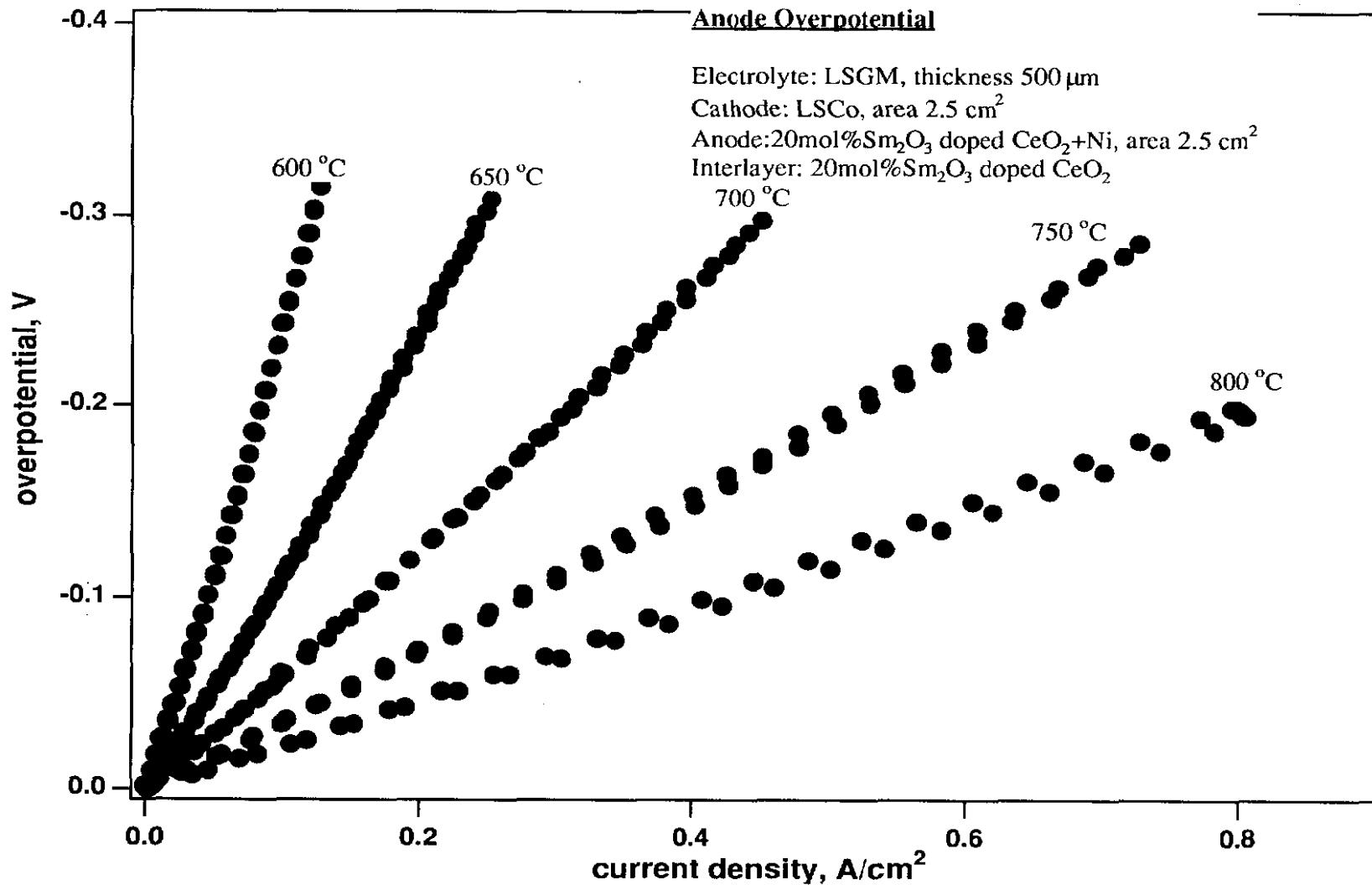
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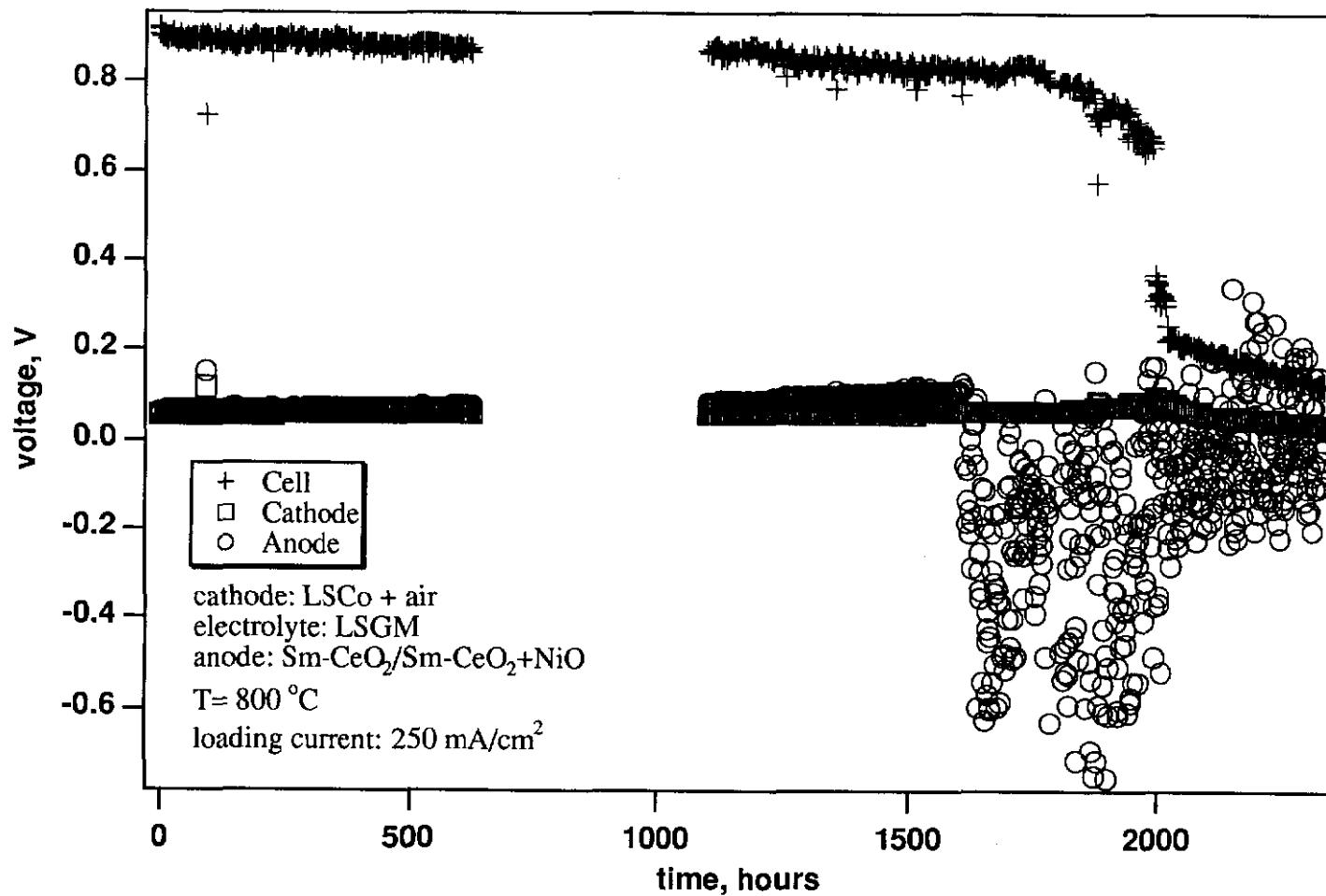
# Performance of Fuel Cell with Buffer-layer



# Anode Overpotential *vs* Current Density



# Preliminary Life Test for 500 $\mu\text{m}$ LSGM Electrolyte



# CONCLUSION

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With the addition of a doped ceria anode buffer layer, the realization of a SOFC based on an LSGM electrolyte operating in the range  $600^{\circ}\text{C} < T_{\text{op}} < 800^{\circ}\text{C}$  is a realistic goal.

# Solid Oxide Fuel Cell Performance Studies: Anode Development

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## Contractor

Board of Curators  
University of Missouri System  
204 Parker Hall  
Rolla, MO 65401

## 1.0 INTRODUCTION

Materials research focused on solid oxide fuel cells (SOFC) is driven by the recognition that processing and operating at lower temperatures would directly address many reliability problems. Hence numerous research groups around the globe are in pursuit of alternate materials for all four SOFC components with higher conductivities at lower temperatures, mixed-conducting cathodes, novel synthesis techniques (for powders and thin films), controlled and stable microstructures, and chemical, mechanical, and electrical stability under the temperature/time/atmosphere conditions of cofiring and operation.

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 overall aim is to fabricate SOFC's with controlled microstructures utilizing  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  (LSM), yttria stabilized zirconia (YSZ), and Ni-YSZ composites as the cathode, electrolyte, and anode, respectively. Ideally, the electrode materials would be tailored for an increased reaction rate (grain size  $\leq 1 \mu\text{m}$ ), be stable with time ( $> 10,000 \text{ h}$ ), have a thermal expansion match to YSZ ( $\alpha \sim 11 \times 10^{-6}/^\circ\text{C}$ ), show limited chemical interaction with the electrolyte, and show no degradation in electrical performance. This paper describes anodic studies, including starting powder characteristics, electrical conductivity and overpotential measurements, and resultant microstructures as a function of processing conditions (i.e. powder calcination temperature, and annealing temperature) and composition.

## 2.0 OBJECTIVES

The University of Missouri-Rolla is in the last year of a 5 year research program with two primary objectives: 1) developing  $\text{LaCrO}_3$ -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. The latter is the focus of this paper. 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 develop processing ↔ microstructure ↔ property relations of the anode and its corresponding interfacial reactions.

### 3.0 Experimental Procedure

#### 3.1 Apparatus Construction

The system constructed for measuring cell performance is a Kanthal-heated, three-zone, half-shell furnace (1100°C). The self supporting electrolyte is mounted and sealed to two mullite tubes using a ceramic adhesive/sealant, Aremco 503. This sealant is an alumina-based cement with a working temperature up to 1650°C. For all studies, planar SOFC's were heated at ~ 2°C/min and held at 1000°C until all characterization was completed. Samples could not be thermally cycled due to the difference in thermal expansion, 7.5 and 10.5 ppm/°C for the cement and YSZ electrolyte, respectively. Both mullite tubes were sealed to the ends of an outer silica tube of 63.5 mm (2 1/2") OD by use of aluminum faceplates, silicone O-rings, and a Vacoa seal. Gas inlet and outlet ports were drilled into the aluminum faceplates and sealed using Swagelock connectors. The outer end of the mullite tubes were also sealed using stainless steel faceplates and silicone O-rings. Pt and Ni wires for cathode and anode sensing, respectively, were inserted into the alumina tubes to make electrical connection to the YSZ electrolyte. Baffles made of sali board were placed inside the mullite tubes to help mixing of the gases and also acted as positioners for the alumina thermocouple tubes to ensure proper electrical connection.

Both oxidant and fuel flow rates were controlled using Tylan Model FC-260 mass flow controllers. Air was used as the oxidant in the majority of experiments, with a flow rate of 200 sccm. Pure oxygen was also used in some experiments and was delivered at a rate of 200 sccm. A mixture of forming gas, FG, (90% N<sub>2</sub> - 10% H<sub>2</sub>) and CO<sub>2</sub> was used as the fuel and had corresponding flow rates of 200 and 2 sccm, respectively. The FG-CO<sub>2</sub> mixture gave Nernst potentials of 1.00 and 1.05 V with air and O<sub>2</sub>, respectively. The FG-CO<sub>2</sub> mixture had a pO<sub>2</sub> equal to 10<sup>-16.5</sup> at 1000°C. Nitrogen was delivered into the silica to sweep away any oxidant or fuel leaking out of the active region of the cell. In all experiments air was introduced in the top tube, fuel to the bottom tube, and the sweep gas was fed to the bottom and exited the top.

#### 3.2 Materials Selection, Preparation and Characterization

The purpose of this investigation was to study the influence of microstructure and composition on the performance of anodes in single SOFCs. Commercially available Y stabilized ZrO<sub>2</sub> (YSZ) powders were used in this study for the electrolyte and as a major constituent in the anode. Anode [YSZ-Ni<sub>1-x</sub>Mg<sub>x</sub>O (x = 0.0, 0.1)], compositions were synthesized by the glycine nitrate method.

The commercially-available Y-doped ZrO<sub>2</sub> (Zirconia Sales of America Inc.) is a fully stabilized (8 mole % Y<sub>2</sub>O<sub>3</sub>), co-precipitated powder. This particular powder was chosen because of its low cost, ~\$70/kg, low impurity content and low densification temperature, ~1400°C. The YSZ powder had a primary particle size of approximately 250 nm and a corresponding BET surface area of ~ 8.0 m<sup>2</sup>/g.

NiO, and Mg-doped NiO were synthesized using the glycine nitrate method with Ni(NO<sub>3</sub>)<sub>2</sub>·xH<sub>2</sub>O, MgCO<sub>3</sub>, glycine, and distilled water as the starting materials. The starting raw materials were thermogravimetrically standardized to determine the cation concentration. Gentle heating (>100°C) resulted in the evaporation of excess water, yielding a viscous liquid. Further heating cause the precursor solution to ignite and resulted in the formation of a fine powder. The powder was then calcined in MgO crucibles at temperatures ranging from 700°C - 1000°C for 4 h.

The resultant powder was vibratory milled dry for 4 h with  $\text{ZrO}_2$  media. Powder crystallinity, phase, and surface area were characterized using X-ray diffraction and BET techniques as a function of calcination temperature.

A YSZ-NiO (45 vol % Ni) composition was also prepared in which both components were combusted simultaneously using the glycine nitrate process. Starting raw materials were  $\text{Ni}(\text{NO}_3)_2 \cdot x\text{H}_2\text{O}$ ,  $\text{Y}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$ , and a zirconium citrate complex. Resultant powders were calcined and milled and the crystallinity and phase were characterized using X-ray diffraction as a function of calcination temperature.

A total of three different techniques were used to prepare powder mixtures with the goal of producing different resultant microstructures including: 1) mixing YSZ with  $\text{Ni}_{1-x}\text{Mg}_x\text{O}$  ( $x=0.0, 0.1, 0.2$ ) 2) mixing YSZ with NiO followed by calcination at  $1400^\circ\text{C}$  for 4 h and 3) simultaneous combustion of both components using the glycine nitrate technique.

### 3.3 Single Cell Fabrication

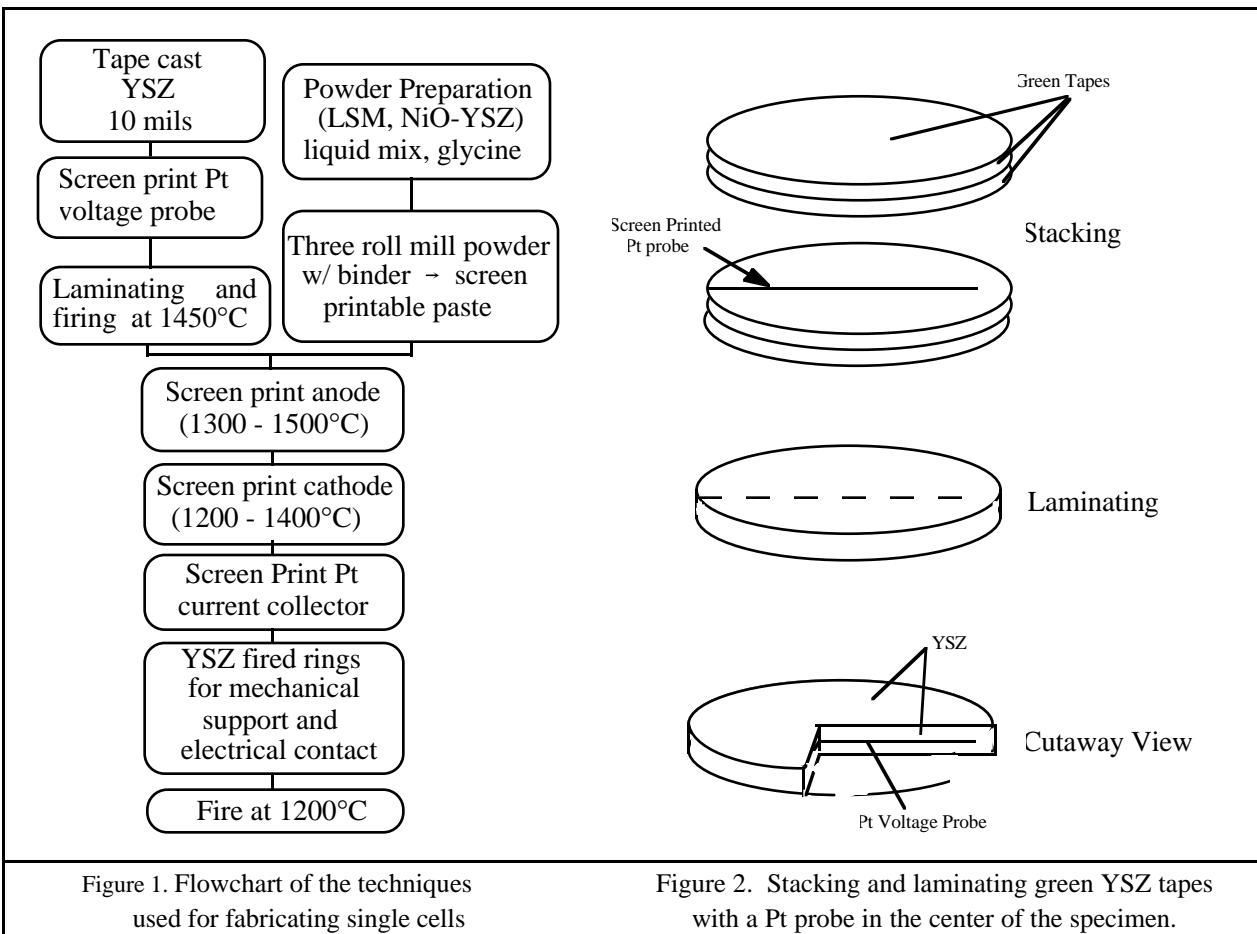
The YSZ electrolyte used in this investigation was self supporting ( $\sim 200 \mu\text{m}$  thick) and the cathode and anode were applied via screen printing onto a pre-sintered dense electrolyte. A flowchart describing the techniques used to fabricate single cells is shown in Figure 1.

#### Electrolyte fabrication

YSZ powders were initially dried at  $150^\circ\text{C}$  to remove any physically bonded water and then mixed with a commercially available binder system from Ferro Corp, B73210. This pre-mixed solution contains binder, plasticizer, solvents, and dispersants with a solids content of 18 wt%. The binder, and solvents are polyvinyl butyral, and ethanol and toluene, respectively. The proportion of YSZ to B73210 by weight was approximately 50/50. A glass release agent, M1111, and a surfactant, M1135, were added to the slip to facilitate removal of the dried tape from the glass and to improve the dispersion behavior of the YSZ powder, respectively. The glass release agent and surfactant were added at a ratio of 1 and 2 wt % of the total organic content, B73210. The slip was ball milled with  $\text{ZrO}_2$  media for 24 - 48 h until the powder was well dispersed. The rheological behavior of the YSZ slip was measured using a HAAKE Viscotester Model VT 500.

The slurry was tape cast using the doctor blade technique at a take-off height of  $\sim 250 \mu\text{m}$  and the resultant dried tapes were  $\sim 50\text{-}75 \mu\text{m}$ . The tape was dried for at least 24 h prior to use to ensure removal of all volatile species. The final proportion of powder to binder (total vol % solids in the tape) was approximately 50/50. Dried tapes were inspected with a light table to discard any defective regions. Circular samples of 2 inch diameter were cut out of the tape and a Pt voltage probe (38 mm long x 0.3 mm wide) was screen printed onto the YSZ tape. The Pt, E1170 (Ferro Corp.), is a fritless, screen-printable paste with a solids content of 70 wt %. A stainless steel 325 mesh screen with an emulsion thickness of 0.1 mil was used. The Pt probe was screen printed to the edge of the YSZ tape for electrical connection. The tape was again dried for at least 24 h prior to use to ensure removal of all volatile species from the Pt paste.

The tapes (six total) were stacked and laminated at 3000 psi for 10 min at  $70^\circ\text{C}$  in such a way as to place the Pt probe in the center of the fired specimen, Figure 2. The thermal processing schedule for binder removal and densification of the YSZ laminates was  $0.5^\circ\text{C}/\text{min}$  to  $350^\circ\text{C}$ , hold for 1 h,  $3^\circ\text{C}/\text{min}$  up to  $1450^\circ\text{C}$ , hold for 2 h, and then cool at  $3^\circ\text{C}/\text{min}$ . Sintered YSZ dimensions were 3.2 cm and  $200 \mu\text{m}$  for the diameter and thickness, respectively. The sintered specimen



underwent a shrinkage of ~25 % (x-y) during densification. The Pt voltage probe was centered internally 100  $\mu\text{m}$  from either surface, and was ~ 5  $\mu\text{m}$  thick and 200  $\mu\text{m}$  wide.

### Anode deposition

The anode powders were mixed with a commercial resin solution, BX018-16, from Ferro Corp. The suspension was mixed using a three roll mill to prepare a well-dispersed paste for screen printing. The wt% loading of powder to binder varied depending on the calcination temperature and resultant surface area of the powder and was therefore performed on a trial and error basis. In all instances the highest amount of powder was loaded into the binder such that the paste was still workable.

Anode compositions were screen printed onto dense YSZ electrolytes and sintered at various temperatures. Stainless steel screens of 165 mesh, 3 mil emulsion thickness were used. Anode compositions were sintered onto the electrolyte between 1300°C and 1500°C in 100°C increments for a 1 h hold, with a heating and cooling rates of 3°C/min. A primary goal of this investigation was to vary the grain size and porosity of electrode microstructures and the their impact on electrode performance, therefore powders were calcined and sintered at various temperatures. Anodes were porous, exhibited grain sizes on the order of 1  $\mu\text{m}$ , and gave resultant dimensions of 0.635cm x 0.635cm and ~ 20  $\mu\text{m}$  thick. Fracture surfaces were characterized by a JEOL Scanning

Electron Microscope to examine resultant microstructures. All specimens were sputtered with Pd/Au before analysis.

#### Mechanical Support and Electrical Connection

A Pt grid (0.2 mm line width and 0.2 mm spacing between lines) was screen printed on the electrodes for cell performance experiments to act as a current collector but also to allow gas diffusion to the electrode/electrolyte interface.

Tape cast and fired YSZ rings ( $\sim 350 \mu\text{m}$  thick) were bonded to both sides of the electrolyte using a ceramic adhesive, Aremco 516, Figure 3. The adhesive is a zirconia based cement which allowed thermal cycling without delamination of the rings from the electrolytes. The rings acted as both mechanical support for the electrolyte and for electrical connection. The mullite tubes used for atmosphere control were cemented to the rings rather than the electrolyte so that no adhesive was in the vicinity of the active region of the cell. The rings were designed with pads which were coated with Pt paste to allow for electrical sensing. Pt wires of 10 mil diameter were bonded from the Pt grid to the Pt pads on the rings using Pt paste, this was done to both the anode and cathode side of the electrolyte. Pt paste was also used to paint a conductive strip from the Pt voltage probe to a pad on the ring for voltage sensing. There are two pads on the cathode side, one for electrical connection to the cathode and one for sensing the voltage probe, whereas only one pad was active on the anode side. The cell was sintered at  $1200^\circ\text{C}$  for 1 h to densify the Pt and achieve a good bond between the YSZ rings and electrolyte.

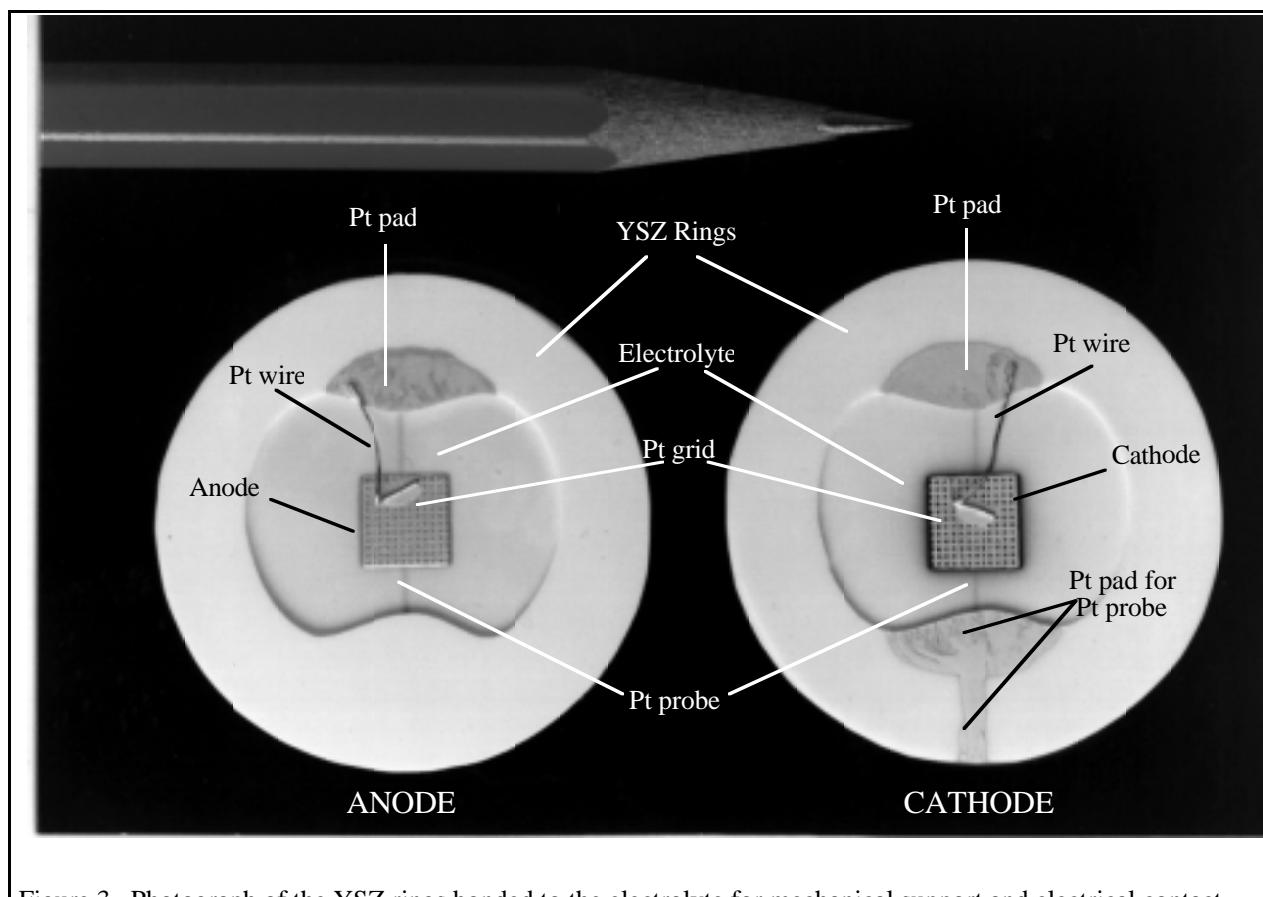


Figure 3. Photograph of the YSZ rings bonded to the electrolyte for mechanical support and electrical contact .

### 3.4 Electrical Characterization

Electrical characterization of single cells utilizing the internal Pt voltage probe was investigated to simultaneously separate the losses attributed to each component (anode, electrolyte, cathode) and their interfaces (cathode/electrolyte and anode/electrolyte) during cell operation. Cell performance studies were focused on the reaction kinetics at the interfaces whereas DC conductivity measurements were performed to investigate the resistive losses of each component as a function of time, composition, and preparation condition.

#### Cell Performance Studies

Electrochemical (I-V) measurements were carried out using a five electrode configuration which allowed for separation of anode and cathode overpotentials during operation, Figure 4. Separate leads were used to carry the current and voltage of the cell to remove the loss associated with the lead wires and allow for a direct examination of the losses attributed to the cell components. The third voltage lead was connected to the voltage probe and was used to monitor the voltage drop between the probe and corresponding electrodes during operation. Pt wires were used on the cathode side, four 20 mil wires for the current lead, and 10 mil wires for both the voltage leads, cathode and internal Pt probe. Ni wires were used on the anode side, two 20 mil wires for the current lead and a 10 mil wire for the voltage lead. Both voltage leads for the cathode and anode were designed to mimic a spring for contact with the Pt grids.

I-V behavior and AC impedance spectroscopy were performed on both half cells and complete cells. The I-V behavior was measured using an Antronics Current/Voltage Control Fuel Cell Testing Module, a Keithley Model 196 Microvolt Meter, and a Fluke 27/FM Multimeter. The Fuel Cell Testing Module was placed in the voltage control mode thus enabling the desired cell voltage and corresponding current to be measured.

After the cell was heated to 1000°C at ~ 2°C/min, air was first fed to the cathode and then the fuel was introduced to the anode. The fuel was delivered to the anode for at least thirty minutes

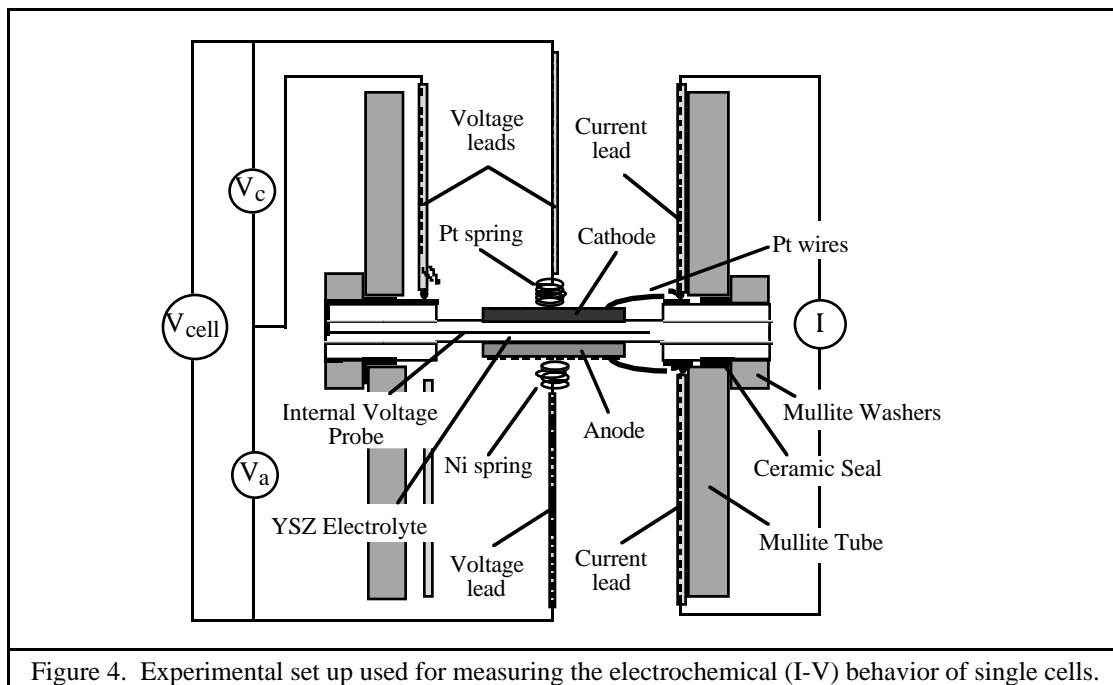


Figure 4. Experimental set up used for measuring the electrochemical (I-V) behavior of single cells.

before electrical contact was made to ensure that the anode (YSZ-NiO) was reduced to the cermet (YSZ-Ni). It could also be determined at this time if the electrolyte was cracked or the sealant had failed. The resistance between the voltage and current leads of each corresponding electrode was measured before testing to ensure that each lead was still connected to the Pt grid and thus the electrode.

After all electrical connections were made the first test was to measure the open circuit, Nernst potential between the anode and cathode. The Nernst potential is very sensitive to the chemical potential gradient and hence reveals whether a small hairline fracture had occurred. It was often difficult to tell this by monitoring the exit ball flow meters and were thus a better test for fractures or leaks. Measurements were carried out from small voltages to larger ones in increments of 25mV and in all cases steady state voltages and currents were measured. Stabilization times were on the order of 2-3 min. The current, total cell voltage ( $V_{cell}$ ), voltage drop from the Pt probe to the cathode ( $V_c$ ) and the anode ( $V_a$ ) were all simultaneously measured. At open circuit and at any given voltage under load the total cell voltage was equal to the two half cell voltages.

## 4.0 RESULTS AND DISCUSSION

The primary focus of this research was to reduce the sintering of the Ni particles, increasing the number of reaction sites, which would result in a lower overpotential. Electrical conductivity and  $\eta$ - $j$  plots were monitored for a 24 h period to better understand the coarsening between Ni particles. Conductivity experiments were used to reveal how the Ni particles were distributed throughout the cermet. Since the conductivity is controlled by the Ni, these experiments help to determine if the YSZ support structure was sufficient in reducing the tendency of the Ni particles to sinter. Electrochemical measurements,  $\eta$ - $j$ , were not only used to determine resultant overpotentials, but also how the Ni-YSZ particle contacts were distributed in the composite.

### 4.1 Resistive (IR) Contribution from the Anode

Four point DC conductivity experiments were performed on anode compositions to determine the resistive loss associated with the anode during cell operation. Anode conductivities ranged from  $\sim 3$  -  $800$  S/cm which depended on the starting raw materials, the fabrication technique, and the sintering temperature. Figure 5 is a typical V-I plot of an anode demonstrating ohmic behavior. Knowing the dimensions of the anode as shown previously for the case of the cathode, area =  $0.4$  cm $^2$  and thickness =  $\sim 20$   $\mu$ m, the resistance was calculated. In the worst case scenario,  $\sigma = 3$  S/cm, the resistance was  $1.7$  m $\Omega$  resulting in a voltage drop of  $0.7$  mV at a current density of  $1000$  mA/cm $^2$ . Similar to the case for the cathode, the ohmic portion of the cathode is very small and cannot be resolved from the total measured voltage. Therefore, the measured voltage as a function of current density between the anode and the Pt voltage probe can be attributed to the resistive loss with the electrolyte ( $100$   $\mu$ m thick) and the overpotential of the anode.

### 4.2 Effect of Vol % Ni

Compositions studied for this experiment ranged from 40-55 vol % Ni in increments of 5 vol %. All samples were prepared from commercially available YSZ and NiO powders synthesized by the glycine nitrate process followed by calcined at  $900^\circ$ C. All samples were fabricated in the form of oxides, sintered on the YSZ electrolyte at  $1400^\circ$ C, and then reduced in situ. After the fuel was introduced to the anode, the cell was allowed to stabilize for  $\sim 1$ - $3$  h before electrochemical

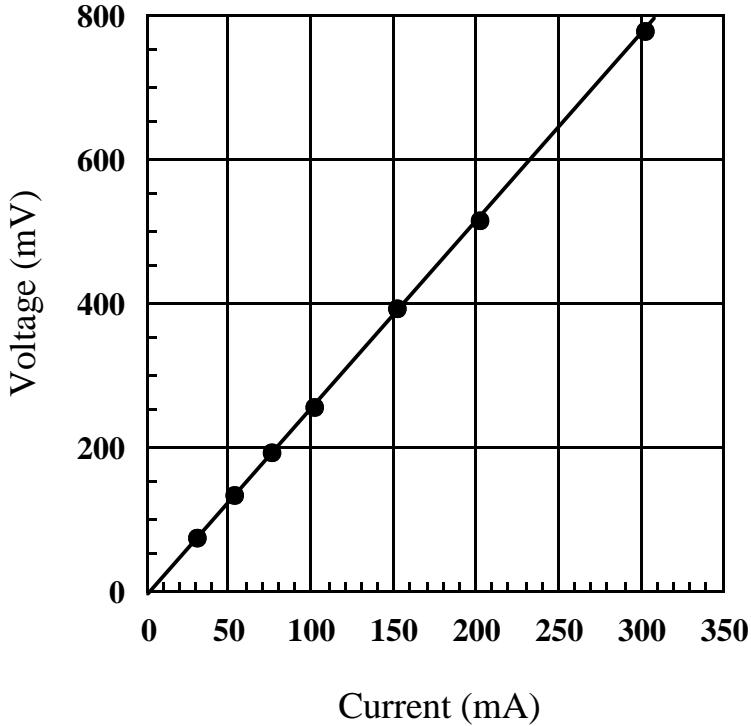


Figure 5 Voltage-current behavior for a typical Ni-YSZ cermet.

measurements were performed. Conductivity experiments were monitored in air, during reduction of NiO to Ni, and under reducing conditions for 24 h.

The electrochemical response of the four compositions is shown in Figure 6. The 40 and 45 vol % Ni samples show similar behavior,  $\sim 220$  mV and  $200$  mV at  $1000$  mA/cm $^2$ , but the 50 and 55 vol % Ni compositions showed much higher overpotentials,  $\sim 280$  and  $370$  mV at  $1000$  mA/cm $^2$ . After 24 h, the overpotentials of all compositions increased, Figure 7. The 40 and 45 vol % Ni samples still show similar behavior,  $\sim 270$  mV at  $1000$  mA/cm $^2$ , and the 50 and 55 vol % Ni compositions had overpotentials of  $\sim 370$  and  $470$  mV at  $1000$  mA/cm $^2$ . The low vol % Ni samples, 40 and 45, have the lowest overpotentials due to the larger YSZ content in the cermet to support the Ni particles. This would effectively reduce the amount of sintering between Ni particles. For compositions with larger vol % Ni (i.e. 55 vol % Ni), less YSZ is available to support the Ni and larger Ni particles would be expected. The resultant microstructures for the four compositions after 24 h of operation are shown in Figures 8 and 9. It is difficult to see any distinguishable difference for the three lowest Ni contents, 40, 45, and 50 %, although, the 55 % composition does show a dissimilarity. The Ni particles are larger and more easily distinguishable from the YSZ support.

The two end members, pure YSZ and NiO, were also examined to demonstrate the extent that the pure Ni particles coarsen and to better understand the Ni-YSZ microstructures. The YSZ and NiO samples were prepared as pastes, screen printed, sintered on the YSZ support at  $1400^\circ\text{C}$ , and reduced at  $1000^\circ\text{C}$  for 24 h. The results are shown in Figure 10. The Ni particles have sintered extensively with grain sizes of  $\sim 5\mu\text{m}$ , and the YSZ has formed a rigid structure for supporting the Ni particles. Comparing Figure 10 with Figures 8 and 9, it has been illustrated the tremendous importance of adding YSZ to the Ni to reduce the sintering between Ni particles. To further demonstrate the large driving force for the Ni particles to densify, the same NiO powder was compacted into a pellet and placed into a furnace at  $1000^\circ\text{C}$  under reducing conditions for 8 hours.

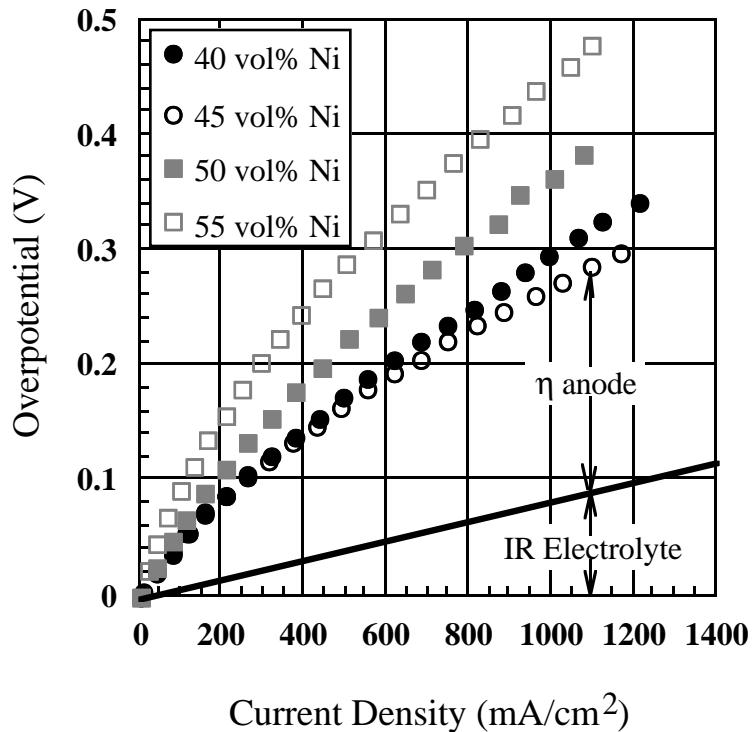


Figure 6. Initial  $\eta$ -j relations of Ni-YSZ cermets with various Ni contents sintered at 1400°C.

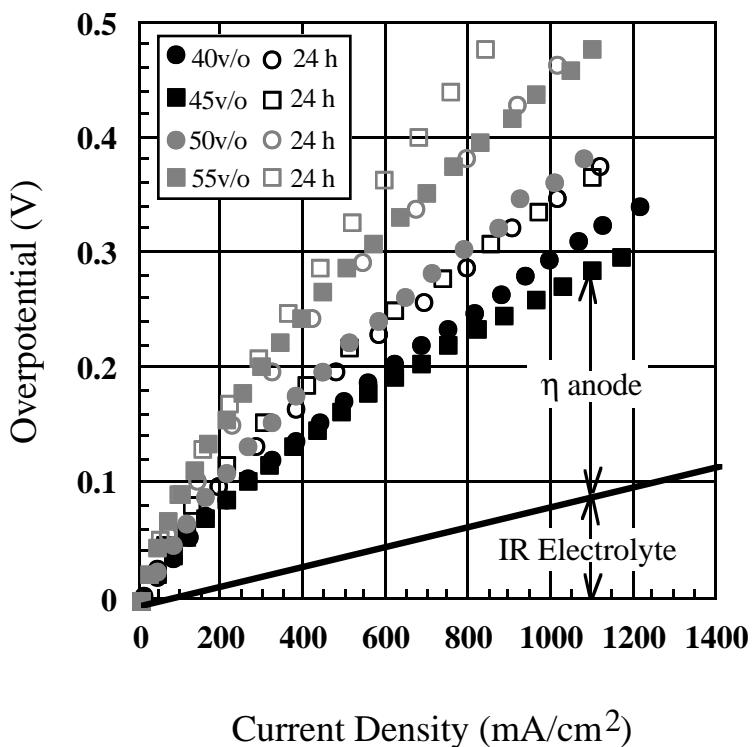


Figure 7.  $\eta$ -j results, initially and after 24 h, for Ni-YSZ cermets with various Ni contents sintered at 1400°C.

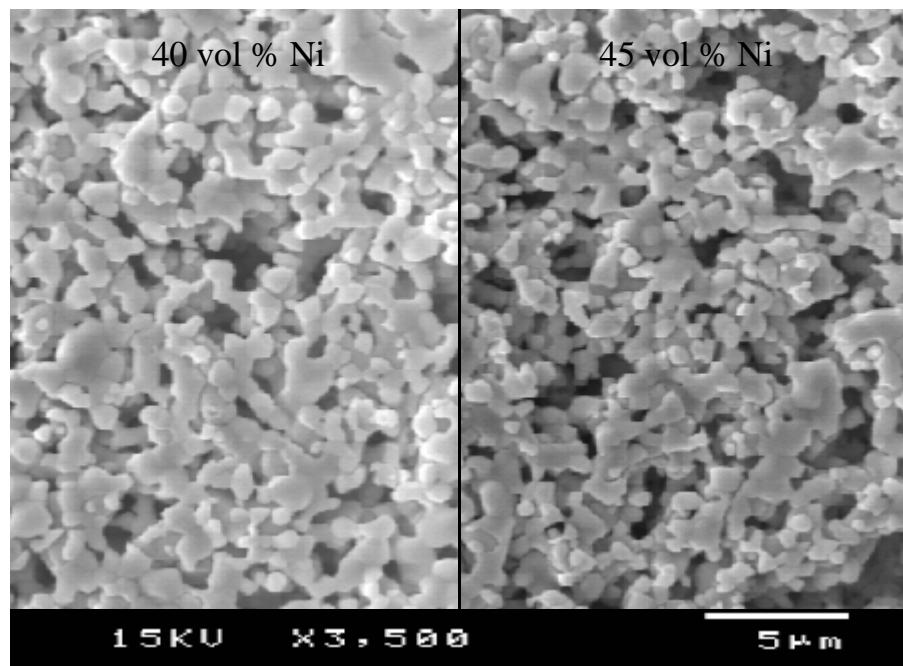


Figure 8. Microstructures of 40 and 45 vol % Ni compositions sintered on the YSZ electrolyte at 1400°C.

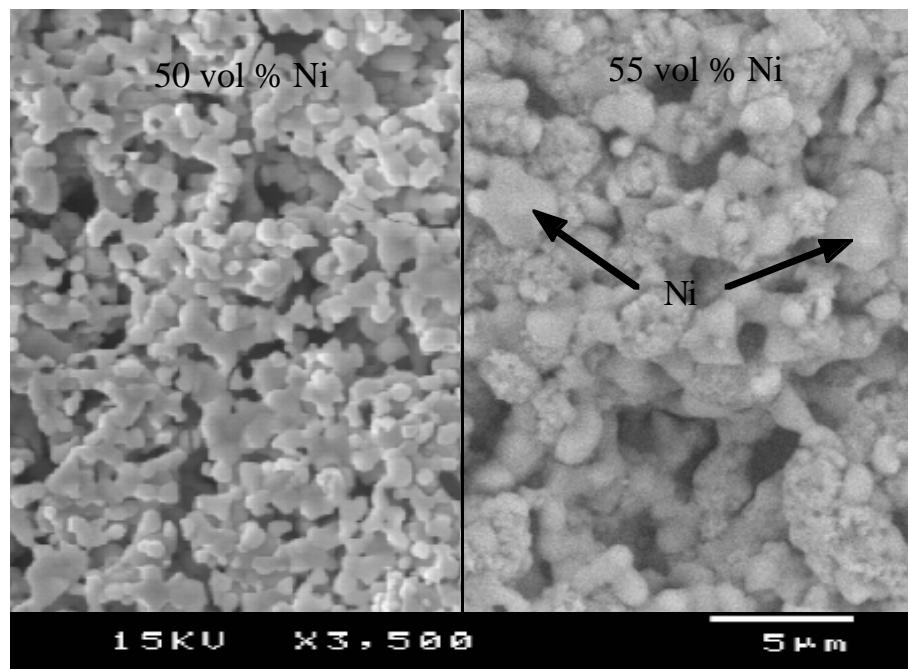


Figure 9. Microstructures of 50 and 55 vol % Ni compositions sintered on the YSZ electrolyte at 1400°C.

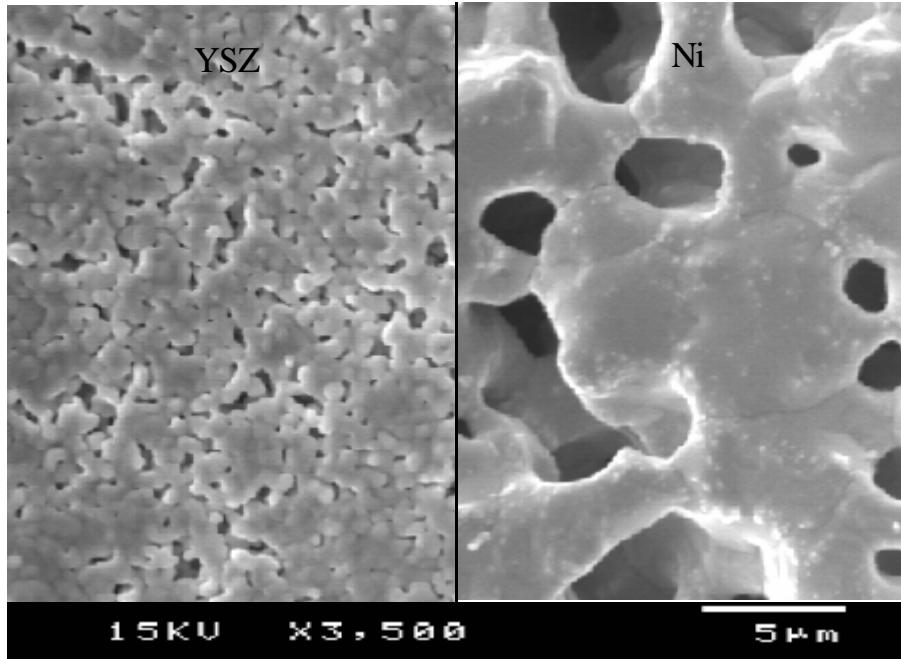


Figure 10. Microstructures of YSZ and Ni sintered on the YSZ electrolyte at 1400°C.

The sample was cooled to room temperature under fuel, and the density was measured. Density measurements revealed the specimen to have a density  $\sim 91\%$  of the theoretical,  $\rho_{Ni} = 8.9\text{g/cc}$ .

Electrical conductivity measurements were also performed on all four compositions for 24 h, Figure 11. Since the samples contain various vol % Ni, a direct comparison cannot be made as to how the Ni particles are distributed within the cermet, although, similar trends were observed. All compositions had similar conductivities in the oxide form,  $\sim 3\text{-}5\text{ S/cm}$ . Upon exposure to the fuel, a large increase in the conductivity occurred, within 5 minutes, then rapidly decreased within  $\sim 3$  hours to a steady state value. The large increase in the conductivity is caused by the reduction of  $\text{NiO}$  to  $\text{Ni}$  metal. The very sharp decrease in conductivity is due to the rapid sintering between Ni particles, and the continued slow decrease in the conductivity (after  $\sim 3$  h) can be attributed to further sintering of the Ni particles.

#### 4.3 Effect of $\text{NiO}$ Starting Raw Materials

Three different  $\text{NiO}$  sources were used in this study to examine the effect of Ni particle size and preparation conditions on the anodic overpotential. All anodes contained 45 vol % Ni, and were fired onto the YSZ electrolyte at 1400°C. The first  $\text{NiO}$  powder was synthesized by the glycine nitrate process and calcined at 900°C as described previously. The remaining two sources were commercially available powders with different primary particle sizes. The first powder had a reported particle size of -325 mesh ( $< 45\text{ }\mu\text{m}$ ) and the second powder was spray dried with a primary particle size less than  $10\text{ }\mu\text{m}$ .

The  $\eta$ - $j$  plots for the three different particle sizes initially and after 24 h are shown in Figure 12. The results of the powder prepared by the glycine nitrate process are the same as reported previously, Figure 7. The -325 mesh powder initially has an overpotential of  $\sim 250\text{ mV}$  at  $600\text{ mA/cm}^2$  but increases to  $\sim 370\text{ mV}$  after 24 h. The spray dried powder has a relatively stable

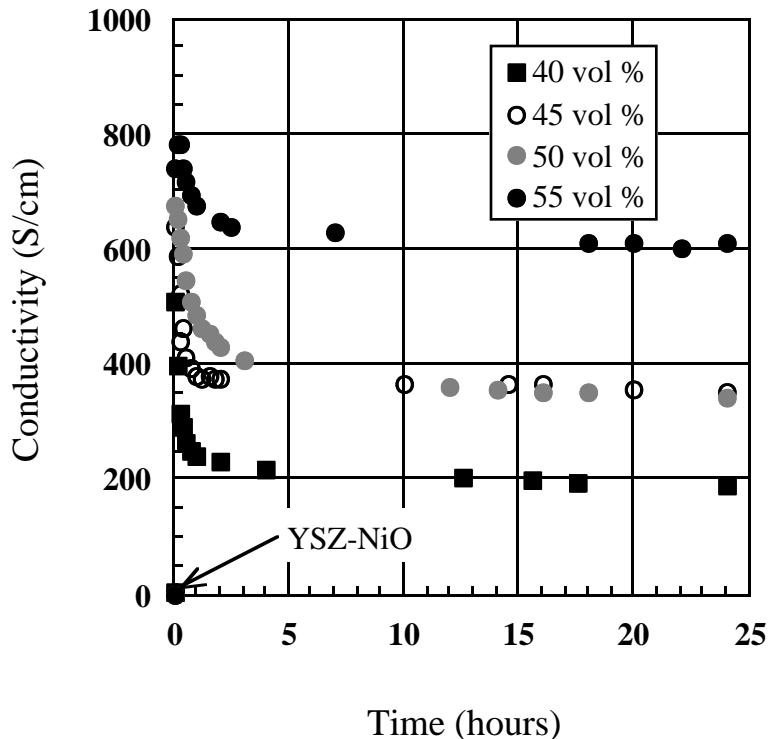


Figure 11. Conductivity versus time for Ni-YSZ cermets prepared with various Ni contents and sintered at 1400°C.

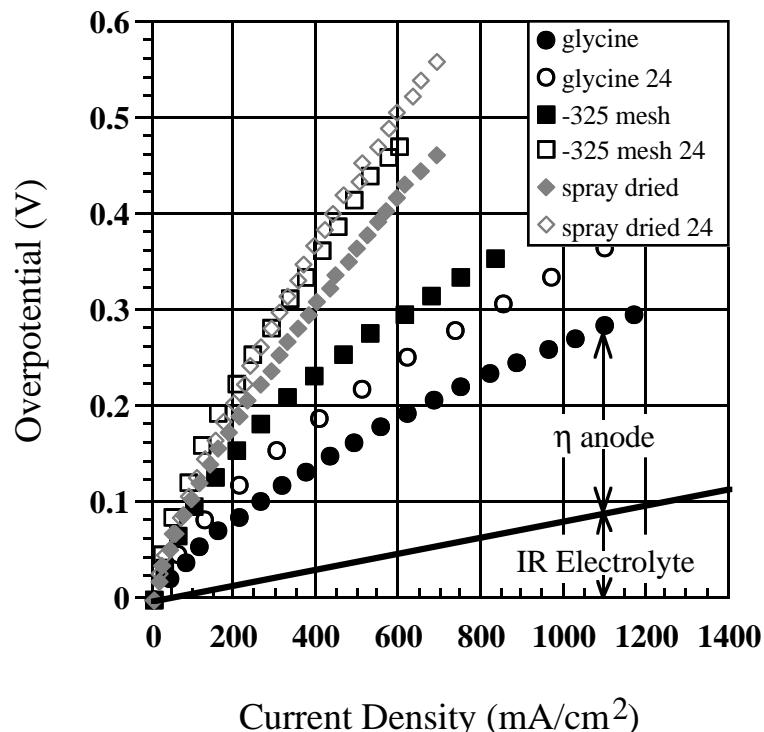


Figure 12.  $\eta$ - $j$  relations of 45 vol % Ni compositions prepared with different NiO sources and sintered at 1400°C.

overpotential but is extremely high,  $\sim 450$  mV at  $600$  mA/cm $^2$ . Examination of the microstructures after 24 h of operation, Figures 13 and 14, reveals large, distinguishable Ni particles for both commercial powders. This can explain the large overpotentials in that the number of reaction sites (Ni-YSZ contacts) has drastically decreased due to the large Ni particle size. The microstructure of the spray dried powder resembles spray dried granules which eventually reduce to Ni spheres.

The conductivity for the three powders also correlates well with the electrochemical results as shown in Figure 15. The conductivity of the spray dried powder was relatively stable upon reduction but was extremely low, comparable to the NiO-YSZ composite ( $\sim 3$  S/cm). This suggests that the Ni particles have rapidly become large and separated from one another decreasing the number of Ni-Ni contacts. The -325 mesh powder initially had a higher conductivity ( $\sim 200$  S/cm) but quickly decrease to values comparable to the spray dried powder. This also suggest that the Ni particles have coarsened and reduced the number of Ni-Ni contacts throughout the cermet. For both commercial powders, the Ni-Ni and Ni-YSZ particle contacts has decreased causing the conductivity to be extremely low and the overpotential to be high.

#### *4.4 Effect of Pre-calcination of Powders*

The influence of calcining NiO and YSZ together at high temperatures ( $1400^\circ\text{C}$ ) before depositing the composition onto the YSZ was next investigated. This approach was examined to provide a more stable anode structure by allowing the YSZ and NiO powders to further densify during calcination. The powders were then milled, deposited onto the YSZ electrolyte, and sintered at  $1400^\circ\text{C}$ . The composition studied was 50 vol % Ni, the  $\eta$ - $j$  results are shown in Figure 16. For comparison, a 50 vol % Ni composition prepared by the conventional technique (described previously with no pre-calcination of NiO and YSZ) is also illustrated. Both the precalcined and conventionally prepared samples have the same starting raw materials, commercial YSZ and NiO prepared by the glycine nitrate process. The pre-calcined powders resulted in a lower and more stable overpotential ( $\sim 240$  mV at  $1000$  mA/cm $^2$ ) than the conventionally prepared composition ( $\sim 380$  mV at  $1000$  mA/cm $^2$ ) after 24 h. The lower and more stable overpotential is believed to be caused by a more rigid and stronger YSZ structure due to the high temperature calcination of the YSZ and NiO. The pre-calcination treatment allows more densification to occur between YSZ particles because the anode is normally constrained to sinter by the YSZ electrolyte during annealing. Therefore, the pre-calcination treatment is more effective than the conventional preparation technique in preventing the Ni from coarsening during operation. The pre-calcined and conventionally prepared anode microstructures after 24 h of operation are shown in Figure 17. The precalcined anode resulted in larger grains ( $\sim 1$ - $2$   $\mu\text{m}$ ) and a coarser microstructure than the conventionally prepared anode due to the high temperature calcination treatment.

#### *4.5 Effect of Annealing Temperature on the Anodic Overpotential*

The temperature at which the anode was sintering on the YSZ electrolyte was investigated to increase the stability of the anode over a 24 h period. Two composition prepared by two different techniques were examined. The first composition contained 50 vol % Ni and the powders were pre-calcined at  $1400^\circ\text{C}$ . The second anode was a 45 vol % Ni composition prepared by the conventional technique. Both composition were fired on the electrolyte at  $1300$ ,  $1400$ , and  $1500^\circ\text{C}$ .

##### 4.5.1 Pre-calcined Powders (50 vol % Ni)

The electrochemical behavior of the pre-calcined 50 vol % Ni composition fired on the electrolyte at  $1300$ ,  $1400$ , and  $1500^\circ\text{C}$  is shown in Figure 18. Both the initial results and those after 24 h of operation are illustrated. The  $1500^\circ$  firing shows the best stability and lowest

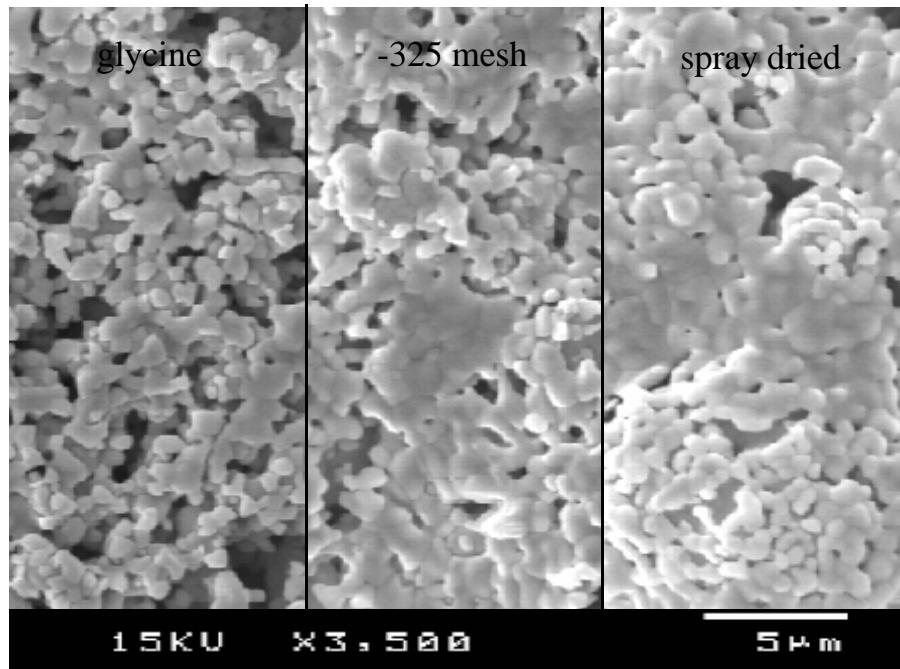


Figure 13. Microstructures of 45 vol% Ni compositions prepared with different NiO sources and sintered on the YSZ electrolyte at 1400°C.

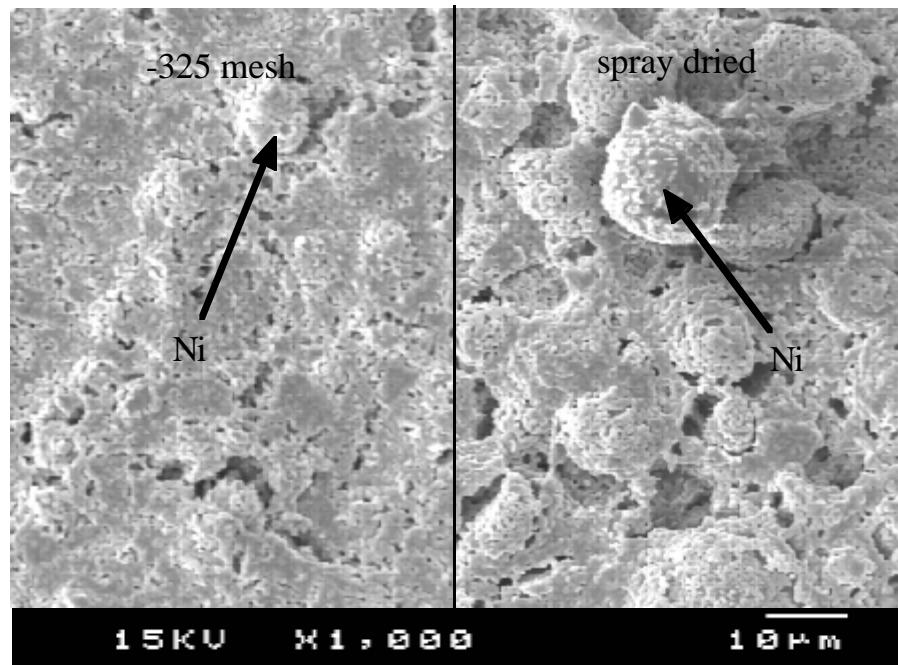


Figure 14. Microstructures of 45 vol% Ni compositions prepared with two different commercial NiO sources and sintered on the YSZ electrolyte at 1400°C.

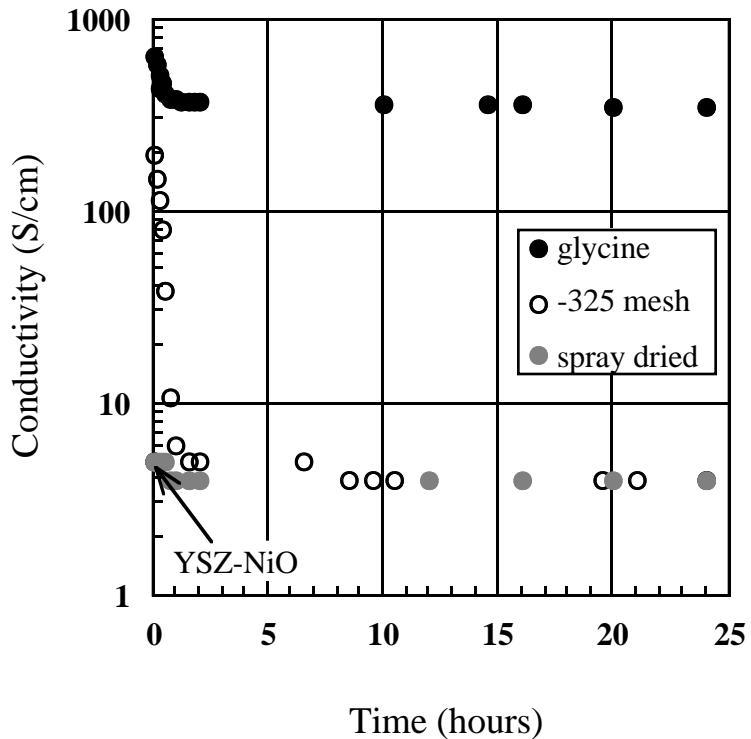


Figure 15. Conductivity versus time of 45 vol % Ni compositions prepared with different NiO sources and sintered at 1400°C.

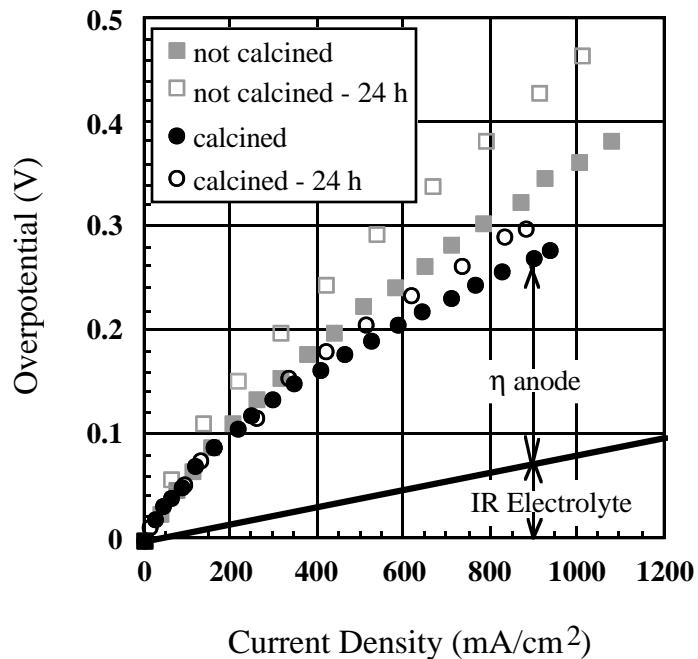


Figure 16.  $\eta$ - $j$  relations of 50 vol % Ni compositions prepared by different techniques, NiO and YSZ calcined together at 1400°C and by the conventional technique, and sintered at 1400°C.

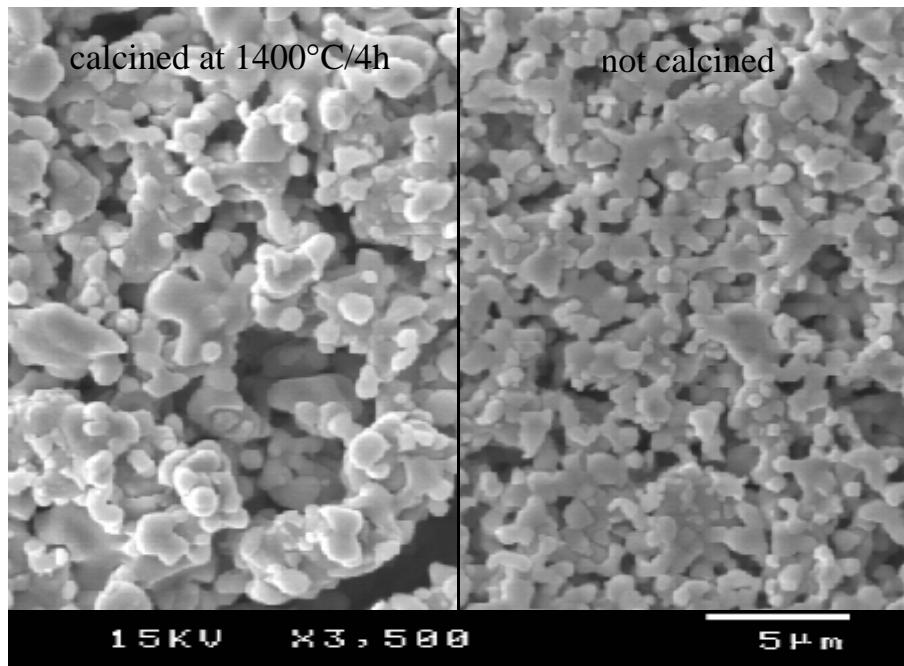


Figure 17. Microstructures of 50 vol % Ni compositions prepared by different techniques, NiO and YSZ calcined together at 1400°C and by the conventional technique, and sintered on the YSZ electrolyte at 1400°C.

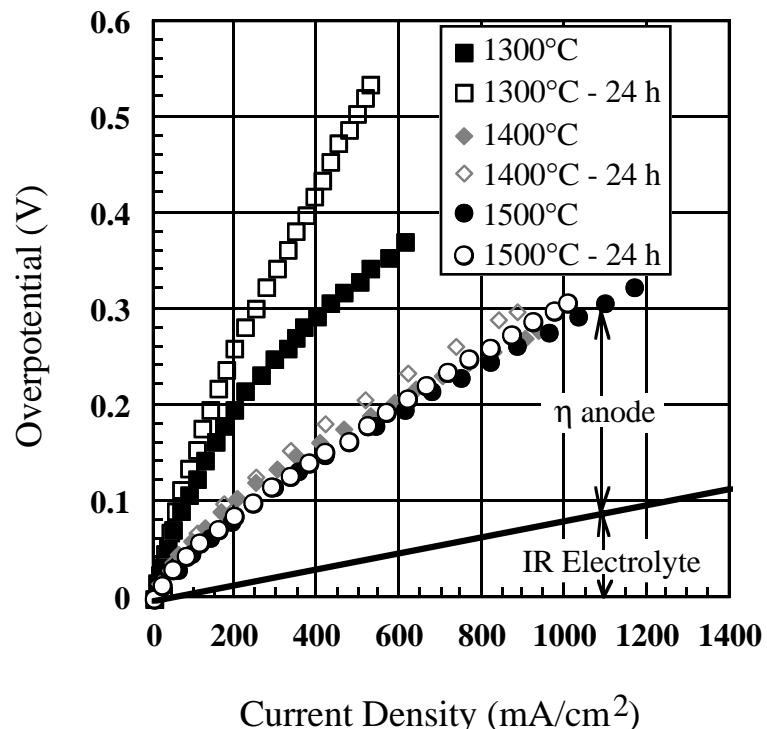


Figure 18.  $\eta$ - $j$  relations of 50 vol % Ni compositions pre-calcined at 1400°C and sintering at various temperatures.

overpotential after 24 h (~ 220 mV at 1000 mA/cm<sup>2</sup>). The anode densified at 1400°C also showed a low overpotential but slightly increased with time (~ 240 mV at 1000 mA/cm<sup>2</sup>). The sample sintered at 1300°C had the highest overpotential and was unstable within the 24 h period (~ 400 mV at 400 mA/cm<sup>2</sup> after 24 h). The improved electrochemical response with higher sintering temperatures is due to the improved sintering of YSZ particles, a more rigid YSZ support will further inhibit the Ni particles from coarsening. The densification of the YSZ support at high temperatures (1300-1500°C) should provide a stable backbone and is not expected to change under fuel cell conditions (pO<sub>2</sub> ~ 10<sup>-17</sup>, 1000°C).

The conductivity and microstructures after 24 h of operation are shown in Figures 19 and 20, respectively. The conductivity results correlate well with the electrochemical response, both suggesting that the sintering between Ni particles has been reduced with higher annealing temperatures. For higher sintering temperature, there should be better Ni-Ni particle contact resulting in more paths for conduction to occur. The microstructures also suggest that the lower firing temperature has a less rigid structure (the grains appear to be smaller and not as well bonded to one another) than the progressively higher annealing temperatures.

#### 4.5.2 Conventional preparation (45 vol % Ni)

The electrochemical behavior of the conventionally prepared 45 vol % Ni composition sintered on the electrolyte at 1300, 1400, and 1500°C is shown in Figure 21. Both the initial and 24 h results are illustrated. The 1500° firing has the best 24 h stability and lowest overpotential (~ 190 mV at 1000 mA/cm<sup>2</sup>). The composition densified at 1400 also showed a low initial overpotential but increased substantially in 24 h (~ 270 mV at 1000 mA/cm<sup>2</sup>). Samples sintered at 1300°C were stable but had high overpotentials (~ 300 mV at 1000 mA/cm<sup>2</sup>). The improved electrochemical response with higher sintering temperatures can also be attributed to the improved densification between YSZ particles as described in the previous section.

The conductivity and microstructures after 24 h of operation are shown in Figures 22 and 23. The conductivity results also correlate well with the electrochemical response, both suggesting that the sintering between Ni particles has been reduced with higher densification temperatures. This results in better Ni-Ni particle contact throughout the cermet, and therefore more paths for conduction. The microstructures also suggest that the higher annealing temperature has a more rigid structure (the grains are larger, ~ 1-2 µm, and bonded well to one another) than the progressively lower annealing temperatures.

Higher annealing temperatures (1500°C) have demonstrated that an improved stability, lower overpotentials, and higher conductivities can be achieved for both the pre-calcined and conventional prepared anodes. A direct comparison cannot be made between the two techniques because the anodes contain different Ni contents. Although, the above results do suggest that a lower vol % Ni (40-45) for the pre-calcined technique should be explored. This could reduce the overpotential and improve the stability at lower annealing temperatures (1400°C).

## 5.0 MAJOR ACHIEVEMENTS DURING THE PAST YEAR

### Anode Studies:

- ◆ YSZ anodes prepared with lower Ni volume fractions (40 & 45%) resulted in lower overpotentials and improved stability.
- ◆ Higher sintering temperatures effectively lowered the overpotential and increased the in-plane conductivity. Due to constrained sintering between the anode and the YSZ electrolyte, higher

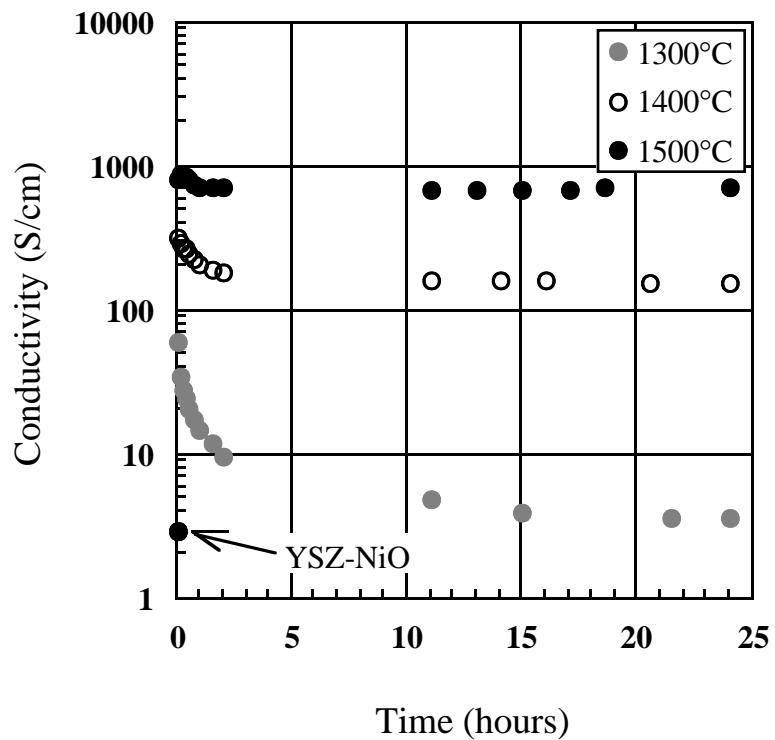


Figure 19. Conductivity versus time for 50 vol % Ni compositions pre-calcined at 1400°C and sintered at various temperatures.

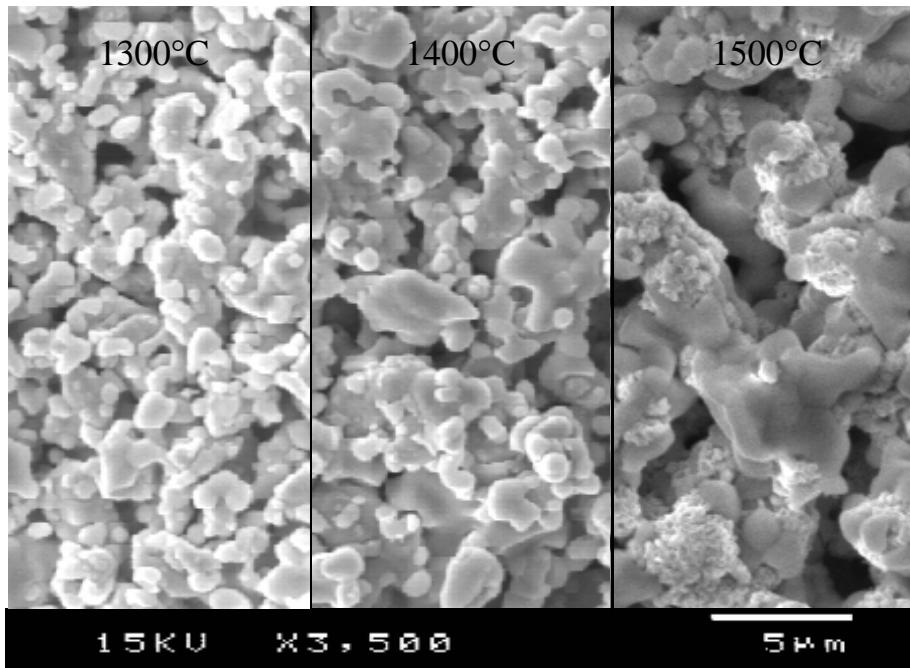


Figure 20. Microstructures of 50 vol % Ni compositions pre-calcined at 1400°C and sintered on the YSZ electrolyte at various temperatures.

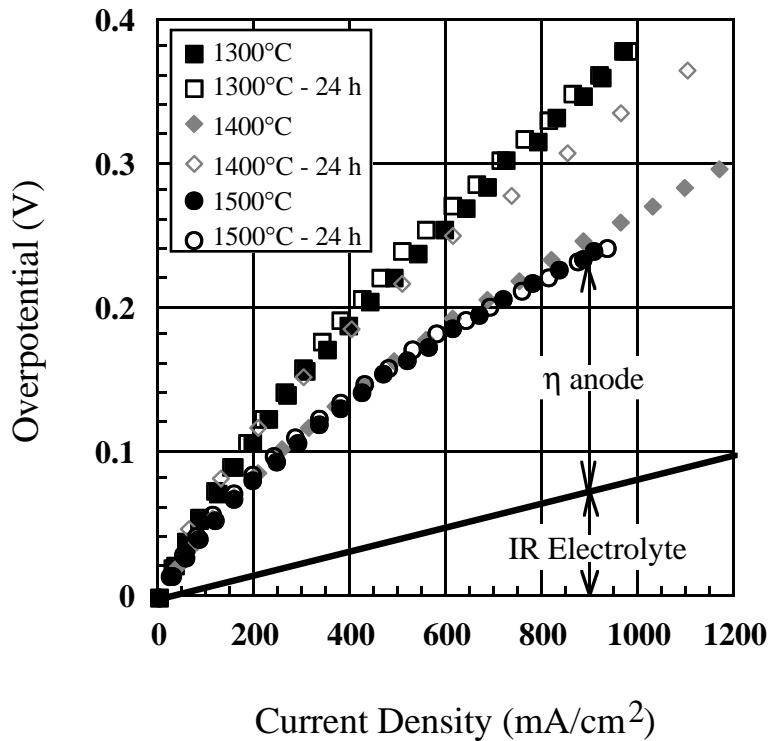


Figure 21:  $\eta$ -j relations of 45 vol % Ni compositions sintered at various temperatures.

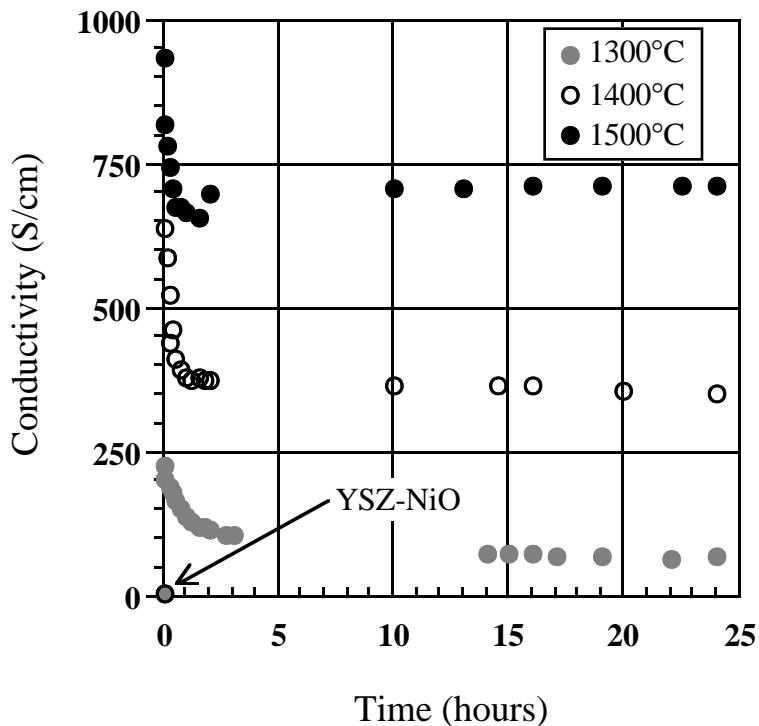


Figure 22: Conductivity versus time of 45 vol % Ni compositions sintered at various temperatures.

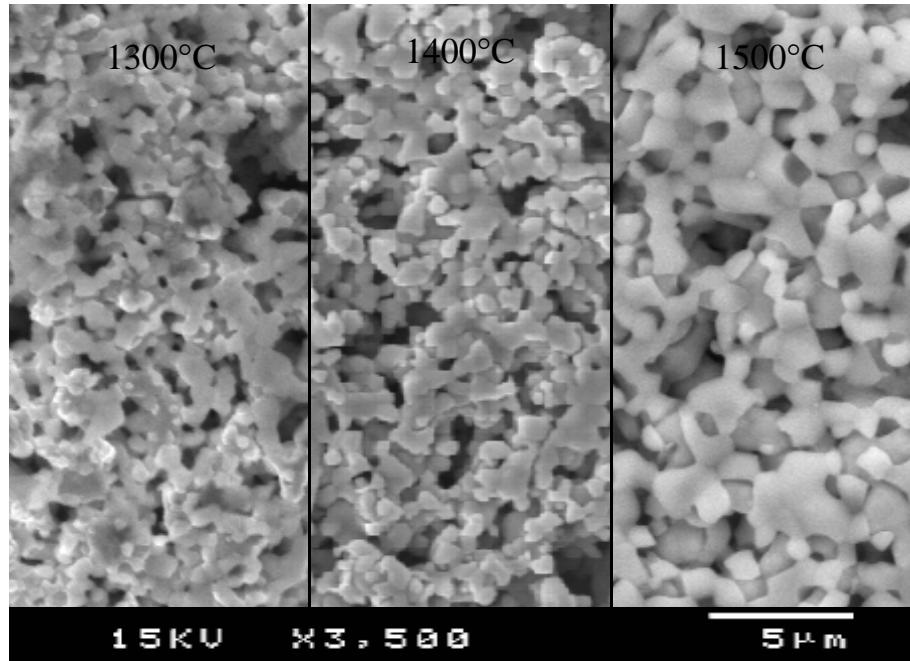


Figure 23 Microstructures of 45 vol % Ni compositions prepared by the conventional technique and sintered on the YSZ electrolyte at various temperatures.

sintering temperatures allowed more densification in the z-direction, resulting in a rigid YSZ structure to support Ni particles

- ◆ By decreasing the densification between Ni grains, there are more paths for conduction (more Ni-Ni contacts throughout the structure and higher conductivities) and a larger number of Ni-YSZ contacts (lower overpotentials).
- ◆ The 45 vol% Ni composition co-fired with the electrolyte had the lowest initial overpotential of any cermet with a similar composition.

## 6.0 FUTURE ACTIVITIES

With respect to the major thrusts of this program:

- ◆ Fabricate additional single cells with controlled microstructures of the anode by co-synthesis of the YSZ and NiO; 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.

Acknowledgement - We gratefully acknowledge the technical interactions with our METC COR, William C. Smith. The period of performance for this program is 4/16/93 through 4/15/98.

## **Planar SOFC Integrated System Technology Development**

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EPRI Program Manager: W. Bakker  
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U.S. ARO Program Manager: R. Paur

### **Introduction and Objective**

Planar SOFC technology development at SOFCo, a McDermott Technology Inc./Ceramatec partnership, has the primary objective of demonstrating compact, high efficiency power generation systems with multi-fuel capability meeting commercial performance goals. Current programs synergistically address this goal through an internally funded program focusing on stack development and system integration for pipeline natural gas (PNG) operation, a DARPA sponsored program to demonstrate logistic fuel operation, and a program to develop cell and stack technology for low temperature operation supported by EPRI.

Recent advances made in planar SOFC stack performance and endurance enabled SOFCo to successfully demonstrate several small scale integrated systems using pipeline natural gas fuel. These advances have enabled verification of system concepts at tall stack scale (more than 60 cells, 4"x4") with commercial level endurance. The system concept using the patented CPn® design, thermally integrates the stack, air preheater, and the reformer by co-locating these components. This platform provides the flexibility to accommodate stacks based on advanced thin film cells currently under development. Multiple system tests enabled development and demonstration of continual improvements at the system level. The design and fabrication of a logistic fuel reformer has been completed in a parallel activity. Process integration of a one kW CPn® system with the logistic fuel reformer has been planned for the third quarter of this year.

## Approach

With the recognition of the system costs and endurance benefits from lower operating temperatures, developmental efforts have been focused in three key areas:

- stack technology development  
(SOFCo)
  - advancement of low temperature electrode technology through materials and process improvement
  - construction and testing of short stacks
  - system verification of tall stacks in the environment of an integrated natural gas reformer and air recuperator
- new materials development  
(EPRI)
  - development and optimization of new electrolyte materials
- fuel processor development  
(DARPA)
  - development and integration of logistic fuel reformation technology

Optimum fuel cell technology development is based on the requirements of integrated power systems, rather than considering only isolated individual cell and stack components and plant equipment. SOFCo developments including the CPn® design and logistics fueled generator are being developed with this system based approach.

The SOFCo CPn® concept evolved through recognition of the impact of balance of plant (BOP) on the economy and efficiency of the total system. The design optimizes the total system and maximizes the efficiency of the system while simultaneously reducing the number of high temperature components peripheral to the stack. The CPn® module consists of a multistack arrangement that allows multistage oxidation of the fuel. Efficiency is enhanced by effective thermal coupling of the stacks with fuel and air processes. The CPn® power system comprises planar SOFC stacks, fuel processor components and the BOP equipment.

The most salient CPn® design feature is the Thermally Integrated SOFC Module, which houses the fuel cell stacks, an air heat exchanger, reforming catalyst, and a spent fuel burner. Air, used as the fuel cell oxidant and coolant, is preheated by heat exchange with the stack air exhaust and delivered to the SOFC stacks. This heat exchanger is incorporated into the wall of the module housing. The fuel processing system is coupled to the stacks within the thermal enclosure accomplishing internal reformation of hydrocarbon fuels. Thermal integration of key process streams within the module provides optimum system performance in a compact, reliable power system.

The key to achieving a low-cost, reliable system is the iterative design and manufacturing development of primary components from a systems perspective, rather than independently developing components separately then combining them into a system. The SOFCo CPn® power system has been successfully demonstrated in a 1.4 kW and several multi-100 W modules.

## Results

### *Cell/Stack Development*

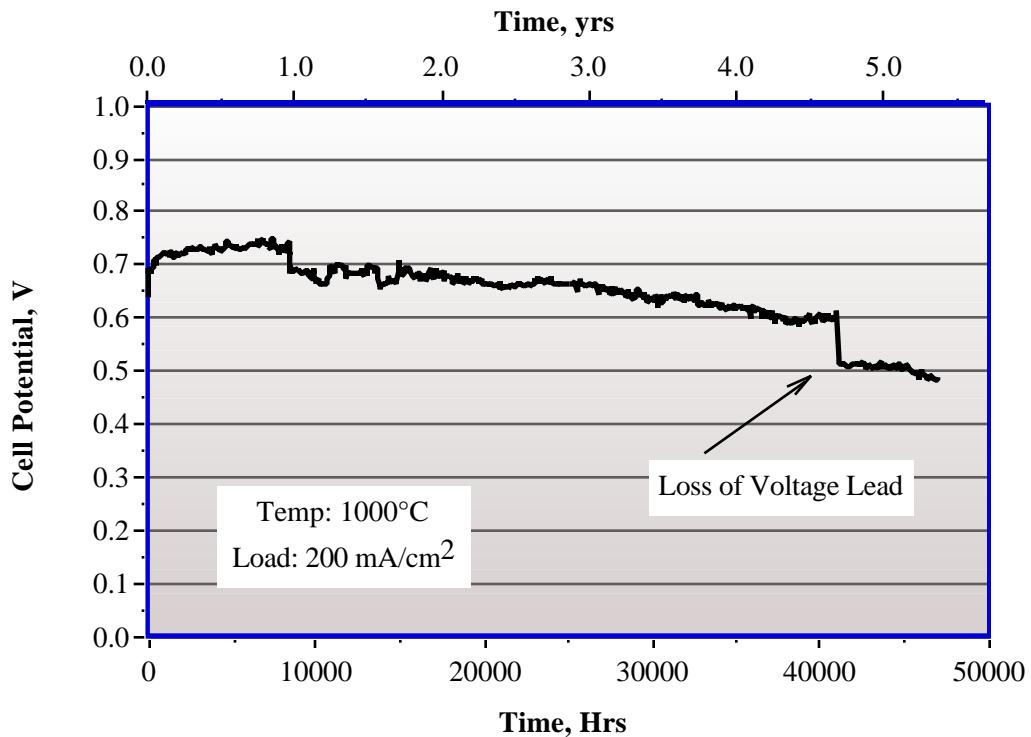
Early developmental activities focused on establishing stable cell operation at 1000 °C. Performance degradation of less than 0.5% per 1,000 hours has been demonstrated for over 46,000 hours of operation (Figure 1). Electrode improvements have established stable stack performance at 850-900°C. The long term performance of 5-cell stacks using the improved low temperature electrodes is shown in Figure 2. Further advances have yielded stacks with demonstrated 800° C operation at an area specific resistance of under 1 ohm.cm<sup>2</sup> (>250 mW/cm<sup>2</sup> peak power density) using 180 micron zirconia electrolyte (Figure 3). Stack performance greater than 95% of single cell performance is now typical.

Single cells have been tested with high sulfur containing fuels (H<sub>2</sub> with 500 ppm H<sub>2</sub>S). Stable cell operation during 150 hours of H<sub>2</sub>S exposure (Figure 4) has been shown. Less than a 30 mV loss was registered while operating on the H<sub>2</sub>S containing fuel.

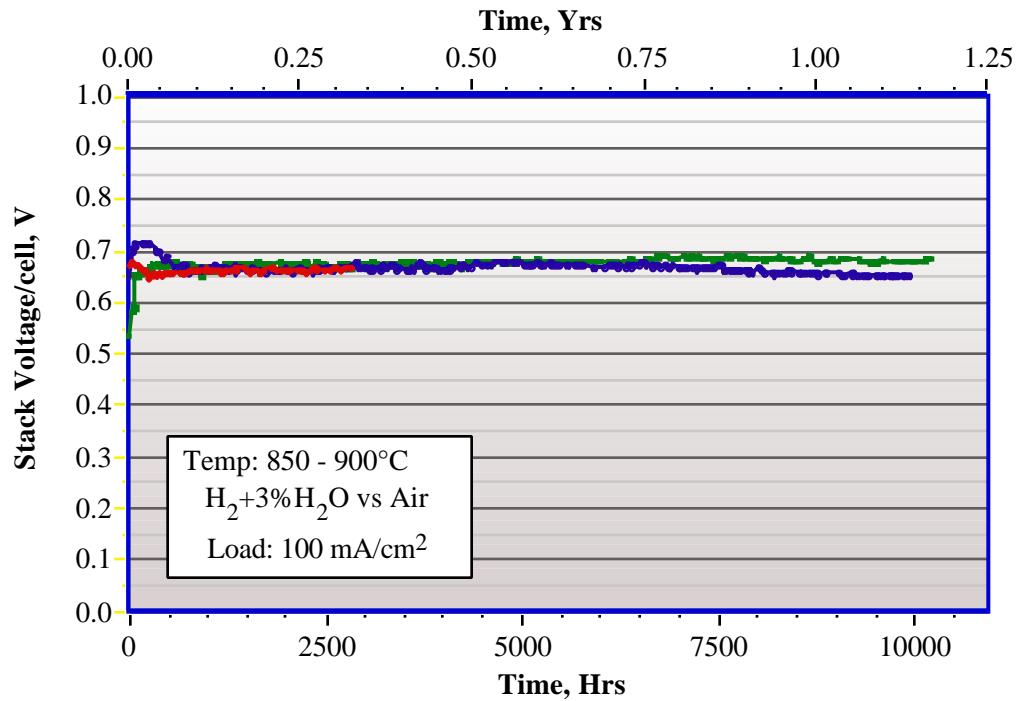
Several integrated system level verifications using pipeline natural gas fuel have been completed. The stack module typically consisted of 60 cells of 4"x4" size. An integrated PNG fuel processor and air preheater were key elements of the system test. The primary objectives of these tests were to establish the stability of stack performance and various sub-systems for multi-100 hours, and to provide experimental verification of system thermal models.

Key Results from these tests are summarized below:

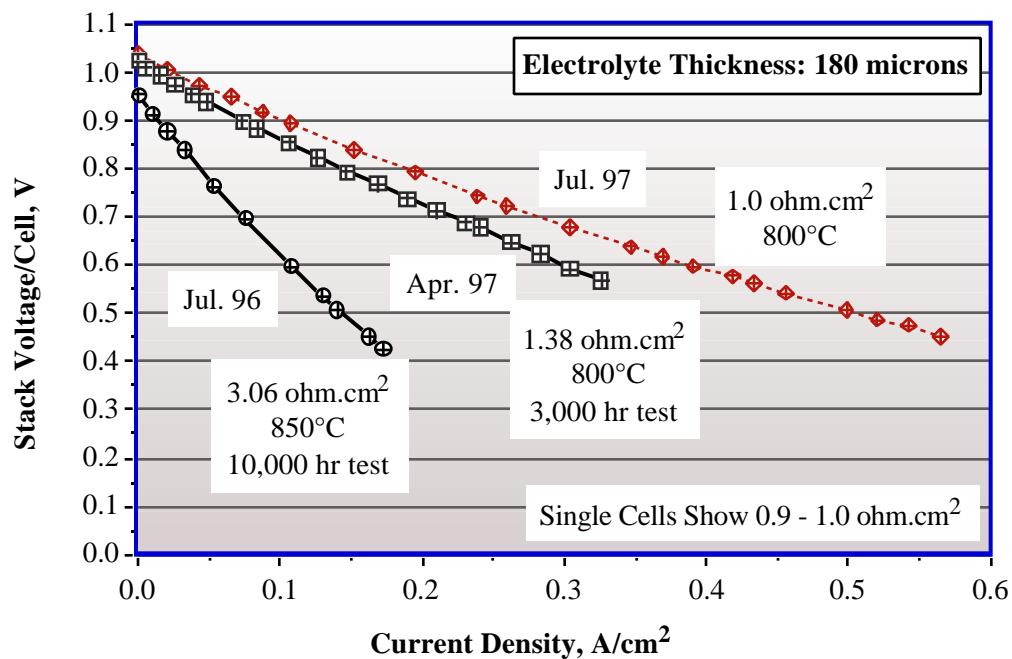
- Theoretical open circuit voltage using both humidified hydrogen and reformed natural gas for a 62 cell stack over four thermal cycles to room temperature.
- Verification of model predicted system performance (stack performance and thermal distribution).
- Performance stability over 250 hours of operation using PNG fuel (Figure 5).



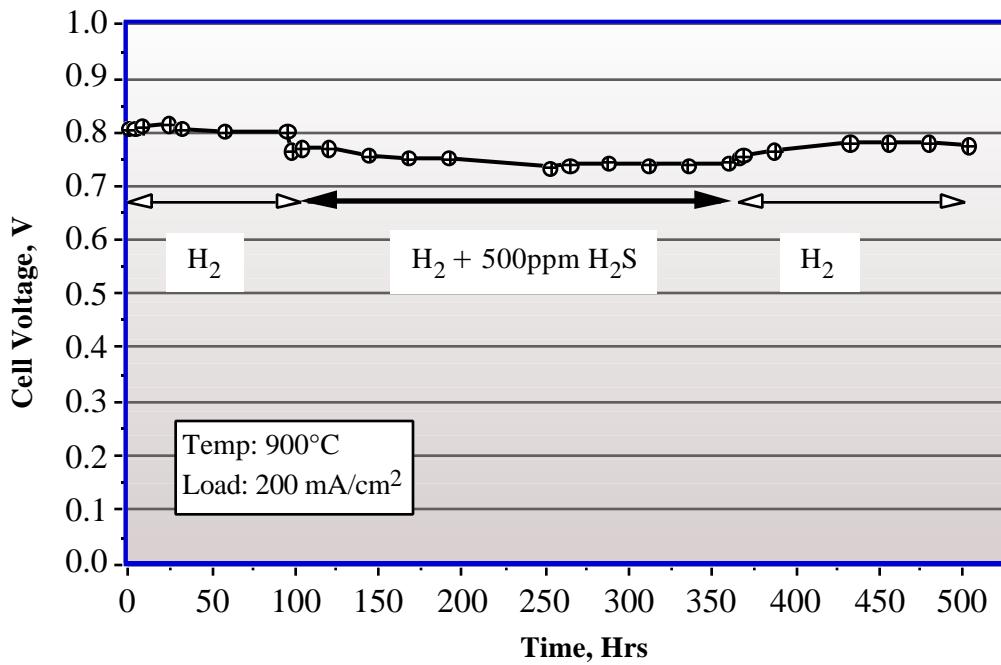
**Figure 1. Single Cell Endurance Test**



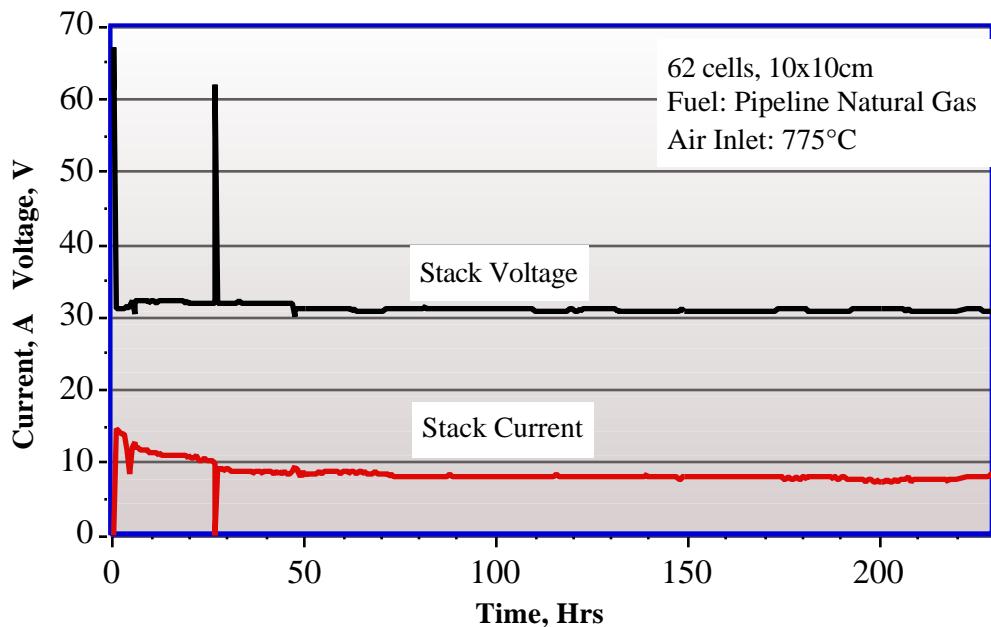
**Figure 2. Stack Endurance Test**



**Figure 3. Stack Performance Improvement**



**Figure 4. Sulfur Tolerance Test**



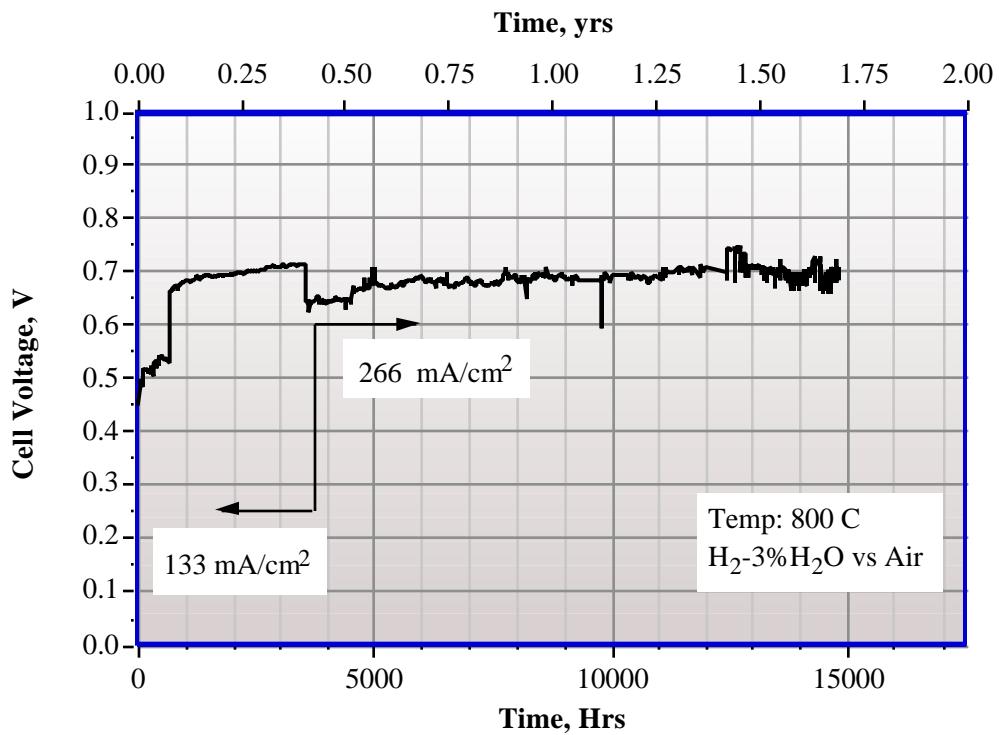
**Figure 5. Tall Stack Test**

### **Low Temperature Electrolyte Development**

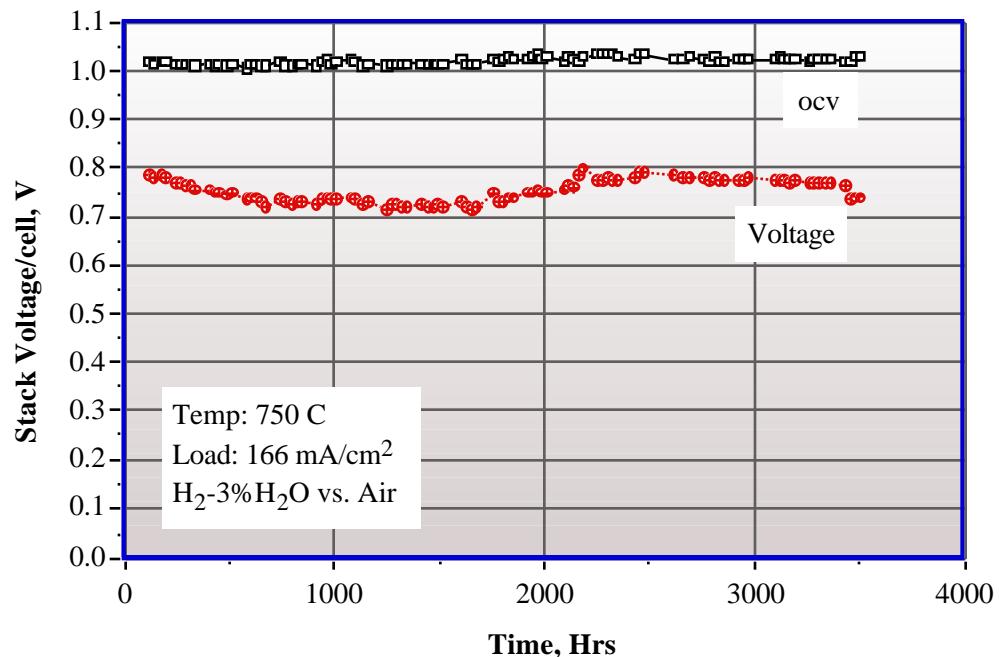
In the area of low temperature electrolyte materials, two classes of materials were evaluated: doped ceria and lanthanum gallate. Both single cells and stacks were fabricated and tested. Although the ceria electrolyte is known to exhibit electronic shorting under fuel cell operating conditions, exceptional performance stability was observed. A ceria electrolyte single cell was operated for 15,000 hours with negligible performance degradation (Figure 6). Performance models incorporating experimental data show stack efficiencies of 45% could be achieved using natural gas fuel. The ability of these cells to operate at temperatures of 600 - 700°C provides an opportunity for lower BOP costs. A lanthanum gallate electrolyte stack (5x5 cm, 5 cells) operating at 750°C has shown very low degradation over the initial 3,000 hours of operation (Figure 7).

### **Logistic Fuel Processor Development**

SOFCo, through sponsorship from DARPA and the U.S. Army, is working on a three-phase, four-year program to demonstrate a mobile electric power fuel cell generator operating on logistic fuel. The program aim is to integrate planar solid oxide fuel cells with a JP-8 fuel processor into a compact generator module. This integration of fuel processing equipment for reforming high sulfur (0.3% by weight) logistic fuel, with SOFC stacks into a compact hardware configuration will represent a significant advance in the state-of-the-art in SOFC system technology.



**Figure 6. Low Temperature Cell Endurance Test  
(Ceria Electrolyte)**



**Figure 7. Low Temperature Stack Endurance Test  
(Lanthanum Gallate Electrolyte)**

The U.S. Army Lightweight Multipurpose Shelter (LMS) was selected as a target application for this project (Figure 8). The LMS generator is housed within a small tunnel, measuring approximately 29" x 25" x 84", near the front of the shelter. The LMS selection was based on the Army's interest in an efficient and quiet power supply to replace the current diesel generator, and the suitable match between the LMS power requirement and the 10 kW demonstration size for the current program.

During June 1996 (Phase 2 in the program) SOFCo demonstrated a 10 kWe partial oxidation (POx) fuel processor in combination with a 100 W solid oxide fuel cell stack (Figure 9). This was the first successful demonstration of a planar SOFC stack operating on JP-8 fuel.<sup>1</sup>

The Phase 2 fuel atmospheric pressure processor used partial oxidation (POx) to reform the JP-8 into a hydrogen-rich synthesis gas (syngas) for use in the fuel cells. While a 100 hour demonstration of the fuel cell/fuel processor system was achieved, further development in the areas of maintenance, portability, and efficiency was planned for Phase 3 before overall system physical integration could begin.

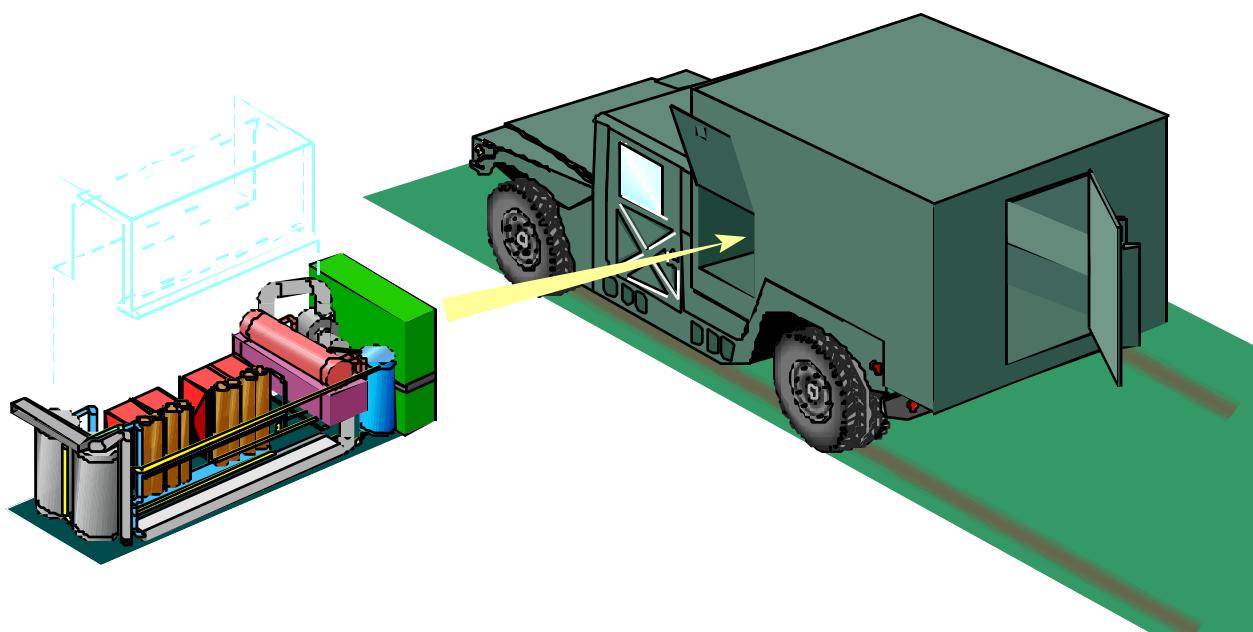
The Phase 3 fuel processor objectives focus on reducing maintenance, improving fuel conversion efficiency, and demonstrating a soot management system compatible with MEP mobility requirements. A new, proprietary POx reactor design is key to achieving these goals. The new design, which is based on proven gasification experience, will accommodate the temperatures and gas residence times required for efficient, long-term operation. In addition, efforts to prevent or minimize coke and soot formation rely on detailed analysis of the reactant mixing and synthesis gas handling. A key challenge is to demonstrate suitable performance in the system which is 1/200th the scale of industrial units.

Proprietary numerical models for the primary partial oxidation reactions were used, together with existing industrial gasifier design and performance correlations to evaluate various POx reactor designs. The POx reaction numerical models include zero, one, and two-dimensional flow representations with quasi-global kinetic modeling. In particular, two-dimensional models, which include full reaction kinetics were used to predict reactor flow patterns, mixing performance, gas residence times, temperature fields and gas composition. The models were benchmarked with data from the Phase 2 fuel processor and then validated using industrial reactor data.

To validate and confirm multi-dimensional numerical models, physical flow visualization tests were conducted. Plexiglas models based on candidate reactor and flow nozzle designs were tested. Air flow rates were adjusted to match stream momentum.

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<sup>1</sup> "Status of SOFCo Planar SOFC Technology Development", R. Privette, M. Perna, K. Kneidel, J. Hartvigsen, S. Elangovan, A. Khandkar, 1996 Fuel Cell Seminar, Orlando, Fla, Nov. 17-20, 1996.



*Figure 8. SOFCo MEP generator conceptual design for Army LMS application*



*Figure 9. Phase 2 breadboard system arrangement*

Smoke particles, illuminated in a two-dimensional laser sheet, were used to evaluate mixing patterns. Figure 10 shows smoke being injected into a model reactor to investigate potential short circuiting of the reactor which could result in unreacted fuel in the product syngas. This approach of combining physical and numerical modeling allowed several different design configurations to be evaluated and optimized.

The Phase 3 fuel processing train has been physically unintegrated to allow optimization of the fuel conversion performance through independent control of operating parameters. The fuel processing train is shown in Figure 11. Included are separate components which accomplish fuel and water vaporization, air preheating, JP-8 partial oxidation, synthesis gas cooling, soot filtering, desulfurization, and reheating. The system is designed to operate over a wide range of temperatures (to 3000 °F) and residence times. It is predicted to achieve a 75% cold gas efficiency<sup>2</sup>. This is significantly higher than the 50-55% conversion level achieved in Phase 2.

Physical and thermal re-integration of the fuel processor will follow subsystem optimization. This integration, including the fuel cell heat and steam byproducts is expected to provide substantial size and weight savings necessary to meet the aggressive constraints of the Army LMS application and will benefit the overall system efficiency.

In addition to increased efficiency and reduced maintenance, another important functional requirement for the Phase 3 fuel processor is the ability to operate with a soot filtering system compatible with the MEP mobility requirements. In the Phase 3 demonstration, an automated system using two parallel soot filters will be used. This will allow continuous on-line soot filtering by cycling between the filters. Cleaning of the off-line filter will be accomplished by injecting air to oxidize the collected soot. The new system requires less space than the soot filter system used during Phase 2.

The feasibility of oxidizing the soot to clean the filter was evaluated with a 1/3rd scale bench test. A sintered metal filter was installed within a vessel and positioned in a furnace to control temperature. Soot collected during Phase 2 testing was first fluidized in a nitrogen stream and applied to the outside surface of the filter. After soot application, a low flow-rate of air was directed through the filter to oxidize the soot. Analysis of the exhaust gas composition (CO and CO<sub>2</sub>) was used to monitor rate of soot oxidation.

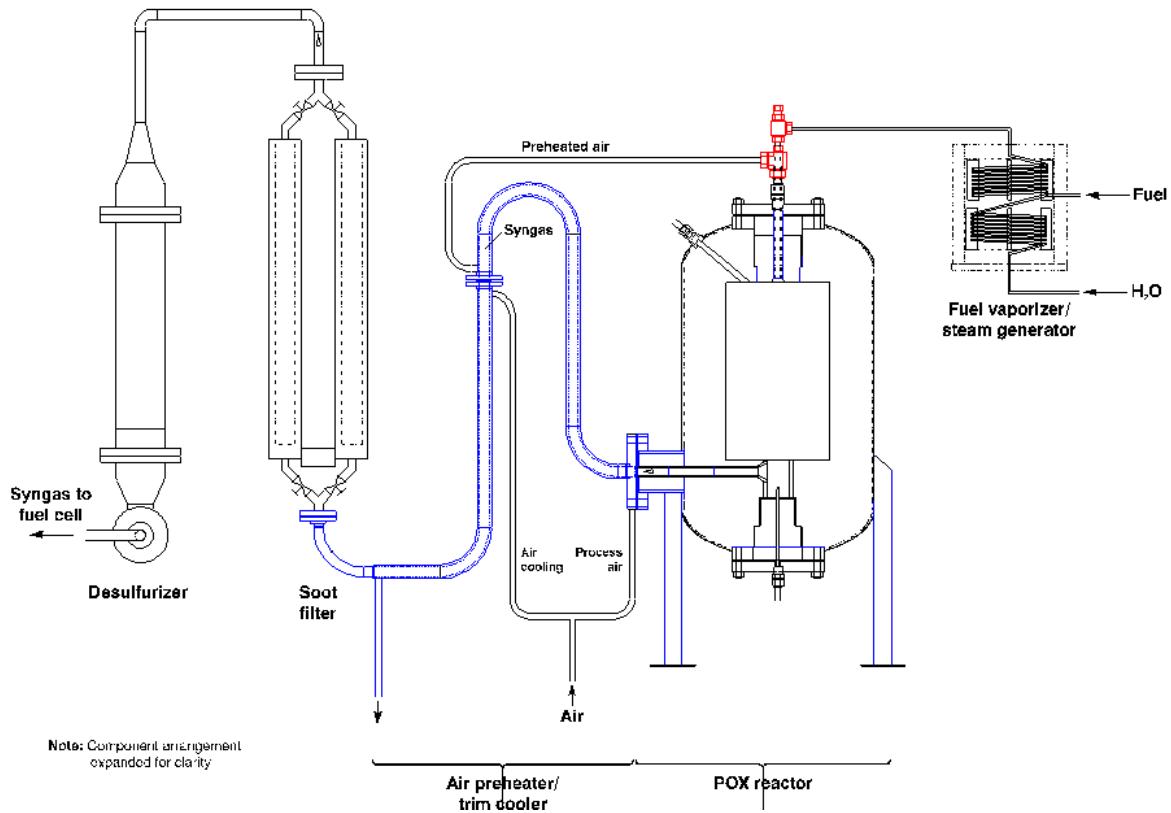
Encouraging results were obtained from the soot filter cleaning tests as shown in Figure 12. These tests demonstrated the ability to clean the sintered metal filter on-line and achieve a clean condition (as determined by pressure drop). The time required to oxidize the soot was determined by monitoring the oxygen concentration in the product gas. Repeated cycles of soot filtering and soot oxidation demonstrated the ability to return to a repeatable filter pressure drop. These results indicate that there will not be a

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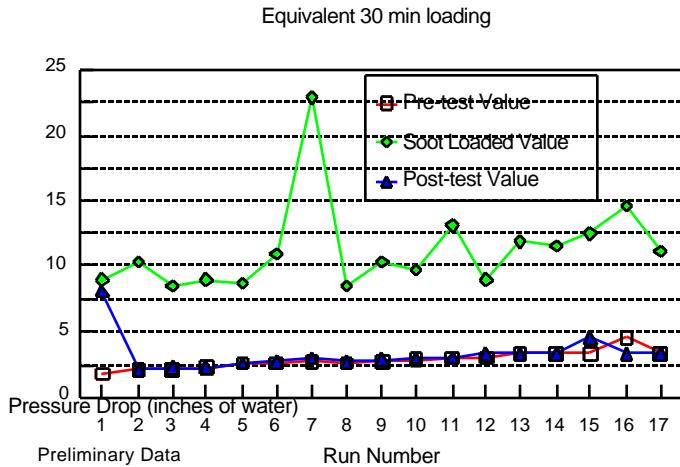
<sup>2</sup> Cold gas efficiency = Higher heating value of (H<sub>2</sub> + CO) products/ Higher heating value of feedstock



*Figure 10. Cold flow visualization studies used to verify numerical modeling results*



*Figure 11. Schematic of unintegrated JP-8 fuel processor*



**Figure 12. Results from soot filter regeneration tests**

gradual increase in pressure drop across the filter from repeated cleaning cycles over the hundreds of hours of fuel processor operation.

System installation is nearing completion in the test facility at the McDermott Technology, Inc. Alliance Research Center in Alliance, Ohio. Fuel cell system demonstration is planned for late September, 1997. Following successful demonstration of the breadboard logistics fueled solid oxide fuel cell system, efforts are planned to address physical integration and transient operation.

## Future Activities

In addition to the EPRI and DARPA/U.S. Army sponsored work, SOFCo also has several initiatives aimed at the commercial development of planar, solid oxide fuel cell technology. The Distributed Power Initiative (DPI) is currently developing a 2 kW natural gas fueled Technology Demonstration Unit (TDU). This TDU will be the first complete integration of a thermally self-sustaining planar SOFC system including the reformer, storage, inverter, controller and heat recovery components. A subsequent 10 kW TDU is also being designed and will be constructed in 1998.

The DPI's ultimate goal is to make use of both previous and current research to develop a commercial system for an initial market entry sized from 10kW-50kW. Field demonstration units will be built for specific high-value applications in an effort to penetrate the commercial market in those areas where higher priced initial units can be

introduced at a premium. Although specific to the market segment in which the units will be introduced, much of the work associated with the DPI is aimed at both standardizing component design and manufacturing. In addition, an aggressive manufacturing cost reduction program also accompanies the DPI wherein high rate/high volume manufacturing techniques are being developed in an effort to reach cost targets derived from market based demand and performance parameters.

## **Acknowledgment**

This work is sponsored, in part, by the Electric Power Research Institute, the U.S. Army Research Office, DARPA, and SOFCo. The content of the information does not necessarily reflect the position or the policy of the U.S. Government, and no official endorsement should be inferred.

# Planar SOFC Integrated System Technology Development

Fuel Cells '97 Review Meeting

August 27, 1997

Morgantown, West Virginia

Work Supported by

EPRI

DARPA

SOFCo

*Clean Energy for the World*



# Outline

## ■ Development Scope Overview

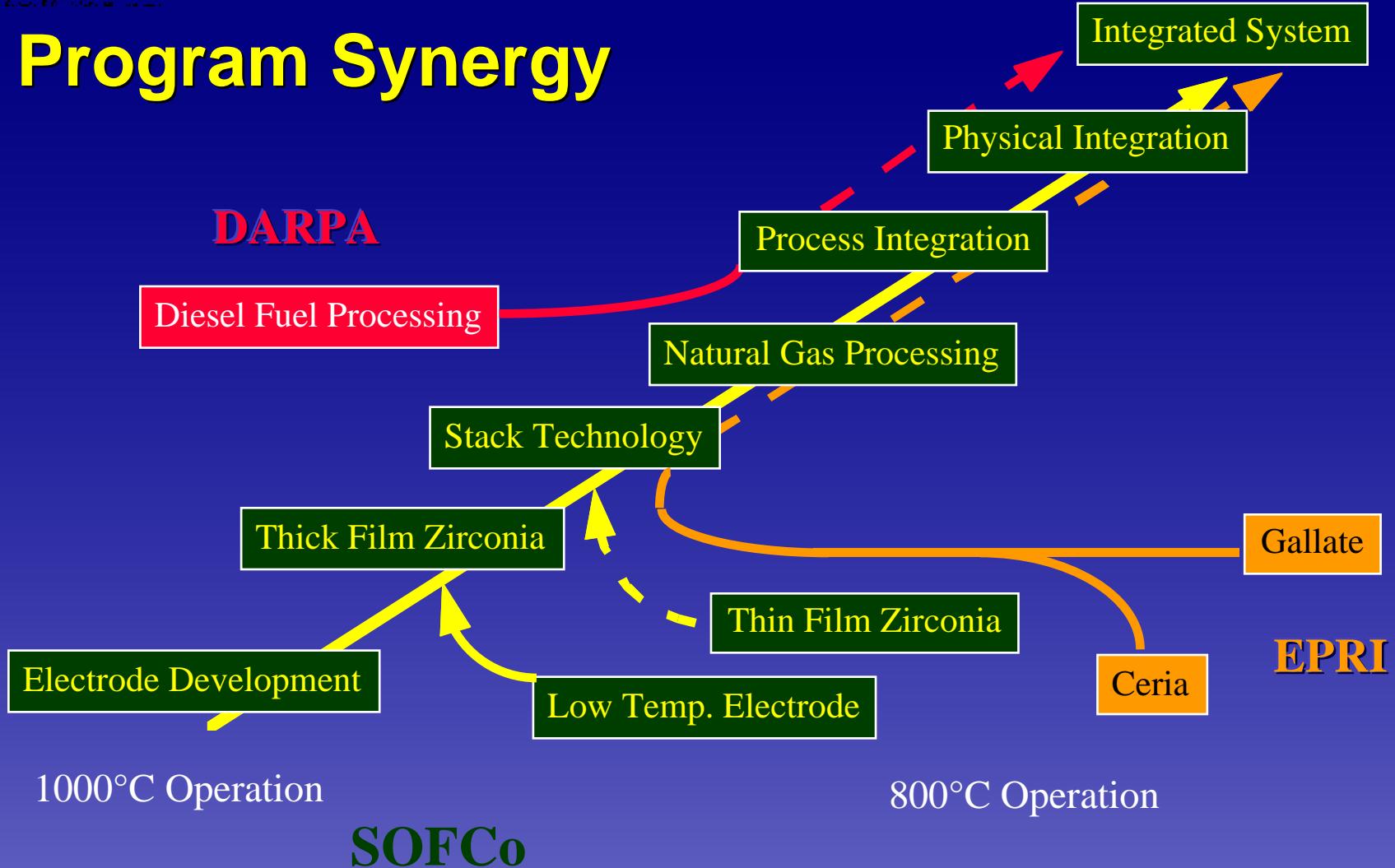
## ■ Significant Results

- cell/stack endurance
- natural gas system endurance
- sulfur tolerance
- low temperature stacks
- fuel processing

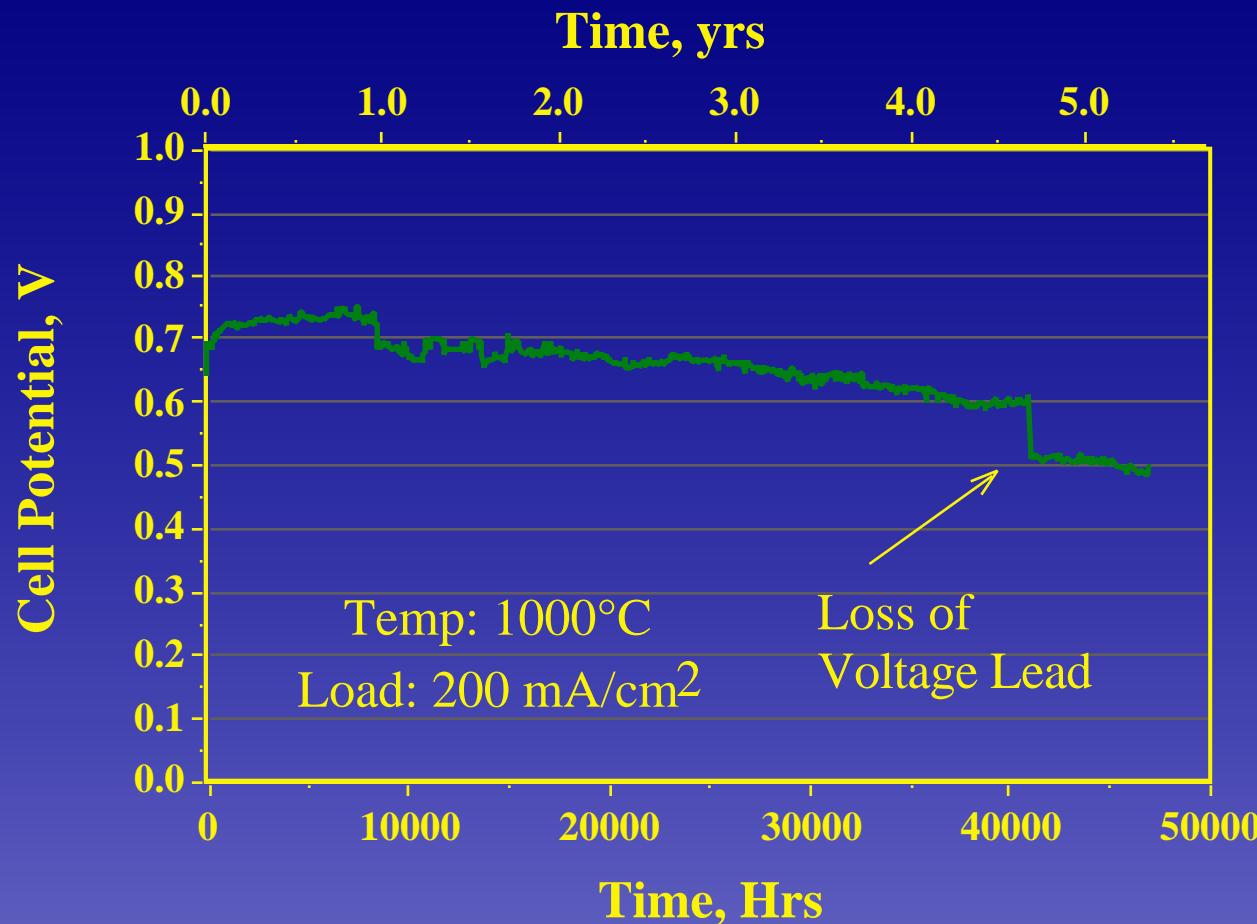
## ■ Future Activities

- ◆ JP-8 combined fuel processor and stack test
- ◆ Technology Demonstration Unit (TDU)

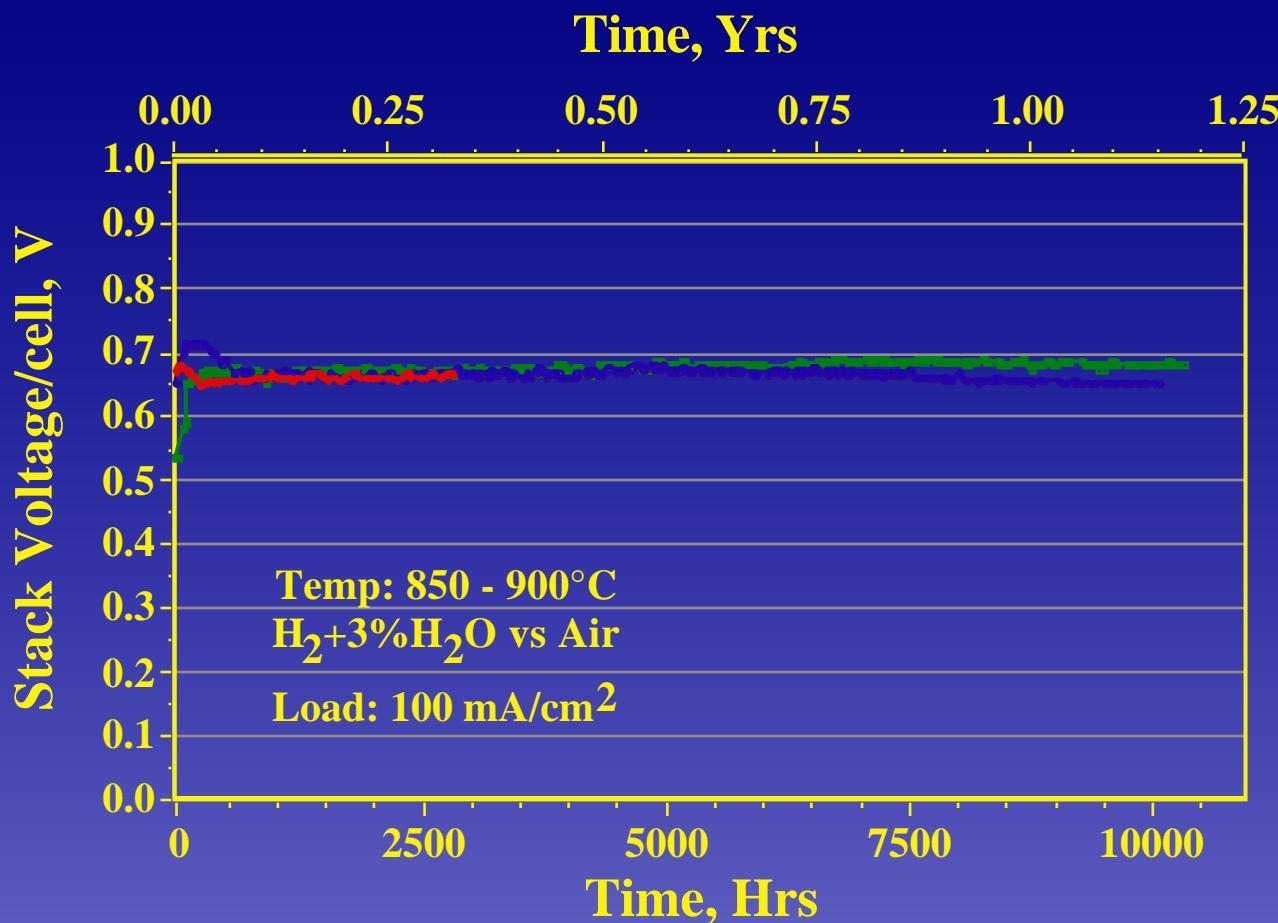
# Program Synergy



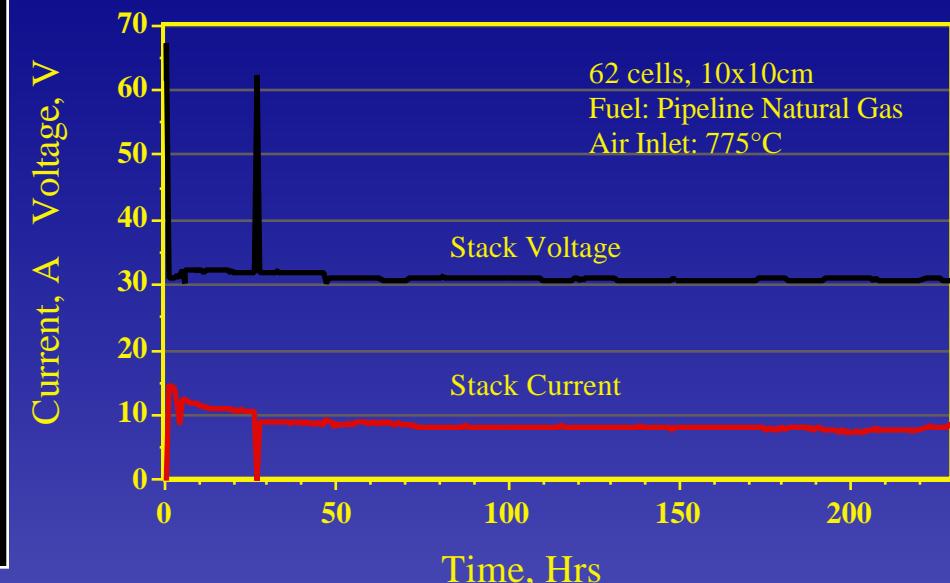
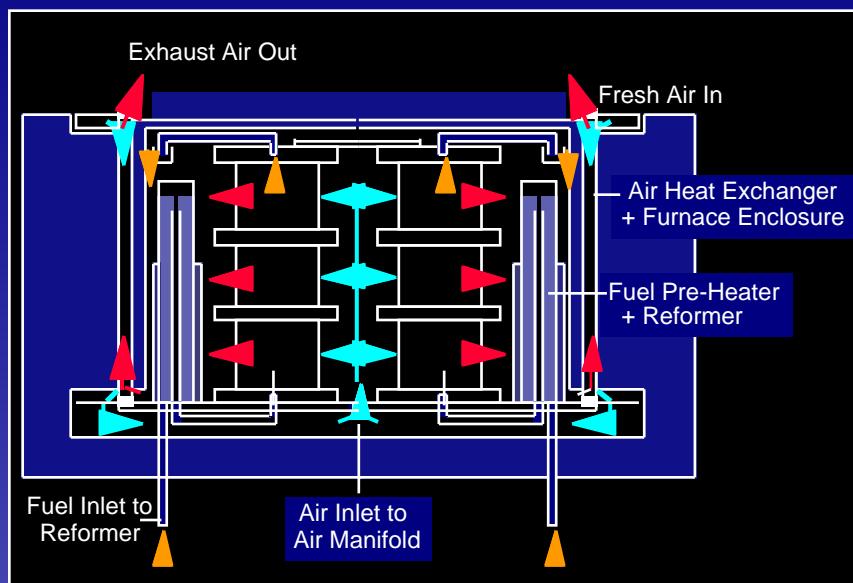
# Cell Technology: Long Term Stability



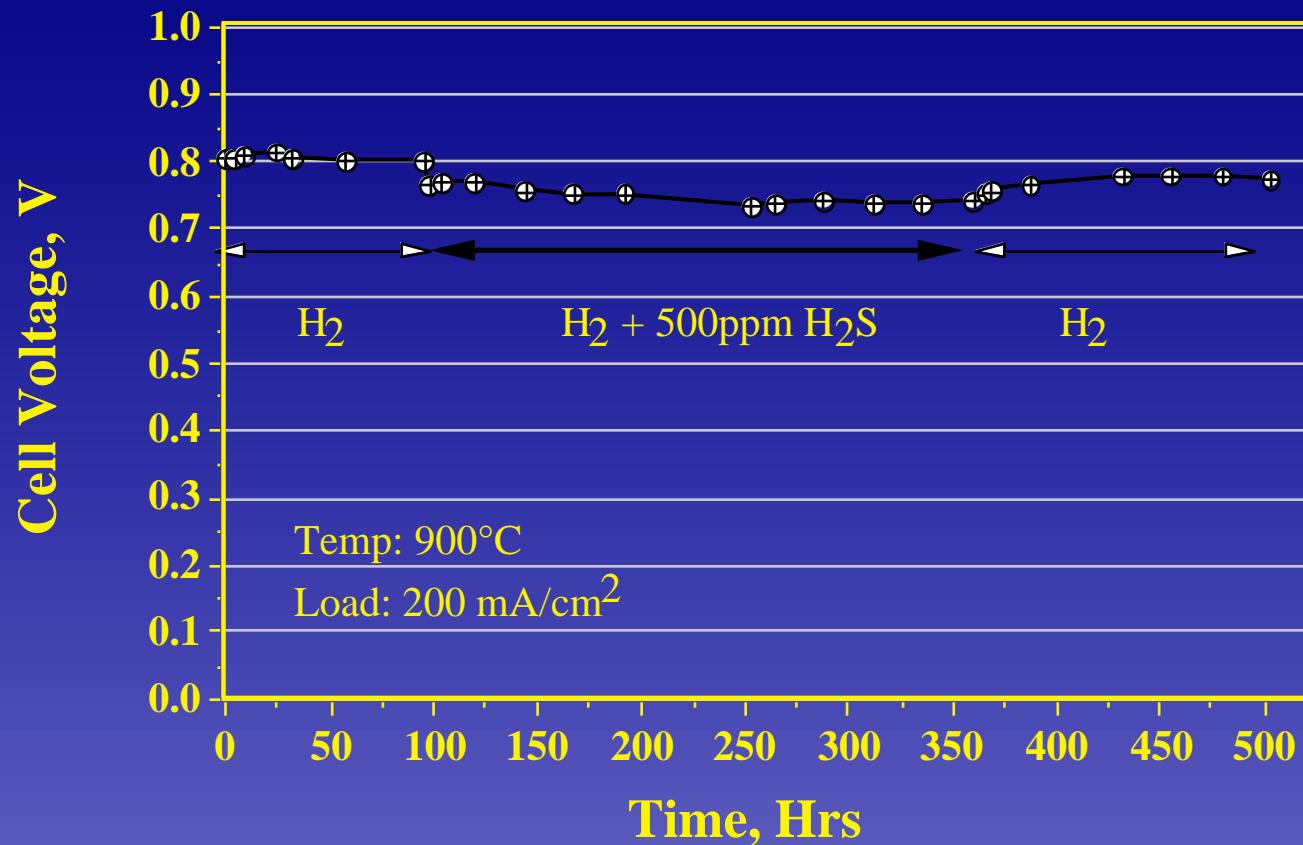
# Stack Technology: Long Term Stability



# System Technology: Tall Stack, Air Preheater, Integrated PNG Reformer



# Sulfur Tolerance Testing

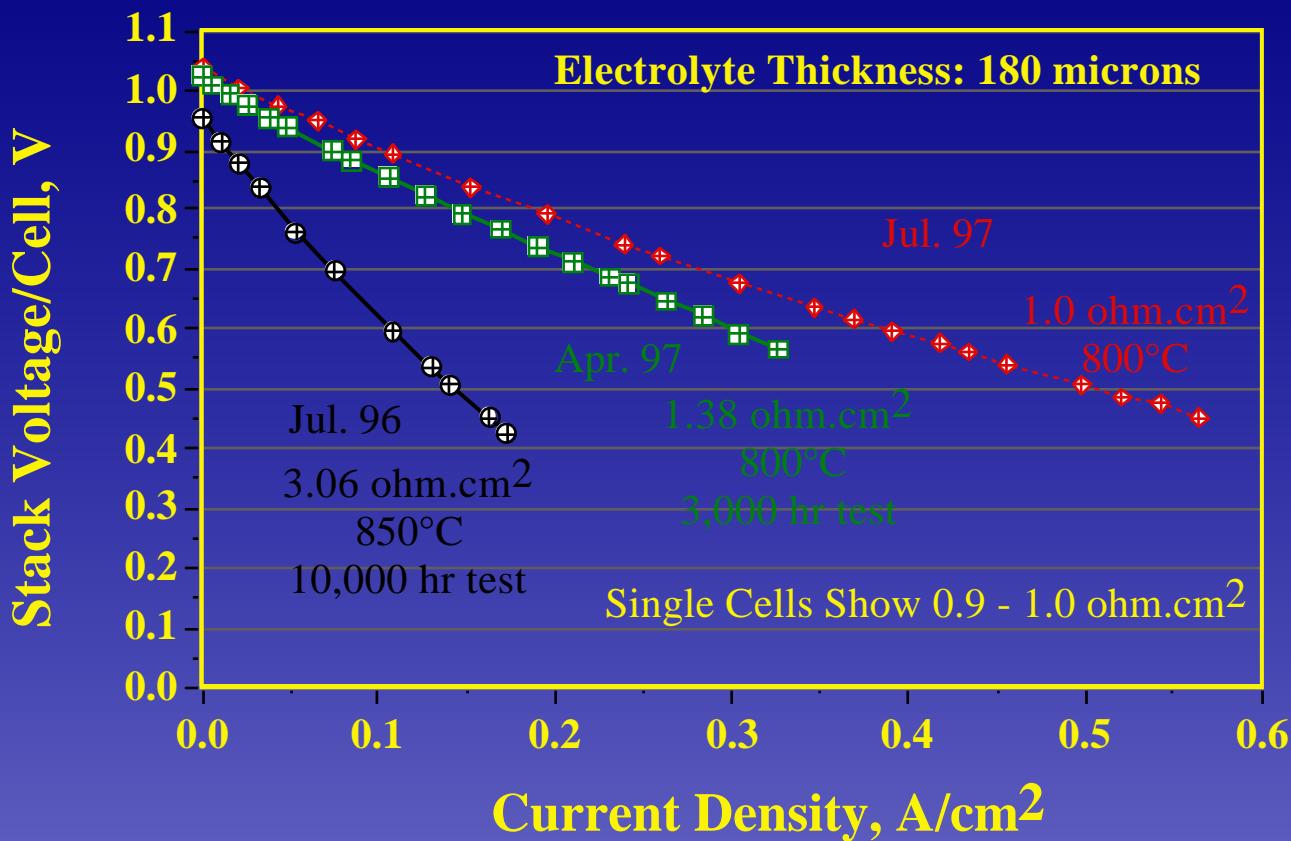


# Low Temperature Stack Technology

- ◆ Approach to temperature reduction
  - electrode/interconnect optimization
    - conventional 180 $\mu$ m YSZ
  - stackable thin film YSZ
  - ceria
  - lanthanum gallate

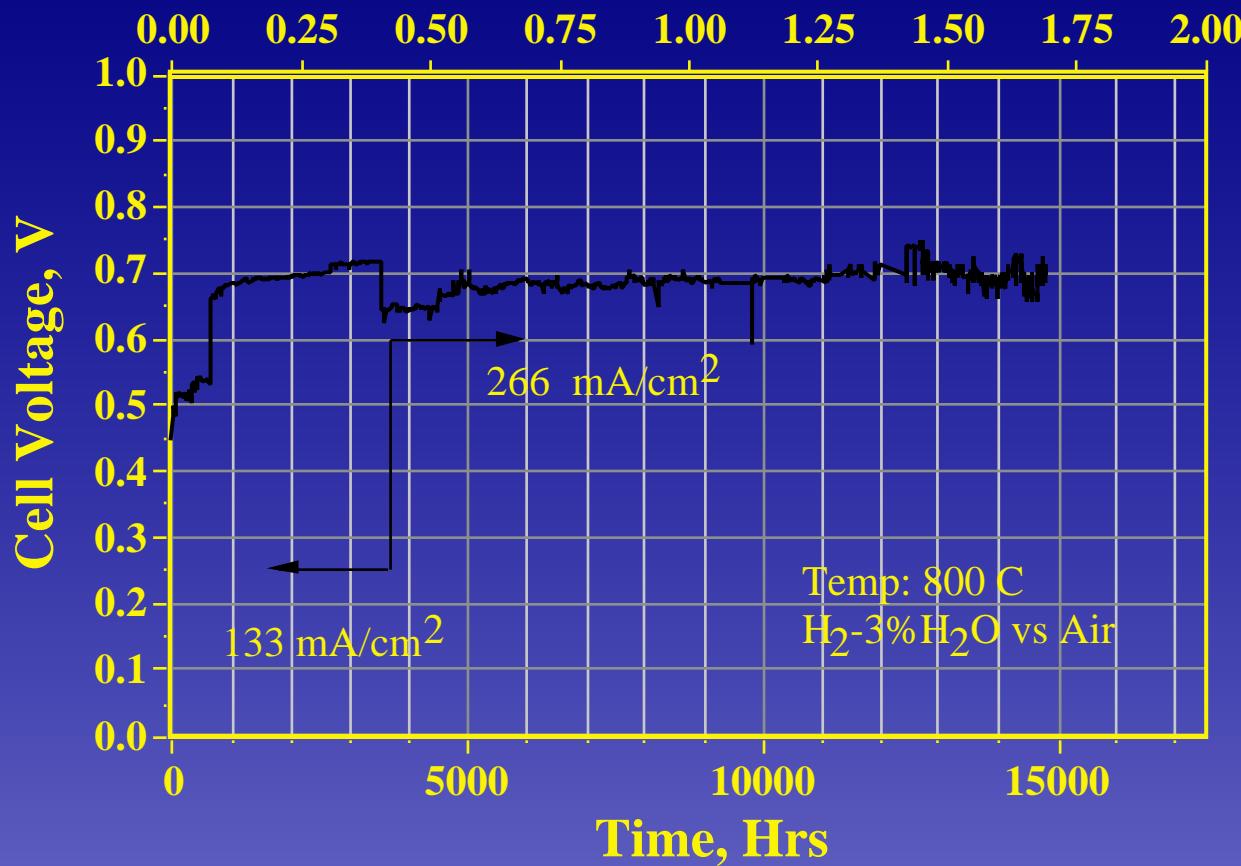
# Temperature reduction:

## Electrode / Interconnect Optimization

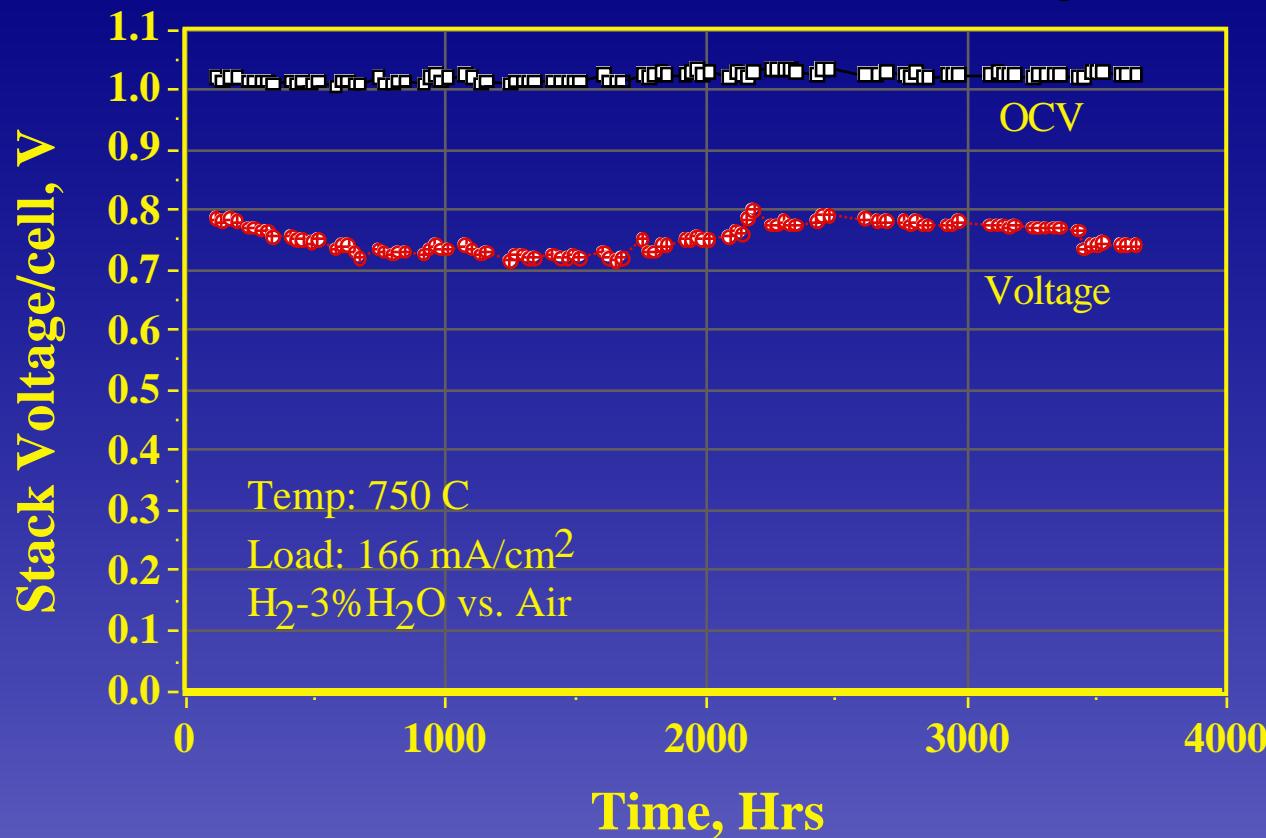


# Temperature reduction: Ceria Electrolyte Cell

Time, yrs



# Temperature reduction: Lanthanum Gallate Electrolyte Stack

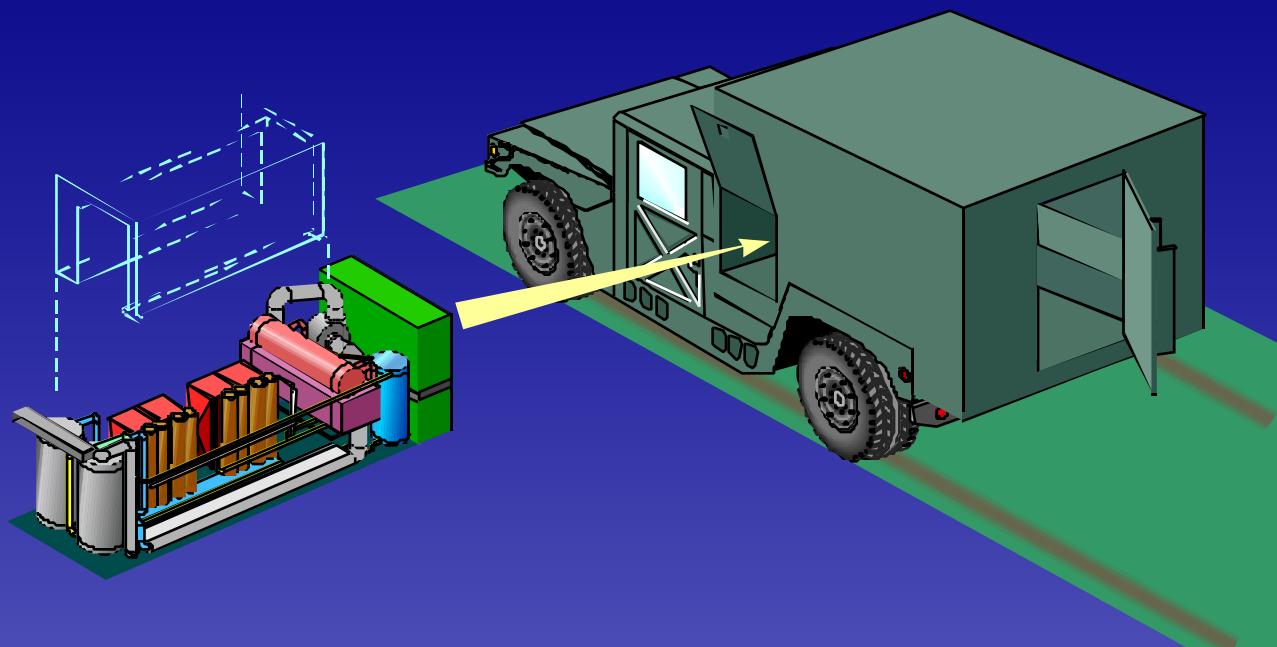


# System Technology Development

## ■ Mobile Electric Power

- ◆ DOD Logistic Fuel Operation
- POx Fuel Processing
- ◆ Process Integration (Breadboard Test)
- ◆ Physical Integration

# DARPA/Army Logistic Fuel Program



# SOFC Mobile Electric Power

- Four-year program sponsored by DARPA and U.S. Army Research Office
- Application
  - ◆ 10 kWe mobile generator for Army Lightweight Multipurpose Shelter
- Key project elements
  - ◆ Compact, lightweight system for mobile applications
  - ◆ JP-8 fuel processor development
  - ◆ Power system integration with SOFCs
  - ◆ Demonstration of multi-kW SOFC power system

## Phase 2 System Demonstration



McDermott Technology, Inc., Alliance, Ohio

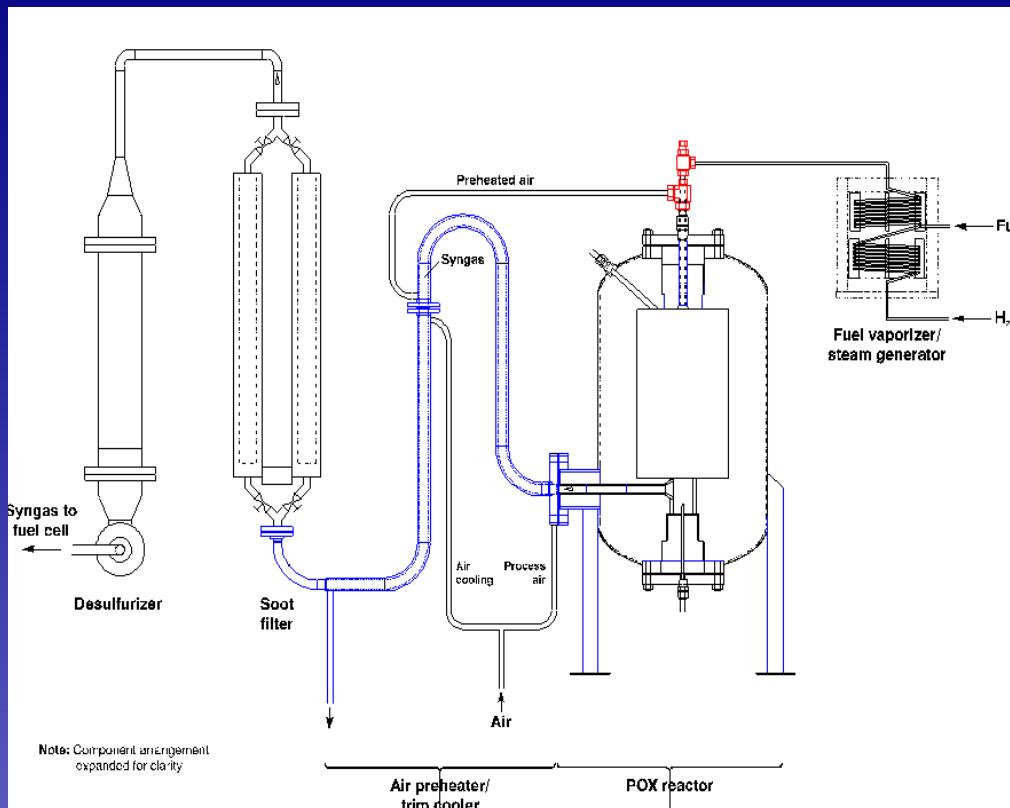
**400 hr operation of  
compact 10 kW<sub>e</sub>  
partial oxidation JP-8  
fuel processor**

**Demonstrated  
integrated system of  
SOFC stack operating  
on JP-8**

## Phase 3 Intermediate Demonstration

- Integrated breadboard system demonstrating prototype-ready subsystem development
- 1 kWe SOFC Stacks
  - ◆ Fuel Utilization >50%
- 10 kWe JP-8 Fuel Processor
  - ◆ 75% fuel conversion to H<sub>2</sub>, CO based on HHV
  - ◆ Uninterrupted operation for 100+ hrs

# Logistics fuel processor

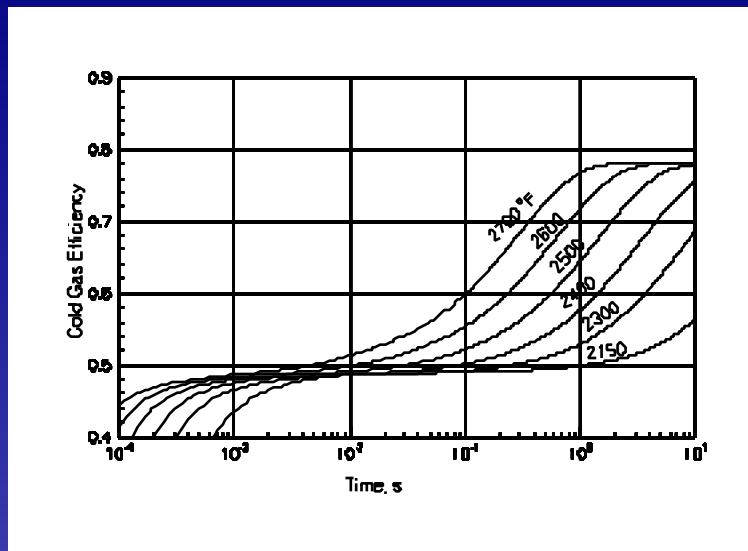


## Features:

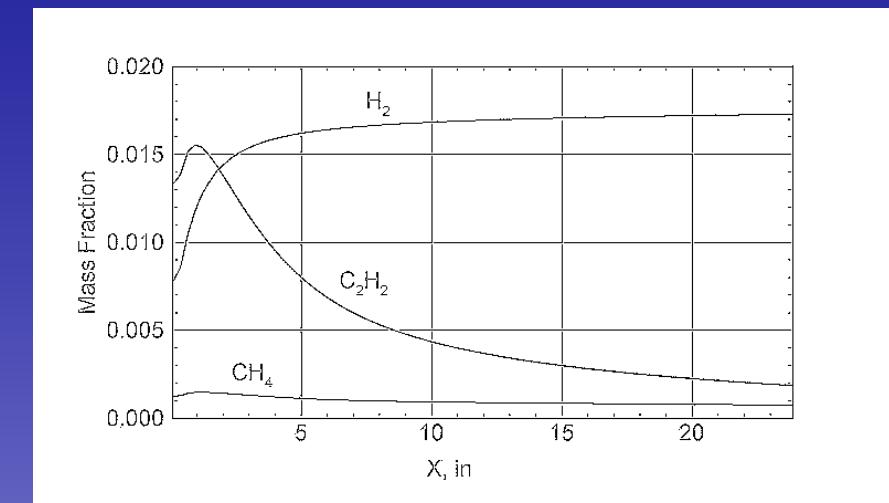
- Efficient fuel conversion
- Low maintenance
- Compact syngas filter

Unintegrated to allow performance optimization through independent subsystem control

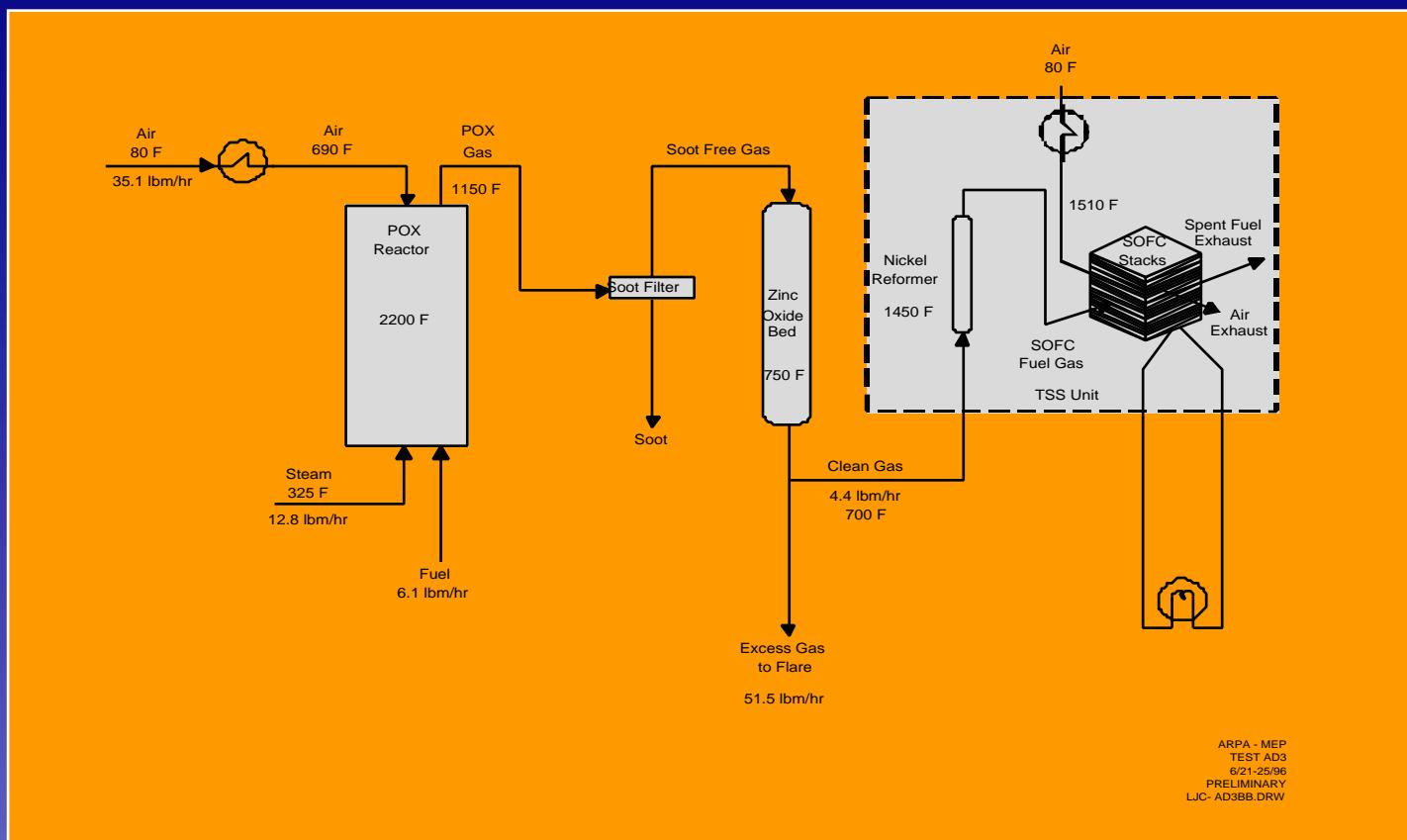
# Dependence on time, temperature



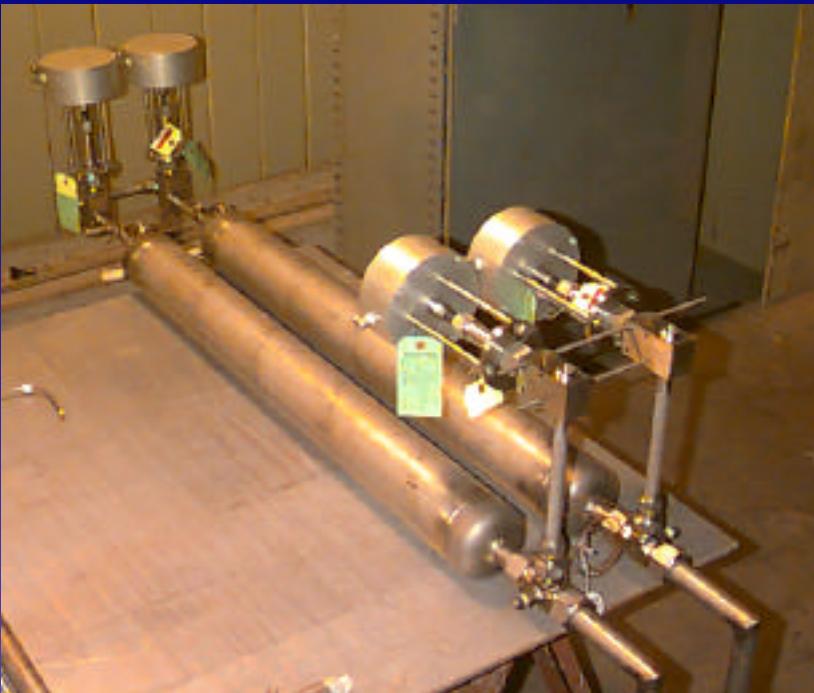
Gas residence time, temperature, and composition are based on numerical modeling of reaction kinetics and benchmarked with industrial gasifier data.



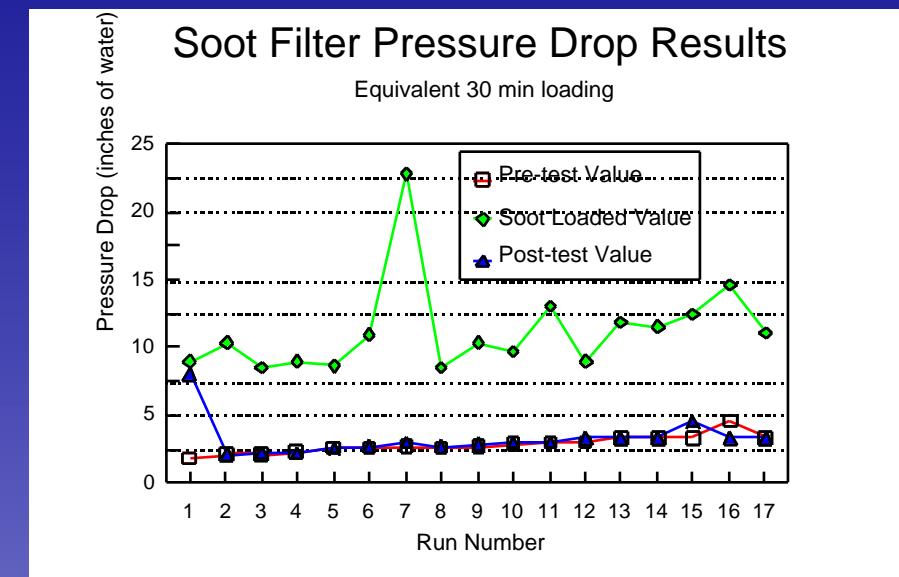
# Phase 2 Integrated Breadboard Demonstration



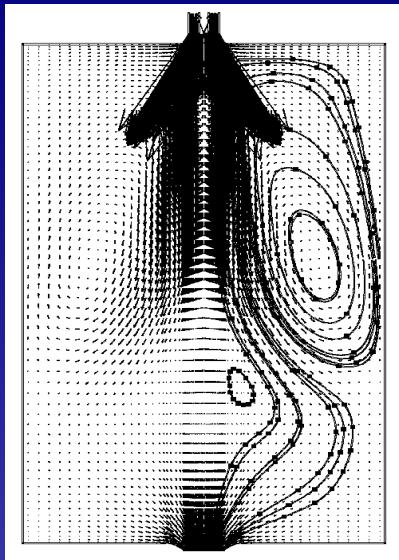
# Regenerable soot filter



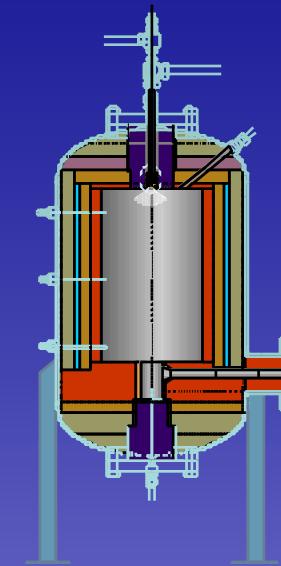
- Automated system for on-line soot filtering
- Reproducible filter regeneration (oxidation) demonstrated



# Reactor design for enhanced mixing



Numerical and physical flow modeling were used to evaluate design options.



# Hardware installation



- System demonstration planned for September, 1997
- Future workscope will address hardware integration and performance mapping

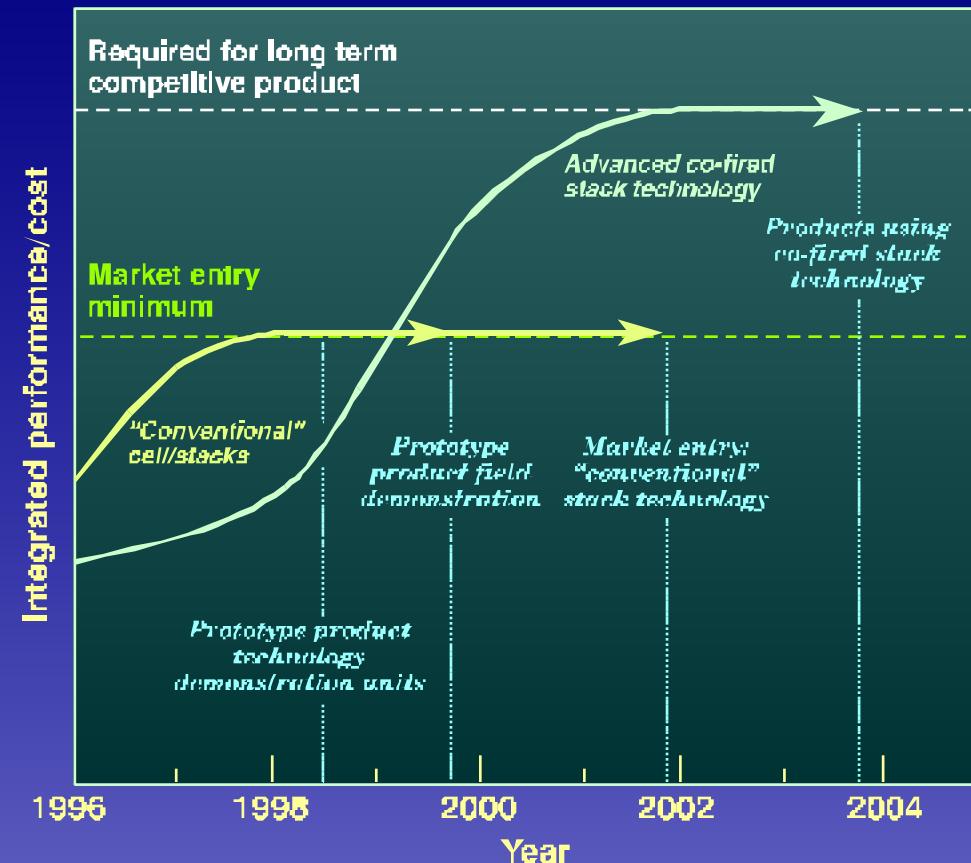
# Technology Demonstration Unit (TDU)

## ■ Technical Objectives

### ◆ integrated package

- 2kW stack output
- pipeline natural gas input
- thermally self sustaining
- electrically self sustaining
- AC output
- waste heat output

# SOFCo Distributed Power Initiative



# Tape-Calendered SOFC Stack Development

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## Abstract

Recent SOFC stack development efforts at AlliedSignal have been focused on demonstrating operation and performance at reduced temperatures (600 to 800 °C). A cost-effective process based on tape calendering has been developed for making reduced-temperature thin-electrolyte cells, and a stack design concept for this application has been evaluated. Use of thin-electrolyte cells reduces stack internal resistances, thus permitting efficient operation at lower temperatures. The proposed stack design incorporates thin-electrolyte cells with metallic interconnect assemblies (made from thin foils) to form a compact, lightweight structure. SOFC stacks based on this design have demonstrated excellent performance and high power densities.

To date, tape-calendered SOFC stacks of up to ten-cell height and 100-cm<sup>2</sup> footprint area have been fabricated. Stacks of various sizes have been tested and have shown excellent performance at reduced temperatures. For example, the power output of a two-cell stack (25-cm<sup>2</sup> footprint area) is about 26 W at 800 °C with hydrogen fuel and air oxidant (power density of 670 mW/cm<sup>2</sup>). A five-cell stack (100-cm<sup>2</sup> footprint area) produces about 270 W at 800 °C (600 mW/cm<sup>2</sup>) and 170 W at 700 °C (375 mW/cm<sup>2</sup>). High stack power densities (1.03 kW/kg and 0.90 kW/L at 800 °C) have been demonstrated.

This paper discusses the development status of the tape-calendering process and the fabrication and operation of tape-calendered stacks at reduced temperatures.



# **Tape Calendered SOFC Stack Development**

**Fuel Cells '97 Review Meeting**  
**FETC, Morgantown, WV**  
**August 26-28, 1997**

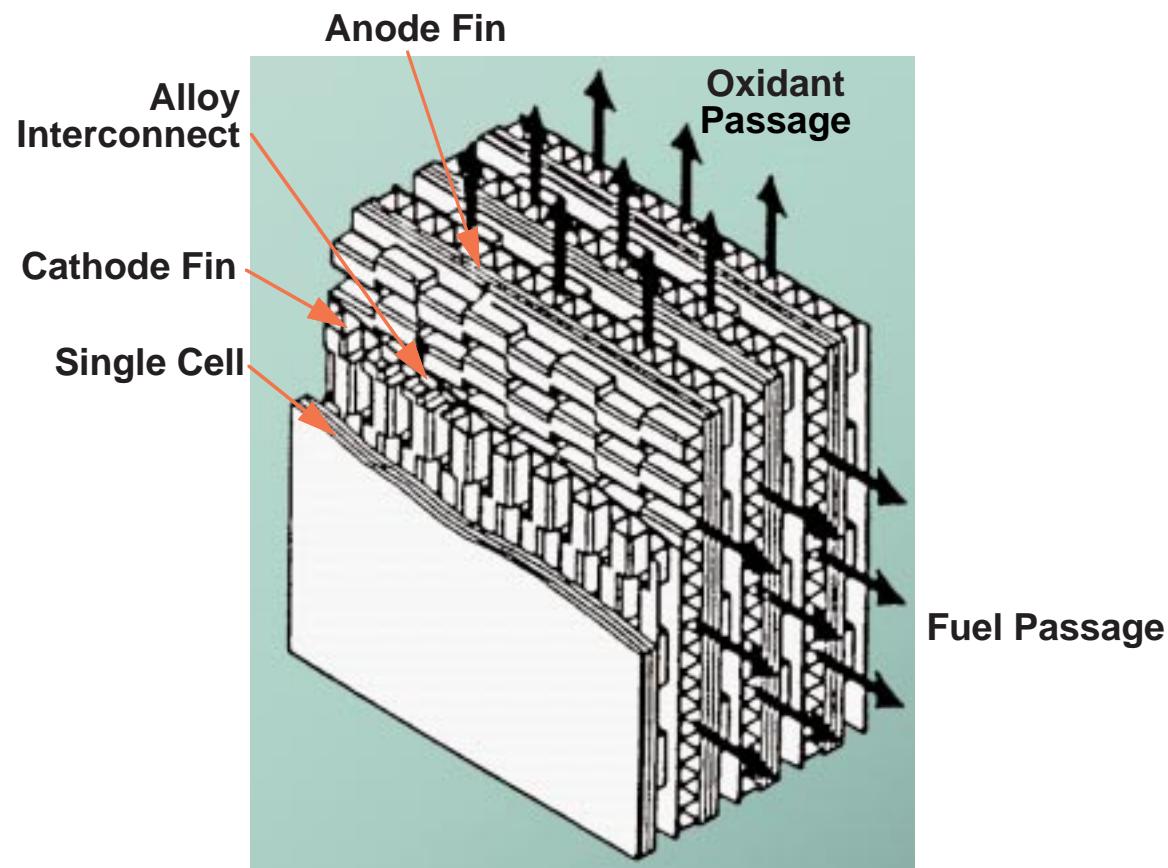
# Tape Calendered SOFC Stack Development

- Approach
  - Reduced - Temperature Operation
  - Tape Calendering for Cell Fabrication
  - Stack Design
    - Low Cost
    - High Performance
    - Light Weight and Compactness
- Funding
  - GRI (Dr. Kevin Krist)
  - DARPA (Drs. Larry Dubois, Robert Rosenfeld, Robert Nowak)

# Reduced-Temperature SOFCs

- **Operating Temperature of 600° - 800°C**
- **Conventional Materials with Thin YSZ Electrolytes**
- **Key Advantages**
  - **Wider Material Choice**
  - **Increased Cell Life**
  - **Reduced Fuel Cell Cost**
  - **Improved Reliability**

# Stack Configuration





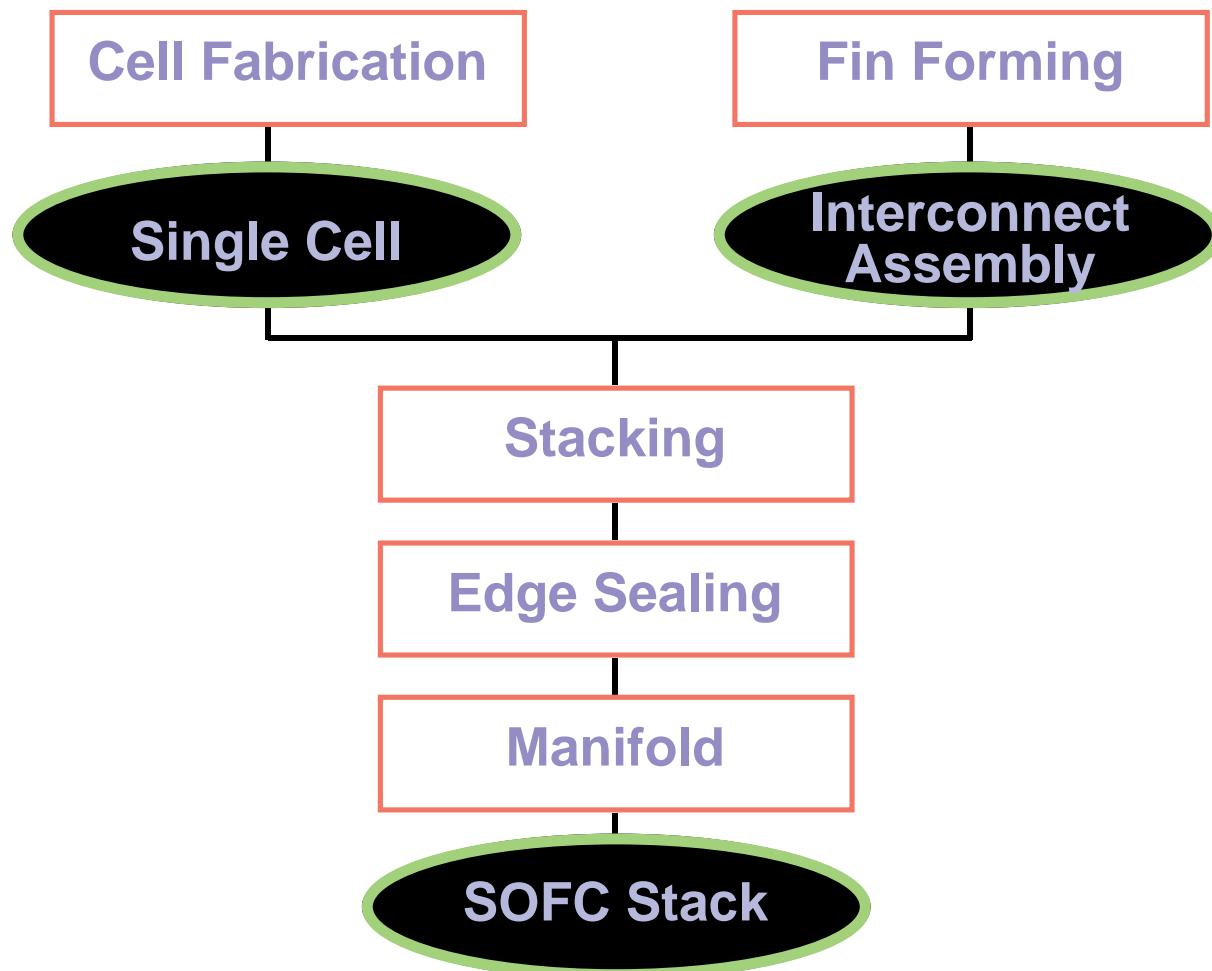
## Design Features

**Thin-Film Electrolytes to Permit Efficient Operation at Reduced Temperatures**

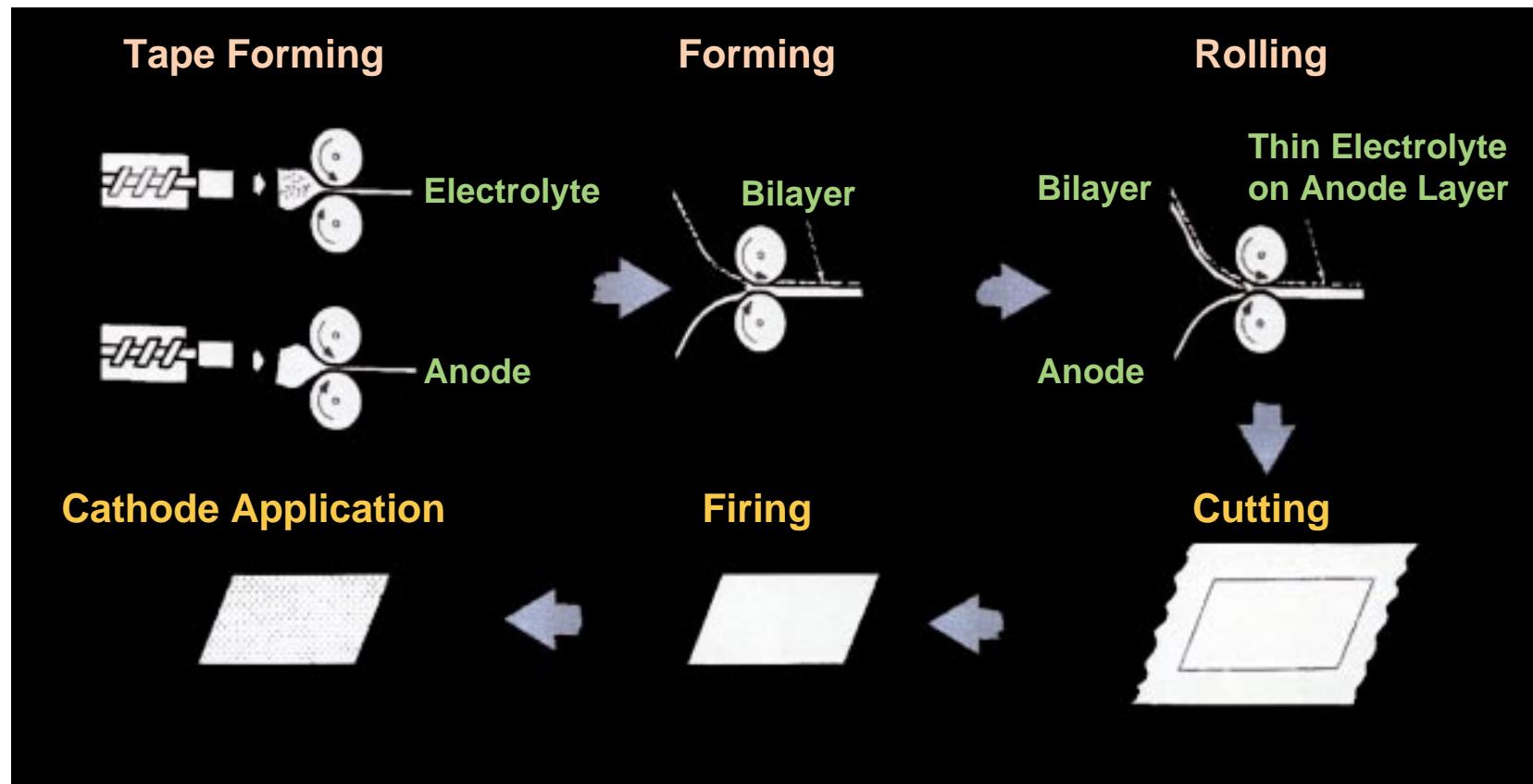
**Lightweight Metallic Structures to Achieve High Power Densities**

**Low-Cost Materials and Fabrication Processes**

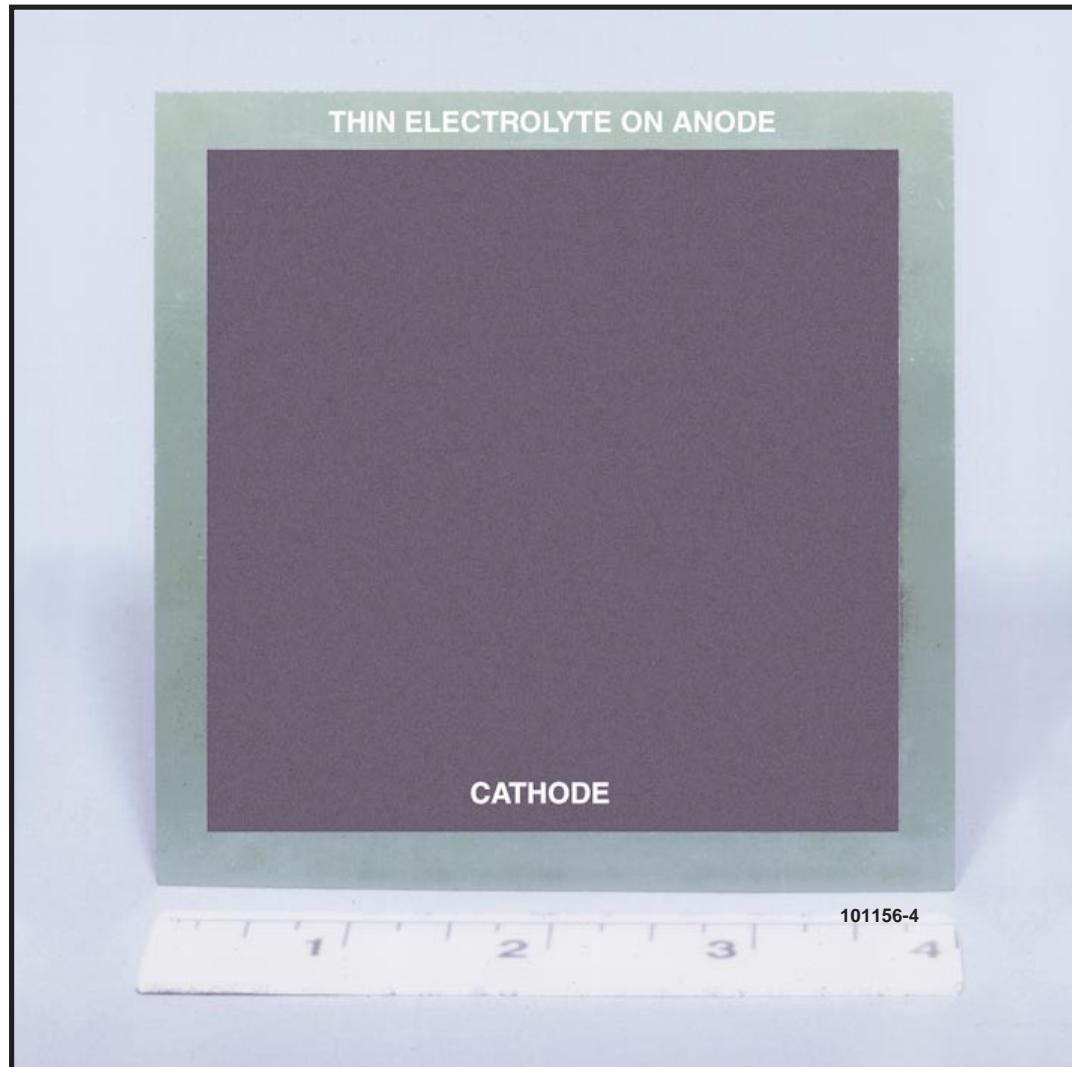
# Stack Assembly Processes



# Thin-Electrolyte SOFC Fabrication Sequence



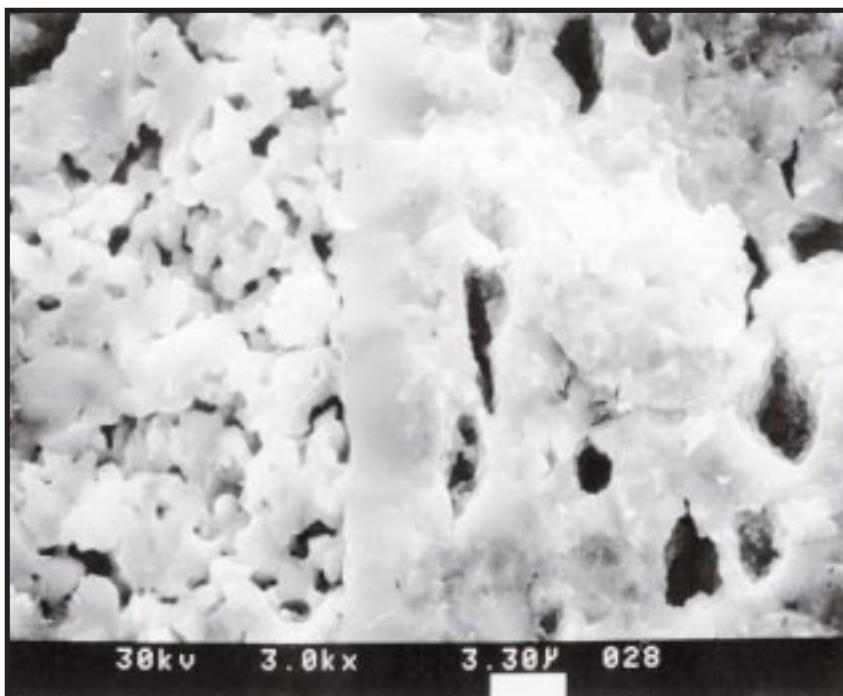
# Photograph of Single Cell



# Thin Electrolyte Cell

# Fracture Surface

# LaMnO<sub>3</sub> Cathode

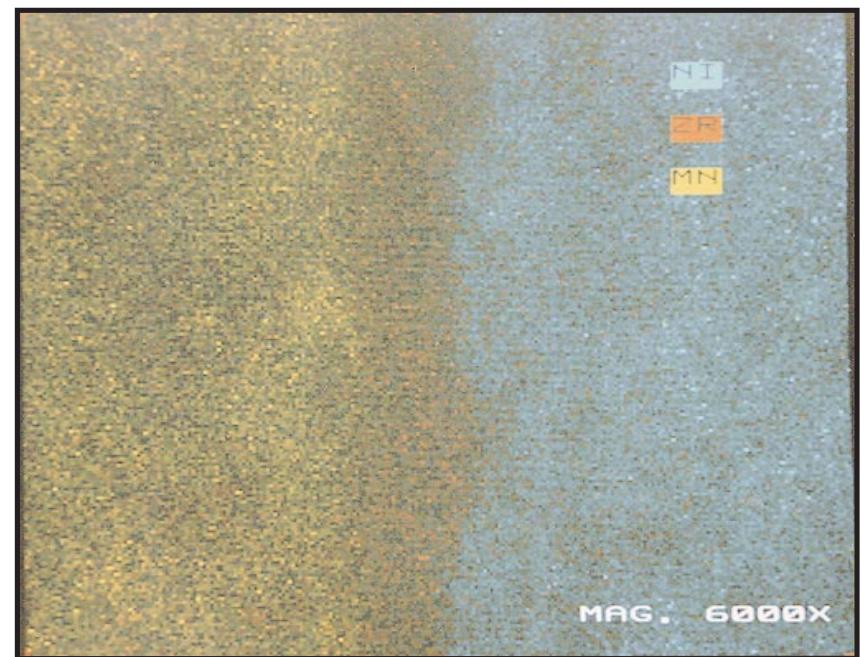


# Element Mapping

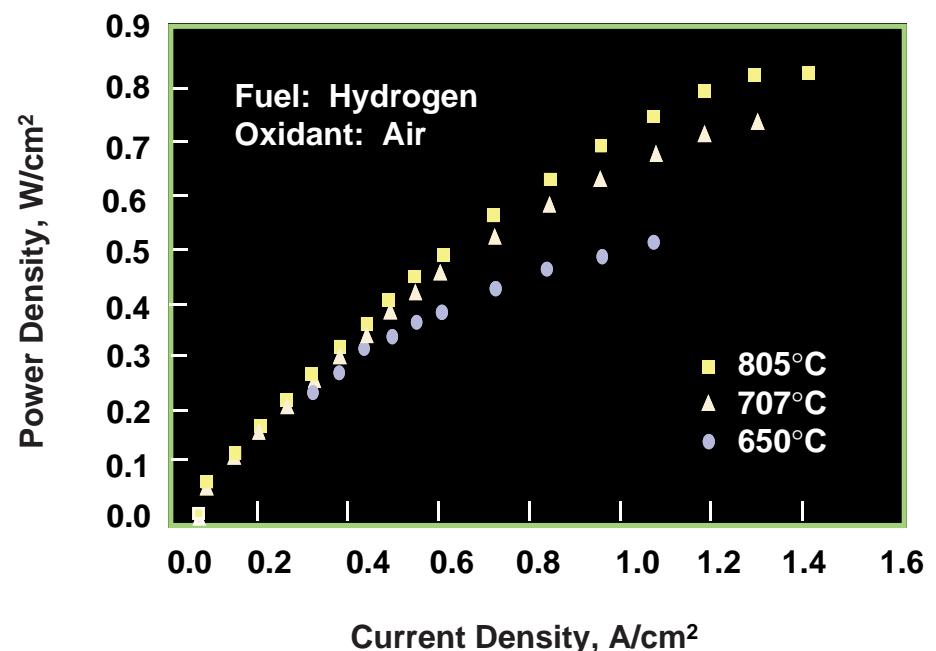
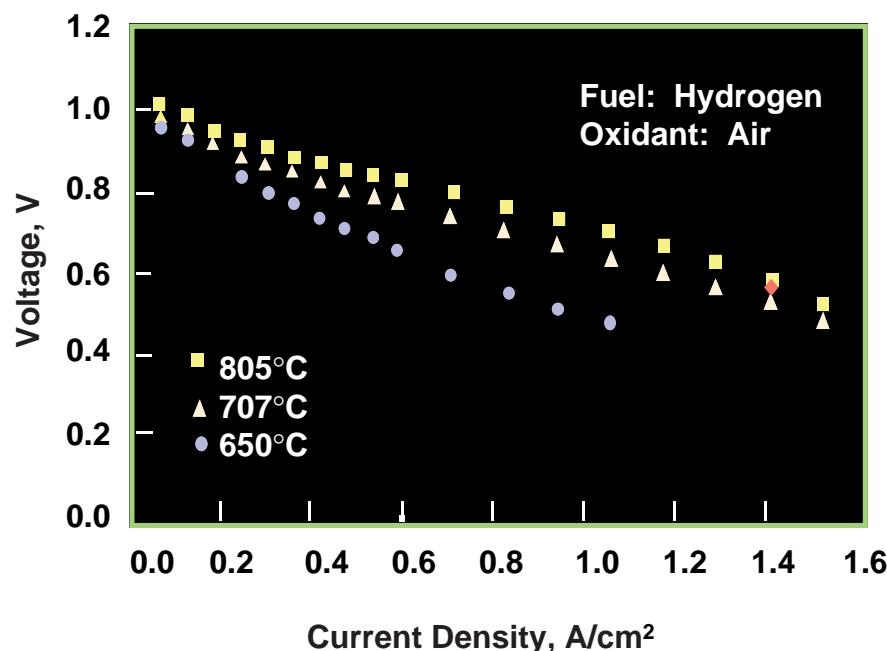
# ZrO<sub>2</sub> Electrolyte

# NiO/ZrO<sub>2</sub>

## Anode

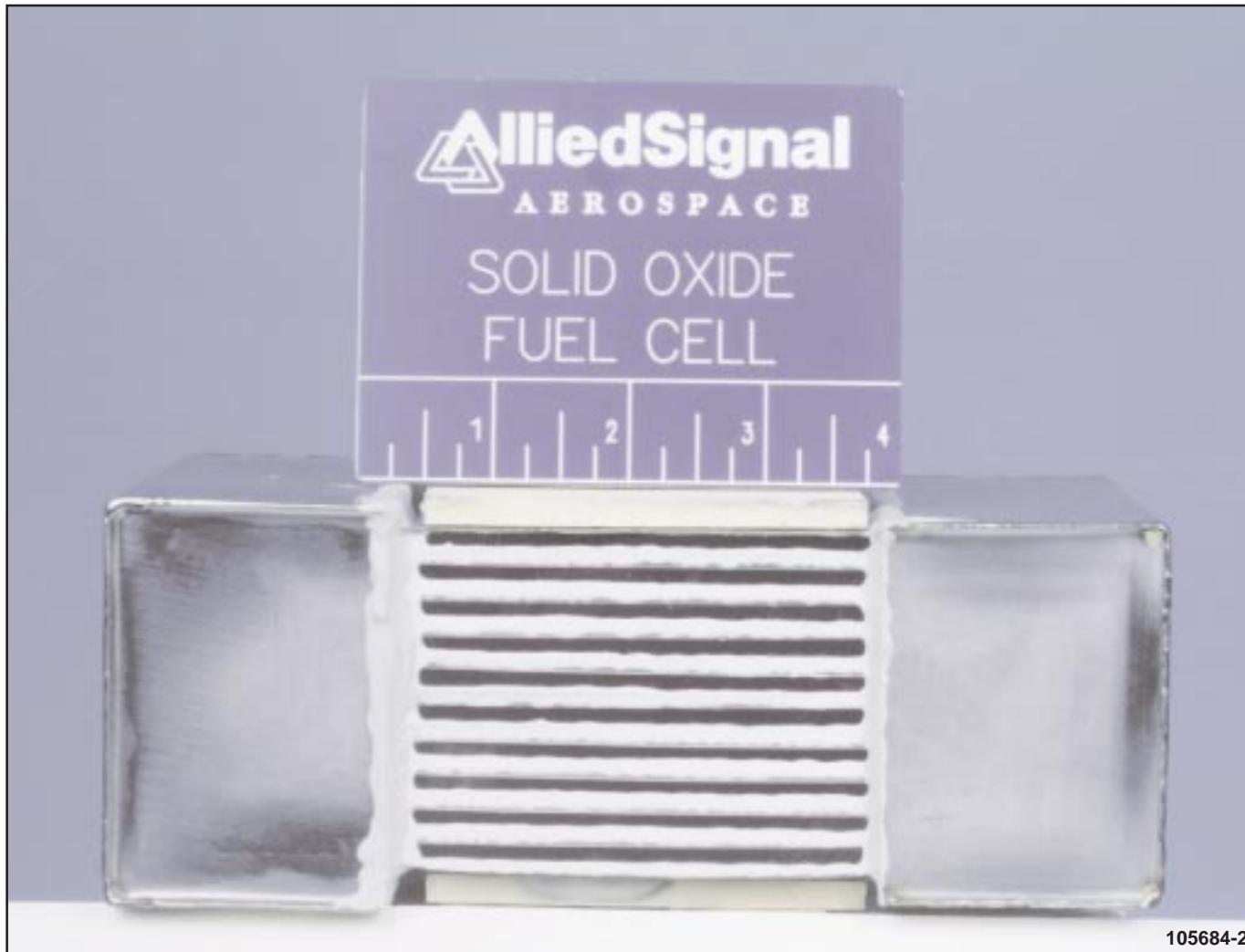


# Performance of Thin-Electrolyte Single Cell

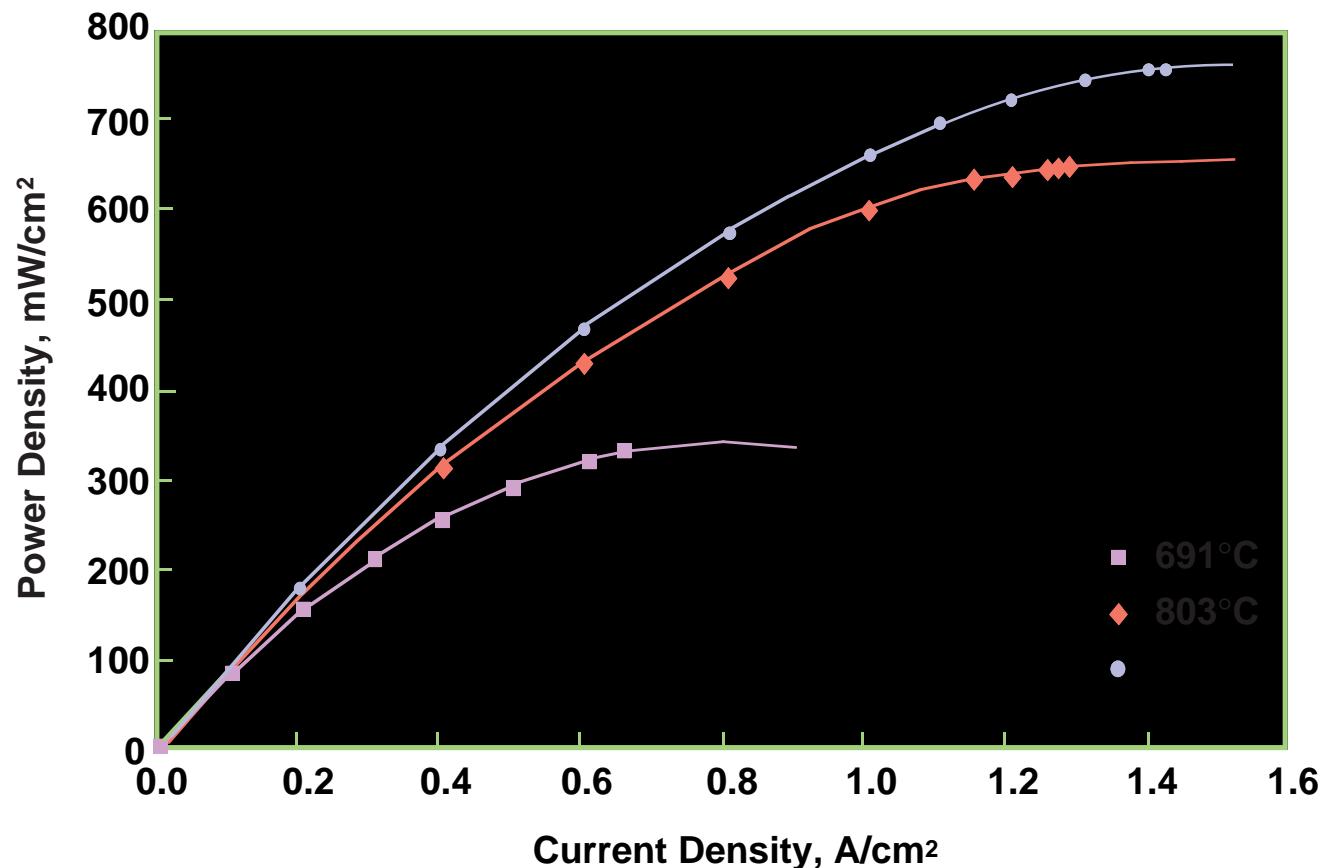




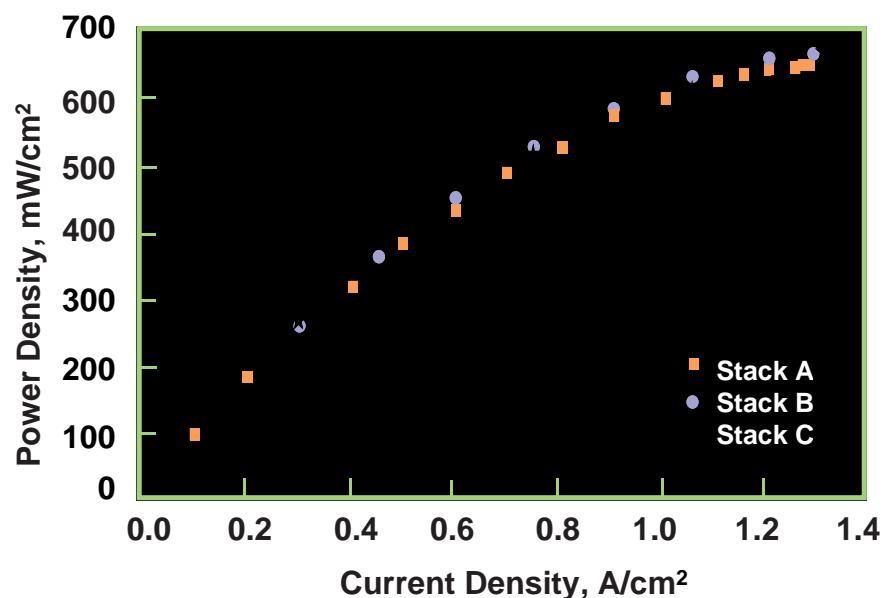
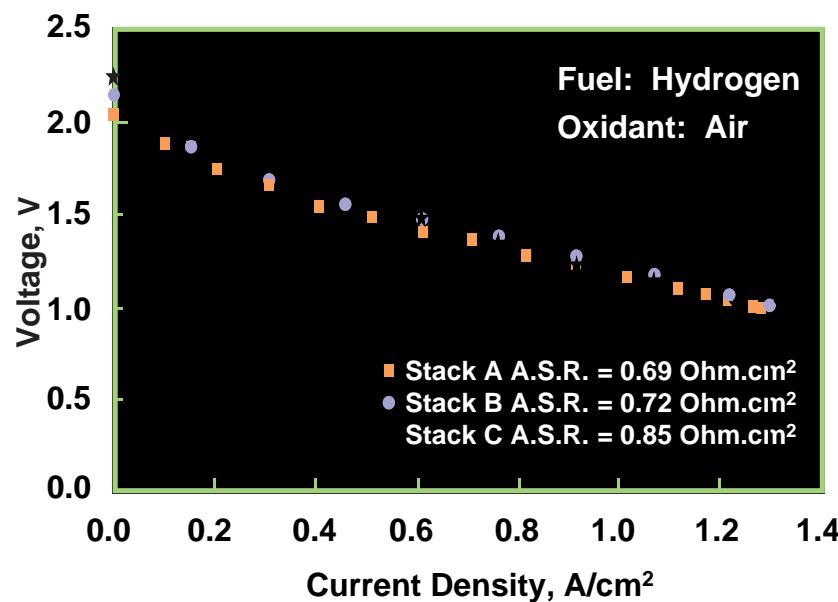
## Ten-Cell Stack



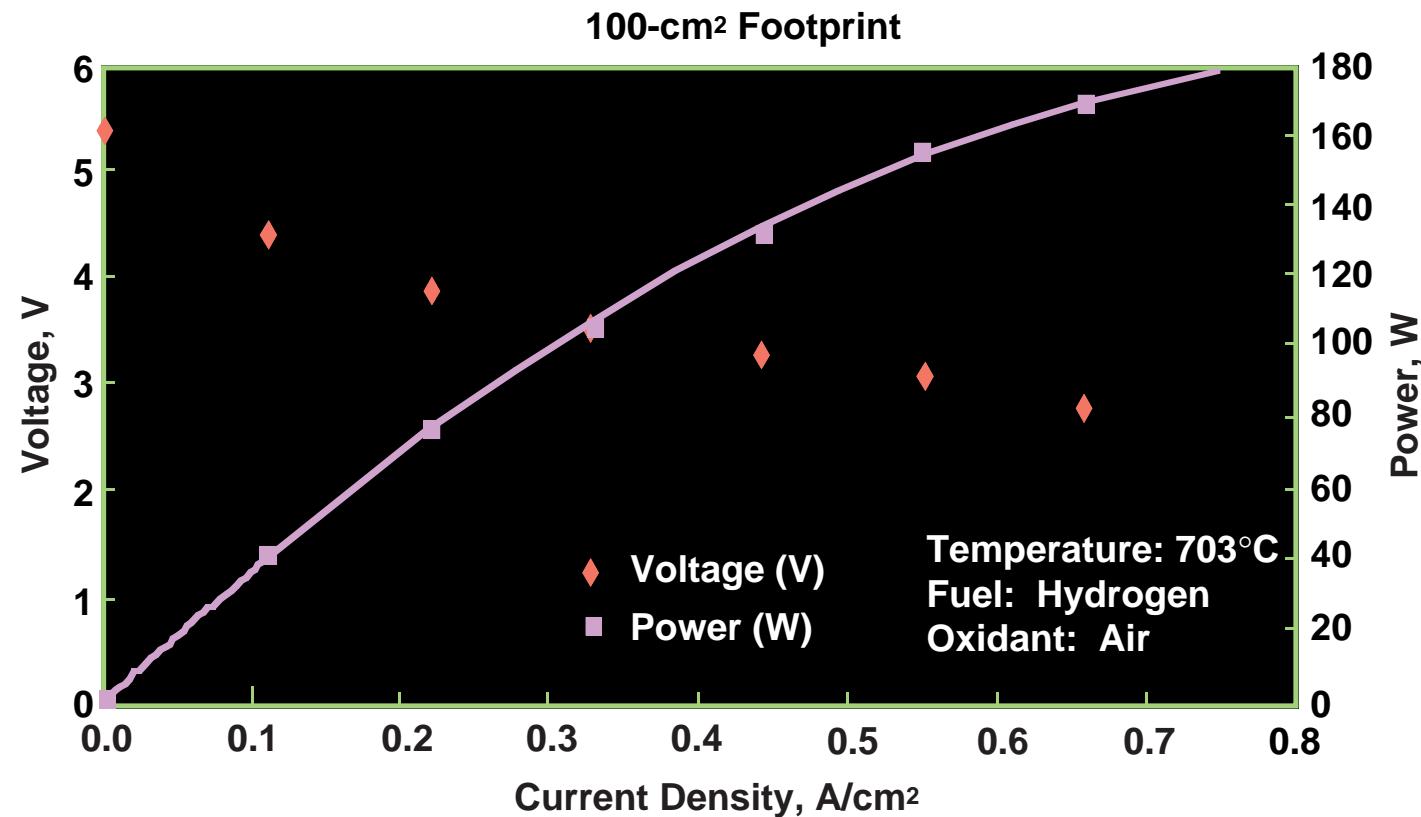
# Power Density Curves of Two-Cell Stack



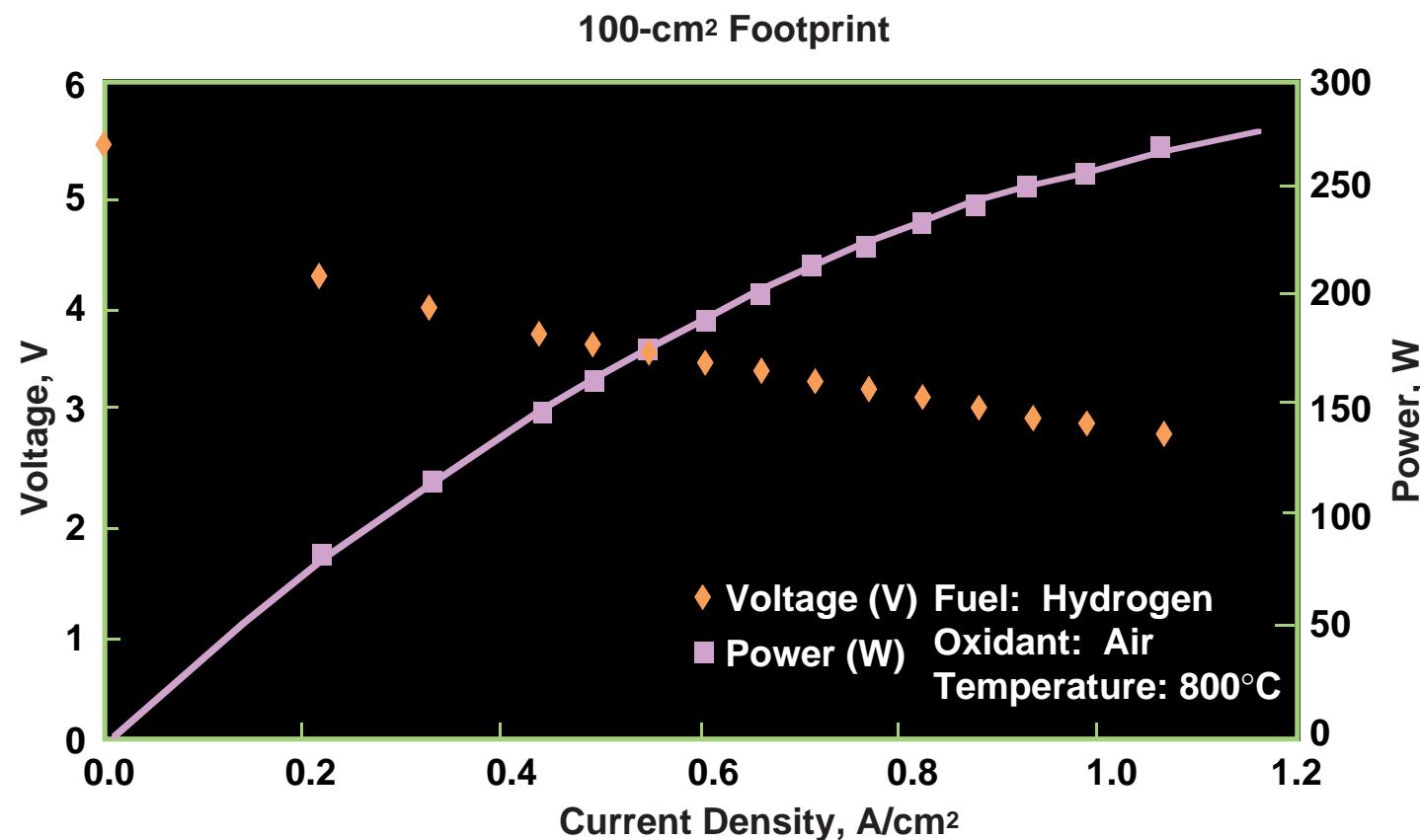
# Performance of Two-Cell Stacks



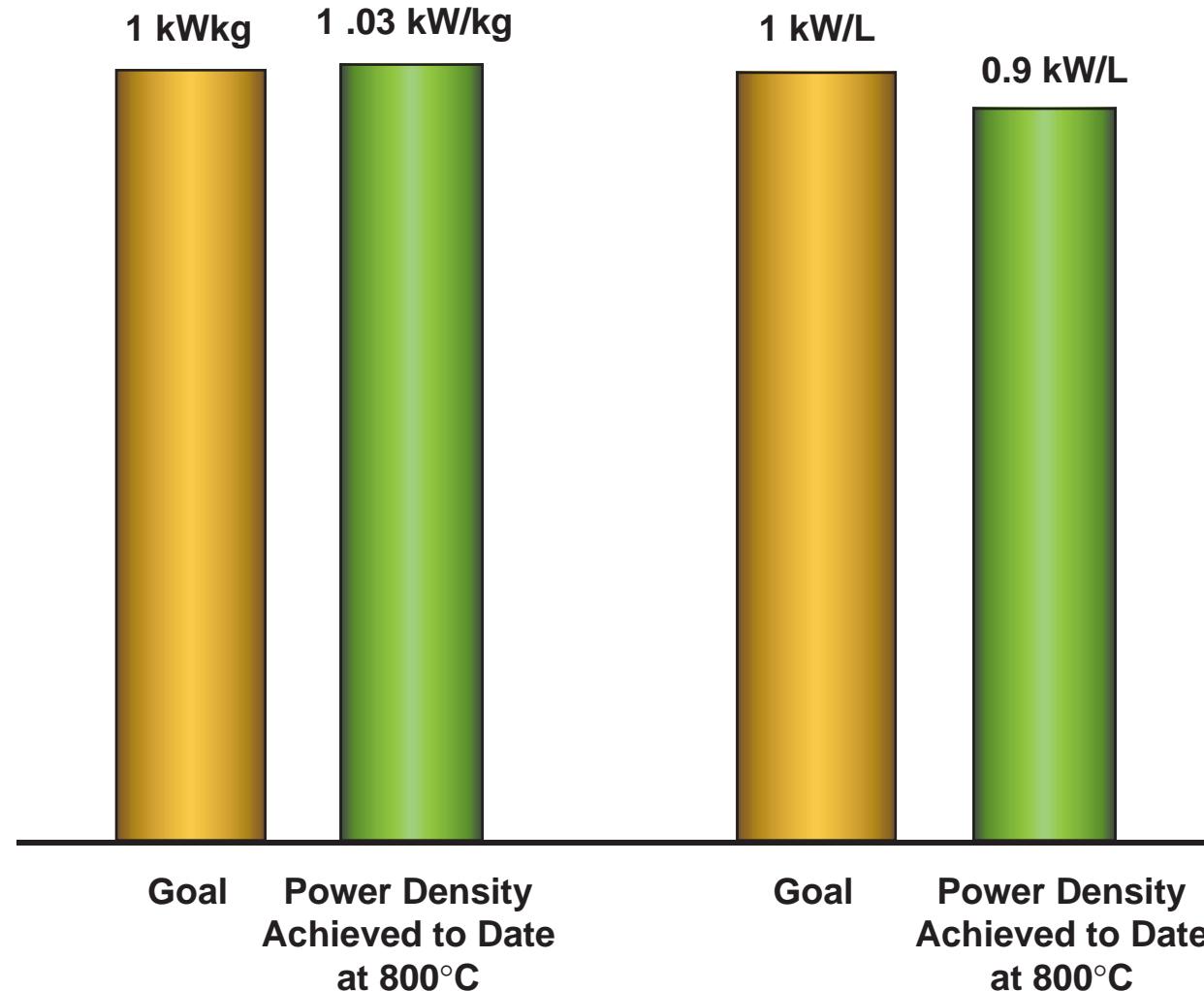
# Performance Curves of Five-Cell Stack



# Performance Curves of Five-Cell Stack



## Stack Power Densities



# Technological Issues

- **Performance Losses in Stacking**
- **Sealing**
- **Thermal Cycling**
- **Life**

## Summary

- **Reduced-Temperature Operation  
Demonstrated (e.g., 5-Cell Stack,  
100-cm<sup>2</sup> Footprint. 270 W at 800°C)**
- **Excellent Stack Performance  
Achieved at Reduced Temperatures  
(e.g., 600 mW/cm<sup>2</sup> at 800°C)**
- **Several Issues to Be Addressed  
(Performance Losses in Stacking,  
Sealing, Thermal Cycling, Life)**

## Solid Oxide Fuel Cells (SOFCs) for the Direct Oxidation of Methane

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### Introduction

This newly funded project is merging the optimization of yttria-stabilized zirconia (YSZ)-based mixed-conducting electrodes with the development of anodes for the direct oxidation of methane. A mixed-conducting YSZ-based electrode has a number of advantages; these include enhanced mechanical and chemical compatibility with the YSZ electrolyte, an increased charge-transfer reaction area resulting in a lower electrode overpotential, and greater tolerances to gas impurities (e. g. sulfur in fuel gases)

Direct oxidation of methane in SOFCs avoids the equilibrium limitations of the steam-reforming reaction for low-temperature operation and the necessity of having a separate steam-reforming reactor. Our ultimate goal in merging developments from these two areas of research is to develop reduced-temperature SOFCs which are robust, inexpensive to fabricate, and can be operated at 600 to 800°C.

### Problems

The work involving mixed-conducting YSZ-based electrodes is at a more advanced stage than the direct-oxidation work. We have demonstrated that the overpotential losses of YSZ-based electrodes can be lower than the electrodes currently used in SOFCs, and that a YSZ-electrolyte cell with YSZ-doped electrodes can exhibit attractive current and power densities. We are now determining the best dopants for optimum cell power densities at reduced temperatures, and are minimizing interfacial resistances and overpotential losses in thin-film YSZ cells having YSZ-based electrodes containing the optimum dopants. We are also collaborating to develop an attractive anode for the direct oxidation of methane.

A key limitation for the direct oxidation of methane, particularly at temperatures below 800°C, is the relatively low reactivity of methane. This is the reason essentially all SOFC designs require that methane first be converted to hydrogen through steam reforming. The question is why can't one carry out the oxidation of methane directly? The rates required in a fuel cell are not extremely high. For a current density of 1 Amp/cm<sup>2</sup>, a porous anode with only 1 cm<sup>2</sup> of catalytic surface area per cm<sup>2</sup> of planar fuel cell (very low for a porous catalyst) would require a turnover frequency of less than 10, a reasonable value for oxidation catalysts.

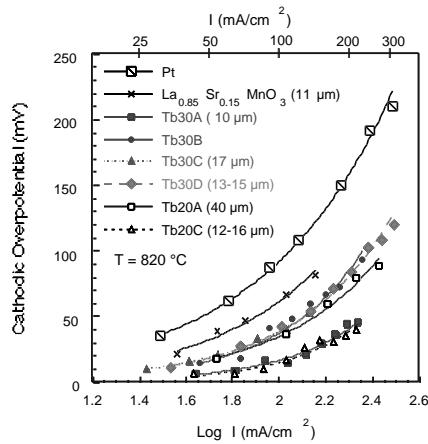
To answer the question posed above, one needs to consider what is actually occurring at the anode in conventional SOFCs and consider the likely reasons why most attempts at direct oxidation of methane have failed. First, while the Ni-zirconia cermet commonly used at the anode is sometimes referred to as an “electrocatalyst”, there is no evidence that it performs this function. The Ni really acts only as a steam reforming catalyst and current carrier. The H<sub>2</sub> formed by steam reforming is so reactive that it can probably be oxidized on the YSZ surface at the typical temperatures used in SOFCs. There is considerable evidence from the literature that oxygen anions from the YSZ electrolyte never migrate to the anode surface when metal catalysts are used. Oxygen anions can diffuse from YSZ to mixed-conducting oxides, such as ceria and bismuth (1); and some success has been achieved for the direct oxidation of methane using these materials. However, no known oxide has achieved acceptable rates.

## Previous Results

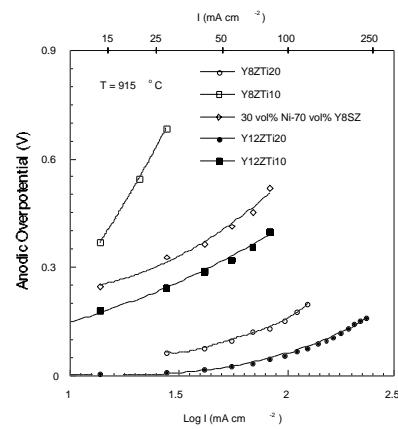
### Mixed-Conducting YSZ-Based Electrodes

In the cathodic environment (air) of a SOFC, the best dopant to produce a YSZ-based electrode is one which provides the highest p-type electronic conductivity with no significant degradation of the oxygen-ion conductivity. We have successfully synthesized mixed-conducting YSZ-based cathodes by dissolving terbia (TbO<sub>1.75</sub>) into YSZ. The Y<sub>8</sub>ZTb<sub>20</sub> (YSZ containing 8 mole % yttria and 20 mole % terbia) and Y<sub>8</sub>ZTb<sub>30</sub> solid solutions have excellent mixed (oxygen-ion and p-type electronic)-conductivities (2, 3). Fig. 1 shows that the overpotentials of six mixed-conducting Y<sub>8</sub>ZTb<sub>30</sub> and Y<sub>8</sub>ZTb<sub>20</sub> cathodes (having different preparation methods and thicknesses) are smaller than those of the similarly prepared platinum and the currently used La<sub>0.85</sub>Sr<sub>0.15</sub>MnO<sub>3</sub> (LSM) cathodes (4, 5). This is believed due to the increased charge-transfer-reaction area for the mixed-conducting Y<sub>8</sub>ZTb cathodes.

We have also dissolved titania in Y<sub>8</sub>SZ and Y<sub>12</sub>SZ to obtain a YSZ-based anode. (6,7). The n-type electronic conductivity of YZTi becomes significant when the oxygen pressure is below  $\sim 10^{-15}$  atm due to the presence of both Ti<sup>+3</sup> and Ti<sup>+4</sup> cations and the associated electron hoping between them in these very low oxygen-pressure environments (7, 8). The n-type electronic conductivity of titanium-doped YSZ increases with decreasing oxygen pressure and with increasing titania concentration and temperature (7, 8). Because typical oxygen pressures at the fuel-gas anode in a SOFC vary from 10<sup>-15</sup> to 10<sup>-22</sup> atm, a YSZ-based anode containing titanium cations should have excellent mixed (oxygen-ion and n-type electronic) conductivity. Fig. 2 shows that the Y<sub>8</sub>/ZTi<sub>20</sub> and Y<sub>12</sub>ZTi<sub>20</sub> anodes have much lower overpotentials than those of a Ni/YSZ cermet anode (4, 7).

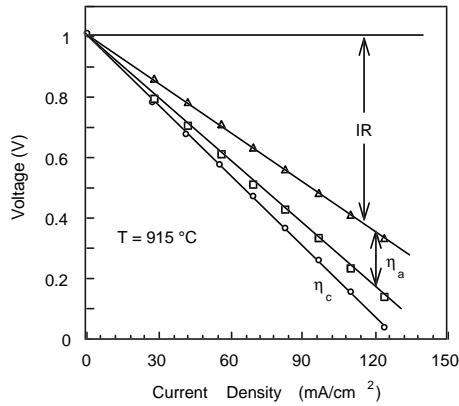


**Fig. 1** Comparison of the overpotentials of the Y8ZTb20 and Y8ZTb30 cathodes with Pt and  $\text{La}_{0.85}\text{Sr}_{0.15}\text{MnO}_3$  cathodes in air.

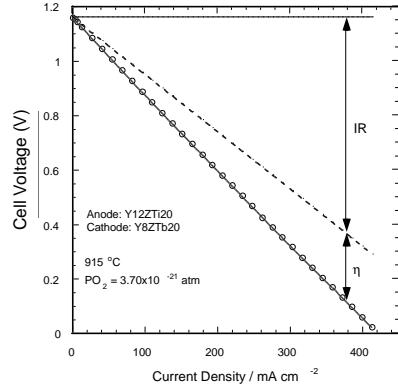


**Fig. 2** Comparison of the overpotentials of the Y8ZTi10, Y8ZTi20, Y12ZTi10, and Y12ZTi20 anodes with that of a Ni(30 vol%)/Y8SZ cermet anode at  $2 \times 10^{-18}$  atm  $\text{PO}_2$

The cell voltage variation with current density for two Y8ZTb/ Y8SZ /Y8ZTi or Y12ZTi cells (4, 7) are shown in Figs. 3 and 4. The improved cell performance shown in Fig. 4 over that shown in Fig. 3 is consistent with the Y12ZTi20 anode overpotentials being lower than those of the Y8ZTi20 anode (as shown in Fig. 2). This research has resulted in two patents (9, 10).



**Fig. 3** Single oxide cell performance at 915°C for the cell: Air, Y8ZTb20 / Y8SZ / Y8ZTi20,  $\text{PO}_2 = 2 \times 10^{-18}$  atm.



**Fig. 4** Cell performance of the single oxide cell: Air, Y8ZTb20 / Y8SZ / Y12ZTi20

The power densities of these cells are not yet comparable to the largest ones reported for thin film (~ 5 -10 mm) YSZ cells, principally because the thickness of our YSZ electrolyte was much larger (~ 1,500 mm). However, our results for single-cells with mixed-conducting YSZ-based electrodes demonstrate that these electrodes can be attractive competitors in the next generation SOFCs. For example, we estimate that the power density of our mixed-conducting YSZ-based electrode cell at 800•C with a thin-film YSZ-electtolyte thickness of 10 mm can be as high as ~ 2.6 W/cm<sup>2</sup>), depending on the thickness and morphology of the electrodes. In obtaining this maximum number, we have assumed that the major cell losses are ohmic ones, which is justified by the lower electrode overpotential ( $\eta$ ) losses illustrated in Figs. 1 and 2. Furthermore, our YSZ-based electrode overpotentials at higher current densities can be reduced by optimizing dopant compositions and morphology. We are also working collaboratively in the develop of new anodes for the direct oxidation of methane.

#### Anodes for the Direct Oxidation of Methane

Automotive, emissions-control catalysts have the properties one is looking for in a SOFC anode. Automotive catalysts contain as much as 40 wt % ceria, with a very small amount of precious metals like Pt, Pd and Rh. The ceria plays the role of oxygen-storage component, a critical component for maintaining the oxygen stoichiometry in the exhaust. To achieve the oxygen-exchange rates needed in automotive applications, ceria must be in intimate contact with the precious metal and it seems clear that oxygen transport between the ceria and the precious metal plays a fundamental part in oxygen storage. For example, in our own work in this area, we have shown that small Rh particles supported on ceria films appear to be oxidized by the ceria in vacuum at temperatures close to room temperature (11). These ideas have obvious application to anode design in SOFCs. High catalytic activity for oxidation of methane in a SOFC anode is not useful if one cannot provide oxygen to the catalyst surface, but the materials used in automotive catalysis provide both high catalytic activities and high oxygen transfer capabilities.

We have used anodes consisting of samaria-doped ceria (SDC), of varying thickness (ie. varying catalytic surface area), with and without Rh added as a dopant (12). The current carrier in our test cells was a Au mesh attached directly to the YSZ electrolyte. Two key features of this cell are noteworthy. First, the catalytic and current carrying functions in our test cell were separate. Au was used to carry current in our studies simply because of its inert catalytic properties. Second, by placing the current carrier next to the electrolyte, electronic and ionic resistances in the anode due to changing the thickness of the catalyst should not strongly affect the performance.

The results obtained using hydrogen as a fuel were largely independent of either anode thickness or presence of the metal at 800•C. The current-voltage characteristics were essentially identical on all four cells tested, even when the thickness of the SDC was increased by a factor of 10, whether or not Rh was added as a dopant. Because the electrolyte in our cells was relatively thick (1.7 mm) it appears that the performance was completely limited by oxygen-ion conductivity in the YSZ. The same was not true when dry methane was used as a fuel. First, increasing the thickness (surface area) of the SDC by a factor of 10 had the effect of increasing the open-circuit voltage from ~0.2 volts to 0.9 volts, as well as increasing the maximum current density from 2 mA/cm<sup>2</sup> to ~10

$\text{mA/cm}^2$ . Both performance measures were less than that obtained using hydrogen as a fuel, where the maximum current density was greater than  $30 \text{ mA/cm}^2$ . More interesting, when Rh was added to the anodes by impregnation of  $\text{Rh}(\text{NO}_3)_3$ , the performance of the cells changed dramatically. Independent of the SDC thickness, the maximum current density and power generation increased to the level found using hydrogen as a fuel.

The above results demonstrate several points but also raise several questions. For the SDC anodes without Rh, it appears that catalytic activities of the anode limited the performance of the cell. With the addition of Rh as a dopant, however, it appears that the performance was no longer limited by the anode. Clearly, further work is needed to determine the point at which anode catalytic properties will limit performance. The results do, however, prove that direct oxidation of methane is feasible with optimized anodes.

### Future Work

Future work will be focussed towards optimising mixed-conducting YSZ-based electrodes and developing anodes for the direct oxidation of methane. Concepts from both areas will be incorporated in developing SOFCs for the direct oxidation of methane.

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# LOW TEMPERATURE, HIGH PERFORMANCE, PLANAR SOLID OXIDE FUEL CELLS AND STACKS

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## ABSTRACT

Conversion of chemical energy of combustion of a fuel into electrical energy by fuel cells continues to be an important thrust area of energy conversion technology. Among these, fuel cells using either molten salt (MCFC) or solid oxide (SOFC) electrolytes are of particular interest because operation at higher temperatures allows for the use of natural gas as a fuel. High temperature fuel cells, however, are subject to materials-related problems such as corrosion which increase with temperature. In addition, the Nernst potential also decreases with increasing temperature. These two factors suggest that lowering of the operation temperature is preferred. However, higher operating temperatures are desirable for internal reforming as well as to minimize losses at electrolyte/electrode interfaces. Based on these considerations, a temperature range between ~600 and ~800°C is considered optimum. The focus of the present work has been on the development of solid oxide fuel cells made of thin electrolyte films supported on a relatively thick anode.

Anode-supported single cells of approximately 3 cm diameter with anode thickness of ~0.75 mm (750  $\mu$ m), YSZ electrolyte thickness of ~10  $\mu$ m, and LSM + YSZ cathode thickness of ~50  $\mu$ m were fabricated. The cell fabrication procedure consists of depositing a thin film of YSZ by dip-coating on a powder compact of a mixture of nickel oxide and YSZ. Densification was achieved at a temperature below 1400°C. Single cells were tested between 650 and 800°C with humidified hydrogen as the fuel and air as the oxidant. Maximum power density at 800°C was

1.8 W/cm<sup>2</sup> (area specific resistance ~0.15 cm<sup>2</sup>) and that at 650°C was 0.8 W/cm<sup>2</sup> (area specific resistance ~0.34 cm<sup>2</sup>). The area specific resistance of the cells obeyed Arrhenius-type behavior with an activation energy of 50 kJ/mol. The observed behavior is in accord with the theoretical analysis of cell performance which takes into account transport of gaseous species through the porous electrodes.

For stack testing, square-shaped cells of dimensions 5 cm x 5 cm x ~2-3 mm were made. Four cell stacks with metallic interconnects were tested at 800°C. The interconnect was configured

from a commercial alloy foil of 5 mil thickness. The interconnect was subjected to surface treatments to improve its performance. No glass seal was used. The edges of the metallic interconnects serve as sealing gaskets. Manifolds were made of commercial alloys. The manifolds are spring-loaded against the stack with electrically insulating gaskets. The absence of a sealing glass allows one to disassemble the stack and reuse the same cells. Often, the same cells were used more than ten times. Also, the presence of a large amount of nickel in the anode renders the cells highly thermal shock-resistant. For example, a cell could be cooled from 800°C to room temperature within a few minutes. The typical repeat unit (anode-electrolyte-cathode-interconnect) area specific resistance was measured to be  $\sim 0.5$   $\text{cm}^2$ . The cells used in this preliminary work were of a thickness much greater than the most optimum. Current efforts are directed towards lowering the thickness down to  $\sim 1.5$  mm.

Funded by Electric Power Research Institute (EPRI), Gas Research Institute (GRI), and the State of Utah.

## I. INTRODUCTION

Fuel cells for direct conversion of chemical energy of a fuel into electrical energy is an important thrust area of energy conversion technology [1-7]. Among these, fuel cells using either molten salt or solid oxide electrolytes operate at high temperatures which allows for the use of natural gas as a fuel unlike low temperature PEM fuel cells which must use hydrogen. High temperature fuel cells, however, are not without drawbacks, the foremost being materials-related problems such as corrosion and/or oxidation. From the standpoint of direct utilization of natural gas as a fuel, there are two conflicting considerations. (1) Higher operating temperatures are preferred for internal reforming as well as for minimization of electrolyte/electrode interfacial losses, and (2) Lower operating temperatures are preferred for attaining a higher thermodynamic efficiency in addition to minimizing corrosion/degradation-related problems. Based on these factors, a temperature range between  $\sim 600$  and  $\sim 800$  °C is considered to be optimum for high temperature fuel cells.

Much work has been reported on solid oxide fuel cells (SOFCs) over the past two decades. The commonly used solid electrolyte in SOFCs is yttria-stabilized zirconia (YSZ) which exhibits excellent stability and remains essentially a pure ionic conductor even in a reducing environment. At a given temperature, ionic conductivity of YSZ is lower than that of bismuth oxide or ceria which necessitates higher operating temperatures. Recently, however, there has been considerable effort devoted towards developing SOFCs for operation at temperatures lower than 800°C. If YSZ is used as the electrolyte, it is imperative that its thickness be made as small as possible. A thickness on the order of 10  $\mu\text{m}$  is considered reasonable. Thin YSZ films, however, are not strong enough to be self-supporting and must be supported on either a porous anode or a cathode. In such cells, the ohmic contribution to the cell resistance can be made small. However, it is necessary that gas transport through the porous electrodes does not significantly add to the overall cell resistance. Thus, in electrode-supported cells, the electrode structure is an important design parameter.

The design of a high performance SOFC requires careful attention to the following parameters: (1) Ohmic contribution of the electrolyte and the electrodes, (2) Charge transfer or activation polarization, and (3) Mass transport through porous electrodes or concentration polarization. In SOFCs employing a thick electrolyte (typically  $> 150$   $\mu\text{m}$ ) as the supporting member, the ohmic contribution is usually a significant part of the overall cell resistance. In addition, the activation overpotential may also be substantial. By contrast, in electrode-supported (either an anode or a cathode as the supporting structure) cells, a thin electrolyte ( $\sim 10$  to 30  $\mu\text{m}$ ) is

deposited directly on a porous electrode. Thick porous electrodes additionally lower the activation overpotential or charge transfer resistance [9-11]. However, concentration polarization can be significant due to resistance to gas transport through porous electrodes. Thus, electrodes must be designed to minimize both activation as well as concentration polarizations through an analysis of charge transfer processes at electrode/electrolyte interfaces and transport of gases through porous electrodes. Such a design strategy was used in the present work for developing cells which are sufficiently thick to ensure excellent strength and durability, while still retaining high performance that can be achieved with thin, fragile cells. Excellent performance at the cell level is a necessary requirement for realizing high performance at the stack level, but by no means is sufficient. Issues regarding the type of interconnect used, interconnect properties concerning oxidation, and stacking strategy are equally important. In this work, efforts were also directed towards developing planar stacks using metallic interconnects.

## II. EXPERIMENTAL PROCEDURE

**Cell Fabrication:** NiO and 8 mol.% yttria-stabilized zirconia (YSZ) powders obtained from commercial sources were mixed in desired proportions in ethanol for 24 hours. For single cell tests, discs of ~32 mm in diameter were die-pressed in a uniaxial press followed by isostatic pressing at pressures up to 200 MPa. For stack tests, square plates of dimensions 6 cm x 6 cm were pressed. Discs and plates were coated using a slurry of YSZ in ethanol, and later sintered in air at 1400°C for 1 hour. LSM powder of stoichiometry  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{(3-)}^{}$  was prepared by calcining a mixture of  $\text{La}_2\text{O}_3$ ,  $\text{SrCO}_3$ , and  $\text{MnO}_2$  in air at 1000°C. The calcined LSM powder was subsequently ball-milled. A mixture containing LSM and YSZ powders in equal amounts (by weight) was made and mixed with an organic liquid to form a paste. The paste was applied over the sintered discs (on the dense YSZ layer). The discs were then heated to 400°C to burn out volatiles. This procedure was repeated until a layer of 50 to 70  $\mu\text{m}$  thickness was formed. The discs were later heated to 1250°C for 1 hour. This thermal treatment was adequate to ensure the formation of a good bond between the porous LSM + YSZ cathode and the underlying dense YSZ layer, but not so severe as to form highly resistive pyrochlore phase,  $\text{La}_2\text{Zr}_2\text{O}_7$ . After electrochemical tests, the cells were fractured and examined under a scanning electron microscope. Selected samples of anodes were made with two types of YSZ powders; one containing 3 mol.%  $\text{Y}_2\text{O}_3$  (tetragonal zirconia, TZP) and the other containing 8 mol.%  $\text{Y}_2\text{O}_3$  (cubic zirconia). The objective was to determine flexural strength. Since the cells in the present work are anode-supported, it is important to ensure that the anode exhibits adequate strength. Samples containing varying amounts of NiO and zirconia were fabricated by conventional die-pressing, pressureless sintering in air followed by reduction in a hydrogen atmosphere at 800°C. Flexural strength was measured in four point bending.

**Single Cell Testing:** The single cell test stand consists of two alumina tubes in between which a cell can be secured. For single cell testing, disc-shaped cells were used. Silver meshes were used as current collectors which were pressed against the cathode and the anode with a thin layer of platinum resinate in between to ensure a good bond between the silver meshes and the electrodes. The cell was secured in two inconel bushings with an electrically insulating gasket. Silver current leads were affixed to the two silver meshes. The cell was placed inside a furnace. The test stand was designed in such a way that the seals (gaskets) were always under a spring loading with springs outside the furnace. Thus, no glass seal was necessary. This permitted repeated heating/cooling of the cells and retesting. Hydrogen as a fuel was bubbled through water maintained at a temperature ~33°C and air was circulated past the cathode. Temperature at the cell was varied between 650 and 800°C. Reduction of NiO into Ni was accomplished in-situ. Cell performance was measured using an electronic load.

**Stack Testing:** Several four cell stacks were assembled using metallic (superalloy) interconnect. Typical procedure for stack assembly and testing was as follows. End plates, which served as current collectors, were also made of a superalloy. The diameter of the current collector rods was 1.27 cm. Three voltage probes were introduced, one each attached to an interconnect. The stack was secured inside a metallic manifold with inert, insulating gaskets as edge seals. In order to improve the sealing, the stack was spring-loaded wherein the springs were outside the hot zone of the furnace. Stack was tested at 800°C with humidified hydrogen as the fuel and air as the oxidant. Reduction of NiO to Ni was achieved in-situ. The active area of the stack was estimated to be between 75 and 80 cm<sup>2</sup>.

### III. RESULTS AND DISCUSSION

**Mechanical Properties of the Anode:** In order that the cells can be handled without breakage, assembled in a stack, and survive during operation, it is imperative that at least one of the components comprising the cell, i.e., cathode, anode, or electrolyte must be of a sufficient thickness. The preferred choices include either the cathode as a support or the anode as a support, but not the electrolyte since the ohmic contribution is substantial, especially with YSZ as the electrolyte and at temperatures below about 950°C. Figure #1 shows the results of strength vs. nickel content of zirconia + nickel bar-shaped samples made with 3 mol.% Y<sub>2</sub>O<sub>3</sub>-doped ZrO<sub>2</sub> (TZP) and 8 mol.% Y<sub>2</sub>O<sub>3</sub>-doped ZrO<sub>2</sub> powder. The numbers in parentheses indicate the porosity. Note that the strength of Ni + TZP is considerably higher than that of Ni + YSZ. Also, it is observed that the strength of Ni + TZP decreases with increasing Ni content while that of Ni + YSZ increases over the range studied. The possible reason is that the intrinsic strength of Ni may be greater than that of YSZ but less than that of TZP. A few samples of LSM with 25% porosity were also tested for strength. Strength of LSM was measured to be about ~50 MPa. Note that the strength of Ni + zirconia is considerably greater than that of LSM. Since the ionic conductivity of YSZ is greater than that of TZP, and that YSZ is not sensitive to low temperature strength degradation, unlike TZP, in the present work anodes were made of Ni + YSZ.

**Issues Concerning Delamination:** From the standpoint of mismatch of thermal expansion stresses, it would appear the choice should be LSM as the support structure. This is because the coefficient of thermal expansion of YSZ is better matched with LSM instead of Ni + zirconia. Fortunately, YSZ has a lower coefficient of thermal expansion compared to Ni + zirconia. Thus, the YSZ film at room temperature is under compression and thus should not crack. It can, however, delaminate. The tendency for delamination is a function of  $\alpha_2 - \alpha_1$ , where  $\alpha$  is the difference in coefficients of thermal expansion and  $T$  is the temperature differential from the temperature at which stresses are zero (stress relief temperature) and the test temperature, and the thickness of the film. It can be shown that the thinner the film, the less likely it is to delaminate [8,9]. In addition, at the operating temperature  $T$  is rather small. Propensity for delamination can be assessed via a schematic shown in Figure #2. Since the cathode is highly porous, its effect on the delamination calculations can be ignored in a first order calculation. It can be shown that delamination due to thermal stresses can occur provided [8,9],

$$(\alpha_2 - \alpha_1)(T_i - T_o) = \frac{\sqrt{2 \frac{f_m}{t} (1 - \frac{\alpha_2}{\alpha_1})}}{t E_1} \quad (1)$$

where  $\alpha_2$  and  $\alpha_1$  are thermal expansion coefficients of the Ni + YSZ anode and YSZ film, respectively,  $T_i$  is the temperature above which stress relief by creep can readily occur,  $T_o$  is room temperature (for delamination at room temperature) or service temperature (for delamination at service temperature),  $f_m$  is the interfacial fracture energy,  $\alpha$  is the Poisson's

ratio of YSZ (film),  $E_l$  is the Young's modulus of YSZ, and  $t$  is the YSZ film thickness. Alternatively, YSZ films of a thickness smaller than a critical thickness,  $t_{cr}$ , given by

$$t_{cr} = \frac{2 \frac{f_m}{E_l} (1 - \frac{2}{T})^2}{(1 - \frac{2}{T})^2} \quad (2)$$

will not delaminate. The preceding assumes that the thickness of the Ni + YSZ anode is much greater than that of the YSZ film, i.e.  $d > t$ . Based on the known values of thermal expansion coefficients, elastic properties of nickel and zirconia, and an assumed interfacial energy of 20 J/m<sup>2</sup>, it is estimated that  $t_{cr}$  is at least on the order of 30 to 40  $\mu\text{m}$  for volume fractions of nickel and YSZ in the anodes of the present work. Thus, as long as the YSZ film thickness is smaller than 30 to 40  $\mu\text{m}$ , delamination is not expected at room temperature. In the present work, typical thickness of the YSZ film was 10 to 15  $\mu\text{m}$ . No delamination has been observed, in accord with calculations.

Issues Concerning Electrode Thickness: Effective Charge Transfer Resistance: Both electrodes in the present work consisted of a mixture of an electrocatalyst (Ni for the anode, LSM for the cathode) and YSZ. It is known that composite electrodes exhibit lower overpotentials [10-12]. The presence of YSZ in addition to the electrocatalyst increases the three phase boundary (TPB) length, provides parallel path for oxygen ions and thereby spreads the reaction zone into the electrodes. Recent work has shown that the effective charge transfer resistance can be given in terms of the various parameters of the electrodes [12]. Figure #3 shows a schematic of a cell in which porous composite electrodes are shown separated by a dense YSZ electrolyte membrane. The effective charge transfer resistance,  $R_{ct}^{eff}$ , is given as a function of electrode thickness,  $h$ , by [12]

$$R_{ct}^{eff} = \frac{BR_{ct}}{\frac{1 + e^{-\frac{h}{2}}}{1 + e^{-\frac{h}{2}}} (1 - \frac{h}{2})Be^{-\frac{h}{2}} + \frac{1 + e^{-\frac{h}{2}}}{1 + e^{-\frac{h}{2}}} (1 - e^{-\frac{h}{2}}) + B} \quad (3)$$

where

$$= \sqrt{\frac{1}{i} (1 - \frac{h}{2}) R_{ct}} \quad \text{and} \quad = \frac{i R_{ct} -}{i R_{ct} +} \quad (4)$$

It can be shown that as  $h \rightarrow 0$ , the  $R_{ct}^{eff}$  is given by [12]

$$R_{ct}^{eff} \approx \sqrt{\frac{BR_{ct}}{\frac{1}{i} (1 - \frac{h}{2})}} \quad (5)$$

Equation (3) shows that the  $R_{ct}^{eff}$  depends upon a number of parameters including the electrode thickness,  $h$ , in a complicated manner. However, the asymptotic limit given by equation (5) as  $h \rightarrow 0$  has a simple appearance. It is seen that  $R_{ct}^{eff}$  exhibits an inverse square root dependence on the ionic conductivity,  $\sigma_i$ , of the electrolyte (YSZ) part of the electrolyte, square root dependence on the intrinsic charge transfer resistance,  $R_{ct}$ , and square root dependence on the microstructural parameter,  $B$ , of the electrode. Calculations show that typically the asymptotic limit is reached in about 30 to 100  $\mu\text{m}$  of thickness,  $h$ . Calculations further show that for an electrode of  $h = 50 \mu\text{m}$ , with an  $R_{ct} = 2.4 \text{ cm}^2$ , it is possible to achieve  $R_{ct}^{eff}$  values on the order of 0.1  $\text{cm}^2$ , i.e., over twenty times reduction in charge transfer resistance. These considerations were used in the design of electrodes.

Issues Concerning Electrode Thickness: Concentration Polarization: Mass transport of gaseous species through the electrodes was carefully analyzed in order to achieve the desired porosity and electrode morphology. These considerations were important in designing electrodes to minimize concentration polarization.

Single Cell Testing: Typical thicknesses of cathode, electrolyte, and anode were respectively  $\sim 50 \mu\text{m}$ ,  $\sim 10 \mu\text{m}$ , and  $\sim 750 \mu\text{m}$ . The anode porosity was measured to be 38 vol.%. Cathode porosity was not measured on this cell but was estimated to be about 30 vol.% based on measurements made on thicker LSM + YSZ cathodes made separately. Figure #4(a) shows the results on one of the cells in which the voltage is plotted vs. current density at four temperatures; 650, 700, 750, and  $800^\circ\text{C}$ . The short circuit current density at  $800^\circ\text{C}$  was in excess of  $5.5 \text{ A/cm}^2$  and that at  $650^\circ\text{C}$  was in excess of  $3 \text{ A/cm}^2$ . The voltage vs. current density trace exhibits an initial concave-up curvature followed by an approximately linear behavior, followed by a convex-up curvature at higher current densities. This trend was particularly more pronounced at higher temperatures. Slopes of linear regions of the voltage vs. current density plots were estimated at the four temperatures. This slope yields effectively the area specific cell resistance,  $R_{\text{cell}}$ , predominantly the ohmic contribution. The  $R_{\text{cell}}$  should contain at a minimum the area specific resistance of the electrolyte,  $\rho_t$ , where  $\rho_t$  which is

$= \frac{1}{\rho_t t}$ , is the electrolyte resistivity and  $t$  is the electrolyte thickness. Table 1 gives the

$i$  experimentally measured conductivity of a YSZ disc of 2 mm thickness at the four temperatures at which the cell performance was measured. Also given in the table is the estimated electrolyte contribution to the  $R_{\text{cell}}$  as well as the measured (minimum)  $R_{\text{cell}}$  from linear parts of the voltage vs. current density plots.  $R_{\text{cell}}$  would also be expected to contain the ohmic part of the activation overpotential or the charge transfer resistance in addition to the ohmic part of the concentration polarization. The inset shows a plot of  $\ln(R_{\text{cell}} / T)$  vs.  $1/T$ , where  $T$  is the temperature in K, from the slope of which, an activation energy,  $Q_{\text{cell}}$  of  $\sim 50 \text{ kJ/mol}$ . is measured. The difference between  $R_{\text{cell}}$  and  $\rho_t t$  is approximately the contribution of charge transfer and concentration polarization. This is also listed in Table 1. It is seen that  $(R_{\text{cell}} - R_{\text{electrolyte}})$  over a range of temperatures from 700 to  $800^\circ\text{C}$  is between  $\sim 0.04$  and  $0.07$

$\text{cm}^2$ , showing that the combined contribution of activation and concentration polarizations is typically less than the ohmic contribution. At  $650^\circ\text{C}$ , the estimated  $(R_{\text{cell}} - R_{\text{electrolyte}})$  is only about  $0.007 \text{ cm}^2$ . We believe such a low value simply reflects an error due to possible differences between the actual ohmic contribution in the cell and that estimated based on measurements of conductivity made on a thick pellet. The ionic conductivity of the thin film YSZ in the actual cell may have been higher than the thick disc due to some differences in the microstructure, e.g. the grain size or differences in the grain boundary structure/chemistry. Nevertheless, the results show that in the anode-supported cells made in the present work, the combined contribution of activation and concentration polarizations has been substantially lowered compared to some of the work reported on electrolyte-supported cells.

Figure #4(b) shows the corresponding plots of power density vs. current density at various temperatures. The maximum power density measured at  $800^\circ\text{C}$  was  $\sim 1.8 \text{ W/cm}^2$  and that measured at  $650^\circ\text{C}$  was  $\sim 0.82 \text{ W/cm}^2$ . It is seen that power density vs. current density plot becomes increasingly nonsymmetric with increasing temperature. This behavior is consistent with calculations based on transport of gaseous species through porous electrodes.

In order to assess the thermal shock behavior of anode-supported cells, the following experiments were performed. Some of the cells of dimensions  $\sim 5$  cm x  $\sim 5$  cm and  $\sim 2$ - $3$  mm in thickness, that had been heat treated in hydrogen to convert NiO into Ni, were heated in an inert atmosphere to  $800^{\circ}\text{C}$  (to prevent the oxidation of Ni). Then, while still hot, the cells were removed from the furnace and placed on a ceramic plate at room temperature. The cells did not crack and the YSZ electrolyte did not debond indicating that the thermal shock resistance of these anode-supported cells is excellent.

**Stack results:** Stack testing was conducted at  $800^{\circ}\text{C}$  using cells of typical dimensions  $\sim 5$  cm x  $\sim 5$  cm x  $\sim 2$ - $3$  mm. Each stack tested had four cells separated by metallic interconnects. The interconnect was subjected to surface treatments to minimize the effects of oxidation. The best interconnect material explored to date exhibits an area specific resistance of  $\sim 0.08$   $\text{cm}^2$  after 5,000 hours at  $800^{\circ}\text{C}$ . Performance is expected to be better at a lower temperature such as  $650^{\circ}\text{C}$ . Voltage probes were introduced between each repeat unit. No glass was used; the edges of the interconnects themselves served as seals. Manifolds were made of a metallic alloy which were attached using electrically insulating inorganic gaskets.

Figure #5 shows voltage vs. current for the four repeat units of the stack. Note that the repeat unit area specific resistance, including the interconnect, is as low as  $\sim 0.49$   $\text{cm}^2$ . Figure #6 shows a plot of the total power vs. current. The maximum power measured was  $\sim 33$  watts. The thickness of the repeat unit was about 3 mm. This translates into a volumetric power density of  $1.1$  kW/liter (excluding manifolding) at  $800^{\circ}\text{C}$ . The cells used were not the most advanced ones incorporating recent improvements in electrode design. It is anticipated that with further improvements in cell fabrication procedures and interconnect, significantly higher stack performance should be possible. Figure #7 shows stack performance history of our work.

## SUMMARY

Anode-supported single cells with YSZ as the electrolyte and LSM + YSZ as the cathode were fabricated and tested. Maximum power densities as high  $1.8$  W/cm $^2$  at  $800^{\circ}\text{C}$  and  $0.82$  W/cm $^2$  at  $650^{\circ}\text{C}$  were measured with humidified hydrogen as the fuel and air as the oxidant. At  $800^{\circ}\text{C}$ , the lowest area specific resistance measured was  $\sim 0.13$   $\text{cm}^2$  in the ohmic regime. The design of the electrode involved attention to both charge transfer processes at electrolyte/electrode interfaces as well as gas transport through porous electrodes. Four cell stacks using square cells of dimensions 5 cm x 5 cm were assembled and tested using metallic interconnects. Critical issues concerning the metallic interconnect involved oxidation resistance of the alloys and the electronic conductivity of the oxide scale. Four cell stacks tested exhibited area specific repeat unit as low as  $0.5$   $\text{cm}^2$  at  $800^{\circ}\text{C}$ . The cells tested were not the highest performing cells. Even with these cells of intermediate level performance, stack power of 33 watts was measured which translates into  $\sim 1.1$  kW/liter at  $800^{\circ}\text{C}$ .

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Table 1

Temperature (°C)	Electrolyte Resistivity, $\rho$ ( cm)	Calculated $R_{electrolyte} = \rho t$ ( cm <sup>2</sup> )	Measured $R_{cell}$ ( cm <sup>2</sup> )	$R_{cell} - R_{electrolyte}$ (Contributions from activation and concentration polarizations)
650	248	0.248	0.255	0.007
700	147	0.147	0.189	0.042
750	91	0.091	0.151	0.06
800	61	0.061	0.130	0.069

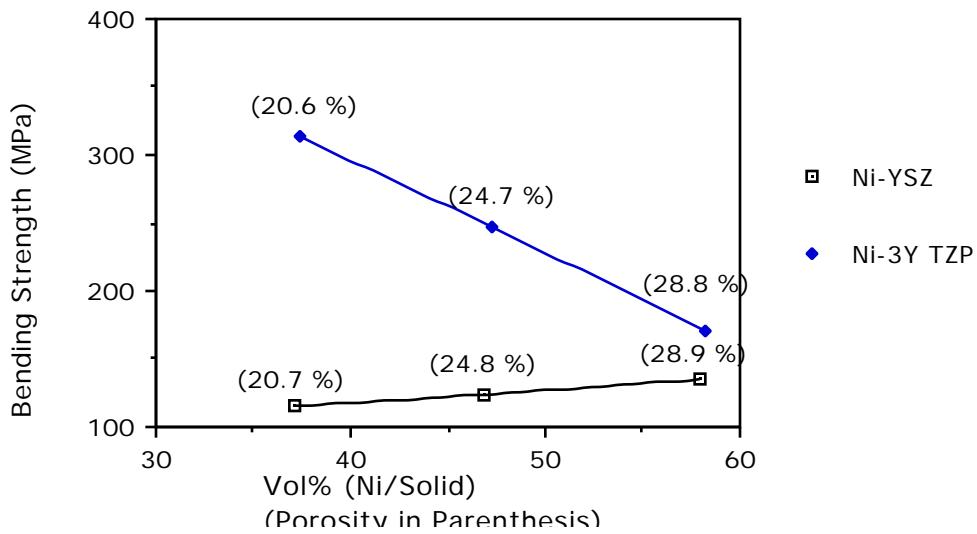


Figure #1: Flexural strength measured on Ni + 8 mol.%  $\text{Y}_2\text{O}_3$ -doped  $\text{ZrO}_2$  (YSZ) and Ni + 3 mol. %  $\text{Y}_2\text{O}_3$ -doped  $\text{ZrO}_2$  (TZP) as a function of vol.% Ni. The porosity is given in parentheses.

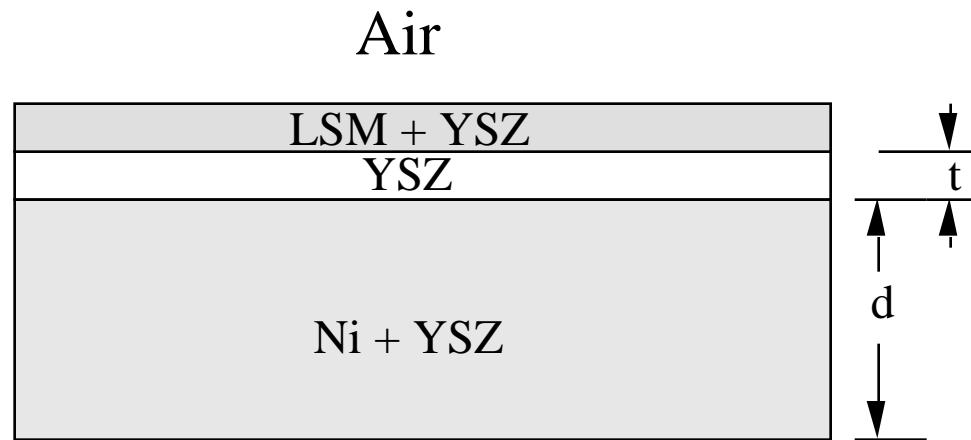


Figure #2: A schematic showing an anode-supported cell. Since the LSM + YSZ cathode is porous, its Young's modulus is rather small. Thus, calculations of stresses to assess potential for delamination was based on YSZ film thickness alone.

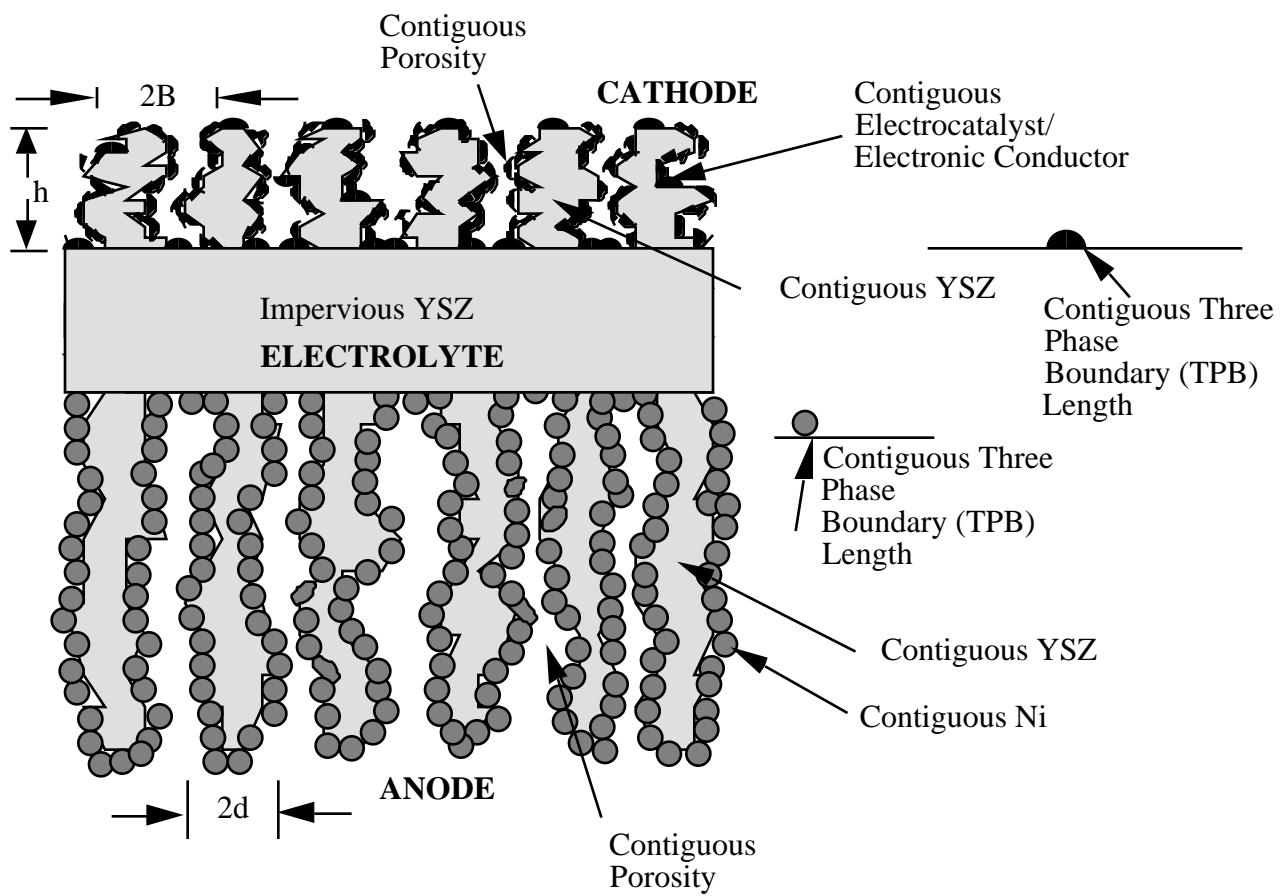
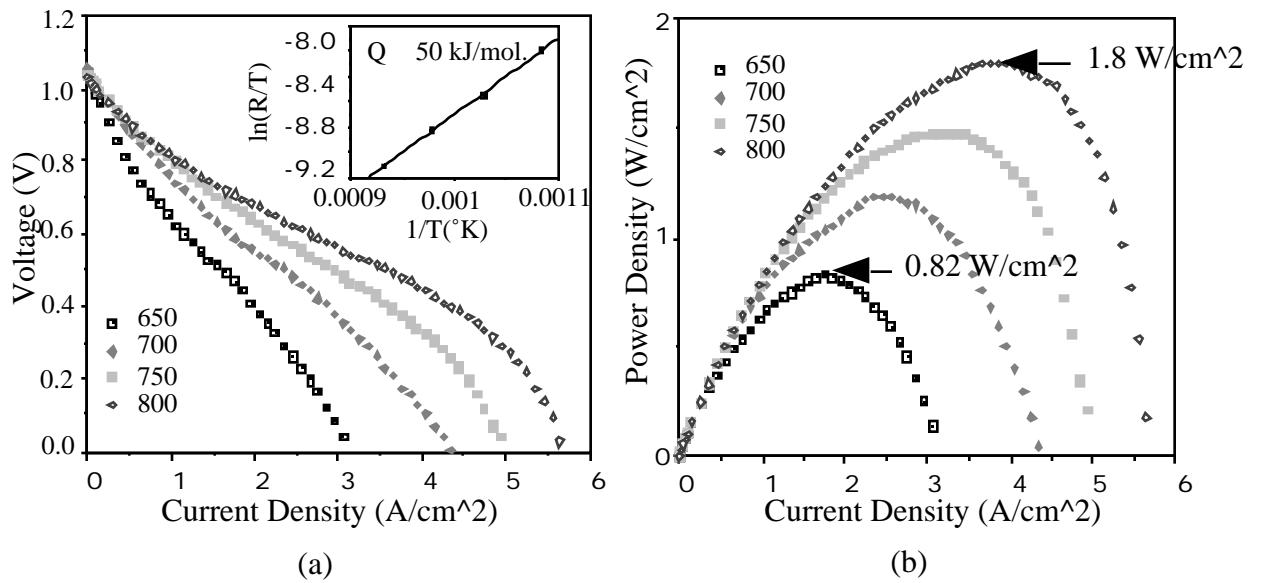
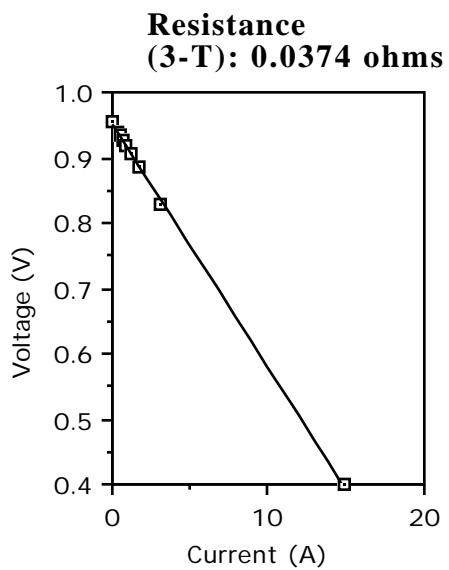
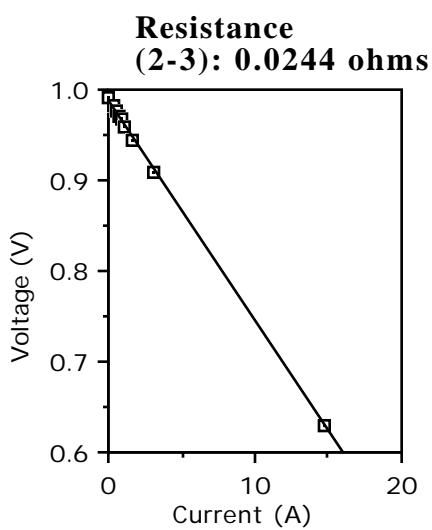
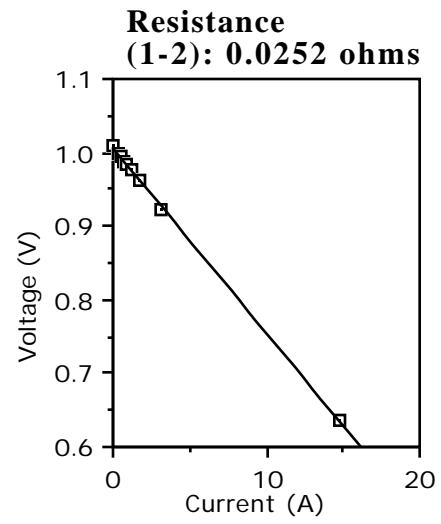
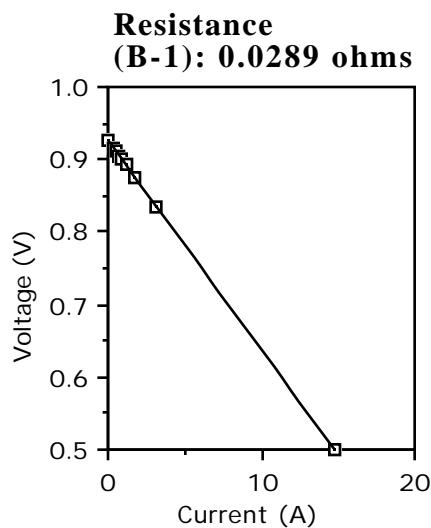


Figure #3: A schematic of an anode-supported cell showing contiguous YSZ in both electrodes, contiguous electrocatalyst in both electrodes, contiguous porosity in both electrodes, and a large TPB length in both electrodes.



**Figure :#4** Voltage and power density plots on a single cell at various temperatures. The maximum power densities at 650 and 800 °C measured were  $\sim 0.82$  and  $\sim 1.8$  W/cm<sup>2</sup> with humidified hydrogen as the fuel and air as the oxidant. Cell dimensions: Cathode thickness 50  $\mu\text{m}$ , electrolyte thickness 10  $\mu\text{m}$ , and the anode thickness 750  $\mu\text{m}$  (0.75 mm). Inset in (a) shows an Arrhenius plot of the cell resistance (determined from the linear part of the voltage vs. current density traces) as a function of temperature. The overall cell resistance obeys an Arrhenius behavior with an activation energy,  $Q = 50$  kJ/mol.



**Figure #5:** Voltage vs. current traces for the repeat units of a four cell stack tested at 800°C with humidified hydrogen as the fuel and air as the oxidant. Area per cell was  $\sim 20 \text{ cm}^2$ . The area specific resistance for (1-2) and (2-3) repeat units were  $0.5 \text{ cm}^2$ . Slightly higher values for the bottom current collector and cell #1 (B-1) and top current collector and cell #3 (T-3) are due to somewhat poorer contact between the cell and the current collectors. The above shows that the cell to cell contact (through the metallic interconnect) is quite good.

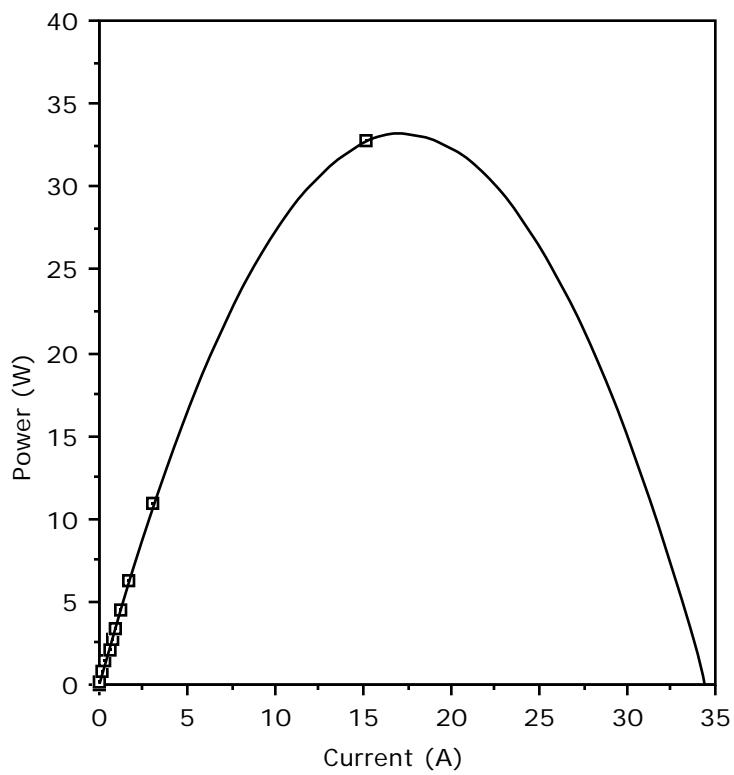


Figure #6: Power vs. current for a four cell stack tested at 800°C with a metallic interconnect.

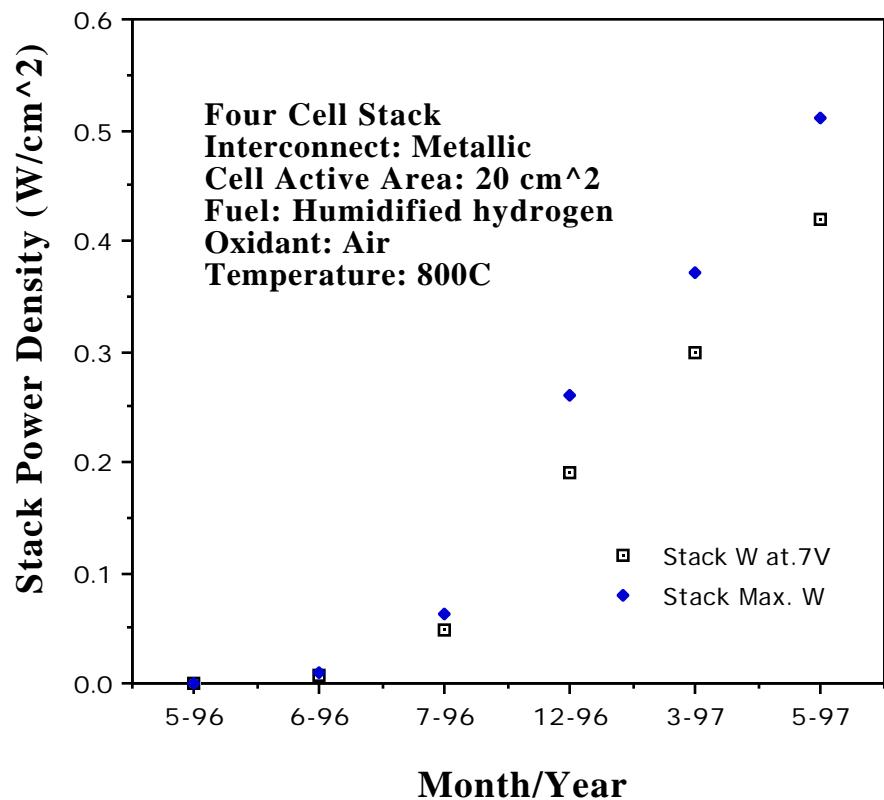


Figure #7: Stack performance history.

# Application of Ceria Layers to Increase Low-Temperature SOFC Power Density

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## Abstract

The main aim of this project has been to achieve solid oxide fuel cells (SOFCs) that can provide high power densities at relatively low temperatures; i.e., down to 600 °C or lower. In order to achieve this, we have developed thin YSZ and YSZ/YDC electrolyte SOFCs and have used interfacial layers to reduce interfacial resistances.

The interfacial resistance of Ni-Y<sub>2</sub>O<sub>3</sub>-stabilized ZrO<sub>2</sub> (Ni-YSZ) anodes have been reduced by inserting mixed-conducting layers of TiO<sub>2</sub>-doped YSZ (YZT) or Y<sub>2</sub>O<sub>3</sub>-doped CeO<sub>2</sub> (YDC) between the Ni-YSZ and YSZ electrolytes.

The effects of interlayer type, thickness, and YSZ surface condition on the interfacial resistance were investigated using Impedance Spectroscopy at temperatures ranging from 600 to 750 °C and three different Ps (H<sub>2</sub>). Two arcs, denoted as high frequency arc (HFA) and low frequency arc (LFA), were normally found in the impedance spectra. The HFA, which has been associated with the charge transfer process, was reduced by the addition of either interlayer.

The decrease in the size of HFA apparently results from the large reaction area within the porous interlayers. The LFA, which is believed to be associated with mass transport processes, decreased with increasing YSZ surface roughness, but increased with increasing interlayer thickness. The LFA was associated with a thermally activated process with an activation energy of  $\approx$  0.45 eV for Ni-YSZ/YDC, and  $\approx$  0.72 eV for Ni-YSZ/YZT. Under 97 percent H<sub>2</sub> + H<sub>2</sub>O, the lowest interfacial resistances of Ni-YSZ with a 0.5  $\mu$ m thick YDC interfacial layer were 0.13  $\Omega$ -cm<sup>2</sup> at 750 °C and 0.29  $\Omega$ -cm<sup>2</sup> at 600 °C.

SOFCs with electrolytes that provide high open-circuit voltage (OCV) and low ohmic loss down to 550 °C are described. The electrolytes were bi-layers consisting of a 4 to 8  $\mu$ m thick Y-doped ceria (YDC) layer with a 1. to 1.5  $\mu$ m thick Y-doped zirconia (YSZ) layer on the fuel side. The cathode/supports were La<sub>0.85</sub>Sr<sub>0.15</sub>MnO<sub>3</sub>-YSZ. The anodes consisted of thin YDC and Ni-YSZ layers. The YDC/YSZ electrolyte SOFCs yielded 85 to 98 percent of the theoretical OCV, compared with  $\approx$  50 percent for YDC electrolyte SOFCs. The cathode overpotential, which

was a main factor limiting SOFC power density, was lower for YDC/YSZ than YSZ electrolytes. The maximum power density at 600 °C, 210 mW/cm<sup>2</sup>, is higher than previously reported SOFCs.

SOFC power densities typically drop rapidly as the operating temperature is decreased, because of electrolyte ohmic losses and/or electrode overpotentials. In this paper, we describe fuel cells utilizing 8  $\mu$ m-thick yttria-stabilized zirconia (YSZ) electrolytes to provide low ohmic loss. Adding thin porous yttria-doped ceria (YDC) layers on either side of the YSZ yielded much reduced interfacial resistances at both the LSM cathodes and Ni-YSZ anodes. The cells provide higher power densities than previously reported at below 700 °C, e.g., 300 and 480 mW/cm<sup>2</sup> at 600 and 650 °C, respectively (measured in 97 % H<sub>2</sub> + 3 % H<sub>2</sub>O and air), and also provide high power densities at higher temperatures, e.g., 760 mW/cm<sup>2</sup> at 750 °C.

# **Increased Solid Oxide Fuel Cell Power Density Using Interfacial Ceria Layers**

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# Introduction

- SOFC power density is reduced at low temperatures due to:
  - electrode overpotentials and/or
  - electrolyte ohmic losses
- Improve low-T performance using Yttria Doped Ceria (YDC)
- Low electrolyte loss using thin YSZ-YDC electrolytes
- Low overpotentials obtained by using:
  - LSM/YDC cathode interface and
  - Ni-YSZ/YDC anode interface
- Can low-T SOFCs internally reform hydrocarbons?
  - demonstrate low-T SOFC operation on 97% methane
  - no C deposition

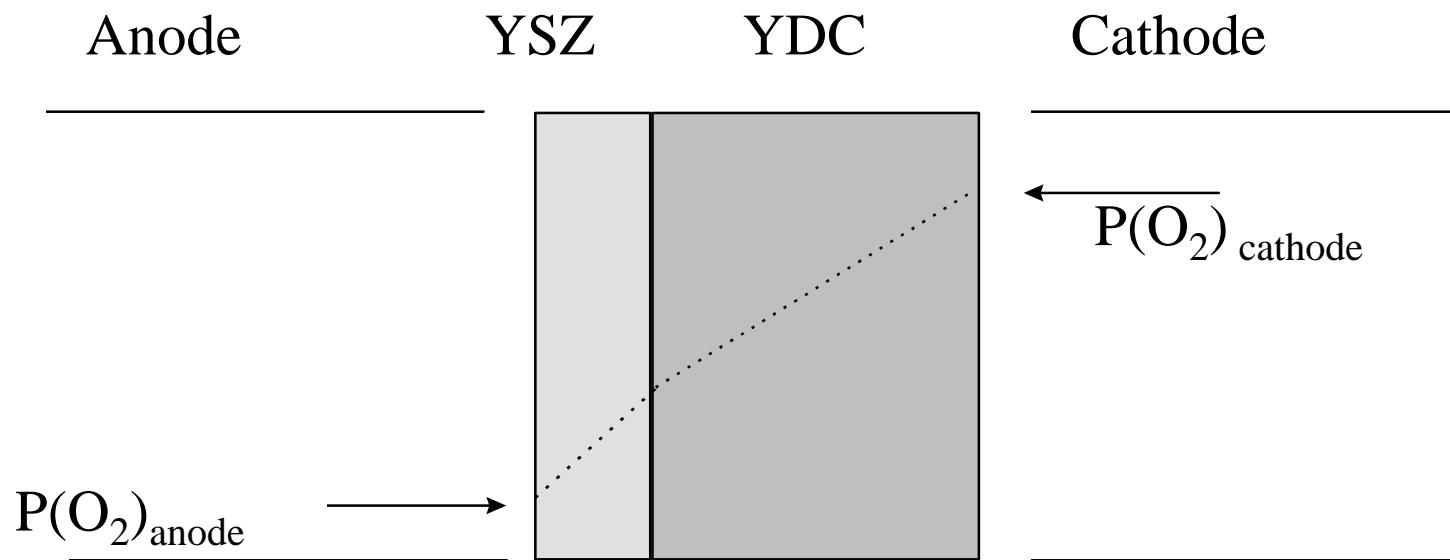
# Experimental

- LSM cathodes by pressing and sintering
- Reactive sputtering from metal targets:
  - dense YSZ and YDC electrolyte layers using DC substrate bias
  - porous YDC interfacial layers and Ni-YSZ anodes
- Standard single-cell tests and impedance spectroscopy
  - $T = 550 - 800^\circ\text{C}$
  - cathode: air
  - anode:  $97\% \text{ H}_2 + 3\% \text{ H}_2\text{O}$   
 $97\% \text{CH}_4 + 3\% \text{ H}_2\text{O}$

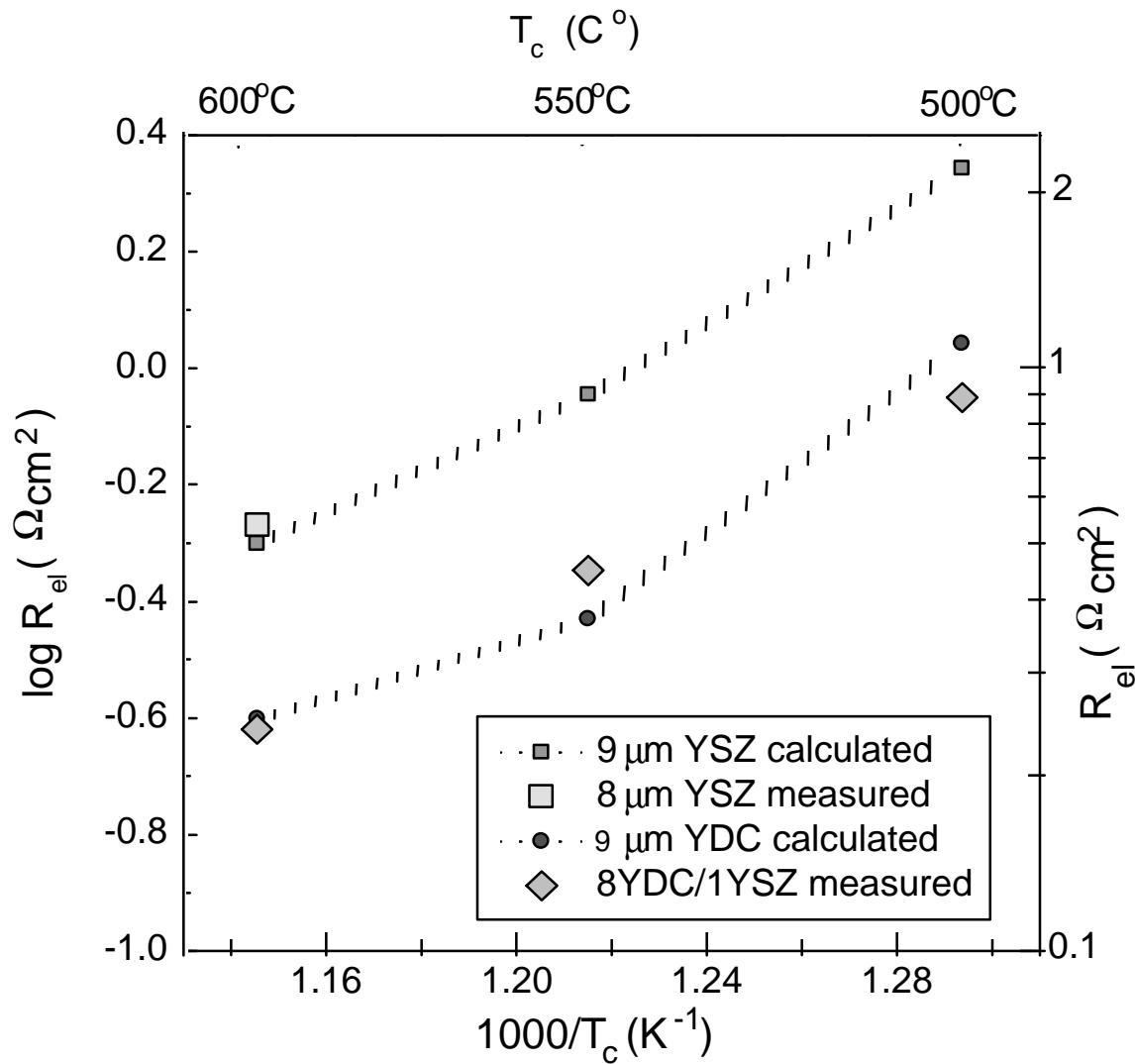
# Decrease electrolyte resistance using YSZ-YDC bi-layer

Thin anode-side YSZ layer:

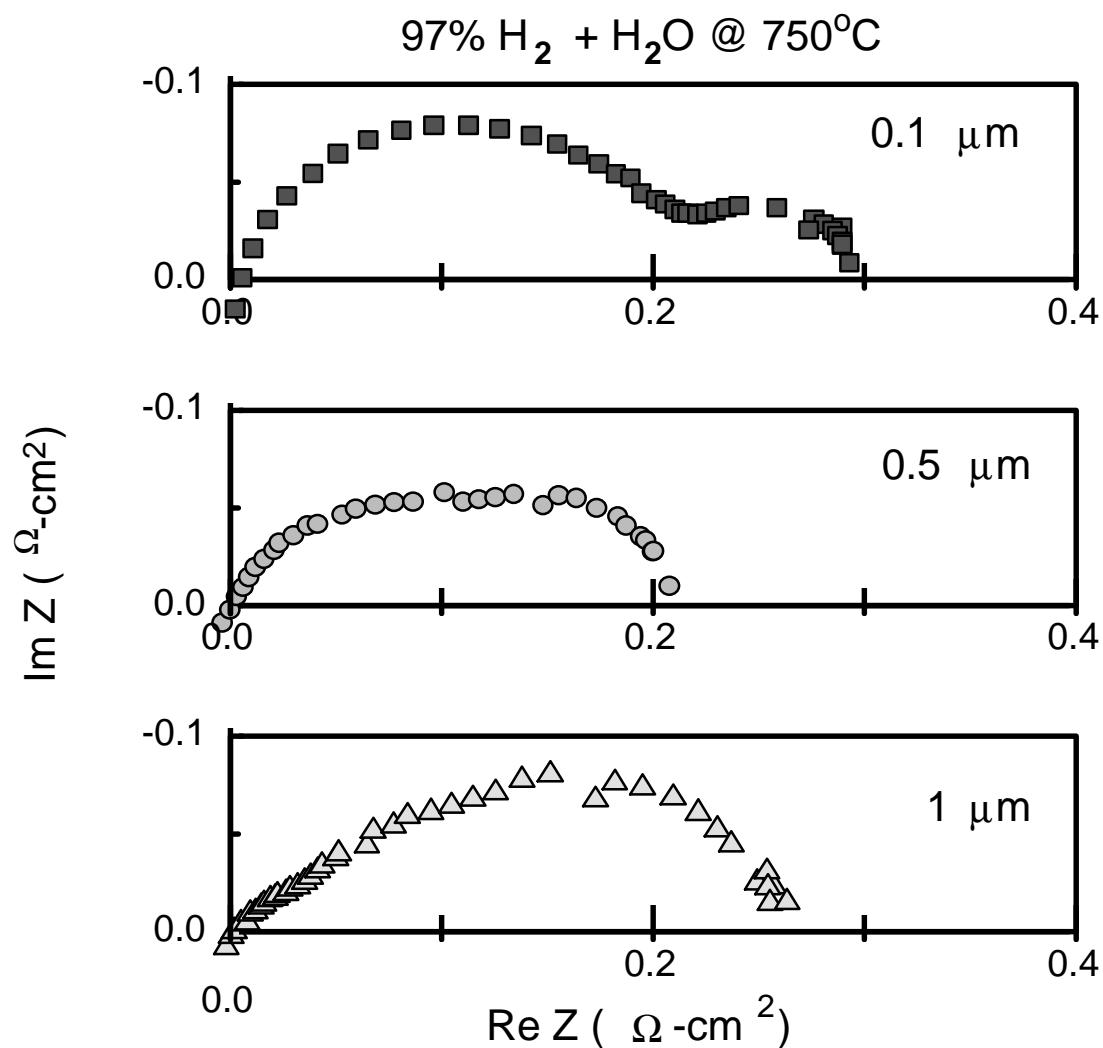
- protects YDC from reduction
- blocks electron current flow



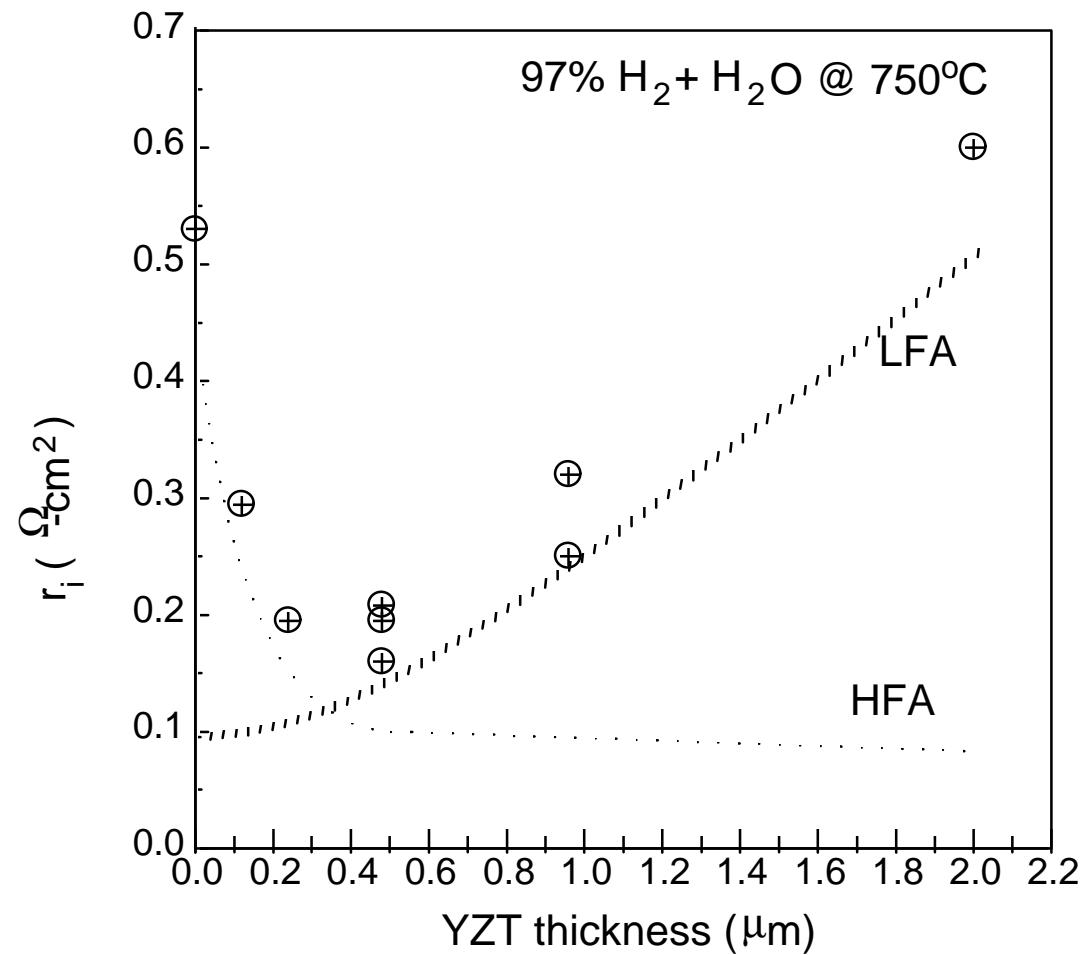
# Electrolyte Resistance: Measured vs. Predicted



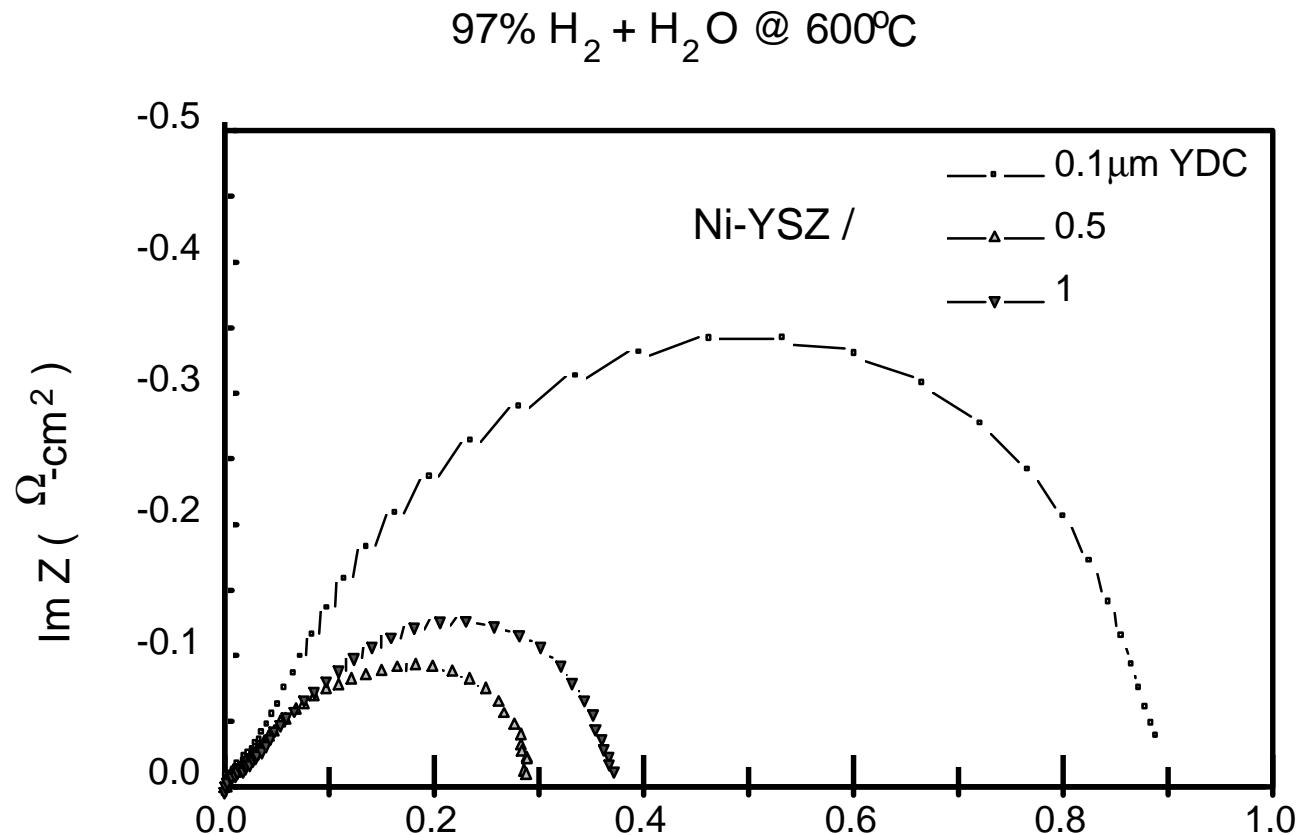
# YZT Thickness Effect



# YZT Thickness Effect



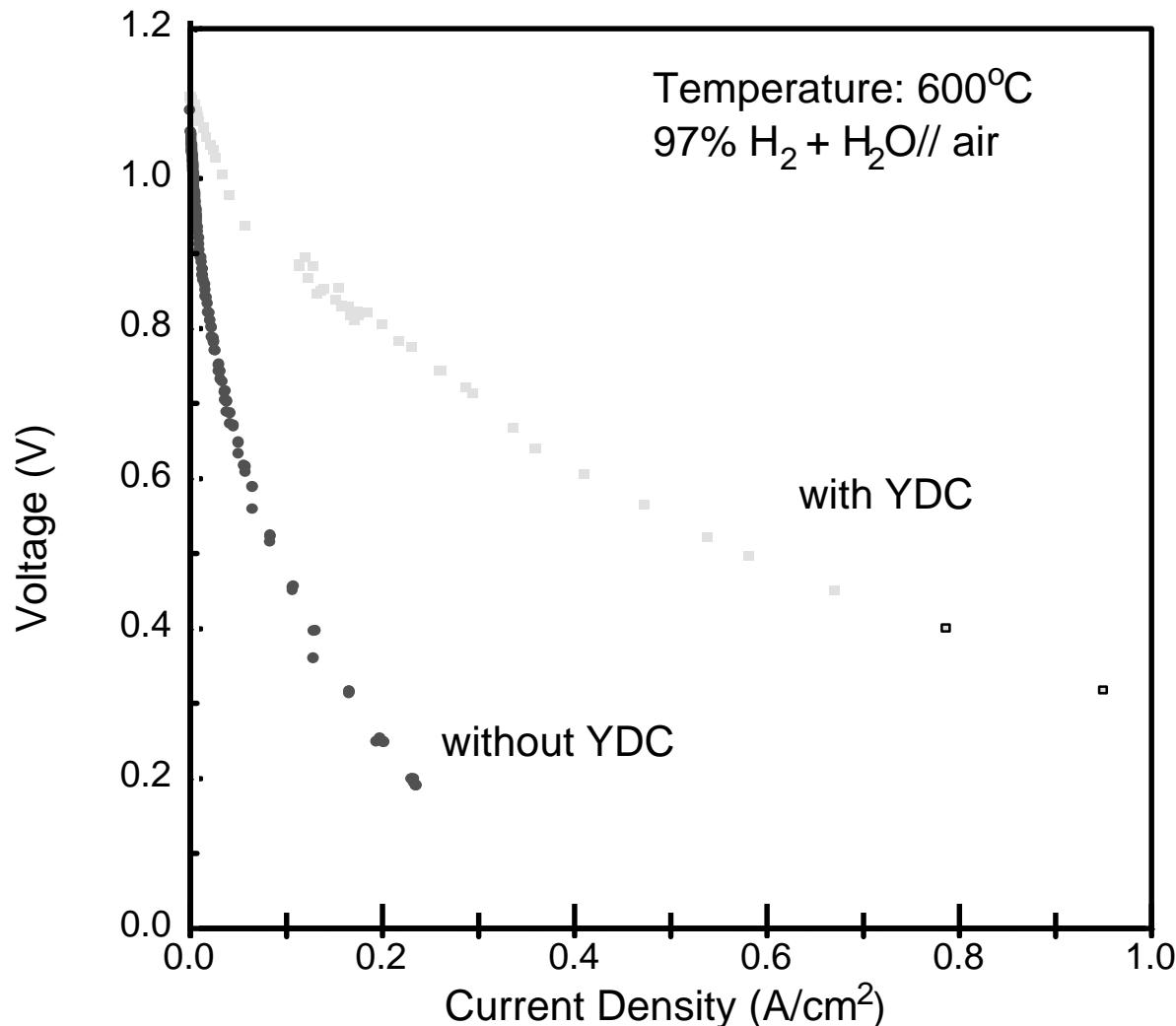
# YDC Thickness Effect on Ni-YSZ anode



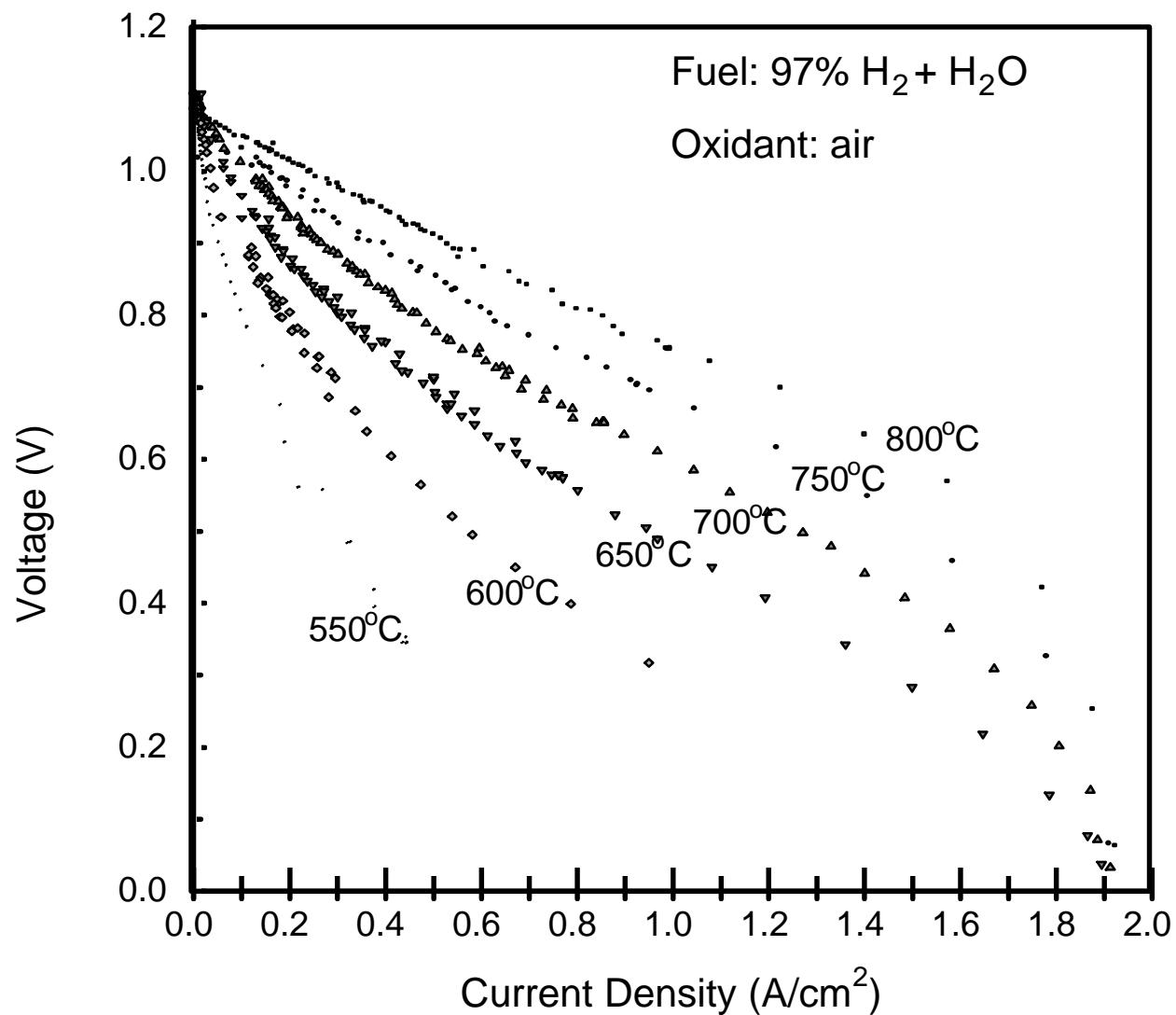
# Cell Performance:

## LSM/YSZ/YDC/Ni-YSZ

## LSM/YDC/YSZ/YDC/Ni-YSZ

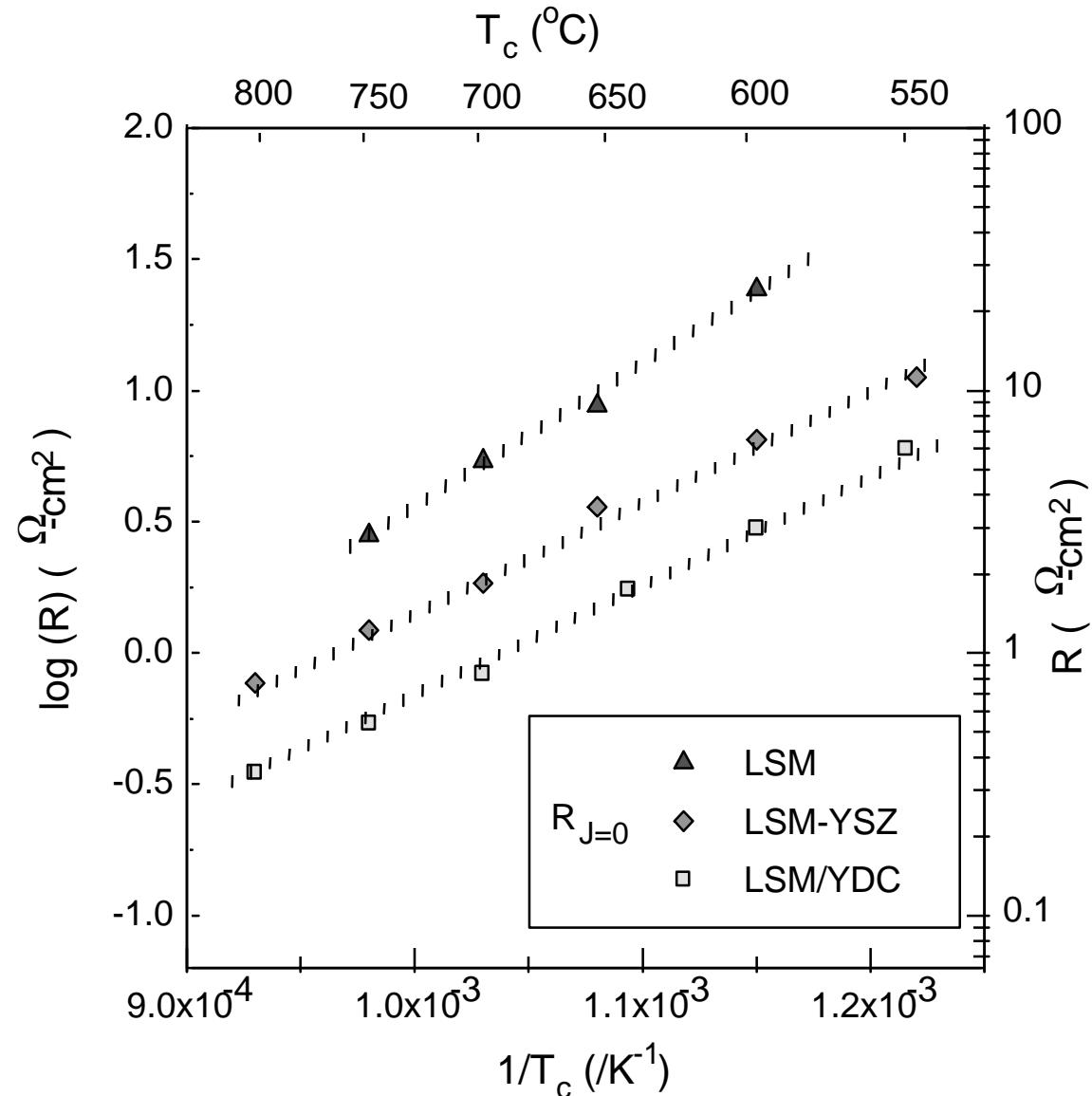


# Cell Performance: LSM/YDC/YSZ/YDC/Ni-YSZ

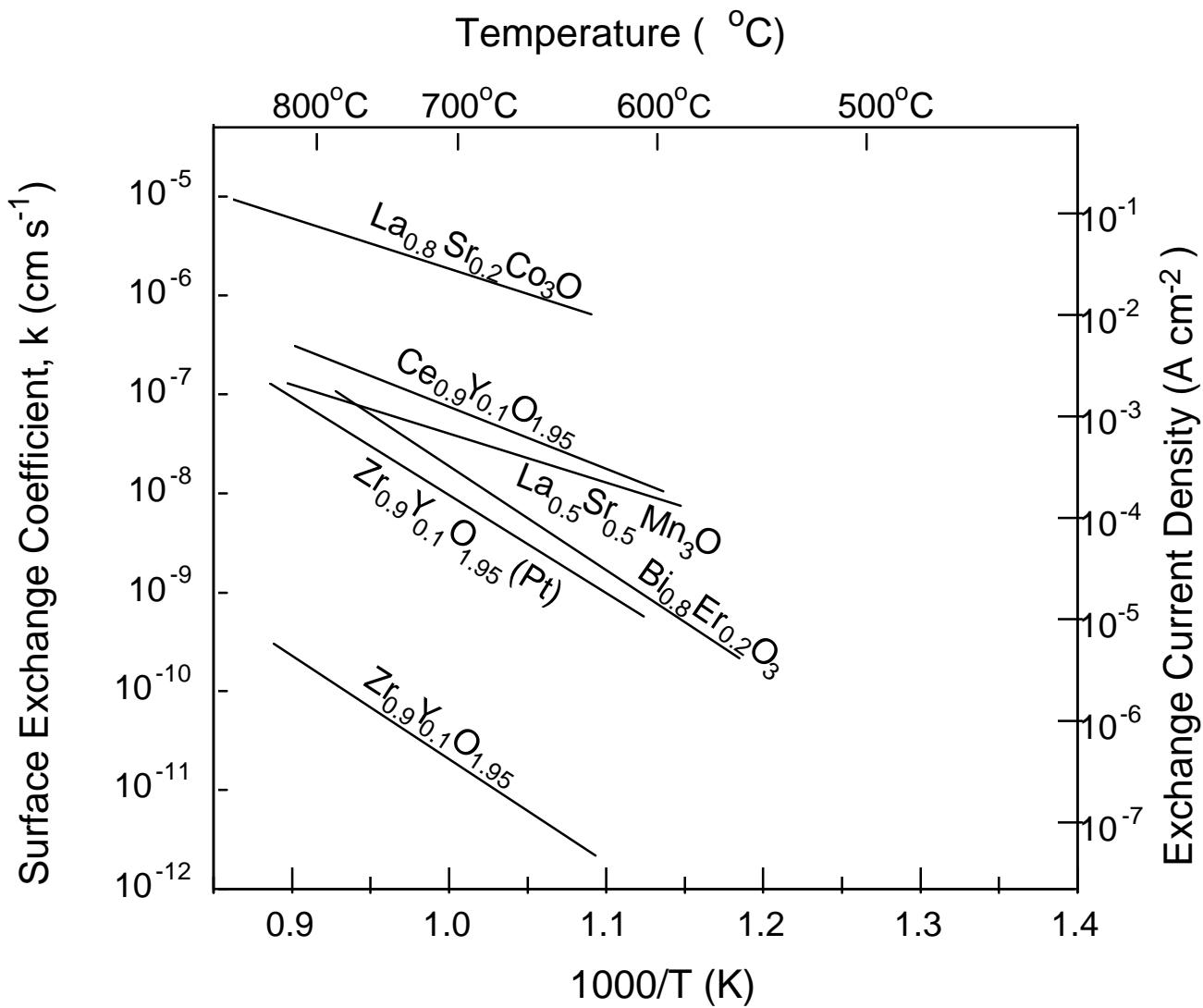


# Cell Resistance

## Cathode/YSZ/YDC/Ni-YSZ

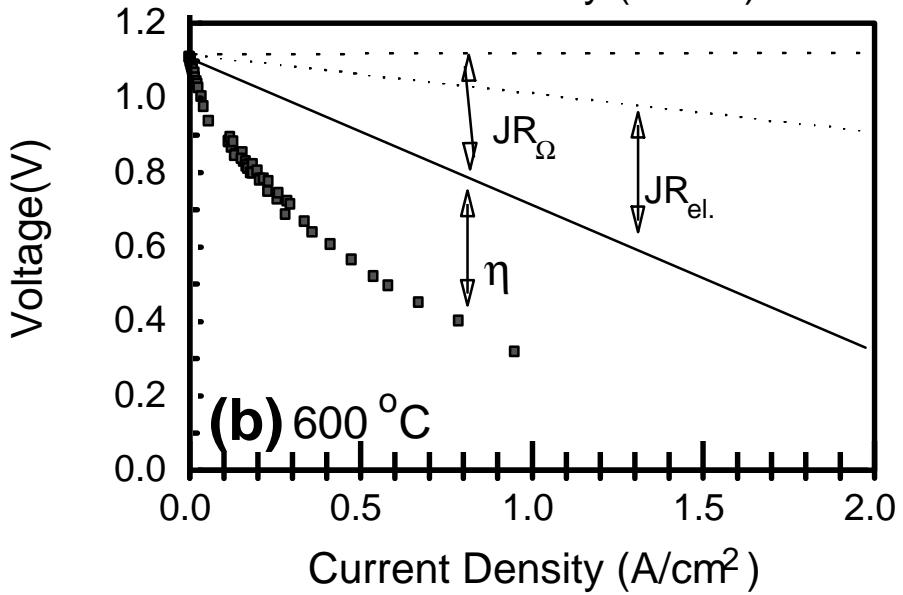
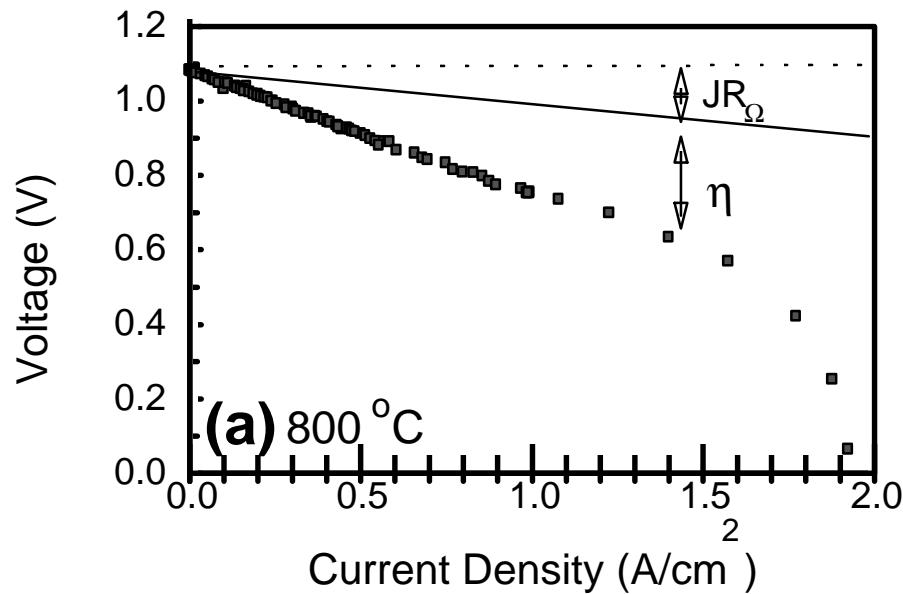


# Oxygen Surface Exchange Coefficient



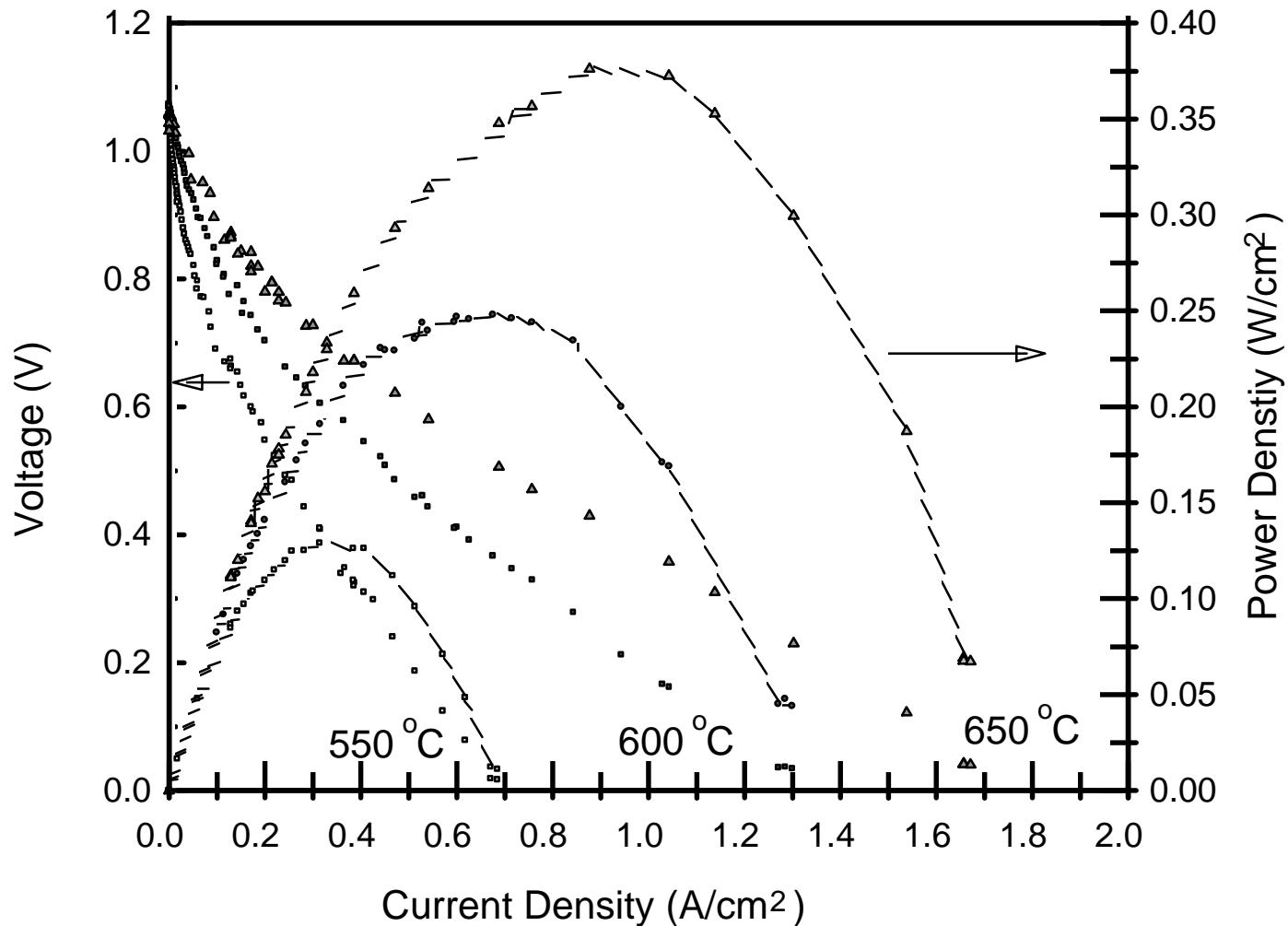
\* B.C.H. Steele, *Solid State Ionics*, **75** (1995) 175.

# Loss Analysis for 600 and 800°C



**Cell: Ni-YSZ/ YDC/ YSZ /YDC/ LSM**

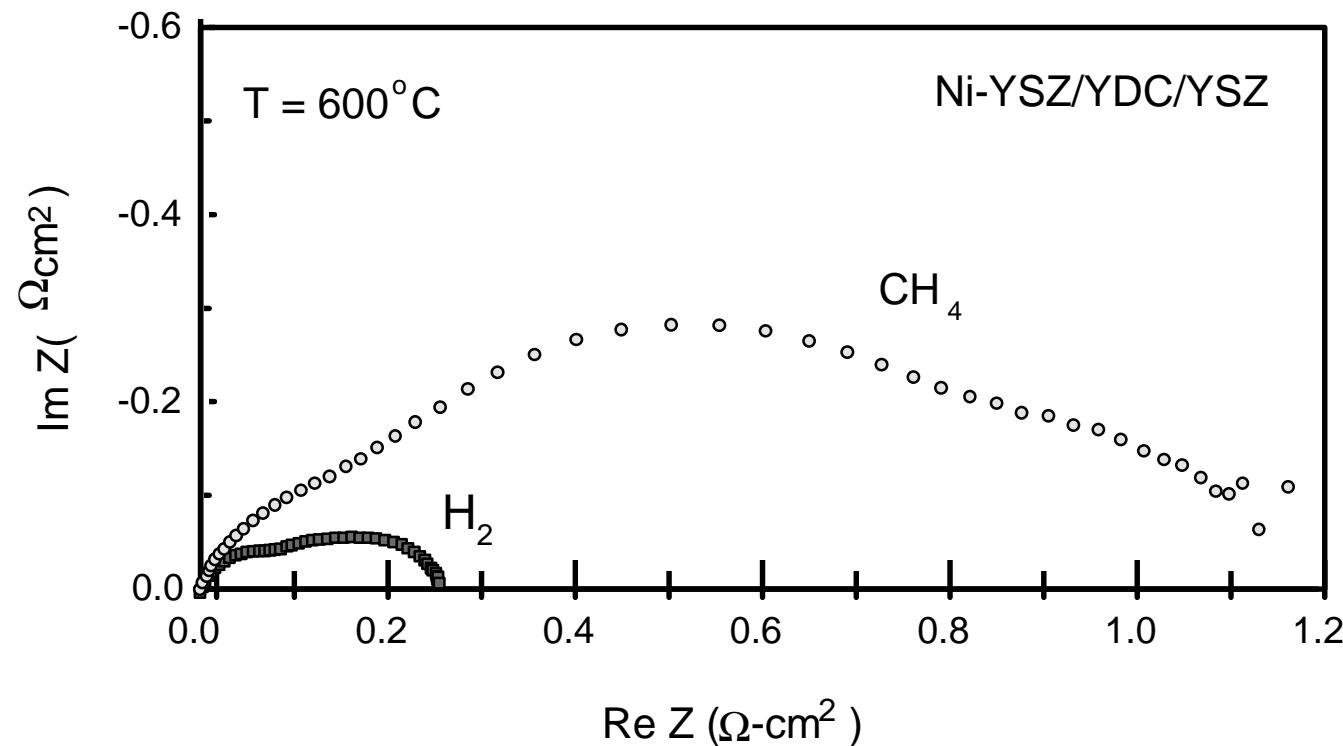
Fuel: 97%  $\text{CH}_4$  + 3%  $\text{H}_2\text{O}$  // Oxidant: Air



## Anode Performance with Hydrogen and Methane

Gases: 97% Hydrogen + 3% Steam

97% Methane + 3% Steam



## Conclusions

- Low-T electrolyte resistance reduced using YSZ-YDC bi-layers.
- On the anode side, YDC enhances anode performance via mixed conductivity and/or increased redox reaction rate.
- On the cathode side, the YDC apparently provided a higher oxygen surface exchange coefficient.
- Low-T SOFC performance:
  - 600°C 300 mW/cm<sup>2</sup>
  - 700°C 630 mW/cm<sup>2</sup>
  - 800°C 870 mW/cm<sup>2</sup>
- Good single-cell performance without carbon deposition with methane fuel

# **SOFC Stack Performance Goals**

**John D. Wright**

**Presented at  
Fuel Cells '97**

**Morgantown, West Virginia**

**TDA Research Inc. • Wheat Ridge, CO 80033 • [www.tda.com](http://www.tda.com)**

# Objective

- **To Understand:**

- What performance is required for a SOFC to compete
- How stack performance affects the Cost of Electricity (COE)
- What are reasonable research goals

# Stack Parameters Affecting COE

- **Area-Specific Resistance**

- Stack cost
- Optimum power density/electrical efficiency

- **Excess Air Ratio**

- BOP costs
- Parasitic power

# Area-Specific Resistance Determines Optimum Power Density, Stack Cost, BOP Cost, and Electrical Efficiency

Base Case:

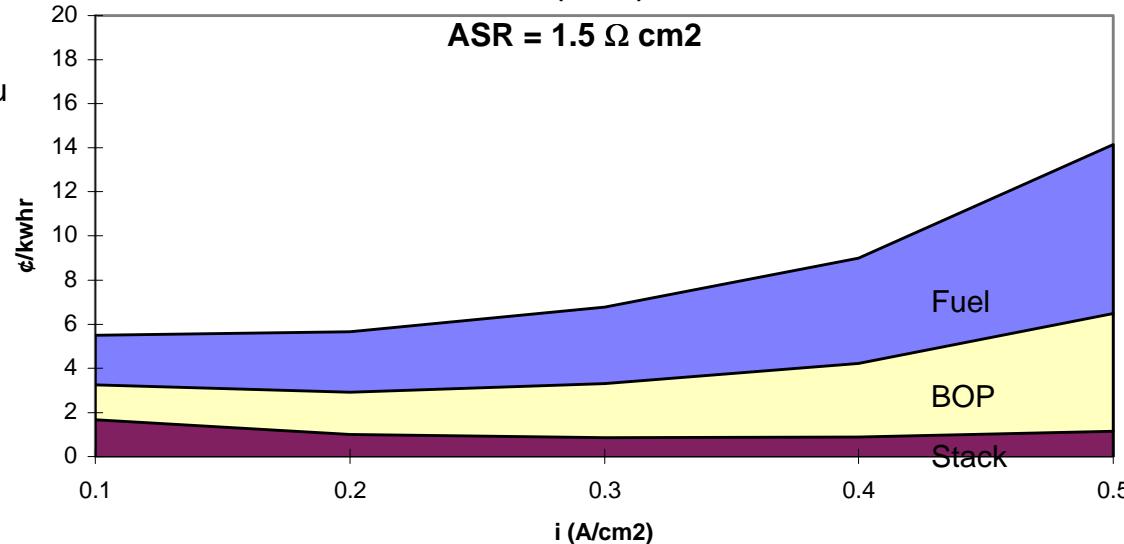
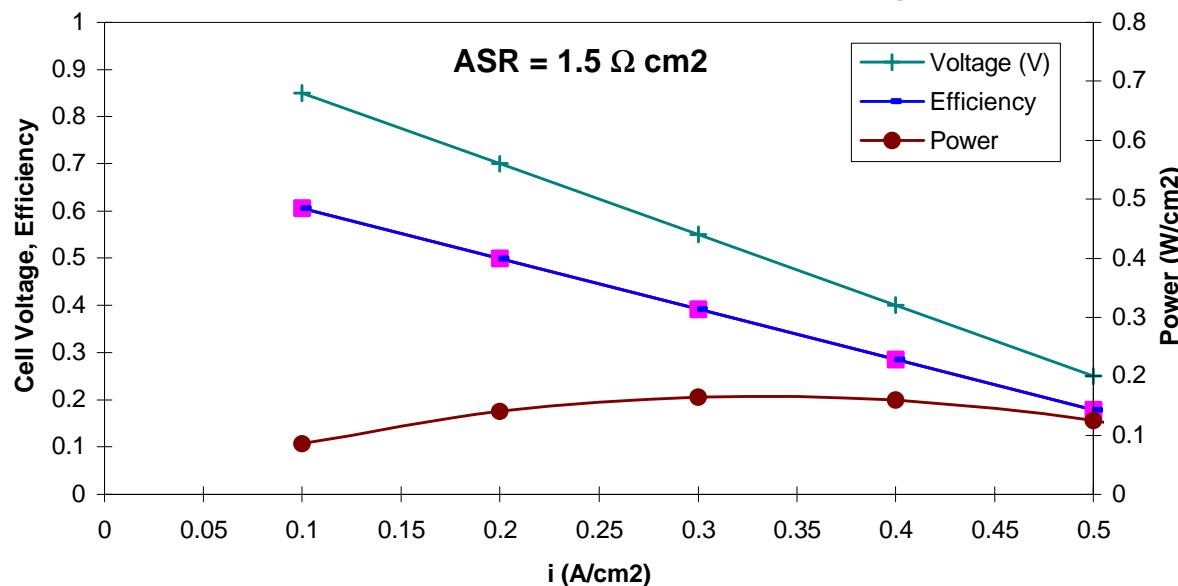
Efficiency: 50%

Stack Cost: \$500/m<sup>2</sup>

BOP: \$667/kW

CRF: 0.25/yr

Gas Cost: \$4.00/MMBtu



# Area-Specific Resistance Determines Optimum Power Density, Stack Cost, BOP Cost, and Electrical Efficiency

Base Case:

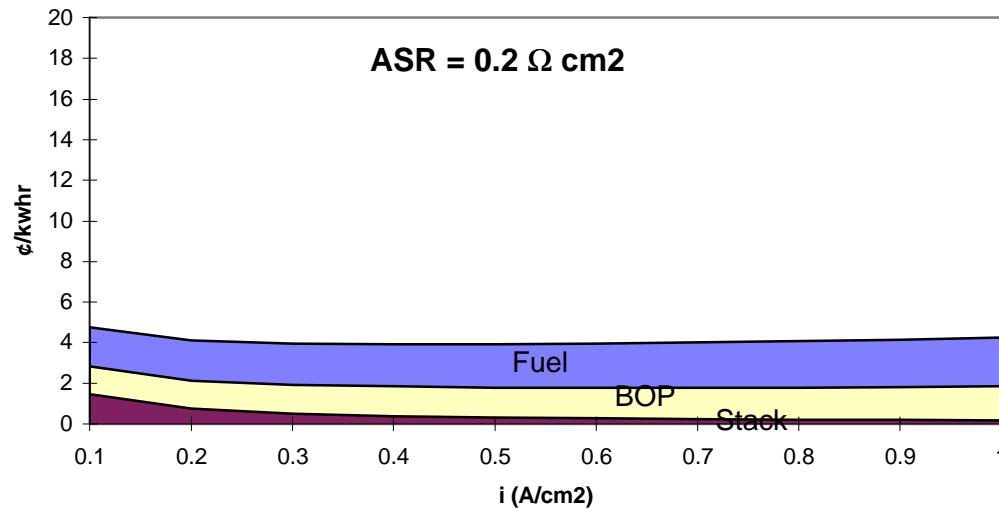
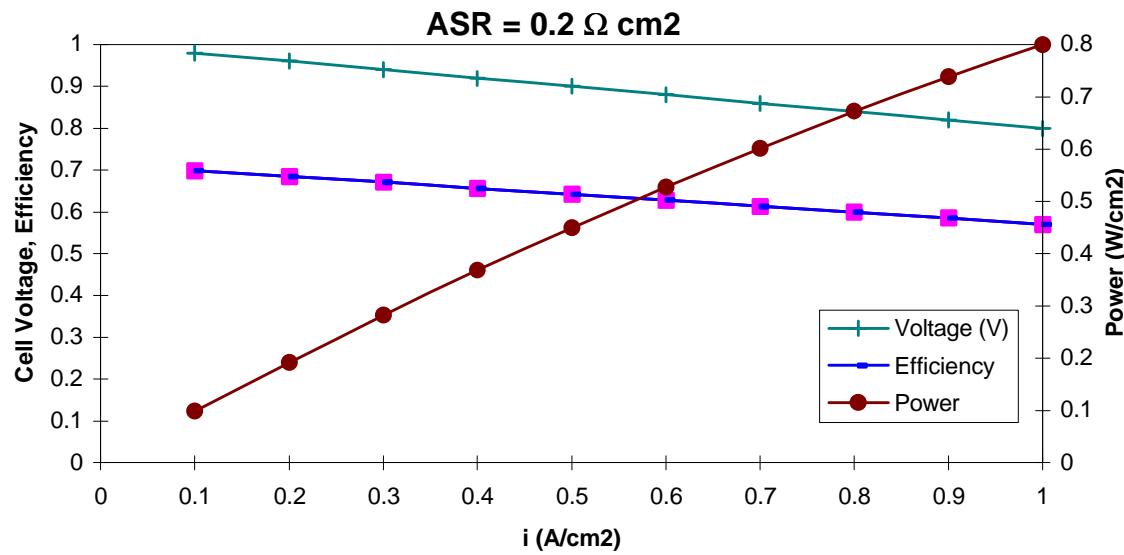
Efficiency: 50%

Stack Cost: \$500/m<sup>2</sup>

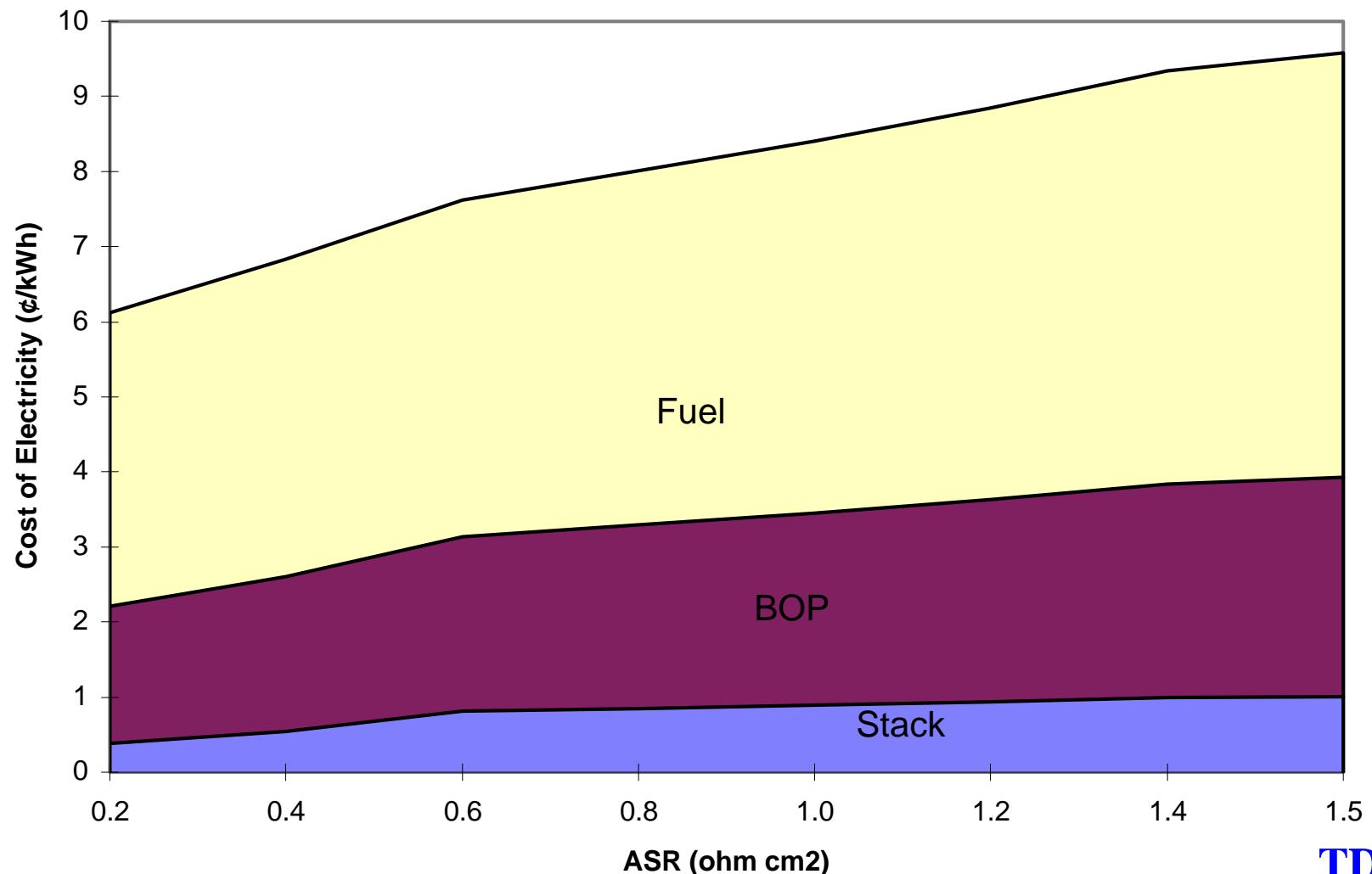
BOP: \$667/kW

CRF: 0.25/yr

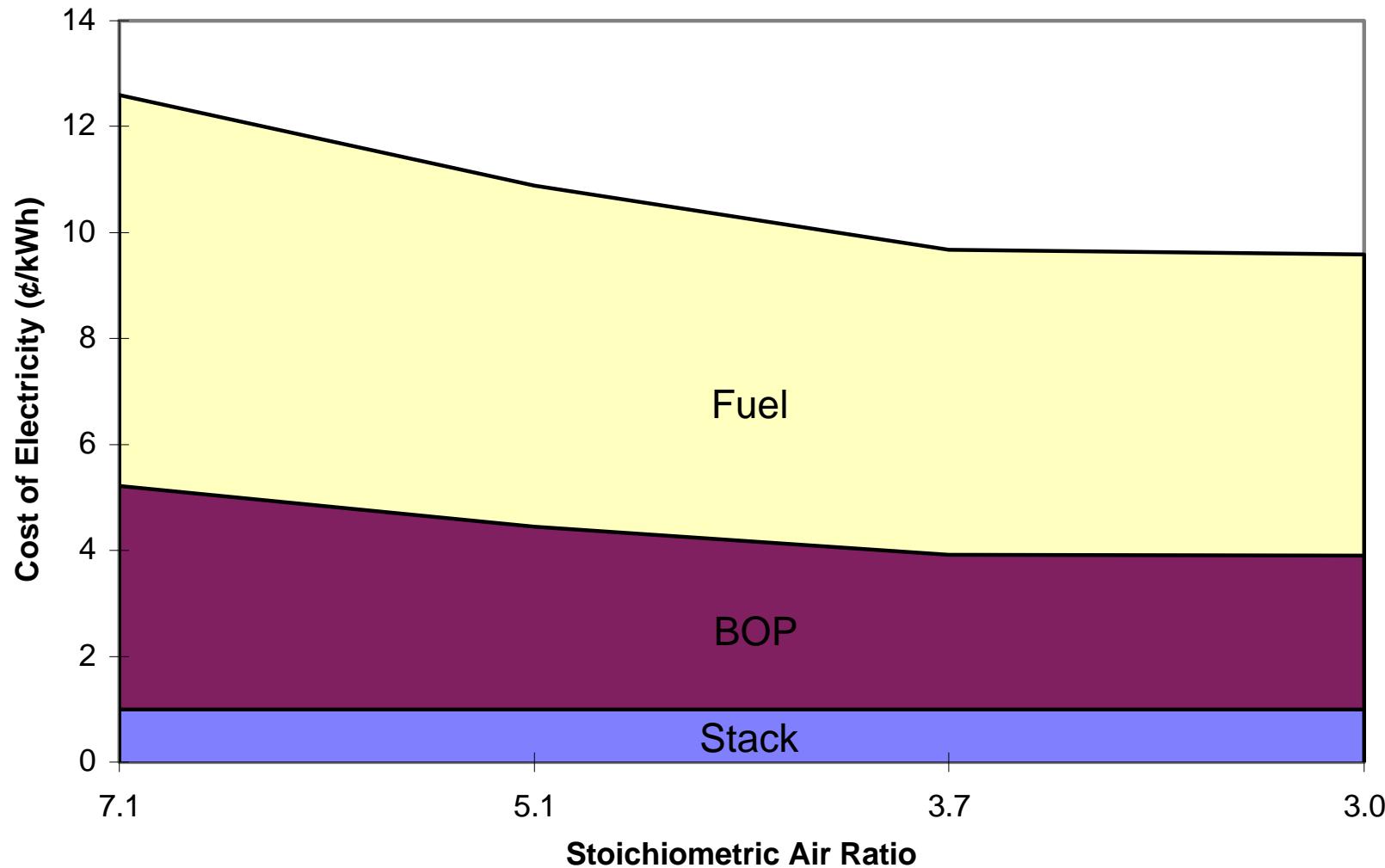
Gas Cost: \$4.00/MMBtu



# COE As a Function of Area-Specific Resistance



# Excess Air Affects Preheater and Compressor Capital Costs and Energy Efficiency



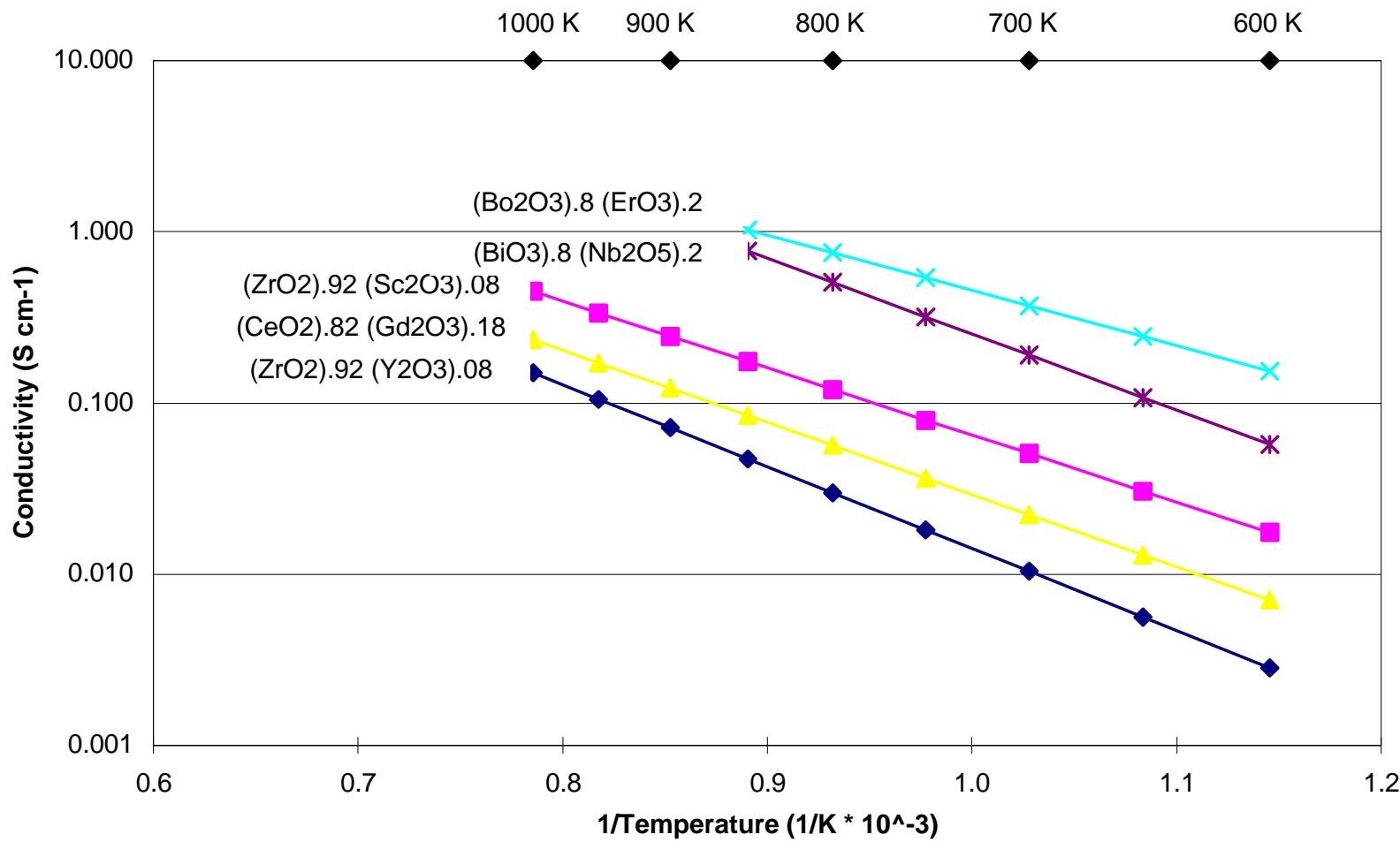
# Goal

- **Fuel Cell with ASR =  $0.2 \Omega \text{ cm}^2$** 
  - Cathode:  $0.07 \Omega \text{ cm}^2$
  - Electrolyte:  $0.06 \Omega \text{ cm}^2$
  - Anode:  $0.07 \Omega \text{ cm}^2$

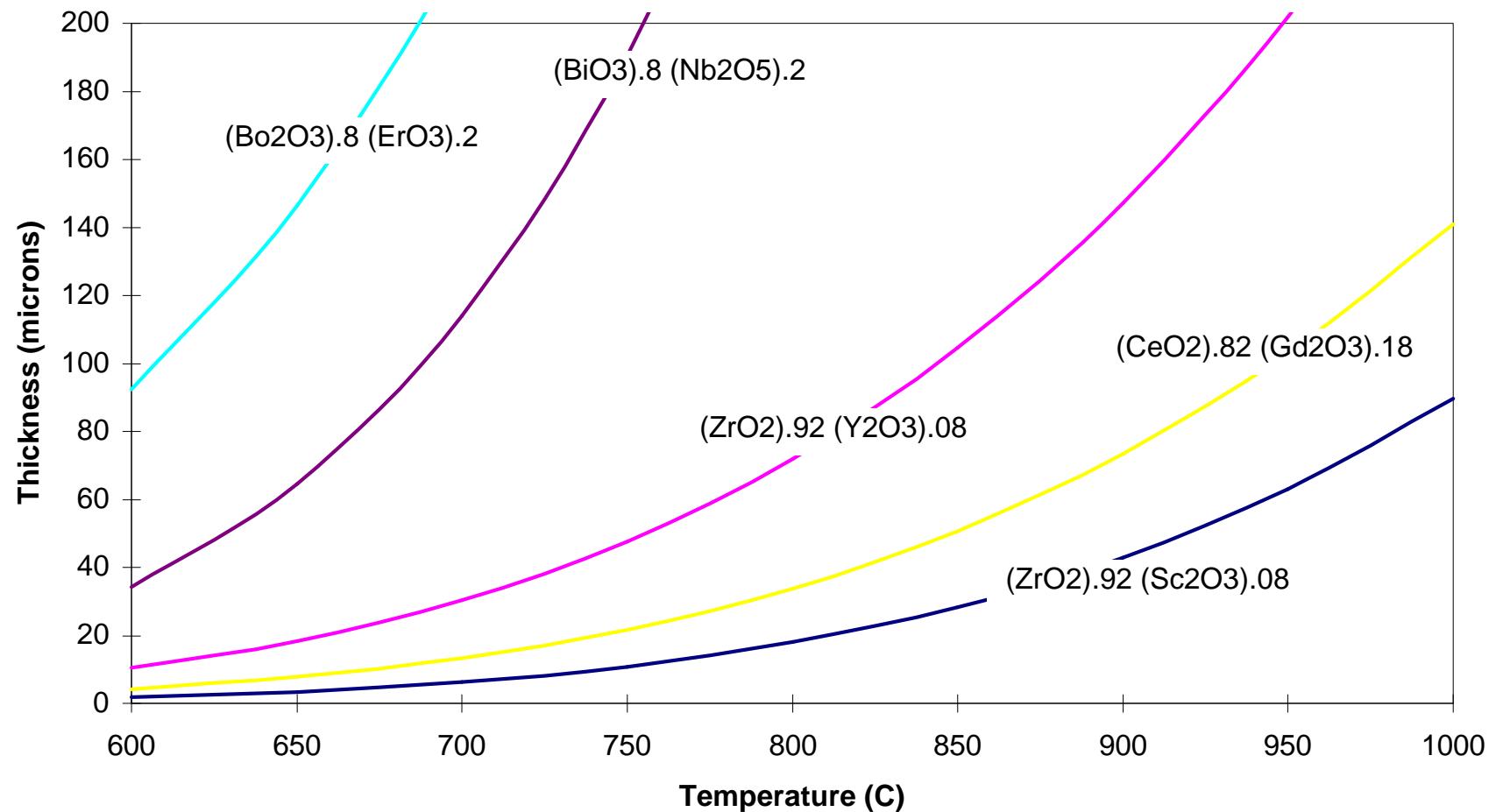
# Electrolyte

- **How Thin Is Thin Enough?**
  - As a function of operating temperature

## Electrolyte Conductivity vs Temperature



## Required Electrolyte Thickness as a Function of Temperature



# Electrodes

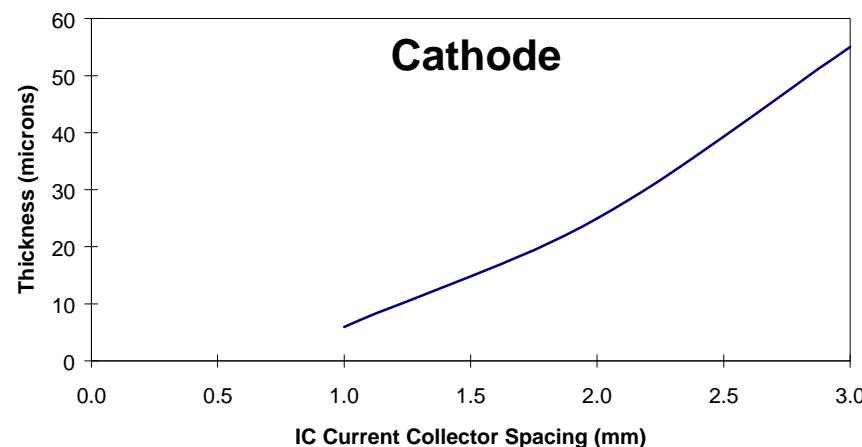
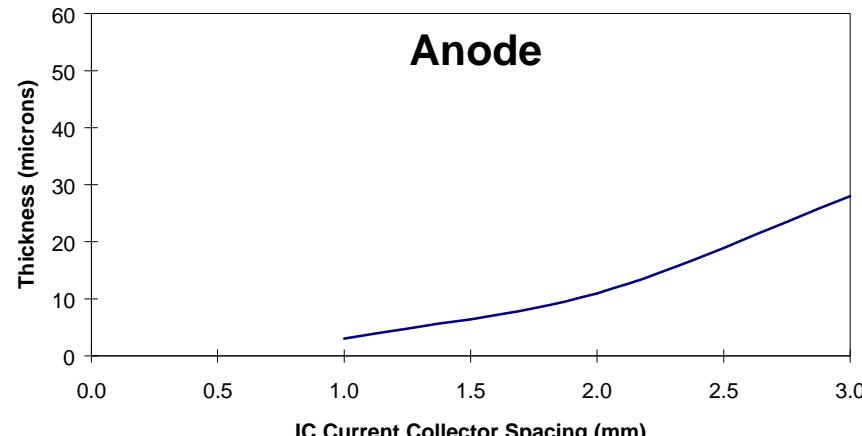
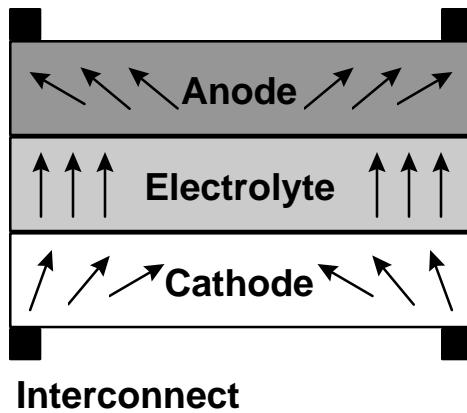
## Mass Transfer Limitations

- ASR Budget =  $0.01 \Omega \text{ cm}^2$
- Allowable Cathode Thickness =  $400 \mu\text{m}$   
Allowable Anode Thickness =  $2000 \mu\text{m}$

# Electrodes

$$\text{Electrical Resistance} = 0.01 \Omega\text{cm}^2$$

Minimum Thickness Required to Control Lateral Resistance



# Electrode Thickness Requirements

	<u>Electrical Conduction</u>		<u>Mass Transfer</u>
Anode	3-30 $\mu\text{m}$		2200 $\mu\text{m}$
Cathode	6-60 $\mu\text{m}$		400 $\mu\text{m}$

# Electrode Performance

- Dominated By Electrocatalytic Performance
- Overall Modeling Such As This Does NOT Provide Guidance

# Conclusions

- **Performance Budgets Can Provide Useful Guides for Setting R&D Goals**
- **Stack Performance Affects Overall Cost By Controlling:**
  - Electrical (ASR and excess air) efficiency
  - Stack cost
  - BOP cost
- **Several Methods Are Available to Prevent YSZ Electrolyte Conductivity From Being Rate Limiting at 700-800°C**
- **Mass Transfer and Ohmic Resistance Rarely Limit ASR in a Planar Cell**

## **Evaluation of Small-Particle Plasma Spray for SOFC Electrolyte Deposition**

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Evanston, IL 60208

### **Introduction**

The commercialization efforts for solid-oxide fuel cell (SOFC) systems are beginning to emphasize cost reduction in order to compete more effectively with other power generating methods. Several aspects of the SOFC stack have attracted particular attention for cost reduction efforts, with electrolyte deposition being one area of focus. Currently, SOFC electrolytes are processed using a range of methods, including tape casting methods, slurry/sol-gel, and electrochemical vapor deposition (EVD). Of these methods, tape casting is reportedly the most economical while EVD has higher costs. Atmospheric- plasma spray (APS) is likely to offer a cost competitive alternative to tape casting, but thin and dense electrolyte layers cannot be produced readily with conventional equipment. Small-particle plasma spray (SPPS) is a modified APS process which allows the deposition of thin and dense coatings by thermal spray.

### **Objective**

The objective of this project is to demonstrate the feasibility of depositing yttria-stabilized zirconia (YSZ) electrolytes onto lanthanum strontium manganite (LSM), and other test substrates. This includes characterizing the effect of process parameters and YSZ powder size on cell performance and electrolyte morphology. The goals of this effort are to demonstrate reliable deposition of dense and crack-free YSZ electrolytes on LSM by SPPS and to evaluate the performance of these electrolytes on both planar and tubular cathodes

### **Approach**

The initial tasks of the program were to demonstrate a preheat cycle which would allow deposition of YSZ onto LSM,

and screening of the deposition parameters using Hastelloy C substrates. Once process parameters are developed on the planar substrates, the process will be transferred to tubular substrates provided by Westinghouse. Modification of the process parameters is expected when the process is transferred to the tubular substrates, and developmental samples will be provided to Westinghouse for evaluation.

### **SPPS Technology**

SPPS technology allows the use of very fine ( 0.2 micron or larger) discrete powders. SPPS consists of both powder injection and feed technology, and is currently in the final phases of the patent process. SPPS can be retrofitted to any commercial plasma spray gun. Through the use of fine powders, SPPS has produced dense (>98%) yttria-stabilized zirconia (YSZ) coatings as thin as 5 micron for use in electrochemical sensors. An initial demonstration of SPPS for SOFC fabrication successfully produced SOFC tri-layers for electrochemical testing which resulted in an open circuit voltage approximately 80% of the theoretical value.

### **Results**

In previous efforts to document the characteristics of SPPS for thick (25 micron to 100 micron) films, deposition rates of up to 5.5 pounds per hour of 5 micron  $\text{Al}_2\text{O}_3$  and a deposition efficiency of approximately 65% were measured. For this effort, coating thicknesses of 15 to 20 micron were targeted and tests performed to characterize the process for producing thin films. Producing thinner films requires a slower powder feed rate and the use of finer powder. Using the 3 micron YSZ, coating rates between 0.13 to 0.60 microns/pass were studied.

For initial screening of deposition parameters, disk substrates approximately 1.5 cm in diameter were used. Hastelloy C substrates were selected solely because of their ready availability. LSM substrates were prepared by uniaxially pressing and then sintering. Two substrate heaters were evaluated for the LSM substrate; one with a maximum temperature of  $850^\circ\text{C}$  while the other was capable of  $1200^\circ\text{C}$ . Disks of LSM were heated to a selected temperature, and then the thermal gun was cycled across the face of the sample at the expected deposition conditions. After exposure, the disks were visually inspected for damage. Preheat temperatures above  $900^\circ\text{C}$  were found to be suitable, and the substrate holder is being modified for this temperature regime.

Using the Hastelloy C substrates, a basic set of plasma spray parameters were investigated in the first phase of experiments. Initial coatings were deposited on thermal shock resistant 26% porous Hastelloy substrates. The substrates were cleaned by sequential immersion in acetone and isopropanol in an ultrasonic bath. The back of the substrates were exposed to Argon during the coating process to reduce oxidation. YSZ coatings were deposited by a robot controlled plasma spray system (Plasma Technik A-3000S). Eight mole % YSZ powder (Advanced Technology Powders) with a 3.0 micron particle size was used in all experiments. The spray distance was kept constant at 5 cm and the substrate was preheated with five passes of the plasma gun. The plasma power, hydrogen flow rate, powder feed rate, injector parameters, and plasma gun traverse speed were selectively varied.

Coatings deposited at the lowest plasma power (36.5 kW) showed no indication of unmelted particles in the microstructures and a low fraction of porosity. Increased plasma power (45.5 kW), however, produced coatings with finer porosity. Modifying the injector parameters significantly reduced the coating porosity and increased the deposition rate twofold. The powder feed rates and hydrogen flow rates examined in this study did not significantly influence the coating morphology. However, high plasma power with low powder feed rates yielded coatings with excellent conformal coverage as the molten splat was able to spread over the porous substrate surface. Slowing the gun traverse speed from 350 mm/s to 245 mm/s, and, thereby, the plasma residence time, created both lateral and longitudinal cracks in the coatings while only slightly increasing the deposition rate. In addition, a greater amount of substrate oxidation occurred.

Scanning electron microscopy (SEM) was performed on polished cross sections of the samples. Very little difference in the microstructure of the YSZ coatings was observed, with all of the coatings showing evidence of grain pullout and small amounts of porosity. The YSZ coatings adhered well to the Hastelloy substrates and no delamination was observed. Inspection of the coatings revealed good thickness uniformity and no evidence of intergranular cracking was observed at the magnifications used (2 kX). With the Hastelloy substrate serving as an anode, silver paste cathodes were attached to samples with promising cell microstructures and the open circuit voltage evaluated at 750°C with air and 3% H<sub>2</sub> - 97% Ar at the respective electrodes. Cell tests samples were fabricated by first depositing a YSZ coating at low powder feed rate to obtain good surface conformal coverage, and then building up the YSZ layer at higher powder feed rates. SEM showed the coatings to be approximately 50 microns thick with

a high visible density. These cells displayed an open circuit voltage of 80% theoretical.

### **Application**

In addition to SOFC electrolyte fabrication, SPPS is also being evaluated for the formation of advanced thermal barrier coatings within the AGTSR program, the replacement of hard chrome plating for corrosion protection in Naval applications (DARPA), and other membrane applications. The very high bond strength achieved with SPPS, 10 KSI with 220 or 400 grit surface preparation, coupled with the ability to produce thin and dense coatings is also creating opportunities for dielectric, wear, and metallization.

### **Future Activities**

In the next months, further development of the SPPS process will be done with a strong emphasis on the LSM substrates. Process improvement will be based on a design-of-experiments approach on the flat substrates. This process improvement effort will drive the final design of a lathe system for processing tubular LSM substrates for process demonstration. We will also continue a parallel effort on the development of spray pyrolysis as an alternative electrolyte deposition process for YSZ.

### **Acknowledgments**

This research is sponsored by Battelle - Pacific Northwest Laboratory under order# 268884-AF2 with Northwestern University, 633 Clark Street, Evanston IL 60208-1110, Telefax (847)491-4800. The project monitor at Battelle - Pacific Northwest Laboratory is Dr. T. Armstrong. The period of performance 10/96 to 9/97.

# NONSEGREGATING ELECTROLYTES FOR MOLTEN CARBONATE FUEL CELLS

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## INTRODUCTION

In the current technology, molten carbonate fuel cells (MCFCs) employ a lithium-potassium carbonate mixture. The mixture, usually 62 m/o  $\text{Li}_2\text{CO}_3$  and 38 m/o  $\text{K}_2\text{CO}_3$  (abbreviated as 62/38 Li/K), melts below about 550°C and, when mixed with lithium aluminate, serves as both the electrolyte and gasketing for the cell and cell stack. Electrolyte segregation has been reported [1] to occur within both the cell and stack. In the cell, the segregation increases the potassium concentration near the cathode and leads to increased cathode solubility and performance decline. In the stack, the high potential difference on the gaskets causes electrolyte segregation, which in turn leads to severe performance decline of the end cells.

Argonne National Laboratory is developing molten carbonate electrolyte compositions which have minimal segregation in the individual fuel cell and cell stack under an electric field. The Li/Na carbonate electrolyte is receiving increased interest [2-5] by MCFC developers as a replacement for Li/K electrolyte. Our approach is to characterize Li/Na carbonate mixtures in terms of their segregation properties in an electric field and, if necessary, to modify the observed segregation by adding Ba and Ca carbonates.

Results of gasket strip (20 V) screening studies, as well as those from cell tests, will be discussed. We found that MCFC performance is linked to the nonsegregating property of the electrolyte. Cation distribution from segregation tests was generally linear and could be expressed as a segregation factor. A reduction in electrolyte segregation factor reduced cell polarization proportionally. The reduced cell polarization exceeds improvements that could be attributed to increased electrolyte conductivity.

## OBJECTIVE

We are identifying nonsegregating Li/Na carbonate electrolyte compositions because others have shown [2-5] that the Li/Na electrolytes show promise in extending the operating life of MCFC. We correlated cell performance with electrolyte segregation behavior. Our further objective is to

overcome problems related to electrolyte segregating and thus improve MCFC performance. A quaternary electrolyte BaCaNaLi shows promise for further improved MCFC life/performance and for reducing temperature sensitivity of Li/Na carbonate electrolyte.

## APPROACH

We use a carbonate-wetted  $\text{LiAlO}_2$  strip test to screen prospective electrolyte compositions for cation segregation and electrolyte migration. When the two cations have equal mobilities (the *Chemla Effect*), an electrolyte is considered nonsegregating and electrolyte migration should be minimized. As more convincing evidence of electrolyte performance, we test them in 100- $\text{cm}^2$  cells at 320  $\text{mA/cm}^2$ .

## PROJECT DESCRIPTION

We screened electrolyte compositions for segregating behavior by using 12-cm-long strips, wetted with carbonate and exposed to 5- to 20-V potential gradients. These conditions simulate the gasketing strip of an externally manifolded MCFC. In our case, strips of carbonate-wetted  $\text{LiAlO}_2$  (Cyprus-Foote) were made from cold-pressed powders (150 MPa). The strip was purged with a 1:2  $\text{O}_2:\text{CO}_2$  gas mixture (500 ml/min) at 655 °C. After 72 h at 20 V, the potential distribution in the strip reached equilibrium. The strips were quenched under load and later examined by metallography. These strips were sectioned for chemical analysis by inductively coupled plasma/atomic emission spectroscopy (ICP/AES). From the ICP/AES results, cation ratios and electrolyte fill in each of the sections were calculated. The variation in cation composition vs. strip length was linear; therefore, these deviations were represented as a single cation separation value, or "segregation factor," given in terms of (m/o)/cm. When the ICP/AES results of electrolyte segregation were correlated to the Li/Na carbonate composition, it was found that the nonsegregating range was 65-75 m/o Li. Surprisingly, the eutectic 52/48 Li/Na carbonate displays the greatest cation segregation. Even the Na-rich side of the eutectic composition, 40/60 Li/Na carbonate, exhibits lower cation segregation than does the eutectic.

Exploratory work with the quaternary Li/Na/Ca/Ba carbonate showed a suppression of melting point due to equimolar addition of Ba/Ca to Li/Na electrolyte. Electrolyte compositions containing 2-10 m/o Ba/Ca additions were also evaluated for cation segregation.

Bench-scale (100- $\text{cm}^2$ ) MCFC tests using off-eutectic Li/Na electrolytes show increased performance at 160  $\text{mA/cm}^2$ , as compared with the baseline cell containing the Li/Na eutectic composition. Increases in electrolyte lithium content from 52/48 to 71.5/28.5 Li/Na improved the cell potential by 75 mV. Cell tests were completed with six Li/Na electrolyte compositions: 40/60, 52/48, 60/40, 67/33, 71.5/28.5, and 75/25 (in m/o). The results show a strong correlation between nonsegregating characteristics and improved cell performance. Nonetheless, these cell tests indicate that electrolytes in the composition range of 65-75 m/o Li improve cell performance over the 52/48 Li/Na eutectic that is commonly used.

Cell area-specific impedances, a combination of IR losses and electrode polarization, for the Li/Na electrolyte cell tests show a strong correlation with the segregation factor from the strip screening tests. This correlation suggests that the nonsegregating property of the carbonate and the composition of the carbonate at the electrodes play a significant role in the MCFC performance.

The recent literature has provided comparisons of Li/K and Li/Na carbonates for performance and life. Generally, the findings favor the Li/Na carbonate [6], but an apparent drawback is its greater temperature sensitivity [7].

We have identified a range of quaternary electrolyte compositions, Li/Na/Ca/Ba carbonates, which should yield general improvement of MCFC operation and performance. These improvements include high current-density operation (250-360 mA/cm<sup>2</sup>), reduced temperature sensitivity, and anticipated extended life. Transfer of this technology to commercial developers will aid its adaptation.

## RESULTS

### The Strip "Gasket" Test

As previously described, we screen electrolyte compositions for nonsegregation using carbonate-wetted strips, which are submitted to 5 to 20 V potential gradients. After 72 h at 20V, the potential distribution in the strip has reached equilibrium. The strips were quenched under load and later examined by metallography. These strips were sectioned for chemical analysis by inductively-coupled atomic emission spectroscopy (ICP/AES). Here, 2-g samples were removed from several regions along the 12-cm strip length. From the ICP/AES results, cation ratios and electrolyte fill in each of the spots were calculated.

Based on the published Li<sub>2</sub>CO<sub>3</sub>-Na<sub>2</sub>CO<sub>3</sub> phase diagram, we chose the 52/48 m/o Li/Na eutectic composition as a baseline for the study. Figure 1 shows the ICP/AES results of samples taken at specified positions on the strips. The 52/48 Ni/Na results are compared with those of the 67/33 Li/Na composition. The variation in cation composition vs. strip length was linear; therefore, these deviations were represented as a single cation separation value or segregation factor given in terms of (m/o)/cm.

The ICP/AES results are presented in a plot of electrolyte segregation factor vs. Ca/Ba carbonate composition (see Fig. 2). The plot shows that the nonsegregating range is 67-75 m/o Li. Surprisingly, the eutectic 52/48 m/o Li/Na carbonate displays the greatest cation segregation. Even on the Na-rich side of the eutectic composition, 40/60 Li/Na carbonate exhibits lower cation segregation than does the eutectic.

The trend in electrolyte fill vs. composition is analogous to that of electrolyte segregation. The greatest change occurs with the eutectic Li/Na carbonate, which also displays the greatest cation segregation. We found that Li/Na carbonate electrolyte migration tends toward the cathode. This migration pattern is the opposite of what is found using Li/K carbonates [8]. Indeed, post-test examinations of bench-scale Li/Na cell tests have shown flooded cathodes.

The quaternary BaCaNaLi carbonates (5% and 3%) were also examined in strip tests. Cation composition vs position in the test strips is presented in Fig. 3. For both compositions, a small degree of cation segregation is apparent, with Ba/Ca concentration increasing at the positive end of the strip. The 3% BaCaNaLi is very close to a nonsegregating electrolyte with approximately 90% of the strip consisting of the starting BaCaNaLi composition.

### **Bench-scale Cell Tests**

Bench-scale 10 x 10-cm MCFC tests were assembled with standard Ni anodes (6% Cr) and Ni cathodes. The electrolyte tiles were formed by hot-pressing. The anode wet seal was aluminized with aluminum foil. Six Li/Na compositions and five BaCaNaLi compositions in m/o (10,5,4,3, and 2 Ba/Ca) were tested. All cells were operated for at least 500 h at 650 °C. To investigate the electrolyte segregation phenomenon, the cells were operated at least 320 mA/cm<sup>2</sup> for at least 1/2 h at the end of the life test. The cells were rapidly quenched to fix compositional gradients. Metallographic and chemical analysis were used to determine composition and amount of carbonate in the MCFC components.

Performance data of the bench-scale tests were used to compare the performance of Li/Na electrolyte compositions. Data on cell impedance, cell voltage, electrode performance at various utilizations and current density operations were collected as a function of time at 1 atm with laboratory standard oxidant (air + 28% CO<sub>2</sub>) and humidified fuel (80% H<sub>2</sub> + CO<sub>2</sub>).

Figure 4 shows the polarization behavior of 6 MCFC cell tests using Li/Na electrolyte compositions. These tests were conducted with fuel and oxidant utilizations of 60% and 40%, respectively, at 320 mA/cm<sup>2</sup> and 650 °C. Cell area-specific impedances, a combination of IR losses and electrode polarization, for Li/Na electrolyte cell tests are shown in Fig. 5. The cell tests displayed improved performance as the electrolyte composition approached the nonsegregating range. Compared to the baseline 52/48 carbonate composition, MCFC voltage increased about 100 mV at 160 mA/cm<sup>2</sup> for the off-eutectic Li/Na carbonate electrolyte compositions, 65-75 mol% Li<sub>2</sub>CO<sub>3</sub>.

High current density operation in Li/K carbonate cells leads to early cell shorting [9]. Electrolyte segregation phenomena or electrolyte freezing presents problems for high current-density operation. The 71.5/28.5 Li/Na composition exhibited a cell potential of 0.76 V at 320 mA/cm<sup>2</sup>, but this appears to be the upper limit of improved cell performance due to higher Li content electrolytes. The cell performance of the 75/25 Li/Na electrolyte composition (0.71 V at 320 mA/cm<sup>2</sup>), although still much improved compared to the 52/48 LiNa, is lower than that of the 71.5/28.5 Li/Na electrolyte cell. Nonetheless, these cell tests suggest electrolytes in the composition range of 65-75 m/o Li improve cell performance over the 52/48 Li/Na eutectic that is commonly used.

We find an apparent correlation between improved MCFC performance and nonsegregating electrolyte compositions. The polarization behavior of 6 MCFC cell tests using Li/Na electrolyte compositions is taken from Fig. 5. These tests are conducted with fuel and oxidant utilizations of 60% and 40%, respectively, at 320 mA/cm<sup>2</sup> and 650 °C. As in Fig. 3, these deviations were represented as a single cation separation value or segregation factor, given in terms of (m/o)/cm. Cell area-specific impedances, a combination of IR losses and electrode polarization, for Li/Na electrolyte cell tests are now correlated with a segregation factor from the strip screening tests (see Fig. 6). This correlation

suggests that the composition of the carbonate at the electrodes plays a significant role in the MCFC performance. Only a minor portion of the reduced cell polarization can be attributed to increased electrolyte conductivity that is associated with the increased  $\text{Li}_2\text{CO}_3$  content of the nonsegregating Li/Na composition. Post-test examination of MCFC cell tests will investigate this observation in support of our continued electrolyte development work.

## APPLICATION

The quaternary carbonate electrolyte Ba/Ca/Na/Li combines a nonsegregating property with a reduced melting point. Differential thermal analysis (DTA) shows a 100°C lower melting point for the BaCaNaLi electrolytes (450°C) than the Li/Na carbonate eutectic.

The results of 10 x 10-cm MCFC test with quaternary BaCaNaLi carbonate electrolytes showed a prospect for reduced temperature operation (Fig. 7). These cells displayed stable cell potentials for more than 1000 h at 160 mA/cm<sup>2</sup> (60/40 % fuel/oxidant utilization). Cell performance and polarization curves were taken under conditions similar to those of the 52/48 Li/Na carbonate cell. The five cells were operated over a temperature range of 550-700°C to establish performance vs. temperature characteristics. Although not shown, the baseline 52/48 Li/Na and 10% BaCaNaLi have very similar temperature characteristics. At 550°C, the 3% BaCaNaLi carbonate cell had a stable voltage 75 mV greater than that of the baseline 52/48 Li/Na. At 550°C, the 2% BaCaNaLi carbonate cell exhibits a 25 mV improvement. Unlike some of the earlier quaternary BaCaNaLi carbonate electrolytes, both 2% and 3% BaCaNaLi could be operated at 320 mA/cm<sup>2</sup>. The quaternary 3% BaCaNaLi carbonate electrolytes shows promise for improved cell stability at 320 mA/cm<sup>2</sup> and a 50-75°C lower operating temperature.

## FUTURE WORK

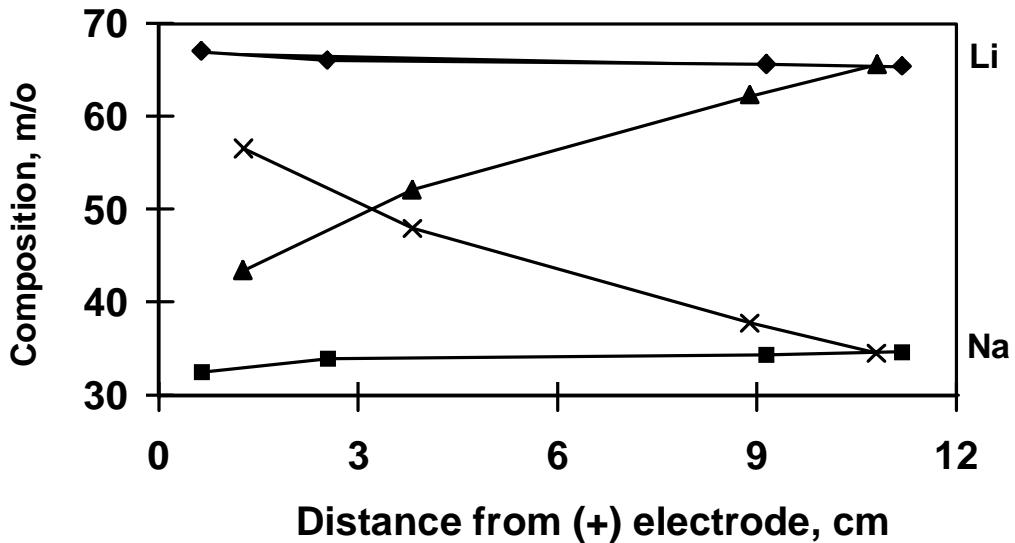
Future work will concentrate on life testing of bench-scale MCFCs for a selected range of quaternary electrolyte compositions. We will continue our investigation of the carbonate composition at the electrode/electrolyte interface with postoperative analysis. The primary focus will be to develop MCFCs with advanced quaternary compositions to learn how they affect MCFC performance. The next step in our advanced electrolyte development is to provide the physical properties to assist with the MCFC design (i.e, wetting properties for proper electrode microstructure development). To facilitate DOE-contractor adaptation of our electrolyte, we are modifying our MCFC testing to accommodate tape-cast components. Having comparable MCFC tests will aid us in addressing specific application details.

## ACKNOWLEDGMENT

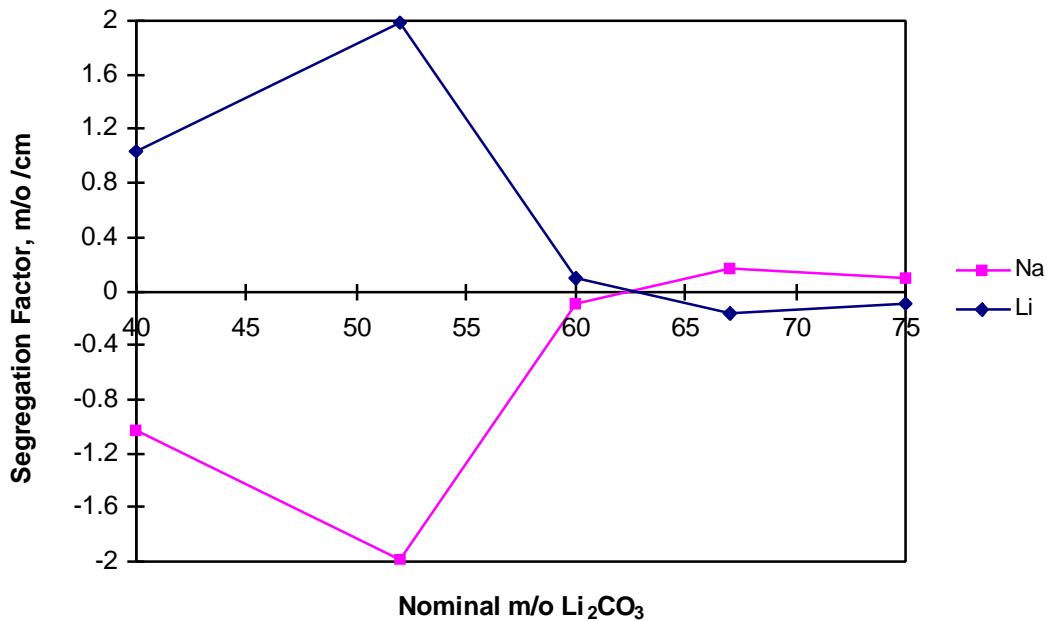
Research sponsored by the U.S. Department of Energy through the Federal Energy Technology Center under Contract No. W-31-109-ENG-38 with FETC Project Manager, Richard Johnson; Contractor Project Manager, Kevin M. Myles, and Principal Investigator, Michael Krumpelt. This contract covers work done in FY 1997.

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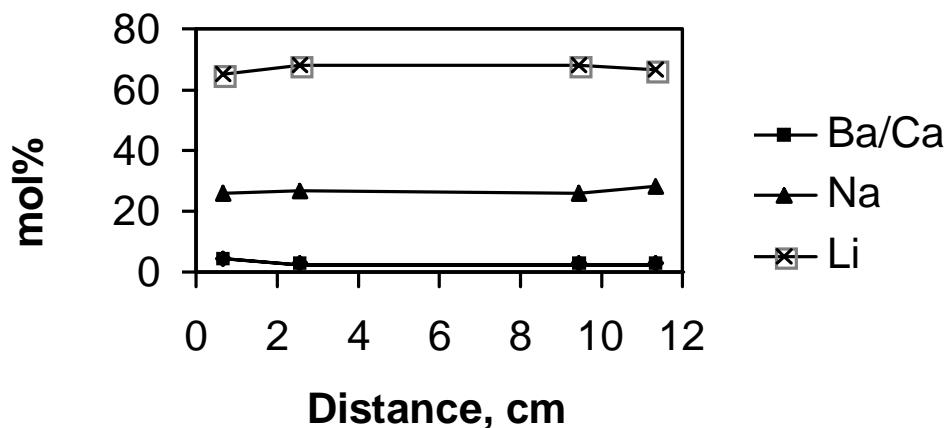


**Fig. 1. Fully-developed cation composition distribution of 12-cm-long strips (with 20V applied) for two Li/Na carbonate compositions: 52/48 and 67/33.**

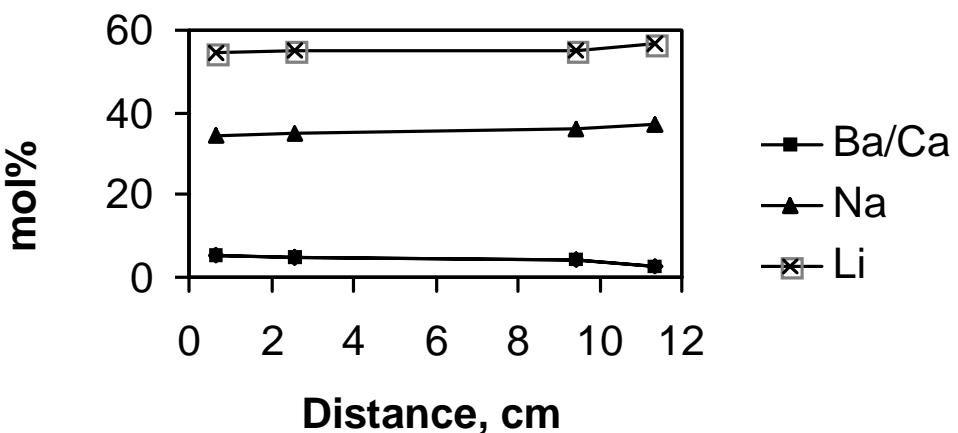


**Fig. 2. Segregation factors versus composition for a range of Li/Na carbonate electrolytes.**

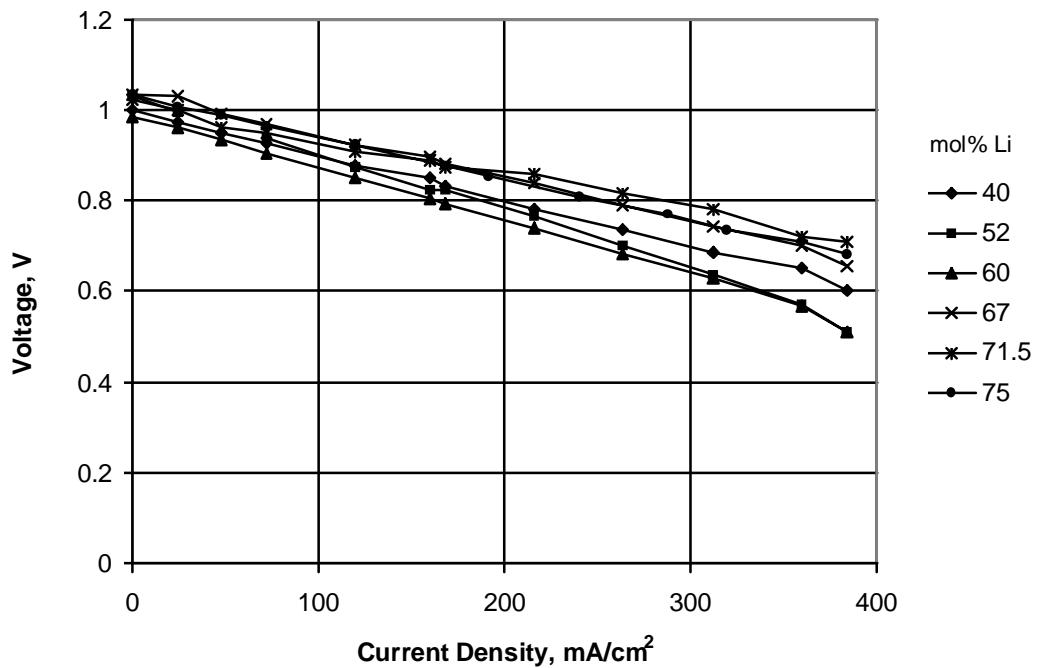
**(a) 3% BaCaNaLi Strip Test Composition**



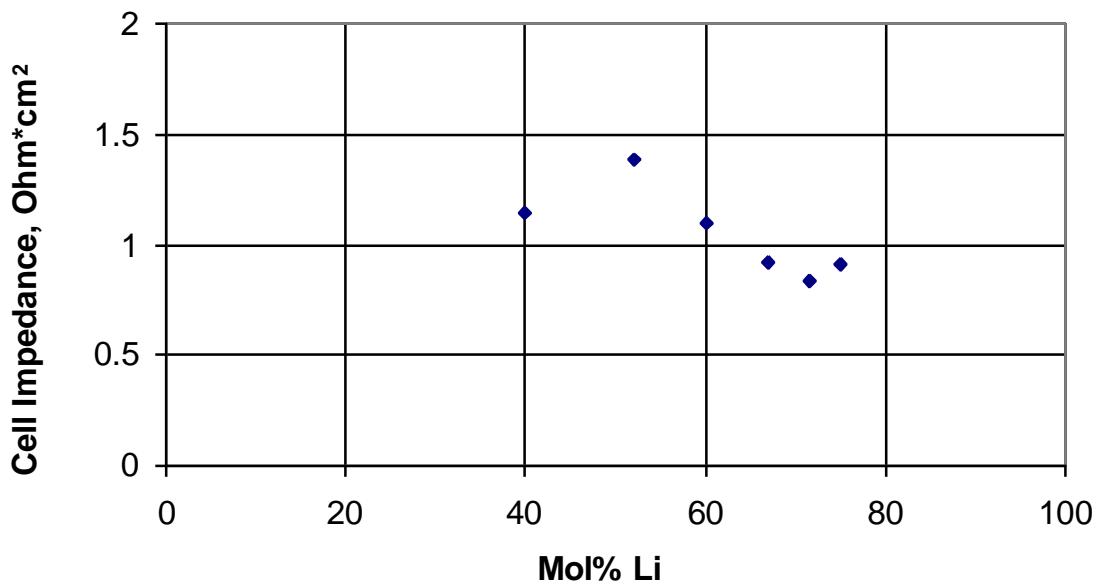
**(b) 5% BaCaNaLi Strip Test Composition**



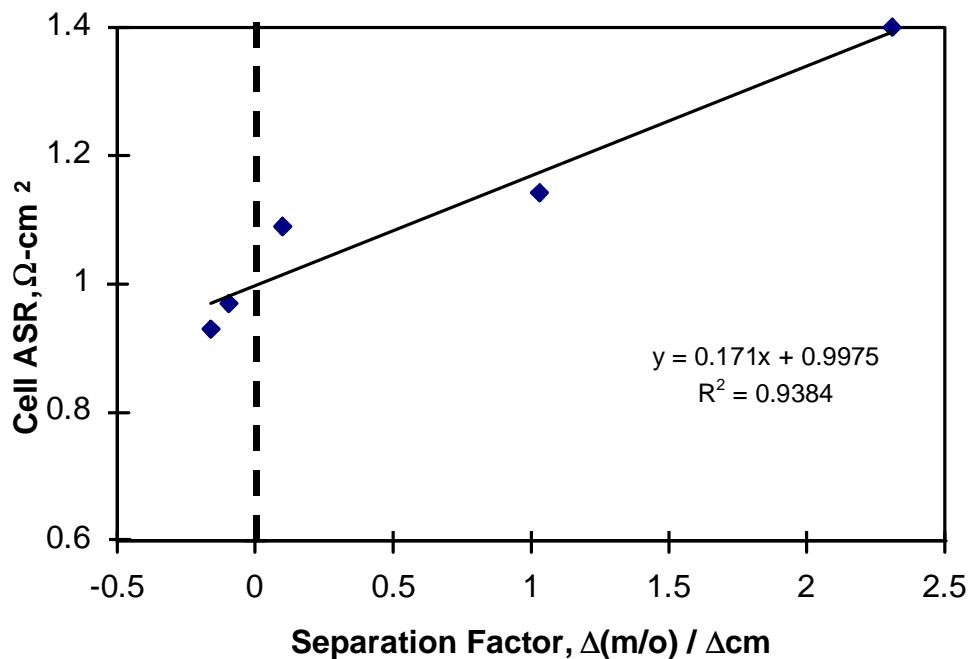
**Fig. 3. Fully-developed cation composition distribution of 12-cm-long strips (with 20 V applied); with (a) 3% BaCaNaLi (3/3/28/66 m/o carbonate) and (b) 5% BaCaNaLi (m/o 5/5/38/52 carbonate) electrolyte.**



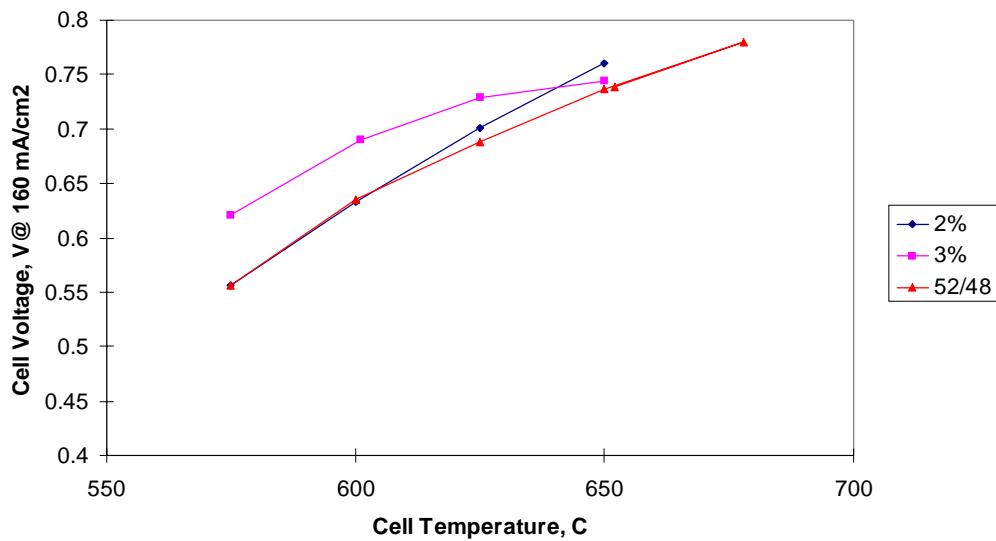
**Fig. 4.** Polarization curves indicating cell impedance as related to electrolyte composition: 6 Li/Na compositions (in m/o) 40/60, 52/48, 60/40, 67/33, 71.5/28.5 and 75/25.



**Fig. 5.** Cell impedance as related to electrolyte composition: 6 Li/Na compositions (in m/o) 40/60, 52/48, 60/40, 67/33, 71.5/28.5 and 75.25.



**Fig. 6.** MCFC cell impedances for range of Li/Na carbonate composition: (40/60 to 75/25 mol%) compared to separation factors established from our strip segregation-screening tests.



**Fig. 7.** Temperature sensitivity of three MCFCs having various electrolyte compositions: 2% and 3% BaCaNaLi, along with baseline 52/48 Li/Na carbonate. Cells operated under standard conditions at 160 mA/cm<sup>2</sup>.

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## 200 kW B Anaerobic Digester System

### ABSTRACT

More than 90% of the wastewater treatment plants (WWTPs) in the United States generate anaerobic digester gas (ADG) as a by-product of their operation. ADG is a mixture of gases, mainly methane (generally 60%) and carbon dioxide. When ADG is released uncombusted, it contributes significantly to the greenhouse effect, as methane traps 10 to 20 times more heat than carbon dioxide does. Therefore, excess ADG is typically flared in flame towers. However, flaring is only a partial solution, since ADG combustion generates photoreactive ozone precursors such as nitric oxides (NOx) and volatile organic compounds (VOC). As a result, WWTPs are regulated as stationary sources of air pollution under the Clean Air Act. WWTPs situated in heavily urbanized areas in severe ozone non-compliance areas such as Southeastern New York State and emitting in excess of 25 tons/year of NOx and VOC are designated and regulated as major sources. This designation requires installation of extensive control and monitoring technologies. The Authority serves at least 14 WWTPs in New York City and Westchester County that fall into this category.

Fuel cells (FCs) provide effective solutions to these problems by efficiently generating premium quality electricity and much-needed heat while consuming ADG and emitting orders-of-magnitude smaller amounts of NOx and VOC. Additionally, integration of FCs with an inexpensive or free renewable fuel resource such as ADG could be a major step in reducing FCPP electricity costs and expediting commercialization of FC technology.

Many of the Authority's WWTP customers could be equipped with multiple FC power plants (FCPPs) with a capacity of approximately 200 kW at each site. Total production potential is estimated at between 5 and 10 MW. FCs have a potential to generate valuable emission credits, assure compliance with present and future Clean Air Act requirements, and substantially reduce air pollution from WWTP flares in Southeastern New York State. This would be an important service to wastewater treatment customers, reducing their operating costs and regulatory burdens. The FC program would also benefit New York State industries, since New York companies would manufacture at least 29% of the equipment.

In its first FC project, the Authority is joining forces with Westchester County to install a FCPP at the Yonkers Joint Wastewater Treatment Plant. The U.S. Department of Energy (US DOE) the New York State Energy Research and Development Authority and the Electric Power Research Institute have helped the Authority to develop and finance the project, which is the first installation of its kind nationally. The project received the \$200,000 grant # DE-FG21-96MC33354 from the US DOE Climate Change Fuel Cell Program administered by Morgantown Energy Technology Center officers Ms R. Diane Manilla and Ms Diane Hooie. International Fuel Cells Corporation (IFC) manufactured the FCPP of Connecticut. It commenced operation in April, 1997. The standard IFC PC25C power plant was modified to accept a lower BTU ADG and to pass through up to 40% of carbon dioxide. Also, an ADG pretreatment system to remove water, sulfur bearing compounds, and halogens was developed with the US DOE assistance.

ADG is partially utilized and partially flared at the Yonkers Joint Sewage Treatment Plant. An FCPP captures a portion of the otherwise flared ADG, use it as fuel and produce approximately 200 kW of electricity and an ample amount of heat for various uses at the host plant. One or two more fuel cells could be installed at this site in the future to capture 100% of the otherwise wasted ADG.

Potential FC benefits extend far beyond WWTPs. FC development is reaching its maturing status at a time of emerging deregulation in the electric utility industry. FCs offer high efficiency, very low emissions, available thermal energy, fuel flexibility, potential mobility and premium quality electricity in distributed generation applications. FCs may allow the Authority to reach customers directly, bypassing competitors distribution systems and avoiding their delivery charges. For these reasons, the Authority is gaining hands-on operating experience and assessing the role FCs may play in distributed generation applications and in the rapidly unfolding competitive energy markets.

# An Integrated Fuel Processor For PEM Fuel Cells

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## Introduction

Proton-exchange membrane fuel cells (PEMFCs) are of interest for many applications including transportation, commercial vehicles, residential and commercial power generation, emergency power supply, and small portable power supplies. In some cases, commercial utility can be realized by supplying hydrogen to the PEMFC stacks from a cylinder of compressed gas. However, for many applications, economic feasibility depends on the successful development of a practical and low-cost fuel processor. Despite extensive efforts to develop partial oxidation (POX) and autothermal reforming for this application (Kumar et al, 1996; and Recupero et al, 1996), these fuel processing methods have not proven to be entirely satisfactory and a new approach is needed.

Northwest Power Systems (NPS) is developing a family of versatile fuel processors based on a novel design that combines steam reforming with hydrogen purification. The resulting integrated fuel processor promises to be affordable, highly compact, and lightweight. Moreover, the NPS fuel processor offers advantages over alternative conventional processes including POX, autothermal reforming, and steam reforming.

## Objective

The goal of this program is to design, build, and test a prototype fuel processor that meets the following requirements:

1. capable of recovering 70% to 80% of the available hydrogen;
2. good load-following characteristics;
3. capable of delivering hydrogen containing <10 ppm CO; and
4. low cost.

Furthermore, it is preferable that the fuel processor be compact in size and lightweight. High-purity product hydrogen from the fuel processor (>99% pure) will allow the fuel cell stack to deliver the highest possible power density for a given set of operating conditions (Inbody et al, 1996).

## Approach

To achieve these goals, NPS has completed the design of an integrated fuel processor that combines steam reforming, heat production, and hydrogen purification into a single device. The key features of the integrated fuel processor are shown in Figure 1. High-pressure steam reforming, rather than POX or autothermal reforming, is used for the production of hydrogen from a feedstock for the following reasons:

1. hydrogen yields are significantly greater;
2. hydrogen purification is more readily accomplished since POX and autothermal reforming are relatively low pressure operations that result in significant dilution of the product hydrogen by nitrogen (from air); and
3. overall energy efficiencies typically are highest for steam reforming.

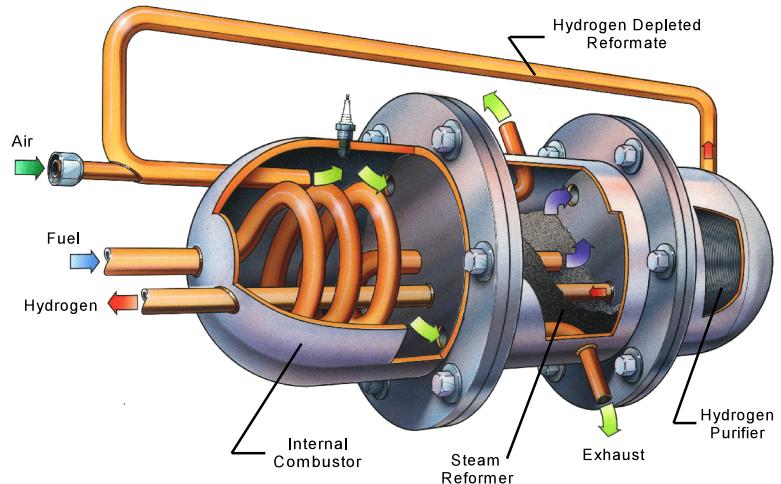


Figure 1. Cut away view of the NPS integrated fuel processor.

Steam reforming is conducted at elevated pressures (about 100 psig to 250 psig) using commercial catalysts and temperatures in the range of 300°C to 600°C. The reformate stream is purified using a two-stage purifier that also operates within the same temperature range, allowing direct integration of the reformer and purifier without the need for intermediate heat exchange. The first stage of the purifier performs a bulk separation of hydrogen from reformate and consists of a hydrogen-permeable metal membrane. The second stage of the purifier serves to reduce the CO and CO<sub>2</sub> content of the hydrogen to very low levels. Thus, the product hydrogen is >99% pure and contains <10 ppm CO and <50 ppm CO<sub>2</sub>.

A unique feature of the NPS integrated fuel processor is that the two-stage hydrogen purifier is minimized in size and cost by limiting hydrogen recovery to only 70% to 80% of the hydrogen generated by steam reforming. The balance of the hydrogen, along with other byproduct gases and unreacted feedstock, is then conveniently used as a gaseous fuel to provide the required heat for (a) vaporizing the liquid feedstock and water, (b) heating the vaporized feedstock to the reforming temperature, and (c) providing heat to the catalytic reforming bed to satisfy the steam-reforming reaction enthalpy. This approach eliminates the requirement for an external burner fired with unreacted feedstock. The net result is high overall conversion to hydrogen at a reduced cost.

## Results

The technical feasibility of this integrated fuel processor has been demonstrated at the bench scale by generating 2.5 L/min of product hydrogen. The product hydrogen was >99% pure and contained no detectable CO and CO<sub>2</sub> (<2 ppm CO and <2 ppm CO<sub>2</sub> given the limits of detection). With this bench-scale prototype fuel processor we have demonstrated overall energy efficiencies (using methanol/water mix as the feedstock) of 70% to 75% (HHV). Efforts are presently underway to scale up the fuel processor to deliver 50 L/min product hydrogen, sufficient for a nominal 5 kW PEMFC stack.

It has been necessary to iterate the design of the combustor and hydrogen purifier as part of the scale-up effort. In particular, the design of the combustor has been improved with respect to heat transfer and low pressure drop. The previous generation of combustor utilized a long tube arranged as a spiral and placed within the catalytic reforming bed. However, this design suffered from moderately high pressure drop and large (about 100°C to 200°C) temperature gradients. We now favor a new design as shown in Figure 1 in which a combustion chamber vaporizes and superheats the feedstock and then exhausts through one or more straight tubes that pass through the reforming catalyst bed.

The two-stage hydrogen purifier is currently being designed as a compact brazed-plate module. Previously we were utilizing a tubular design, but this proved to be too bulky.

Steam reforming must be conducted at pressure to provide the driving force necessary for hydrogen separation using the first stage (membrane) of the purifier. Figure 2 shows the relationship between the reforming pressure and the required membrane area. Since the membrane is composed of an alloy of palladium, it is important to minimize membrane area to achieve acceptable costs.

There is reduced benefit to operating the reforming reactions at pressure much greater than about 250 psig, and the parasitic power load for pumping the liquid feedstock to this pressure is insignificant. Therefore, this is the target operating pressure for the fuel processor. Fortunately, nearly all commercial feedstocks are available as liquids, including methanol, ethanol, propane, gasoline, diesel, and jet fuel. The only commonly used feedstock that is not readily available as a liquid is natural gas. So, for natural gas the optimum reforming pressure is likely to be much less than 250 psig, perhaps about 100 psig, although an optimized system design for natural gas has not been completed.

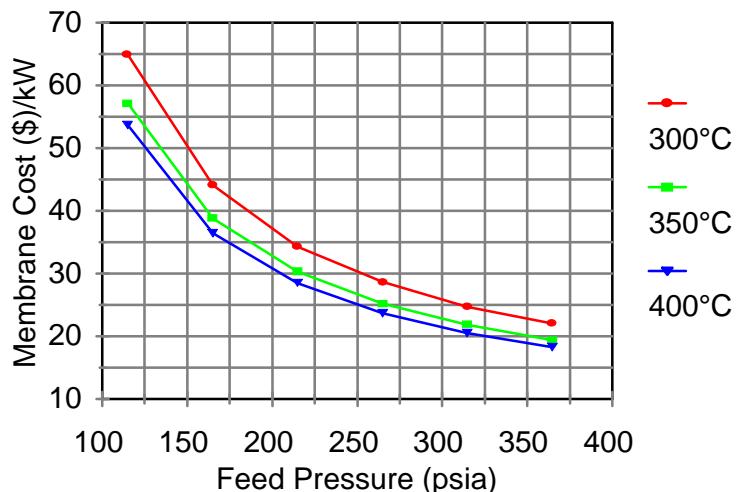


Figure 2. Required area of palladium-alloy membrane as a function of the reforming pressure and operating temperature. Basis: 65% hydrogen in reformat, 75% hydrogen recovery, product hydrogen at ambient pressure.

## Applications and Benefits

Obviously, it is important to evaluate more than just the technical merits of a potential fuel processor for use in PEMFC systems--the economics of the fuel processor must also be compared and contrasted to the economics of other fuel processor options. More to the point, the economics of the entire fuel cell system should be evaluated to determine the impact of the choice of fuel processor on the capital and operating costs to

the end user. In this type of analysis the NPS integrated fuel processor shows significantly lower capital and operating costs in comparison to other conventional fuel processing methods. For example, Table I qualitatively compares and contrasts the NPS integrated fuel processor and a typical POX reactor using methanol as the feedstock (similar results are obtained when this evaluation is conducted using propane, ethanol, or other feedstocks in place of methanol).

Table I. Characteristics affecting the economics of the NPS integrated fuel processor and a typical POX reactor, both operating on methanol.

Characteristic	NPS Integrated Fuel Processor	POX Reactor
Process	Steam Reforming	Partial Oxidation in Air
H <sub>2</sub> Concentration in Reformate	65% to 75%	30% to 35%
Subsequent Purification Operations	None Required	Low Temperature WGS, Selective CO Oxidation
Product H <sub>2</sub> Purity	>99%	40%
Moles H <sub>2</sub> Produced/Mole Methanol Consumed	3	2
H <sub>2</sub> Recovery	>70%	100%
H <sub>2</sub> Utilization	98%	60%

The primary difference between the operation of the NPS integrated fuel processor and the operation of a POX reactor is in the purity of hydrogen produced by each method. As shown in Table I, the purity of hydrogen produced by the NPS fuel processor is very high, whereas the POX reactor cannot deliver high purity hydrogen. The low cost of the POX reactor, an often stated advantage, is offset by the requirement of subsequent purification steps (WGS reactor and selective oxidation) and heat exchangers. The requirement for these subsequent operations also makes the POX reactor considerably larger, heavier, and more complex than the NPS fuel processor.

An improvement to POX is autothermal reforming, in which a reforming catalyst is placed within the reactor and supplemental water is injected to achieve slightly higher hydrogen concentrations in the product stream. However, while this approach raises the hydrogen content from about 40% to 50% (Kumar et al, 1996), it has the disadvantage of increasing the cost and complexity of the system by requiring the addition of a catalyst bed and water injection.

Another significant economic advantage of the NPS integrated fuel processor is that it has a high yield of hydrogen per unit of feedstock consumed. This results from the use of water as the oxidant--water contains chemically bound hydrogen that is released as product hydrogen during the steam reforming process.

Now, turning our attention to the integration of these two fuel processing methods into a standard PEMFC system rated to deliver nominally 5 kW, we see another economically significant difference between the two approaches. Specifically, the low purity hydrogen delivered by the POX reactor yields (a) relatively low power output from the PEMFC stack, and (b) low utilization of the hydrogen fed to the stack. The low power output from the fuel cell stack is caused by both the low hydrogen partial pressure (given a fixed total anode gas pressure) and mass transfer resistance that appears as hydrogen is consumed at the anode. Since the hydrogen partial pressure continues to decrease as hydrogen is consumed, and mass transfer resistance continues to increase, the net result is relatively low hydrogen utilization. This problem is manifested in the form of an increased system cost (since the fuel cell stack must be increased in size in the case of POX) and increased feedstock utilization rate. These results are summarized in Table II.

The analysis in Table II leads us to conclude that the NPS fuel processor will lead to lower overall operating costs due to a higher energy efficiency, which in turn results from higher purity of hydrogen delivered to the PEMFC stack. However, it is also expected, based on this analysis, that the capital cost of a PEMFC system using the NPS integrated fuel processor will be less than that for a system using POX or related fuel processing methods. The difference in capital cost is most directly attributed to the difference in PEMFC stack gross power rating (i.e., stack size). For this analysis, the estimated cost for the system assumes the cost of the stack and all supporting hardware (excluding fuel processor) is \$1,500/kW. The NPS fuel processor is projected to cost \$400/kW, and the POX reactor with subsequent WGS reactor, selective oxidizer, and heat exchanger, is estimated to cost about \$550/kW.

## **Future Activities**

Activities are currently in progress to scale up the integrated fuel processor to 5 kW. A prototype 5 kW fuel processor is anticipated by the end of the year. Since the integrated fuel processor can utilize a range of different feedstocks in addition to methanol, NPS is also directing a portion of its effort at producing hydrogen from propane and other selected feedstocks with the goal of demonstrating, during 1998, a family of fuel processors operating on a range of feedstocks.

Table II. Performance and economic comparison of two 5 kW (net) PEMFC systems based on the NPS fuel processor and a POX reactor, assuming methanol is the feedstock.

Parameter	NPS Integrated Fuel Processor	POX Reactor
PEMFC Gross Power	5.85 kW	7.25 kW
Parasitic Load (Total)	about 17%	about 16%
H <sub>2</sub> Utilization	98%	60%
Feedstock Efficiency (@ 0.6V/cell)	0.128 Gal. Methanol/kW (Gross)	0.219 Gal. Methanol/kW (Gross)
Feedstock Consumption Rate (5 kW Net)	0.75 Gal. Methanol/Hr.	1.6 Gal. Methanol/Hr.
System Cost (Est.)	\$11,000	\$15,000

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# RESEARCHES AND DEVELOPMENT OF A FUEL CELL WITH PROTONIC CONDUCTIVITY

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## ABSTRACT

Technology and research results of single fuel cells (SFC) on the basis of  $\text{BaCe}_{0.9}\text{Nd}_{0.1}\text{O}_{3-\delta}$  (BCN) electrolyte with the 78 % Ni + 22 % BCN anode and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  cathode, and also of experimental FC stack, assembled from six SFC, are stated.

## INTRODUCTION

Development and perfection of high efficient and low emission energy sources on the basis of fuel cells, realizing a principle of direct electrochemical transformation of hydrocarbon natural fuel to electricity, is a dominant scientific and technical problem in power generation and has primary practical application.

Operation experience of power plants on the basis of various FC types (alkaline, phosphoric acid, molten carbonate) alongside with advantages demonstrates technical shortcomings, main of which is the presence of liquid components in FC systems. Therefore solid electrolytes on the basis of complex oxide compositions of rare earth metals have the doubtless prospect.

High efficient SOFC-based energy sources are possible, if SOFC materials with opportunities of fast ion transport, low power barriers for transition through "gas-electrolyte" interface and sufficient operational reliability could be developed. Alongside with common materials, such as stabilized zirconium dioxide (YSZ), alternative materials on the basis of cerates and, first of all, barium cerates (BC) are of significant interest for the researchers. The possibility of essential decrease of BC-based fuel cell operation temperature has predetermined outburst of interest to the development of SOFC with new electrolyte.

The work performed at the Russian Federal Nuclear Center under the contract with EPRI is devoted to the research and development of a fuel cell on a basis of  $\text{BaCe}_{0.9}\text{Nd}_{0.1}\text{O}_{3-\delta}$ .

The fragment of this work, covering SFC research, made under the scheme: 78%Ni + 22 % BCN /  $\text{BaCe}_{0.9}\text{Nd}_{0.1}\text{O}_{3-\delta}$  /  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ , and also characterization of experimental FC stack, assembled from six SFC is reported.

## TECHNOLOGY AND TECHNIQUE OF SFC RESEARCHES

The electrolyte membranes were made from powders of barium carbonate ( $\text{BaCO}_3$ ), cerium dioxide ( $\text{CeO}_2$ ) and one and a half neodymium oxide ( $\text{NdO}_{1.5}$ ). The weighed components were mixed and rubbed with ethyl alcohol, and then dried at 150-200°C during 2 hours. To synthesize preliminary perovskite structure the obtained mixture was calcined in two steps at temperatures 900 and 1200 °C during 1 hour in platinum crucibles. Powder was dry milled after each calcination.

Then a powder was entered at hashing into 10 % solution of polyvinyl butyral in ethyl alcohol with dibutyl phthalate, solvent was removed and received paste was progressively rolled to reach required thickness. Membrane samples were cut out from green rolled sheets, which after initial annealing (500 °C) were sintered in air at 1490-1500°C during 2 hours. Thickness of received membranes was 0,5 mm.

Anodic paste consisting of 78% Ni + 22% BCN (19-23 mg/cm<sup>2</sup>) was put on one side of a membrane and it was fired to electrolyte in air at 1350°C for 2 hours, and cathodic paste consisting of  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  (76-94 mg/cm<sup>2</sup>) was put on the other side and fired at 1250°C for 2 hours.

Then SFC was placed in a measuring unit and reduction of nickel oxide to metal on anodic coating was fulfilled. Characteristics were investigated at supply of humidified hydrogen to anode (consumption 5 l/hr), and humidified air - to cathode (consumption 24 l hr).

### RESULTSFSCRESEARCH

Voltage-current SFC characteristics in a temperature range 700-900°C are given in Fig.1 and have linear nature. The calculated ion transport numbers ( $t_i$ ) vary from 0,9 at 900 °C to 0,94 at 700 °C. Current density at  $U = 700$  mV and temperature 800 °C lays in the range 50-55 mA/cm<sup>2</sup>.

SFC power as a function of current density is given in Fig.2. It can be seen, that I-P curves pass through a maximum. The maximum power increases with temperature growth and makes up 63-65 mW/cm<sup>2</sup> at 900 °C.

SFC ohmic losses at different temperatures are given in Fig.3.

It was not possible to measure anodic polarization with satisfactory accuracy. But we established, that maximum values of anodic overvoltage did not exceed 30mV. The main contribution to SFC polarization losses is due to cathodic overvoltage (Fig.4).

Researches of BCN electrolyte serviceability in various fuel mixes, containing 20, 30, 40 vol. %  $\text{CO}_2$  in fuel cell:  $\text{H}_2 + \text{H}_2\text{O} + \text{CO}_2$ , Pt/BCN/Pt, air, were also carried out. In this case we used  $\text{BaCe}_{0.9}\text{Nd}_{0.1}\text{O}_{3-\delta}$  membrane 1 mm thick. Porous platinum was used as electrodes. Working area of SFC was 1,54 cm<sup>2</sup>. SFC total power as a function of fuel composition at 600-800 °C is represented in Fig.5. SFC electrical characteristics at long-term operation with pure humidified hydrogen, and with fuel mix, containing 20%  $\text{CO}_2$  after 130 minutes of work, are shown in Fig.6.

The researches testify, that BCN-electrolyte with platinum electrodes keeps long-term serviceability in pure humidified hydrogen, but the  $\text{CO}_2$  presence in fuel mix leads to irreversible deterioration of its electrochemical properties in a short interval of time. It result in a complex cell degradation: formation of barium carbonate in "electrolyte-electrode" interface, change of electrolyte structure, elamination of electrode coating.

Study of SFC electrical characteristics stability in time was carried out at temperature 905±6°C under the scheme:  $\text{H}_2 + \text{H}_2\text{O}$ , Ni + BCN/BCN/MLS, air. The resistance of an external load was chosen so that SFC gave out the maximum power. The electrical SFC characteristics (Fig.7) demonstrate stable work during 106 hours.

## EXPERIMENTAL SFC STACK AND ITS CHARACTERISTICS

The doubtless interest in FC researches is the study of not only SFC behavior, but also, as the general purpose - research of SFC set, assembled in stack.

For realization of such researches the round BCN plates 26 mm in diameter and 1 mm thick were made by pressing (Fig.8). Electrodes (anode: 78 % Ni + 22% BCN, cathode:  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ ) were put on and fired to plates surfaces. Platinum current collectors were fired by means of current-conducting glass to cathodic and anodic surfaces. SFC stack is a series of alternating anodic and cathodic gas cavities, separated by solid electrolyte. Channels for gas input and output are located perpendicularly to each other. SFC number in a stack is 6 pieces (Fig.9).

The SOFC stack is located in the ceramic frame with covers fixed in ceramic casing, in which supply of fuel and oxidizing gases to stack and removal from it are made (Fig.10).

The metal casing with SOFC stack is placed in the central part of electrical heater of the experimental block, the space between the stack frame and heater is filled with heat insulation (Fig.11, 12).

The researches were carried out with  $\text{H}_2 + 2,5$  volume%  $\text{H}_2\text{O}$  as a fuel (consumption 4 l/hour) and with air as an oxidant (consumption 40 l/hour).

The individual characteristics of SFCs assembled in stack are represented in Fig.13, 14. Characteristics of FC stack are shown in Fig.15, 16.

Characteristics of FC stack, consisting of six connected in parallel SFC (total area 18,8 cm<sup>2</sup>) at temperature 650 °C were received as follows:

open circuit voltage  $E = 0,866$  V, maximum current  $I_{\max} = 0,418$  A,  
current density  $J_{\max} = 22,2$  mA/cm<sup>2</sup> at  $U = 0,44$  V,  
maximum power  $P_{\max} = 0,184$  W, power density - 9,8 mW/cm<sup>2</sup>.

## CONCLUSION

Carried out researches of fabrication technologies and characteristics of FC on the basis of BCN-ceramics ( $\text{BaCe}_{0,9}\text{Nd}_{0,1}\text{O}_3\delta$ ) allowed to obtain additional scientific data, which testify the possibility of using investigated compositions in medium temperature solid oxide fuel cells with protonic conductivity, taking into account limited  $\text{CO}_2$  content in a fuel gas.

## ACKNOWLEDGEMENTS

The authors thank Electric Power Research Institute (USA) and Project Manager Dr. R. Goldstein for financing of this work and development of friendly scientific relationships between EPRI and Russian Federal Nuclear Center for the sake of peaceful interests of two countries.

## REFERENCE

1. Litvinov B.V., Kleshev Yu.N. et al. Researches and Development of a Fuel Cell with Protonic Conductivity. The reports: Stage 1 and Stage 2, RFNC - VNIITF, 1995/1996., Under contract № WO1676-20/8062-10 with EPRI (306 pages, 174 figures, 37 tables, 159 references).

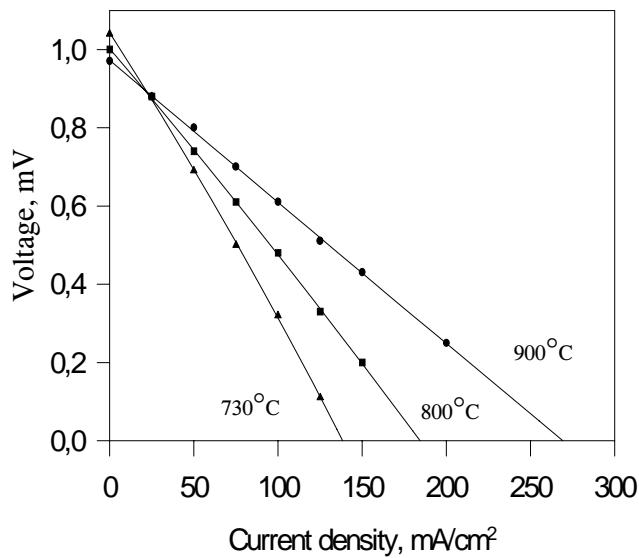


Fig. 1. SFC voltage-current characteristics

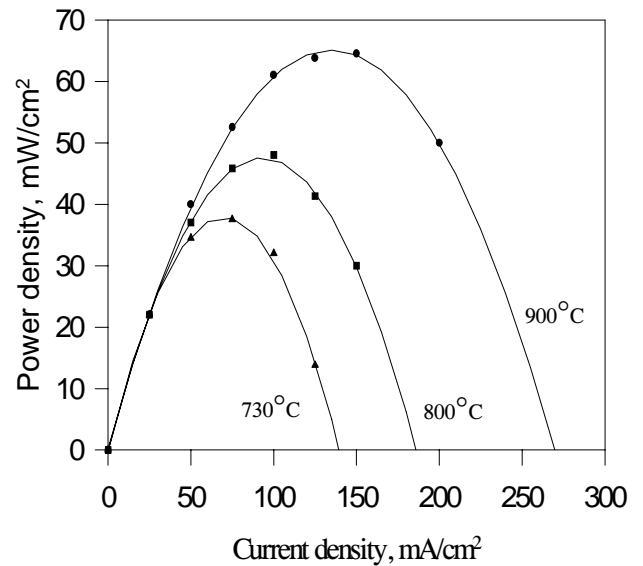


Fig. 2. SFC power vs current density

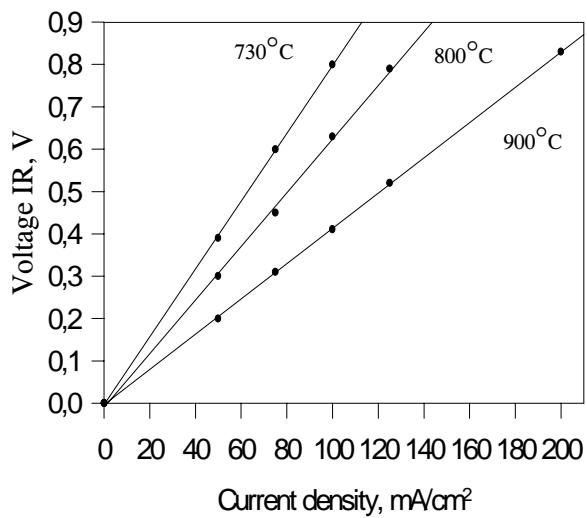


Fig. 3. SFC ohmic losses vs current density.

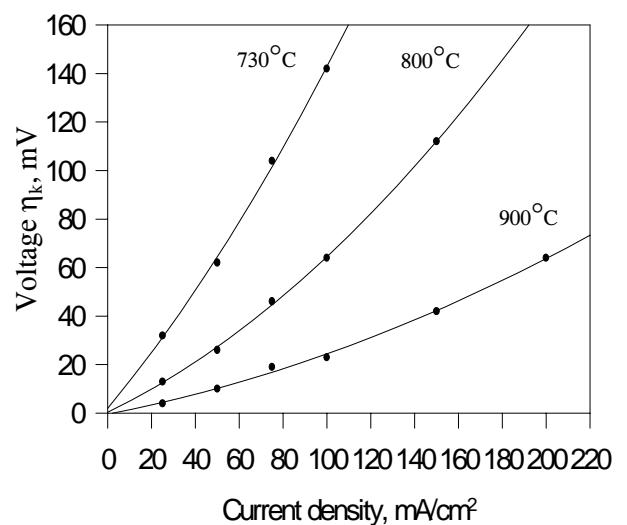


Fig. 4. SFC cathodic polarization.

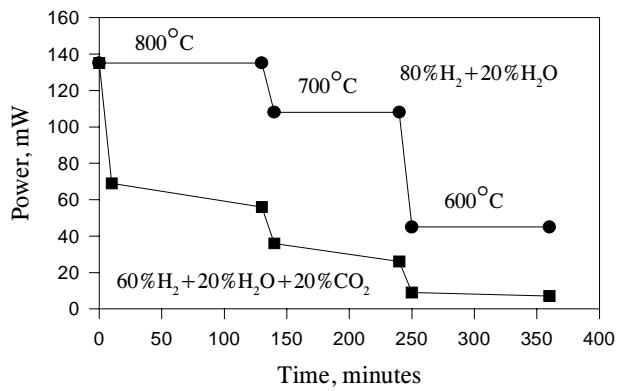


Fig. 5. SFC maximum power vs temperature and operation time in different fuel mixes

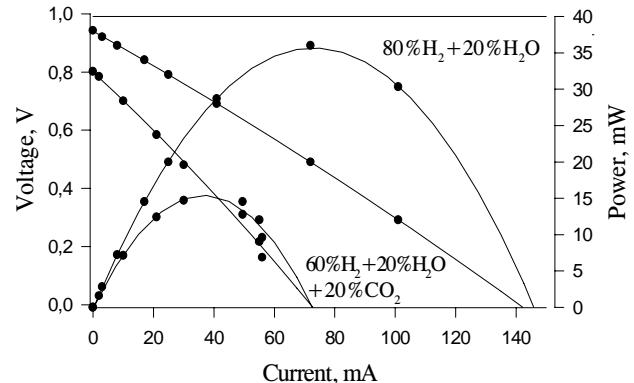


Fig. 6. Characteristics of SFC long-term operation with pure humidified hydrogen and with  $\text{CO}_2$ -containing mix after 130 minutes operation

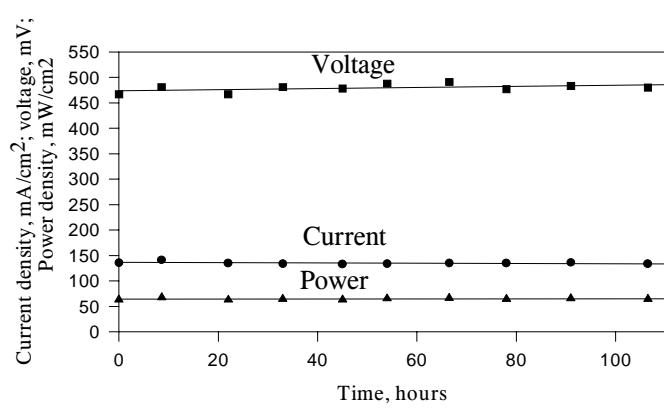


Fig. 7. SFC electrical characteristics vs. time

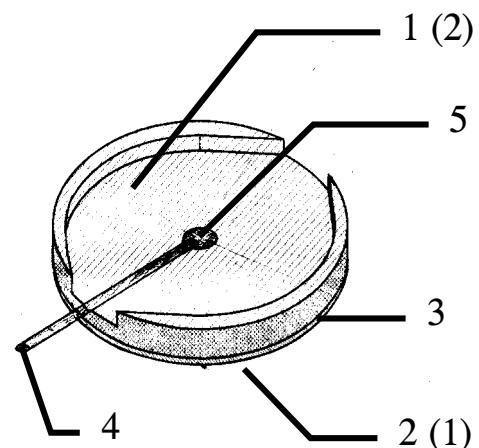


Fig. 8. SFC design  
 1 - anode, 2 - cathode, 3 - BCN-electrolyte,  
 4 - platinum current collector,  
 5 - current conducting glasses

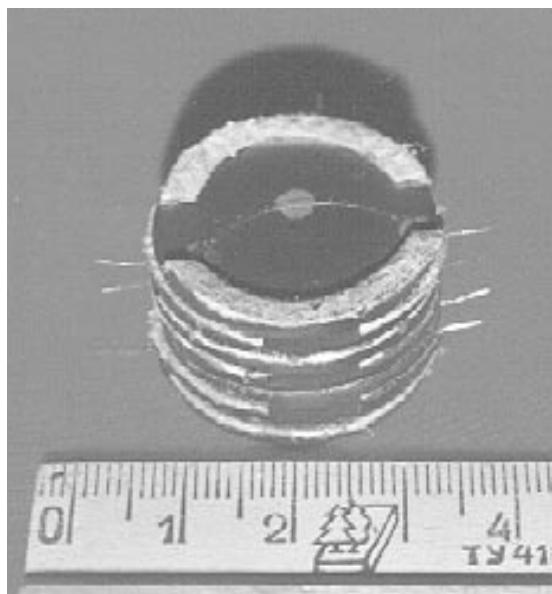


Fig. 9. Fuel cell stack

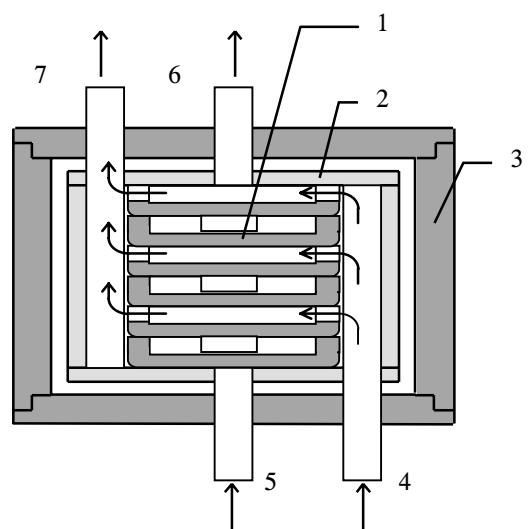


Fig. 10. Experimental stack design:  
 1 - SOFC stack, 2 - ceramic frame,  
 3 - heater frame, 4 - air,  
 5 - hydrogen, 6 - anode gas outlet,  
 7 - cathode gas outlet

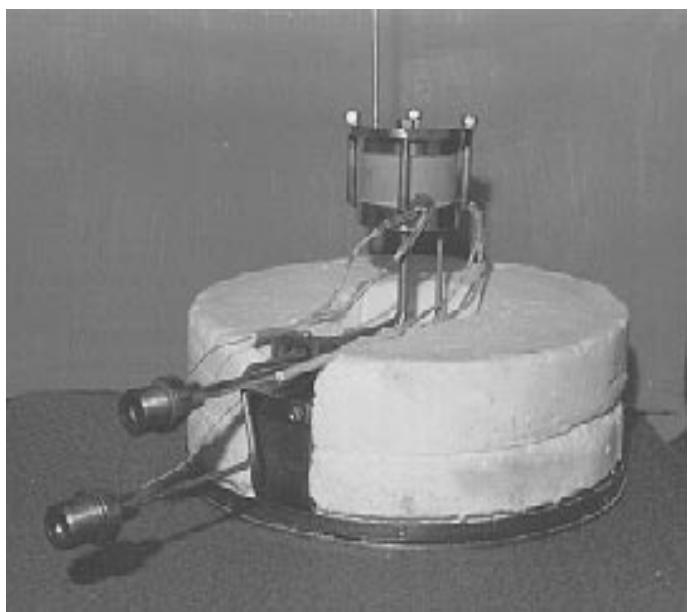


Fig. 11. Casing assembled with SOFC stack



Fig. 12. Experimental stack

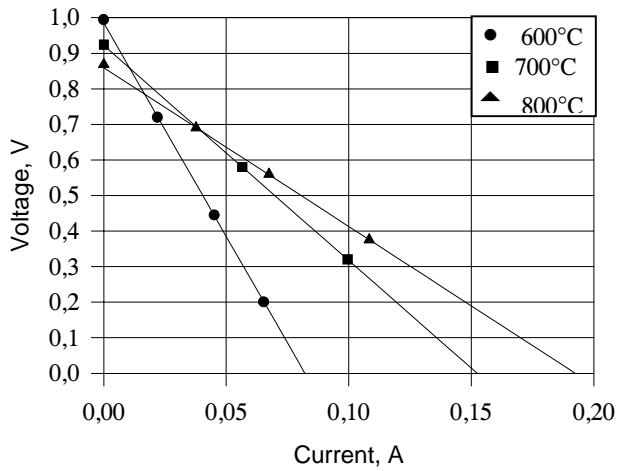


Fig. 13. SFC voltage-current characteristics

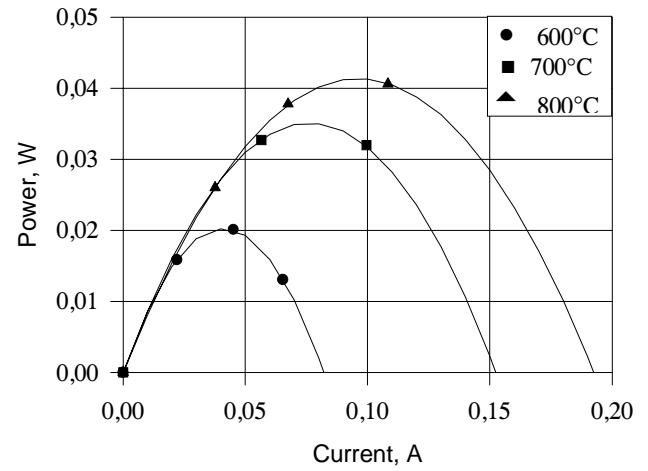


Fig. 14. SFC power vs. current

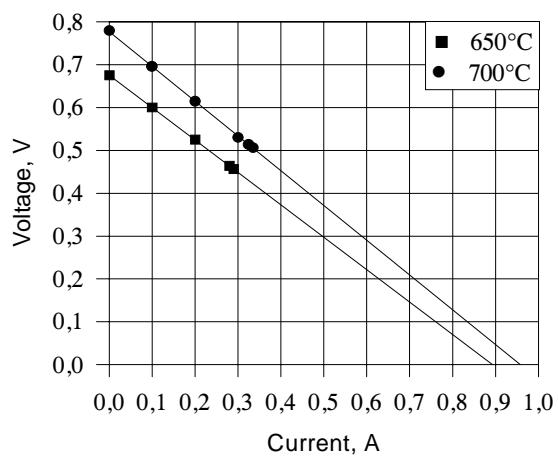


Fig. 15. SFC stack voltage vs. current

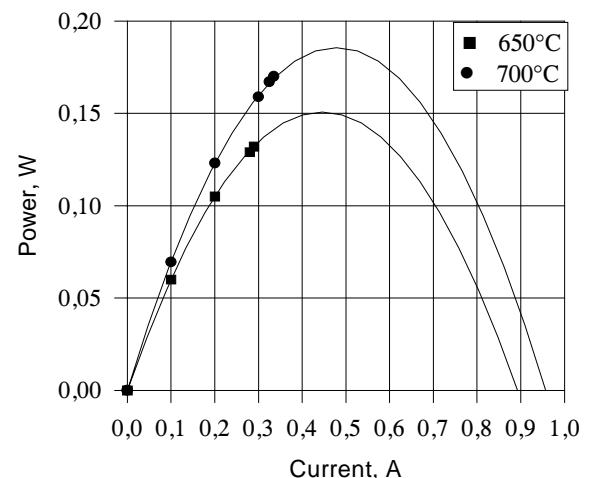


Fig. 15. SFC stack voltage vs. current

# Results of FETC Pre-Workshop Survey

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This survey was prepared to focus discussion at the Fuel Cell Advanced Research and Technology Development (AR&TD) Workshop, to be held on August 28, 1997, in conjunction with the 1997 Fuel Cell Program Review Meeting, August 26-28, 1997, at the Federal Energy Technology Center (FETC) facilities in Morgantown, West Virginia.

The survey included separate sections for molten carbonate and solid oxide fuel cells and was sent to 40 representatives of the fuel-cell community as selected by the project managers of fuel cell cooperative agreements at FETC.

Twenty-one responses were received: 17 responses to the molten carbonate section and 15 responses to the solid oxide section (many individuals responded to both sections). The number associated with each research topic below indicates the average priority assigned the research topic, according to the following scale:

**Low Priority**  $\leftarrow$   $\rightarrow$  **High Priority**

Each topic area in the survey also provided the opportunity for respondents to suggest other research topics and to offer comments. All the respondent-suggested research topics and comments are listed below in the appropriate topic area. The responses were anonymous, but respondents indicated their relationship to the fuel-cell program. The tally of these relationships may be found at the end of each section.

## A. Molten Carbonate Fuel Cells (MCFCs)

## 1.0 Electrodes (Anode and Cathode)

<b>1.0 Electrodes (Anode and Cathode)</b>		<b>Average Priority</b>
1.1	Develop new electrode materials . . . . .	2.88
1.2	Reduce contact resistance . . . . .	4.18
1.3	Increase power density . . . . .	4.19
1.4	Improve endurance . . . . .	4.25
1.5	Understand physical properties and reaction mechanisms . . . . .	2.25

1.6 Additional suggested topics related to electrodes:

- Improve cathode strength (3 responses)
- Mechanical strength
- Reduce anode creep
- Creep-resistant cathode
- No new electrode materials are necessary. We need to understand how their mean pore sizes affect the carbonate inventory, etc.
- Internal reforming kinetics and relationship to electrolyte contact
- 1.1, 1.2, and 1.5 connected with improved S tolerance; low cost important!

<b>2.0 Electrolyte</b>	<b>Average Priority</b>
2.1 Develop alternative electrolytes .....	3.73
2.2 Improve understanding of migration phenomena .....	4.06
2.3 Improve reactivity .....	3.00
2.4 Lower/higher operating temperatures .....	3.47
2.5 Understand physical properties and reaction mechanisms .....	2.93
2.6 Additional suggested topics related to electrolytes:	
- Understand wetting of oxide surface by carbonate and means to minimize (2 responses)	
- Wetting of electrolyte on various materials (wetting angle)	
- How wetting and contact angles change with time and gas composition/gas conversions	
- Improve understanding of electrolyte role in high voltage dielectric breakdown	
- Lower ohmic losses	
- Understand and reduce migration	
- Alternate support materials	

**Note:** a number of responses indicated that 2.2 above should include segregation in addition to migration.

<b>3.0 Separator Plate/Wet Seal</b>	<b>Average Priority</b>
3.1 Develop improved aluminization methods to reduce cost and/or improve endurance .....	3.19
3.2 Develop alternatives to aluminized coating .....	3.44
3.3 Develop new separator plate materials .....	3.31
3.4 Develop new high-temperature wet-proofing materials .....	3.38
3.5 Additional suggested topics related to the separator plate or wet seal:	
- Reduce cost of nickel-clad SS	
- Develop a low-cost nickel-clad bipolar plate/anode current collector	

<b>4.0 Other</b>		<b>Average Priority</b>
4.1	Develop new processes to reduce manufacturing cost . . . . . What is the most important fuel cell system component for cost reduction? - Balance of plant (2 responses) - Inverter - System piping - Separator/Bipolar plate (6 responses) - Current collector (2 responses; one added “takes up space/produces no power”) - Reducing number of SS layers and the contact IR	4.06
4.2	Develop standards for accelerated testing . . . . .	3.12
4.3	Develop procedures for non-destructive testing . . . . .	2.88
4.4	Additional suggested topics for MCFC Advanced Research: - Higher cross-pressure tolerance in electrolyte, wet seals, and manifold - Develop means to minimize electrolyte migration and loss from MCFC - Understand mechanism of phase transformation and particle growth of matrix - Approach to fuel processing: external vs. internal; ability to use multiple fuels - Protection of anode current collector with nickel - Distributed manifold and other advanced stack configurations - Impacts of cycling temperatures	

**Comments:**

- Once stack durability and reliability are ensured (doing this should be a high priority) then reducing the decay rate by reducing SS layers becomes important economically. This will require component design and perhaps material changes.
- Stack life and cost are the critical issues!

**The primary relationships of the MCFC respondents to the DOE Fuel Cell Program are:**

8	Fuel Cell Developers
3	AR&TD Researchers
1	Fuel cell component supplier
1	Other sponsor of fuel cell research
4	Other: One retired developer/funder and three consultants for sponsors or developers

## **B. Solid Oxide Fuel Cells (SOFCs)**

### **1.0 Cathode (Air Electrode)**

	<b>Average Priority</b>
1.1 Cathode-electrolyte interface characteristics .....	4.14
1.2 Mechanical and electrochemical properties of cathode .....	3.00
1.3 Alternative cathode materials .....	2.86
1.4 Lower cost cathode fabrication .....	3.73
1.5 Additional suggested topics related to the cathode:	
- New electrodes for low-temperature electrolytes	

### **2.0 Anode (Fuel Electrode)**

	<b>Average Priority</b>
2.1 Sulfur tolerance of anode .....	3.38
2.2 Mechanical and electrochemical properties of anode .....	3.15
2.3 Alternative anode materials .....	2.85
2.4 Lower cost anode fabrication .....	3.64
2.5 Additional suggested topics related to the anode:	
- New low-temperature anodes; interfacial phenomena for low temperature cells	
- Anode/electrolyte interface	

### **3.0 Electrolyte**

	<b>Average Priority</b>
3.1 Mechanical and electrochemical stability of electrolyte .....	2.85
3.2 Alternative electrolyte materials .....	3.29
3.3 Lower cost/alternative electrolyte fabrication methods .....	4.07
3.4 Additional suggested topics related to the electrolyte:	
- Low temperature electrolytes (2 responses)	

### **4.0 Interconnects and Seals**

	<b>Average Priority</b>
4.1 Mechanical and electrochemical stability of interconnect .....	4.21
4.2 Ceramic and/or ceramic metal seals .....	4.60
4.3 Additional suggested topics related to interconnects and seals:	
- Metallic interconnects	
- If planar, approach to separator plates	

		Average Priority
5.1	System scale-up . . . . .	4.33
5.2	Develop materials to operate in an alternate temperature range . . . . .	4.14
	What temperature range?	
-	500 to 600 °C	
-	500 to 750 °C	
-	500 to 800 °C (2 responses)	
-	600 to 700 °C	
-	700 °C	
-	700 to 800 °C	
-	less than 800 °C	
-	about 800 °C	
-	planar: 800 °C; 1,000 °C for tubular	
	What is the most critical component that requires development for operation at these temperatures?	
-	Anode	
-	Cathode (3 responses)	
-	Electrodes (2 responses)	
-	Electrolyte (2 responses)	
-	Interconnect (3 responses)	
-	Cathode/Electrolyte Interface	
5.3	Develop standards for accelerated testing . . . . .	3.08
5.4	Develop procedures for non-destructive testing . . . . .	3.31
5.5	Develop operational controls for load following . . . . .	2.62
5.6	Evaluate the effects of pressurization . . . . .	3.62
5.7	Investigate internal fuel reforming (natural gas and other fuels) . . . . .	3.36
5.8	Develop lower cost cell fabrication and assembly methods . . . . .	4.50
5.9	Additional suggested topics for SOFC Advanced Research:	
-	SOFC Applications	

#### Comments:

- The U.S. needs a second generation SOFC Program. The Westinghouse technology is being commercialized.
- The question on SOFC depends critically on configuration; i.e., tubular vs. planar.

#### The primary relationships of the SOFC respondents to the DOE Fuel Cell Program are:

- 6 Fuel Cell Developers
- 3 AR&TD Researchers
- 2 Other sponsors of fuel cell research
- 4 Other: all four provide support or consulting services for sponsors or developers

# **EPRI Assessment of Fuel Cell R&D Needs**

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## **PAFC Needs**

- Higher stack power density to reduce cost.
- Added electrolyte inventory or reduced electrolyte loss to ensure long cell stack life.
- Continued reduction in separator plate costs.
- Advanced water treatment system.
- Improved cooling system reliability.

## **MCFC Needs**

- Alternative separator plate material to eliminate corrosion and reduce cost.
- Eliminate electrolyte migration in external manifold MCFC design.
- Creep resistant electrodes.
- Alternative electrolytes may reduce corrosion.
- Eliminate or simplify aluminization.
- Eliminate or simplify nickel coating.

## **SOFC Needs**

- Establish a major DOE-supported planar SOFC program.
- Develop techniques to scale-up SOFC cells to larger sizes to reduce costs.
- Sulfur tolerant anodes.
- Improved conductivity metal interconnectors.
- Improved seals for metal interconnectors.
- Improved seals for external manifolds.
- System development.
- Improved, reduced temperature electrolytes.

## **PEM Fuel Cells**

- Lower cost membrane.
- Higher temperature membrane (250 - 300 °F).
- Improve membrane tolerance to diffusion of methanol.
- CO tolerant anode catalyst.
- Improved membrane water tolerance to dry out.
- Improved fuel processing systems.
- Inexpensive separator plates.

# **GRI Basic Solid Oxide Fuel Cell Research**

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**Objective:** Total system cost , \$600/kW

## **Phase I (1990-1996)**

- Planar single cells, operating at <800 °C

## **Phase II (1997-2000)**

- Reliable, high performance planar stacks, operating at <800 °C
- Tubular SOFC Improvements

## **Budget** (Thousands of dollars)

1990	\$300
1991	\$300
1992	\$300
1993	\$650
1994	\$650
1995	\$650
1996	\$600
1997	\$1,000
1998	\$1,000
1999	\$1,000
2000	\$1,000

## **Summary**

### **Reduced-Temperature, Planar SOFCs**

- Technically Challenging, Longer-Term Option
- Potential for <\$700/kW Capital Cost
- High Power Density, Small Size
- Good Manufacturability

### **Tubular SOFCs**

- Ready for Commercialization in about 3 Years
- Seal-Less Design, Tolerance to Thermal Stress
- Recent Technical Progress
- Operability in High-Efficiency, Pressurized SOFC/Turbine Cycles

# **The DOE Fuel-Cell AR&TD Program**

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## **Role of DOE Fuel-Cell AR&TD**

- To Support Major Fuel Cell Developers
  - New Materials
  - New Manufacturing Processes
- To Identify Breakthroughs That Lead to New Technologies
- To Verify R&D Claims
- To Maintain a Broad U.S. Technology Support Base
  - Equipment
  - Personnel

## **AR&TD Issues**

- Declining R&D Funding by Government and Industry
- Shrinkage of Number of Universities Conducting Research in Some Fuel Cell Areas in Some Countries
- Lack of Forums for Communicating Results to Industry
- Short Time Horizon for Research Results
- Lack of Fuel-Cell Research Associations in Some Countries

## **Some AR&TD Funding Mechanisms**

- Direct Contract to a Funding Agency
- University Coal Research
- Subcontract to DOE Developer
- SBIRs
- Consortia

## **Fuel Cell R&D Areas for Consortia, Pre-Commercial R&D**

- Advanced New Manufacturing Processes That Reduce Environmental and Cost Problems
- Advanced Materials That Promise Better Properties and Performance

## **Solid Oxide Fuel Cells: Developmental Issues**

- Lower Cost Materials and Manufacturing Processes
- Sealing in Planar Designs
- Gas Pre-Heating
- Material Corrosion: Changes in Composition, Porosity, Density, Phase, etc., Over Time
- More Compatible Materials for Interconnect, etc.
- Lower Temperature Materials
- Thinner Components
- Thermal Cycling

## Carbonate Fuel Cells: Technical Issues

- Life and Endurance Testing
- Thermal Cycling
- Lower Cost Aluminization
- Higher Power Density
- Improved Stability: Corrosion and Electrolyte Loss
- Develop Environmentally Friendly, Low-Cost Compositions/Fabrication Processes

### FY 1997 Agency Funding (\$ millions)

	<b>PEM</b>	<b>SOFC</b>	<b>MCFC</b>	<b>PAFC</b>
<b>DOE, FE</b>	--	13.0	37.0	--
<b>DOE, EE</b>	21.0	--	--	--
<b>DOD, ES</b>	--	--	--	8.0
<b>DARPA</b>	3.0	1.0	2.0	--
<b>GRI</b>	--	1.0	--	--
<b>EPRI</b>	--	1.0	--	--
<b>DOT</b>	2.5	--	--	2.5
<b>Total</b>	<b>26.5</b>	<b>16.0</b>	<b>39.0</b>	<b>10.5</b>

# **Fuel Cell Opportunities in the Division of Materials Sciences, Office of Basic Energy Sciences, U.S. Department of Energy**

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## **Outline**

- I. Budget
- II. Areas of Research
  - Fuel Cells
- III. Opportunities
  - Where to get more information

## **Office of Basic Energy Sciences**

### **Examples of Areas of Research that Underpin the Mission**

- ★ Analytical chemistry
- ★ Atomic, Molecular, and Optical Physics
- ★ Batteries and Fuel Cells
- ★ Bioenergetics
- ★ Biomaterials and Biocatalysis
- ★ Catalysis
- ★ Chemical Kinetics
- ★ Chemical Physics
- ★ Ceramics
- ★ Combustion Dynamics
- ★ Condensed Matter Physics
- ★ Corrosion
- ★ Earth Structure
- ★ Electrochemistry
- ★ Fermentation Microbiology
- ★ Geophysics and Geochemistry
- ★ Heavy Element Chemistry
- ★ Instrumentation Development
- ★ Intermetallic Alloys
- ★ Magnetism and Magnetic Materials
- ★ Materials Physics and Chemistry
- ★ Metals Forming
- ★ Materials Welding and Joining
- ★ Metallurgy
- ★ Mineral Reactions
- ★ Organometallic Chemistry
- ★ Photochemistry
- ★ Photosynthetic Mechanisms
- ★ Plant and Microbial Sciences
- ★ Plant Genomics
- ★ Polymer Science
- ★ Process Engineering
- ★ Radiation Effects
- ★ Robotics and Control Systems
- ★ Rock Deformation
- ★ Rock-Fluid Dynamics
- ★ Separations Science
- ★ Solar Energy Conversion
- ★ Solid Dynamics
- ★ Solid State Physics and Chemistry
- ★ Structural Characterization
- ★ Superconductivity
- ★ Surface Science
- ★ Synthesis and Processing Science
- ★ Thermophysical Properties

## **Office of Basic Energy Sciences**

### **Examples of PIs Currently Supported by BES in Fundamental Research Areas That Underpin Electrochemical Technologies**

Bates (ORNL)

Angell (ASM)

Licht (Clark)

MacDonald (Penn State)

Ocko (BNL)

Ross (BNL)

McBreen (BNL)

Smyrl (Minn)

Maroni, Nagy, Melendres (ANL)

Tuller (MIT)

Fultz (Cal Tech)

Halley (Minn)

## **Opportunities**

More Information

www

Search

DOE Energy Research

Click

BES

- Abstracts of all National Lab, University, Industry Projects
- Request for Proposals

PAIR

- Partnership for Academic-Industry Research
- Preproposal Deadline October 1, 1997

E-mail: Richard.Kelley@oer.doe.gov



# Office of Basic Energy Sciences

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Deputy Associate Director  
Iran L. Thomas

*Robert Astheimer*  
Research Coordinator

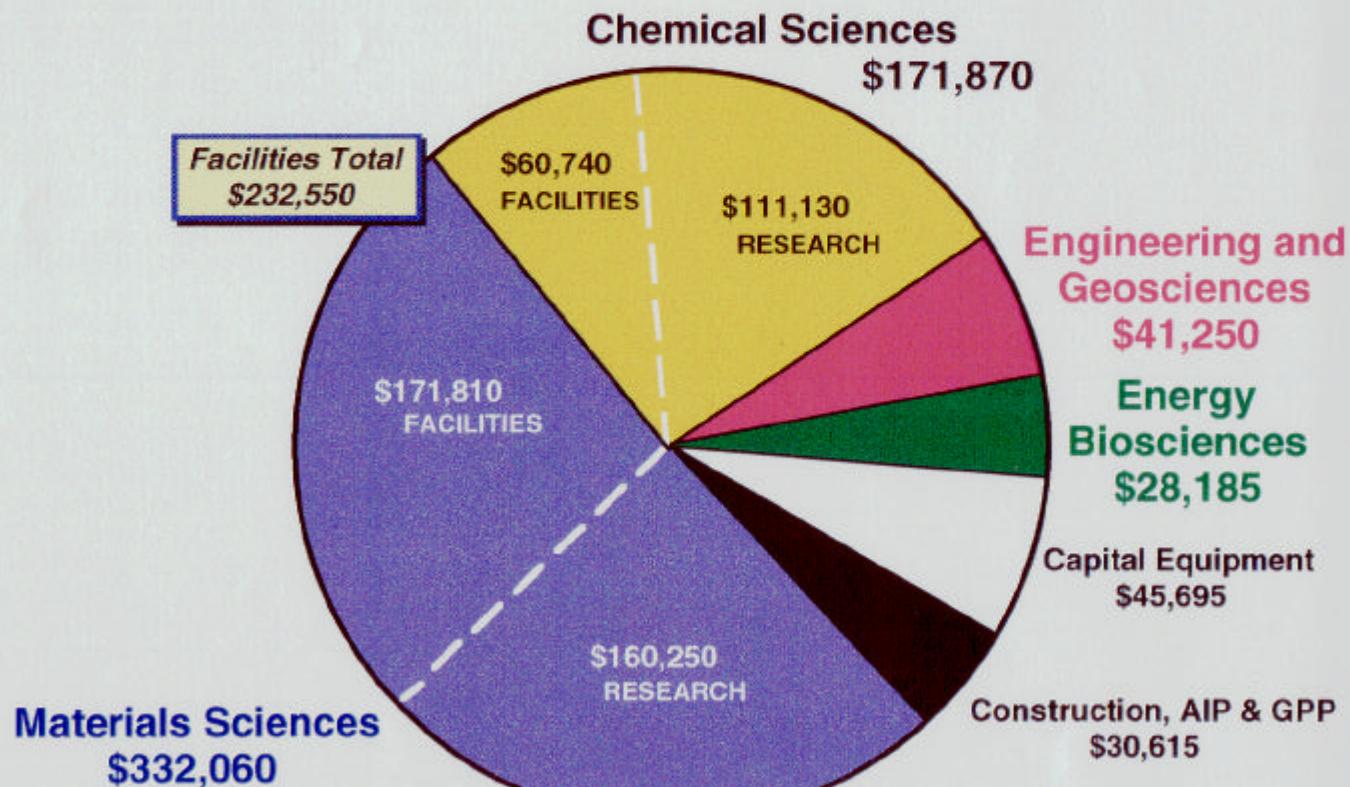
Materials Sciences  
Iran L. Thomas  
Director

Chemical Sciences  
Robert Marianelli  
Director

Engineering &  
Geosciences  
Iran L. Thomas  
Acting Director

Energy Biosciences  
Gregory Dilworth  
Director

FY 1997  
Budget Appropriation  
\$649,675





# Office of Basic Energy Sciences

## Scientific User Facilities

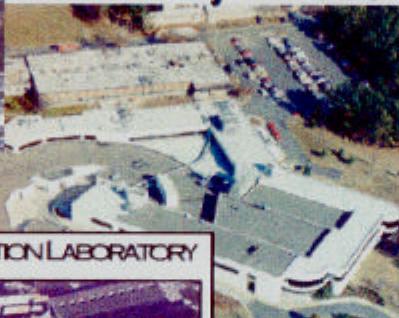
Advanced

### Light Sources



Light Source

National Synchrotron



Light Source

STANFORD SYNCHROTRON RADIATION LABORATORY



Advanced



Photon Source

And...

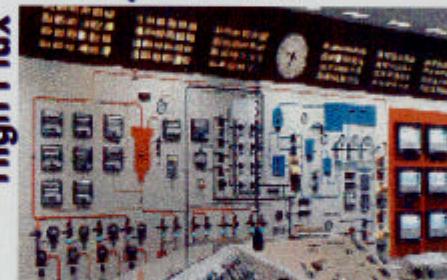
National Center for Electron Microscopy



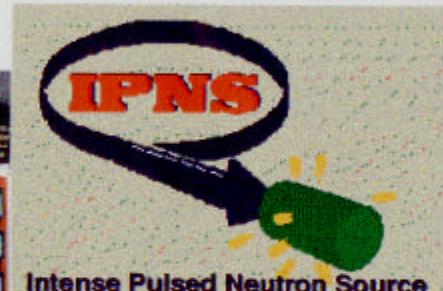
MATERIALS  
PREPARATION  
CENTER

### Neutron Sources

Isotope Reactor



High Flux



Intense Pulsed Neutron Source



Beam Reactor

**LANSCE** High Flux

Combustion  
Research  
Facility



# DARPA Advanced Energy Technologies

Robert Nowak

(Phone: 703-696-7491; Fax: 703-696-3999; E-mail: rnowak@darpa.mil)

DARPA/DSO

3704 North Fairfax Drive

Arlington, VA 22203-1714

## Portable Power — Opportunities

- ★ **Batteries:** significant improvements over existing systems;  
>> 2X Specific Energy/Power

- ★ **Fuel Cells, Direct Methanol**

Methanol Crossover

Cathode Catalyst Activity and Methanol Tolerance

Anode Catalyst Activity

Membrane Electrode Assembly Processing

- ★ **Fuel Cells, Alternative Fuel Options and Concepts**

- ★ **TPV and AMTEC**

Low Cost and Efficient PV Cells

Efficient TPV Cavity Designs

Compact AMTEC Designs

Efficient Fuel Combustion at Low Flow Rates

High Temperature Heat Recuperation

## **Mobile Electric Power — Opportunities**

### **★ Fuel Reforming**

- Size
- Efficiency
- Sulfur Removal and/or Tolerance
- Hydrogen Purity
- Fuel Cell Integration

### **★ Customize for the Military**

- System Size and Weight
- System Efficiency
- Environmental Issues:
  - shock, vibration, temperature, altitude, salt spray, dust, etc.
- Signature:
  - acoustic, thermal, etc.
- Operation and Maintenance



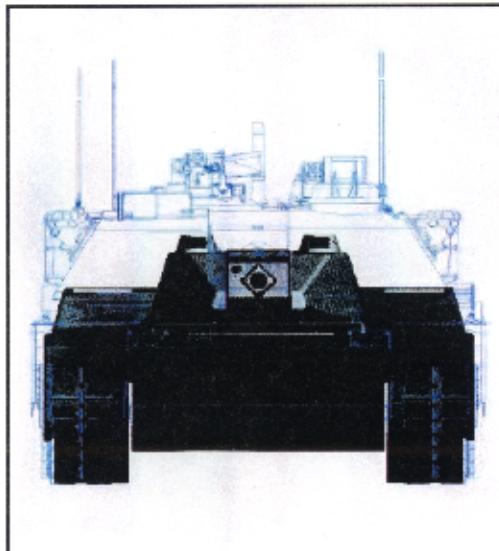
# Advanced Energy Technologies



Defense Sciences Office

## Power for the Military

**Mobile Electric Power**  
**2 - 100 kW**

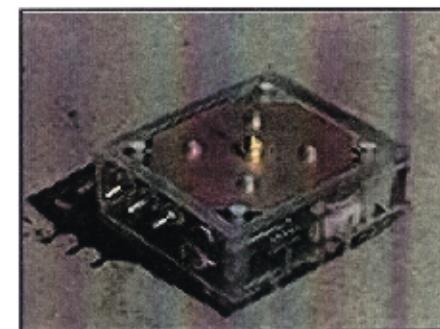


**Portable Power**  
**50 - 500 W**



**Energy Harvesting**  
**< 5 W**

**Micro - Internetted Unattended  
Ground Sensor**



↔

~ 1 "

- Silent Watch
- Field Power Stations

- Battery Replacement
- Micro-Climate Cooling
- Battery Charging

- Ground Sensors
- Micro - Robots



# Mobile Electric Power 2 - 100 kW Fuel Reformer Demonstrations

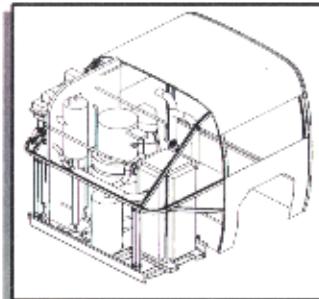


Defense Sciences Office

## 100 kW PAFC

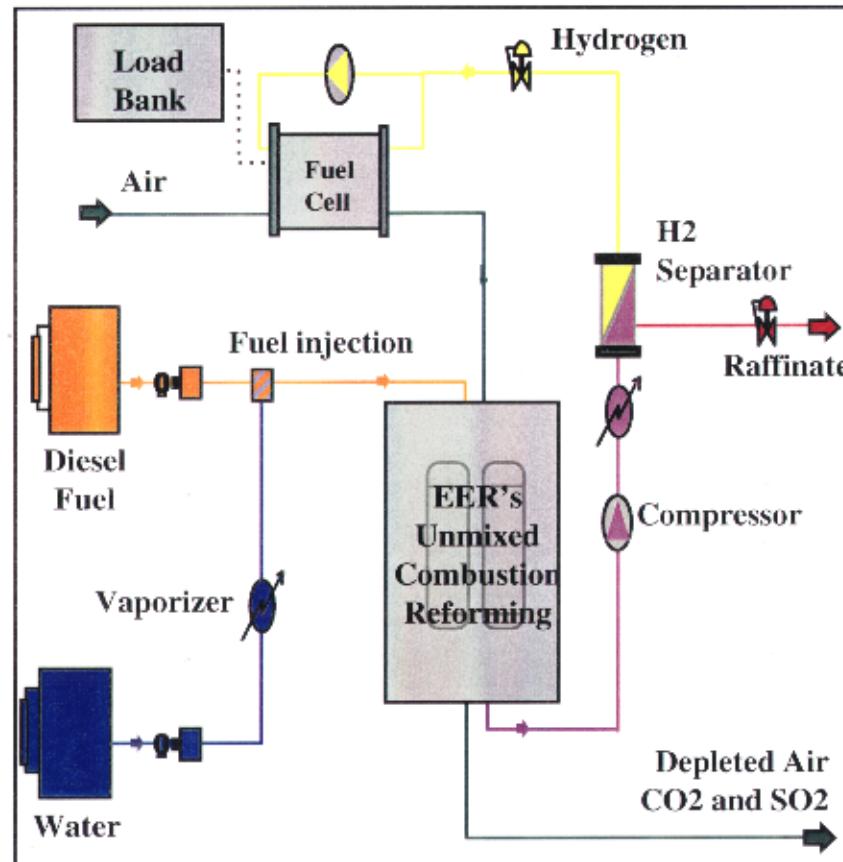


- 14 kW Tested
- 100 kW Fabrication



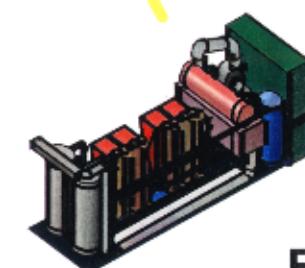
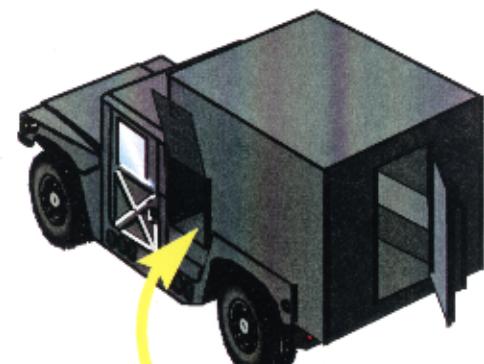
- Georgetown U. Bus

## 20 kW PEMFC



## 10 kW SOFC

### Multipurpose Shelter



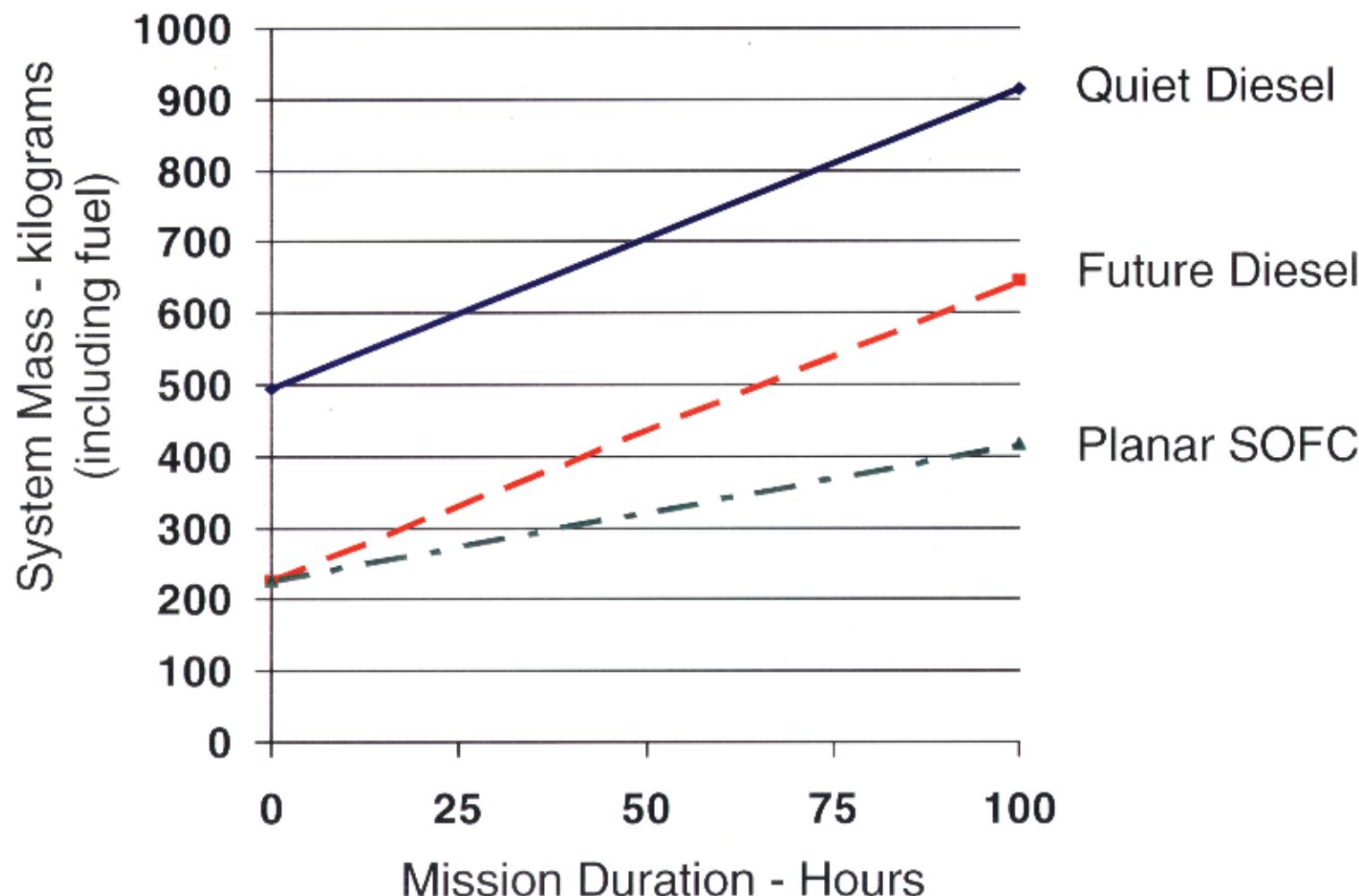
Fuel Cell



## System Mass vs. Mission Duration Mobile Electric Power - 10 kW (Logistics Fuel)



Defense Sciences Office

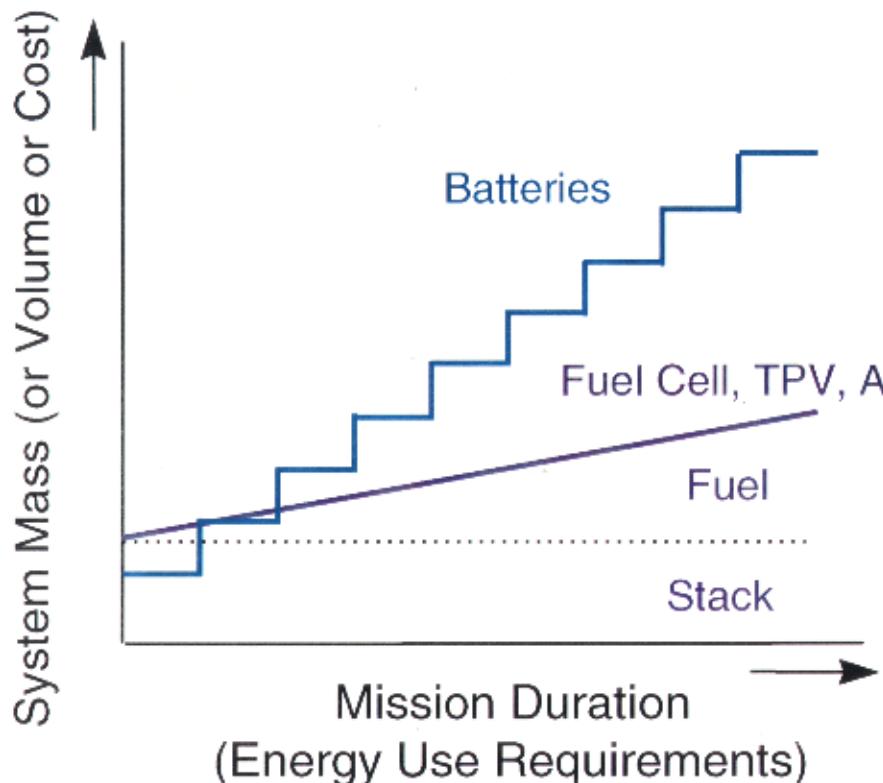




# Energy Conversion vs. Energy Storage



Defense Sciences Office



## Energy Density of Selected Fuels and Batteries

Fuel:	<u>Energy Density</u>
Diesel Fuel/Jet Fuel	12,000 Wh/kg
Methanol	5,000
High Explosive	1,000

Battery:	
Primary Battery (est. max.)	500
Rechargeable (est. max.)	200
Li/SO <sub>2</sub> Battery (primary)	176
Alkaline Battery (primary)	80
Nickel-Cadmium (secondary)	40

**Driving Force:** Substantially decreased size, weight, and cost with improved safety and environmental compliance → Increased force mobility



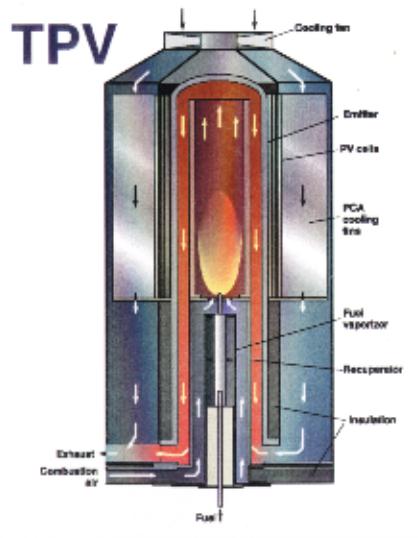
# Portable Power

## 50 - 500 W

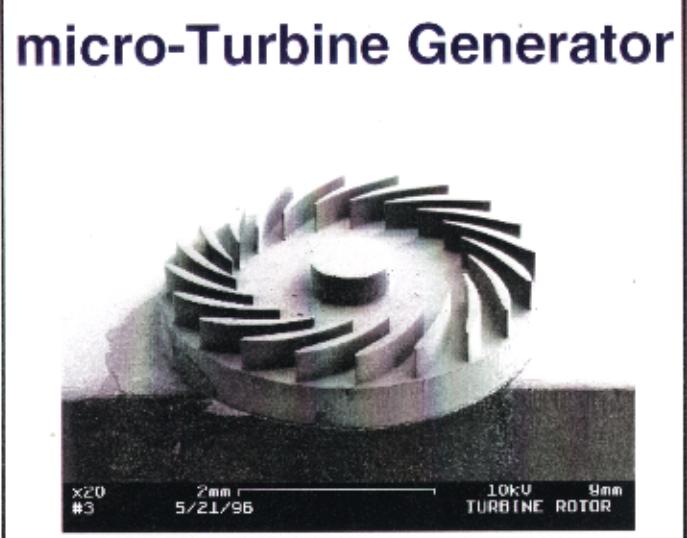


Defense Sciences Office

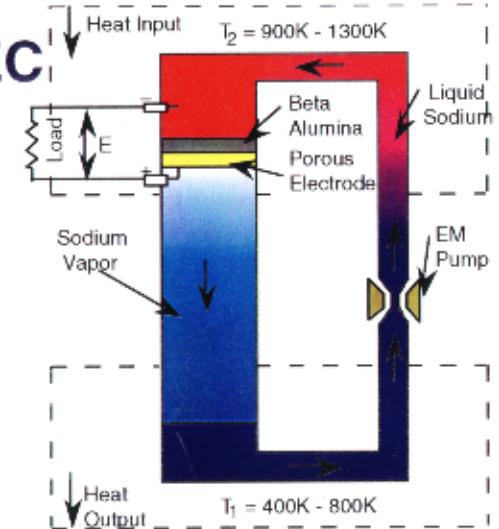
TPV



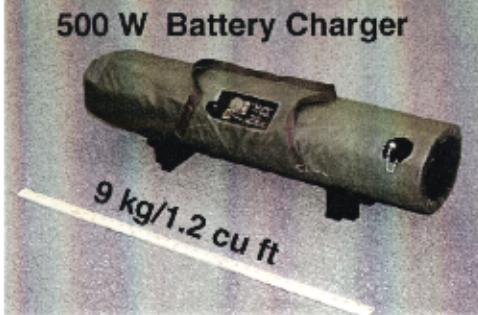
micro-Turbine Generator



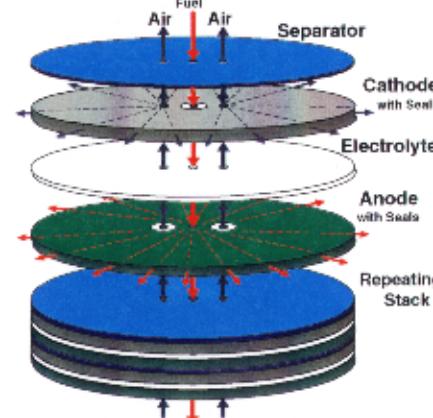
AMTEC



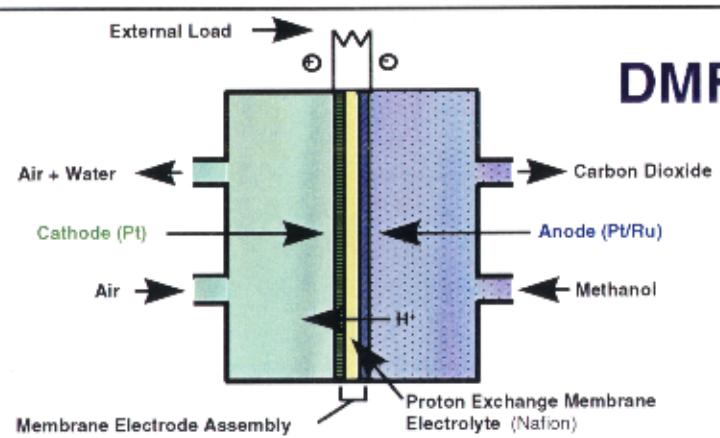
500 W Battery Charger



Planar SOFC



External Load



DMFC

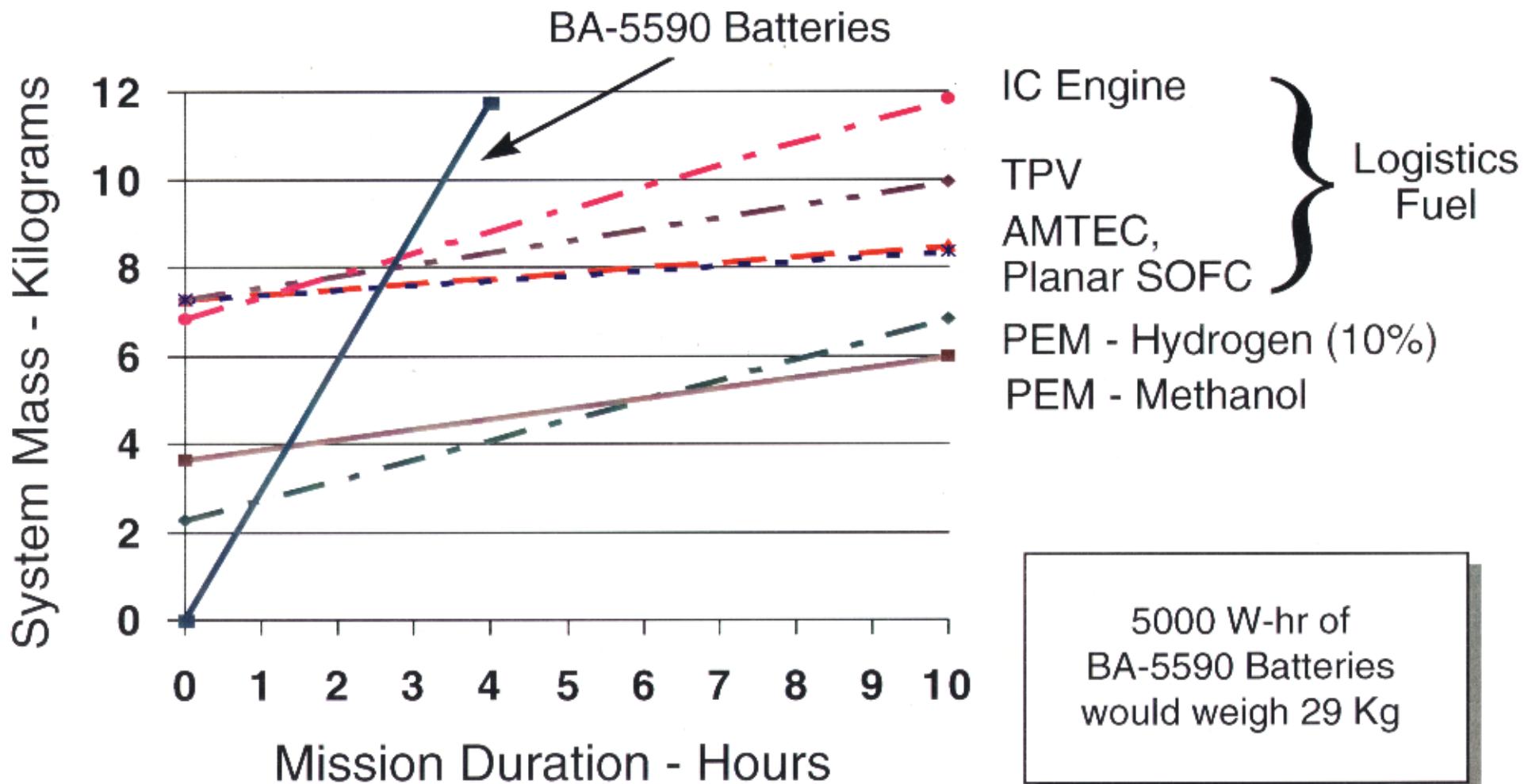


# System Mass vs. Mission Duration

## Portable Power - 500 W



Defense Sciences Office

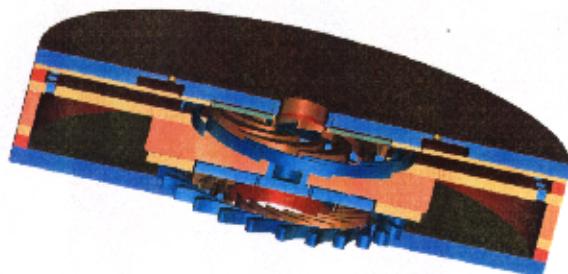




# MIT Micro Turbine Generator Performance Comparison



Defense Sciences Office



## $\mu$ Turbogen\*

## BA-5590 (LiSO<sub>2</sub>) Battery

Power Output  
Energy Content  
Weight  
Volume

50 W  
175 W hr  
50 g  
50 cc

50 W  
175 W hr  
1100 g  
880 cc

Specific Energy  
Energy Density

3500 W hr/kg  
3 W hr/cc

175 W hr/kg  
0.2 W hr/cc

\* Effort lead by MIT; all values include fuel





# The Portable Power Burden for 10 kW-hr of Electrical Energy



Defense Sciences Office

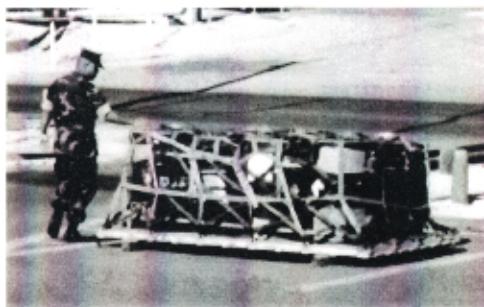
1996  
60 Kg



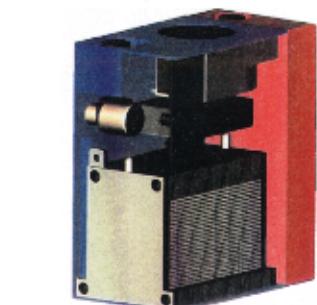
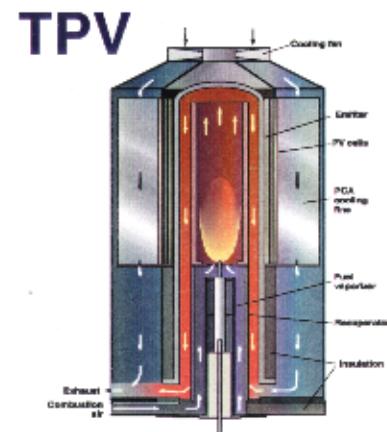
2002  
15 Kg



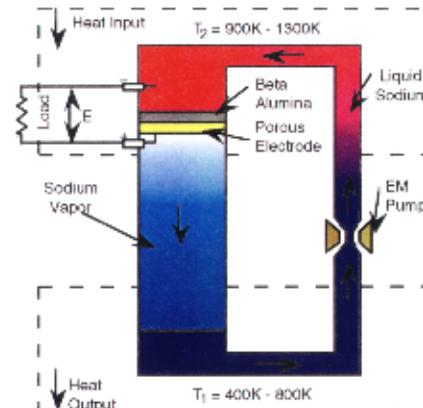
2010  
4 Kg



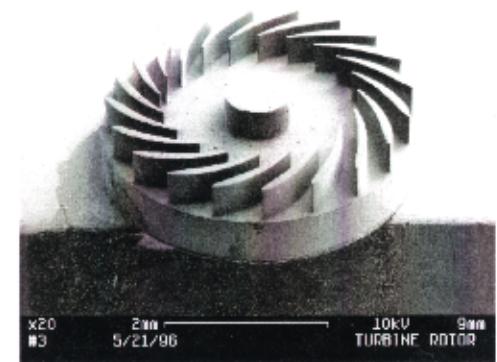
Batteries



DMFC  
(BA-5590 Equivalent)



AMTEC



Microturbines

# **U.S. Department of Agriculture Small Business Innovation Research Program**

Charles F. Cleland and Ruth Lange

(Phone: 202-401-4002; Fax: 202-401-6070; E-mail: [ccleland@reeusda.gov](mailto:ccleland@reeusda.gov))

Room 328, Aerospace Center

901 D Street, S.W., Stop 2243

Washington, DC 20250-2243

## **Features of USDA SBIR Program**

- Award Grants only; awards based on scientific/technical merit; ideas investigator-initiated.
- Nine broad topic areas.
- Funds allocated to topic areas in proportion to number of proposals received.
- Phase I Grants: 6 months at \$65,000. Phase II Grants: 2 years at \$250,000.
- Proposals reviewed by confidential peer review using outside experts from non-profit organizations.
- All applicants receive verbatim copies of reviews.
- Follow-on funding commitment strongly encouraged.

## **Topic Areas**

Forests and Related Resources  
Plant Production and Protection  
Animal Production and Protection  
    Air, Water, and Soils  
Food Science and Nutrition  
Rural and Community Development  
    Aquaculture  
Industrial Applications  
Marketing and Trade

## **Rural and Community Development**

1. New Agricultural Enterprises
2. Transportation
3. Education
4. Health Care
5. Information Services
6. Telecommunications

## **Marketing and Trade**

1. Development of Marketing Systems
2. Development of Innovative Real-Time/Near-Time Information Systems
3. Assessments and Specification of Marketing Opportunities

### **Example of Winning Proposal**

Ms. Marcia Smith, Columbia Cascade, Inc., Reston, VA  
International Trade Assistance for Rural Areas Using Expert System Technology

## History of USDA SBIR Funding

<b>FY</b>	<b>Budget (x 10<sup>6</sup>)</b>	<b>Phase I</b>	<b>Phase II</b>
90	4.11	32/314	13/17
91	4.89	36/296	16/22
92	5.63	44/346	19/30
93	7.02	53/380	23/35
94	7.17	60/443	22/36
95	9.29	72/445	27/41
96	9.10	63/428	33/60
97	11.40	72/401	29/47

## Geographical Distribution of USDA SBIR Winners, FY 83-FY 97

<b>California</b>	<b>West</b>	<b>Northeast</b>	<b>North Central</b>	<b>South</b>
CA-117	WA-43	MA-52	MN-31	FL-23
	OR-40	PA-38	MI-31	TX-23
	CO-29	NY-31	OH-20	VA-21
	AZ-20	NJ-19	WI-16	NC-19
	ID-19	MD-15	IL-15	TN-9
	HI-18	CT-14	ND-13	LA-9
	MT-12	DC-6	NE-11	GA-8
	UT-10	VT-6	SD-10	OK-6
	NM-10	NH-4	IA-9	MS-5
	AK-6	WV-1	IN-9	AR-3
	WY-4	RI-0	KS-9	SC-3
	NV-0	ME-0	MO-8	AL-2
		DE-0		KY-1
				<u>VI-1</u>
117 (14.1%)	211 (25.5%)	186 (22.4%)	182 (22.0%)	133 (16.0%)

## **Solicitation/Proposal Schedule, FY 1998**

1.     Solicitation released on 6/1/97
2.     Proposal due date of 9/4/97
3.     Panels meet in January and February of 1998
4.     Decisions made by 3/1/98
5.     Phase I Grant Period is from 5/15/98 to 11/30/98
6.     Phase II Application Deadline is 2/12/98

## **Internet Sites of Interest**

**SBA**           <http://www.sbaonline.sba.gov>

**USDA**           <http://www.usda.gov>

**USDA/SBIR** <http://www.reeusda.gov/sbir/sbir.htm>

**CRIS**           <http://cristel.nal.usda.gov:8080>

## **Commercialization Results**

<b>Year</b>	<b>No. Awards</b>	<b>Positive Impact</b>	<b>Percent</b>	<b>Sales</b>	<b>Percent</b>
1988	14	10	71	8	57
1999	13	9	69	5	38
1990	13	9	69	6	46
1991	16	12	75	10	63
1992	19	14	74	12	63
1993	23	21	91	13	57
1994	22	20	91	13	59
<b>Total</b>	120	95	79	67	56

### **Distribution of Sales for Phase II Awardees, FY 1988 - 1994**

<b>Range</b>	<b>Number</b>
< \$10,000	3
\$10,000-99,999	7
\$100,000-499,999	14
\$500,000-999,999	4
> \$1 million	7

### **University Involvement in USDA SBIR**

1. Strongly encouraged.
2. University faculty can serve as consultants or can receive a subcontract (in both cases, limited to no more than 1/3 of Phase I award or 1/2 of Phase II award) and continue to work full-time at university.
3. University faculty can serve as principal investigator on the grant, if they reduce employment at the university to 49 percent for duration of grant, and if the SBIR research is performed some place other than their research lab.
4. It is usually not acceptable for university faculty to serve as consultants and have all the research done in their lab.

## **USDA SBIR Review Process for Phase I**

1. There is a different review panel for each topic area.
2. An outstanding research scientist is selected as topic manager for each review panel.
3. Proposals undergo initial screening and then are assigned to the appropriate topic area.
4. Each proposal is sent to six ad-hoc reviewers who mail in written reviews.
5. Each proposal is reviewed by two members of the review panel.
6. Based on both the panel and ad-hoc reviews and the panel discussion, each proposal is ranked and the top ranked ones are recommended for award.
7. The SBIR program follows the panel recommendations very closely, and allocates funds to each topic area in proportion to the number of proposals submitted.
8. Those proposals recommended for funding undergo an administrative review prior to the grant being awarded.
9. A panel summary plus verbatim copies of the reviews, minus the score and name of the reviewer, are sent to the principal investigator for all proposals, funded or not.

## **USDA SBIR Review Process for Phase II**

1. Each proposal is sent to six to eight ad-hoc reviewers who are experts on some aspect of the proposal.
2. The ad-hoc reviews for all proposals in a given topic area are sent to the topic manager who provides a rank order for the proposals, based on his/her reading of the ad-hoc reviews and of the proposals.
3. The rankings from each topic manager are presented to an internal panel consisting of program managers from the National Research Institute. Based on their reading of the proposals, the ad-hoc reviews and justifications from the topic managers for their rankings, the panel establishes the final rank order for the proposals in each topic area.
4. The SBIR program uses these rankings to determine which proposals should be funded and at what dollar level. Other factors that are considered at this point include follow-on funding agreements and prior success in commercializing technologies developed SBIR support.

## **Evaluation Criteria**

1. Scientific/technical merit.
2. Degree to which Phase I objectives were met and feasibility demonstrated (Phase II only).
3. Importance of problem to American agriculture or rural development.
4. Probability of commercial success.
5. Adequacy of research objectives.
6. Adequacy of research plan.
7. Qualifications of Principal Investigator and other key personnel.
8. Adequacy of facilities.
9. Qualifications of consultants.
10. Letters from consultants indicating their willingness to work on project are included as part of the proposal.
11. Adequacy of bibliographies for the Principal Investigator, other key personnel, and consultants.

## **Elements Common to Successful Proposals**

1. Well written, succinct, and logical.
2. Thorough literature review.
3. Addresses important problem.
4. Innovative approach.
5. Well designed and detailed experimental plan.
6. If successful, would have good commercial potential.

## **Common Proposal Criticisms**

1. Poorly written and presented.
2. Principal Investigator lacks necessary technical expertise.
3. Insufficient literature review.
4. Insufficient technical information.
5. Cannot be completed in 6 months.
6. Inadequate bibliographical information.
7. Lacks letters from consultants.
8. Research already done by others.
9. Too vague and unfocused.
10. Failure to indicate where project would go in Phase II.
11. Poor commercialization potential.
12. Doubtful economic prospects.
13. Inadequate detail in experimental plan.
14. Too much research done at university.
15. Need to engage consultants to add expertise in area where Principal Investigator is deficient.

# **Identification of Breakout Topics, and Initiation of Breakout Group Discussions**

## **A. Session on MCFC-Stack AR&TD Issues**

**Facilitator:** *Julianne Klara (FETC)*

**Scribe:** *Diane Hooie (FETC)*

### **Summary**

Fourteen people were present in this session, representing manufacturers, research and development institutions, and consultants. All attendees, including the facilitator and scribe, were engineers. In addition, eight were foreign visitors.

The format of the session consisted of a brainstorming session and then the nominal group technique was used to prioritize the issues. This technique involves each participant voting for the his/her top three issues, and three points are assigned for the top issue, two points for the second, and one for the third. The votes are then tallied, with the most votes going to the issues that should have the highest priority.

***Cell conversion efficiency*** was identified as the most important issue. This involves the reduction of polarization losses as well as improved heat removal from the cells. The second priority identified was ***separator plate materials***. Corrosion needs to be reduced as well as improved active area and wet seals needs to be developed. The third key item identified was ***improvement of the electrolyte***. This includes reduction of several key mechanisms: creep, evaporation, migration, segregation, ionic degradation, and nickel oxide solubility.

The focus of this session were those advanced research and development issues related only to the molten carbonate stack. However, in the process of developing the key issue list, other issues were identified. At the request of the participants, these were also prioritized using the same technique. The top issues include improved robustness of the stack, commercial availability of components and materials, seal integrity, and thermal cycle ability.

### **Discussion**

Participants identified five key areas that need improvement for the fuel cell stack. They then identified specific stack issues that would improve those areas. These were further divided into advanced research and development needs or product development/improvement needs that would probably not be addressed through an AR&TD program. The stack issues were then ranked according to the highest priority. These are shown below.

<b>Advanced Research and Technology Development Issues</b>	
Code	
	<i>Durability</i>
'D1	<p>Separator plate materials</p> <ul style="list-style-type: none"> <li>- Reduce corrosion</li> <li>- Improve active area/wet seals</li> </ul>
'D2	<p>Electrolyte (match to operating conditions)</p> <ul style="list-style-type: none"> <li>- Reduce creep</li> <li>- Reduce evaporation</li> <li>- Reduce migration (definitely not a problem for internally manifolded, externally manifolded reported it is also not a problem)</li> <li>- Reduce segregation (gradation of two ion species)</li> <li>- Reduce NiO solubility</li> </ul>
'D3	<p>Lack of methodology for accelerated long-term testing</p>
	<i>Reliability</i>
	<p>No AR&amp;TD issues identified.</p>
	<i>Robustness</i>
'R1	<p>Negative Cells operation</p> <ul style="list-style-type: none"> <li>- Examine effect on other cells</li> <li>- Understand mechanism</li> </ul>
	<i>Cost and Performance</i>
'P1	<p>Cell conversion efficiency</p> <ul style="list-style-type: none"> <li>- Reduce polarization</li> <li>- Improve heat removal/thermal management</li> </ul>
'P2	Improve volumetric density (W/ft <sup>3</sup> )
'P3	Improve footprint (W/ft <sup>2</sup> )
'P4	Develop thinner cells
	<i>Looking Toward the Future</i>
'N1	Develop new and innovative ideas. Future generation.

These were ranked in the following table. Note that “thinner cells” received no votes and was not ranked.

<b>AR&amp;TD Issues by Priority</b>		
<i>Rank</i>	<i>Issue</i>	<i>Total Points</i>
1	P1 - Cell conversion efficiency	33
2	D1 - Separator plate materials	18
3	D2 - Electrolyte (match to operating conditions)	16
4	D3 - Lack of methodology for accelerated long-term testing	6
4	P2 - Improve volumetric density (W/ft <sup>3</sup> )	6
6	P3 - Improve footprint (W/ft <sup>2</sup> )	6
7	N1 - Develop new and innovative ideas. Future generation.	2
8	P4 - Negative cells operation	1

<b>Non-AR&amp;TD Issues</b>	
Code	
	<i>Durability</i>
‘D4	Seal integrity
‘D5	Reuse/recycle materials and components
‘D6	Repairableness
‘D7	Stability of reforming catalysts
‘D8	Thermal cycle ability
	<i>Reliability</i>
‘R2	Defect-free manufacturing repeatability
	<i>Robustness</i>
‘R3	Ability to operate under off-design conditions
	<i>Cost and Performance</i>
‘C1	Improve manufacturing processes and assembly
‘C2	Commercially available components and materials
‘C3	Design simplification
‘C4	Compact/portable system/develop other applications
	<i>Looking Toward the Future</i>
	Nothing included in this topic.

These were ranked in the following table. For these issues, there was little difference in most of the topic areas. However, “stability of reforming catalysts” received no votes and was not ranked.

Non-AR&TD Issues by Priority		
Rank	Issue	Total Points
1	R3 - Ability to operate under off-design conditions	10
1	C2 - Improve manufacturing processes and assembly	10
3	D8 - Thermal cycle ability	7
3	D4 - Seal integrity	7
5	D5 - Reuse/recycle materials and components	6
6	D6 - Repairability	5
6	R2 - Defect-free manufacturing repeatability	5
6	C3 - Design simplification	5
9	C1 - Improve manufacturing processes and assembly	3
10	C4 - Compact/portable system/develop other applications	2

## B. Session on SOFC-Stack AR&TD Issues

Facilitator: *Janice Murphy (FETC)*

Scribe: *William Cary Smith (FETC)*

### Summary

A group of manufacturers, developers, researchers, and industry personnel gathered to discuss the specific research needs for solid oxide fuel cell advanced research and technology development. The group identified 13 specific areas of interest. Each participant then voted for the two most critical issues from their viewpoint. These votes were tabulated and are shown in the following table.

## Specific Needs for SOFC AR&TD

	Area	Votes Cast
1	Seals for planar in short term, and tubular in long term	4
2	Low-cost fabrication including thin films for electrolyte and interconnect	11
3	Stack development for low-temperature planar material	6
4	Interconnect issues, stability, material selection (metallic/ceramic)	3
5	Hydrocarbon operation of low-temperature fuel cells	4
6	Thin film technologies with low cost	0
7	New materials to enhance chemical capabilities	0
8	Availability needs (when will they be ready?)	0
9	Funding on development scale for planar	9
10	Balance between power density and efficiency (optimize life cycle costs; area of specific resistance)	0
11	Reliability/maintenance/endurance from utility standpoint	4
12	Thermal cycling (tubular and especially planar)	0
13	De-emphasize multi-function fuel cells (modular design for low cost)	1

## Discussion

The top three categories: ***low-cost fabrication, funding, and stack development at low temperature***, were topics discussed in more detail.

There was general agreement that there needed to be more work conducted in electrolyte and interconnect fabrication emphasizing low fabrication costs.

The group would like to see DOE funding increased for intermediate temperature planar fuel cells. However, the group did not have a plan on how this might be done. Other sources of funding were not suggested. An alternative of recoupment was explored and the majority (11 to 1) of those voting favored this approach.

The third priority was the development of stacks for low temperatures in the 600 to 800 °C range. This should include system integrations of existing technology, modeling component and complete power systems, hydrocarbon operation results, and critical data base development.

## **C. Session on Other Fuel Cell Technologies and Applications**

**Facilitator: *Daniel Rastler (EPRI)***

**Scribe: *Damon Benedict (FETC)***

### **Summary**

About 13 people attended this breakout session, representing manufacturers, developers, researchers, and government. The discussion primarily related to Proton Exchange Membranes (PEMs), including PEM applications, R&D needs, and approach to meet these R&D needs. Specific suggestions included:

#### **PEM Applications:**

- Portable Power (about 200 kW)
- Small Stationary Power (2 -1,000 kW)
- Transportation
- Auxiliary Power Units (2 -60 kW)
- Emergency Applications
- Biomass, Industrial, or Rural Utilization

#### **PEM R&D Needs:**

- Fuels Logistics
- Fuel Processing
- Fuel Cell Concerns, including
  - effects of sulfur and trace organics
  - operation at higher temperatures
  - new catalysts
- Hydrogen Storage

#### **PEM R&D Approaches:**

- Define Needs and Problems
- Present the Needs and Problems to Other Disciplines (e.g., at conferences, etc.)
- Good Front-End Coordination Utilizing Consortia via the ATS Experience
- Develop Transition Plan
- Identify High-Value Market Entry

Other fuel cell applications that were discussed included utilization of biomass gas and logistic fuels, regenerative systems, and hybrid systems.

## **D. Fuel Cell Balance-of-Plant Session**

**Facilitator:** *John Wimer (FETC)*

**Scribe:** *Kevin Krist (GRI)*

### **Attendees**

Around 15 people attended the session, including but not limited to representatives from fuel cell and turbine manufacturers, public utilities, research organizations, architecture-engineering firms, and government agencies. An attendance list was distributed and taken up by the session organizer.

**Objective:** To assemble and prioritize advanced research and technology development (AR&TD) ideas for the fuel cell balance-of-plant (BOP).

**Agenda:** (1 hour session)

- Introduction (5 minutes)
- Brainstorm ideas (20 minutes)
- Discuss pros and cons of each idea (20 minutes)
- Prioritize ideas (10 minutes)

### **Brainstorming**

During brainstorming, the following areas, listed in no particular order, were suggested for AR&TD work on the fuel cell BOP. It was noted that, depending on the definition of AR&TD, some of these areas may not be appropriate for AR&TD.

- A. Power Conditioning
  - 1. cost effective inverters
  - 2. voltage stabilizers
  - 3. transformers
- B. Fuel Processing
  - 1. reformer (with and without shift)
  - 2. multi-fuel capability
- C. Cleanup of Fuel, Air and Water
  - 1. sulfur removal
- D. Siting Requirements
  - 1. climate proofing for marine environments (salt ingestion)
  - 2. siting codes and standards, e.g., indoor/outdoor operation, operation in enclosed spaces

- E. Controls
- F. Sensors
  - 1. long-term (8,000 - 10,000 hours) stability of sensors and transmitters
- G. Steam Turbines
- H. Turbomachinery, e.g., Gas Turbines
- I. Modular Packaging
- J. Standardization
- K. BOP Economics
  - 1. identify cost reduction opportunities by benchmarking BOPs of competing power technologies
- L. System Integration/Modeling
  - 1. combinations of different fuel cells, e.g., high and low temperature cells
- M. Insulation (thermal/electrical)
  - 1. highly effective, low cost
- N. Heat Exchangers
  - 1. options to eliminate
- O. Systems Development for New Applications and Market Studies
  - 1. develop BOP for small capacity FC systems
  - 2. mobilization, including motion sensitivity and high shock capability
  - 3. provisions for cogeneration
  - 4. potable water
- P. Cost Reduction for Installation and Operation and Maintenance
  - 1. make the BOP user friendly
- Q. Designs for Different Load Profiles, e.g., Peak, Intermediate, Base

## **Prioritization of AR&TD Areas for the Fuel Cell BOP**

The 17 areas listed above, lettered A-Q, were prioritized using a multi-voting technique. Each attendee had three “votes” to distribute as he or she wished among the 17 areas, including assigning multiple votes to a single area. The results of the prioritization are shown in the following table.

Priority	AR&TD Area for Fuel Cell BOP	Votes Received
1	Fuel Processing	7
1	Power Conditioning	7
3	Siting Requirements	5
4	Insulation	4
4	System Integration/Modeling	4
4	Turbomachinery	4
7	Cleanup of Fuel, Air, and Water	2
7	Systems Development for New Applications and Market Studies	2
9	Cost Reduction for Installation and Operation and Maintenance	1
9	Modular Packaging	1
10	BOP Economics	0
10	Controls	0
10	Designs for Different Load Profiles	0
10	Sensors	0
10	Standardization	0
10	Steam Turbines	0

## CLIMATE CHANGE FUEL CELL PROGRAM

New Program Boosts Fuel Cell Industry

### Project Benefits

- Fuel cells are similar to batteries in that they both produce a DC current by using an electrochemical process. Two electrodes, an anode and a cathode, are separated by an electrolyte.
- Like batteries, fuel cells are combined into groups, called stacks, to obtain a usable voltage and power output.
- Unlike batteries, fuel cells do not "run down" because they receive their fuel source from outside the cells. Instead, they convert the energy from a hydrogen-rich fuel directly into electricity. They will continue to operate as long as they are supplied with fuel and air.
- Fuel cells emit almost none of the sulfur and nitrogen compounds released by conventional generating methods, and can utilize a wide variety of hydrogen-rich fuels: natural gas, coal-derived gas, landfill gas, biogas, or alcohols.

### Three types of fuel cells are targeted for stationary power generation:

- Phosphoric Acid Fuel Cells (PAFC)** are the most mature commercially available fuel cell technology. They operate at about 200°C (400°F), and their electrical efficiency can exceed 40 percent (LHV). With the use of by-product heat, total efficiency can reach 80 percent.
- Molten Carbonate Fuel Cells (MCFC)** are now being tested in full-scale demonstration plants. They offer higher fuel-to-electricity efficiencies (approaching 60 percent LHV), and operate at higher temperatures (about 650°C, or 1200°F). When the by-product heat is used, total efficiencies can approach 85 percent.
- Solid Oxide Fuel Cells (SOFC)** are currently being tested in a 100-kilowatt plant. They offer the stability and reliability of all-solid-state ceramic construction. Their high-temperature operation (up to 1000°C, or 1800°F) allows more flexibility in the choice of fuels. Like MCFC, SOFC approach 60 percent electrical efficiency LHV, and 85 percent total efficiency.

### Cost Profile

(Dollars in Millions)

	FY95	FY96	FY97	FY98	FY99
<b>Department of Defense*</b>	\$8.4	—	—	—	—
<b>Private Sector Partners</b>	—	\$4.1	\$21.2	\$8.7	\$0.9

\* Appropriated Funding

### Key Milestones

FY95	FY96	FY97	FY98	FY99	FY00	FY01
Funding	Solicitation	Acceptance Testing		Operation Testing		
Funding authorized	Solicitation completed		Acceptance testing completed		Operations testing completed	

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# PROJECT facts

DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
FEDERAL ENERGY TECHNOLOGY CENTER

NATURAL gas  
RESEARCH

NG001.0897M

## CLIMATE CHANGE FUEL CELL PROGRAM

New Program Boosts Fuel Cell Industry

### Project Description

Fuel cells have emerged in the last decade as one of the most promising new technologies for meeting the Nation's energy needs well into the 21st century. Unlike power plants that use conventional technologies, fuel cell plants that generate electricity and usable heat can be built in a wide range of sizes—from 200-kilowatt units suitable for powering commercial buildings, to 100-megawatt plants that can add baseload capacity to utility power plants.

The Climate Change Fuel Cell Program is a joint effort of the U.S. Departments of Defense (DoD) and Energy (DOE) that was authorized by Congress in the fiscal year 1995 Defense appropriations bill. The DOE Federal Energy Technology Center (FETC) is implementing and managing the program, providing financial assistance to buyers of fuel cells who are energy-service providers, utilities, or end users.

Fuel cells offer extremely high efficiency in the production of electricity, yet very low pollutant emissions. The objectives of the Program are to reduce greenhouse emissions, accelerate fuel cell commercialization, and satisfy DoD goals for the environment and U.S. economy.

Congress authorized additional funding for fiscal years 1996 and 1997 through the Defense appropriations bill. It is expected an additional \$14 million will be awarded through this program.

### Program Goal

Thirty-three awards were made to utility, end user, and energy service providers for utility, commercial, or industrial use of fuel cells in the U.S. and abroad. Most of the projects are for 200-kilowatt fuel-cell installations, but several are for 400-, 600-, or 800-kilowatt operations. All grantees are purchasing fuel cells from the ONSI Corporation of South Windsor, Connecticut. ONSI is the marketing subsidiary of International Fuel Cells.

The 33 grantees are purchasing a total of 42 ONSI 200-kilowatt units, to be used as single units or multiple arrays of the 200-kilowatt module.

The program is a key element of the Federal Administration's Climate Change Action Plan, which is an effort to curb emissions of greenhouse gases in part through the development of environmentally clean energy technologies. The intent of the program is to satisfy DoD goals for the environment, readiness, and economy through activities that would stimulate end-user applications. This would stimulate a major expansion of fuel-cell manufacturing capability in the U.S. and, in turn, bring mass-produced fuel cells into a cost range that is competitive with more traditional sources.

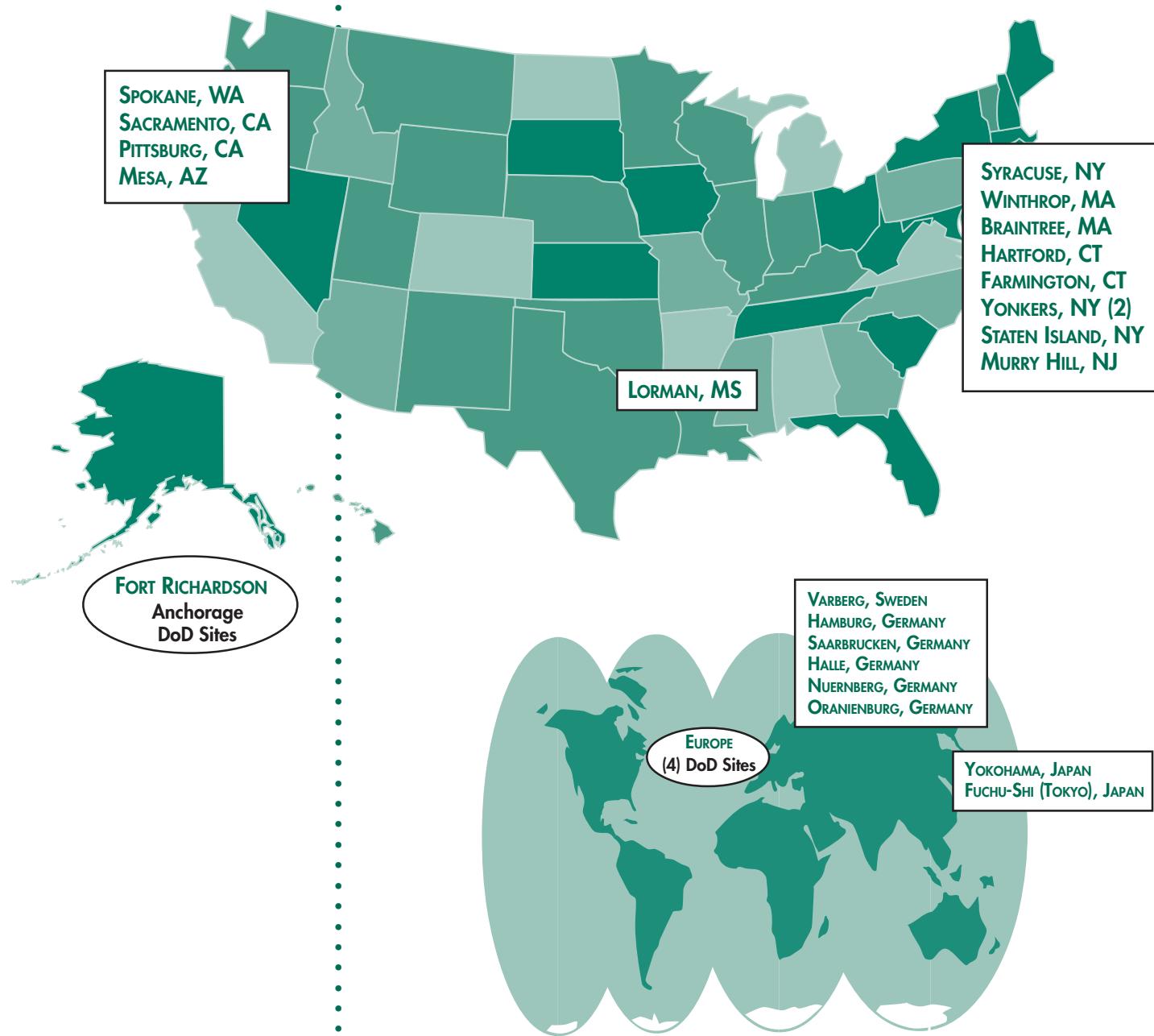
Since fuel cells generate electricity electrochemically, rather than mechanically, they are more efficient over a wider load factor and can cut greenhouse gases by over 50 percent.



## PROGRAM FACTS

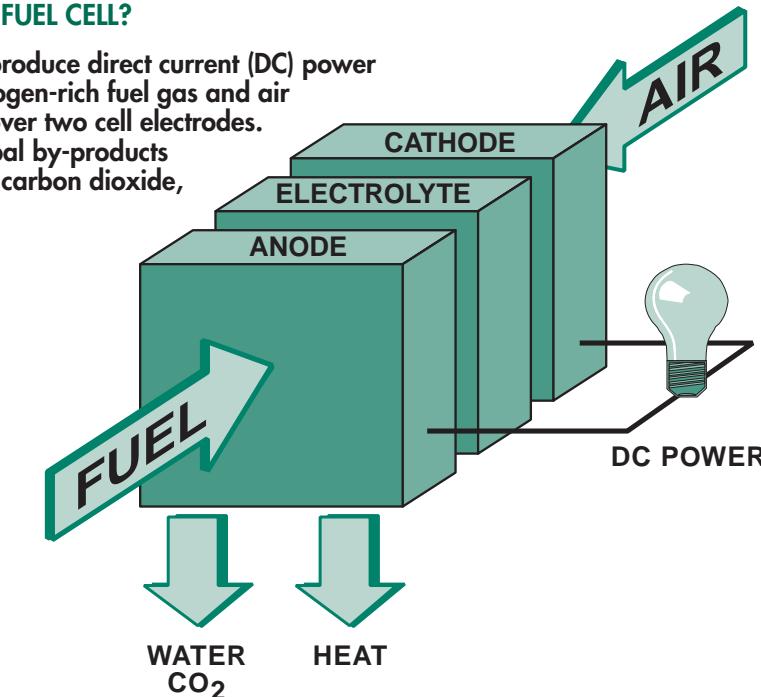
ACCEPTANCE TEST	TYPE OF APPLICANT	TYPE OF USE
1996 = 5	Utility = 12	Utility = 6
1997 = 24	End User = 10	Industrial = 8
1998 = 4	Energy Service = 7	Commercial = 18
	Mixed (End User + Utility or Utility + Electric) = 4	Mixed (Utility/Commercial) = 1

## FUEL CELLS IN THE U.S. AND ABROAD



## WHAT IS A FUEL CELL?

Fuel cells produce direct current (DC) power from hydrogen-rich fuel gas and air that flow over two cell electrodes. The principal by-products are water, carbon dioxide, and heat.



## SPECIFIC USES:

- Heating District Building/Network
- Hotel
- Laboratory/Processing Plant
- Manufacturer
- Medical Facility
- Military Housing/Facility
- Office Building
- University/School
- Wastewater Treatment

## WHY CHOOSE THE ONSI FUEL CELL?

The ONSI PC25™ is a phosphoric acid fuel (PAFC). PAFC technology is already a commercial reality. ONSI's 200-kW modular unit was first installed in 1992 and since then, over 100 units have been sold worldwide.

## UNIQUE FEATURES:

- Transportable (1)
- Premium Power (2)
- Anaerobic Digester (2)
- Anaerobic Gas (1)
- Propane (1)
- Landfill Gas (1)
- Hydrogen Fueled (1)

# PROJECT facts

DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
FEDERAL ENERGY TECHNOLOGY CENTER

ADVANCED CLEAN/EFFICIENT  
POWER systems

PS023.0897M

## PRIMARY PROJECT PARTNER

**Energy Research Corporation**  
Danbury, CT

## MAIN SITES

**Santa Clara, CA**  
(Demonstration)

**Torrington, CT**  
(Manufacturing)

## TOTAL ESTIMATED COST

### Product Design Improvement

**\$139,214,992**

### Santa Clara Demonstration

**\$53,388,290**

## COST SHARING

### Product Design Improvement

**DOE            \$99,571,776**

**Non-DOE    \$39,643,216**

### Santa Clara Demonstration

**DOE            \$28,608,748**

**Non-DOE    \$24,779,542**

## DEVELOPING THE SECOND-GENERATION FUEL CELL — THE ENERGY RESEARCH PROJECT

### Project Description

The Energy Research Corporation (ERC) in Danbury, Connecticut, is one of the Nation's principal developers of stationary fuel cell power plants — a technology that is among the cleanest and most efficient available for the 21<sup>st</sup> century.

Sharing costs with the Department of Energy's Office of Fossil Energy, ERC demonstrated a carbonate fuel cell power system at the

Santa Clara, California. This power plant operated on pipeline natural gas in a grid-connected mode for 4,000 hours, delivering 2,500 MWh of energy to the City of Santa Clara grid, a record for a first-of-a-kind MW size fuel cell power plant. The project was jointly funded by FETC and the SCDP (EPRI, NRECA, City of Santa Clara Electrical Department and six utilities from California and Arizona: City of Los Angeles Department of Water and Power, City of Vernon Light and Power Department, National Rural Electric Cooperative Association, Sacramento Municipal District, and Southern California Edison Company). Equipped with the Santa Clara experience, ERC is currently defining the commercial system design. ERC plans to soon implement a second demonstration of this power plant which is expected to deliver similar power levels in a plant one-tenth the size. This 2 MW second demonstration is a key milestone in the commercial entry of second-generation fuel cell technology.

Carbonate fuel cells offer higher fuel-to-electricity efficiencies than the fuel cell systems now being marketed, and they are also expected to have much lower capital costs. Quiet and virtually pollution-free, they are scheduled to become commercially available by the year 2002. The commercial system is expected to consist of 375-kilowatt stacks, with four stacks per module. Initially, the cells will be fueled by natural gas; later, as the technology advances, fuel sources could be expanded to include gas made from coal or biomass.

ERC has constructed a manufacturing facility in Torrington, Connecticut, to fabricate full-size stacks for design verification and demonstration units. The current capacity of this facility of 17 MW/yr is being expanded to 50 MW/yr to manufacture early market products. The company received an endorsement from the American Public Power Association and the Electric Power Research Institute, and a buyers' group has been actively collaborating with ERC to provide user input.

ERC has also joined forces with Fluor Daniel, Inc., a leading international supplier of services to the power industry, and MTU (Germany), an affiliate of Daimler Benz to explore worldwide opportunities.



## DEVELOPING THE SECOND-GENERATION FUEL CELL — THE ENERGY RESEARCH PROJECT

### CONTACT POINTS

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### Project Partners

**ELECTRIC POWER RESEARCH INSTITUTE**  
Palo Alto, CA

**NATIONAL RURAL ELECTRIC  
COOPERATIVE ASSOCIATION**  
Washington, DC

**THE CITY OF SANTA CLARA  
THE SACRAMENTO MUNICIPAL  
UTILITY DISTRICT**  
Sacramento, CA

**SOUTHERN CALIFORNIA EDISON**  
Rosemead, CA

**THE LOS ANGELES  
DEPARTMENT OF WATER  
AND POWER**  
Los Angeles, CA

**THE CITY OF VERNON, CA**  
Vernon, CA

### Development Partners

**FUEL CELL ENGINEERING**  
Danbury, CT

**FUEL CELL MANUFACTURING  
CORPORATION**  
Torrington, CT

**FLUOR DANIEL, INC.**  
Irvine, CA

**JACOBS APPLIED TECHNOLOGY**  
Orangeburg, SC

**MOTERN-UND TURBINEN-UNION (MTU)**  
Munich, Germany

**FUEL CELL COMMERCIALIZATION GROUP**  
Washington, DC

**SANTA CLARA DEMONSTRATION PROJECT**  
Santa Clara, CA

### Program Goals

- Fuel cell technology is modular and lends itself well to dispersed power generation.
- Fuel cells could be sited at electrical substations or at the point of end use, such as a hospital or shopping mall. ERC's carbonate fuel cell system features a unique Direct Fuel Cell concept, which eliminates auxiliary equipment and simplifies the power system.
- ERC's Santa Clara power unit will be the technological forerunner of the company's commercial product line. This demonstration, coupled with the company's continued product development and commercial system demonstrations, will maintain the United States' position as world leader in advanced fuel cell technology.
- The program goals are to commercialize the tubular SOFC by 2002. Commercialization of the technology supports DOE goals for emissions reduction and energy security.

### Project Benefits

- Fuel cells have emerged as one of the most promising new power-generation technologies for the 21<sup>st</sup> century. Endorsed by President Clinton's Climate Change Action Plan, fuel cells are an environmentally clean, quiet, and highly efficient method for generating electricity and heat from natural gas and, potentially, other fuels.
- Carbonate fuel cell technology is increasingly attractive because it offers several advantages over conventional power plants, as well as today's market-entry (phosphoric acid) fuel cell systems:
  - Fuel-to-electricity efficiencies can exceed 60%, well above the 33% to 35% efficiencies of today's conventional power plants and the 40% to 45% efficiencies of phosphoric acid systems; when the waste heat is utilized, total thermal efficiencies can approach 85%.
  - Higher operating temperatures (approximately 650°C compared with 200°C for first-generation systems) make the molten carbonate fuel cell a better candidate for combined-cycle applications (where exhaust heat is used in a steam cycle to generate additional electricity).
  - The projected cost of the technology is competitive.
  - The technology exceeds all current and envisioned environmental regulations, producing water and carbon dioxide as the only emissions (the amount of carbon dioxide released per unit of electricity is considerably less than current power-generating technologies, because of the higher efficiencies).

# PROJECT facts

ADVANCED CLEAN/EFFICIENT  
POWER systems

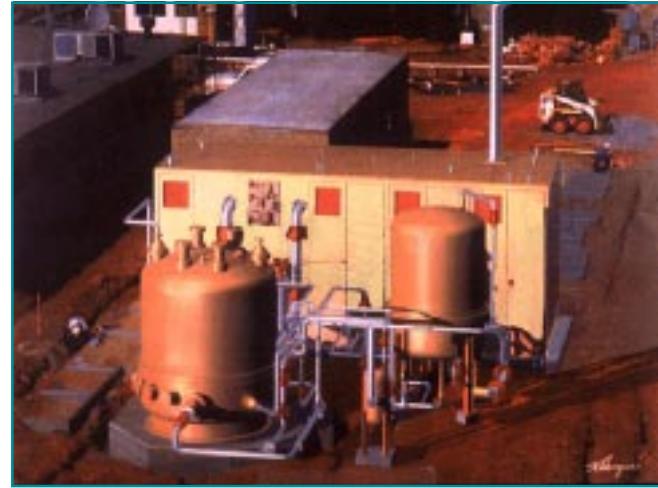
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DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
FEDERAL ENERGY TECHNOLOGY CENTER

## DEVELOPING THE SECOND-GENERATION FUEL CELL — THE M-C POWER PROJECT

### Project Description

The M-C Power Corporation, headquartered in Burr Ridge, Illinois, has tested the prototype of the next generation of fuel cell technology in San Diego, California, and is now in a 5-year development effort (1994-1999) to improve the design and reduce costs for the first market-entry units.



### PRIMARY PROJECT PARTNER

**M-C Power Corporation**  
Burr Ridge, IL

### MAIN SITES

**San Diego, CA**  
(Demonstration)

**Burr Ridge, IL**  
(Manufacturing)

### TOTAL ESTIMATED COST

#### Product Design Improvement

**\$104,216,627**

#### San Diego Demonstration

**\$29,326,821**

### COST SHARING

#### Product Design Improvement

**DOE        \$70,861,366**

**Non-DOE    \$33,355,261**

#### San Diego Demonstration

**DOE        \$16,571,222**

**Non-DOE    \$12,755,599**

Molten carbonate fuel cells operate at higher temperatures (in excess of 200°F) than first-generation phosphoric acid systems (which run at about 400°F). Higher operating temperatures increase power-generating efficiencies and correspondingly decrease emissions of carbon dioxide, a greenhouse gas. A molten carbonate fuel cell operates at nearly twice the efficiency of a coal combustion plant, reducing carbon dioxide emissions by nearly 50%.

The unit installed in San Diego, at the Miramar Naval Air Station, generated 250 kilowatts of electricity. Also, heat produced by the fuel cell was used in adjacent buildings at the site. This cogeneration approach raises overall efficiencies to nearly 85% — meaning that 85% of the energy value of the fuel is used.

Because fuel cells produce electricity and heat by an electrochemical process — like a battery — they emit virtually none of the sulfur and nitrogen pollutants associated with combustion processes.

Prior to the San Diego test, M-C Power tested its configuration in four 11-square-foot stacks, the largest size to be fabricated to date. In the 250-kilowatt stack, 250 of these full-size cells are assembled into a compact module.

The first project began in September 1990, and the latest in December 1994. The total effort is expected to end in December 2000.

The first unit was installed in Brea, California, and was operated in 1995. The second unit was operated at Miramar Naval Air Station in San Diego in 1997. This unit operated for 2,350 hours and delivered 158 megawatts per hour of direct current output and 296,500 pounds of 110 psig steam to the base.

This program consists of three separate projects. The first is a \$60.5 million contract, which includes \$18.1 million from M-C Power. The second is for \$30.8 million, including \$14.2 million from M-C Power. The third is a \$104 million product development and improvement project, with \$33 million from M-C Power.

## DEVELOPING THE SECOND-GENERATION FUEL CELL — THE M-C POWER PROJECT

### Program Goals

The M-C Power demonstration in San Diego provided valuable experience in installing and operating a molten carbonate fuel cell in a commercial cogeneration application. Fuel cell modules comparable in size to those installed at the demonstration site will be linked together for larger power plant applications. The demonstration, coupled with the company's continued product development, will keep the United States the world's leader in advanced fuel cell technology.

The program goals are to commercialize the tubular MCFC by 2002. Commercialization of the technology supports DOE goals for emissions reduction and energy security.

### Project Benefits

Fuel cells have emerged as one of the most promising new power-generating technologies.

Endorsed by President Clinton's Climate Change Action Plan, fuel cells are an environmentally clean, quiet, and highly efficient method for generating electricity and heat from natural gas and potentially other fuels.

Molten carbonate fuel cell technology is becoming increasingly attractive because it offers several advantages over today's market-entry (phosphoric acid) fuel cell systems:

- Fuel-to-electricity efficiencies can exceed 55%, well above the 33%-35% efficiencies of today's conventional power plants and the 40% to 45% efficiencies of phosphoric acid systems; when the waste heat is utilized, total thermal efficiencies can approach 35%.
- The higher operating temperatures (approximately 1,200°F, compared with 400°F for first-generation systems) make the molten carbonate fuel cell a better candidate for combined-cycle applications (where exhaust heat is used in a steam cycle to generate additional electricity).
- The technology is expected to be much lower in capital cost, approaching \$1,200 per installed kilowatt, or less than today's conventional coal-fired power technology.
- The technology exceeds all current and envisioned environmental regulations, producing water and CO<sub>2</sub> as the only emissions (the amount of CO<sub>2</sub> released per unit of electricity is considerably less than current power-generating technologies because of the higher efficiencies).

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### Project Partners

**SAN DIEGO GAS AND ELECTRIC**  
San Diego, CA

**GAS RESEARCH INSTITUTE**  
Chicago, IL

**BECHTEL CORPORATION**  
San Francisco, CA

**STEWART AND STEVENSON**  
Houston, TX

**ELECTRIC POWER RESEARCH INSTITUTE**  
Palo Alto, CA

**SOUTHERN CALIFORNIA GAS**  
Los Angeles, CA

**INSTITUTE OF GAS TECHNOLOGY**  
Mt. Prospect, IL

# PROJECT facts

ADVANCED CLEAN/EFFICIENT  
POWER systems

PS027.0897M

DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
FEDERAL ENERGY TECHNOLOGY CENTER

## PHOSPHORIC ACID FUEL CELL COMMERCIALIZATION

### Project Description

The ONSI Corporation of the U.S. is the only manufacturer of commercial phosphoric acid fuel cell (PAFC) units in the world. This company has the capability to produce 200 PC25™ units per year in its facility using robotics manufacturing and automated assembly techniques. The PC25™ converts 1900 SCF per hour of natural gas into 200 kW of grid-connected or grid-independent premium power and up to 750,000 Btu/hr of useful thermal energy at up to 250°F.

### Program Goals

More than 400 PC25 fuel cell power plants have been ordered from ONSI as of May 1997. Eighty-two units are now operational at sites in the United States, Europe, and Asia.

Eight units have operated more than 30,000 hours with the first unit expected to surpass the 40,000 cell stack design life in late 1997. The longest continuous operation record has been set, and then surpassed by two units operating in the Tokyo Gas Company service territory. The first company record of 9,477 hours was set in September 1996. This was followed by a second Tokyo Gas Company unit which operated for 9,500 hours before being shut down for an inspection required by Japanese regulations.



The fleet continues to show an impressive availability, above 95%. The latest PC25™ design, the Model C, has entered service world-wide. Twenty-five Model C units are operating on three continents as of May 1997, and the world-wide fleet has now operated for a total of 1,450,000 power plant operating hours.

ONSI continues to focus on PC25™ manufacturing cost reduction, with lower costs expected as higher production volumes are achieved.

DOE's Office of Fossil Energy was instrumental in supporting the development of PAFC stack and system technology.

Continued efforts include a cooperative government/private sector partnership at no additional cost to the government.

# PHOSPHORIC ACID FUEL CELL COMMERCIALIZATION

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## Project Benefits

The PC25 demonstrates the technical viability and environmental cleanliness of fuel cell technology. These fuel cells have been sited, permitted, installed, started, operated, and maintained in a real world environment. The fleet continues to demonstrate reliable, safe operation in a variety of climates, applications, and service scenarios. Ambient temperatures range from sub-zero plus 100°F. The table on the overleaf shows the results from PC25 operation by U.S. owners. A significant premium power market is emerging for the PC25, in which the fuel cell's stable, and quality electric output is providing energy service to critical loads in commercial buildings. PC25s have been installed as back-up generators, as well as the primary source of supply for dedicated loads, replacing conventional uninterruptable power supplies. Other units are demonstrating the fuel flexibility of fuel cell power plants by operating on biomass fuels from landfills and anaerobic digester plants. The success of the PC25 is best illustrated by two recent awards:

- 1995 Cogeneration Project of the Year (Brooklyn Union, NY Installation) by the Cogeneration and Competitive Power Institute.
- 1994 Efficient Building Award for Energy and the Environment (Kaiser Permanente Medical Center Installation) by Energy User News.

## Major North American PC25 Owners' Findings

Category	Result	Comment
Target Installation Cost	\$50,000 to \$100,000	Simple and short interconnects
Permitting	Nothing unusual	Units have AGA approval/seal, some states have blanket EPA permits
Electrical Interfaces	Grid Connected and/or Grid Independent parallel	Widely accepted by electric utility
Maintenance	Over 9000 hours between major maintenance demonstrated	PC25C annual maintenance ONSI or other contractors available for maintenance
Availability	PC25A = 88.7% raw	Estimate 95+% with local parts and service PC25C expected to be 95+% raw

# PROJECT facts

ADVANCED CLEAN/EFFICIENT  
POWER systems

PS025.0897M

DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
FEDERAL ENERGY TECHNOLOGY CENTER

## DEVELOPING THE SOLID OXIDE FUEL CELL

### Project Description

Westinghouse Electric Corporation is developing the tubular ceramic-based solid oxide fuel cell — potentially one of the simplest, cleanest, most efficient, and most versatile technologies on the power generation horizon. Westinghouse is widely recognized as the world leader in this promising new technology.

The \$173-million effort — 48% of the funding is coming from the private sector — is a six-year development project aimed at moving this 21<sup>st</sup> century technology up to the threshold of commercial use.

Like a battery, the solid oxide fuel cell generates power electrochemically avoiding the air pollutants and efficiency losses associated with combustion processes. Unlike batteries fuel cells operate continuously, generating power as long as natural gas, clean-coal-derived gas, or other hydrocarbon fuels are supplied. The solid electrolyte allows for the simplest of fuel cell plant designs, and requires no external fuel reforming.

The solid oxide concept uses ceramics, which allows the cells to operate at higher temperatures than other fuel cells, producing more energy per unit of fuel and far less carbon dioxide (a greenhouse gas). The high exhaust temperature and pressurization potential of the Westinghouse design make it particularly suited for multiple combined cycles and high efficiencies.

Westinghouse's configuration is a tube made up of multiple ceramic layers bonded together. Multiple tubes link to form power modules; modules link to form generators or larger power plants.

The development effort culminated in tests of two 25-kilowatt generators, and the building of a 100-kilowatt generator. The megawatt-class plant will incorporate as many as 5,000 individual solid oxide fuel cells. The modules will be tested at utility and industrial sites.

### PRIMARY PROJECT PARTNER

**Westinghouse Electric Corporation**  
Pittsburgh, PA

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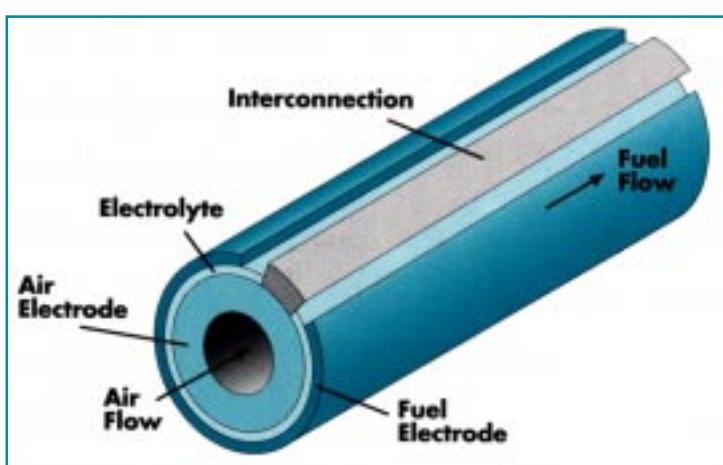
**Westinghouse Science and Technology Center**  
Pittsburgh, PA

### COST

**\$173,800,000**

### COST SHARING

<b>DOE</b>	<b>\$89,300,000</b>
<b>Non-DOE</b>	<b>\$83,700,000</b>



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## Project Partners

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**ELECTRIC POWER RESEARCH  
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## DEVELOPING THE SOLID OXIDE FUEL CELL

### Program Goals

The clean environmental performance of solid oxide fuel cells makes them especially well-suited for areas with strict air-quality requirements. Future units could cogenerate electricity and steam for hospitals, shopping malls, and large residential or commercial complexes. Both urban centers and remote sites (for example, those with relatively low-cost fuel sources such as coal-bed methane operations) could be candidates for solid oxide fuel cells.

Commercialization of the Westinghouse solid oxide fuel cell could offer a new approach to generating power in the United States and worldwide. It could create a new solid state manufacturing industry, employing skilled workers to design and fabricate power technologies for tomorrow's energy needs.

### Project Benefits

The solid oxide fuel cell is one of the cleanest, most efficient power-generating technologies now being developed.

Capable of using either natural gas or cleaned coal gas, it emits no sulfur pollutants and as much as 60% to 65% less carbon dioxide (a greenhouse gas) than a conventional coal-burning plant.

It is also one of the most efficient means for generating electricity and usable heat. As a simple cycle power generator, it can convert more than 55% of the energy in its fuel source to electricity (conventional coal plants, for example, operate at efficiencies of only 33% to 35%). When the high-quality waste heat from the electrochemical process is used, overall efficiencies could reach 85%. When utilized with a gas turbine in a combined power system, efficiencies over 70% can be achieved.

Because they involve no liquid or moving parts, solid oxide modules are expected to operate reliably for many years. A unit that can generate 2 megawatts of electricity, enough for a small substation, can fit on less than one-tenth of an acre. Thus it can be placed close to power needs, avoiding long transmission lines.

With a simple adjustment of air and fuel flows — much as a gas pedal is used in a car — a solid oxide fuel cell can easily follow changing demands for electricity, boosting output when necessary, then cycling down when demand is low.

The all-solid-state composition of these fuel cells promises to bring to the electric power sector the mass-production processes that have reduced costs in the electronics industry.

# PROJECT facts

ADVANCED CLEAN/EFFICIENT  
POWER systems

PS026.0897M

DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
FEDERAL ENERGY TECHNOLOGY CENTER

## SOLID OXIDE FUEL CELL PROJECT GENERATING TOMORROW'S ELECTRICITY CLEANLY

### Project Description

Westinghouse Electric Corporation is developing the tubular ceramic-based oxide fuel cell, which are one of the simplest, cleanest, most efficient, and most versatile technologies on the power-generation horizon. Westinghouse is widely recognized as the world leader in this promising new technology.

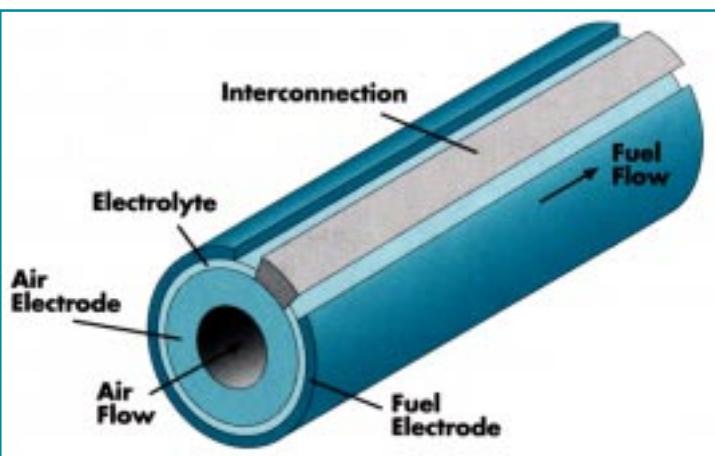
The \$200-million effort — 59% of the funding is coming from the private sector — is a 5-year development project aimed at moving this 21<sup>st</sup> century technology up to the threshold of commercial use.

Like a battery, the solid oxide fuel cell generates power electrochemically, avoiding the air pollutants and efficiency losses associated with combustion processes. Unlike batteries, fuel cells operate continuously, generating power as long as natural gas, clean coal-derived gas, or other hydrocarbon fuels as supplied. The solid electrolyte allows for the simplest of fuel cell plant designs, and requires no external fuel reforming.

The solid oxide concept uses ceramics, which allows the cells to operate at higher temperatures than other fuel cells, producing more energy per unit of fuel and far less carbon dioxide (a greenhouse gas). The high exhaust temperature and pressurization potential of the Westinghouse design make it particularly suited for multiple combined cycles and high efficiencies.

Westinghouse's configuration is a tube made up of multiple ceramic layers bonded together. Multiple tubes link to form power modules; modules link to form small generators or submodules for larger power plants.

The development effort will culminate in tests of a 250-kilowatt fuel cell-micro turbine combined cycle power plant, a 800-kilowatt fuel cell-turbine combined cycle power plant, a 1,300 kilowatt fuel cell-turbine combined cycle power plant, and a 2500-kilowatt fuel cell-turbine combined cycle power plant. These modules will be tested at utility and industrial sites.



### PRIMARY PROJECT PARTNER

**Westinghouse Electric Corporation**  
Pittsburgh, PA

### MAIN SITES

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Pittsburgh, PA

**Southern California Edison**  
Los Angeles, CA

**Environmental Protection Agency**  
Forte Meade, MD

**Ontario Hydro**  
Ontario Canada

### COST

**\$202,550,978**

### COST SHARING

DOE	\$82,903,796
Non-DOE	\$119,647,182

## SOLID OXIDE FUEL CELL PROJECT

GENERATING TOMORROW'S ELECTRICITY CLEANLY

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### Program Goals

Commercialization of the Westinghouse concept — the only fuel cell type in which American technology clearly leads the world — could offer a new approach to generating power in the United States and worldwide. It could create a new solid state manufacturing industry, employing skilled workers to design and fabricate power technologies for tomorrow's energy needs.

The program goals are to commercialize the tubular SOFC by 2002. Commercialization of the technology supports DOE goals for emissions reduction and energy security.

### Project Benefits

The solid oxide fuel cell is one of the cleanest, most-efficient power-generating technologies now being developed.

Capable of using either natural gas or clean coal gas, it emits no sulfur pollutants and as much as 60%-65% less carbon dioxide (a greenhouse gas) than a conventional coal-burning plant.

It is also one of the most efficient means for generating electricity and usable heat. As simple cycle power generator, it can convert more than 55% of the energy in its fuel source to electricity (conventional coal plants, for example, operate at efficiencies of 33%-35%). When the waste heat from the electrochemical process is used, overall efficiencies could reach 85%. When utilized with a gas turbine in a combined power system, efficiencies over 70% can be achieved.

Because they involve no liquid or moving parts, solid oxide modules are expected to operate reliably for many years. A unit that can generate 2 megawatts of electricity, enough for a small substation, can fit on under one-tenth of an acre, allowing it to be placed close to power needs, avoiding long transmission lines.

With a simple adjustment of air and fuel flows — much as a gas pedal is used in a car — a solid oxide fuel cell can easily follow changing demands for electricity, boosting output when necessary, then cycling down when demand is low.

The all-solid-state composition of these fuel cells promises to bring to the electric power sector the mass-production processes that have reduced the costs in the electronics industry.

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