
Advances in Solid Oxide Fuel Cells

*A collection of papers presented at the
29th International Conference
on Advanced Ceramics and Composites,
January 23-28, 2005,
Cocoa Beach, Florida*

Editor
Narottam P. Bansal

General Editors
Dongming Zhu
Waltraud M. Kriven



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Preface

The second international symposium "Solid Oxide Fuel Cells: Materials and Technology" was held during the 29th International Conference on Advanced Ceramics and Composites in Cocoa Beach, FL, January 23-28, 2005. This symposium provided an international forum for scientists, engineers, and technologists to discuss and exchange state-of-the-art ideas, information, and technology on various aspects of solid oxide fuel cells. A total of 88 papers, including three plenary lectures and seven invited talks, were presented in the form of oral and poster presentations indicating strong interest in the scientifically and technologically important field of solid oxide fuel cells. Authors from 16 countries (Canada, China, Denmark, England, France, Germany, India, Italy, Japan, Russia, South Korea, Spain, Switzerland, Taiwan, Ukraine, and U.S.A.) participated. The speakers represented universities, industries, and government research laboratories.

These proceedings contain contributions on various aspects of solid oxide fuel cells that were discussed at the symposium. Thirty six papers describing the current status of solid oxide fuel cells technology and the latest developments in the areas of processing, fabrication, manufacturing, characterization, testing, performance analysis, long term stability, anodes, cathodes, electrolytes, interconnects, sealing materials and design, interface reactions, mechanical properties, fuel reforming, etc. are included in this volume. Each manuscript was peer-reviewed using The American Ceramic Society review process.

The editor wishes to extend his gratitude and appreciation to all the authors for their cooperation and contributions, to all the participants and session chairs for their time and efforts, and to all the reviewers for their useful comments and suggestions. Financial support from The American Ceramic Society is gratefully acknowledged. Thanks are due to the staff of the meetings and publications departments of The American Ceramic Society for their invaluable assistance. Advice, help and cooperation of the members of the symposium's international organizing committee (Tatsumi Ishihara, Tatsuya Kawada, Nguyen Minh, Mogens Mogensen, Nigel Sammes, Prabhakar Singh, Robert Steinberger-Wilkens, Jeffrey Stevenson, and Wayne Surdoyal) at various stages were instrumental in making this symposium a great success.

It is our earnest hope that this volume will serve as a valuable reference for the engineers, scientists, and other technical people interested in different aspects of materials, science and technology of solid oxide fuel cells.

Narottam P. Bansal

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Overview and Current Status

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WORLDWIDE SOFC TECHNOLOGY OVERVIEW AND BENCHMARK *

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ABSTRACT

Solid Oxide Fuel Cells (SOFC) are generally considered a promising future electricity generation technology due to their high electrical efficiency. They also display a multi-fuel capability (hydrogen, carbon monoxide, methane etc.), may play a role in carbon sequestration strategies and render the highest electricity generation efficiency in power station design if coupled with a gas turbine. Still, their development is faced with various problems of high temperature materials, design of cost effective materials and manufacturing processes and efficient plant design.

This paper will summarize the world wide efforts in the field of SOFC, presenting an overview of the main existing SOFC designs and the main developers active in this field. Based on data published in proceedings of international conferences during the last years, a comparison is made of the results achieved in cell, stack and system development.

INTRODUCTION

Within the last ten years, SOFC development has made big progress which can clearly be seen from the tenfold increase in power density. A declining interest in SOFC could be observed towards the end of the last century, when several of the leading companies terminated their activities, amongst them Dornier in Germany and Fuji Electric in Japan. Nevertheless a tremendous increase in activities occurred during the last years with companies re-starting their activities and new industry and research institutions starting SOFC-related work.

This report tries to give an overview of the main development lines and summarizes the development status, reached at the end of 2004, by presenting a comparison of obtained results in cell, stack and system development. The authors concentrate on the published results of industry and the larger research centers. The numerous activities at universities are not taken into account in order to facilitate the overview. In the following chapters the various design variants are presented, followed by a description of the main companies involved and by the status of cell, stack and system technology.

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DESIGN CONCEPTS

There are two main concepts under development – the tubular and the planar design. As far as proof of long term stability and demonstration of plant technology are concerned, the tubular concept is far more advanced, while the planar design offers higher power density.

Tubular Concepts

The most advanced tubular SOFC is being developed by Siemens Westinghouse Power Corporation (SWPC). Their concept is based on a porous cathode tube, manufactured by extrusion and sintering. The tube length is 1.8 m with a wall thickness of 2 mm and an outer diameter of 22 mm (see figure 1). The active length is 1.5 m, which is coated by atmospheric plasma spraying first with a ceramic interconnect, then with zirconia electrolyte originally deposited by EVD (Electrochemical Vapor Deposition) and with an Ni-YSZ anode¹. The cells are connected to bundles via nickel felts. The high ohmic resistance of this concept requires an operating temperature between 900 and 1000°C to reach power densities of about 200 mW/cm². To overcome this problem SWPC is working on a modified concept, using flattened tubes with internal ribs for reduced resistance (“High Power Density” (HPD) tubes, see also figure 1). A similar design, but anode-supported, is being developed by Kyocera²².

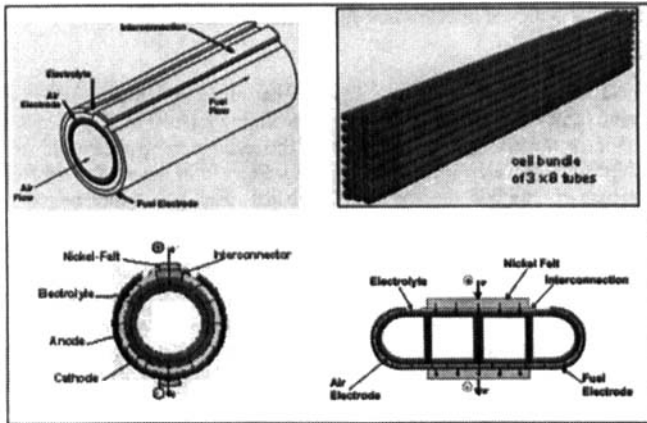


Figure 1: SWPC – tube design, cell bundle and flattened tube¹

The Japanese company TOTO uses the standard tubular design, but started earlier with the implementation of cheaper manufacturing technologies². They chose shorter tubes of 0.5 m length with an outer diameter of 16 mm. The US company Acumentrics is developing anode supported tubes with a length of 45 cm with an outer diameter of 15 mm¹⁷. A different tubular design is pursued by Mitsubishi Heavy Industries (MHI/Japan). The single cells are positioned on a central porous support tube and connected electrically in series via ceramic interconnector rings, which leads to an increased voltage at the terminals of a single tube. Fuel is supplied to the inside of the tube and air to the outside^{2,3}.

Planar Concepts

Within the planar concepts, the electrolyte supported concept and the electrode supported concept have to be distinguished. The former generally uses an electrolyte made of yttrium stabilized zirconia (YSZ) with a thickness of about 100 to 200 μm and an area of $10 \times 10 \text{ cm}^2$ (sometimes larger) as the supporting part of the cell. Typical operating temperatures of this concept are 850 to 1000°C due to the relative high ohmic resistance of the thick electrolyte. In case of operation at very high temperatures, ceramic interconnects made of lanthanum-chromite have been used. Since these ceramic plates are restricted in size, require high sintering temperatures, have different thermal expansion behavior in oxidizing and in reducing atmosphere and have comparatively bad electrical and thermal conductivity, there is an obvious trend to metallic interconnect plates. The advantage of ceramic plates is the negligible corrosion and therefore low degradation which sustains the interest in this material. The metallic interconnect plates allow on one hand the reduction of operating temperature and on the other hand an increase in size. The good thermal conductivity reduces the temperature gradients in the stack and allows larger temperature differences between gas inlet and outlet, which reduces the necessary air flow for cooling. Since the thermal expansion coefficient of conventional high temperature alloys is significantly higher compared to zirconia, a special alloy was developed (chromium with 5% iron and 1% yttria) by the Austrian company Plansee, which was used by Siemens and is still being used by Sulzer Hexis. In the Sulzer Hexis design, fuel is supplied to the centre of the electrolyte supported circular cells (having a diameter of 120 mm) and flows to the outer rim of the cell, where the fuel gas, which has not reacted within the cell, is burned. Air is supplied from the outside and heats up, while flowing towards the centre (two-layer interconnect design, see figure 2). The stack is typically operated at 950°C. Up to 70 cells are stacked together, delivering 1.1 kW⁴. Recently, Sulzer changed the design to a single plate concept in order to reduce manufacturing costs.

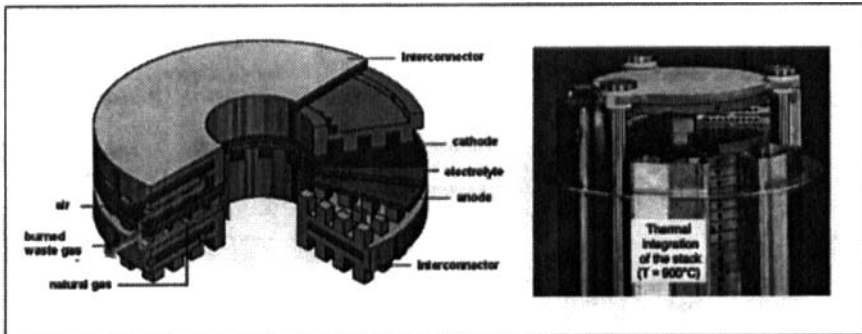


Figure 2: Sulzer Hexis Cell and Stack⁴

A joint development between Mitsubishi Heavy Industries (MHI) and Chubu Electric Power Company is the so called MOLB-Type (Mono-block Layer Built) planar SOFC. The cells are manufactured up to a size of 200 x 200 mm², based on a corrugated electrolyte layer. In this way the electrolyte also contains the gas channels, which simplifies the design of the interconnects, for which planar ceramic plates are used (see figure 3). The biggest stack of this type was built of 40 layers, delivering 2.5 kW at 1000°C.⁵

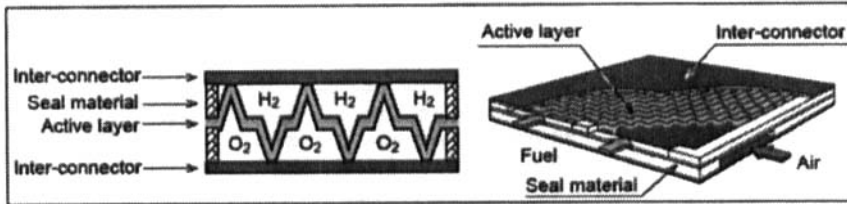


Figure 3: MHI and Chubu - MOLB Design^{3,5}

Since the electrolyte resistance is the most important obstacle on the way to further reducing the operating temperature the production of thinner electrolytes is a major challenge. This can be done by shifting the function of mechanical stabilization from the electrolyte to one of the electrodes. In this approach the anode is mostly favored, because it generically has a good electrical conductivity. Therefore no increase in ohmic resistance is incurred by increasing the electrode thickness. Also, the nickel cermet has a good mechanical stability, which allows the manufacturing of larger components than with ceramic electrolyte substrates (see figure 4).

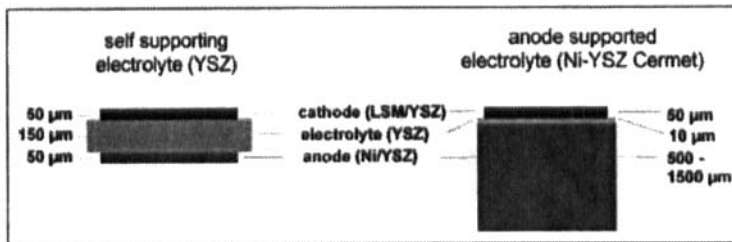


Figure 4: Anode supported cell concept (right) compared to electrolyte supported (left)

As one of the first institutions, this development was started in 1993 at Forschungszentrum Jülich and in the meantime is favored by many developers throughout the world as the 'next generation' of SOFC. This concept allows reducing the operating temperature down to the range of 700 to 800°C whilst retaining the same power density as electrolyte supported cells at 950°C. At the same time this design allows the use of ferritic chromium alloys for interconnects, because its thermal expansion coefficient corresponds to that of the anode substrate.

At Forschungszentrum Jülich, anode substrates are manufactured by warm pressing with a thickness of 1 to 1.5 mm on which an electrolyte made by vacuum slip casting with a thickness of 5 to 10 μm is applied. The stack design is based on a co- or counter- flow arrangement. The latter is favored in case of natural gas operation with internal reforming. Figure 5 shows the stack design and a 60 layer stack delivering 11.9 kW at 800°C with methane and internal reforming. ¹²

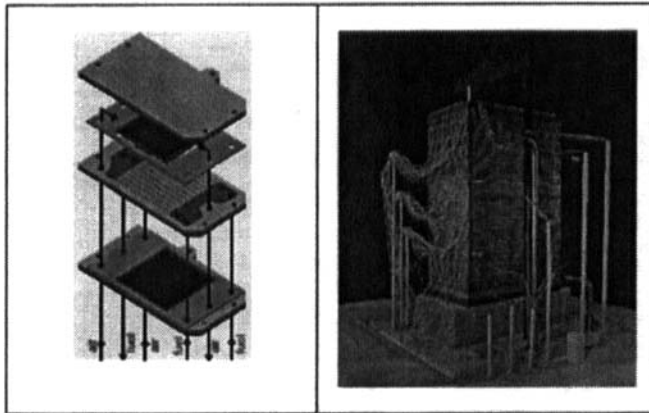


Figure 5: FZJ - Stack design and 10 kW stack ¹²

Similar concepts are pursued for instance by Global Thermoelectric, Delphi/PNNL and Haldor Topsø/Risø National Laboratory. ECN and its spin-off company InDEC (Innovative Dutch Electrochemical Cells, now part of H.C. Starck) manufacture electrolyte supported cells as well as anode supported cells.

Other institutions, like the DLR in Stuttgart, have developed concepts using pure metal substrates instead of the anode cermet to improve mechanical and redox stability. Up to now they have realized stacks with three to four layers.

A completely different design has been developed by Rolls Royce. Short electrode and electrolyte stripes are applied onto a porous ceramic substrate, which functions as mechanical supporting element. The single cells are connected electrically in series using short stripes of ceramic interconnects and are operated at about 950°C ⁶. Rolls Royce is currently working on the realization of multiple kW stack units. Kyocera has started working on a similar concept ²³.

IMPORTANT DEVELOPERS

At the end of the nineties, some of the most important developers in Europe, Daimler-Benz/Dornier and Siemens, terminated their activities in planar SOFC. After an interim phase, the number of companies engaged in SOFC development has again increased.

The following tables list the most important developers.

Table I: Developers in Europe

Country	Institution/company	Concept	Main focus in development
Denmark	Haldor Topsoe together with Risoe	planar: until 1999: ceramic IC, electrolyte substrate since 2000: metallic IC, anode substrate	system, reformer materials, cells, stack
Finland	VTT	—	fuel conditioning, cell and stack testing, modeling
	Wärtsilä	—	system
Germany	BMW	planar, metallic IC, metallic substrate	stack, system
	DLR-Stuttgart	planar, metallic IC, metallic substrate	materials, cells, stack
	FZJ	planar, metallic IC, anode substrate	materials, cells, stack, system, modeling
	H.C. Starck/Indec (NL)		powders, cell manufacturing
	IKTS-Dresden	planar, metallic IC, electrolyte substrate	stack
	Siemens	tubular, "flat tube"	materials, manufacturing
	Webasto	planar, metallic IC, electrolyte substrate	stack, system
France	EDF/GDF	—	fuel conditioning, testing
	CEA; Fuel Cell Network	—	materials
Great Britain	Ceres Power	planar, CGO electrolyte for 550°C, metallic IC, metallic substrate	materials, cell, stack, system
	Rolls Royce	planar, on porous ceramic substrate	materials, cells, stack, system
Netherlands	ECN	—	materials, cells, stack,
Switzerland	Sulzer Hexis	planar, metallic IC, electrolyte substrate anode substrate	materials, cells, stack, system
Europe		total employees (ca.)	450 - 500

Table II: Developers in North America

Country	Institution/ company	Concept	Main focus in development
USA	Acumentrics	tubular (anode substrate)	cells, stack, system
	ANL (Argonne National Lab.)	planar, metallic IC, electrolyte substrate	materials, cells, modeling
	Cummings/SOFCo	planar, ceramic IC, electrolyte substrate	materials, cells, stack, system
	Delphi Automotive Systems (collaboration with PNNL)	planar, metallic IC, anode substrate	cells, stack, system/APU
	GE (former Honeywell former Allied Signal)	planar, metallic IC, anode substrate	materials, cells, stack, reformer,
	LLNL (Laurence Livermore National Lab.)	planar, metallic IC, anode substrate	cells, stack
	NETL (National Energy Technology Lab.)		
	PNNL (Pacific Northwest National Lab.)	planar, metallic IC, anode substrate	materials, cells, modeling
	SWPC	tubular (cathode substrate); "flat tube"	materials, cells, system
	TMI (Techn. Managem. Ing.)	planar	cells, Stack, System
ZTek	planar, metallic IC	cells, stack, system	
Canada	Global Thermoelectric (now part of Versa Power)	planar, metallic IC, anode substrate	materials, cells, stack, system
	FCT (Fuel Cell Technology) together with SWPC	—	system
North America		total employees (ca.)	450 - 500

Table III: Developers in Asia and Australia

Country	Institution/company	Concept	Main focus in development
Japan	Kyocera with Tokyo Gas and Osaka Gas	cylindrical planar flat tubular (anode substrate) "horizontal pattern"	materials, cells, stack, system
	Mitsubishi Heavy Industries (MHI) with Chubu EPCo (CEPCo)	MOLB Design: planar, ceramic IC, electrolyte substrate	materials, cells, stack, system
	Mitsubishi Heavy Industries (MHI) with EPDC	tubular (porous support tube, serial connection)	materials, cells, stack, system
	Mitsubishi Materials (MMC) with Kyushu EPCo (KEPCo)	Gallate electrolyte, 800°C planar, metallic IC, electrolyte substrate	materials, cells, stack, system
	Nihon Gaiishi (NGK)	planar, anode substrate	materials, cells
	Nippon Shukubai	planar, electrolyte substrate	materials, cells
	Toho Gas	planar, metallic IC, electrolyte substrate	materials, cells, stack, system
	TOTO with Kyushu EPCo (KEPCo)	tubular (cathode substrate)	materials, cells, stack, system
	Tokyo Gas	planar, metallic IC, anode substrate	materials, cells, stack, system
Korea	KIER (Korean Institute of Energy research)	anode supported flat tube	stack, system (pressurized)
Australia	CFCL	Planar, electrolyte substrate, since 2001 shift to ceramic IC	materials, cells, stack, system
Asia, Australia		total employees (ca.)	350 - 400

In the last two decades of the last century, Westinghouse (now Siemens Westinghouse Power Corporation SWPC) dominated the development in the USA. Since the "SECA" – program was started, the situation has completely changed, and several consortia have been formed and activities re-started in the field of planar SOFC.

In Japan in the nineties, more than 10 companies were involved in planar SOFC development. After the goals of the "Shunshine" project of Nedo could not be achieved completely, a re-orientation took place with additional companies starting development.

DEVELOPMENT STATUS

Cells

In the field of cell development, many activities are ongoing. Therefore, it is quite difficult to compile overview data, especially if they are supposed to be based on comparable operating conditions. In figure 6, this has been attempted for different types of cells at 0.7V cell voltage, (with the most common cathode materials indicated): anode supported cells at 750°C operating temperature, electrolyte supported cells at 800 to 900°C and tubular cells at 900 to 1000°C. Although direct comparison is difficult because of differing operating conditions and fuel gases, it is obvious that the highest power densities are achieved using anode supported cells, preferably with LSF (lanthanum strontium ferrite) cathodes.

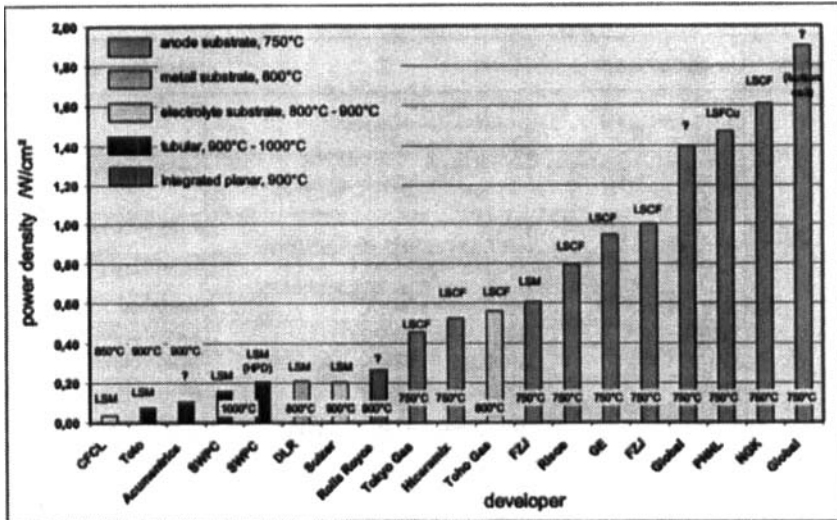


Figure 6: Power density at 0.7 V for different types of cells at the relevant operating temperature

Besides the power density, the producible cell size is an important feature in characterizing the potential of the technology. Achieved values of the active electrode area are given in figure 7.

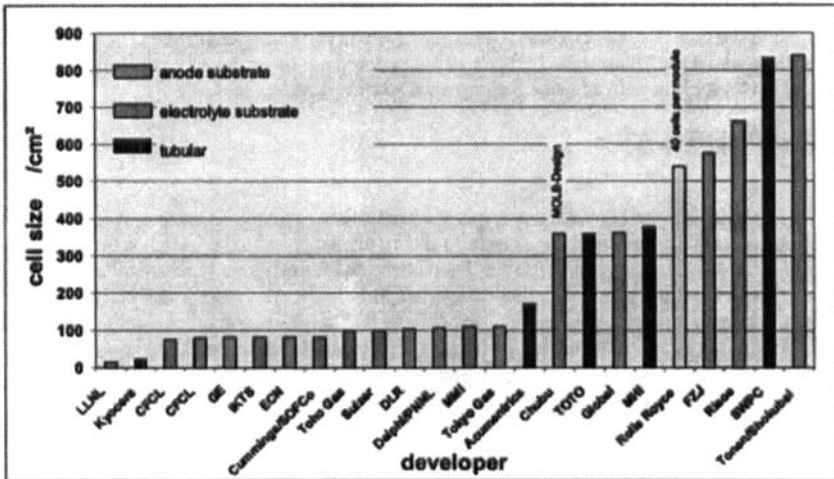


Figure 7: Maximum active cell size manufactured by various companies⁶⁻¹⁵

Meanwhile, the degradation rates of planar cells are approaching the same range as the tubular cells of SWPC. At the same time, the demonstrated operation times have clearly increased. Both properties are depicted in Figure 8.

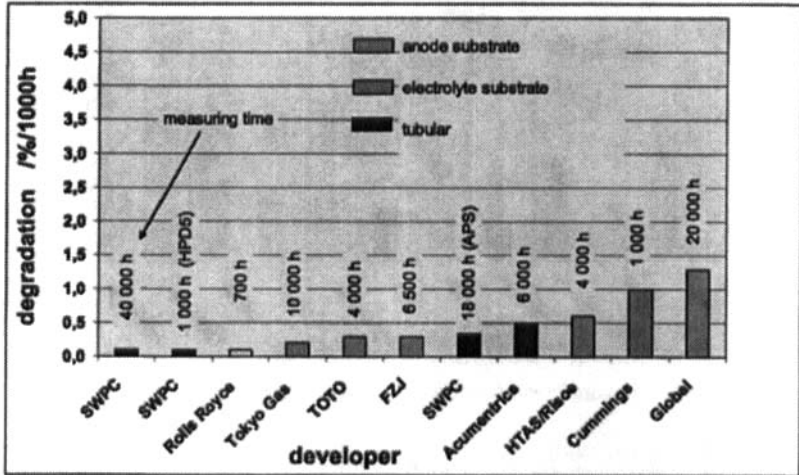


Figure 8: SOFC cells - degradation rates in inert environment (except for Global, who measured in metal housings) ^{4, 9, 11, 14, 15, 17}

Stacks

Compared to the situation a few years ago, there are many more developers with proprietary stack technology. Some of them have changed the design in recent years, restarting developments at lower power. The achieved long term stability of stacks with at least two cells containing all relevant materials is shown in Figure 9, and the maximum power output achieved is shown in Figure 10.

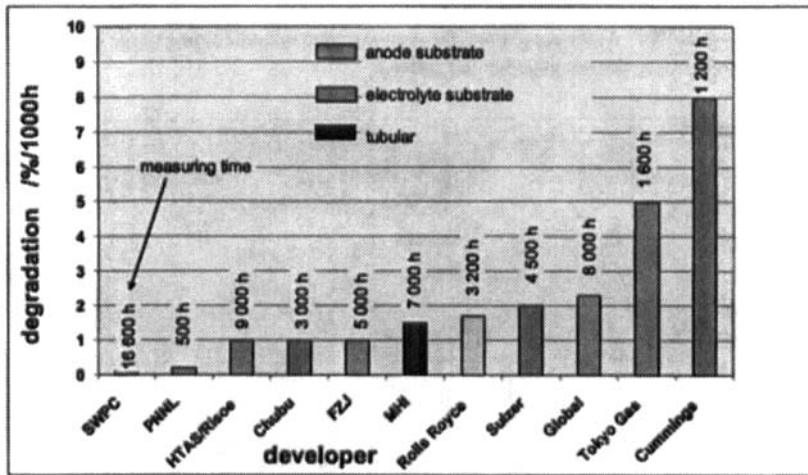


Figure 9: SOFC stacks - degradation rates ^{1, 5, 6, 9, 15 - 19}

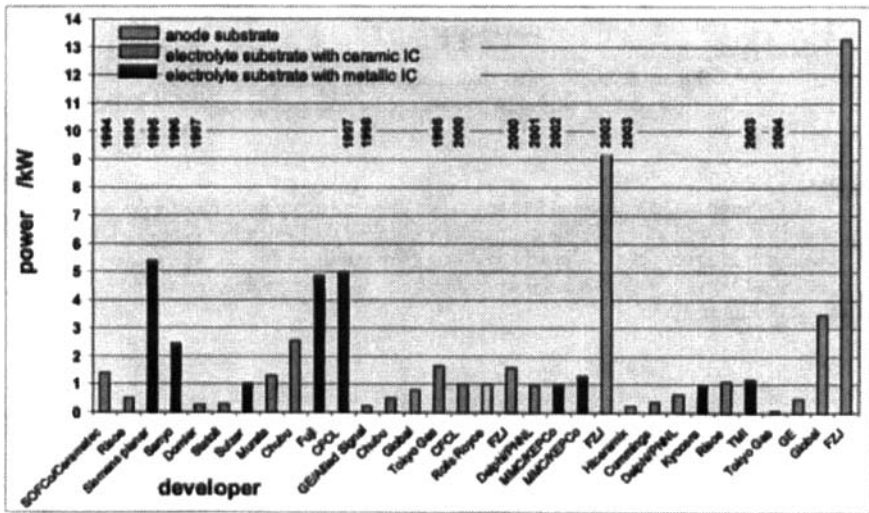


Figure 10: SOFC stacks - maximum power ^{6 - 15, 17, 18}

System

During the last two years, quite a lot of companies claimed to have established and tested complete SOFC systems. Most of them are first lab-test systems; however, this demonstrates the impressive progress that has been made during the last years. Most experience is available at Sulzer Hexis (small residential systems) and Siemens Westinghouse (medium sized CHP plants). However, field tests have shown that there is still a lot of improvement necessary to approach reliable and cost competitive systems. This is even more the case for the other developers, as can be seen in Table IV. So a lot of work still has to be done to reach a reliable status, which can be called "prototype" or "pre-commercial".

Table IV: Status of system development and testing (ranked by system power)

company	achieved power [kW]	power density [W/cm ²]	number of stacks/cells	temperature [°C]	operation time [hours]	year	total No. of systems (also smaller)	Ref.
Cummings			2 / ?			2004	1	12
CFCL	1,00	0,105	1 / 140	900	1900	2004	2	16
Sulzer Hexis	1,00	0,180	1 / 50	950	4500	2004	~130	19
Toho Gas	1,03	0,152	1 / 68	800		2004	1	23
TMI	1,20					2003	1	11
MMC/KEPCo	1,25	0,220	1 / ?	770		2004	1	16
Kyocera	1,50	0,300	4 / 50	780	210	2003	1	22
Delphi/PNNL	1,60		2 / 30	750		2004	4	17, 18
Global (Versa Power Systems)	2,30	0,320	1 / 80	750	1100	2004	5	17, 18
MMC/KEPCo	2,97	0,214	3 / 41	780	2300	2004	1	16
TOTO	3,20	0,077		1000		2002	1	9
FCT/SWPC	4,36	0,120	88 tubes with 75 cm	950	4550	2004	4	18
Acumentrics	5 kW class						>5	17
Chubu	15,00	0,240	30 / 10; no complete system	1000	7500	2000	1	9
MHI	21,00	0,180	no complete system	900			1	5
SWPC	110,00	0,130	1 module / 1152	950	20400	2001	12	17
	191,00	0,120	2 modules / 1152	950	1100	2003		

SUMMARY AND OUTLOOK

As far as the development status of system technology and long term stability are concerned, the tubular design of SWPC still plays a leading role in the SOFC field. Nevertheless, most developers today see a clear advantage in the cost reduction potential of the planar technology. This is on one hand due to the more cost efficient manufacturing technologies and on the other hand due to the higher power density. In this relation, there is a clear trend towards anode supported design, using ferritic chromium steel as interconnect material. Besides increased power density, this concept also provides the chance of reducing the operating temperature below 800°C. Although the development status of the planar design is clearly behind the tubular,

considerable progress could be achieved during the recent years. A consolidation of activities can be observed, especially in the USA, driven by the SECA program and also in Japan by increased engagement of industry. A great push was created by the envisaged application as APU (auxiliary power unit), especially in Germany and in the USA. In the stationary field, the main focus is on small units in the kW range for residential energy supply and up to several 10 kW for small to medium sized CHP applications.

REFERENCES

- ¹Kabs, H.; "Advanced SOFC Technology and its Realization at Siemens Westinghouse", *Bilateral Seminars 33, Materials and Processes for Advanced Technology: Materials for Energy Systems, Egyptian-German Workshop, Cairo 7.-9. April 2002*, ISBN 3-89336-320-3, edited by D.Stöver, M.Bram, 91-101 (2002)
- ²Fujii, H.; "Status of National Project for SOFC Development in Japan", *Solid Oxide Fuel Cells Meeting*, 18. November 2002, Palm Springs, USA, (2002)
- ³Fujii, H.; Ninomiya, T.; "Status of National Project for SOFC Development in Japan", *European Solid Oxide Fuel Cell Forum, Vol. 2, Lucerne, Switzerland 1.-5. Juli 2002, Proceedings*, edited by J. Huijsmans, 700-707 (2002),
- ⁴Schmidt, M.; "The Hexis Project: Decentralised electricity generation with waste heat utilisation in the household", *Fuel Cells Bulletin 1, No 1*, 9-11 (1998)
- ⁵Nakanishi, A.; Hattori, M.; Sakaki, Y.; Miyamoto, H.; Aiki, H.; Takenobu, K.; Nishiura, M.; "Development of MOLB Type SOFC", *Fifth European SOFC Forum, Vol. 2, Lucerne, Switzerland 1.-5. Juli 2002, Proceedings*, edited by J. Huijsmans, 708-715 (2002),
- ⁶Gardner, F. J. et al, SOFC Technology Development at Rolls Royce, *Journal of Power Sources* 86, 122 – 129 (2000)
- ⁷6th Int. Symposium Solid Oxide Fuel Cells (SOFC VI), The Electrochemical Society, Pennington, NJ, USA, *Proceedings* (1999)
- ⁸7th Int. Symposium Solid Oxide Fuel Cells (SOFC VII), The Electrochemical Society, Pennington, NJ, USA, *Proceedings* (2001)
- ⁹8th Int. Symposium Solid Oxide Fuel Cells (SOFC VIII), The Electrochemical Society, Pennington, NJ, USA, *Proceedings*, (2003)
- ¹⁰Fuel Cell Seminar, Portland, USA, *Proceedings* (2000)
- ¹¹Fuel Cell Seminar, San Diego, USA, *Proceedings*, (2003)
- ¹²Fuel Cell Seminar, San Antonio, USA, *Proceedings*, (2004)
- ¹³Fourth European SOFC Forum, Lucerne, Switzerland, *Proceedings*, (2000)
- ¹⁴Fifth European SOFC Forum, Lucerne, Switzerland, *Proceedings*, (2002)
- ¹⁵Sixth European SOFC Forum, Lucerne, Switzerland, *Proceedings*, (2004)
- ¹⁶The Fuel Cell World, Lucerne, Switzerland, edited by U. Bossel, *Proceedings*, (2004)
- ¹⁷US DOE, 2004 Office of Fossil Energy Fuel Cell Program Annual Report, (2004)
- ¹⁸International Energy Agency, Annex XVIII (SOFC), San Antonio, USA, (2004)
- ¹⁹f-cell , Stuttgart, Germany, *Proceedings* (2004)
- ²⁰www.ngk.co.jp/infor/develop/topics4/index.html
- ²¹www.shokubai.co.jp/main/06kaihat/sofc/sofc_top.html
- ²²<http://global.kyocera.com/news/2003/1205.html>
- ²³www.tohogas.co.jp

U.S. DOE SOLID OXIDE FUEL CELLS: TECHNICAL ADVANCES*

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ABSTRACT

The U.S. Department of Energy's (DOE) Office of Fossil Energy's (FE) National Energy Technology Laboratory (NETL), in partnership with private industries, is leading the development and demonstration of high efficiency solid oxide fuel cells (SOFCs) and fuel cell turbine (FCT) hybrid power generation systems for distributed generation (DG) markets. The DOE FE DG program has three aspects: the Solid State Energy Conversion Alliance (SECA), Central Power Systems and the High Temperature Electrochemistry Center (HiTEC). NETL is partnering with Pacific Northwest National Laboratory (PNNL) in developing new directions in research under SECA for the development and commercialization of modular, low cost, and fuel flexible SOFC systems. The SECA initiative, through advanced materials, processing and system integration research and development (R&D) will bring the fuel cell cost to \$400 per kilowatt (kW) by 2010 for stationary and auxiliary power unit (APU) markets. The SECA program is currently structured to include six competing industry teams supported by a crosscutting core technology program (CTP). DOE is ultimately concerned with coal-based central power plants. Advanced aspects of solid oxide technology are part of HiTEC R&D.

INTRODUCTION

SOFCs use a ceramic electrolyte that results in a solid state unit, an important aspect. The conduction mechanism is solid state conduction of O^{2-} ions. The reaction is completed by the reaction of oxygen ions and hydrogen to form water. SOFCs can extract hydrogen from a variety of fuels using either an internal or external reformer. They are also less prone to CO poisoning than other fuel cells and thus are attractive for coal-based fuels. SOFCs work well with catalysts made of nickel, which is much less expensive than platinum. SOFCs can achieve efficiencies of 60 percent stand-alone, or over 80 percent (net) if the waste heat is used for cogeneration. Currently, demonstration units exist up to 2 megawatts (MW). Challenges with SOFCs are development of high power density, reducing cost, and better seals and metallic interconnects.¹

For SOFCs, conventional fuels can be used now, and hydrogen can be used in the future. Like all fuel cells, SOFCs will operate even better on hydrogen than conventional fuels. Therefore, the commercialization path for fuel cells is through portable and stationary markets using today's conventional fuels and then transportation markets using hydrogen. Each market offers progressively lower cost potential.^{2,3}

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In the U.S., we let the market make eventual choice among technology alternatives. Less expensive materials, simple stack and system design, and high volume markets are the three criteria that must be met by a fuel cell system to compete in today's energy market. These criteria form the basis for SECA's common sense goal of lowering fuel cell costs.

High temperature SOFCs have high electricity conversion, superior environmental performance, combined heat and power, fuel flexibility, size and siting flexibility, and transportation and stationary applications attributes as noted in Table I. These attributes hold promise for worldwide stationary industrial and residential power generation, APUs in trucks and cars, and a range of military applications.

Table I. Solid Oxide Fuel Cells – Attributes	
High electric conversion efficiency	<ul style="list-style-type: none"> • Demonstrated - 47% • Achievable - 55% • Hybrid – 65% • CHP – 80%
Superior environmental performance	<ul style="list-style-type: none"> • No NO_x • Lower CO₂ emissions • Sequestration capable • Quiet; no vibrations
Cogeneration – combined heat and power (CHP)	<ul style="list-style-type: none"> • High quality exhaust heat for heating, cooling, hybrid power generation, and industrial use • Co-production of hydrogen with electricity • Compatible with steam turbine, gas turbine, renewable technologies, and other heat engines for increased efficiency
Fuel flexibility	<ul style="list-style-type: none"> • Low or high purity H₂ • Liquefied natural gas • Pipeline natural gas • Diesel • Coal gas • Fuel oil • Gasoline • Biogases
Size and siting flexibility	<ul style="list-style-type: none"> • Modularity permits wide range of system sizes • Rapid siting for distributed power
Transportation and stationary applications	<ul style="list-style-type: none"> • Watts to megawatts

This paper begins with a discussion of the U.S. DOE role in SOFC R&D under SECA, the hurdles it seeks to accomplish, recent advances and funding levels. Central power concepts including FutureGen with hydrogen are detailed in the next sections. Then other technologies that are also impacted by solid oxide R&D are discussed under HiTEC.

SOLID-STATE ENERGY CONVERSION ALLIANCE (SECA) PROGRAM

The SECA Program is the main thrust of the DOE FE DG Fuel Cell Program. It is dedicated to developing innovative, effective, low-cost ways to commercialize SOFCs. The

program is designed to move fuel cells out of limited niche markets into widespread market applications by making them available at a cost of \$400/kW or less through the mass customization of common modules by the year 2010. SECA fuel cells will operate on today's conventional fuels such as natural gas, diesel, as well as coal gas and hydrogen, the fuel of tomorrow. The program will provide a bridge to the hydrogen economy beginning with the introduction of SECA fuel cells for stationary (both central generation and DG) and APU applications. ^{4, 5, 6, 7, 8, 9, 10}

The SECA program is currently structured to include competing industrial teams supported by a crosscutting core technology program (CTP). SECA has six industrial teams, Cummins-SOFCo, Delphi Battelle, General Electric (GE), Siemens Westinghouse (SW), Acumentrics, and FuelCell Energy (FCE), working on designs and manufacturing as shown in Table II that can be mass-produced at costs that are ten-fold less than current costs. Figure 1 shows planar configurations.

Table II. SECA Industrial Team Design & Manufacturing		
Team	Design	Manufacturing
Cummins-SOFCo	Electrolyte supported-planar 825° C Thermally matched materials Seal-less stack	Tape casting Screen printing Co-sintering
Delphi-Battelle	Anode supported-planar 750° C Ultra compact Rapid transient capability	Tape casting Screen printing 2-stage sintering
General Electric Company	Anode supported-radial 750° C Hybrid compatible Internal reforming	Tape calendaring 2-stage sintering
Siemens Westinghouse Power Corp.	Cathode supported-flattened oval 800° C Seal-less stack	Extrusion Plasma spray
Acumentrics Corporation	Anode supported-microtubular 750° C Thermally matched materials Robust & rapid start-up	Extrusion Dip processing Spray deposition Co-sintering
FuelCell Energy, Inc.	Anode supported-planar < 700° C Low cost metals Thermal integration	Tape casting Screen printing Co-sintering Electrostatic deposition

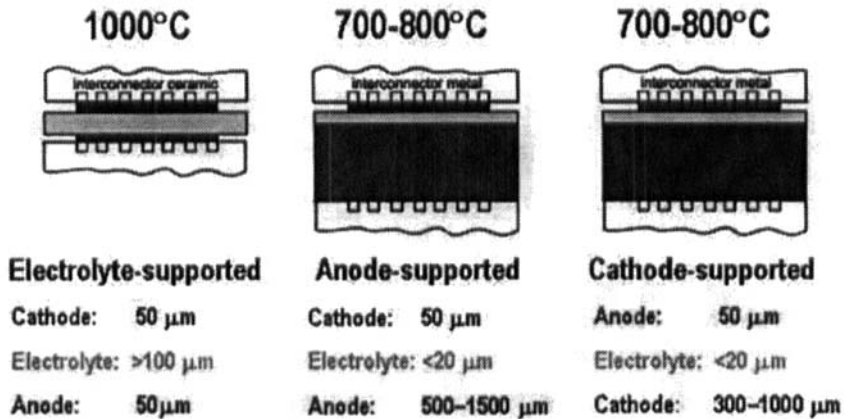


Figure 1. Planar Cell Designs

All of the industry teams made exceptional progress in FY2004 by completing conceptual designs and testing stack configurations showing increased power densities and fuel utilization, control system reference designs, and reformers that demonstrate sustained output and thermal cyclic capability. Acumentrics field-tested a unit that includes some advancements developed under SECA in Alaska on propane in May. SOFCo achieved 2,000 hours of uninterrupted, steady-state operation while fueling a 1 kW SECA unit with natural gas. SW overcame a major fabrication hurdle by applying a doped lanthanum gallate electrolyte material to the cathode using plasma spray technique. This eliminated fabrication imperfections and led to a thinner, more conductive electrolyte layer that could reduce temperature while increasing power for increased lifetime and reduced cost. GE achieved a power density of over 0.4 W/cm² at 0.7 V, while maintaining a fuel utilization of 88 percent. This is over 0.1 W/cm² more than what is mandated by the Phase I requirements, an indicator of progress to come. Cummins demonstrated sustained output and thermal cyclic capability of their reformer process. The reformer operated for 2900 hours at steady state and cyclic operation, corresponding to all Phase I requirements. At the beginning of FY 2005, FCE combined its Canadian SOFC operations, formerly known as Global Thermolectric Corporation, into its lead product development sub-contractor, Versa Power Systems. This consolidation into a single entity provides a greater opportunity to commercialize SOFC products under SECA. Delphi demonstrated its SECA Generation-3 SOFC using fuel gas extracted from coal at the Power Systems Development Facility (PSDF) coal-gasification plant in Wilsonville, Alabama, in July 2004. This is the second test at the PSDF aimed at demonstrating that high efficiency SECA fuel cell technology can successfully use coal gas to produce power cleanly and efficiently. Overall, the SECA Program is progressing extremely well leading up to Phase I prototype testing starting in FY 2005.

Major automotive and truck manufacturers, such as BMW and PACCAR, are collaborating with industry teams for fuel cell business ventures to pursue growth in APUs applications. In fact, BMW has an arrangement with Delphi to put a compact fuel cell APU in its trucks by 2007. The National Aeronautics and Space Administration's (NASA) interest in