Overview and Current Status

Advances in Solid Oxide Fuel Cells
Narottam P. Bansal

WORLDWIDE SOFC TECHNOLOGY OVERVIEW AND BENCHMARK *

Ludger Blum, Wilhelm A. Meulenberg, Heinz Nabielek, Robert Steinberger-Wilckens
Forschungszentrum-Jülich GmbH
52425 Jülich, Germany

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.

*This paper has been published in the *International Journal of Applied Ceramic Technology*, Vol. 2, Is. 5. The *International Journal of Applied Ceramic Technology* is the primary citation of record.

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 anode [1]. 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/cm^2. 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 Kyocera [22].

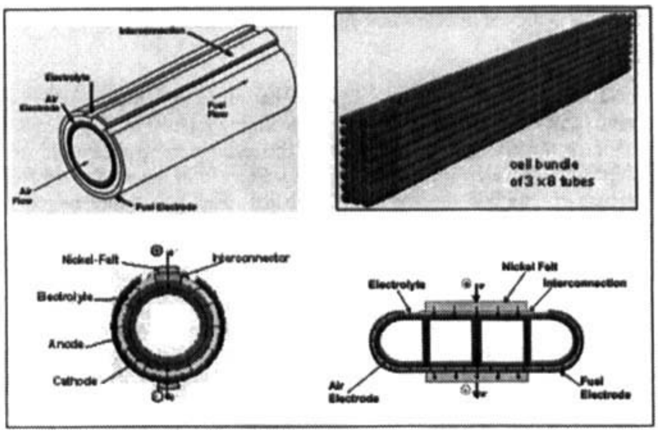

Figure 1: SWPC – tube design, cell bundle and flattened tube [1]

The Japanese company TOTO uses the standard tubular design, but started earlier with the implementation of cheaper manufacturing technologies [2]. 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 mm [17]. 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 x 10 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 kW [4]. Recently, Sulzer changed the design to a single plate concept in order to reduce manufacturing costs.

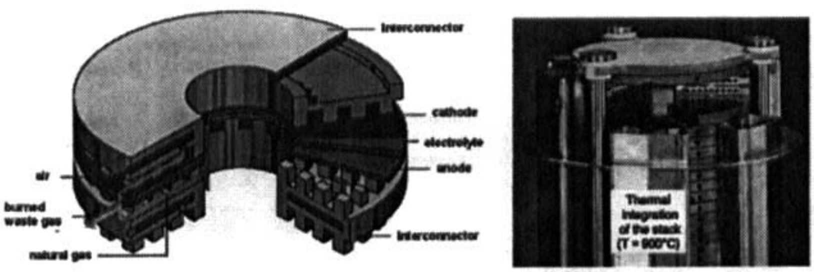

Figure 2: Sulzer Hexis Cell and Stack [4]

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]

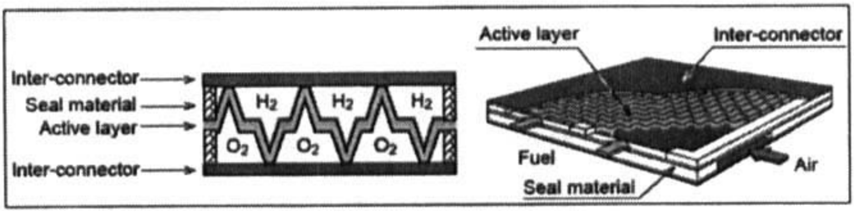

Figure 3: MHI and Chubu - MOLB Design [3, 5]

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**).

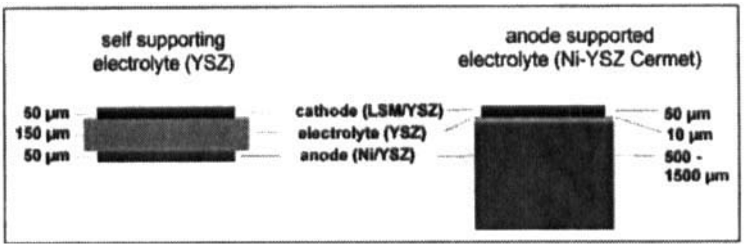

Figure 4: Anode supported cell concept (right) compared to electrolyte supported (left)

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]

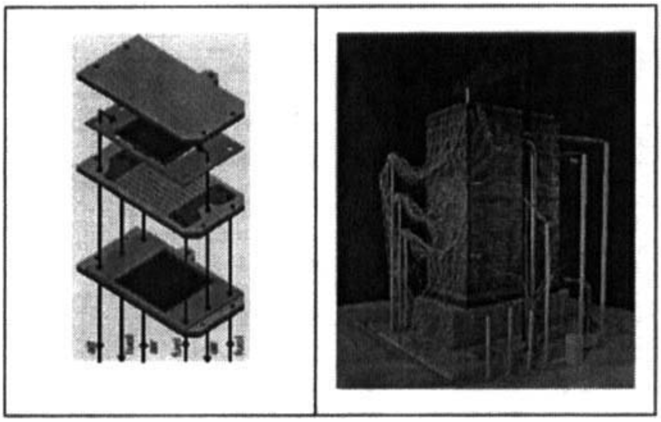

Figure 5: FZJ - Stack design and 10 kW stack [12]

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 [13].

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 Gaishi (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.

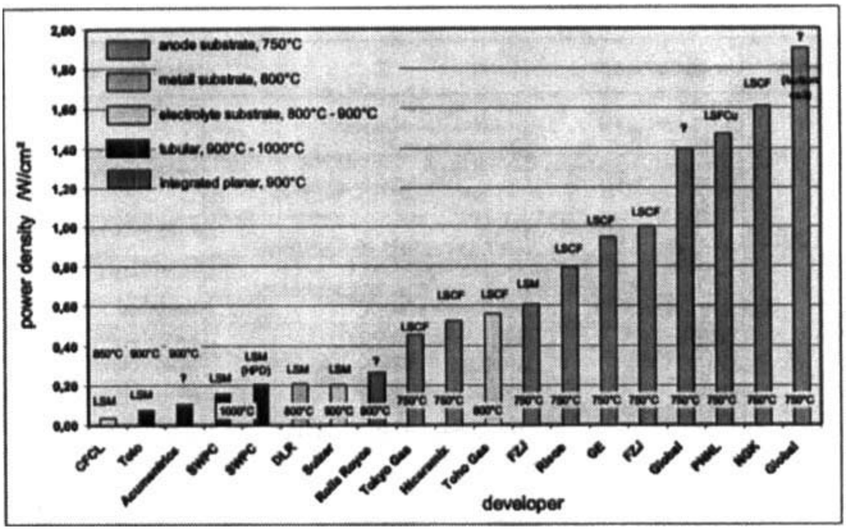

Figure 6: Power density at 0.7 V for different types of cells at the relevant operating temperature

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.

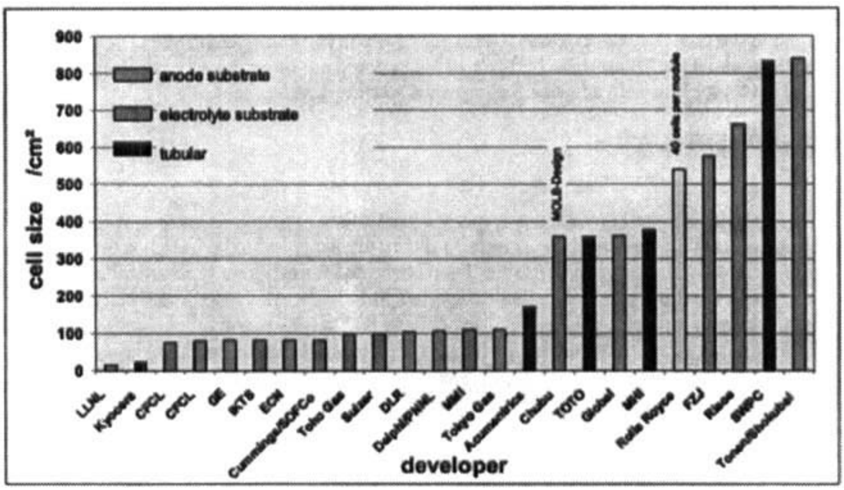

Figure 7: Maximum active cell size manufactured by various companies [6 - 15]

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**.

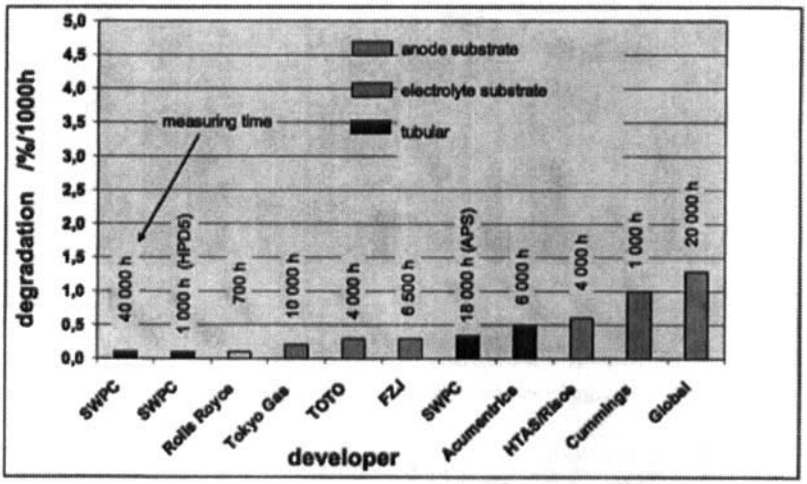

Figure 8: SOFC cells - degradation rates in inert environment
(except for Global, who measured in metal housings) [8, 9, 11, 14, 15, 17]

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**.

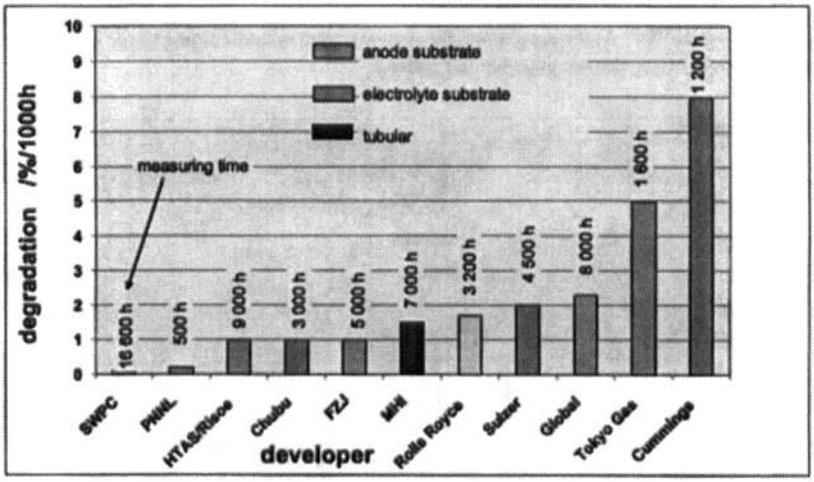

Figure 9: SOFC stacks - degradation rates [1, 5, 6, 9, 15 - 19]

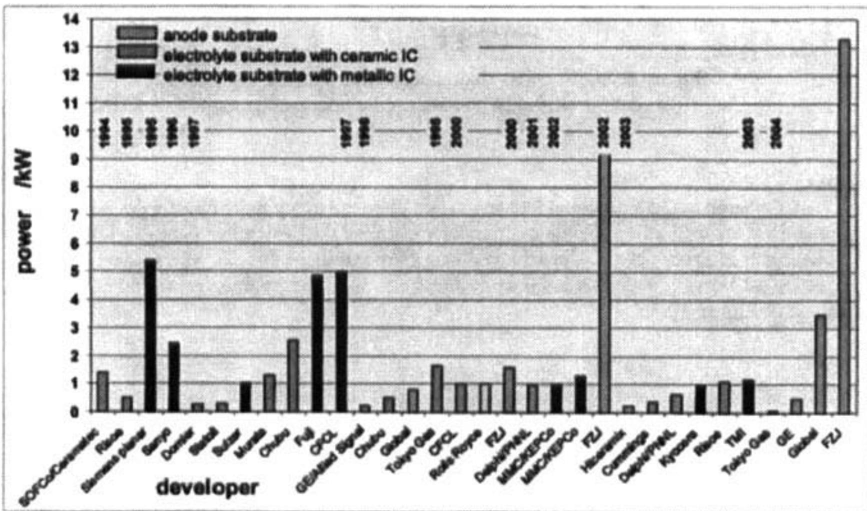

Figure 10: SOFC stacks - maximum power [6 - 15, 17, 18]

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/cm²]	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	/ 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

[1]Kabs, 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)

[2]Fujii, H.;"Status of National Project for SOFC Development in Japan", *Solid Oxide Fuel Cells Meeting,* 18. November 2002, Palm Springs, USA, (2002)

[3]Fujii, 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),

[4]Schmidt, M.; "The Hexis Project: Decentralised electricity generation with waste heat utilisation in the household", *Fuel Cells Bulletin 1, No 1,* 9-11 (1998)

[5]Nakanishi, 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),

[6]Gardner, F. J. et all, SOFC Technology Development at Rolls Royce, *Journal of Power Sources* 86, 122 - 129 (2000)

[7]6th Int. Symposium Solid Oxide Fuel Cells (SOFC VI), The Electrochemical Society, Pennington, NJ, USA, *Proceedings* (1999)

[8]7th Int. Symposium Solid Oxide Fuel Cells (SOFC VII), The Electrochemical Society, Pennington, NJ, USA, *Proceedings* (2001)

[9]8th Int. Symposium Solid Oxide Fuel Cells (SOFC VIII), The Electrochemical Society, Pennington, NJ, USA, *Proceedings,* (2003)

[10]Fuel Cell Seminar, Portland, USA, *Proceedings* (2000)

[11]Fuel Cell Seminar, San Diego, USA, *Proceedings,* (2003)

[12]Fuel Cell Seminar, San Antonio, USA, *Proceedings,* (2004)

[13]Fourth European SOFC Forum, Lucerne, Switzerland, *Proceedings,* (2000)

[14]Fifth European SOFC Forum, Lucerne, Switzerland, *Proceedings,* (2002)

[15]Sixth European SOFC Forum, Lucerne, Switzerland, *Proceedings,* (2004)

[16]The Fuel Cell World, Lucerne, Switzerland, edited by U. Bossel, *Proceedings,* (2004)

[17]US DOE, 2004 Office of Fossil Energy Fuel Cell Program Annual Report, (2004)

[18]International Energy Agency, Annex XVIII (SOFC), San Antonio, USA, (2004)

[19]f-cell , Stuttgart, Germany, *Proceedings* (2004)

[20]www.ngk.co.jp/infor/develop/topics4/index.html

[21]www.shokubai.co.jp/main/06kaihat/sofc/sofc_top.html

[22]http//global.kyocera.com/news/2003/1205.html

[23]www.tohogas.co.jp