MULTI-kW-SOFC DEVELOPMENT AT SIEMENS

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ABSTRACT

The Siemens SOFC development program reached an important milestone early in 1994. A stack operating with hydrogen and oxygen and producing a max. power of 1.8 kW at a current density of 0.8 A/cm² was tested for more than 300h. The SOFC configuration is based on a flat metal separator plate using the multiple cell array design. Improved PENs, functional layer and joining technique were implemented. Based on this concept, the layout of a 100 kW plant was elaborated.

INTRODUCTION

The SOFC development at Siemens has been started in 1990 after a two years preparation phase. The first period with the goal of the demonstration of a 1 kW SOFC stack operation ended in 1993. This important milestone was finally reached in the begin of 1994. The second project phase with the final milestone of a 20 kW module operation will last till end of 1995. This result will form a basis for the next phase in which a 100 kW pilot plant will be established and tested.

BASIC DESIGN

The planar design of the Siemens high-temperature fuel cell combines metallic and ceramic materials and allows high power densities to be achieved.

A fuel cell stack (fig. 1) consists of two metallic end plates and several bipolar plates which direct the process gases to the electrochemically active elements. A main task has been to adapt the thermal expansion coefficient of the metallic plate to that of the
electrolyte made of 8YSZ and at the same time attaining high corrosion resistance. These goals have been reached with a new metal alloy called CrFe5Y2O31, which has been developed together with an Austrian partner, Metallwerke Plansee. This metallic bipolar plate has good electrical conductivity making it especially well-suited for high current densities, and produces a very uniform temperature distribution due to its superior thermal conductivity (1). Its high mechanical strength at increased temperatures permits the manufacture of large plates as well as large-volume stacks consisting of numerous individual cells.

New methods have been investigated to reduce the costs for manufacturing and shaping of plates out of this material. Hot isostatic pressing and sawing are very promising for the manufacturing of smooth thin plates. The electrochemical machining method, which is used to manufacture the grooves for the gas flow in the bipolar plate, is of great interest. The breakthroughs can easily be realized by water jet cutting.

A characteristic of the Siemens design is the multiple cell array concept (2) which depends on the manufacture of large plates as mentioned above. It allows the arrangement of several ceramic single cell elements (so-called PENs) in parallel in one layer as shown in fig. 1. By this measure larger electrode areas can be realized in one stack. The actual bipolar plate has dimensions of 260 x 260 mm². This allows the implementation of 16 PENs of the size 50 x 50 mm² in parallel in one layer. The total electrode area per layer results to 256 cm².

On the anode side a Ni-grid works as functional layer to improve the electrical contact. For this purpose LaCoO₃ is used on the cathode side. This perovskite is wet sprayed as a powder with a certain grain size by an air brush method. It results in a deformable layer compensating the thickness differences of the large number of parts which have to be assembled in one layer in parallel as well as in series. The contact resistivity of the two layers is depicted in fig. 2.

The sealing of the PENs to the plate (inner sealing) and of the plates with each other (outer sealing in the range of the internal manifoulding) is realized by deformable glass-green tape. This can easily be shaped by a stamping tool, which allows low manufacturing costs. The electrical insulation between the bipolar plates is accomplished by using ceramic or glass-ceramic frames. The latter can also be stamped in a green state.

All the manufacturing processes as well as the characteristics of the materials used are described in several specifications. This is part of the product assurance work performed in the whole project as it is necessary for the certification according to DIN ISO 9001.
STATUS OF PEN DEVELOPMENT AND MANUFACTURING

Decreasing the internal cell resistance, mainly by reduction of the polarization losses at the electrode/electrolyte interfaces, has led to a power output of 0.7 W/cm² with single cells (current density of 1 A/cm² at a cell voltage of 0.7 V, measured with hydrogen and oxygen at 975 °C) (3). The operation with air instead of oxygen results in power densities of 0.45 W/cm². This power output is reduced to about 30%, if the operating temperature is 820°C. The different characteristics are illustrated in fig. 3.

Parallel to the improvement of current/voltage characteristics, also the long term behaviour has been investigated. Changes in electrode preparation and microstructure resulted in degradation rates of 3% in 1000h as tested at 950°C in ceramic housing. At 850°C no degradation could be observed over a period of 4500h. In a first long term test at 850°C in a metallic housing, a degradation rate of 2% per 1000h was observed during 2000h of operation.

A very important step towards manufacturing of bigger stacks has been the transfer of the PEN manufacturing from laboratory scale to a pilot plant. In this plant 30000 electrolytes of the size 50 x 50 mm² can be manufactured per year. The capacity of screen printing and sintering of electrodes is 40000 per year. This means PENs for a 100 kW module (operating with air and 80% fuel utilization) can be manufactured per year. In a next step, the manufacturing of PENs with the size 100 x 100 mm² will be implemented.

1.8 kW STACK TEST RESULTS

Based on the design described above, in April 1994 a stack with 13 cell layers has been assembled, each layer consisting of 16 parallel PENs, which means 208 PENs with an electrode area of 3328 cm² in total. The stack had dimensions of 260 x 260 x 50 mm³, including end plates.

It has been operated with pure non humidified hydrogen. The open circuit voltages of all layers have ranged from 1.30 to 1.35 V, which shows that all PENs have been sealed very tight to the bipolar plates. After an operation time of 74h at 950°C the max. output power of 1.76 kW has been measured using hydrogen and oxygen. This corresponds to a power density of about 0.53 W/cm². Four single layers with a higher cell voltage have delivered a power density of 0.6 W/cm². The oxygen utilization has been 46% and the fuel utilization 27%. The cell voltage characteristics of the 13 layers are depicted in fig. 4. A max. current density of 800 mA/cm² at a mean cell voltage of 660 mV has been attained. The differing characteristic of layer one is due to its reduced temperature. The reason lies in the increased hydrogen flow because of discrepant flow dimensions in the base plate.
After 77h of operation, a measurement has been performed using air instead of oxygen. In this case the max. power has been 0.8 kW, which means a power density of 0.24 W/cm². The power/current density characteristics are shown in fig. 5. The oxygen utilization has been 58% and the fuel utilization 14%.

Measurements at reduced temperatures have shown a decrease in power down to 40% at 850°C and to about 25% at 800°C. The corresponding characteristics are outlined in fig. 6.

An increase of fuel utilization from 8% to 64% at a constant current density of 250 mA/cm² at 850°C has caused a decrease in power by 10% as illustrated in fig. 7.

A temperature cycle from 950°C to room temperature and back to 950°C has been performed. Only one layer was worse afterwards, probably because of leakage. The other 12 layers have shown no changes of current/voltage characteristics. This result provides a good basis for future development.

After about 300h of operation time, the test has been terminated to investigate the used parts for further improvement of the following stacks.

100 kW PILOT PLANT

The reported design is used as a basis for the layout of further bigger stacks and modules. It is planned to build up a 100 kW module consisting of four 25 kW stacks till end of 1998. According to the current planning, these stacks will have bipolar plates with the dimensions of about 360 x 360 mm², each with 9 PENs of the size 100 x 100 mm². This results in stacks with a height of about 0.5 m. A sketch illustrating the main dimensions is depicted in fig. 8.

Investigations on system behaviour and system calculations have led to a flow scheme of a 100 kW combined heat and power plant with an electrical efficiency of about 52% and a total efficiency above 90% (fig. 9). These values are based on the use of air and natural gas with internal reforming.

CONCLUSIONS

The test results described above have shown that the straight forward development is successful. It has proved the feasibility of larger stacks based on the multiple cell design using metallic bipolar plates.
The development of PENs with high power density shows the potential of this SOFC technology. The aim must be to reach these values as near as possible under real operation conditions and to further improve the long term stability.

But even in case of current/voltage characteristics, as realized today, high electrical and system efficiencies can be attained: and this is already valid for plants with lower power. Thus it represents a great advantage compared to existing technologies.

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Figure 1: Assembly concept of a stack
Figure 2: Contact resistivity of functional layers

Figure 3: PEN characteristics at different operating parameters
Figure 4: 1.8 kW stack - cell voltage characteristics

Figure 5: 1.8 kW stack - power characteristics with oxygen and air
Figure 6: 1.8 kW stack - power characteristics at different temperatures

Figure 7: 1.8 kW stack - power characteristic at increased fuel utilization at 850°C and 250 mA/cm²
Figure 8: 100 kW module - main dimensions

Figure 9: 100 kW pilot plant - block diagram