SOFC DEVELOPMENT AT SIEMENS

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ABSTRACT

Based on the results of the 10 kW stack, operated for more than 1400 hours in June 1995 the development of components and system was continued. The size of cells was increased to 100 x 100 mm² and integrated in one metal bipolar plate using the multiple cell array design. This leads to an electrode area of more than 700 cm². Improvement of the cathodes let to current densities of 0.8 A/cm² at 0.7 V, using hydrogen with 50 % water and air. As important development goal a 20 kW test facility was designed and constructed. It was set into operation in September 1996, delivering in a first test 7.2 kW with hydrogen/air. Based on this concept, a 100 kW plant is being designed.

INTRODUCTION

The SOFC development at Siemens has been started in 1990 after a two years preparation phase. The first period ended with the demonstration of a 1 kW SOFC stack operation in the begin of 1994. The second project phase should show the operation of a stack of the 10 kW class, which was finally tested in June 1995. The original goal of testing a 20 kW module had to be shifted to the end of 1996. The result of this test will form a basis for the next phase in which a 50 to 100 kW pilot plant will be built and tested.

BASIC DESIGN

The planar design of the Siemens high-temperature fuel cell combines metallic and ceramic materials and allows high power densities. A fuel cell stack consists of two metallic end plates and several bipolar plates containing channels which direct the process gases to the electrochemically active elements. The
design is described in detail in (1). The metallic plate material is the Plansee-alloy CrFe5Y2O31, which has proven its very high corrosion resistance in several long term tests in various atmospheres up to 5000 hours (2). Its good thermal conductivity in combination with its high mechanical strength at elevated temperatures (3) allows temperature differences of 200 to 300 °C between gas inlet and outlet without damage to the stack, which could be shown in several stack tests.

A characteristic of the Siemens design is the multiple cell array concept (4) which depends on the manufacture of large plates, possible by using the metal described above. It allows the arrangement of several ceramic single cell elements (so-called MEAs) in parallel in one layer. By this measure larger electrode areas can be realized in one stack. The largest bipolar plate used up to now had dimensions of 260 x 260 mm². This allows the placement of 16 MEAs of the size 50 x 50 mm² in parallel in one layer. The total electrode area per layer is 256 cm². The actual max. size combines 9 cells of the new type 100 x 100 mm² in one plate of the dimensions 360 x 360 mm². This leads to an active electrode area of 729 cm² in one layer. Stacks using these plates will be tested in the first half of 1997.

By using high temperature alloys, which form chromium oxide layers against corrosion, a new problem appears. At temperatures used in SOFC stacks chromium oxide evaporates from the surface of the bipolar plate and poisons the cathode, which results to an increased degradation rate. To avoid this a protective layer out of LaSrCrO₃ is applied to the surface of the bipolar plate by vacuum plasma spraying (5).

**STATUS OF CELL DEVELOPMENT**

Comparison between cell characteristics measured with oxygen and air showed a much bigger difference than caused by the Nernst potential. To improve the behaviour with air a two-layer cathode with changed micro structure was developed, using a substoichiometric LaSrMnO₃. Decreasing the internal cell resistance, mainly by reduction of the polarization losses at the electrode/electrolyte interfaces, has led to improved cell behaviour with a power output of 0.9 W/cm² (current density of 1.2 A/cm² at a cell voltage of 0.75 V, reacting hydrogen and air at 950 °C). Using a mixture of hydrogen with 50 % water vapour a power density of 0.6 W/cm² was reached. This power output is reduced by about 50 %, if the operating temperature is lowered to 850 °C. This means that the envisaged current density of 300 mA/cm² at 0.75 V can already be reached at 850 °C. This is regarded as great advantage with reference to cost reduction of peripheral components. The different current-voltage relations are illustrated in fig. 1.

Parallel to the improvement of current-voltage relations, also the long term behaviour has been investigated. Changes in electrode preparation and micro structure resulted in degradation rates of less than 2 % in 1000 hours, tested at 950 °C in ceramic housing in...
H₂/H₂O-atmosphere of 1:1. At 850 °C no degradation could be observed over a period of 4500 h. In a first long term test at 850 °C in a metallic housing, a degradation rate of 2 % per 1000 h was observed during 2000 h of operation. First tests at 950 °C, using a protective layer against chromium evaporation showed similar degradation rates as tests in ceramic housings (5).

Also first tests with methane and total internal reforming have been performed. After activation under 3 % water vapour in hydrogen the fuel gas is first changed to a mixture of 21,5 % water vapour in hydrogen, which has the same oxygen activity as methane/water vapour with a steam to carbon ratio of 2, which is used afterwards. The corresponding characteristics are shown in fig. 2. The cell does not reach its thermodynamic value, because the operating temperature inside the cell is reduced by about 40 °C by the internal reforming. But besides this effect no problems occurred during operation with methane.

A very important step towards manufacturing of bigger stacks has been the transfer of the MEA manufacturing from laboratory scale to a pilot plant. In this plant, 30000 electrolytes of the size 50 x 50 mm² or 10000 parts of the size 100 x 100 mm² can be manufactured per year. The capacity of screen printing and sintering of electrodes is slightly higher. This means cells for a module up to 300 kW power (operating with air and 80% fuel utilization) can be manufactured per year. The production of MEAs with the size 100 x 100 mm² has been started in the beginning of 1996.

10 KW STACK TEST RESULTS

Based on the design described above, in June 1995 a stack with 80 cell layers has been assembled, each layer consisting of 16 parallel cells, which means 1280 MEAs with a total electrode area of about 2 m². The stack had dimensions of 260 x 260 x 260 mm³, including end plates.

After brazing and anode reduction the stack performed well. Using dry hydrogen the open circuit voltage of the stack was 104 V. The mean voltage of a single layer was 1.3 V, which indicates that all cells have been sealed gas tight to the bipolar plates. The initial loading of the stack showed a power output of 10.7 kW (corresponding stack voltage 60 V, mean cell voltage 0.75 V ± 50 mV) at 950 °C and gas utilisation of 50 %. Further loading of the stack was limited by the electrical equipment of the test stand. Operating with hydrogen and air gave a power output of 5.4 kW. The power output at 850 °C was still 4.1 kW. The used cells were of an older type than described above. The different current-voltage relations of the stack are plotted in fig. 3.

After a number of different I/U-measurements, the stack was operated at constant load of 200 - 300 mA/cm² for 1000 hours at 850 °C. During this time, the stack showed a relatively high degradation rate and, compared to the initial performance, a power loss of
19 % was observed at the designed load of 270 mA/cm² (no protective layer was implemented in this stack assembly). Afterwards, a thermal cycle of 950 °C/RT/950 °C was run without serious damage to the stack.

20 KW TEST STAND

Based on the results of the 10 kW stack a 20 kW system was designed and built. It is suited for the operation with hydrogen and air (with the possibility to shift to oxygen). It is a self-sustaining system down to a power output of about 10 kW. Therefore, the heat loss of the stack-housing to the environment has to be reduced to about 4 kW. The test facility is designed in a manner that 4 stacks can be operated in parallel. The stacks including thermal insulation are placed on a rack of the dimensions 2.8 x 2.8 x 2 m³. On top of this rack there is a basement out of high temperature concrete of 1 x 1 x 0.5 m³. Beyond this platform the recuperative high temperature heat exchangers and the high temperature tubing is arranged. The test facility is controlled by a stored programme computer. Fuel gas flow is adjusted proportional to the load and the air flow regulated so as to keep the max. stack temperature constant. The gases are preheated by the hot waste gases in specially designed recuperative heat exchangers. A block diagram of the test stand is shown in fig. 4.

The plant was put into operation in September 1996. For this purpose 2 stacks, each consisting of 50 layers were assembled directly in the test facility (this means 1600 cells with a total electrode area of more than 2.5 m²). The two stacks were sealed at 1000 °C. The temperature distribution inside the stacks was as homogeneous as expected and also the reduction behaviour was excellent. The voltages of the controlled cell groups (each five cell layers were measured together as one cell group) showed very similar characteristics. After reduction the mean open circuit voltage was about 1.25 V, using dry hydrogen and air, which means a sufficient tight sealing. The operation temperature of the stacks was 900 °C. During the measurement of a first characteristic using hydrogen and air the two stacks behaved very similar concerning temperature distribution and current produced (the current of each stack could be observed separately). The characteristics are shown in fig. 5 and 6. A power output of 7.2 kW could be reached at a fuel utilisation of 30 % and an air utilisation of 50 %. Starting a measurement with oxygen instead of air the resistance of a part of one stack increased in such a way that the test had to be stopped.

The reasons for this problem are now under investigation. The results of the first operation of the test facility are, that all components of the test stand operated well and that the assembly concept of parallel operated stacks could be proven.
FUTURE WORK

Based on the work described above, it is planned to build up a 50 kW component test stand consisting of 25 kW stacks till end of 1998. This results in stacks with a height of about 0.5 m, using the new design with nine cells of the size 100 x 100 mm² in one layer.

In a next step it is planned to establish a 100 kW demonstration plant in the year 2000, together with a customer, to show the feasibility of SOFC plants for combined heat and power generation.

Investigations on system behaviour and system calculations have led to a flow scheme of such a 100 kW combined heat and power plant with an electrical efficiency of about 50 % and a total efficiency above 90 % (fig. 7). These values are based on the use of air and natural gas with internal reforming.

CONCLUSIONS

The test results described above proved the feasibility of manufacturing and operating larger stacks based on the multiple cell design using metallic bipolar plates as well as the ability to establish and operate systems using these stacks in a multiple stack arrangement.

The development of cells with high power density shows the potential of this SOFC technology. The high current densities, possible with the planar design, were shown with dry hydrogen and oxygen in the beginning of the project. In the last period they were demonstrated also under real operating conditions. Also the possibility to reduce the operating temperature to at least 850 °C, using the available materials, could be shown.

The long term stability has to be further improved using the developed protective layers on the cathode side.

High electrical and system efficiencies can be attained, even for small plants with a power output of 100 kW. This represents a great advantage compared to existing technologies.

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Fig. 1: I/U Curves of Improved Cells with Different Gases, at 850 and 950 °C

- 0.5 l/min anode gasflow
- 78.5% H2, 21.5% H2O
- 0.53 Ohm cm²

- 0.5 l/min anode gasflow
- 33.3% CH4, 66.7% H2O
- 0.40 Ohm cm²

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Fig. 2: I/U Curves Measured with Methane and Comparable Hydrogen/Water-Vapour at 950 °C

- 0.5 l/min anode gasflow
- 78.5% H2, 21.5% H2O
- 0.5 l/min air

- 0.5 l/min anode gasflow
- (S/C = 2)
- 33.3% CH4, 66.7% H2O
- 0.5 l/min air

Fig. 3: I/U Curves of 10 kW Stack

Fig. 4: Block Diagram of 20 kW Test Stand
Fig. 5: I/U Characteristics with Hydrogen/Air at 900 °C in the 20 kW Test Stand

Fig. 6: P/I Characteristics with Hydrogen/Air at 900 °C in the 20 kW Test Stand
Fig. 7: Flow Scheme of a 100 kW CHP Plant with Heat Recovery