HIGH TEMPERATURE MEMBRANE FUEL CELL WITH INTEGRATED HEAT PIPE

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Abstract: This paper presents work towards a novel high temperature electrolyte membrane (PEM) fuel cell stack, based on function integrated innovative components. Silicon as functional material and a membrane adapted from Polybenzimidazole (PBI) allow the operation up to 200°C. Highly efficient heat pipes are produced for the integration into the bipolar plate of the stack to enable a homogeneous and 2-dimensional heat distribution. The reduction of the number of components implies the manufacture of bipolar plates with integrated heat pipe and flow field structures from monolithical silicon. A special equipped test bench has been developed allowing measurements of current density, impedance and temperature on segmented high temperature PEM fuel cells.

Key Words: high temperature PEM fuel cell; heat pipe; silicon; test bench

1. INTRODUCTION

High temperature PEM fuel cells, operating at about 170°C, have many advantages in comparison to standard PEM fuel cells. The major benefit of high temperature PEM fuel cells is the simplification of the water management as only the gas phase of water needs to be considered. Moreover catalyst stability towards carbon monoxide (CO) is enhanced, allowing the application of regenerative hydrocarbons in cogeneration without purification. The increased temperature level of waste heat facilitates an efficient utilization for heating and cooling.

The challenge on PEM fuel cells is the thermal management for heating up to the required working temperature in the initial phase and dissipating waste heat during operation. Furthermore the required heat transfer technology should be capable of being integrated and miniaturizable. An optimal qualified technology for this application is a heat pipe. A heat pipe is a heat transfer technology that can transport a vastly larger quantity of heat compared to an equivalent cross section of solid material. The larger heat flow is achieved by circulation of a working fluid. Inside a heat pipe the fluid turns to vapor at the hot interface. The gas naturally flows and condenses on the cold interface. The so generated condensate is moved by capillary action back to the hot interface to evaporate again and repeat the cycle. With the aid of this mechanism heat pipes enable a homogenous and 2-dimensional heat distribution. Integrated into the bipolar plates of the stack the build-up of a PEM fuel cell with a reduced number of components is possible.

2. THEORY AND EXPERIMENTAL

In the following the novel stack layout as well as the heat pipe concept are presented. Furthermore the developed test bench is introduced which will serve for the characterization and determination of the optimal system management strategies.

2.1 Stack layout

A conventional fuel cell stack is made up of a membrane electrode assembly at which on both electrodes a gas diffusion layer and a gas distribution structure are connected. Typical materials are graphite and micro-structured metal foils respectively. The challenges of the application of these materials are the difficult micro-structuring of graphite and the poor corrosion resistance of metals. Besides, operation temperatures of 170°C demand a heat transfer component and a PBI-based membrane electrode assembly.

The new design implies the assembly of a fuel cell stack with a reduced number of components. We have reported a stack design with a reduced number of components already at the Conference of Microreaction Technology ’00 [1], the Grove

Cell Symposium '01 [2] and HARMST '01 [3]. In our design of the fuel cell gas diffusion layer, gas distribution structure and bipolar plate are combined to one function unit. This as well as the integration of the heat transfer technology into the bipolar plate allows the miniaturization of the stack and a higher volumetric power density. Gas supply and removal is realized by a flow field inserted into the surface of the bipolar plate. The flow field is made-up of a parallel channel array with manifolds for gas inlet and outlet at the ends of each channel. Figure 1 shows a schematic few of the fuel cell entity.

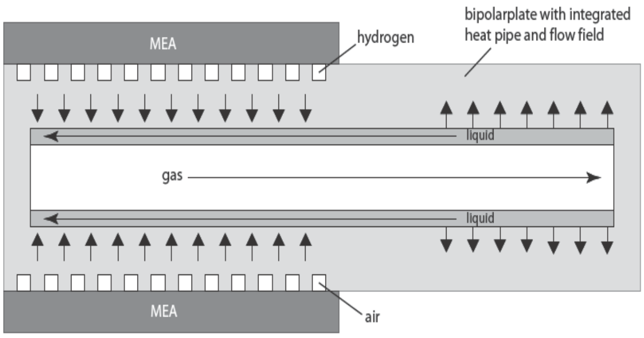

Figure 1: Schematic view of the entity of a fuel cell with integrated heat pipe

The assembly of the fuel cell stack with novel function integrated components requires a material which meets the demands of the heat transfer technology and the fuel cell such as good thermal and electrical properties, corrosion resistance and good suitability for structuring. Silicon shows at operating temperatures of approximately 170°C a good thermal and electrical conductivity. Since the resistivity of silicon strongly decreases with increasing temperature, at 170°C comparable values to that of metals can be achieved. Corrosion resistance is given without expensive coating or alloying.

The micro structuring and bonding of silicon is easy realizable with the techniques of micro system technology. A two-stage anisotropic dry etch process with an etch rate of 2 - 3 µm/min enables a fast manufacture of deep slender channels with an aspect ratio between 15:1 and 30:1. Bonding processes permit a leak proofed connection of two silicon parts.

2.2 Heat pipe

In order to observe the functionality of the heat pipe and hence optimize the geometrical layout first heat pipes are manufactures from a silicon-pyrex bond. The heat pipe consists of a sealed hollow tube with rectangular cross section. On the internal side of the silicon component a parallel groove structure exerts a capillary force on the liquid phase of the working fluid. The grooves are manufactured by the dry etch process mentioned in section 2.1. The orifice for the filling process is lasered or drilled with a diamante point. Anodic bonding enables a leak proofed connection of the silicon and pyrex component.

Which working fluid is chosen depends on the temperature condition, in which the heat pipe must operate. The properties of the working fluid which determine maximum heat transport can be combined to form a figure of merit M [1].

$$M = \frac{\rho_l \cdot \sigma_l \cdot L}{\mu_l} \qquad (1)$$

The fluids with higher figure of merit are more desirable. Figure 2 illustrates the figure of merit of some common working fluids. Water shows very good characteristics in a wide temperature range. Apart from this water facilitates considerably the handling because of its nontoxicity and is therefore chosen as working fluid for the first heat pipes.

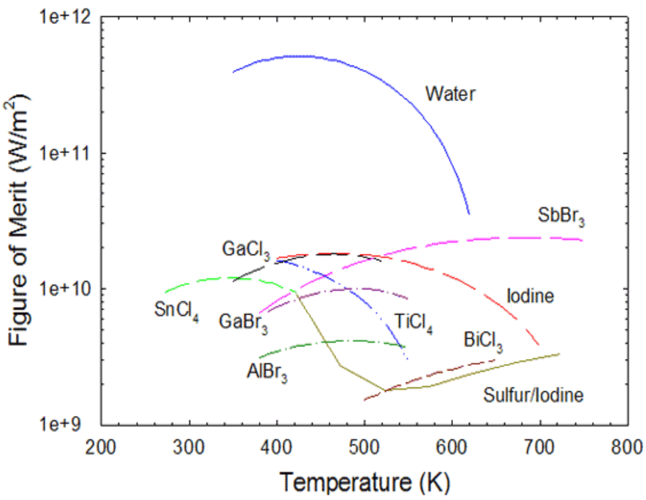

Figure 2: Figure of Merit for selected working fluids [2]

2.3 Test Bench

A test bench was developed, allowing single cell measurements on high temperature PEM fuel cells. Cell temperature can be controlled by a thermo-oil circuit and up to 200°C can be achieved. The anode and cathode gas flow rates are controlled by mass flow controllers. In the anode gas stream different CO concentrations of up to 5% can be added. Additionally, anode and cathode can be purged with nitrogen. On this test bench single cells can be measured in potentiostatic or galvanostatic mode to a maximum current of 50 A.

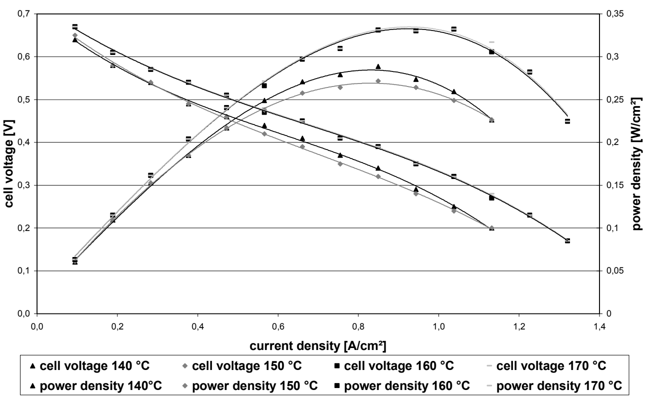

Figure 3: Cell characteristics at different temperatures

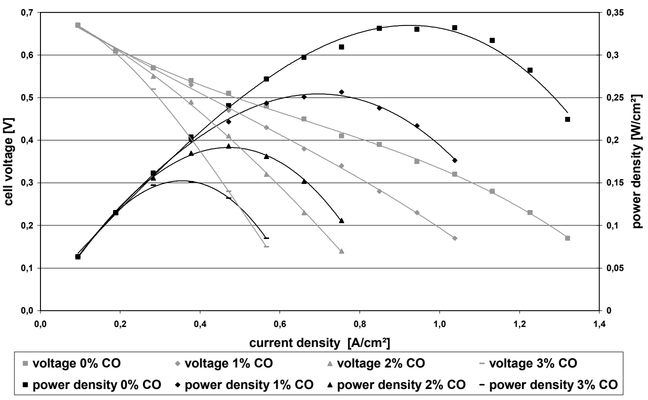

Figure 4: Cell characteristics at different CO fractions in the anode gas (160°C cell temperature)

For first tests a cell without heat pipe with an active area of 25 cm² and a parallel graphite flow field on both sides was used to characterise a PBI-based high temperature PEM membrane electrode assembly. Polarisation curves were measured at different gas flow rates, temperatures and CO rates. Figures 3 and 4 show polarisation curves and power densities of a PBI-based membrane in the test cell at different temperatures and CO concentrations. The stoichiometry was set to 1.2 on the anode and 2 on the cathode.

Figure 3 shows a slight increase of the maximum power density at increasing temperature between 140°C and 170°C. Figure 4 displays a strong dependency of the maximum power on CO concentration. It decreases about by 20% from 0% to 1% CO.

3. DISCUSSION

A fuel cell with silicon as functional material doesn't achieve comparable high area-related power densities to a conventional fuel cell. However the application of silicon makes the integration of the heat pipe and the reduction of the number of components possible and thus a considerably higher volumetric power density. If high doped (> 10^{19} cm^{-3}) silicon is used the electrical conductivity can be increased even more. Current-voltage-measurements indicate that, in contact with a metal layer, ohmic characteristics occur. So also the contact to the metallic endplates in a stack assembly forms no problem.

The filling of a well-defined amount of working fluid into the heat pipe is difficult, because of the enclosed air. The realization of a second orifice allows the air to escape through it and facilitates for this reason the filling. After sealing the first orifice the heat pipe is heated up until boiling starts. Thus noncondensible gases are displaced before also the second orifice is sealed. Hence the filling process can be performed under ambient conditions and no extensive vacuum system is needed.

The mentioned dry etch process enables the manufacture of deep slender channels und thus high capillary forces, which are necessary to transport the condensate in the heat pipe. An improved wetting of the capillary structure by the working fluid can lead to a further increase of heat pipe performance. Thereto the capillary structure is selectively coated with a low temperature PECVD nitride layer. Measurements with water show a decrease in contact angle from 75° on a blank silicon surface to 27° on a silicon nitride surface.

The measurements at the test bench show that the cell is still operable at 3% CO, unlike standard PEM fuel cells that need CO concentrations clearly below 100 ppm. Furthermore variations of flow field geometry, clamping pressure and anode/cathode stoichiometry lead to only small changes in power density. So, simple flow field structures with low pressure drop and low gas flow rates ($\lambda_{anode} = 1.2$, $\lambda_{cathode} = 2$) are possible. This can lead to smaller fan or pump power compared to similar low temperature PEM fuel cells and therefore to a higher system power.

4. CONCLUSION

An assembly with novel function integrated components was presented, which envisions the manufacture of bipolar plates with heat pipe and flow field structures from monolithical silicon under application of micro system technologies. Besides processes were introduced which facilitate the filling procedure and increase the capillary action of the heat pipe.

Furthermore a test bench was presented allowing single cell measurements on high temperature PEM fuel cells. In the measurements an ideal operating range (temperature, carbon monoxide concentration, gas flow rates, start and stop procedure) for high temperature PEM fuel cells was identified. The influence of these different parameters to the cell power density was observed. These results give important information of the construction and dimensioning of a fuel cell system.

The introduced concept promises a reliable and steady operation and has the potential for high energy densities.

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NOMENCLATURE

ρ_l = density of the liquid working fluid
σ_l = surface tension of the liquid
μ_l = viscosity of the liquid working fluid
L = enthalpy of vaporization of latent heat

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