

## Low-cost air-cooled PEFC stacks

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### Abstract

The general objective of the work was to adapt the existing technology of polymer electrolyte fuel cells (PEFC) to the demands for commercial applications and to make it competitive with respect to performance and costs compared to the present day technology of combustion engines. This should be realised by the use of optimised electrodes which enable a highly efficient operation with air at low overpressure and a simplified construction of the electrochemical cell which is capable of mass production and low-cost. The initial project cost goal (materials and fabrication, extrapolated to a production of 100,000 fuel cell systems per year) was 100–200 ECU/kW. The feasibility of this goal was planned to be ascertained by the development of a PEFC stack in the size of 3.5 kW rated power. © 2002 Elsevier Science B.V. All rights reserved.

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### 1. Introduction

The existing status of polymer electrolyte fuel cells (PEFC) proved the reliability of the system in various applications, such as manned space flight or submarine propulsion. However, its costs are far too high to get access to any commercial market. Presently, the costs for hand-made prototype polymer electrolyte fuel cell stacks are in the range of US\$ 20,000/kW<sub>el</sub>, of which about 90% are manufacturing costs and about 10% material costs. Even if the manufacturing cost in a mass market production decreases tremendously to below US\$ 10/kW<sub>el</sub>, the material cost still have to be lowered by two orders of magnitude. Therefore, in a BMBF<sup>1</sup> funded project from 1994 to 1998 different ideas for cost reduction were tested and verified in small single cells. In a subsequent Joule project these improvements were used to build up a fuel cell stack. This stack should have a rated power of 3.5 kW<sub>el</sub> and a peak power of 5 kW<sub>el</sub>. The applied fuel is pure hydrogen or purified hydrogen (CO < 10 ppm) and the oxidant is air at a pressure less than 1.5 bar<sub>a</sub>. The goals for the power/volume and the power/weight were 2.5 l/kW<sub>el</sub> and 2.5 kg/kW<sub>el</sub>, respectively.

To compete with the present day technology of combustion engines the costs for the polymer electrolyte fuel cell stack have to be decreased to less than US\$ 30/kW<sub>el</sub>.

### 2. Improvements for different stack components in regard to cost reduction

In order to reach this restrictive cost goal for commercial applications a lot of different improvements in the stack are necessary. The items for cost reduction and the cost goals are given in Fig. 1.

#### 2.1. Membrane electrolyte assembly (MEA)

The presently used expensive Nafion<sup>®</sup> membrane has to be replaced by a low-cost material. The cost goal of less than US\$ 20/m<sup>2</sup> should be attained, if, for example, non-fluorinated polymers be used. Test runs with various alternative membranes from different suppliers in single cells were performed. One kind of non-fluorinated membrane based on polyetherketone was operated successfully for more than 2000 h ( $T = 80\text{ }^{\circ}\text{C}$ ,  $p = 1.5\text{ bar}_a$ , thermal and load cycling).

The costs for MEAs can be further decreased enormously by reducing the Pt-loading of the electrodes. Initially a Pt-loading of 4 mg/cm<sup>2</sup> for each electrode was required to obtain the desired power output of single cells.

In Fig. 2, results for different Pt-loading are presented. The  $U$ - $j$  characteristics were measured at 80 °C using pure hydrogen as fuel and pure oxygen as oxidant at a pressure of 2 bar<sub>a</sub> for both gases. The tests were run in pure oxygen to eliminate the transport effects caused by the cell design. It can be recognised that with a Pt-loading of 0.15 mg Pt/cm<sup>2</sup> for each electrode similar results were obtained as with the conventional standard electrode containing 2 mg Pt/cm<sup>2</sup>.

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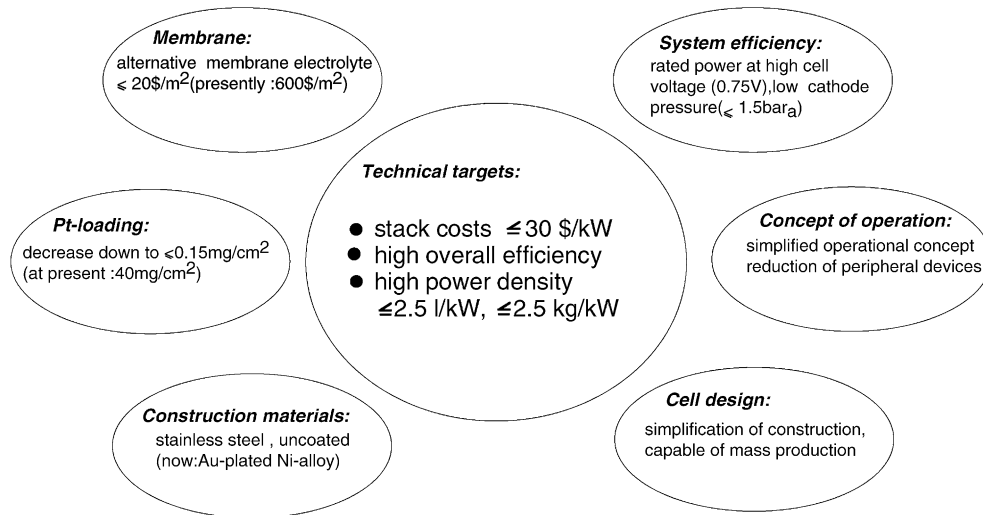


Fig. 1. Targets for a low-cost fuel cell stack and different issues for cost reduction.

Even with a Pt-loading of  $0.07\ \text{mg Pt}/\text{cm}^2$  a sufficiently high power output was observed in respect of the goal (750 mV at  $500\ \text{mA}/\text{cm}^2$ ).

These electrodes were prepared by spraying an ink directly onto the membrane. The ink consists of fine dispersed platinum on carbon in a solution of Nafion<sup>®</sup>, alcohol and water.

In addition, electrodes were fabricated by screen printing. In this case, a Pt paste was used, because in this manufacturing process the material has to have a higher viscosity. For the preparation of the paste the same ingredients were used as in the case of the ink preparation, only in different concentrations.

Applying this method for the electrode preparation the Pt amount could be lowered to  $0.25\ \text{mg Pt}/\text{cm}^2$  without a

significant loss in the power output of the stack. There is still a potential for further Pt reduction in the optimisation of the manufacturing process.

## 2.2. Construction materials for bipolar plates

The presently used materials for the bipolar plate are too expensive in respect of both material costs and manufacturing costs. There are two different kinds of materials which were used as bipolar plate materials in prototype fuel cells: graphite or carbon based composites and metals, such as niobium, titanium or different alloys.

The main disadvantage of graphite and carbon based bipolar plates is the expensive machining process for the generation of the gas channels. In contrast to metals the

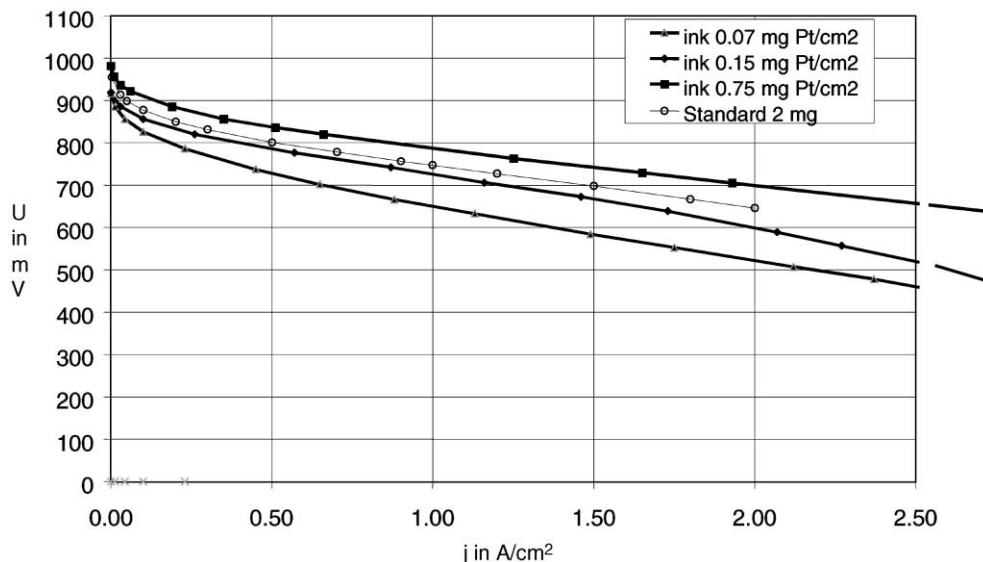


Fig. 2. Effect of Pt-loading on the  $U$ - $j$  characteristic:  $\text{H}_2/\text{O}_2$  operation at  $2.0\ \text{bar}_a$  (humidified);  $80\ ^\circ\text{C}$ .

mechanical strength of graphite is lower and the gas permeability is higher. Therefore, graphite bipolar plates typically require a plate thickness of 1.5–5 mm, whereas metals can be used as thin foils of less than 100  $\mu\text{m}$  thickness. This results in a lower volume density for stacks built with graphite bipolar plate in contrast to stacks built with metallic bipolar plates, provided the power density of the membrane electrolyte membrane is equal.

The processing of the most metals is easy, especially if thin foils are used (see also Section 2.3 cell design). In order to reach the cost goal the metallic bipolar plates are not allowed to be coated with gold or another noble metal. Even due to the costs for a deposition process any coating should be avoided. For this reason two problems arise for the metallic bipolar plates. Most metals will suffer from a corrosion attack, if they are exposed to an electrochemical potential in an acid solution and, due to the formation of a thin oxide layers on the surface the contact resistivity between the metal surface and the current collector from the membrane electrode assembly will exceed the tolerated value of  $10 \text{ m}\Omega \text{ cm}^2$ .

Due to the cost problem the cheapest suitable metal has to be found. Therefore, on different electrochemical corrosion tests were carried out on different pre-selected iron-based alloys electrochemical corrosion tests at room temperature and at  $80^\circ\text{C}$ . In a first attempt, the conditions in a fuel cell were simulated by using an electrolyte containing different sorts of ions (200 ppm  $\text{F}^-$ , 100 ppm  $\text{Cl}^-$ , 200 ppm  $\text{NO}_3^-$  and 300 ppm  $\text{SO}_4^{2-}$ ).

In addition, the uniform corrosion of the iron-based alloys was investigated in a 0.1 M  $\text{H}_2\text{SO}_4$  electrolyte. The most severe condition were tested in a 0.1 M HCl solution, which can cause pitting corrosion. The results obtained on different iron-based alloys are documented in Fig. 3. For four different iron-based alloys pitting corrosion is initiated at potentials of approximately 0.6–0.75 mV. In these cases a significant increase in the current density is observed. These

alloys, including well known stainless steels, such as 316 L (1.4404) and 304 L (1.4306) are not suitable to be used as bipolar material.

It was calculated that the corrosion current density should not exceed  $10^{-6} \text{ A/cm}^2$  for car applications. In this case, a life time of only 4000–5000 h is required. For stationary applications the corrosion current density has to be even lower than  $10^{-6} \text{ A/cm}^2$ , because in this case, the demand on the life time is increased at least by a factor of 10.

In Fig. 3, it is outlined that there are few iron-based alloys, which have a sufficiently low corrosion current at potentials up to 1.1 V (zero electric voltage of a single cell). These materials show a similar behaviour to the usually used, more expensive nickel based alloys.

Due to the commercial availability of semi-finished material in respect of thin metal foils one corrosion resistant iron-based alloy was selected for cell measurements.

In every single cell the contact resistivity between the uncoated metallic bipolar plate and the current collector, consisting of a carbon paper, has to be low. It was calculated that a contact resistivity of  $10 \text{ m}\Omega \text{ cm}^2$  per interface can be tolerated, which causes a voltage drop of 10 mV in the  $U$ - $j$  characteristic at a current density of  $1 \text{ A/cm}^2$ .

The determination of the contact resistivity was done by four point probe measurement in a special device (Fig. 4) at room temperature. According to the conditions in single cells, the samples consist of a bi-layer of a thin metal alloy foil and a carbon paper, which were clamped between two gold-coated stamps. Applying a constant current density of  $500 \text{ mA/cm}^2$  to the samples the voltage drop was registered as a function of the applied mechanical contact pressure. In Fig. 4, the results for different samples are documented. The lowest contact resistivity was obtained using the usual gold-plated Ni-based alloy. At pressures higher than 2 bar the contact resistivity decreased below the critical  $10 \text{ m}\Omega \text{ cm}^2$ . First measurements with the selected iron-based alloy resulted in contact resistivities which were too high about

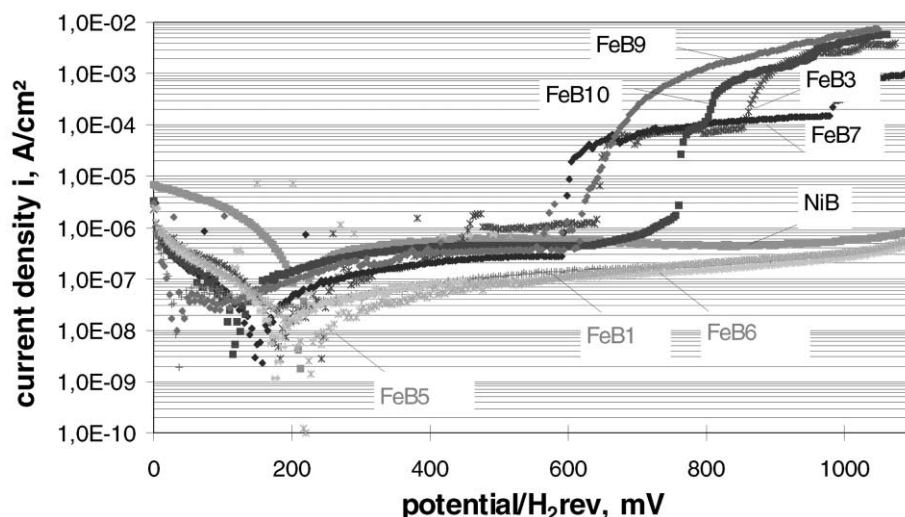


Fig. 3. Screening of possible construction materials: corrosion behaviour examined with the potential step method; electrolyte, 0.1 mol/l HCl.

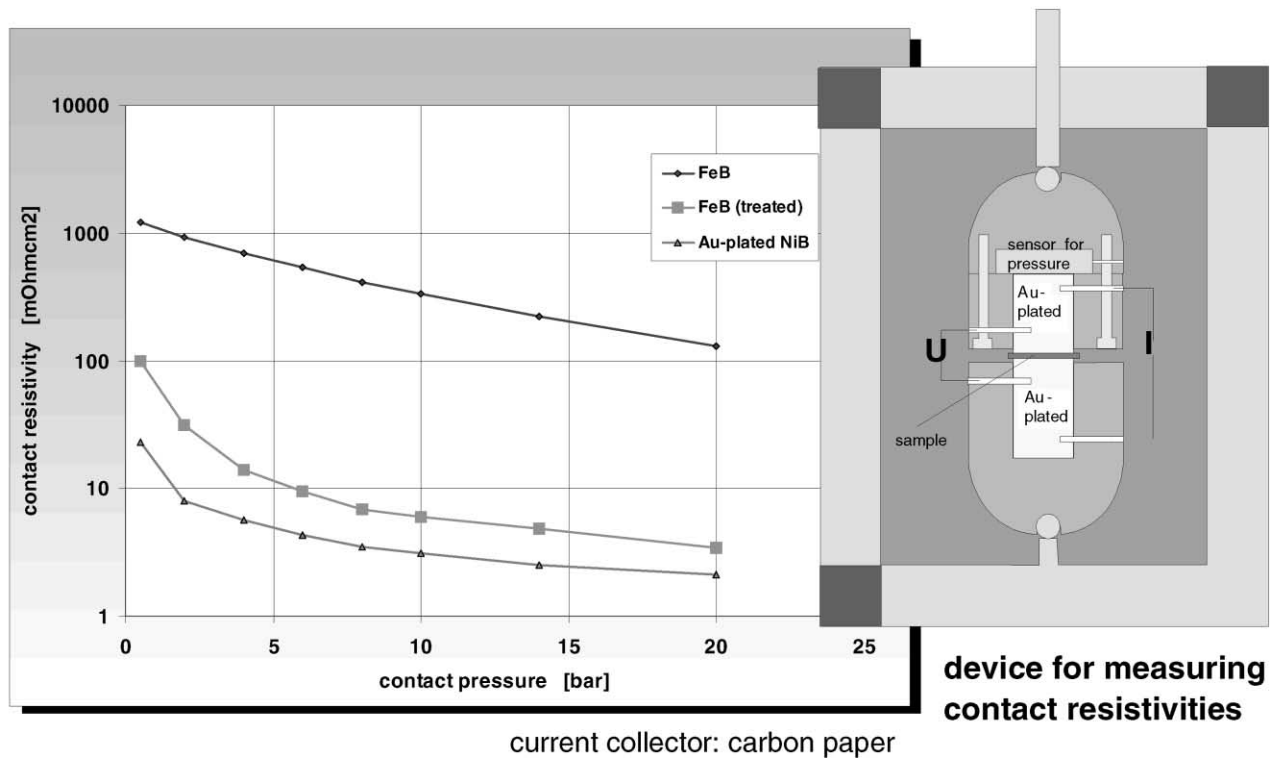


Fig. 4. Measurement of the contact resistivity as a function of the contact pressure: influence of a surface treatment on the contact resistivity of a Fe-based alloy.

two orders of magnitude. Applying a special surface treatment on the metallic foil the contact resistivity could be lowered to reach almost the values obtained with the gold-coated Ni-based alloy. In order to fall below the critical contact resistivity of 10 mΩ cm<sup>2</sup>, in this case a slightly increased contact pressure of approximately 5 bar is necessary. Due to the required cost reduction this treatment consists of no thin layer coating technique.

The stability of the contact resistivity was proven in single cell long term tests for more than 7000 h.

### 2.3. Cell design and concept of operation

Further cost reduction is obtained by the simplification of the cell design using thin sheet metal as construction material. In Fig. 5, the low-cost innovative cell design is presented. The gas channels were easily stamped into the thin metallic foils and the four manifolds for the gas supply at the corner of the cells were created by punching. These are established cheap production processes in the metal industry for mass production. In Fig. 5, the metal thin foils

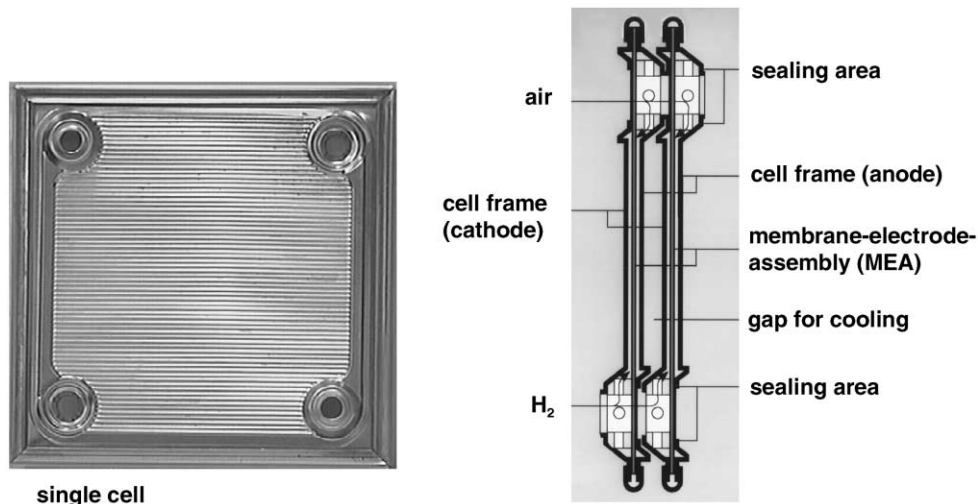


Fig. 5. Schematic view of the Siemens low-cost cell design.

are still coated with a gold layer, which is no longer required.

The membrane electrolyte assembly is located between two metallic foils, which were pressed together by four metal clamps. In this overscan and at hole edges (Fig. 5, right side) the sealing is realised by the membrane itself, thereby avoiding the use of any additional seals or glues.

The disassembly of such a construction for repair or recycling purposes is also easy. Furthermore, this concept allows for a compact lightweight fuel cell stack, which still enables air cooling. In Fig. 5, also the cooling gap between two single cells can be seen. The construction has been patented by Siemens in 1994 (DE 4442 285 C1 and EP 0795 205 B1).

Additional cost reduction is attainable by a simplification of the fuel cell system. The presently used fuel cell stack system requires an expensive water circulation system both for the cooling system and for the humidification of the reactive gases. In the cooling system peripheral components, such as pumps, heat exchangers and condensers, are necessary.

In the simplified stack design no water circulation system is required because an air cooling system is used for the fuel cell stack and no external humidification for the membrane electrode assembly should be necessary. Several fuel cell stacks of different sizes were operated successfully using air blowers for the stack cooling, but up to now the external humidification of the Nafion<sup>®</sup> membranes could not be completely abandoned without significant loss in the cell performance.

The stacks are operated at a low air pressure, typically 1.5 bar<sub>a</sub>. In this case, for the air compression a small and cheap air radial blower is necessary, which requires only a small amount of energy compared to the stack output. It is in the order of 1–3%. Other groups are operating the stacks at a high air pressure of about 3 bar<sub>a</sub> in order to achieve an improvement of the cell performance. In this case, approximately 17% of the stack output is necessary to run the required air compressor. There are already efforts to regain energy by using an additional air expander system. Presently, such compressor/expander systems are in development. An evaluation in respect to costs, local requirements and stack performance has to be done in future.

In a first step, a 100 cm<sup>2</sup> design was used to confirm the fundamental feasibility of the cooling and sealing concept by the operation of single cells and short-stacks. In a second step, a full-size design with an active area of 310 cm<sup>2</sup> was constructed and used for further experiments and building the final demonstrator.

### 3. Innovative fuel cell stacks

Due to the restricted space for the fuel cell system in passenger cars the fuel cell stack has to have a low volume. Using the metal sheet technology the lowest possible volume and weight can be realised.

Table 1

Operational parameters for the 35-cell final demonstration stack

Air pressure (bar <sub>a</sub> )	1.5
Air stoichiometry, $\lambda$	2–3
Air dewpoint (°C)	62
Hydrogen pressure (bar <sub>a</sub> )	1.5
Hydrogen stoichiometry	1.2–2
Hydrogen dewpoint (°C)	68
Inlet cooling air temperature (°C)	25
Outlet cooling air temperature (°C)	50–80

As a concept proof, in a first step, a modular 100 cm<sup>2</sup> cell stack was build, in which the improvements mentioned in Section 2 were realised. Then, the cell design was scaled up to a technically relevant cell size. An active area of 310 cm<sup>2</sup> was chosen as a compromise between voltage and power requirements. A 35-cell demonstrator was assembled and operated.

Based on experiments with single cells and short-stacks of the new 310 cm<sup>2</sup> design, the parameters for the operation of the 35-cell demonstration stack have been derived. The operational parameters are listed in Table 1.

The humidification of the final demonstrator was planned to be part of the test bench, because the use of dry gas was not successful with all types of MEAs tested.

The assembled final stack is shown in Fig. 6. Machined end plates of aluminium, which are not yet weight-optimised, were used. The bundle of thin wires is to detect each individual cell voltage. A commercial blower, used for car ventilation was attached at the stack as ancillary to remove the waste heat. In this demonstrator conventional membrane electrolyte assemblies had to be used because at the very beginning of the stack assembly advanced electrodes and membranes of the required size were not yet available.

The stack performance and, more precisely, parameters like minimum gas stoichiometry are determined by the weakest cells of the stack, and therefore, a good reproducibility of single cells is more important than a good performance which is only reached by some of the cells. Therefore, the voltage of every single cell was registered separately. For the 35-cell stack a standard deviation of the cell voltages of typically 20 mV was reached, and it could be lowered to about 5 mV for a higher lambda setting.

A load line was recorded after 30 h of operation, and parameters according to the above table were used. The performance data of two different 35-cell air-cooled stacks are documented in Fig. 7. The results obtained on both stacks are very similar, which proves a good reproducibility. At the rated point, at a current density of 400 mA/cm<sup>2</sup> an average cell voltage of 725 and 730 mV was obtained for the two stacks, respectively. The power output at 400 mA/cm<sup>2</sup> was 3.1 kW and a peak power of 6.2 kW (all values at 1.5 bar<sub>a</sub>) was obtained. Within the operation time of 300 h no degradation of the stacks could be observed.

Long term tests were performed on short-stacks, which consist of six cells with an active area of 100 cm<sup>2</sup> per cell.

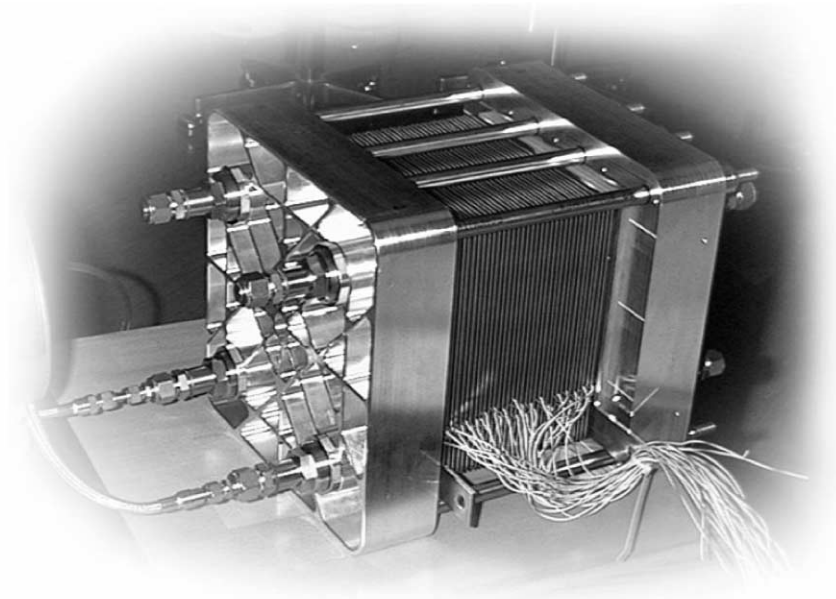


Fig. 6. Air-cooled low-cost fuel cell battery: 35-cell short stack; active cell area, 310 cm<sup>2</sup>.

The materials in these stacks are the same as in the larger stacks. During the long term test primarily a constant current density of 300 mA/cm<sup>2</sup> was applied on the cells and the voltage was recorded. During some time periods an alternating load was applied to the stack, because it is assumed that these are more severe condition in respect of cell degradation. In this case, the current density was varied between 10 and 300 mA/cm<sup>2</sup>. In a test run of 3500 h an average cell degradation of less than 7 μV/h was determined. This value is in the range typical of electrode degradation so

that an influence by corrosion (with metallic ions) can be excluded.

The power/volume and power/weight of the 35-cell stack are only 1.8 l/kW and 2.0 kg/kW, respectively (extrapolated to a 100-cell stack with optimised end plates) even with the cooling gap inserted (which renders an external heat exchanger unnecessary).

At the point of design freeze the qualification of a suitable Fe alloy was not yet completed. In order to enable the hardware demonstration within the project period the final

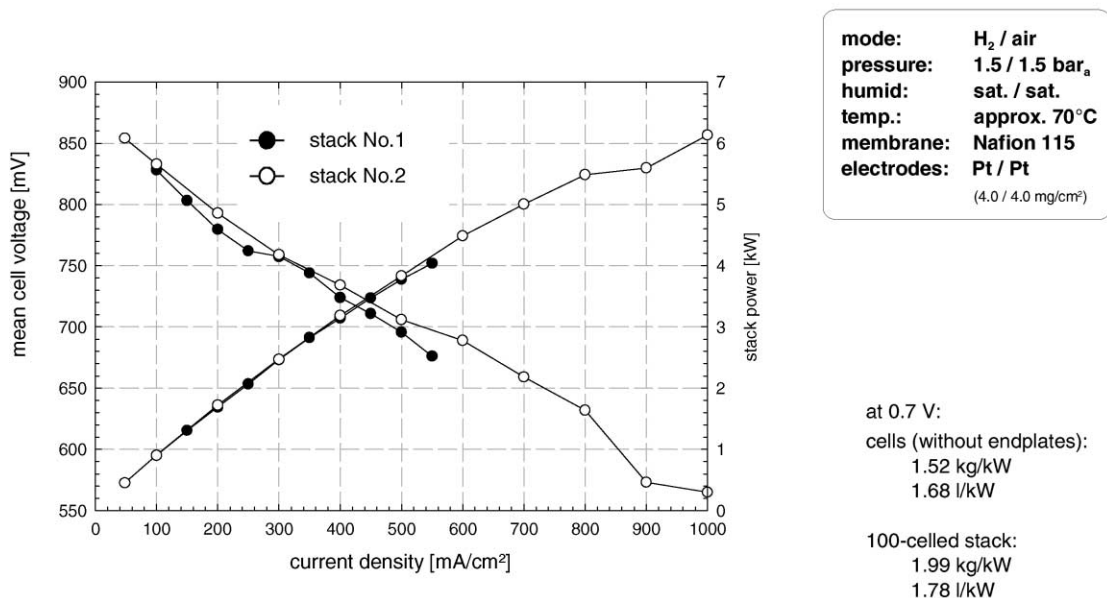


Fig. 7. U–j characteristics and stack power outputs for two different 35-cell stacks.

stack was made with bipolar plates of Ni alloy which still were Au-coated. In order to settle the cost assessment on a confirmed basis, an 8-cell substack with stainless steel and screen printed electrodes was built at the end of the project. The only difference to the earlier stack assemblies was that the mechanical compression had to be increased from approximately 4 to 8 bar. A decrease of approximately 10 mV due to the higher contact resistance can be measured. This is a tolerably low value considering that a tedious and expensive coating can be omitted.

#### 4. Cost considerations

The initially defined cost target of 100–200 ECU/kW can easily be met by the new stack design in combination with alternative materials. Under assumption of the rated performance of  $0.35 \text{ W/cm}^2$  and an automated production of  $10^5$  stacks per year material costs of US\$ 36/kW can be reached. By increasing the power density to  $0.49 \text{ W/cm}^2$  and assuming Pt-recycling the materials costs can be further diminished to US\$ 21/kW. However, current prototypes based on this design will still have material costs around US\$ 2800/kW. The corresponding labour costs will even exceed this value. So the challenge in the future will be to reach high

production numbers even with high-costs at the beginning. For system integration reasons water-cooled types should be developed in a next step.

#### 5. Conclusions

For PEFC a lot of improvements in respect of cost reduction could be realised. By combining all the described single items of cost reduction it is conceivable that stack costs of  $\leq \text{US\$ } 30/\text{kW}$  can be reached. The basis for such an optimistic view is the assumption of an automated mass production with a rate of 1,000,000 units of 50 kW per year. However, the bottleneck for an early market introduction will remain the fuel processing. This still has to be achieved with respect to weight, volume, efficiency, response and start-up time composed to the ICE technology. If this is realised fuel cell vehicles certainly will conquer the road soon.

#### Acknowledgements

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