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IEF-3 Report 2007

From Basic Principles to Complete Systems

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Energy Technology

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Preface

In 2005 and 2006 we successfully continued our application-oriented research into fuel cell technology. The period was characterized by the transition from stack development to system development. After several years of research, it proved possible to upscale the direct methanol fuel cell to a stack size in the range of 2 kW. This size needs a system environment which can only be meaningfully optimized in a practical context. In previous years, light traction had already established itself as an area of application in our analyses, and in 2006 we set up an industry consortium to equip a forklift truck with a direct methanol fuel cell. We were fortunate to have the opportunity to verify the results of our 2006 work at the 2007 Hanover Trade Fair. This is the eighth time in recent years that we have presented this technology and its further developments at the Hanover Trade Fair. This year, for the first time, we were able to show the direct methanol fuel cell drive in a practical application, a Jungheinrich forklift truck. In spite of the considerable progress we have made, several major areas still need to be researched over the coming years. Apart from the cost reduction, the issue of product life is a hot topic.

Interestingly, it is system orientation that leads to the basic research topics, which are chiefly determined by dynamic operation in a real system. In this regard, approval for an analytical laboratory was granted by the Helmholtz Association of German Research Centres (HGF) in 2006. This will allow us to extend our capabilities yet further, from basic principles to whole systems so that we will be able to perform technological procedures, modelling specifications and experimental electrochemical work in close balance.

The analytical laboratory will be mainly devoted to the requirements of direct methanol fuel cells and high-temperature polymer membrane cells, as the corresponding analytical facilities for ceramic fuel cells already exist at Research Centre Jülich. The main difference is the fragility and water content of the layers of the polymer fuel cell in comparison with hard and brittle ceramics. In this laboratory major efforts will therefore concentrate on appropriate preparation methods.

Great advances were made in reformer technology in the period under review. A major breakthrough was the realistic modelling of the mixing chamber of the reformer on the mainframe computer. Achieving a reformer lifetime of 2000 h was also based on highly realistic modelling and innovative design, as was upscaling the reformer from 5 kW to 50 kW, which was verified by using a glass model. The reformer produces an ultrapure reformate throughout its lifetimewith no detectable soot.

Systems technical investigations in the Institute originated with the SOFC. A 20 kW SOFC facility with an innovative design that avoids complex and expensive piping by integrating the hot components of the setup into the stack structure has been built and verified in system tests, and is set to go into operation in 2007. The synergies with direct methanol fuel cells found in systems engineering were exploited successfully, and were extended to include the first system for the high-temperature PEFC.

The Institute will therefore in future become more heavily involved with the DMFC, SOFC and HT-PEFC as well as with middle distillate reformation. One new objective is to develop middle distillate reformation with an HT-PEFC as a system for on-board power supply. As before, we will continue to rigorously pursue our objectives in the SOFC and DMFC field from materials technology up to whole systems.

Jülich, November 2007

Prof. Dr.-Ing. Detlef Stolten

1

Success

Examples of Success

- JuMOVE 2: improved DMFC system
- 20 kW SOFC plant:
  set-up and functional test
- Operation of an autothermal reformer
  with conventional fuels

1.1 JuMOVe 2: Improved DMFC system

The electric-hybrid vehicle JuMOVe (**J**uelich **M**ethanol-**O**perated **V**ehicle), in which the drive energy is provided by a combination of a direct methanol fuel cell (DMFC) and a storage battery, demonstrates the feasibility of using DMFC technology for mobile, dynamic applications on a kW scale.

Fig. 1: DMFC fuel cell system in electric vehicles: JuMOVe from 2005 (left), JuMOVe 2 from 2006 (right)

A very compact construction is required to be able to compete with pure battery-driven systems. The design or packaging must generally be tailored to fit the available space. The 2005 scooter fulfilled this requirement convincingly (see Fig. 1, left) by integrating system components such as the gas separator, pump and liquid container into the fuel cell stack.

An operational analysis of the first-generation system from 2005 revealed that fundamental modifications needed to be made to the whole system in order to meet requirements regarding reliability, cost and lifetime:

- Operation under realistic conditions (cold start, dynamic operation, idling times) led to extremely rapid ageing: after approx. 20 h of operation, maximum stack power had dropped from its original 1.3 kW to about half that value.
- The whole system was not water autonomous, i.e. after a certain time (depending on system load) it had to be refilled with water as well as with methanol.

An analysis of ageing studies yielded valuable information on the reasons for this performance loss (see Chapter 3.1.2.3). The main conclusions of relevance for modifying the operating concept are:

- The stack must not be operated at unlimited load. It is not sufficient to avoid reversing the polarity of individual cells. As a function of the operating temperature, a characteristic map must be implemented in order to limit maximum stack load.
- The anode compartments (or at least the active areas) must be covered with liquid at all times (even when idle) (see Chapter 3.1.2.5).
- Appropriate measures should be taken to limit any contamination of the liquids with substances produced from contact with stack and system materials (e.g. corrosive products) (see Chapter 3.1.2.5).

1.1.1 Hybridization concept and control structure – JuMOVe 2

In JuMOVe the DMFC stack was hybridized with a battery and drive motor via a so-called direct coupling (passive hybrid; see Fig. 2, left), i.e. these three components were connected in parallel without any additional electronics. Only a diode was inserted between the stack and the battery, in order to prevent any current flowing from the battery to the stack (electrolysis). The battery thus determines the scooters's power supply and the stack voltage. Recent findings have shown an excessive load on the stack, particularly during the warm-up phase, leads to an accelerated and irreversible degradation of the cells.

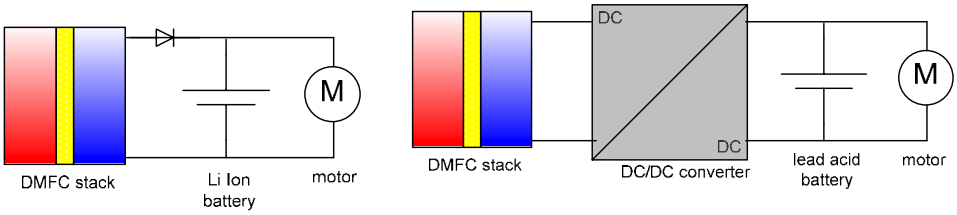


Fig. 2: Hybrid concept JuMOVe (left), JuMOVe 2 (right)

In JuMOVe 2 the stack was actively hybridized (see Fig. 2, right). A DC/DC converter connected between the stack and the battery ensures that the stack can be operated flexibly and the output current can be controlled.

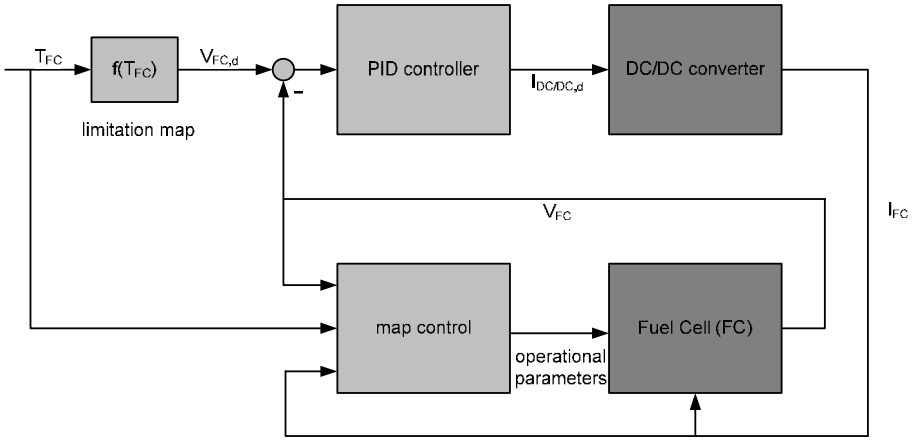


Fig. 3: JuMOVe 2 control structure

In order to protect the fuel cell against accelerated ageing, the fuel cell voltage V_{FC} must not drop below a certain minimum value. This minimum value is stored in the system control as a limitation map: $V_{FC,min} = f(T_{FC})$. This map represents the target value for fuel cell voltage $V_{FC,d}$ for the control structure shown in Fig. 3. This ensures that the fuel cell is operated under optimal and limited load at all times during driving mode. The PID controller adjusts the fuel cell voltage to its target value. The DC/DC converter output current is used as set value. The corresponding fuel cell current is the input parameter for the blocks "mapcontrol" and Fuel

Cell (FC). If the fuel cell is operated at this current, the output value is an actual value for the fuel cell voltage as a function of operating parameters. The PID controller compares the fuel cell voltage with the existing target value. If a difference is observed, the set value is corrected and the loop is repeated.

The objective of map control is to adjust any fuel cell performance that differs from standard values by correcting the operating parameters. The nominal map of a fuel cell without ageing phenomena is stored in the system control. The map describes the current/voltage characteristics as a function of operating parameters. Potential operating parameters are air/fuel ratio, air flow rate, temperature, fuel concentration, fuel mass flow, operating pressure, fuel/air humidification and fuel circulation rate.

1.1.2 JuMOVe 2 water management

In addition to methanol and atmospheric oxygen, the DMFC also requires water as an operation fluid. Water and methanol are converted at the anode into carbon dioxide, protons and electrons. On the other hand, water is created at the cathode as a reaction product – three times as much as is consumed at the anode. Significant water transport also occurs from the anode to the cathode side via the membrane. It is apparent that water management of a DMFC is fairly complex. However, the total water balance is positive (water production), so in general it is possible to build a system that is water autonomous. This is necessary to obtain a high power density and a long operating life.

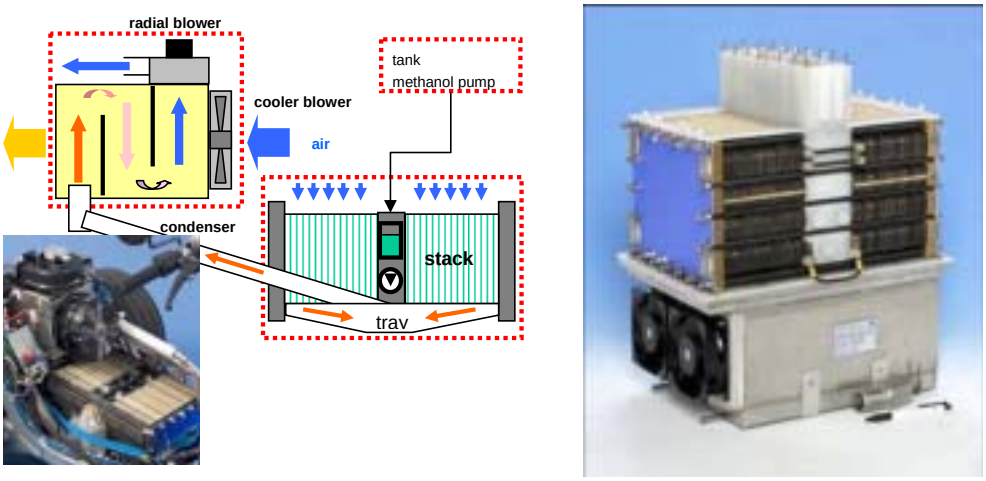


Fig. 4: Stack-condenser connection: JuMOVe (left), JuMOVe 2 (right)

Achieving water autonomy, however, means investing a great deal of time and effort in the process. The cathode air flow removes a substantial part of the water. The positive effect is that this enables heat to be removed from the stack without the need for a separate cooling system. On the other hand, if no further adaptations are made, the whole system is not water autonomous. In JuMOVe and JuMOVe 2, water was recovered by cooling exhaust gases and condensing a part of the contained water (see Chapter 3.1.2.5). Fig. 4 shows the

condensers and how they are coupled to the stack. There are certain similarities with respect to the cooling medium (ambient air) and the flow configuration (cross-counterflow, three direction changes). What is different is the material on the exhaust gas side (coated aluminium in JuMOVe and stainless steel in JuMOVe 2) and the stack connection, which is dictated by the space available (mounted on risers above the stack in JuMOVe and directly under the stack in JuMOVe 2).

The decisive difference between JuMOVe and JuMOVe 2, however, is that only the system of the second-generation device achieves the required water autonomy. This is chiefly thanks to advances in stack development. The stack in JuMOVe had to be driven with a fuel/air ratio of more than 10, whereas the follow-up model can operate with a ratio of 4.

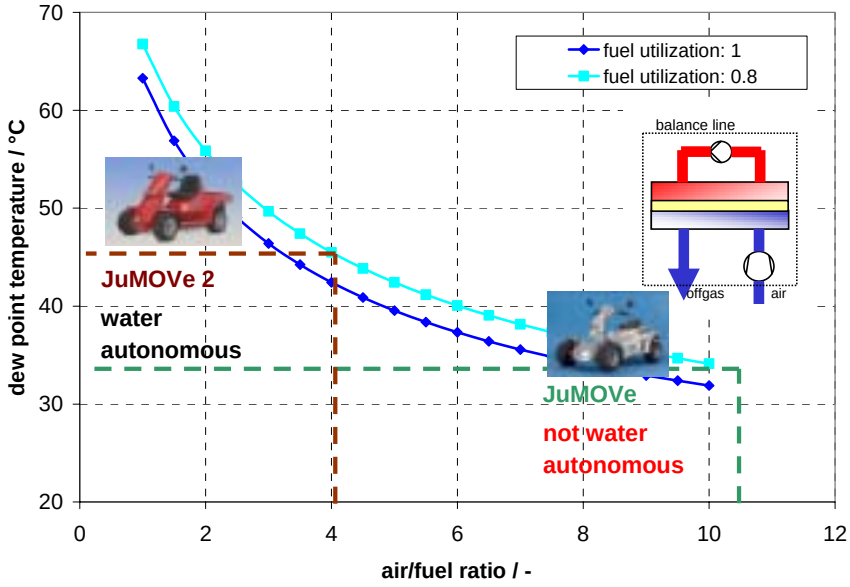


Fig. 5: Limit curves for water-autonomous system operation

Fig. 5 shows how the required system outlet temperature (dew point temperature) changes in order to achieve water autonomy depending on the fuel/air ratio. The diagram results from a simple calculation of how much water is allowed to leave the whole system.

For JuMOVe the exhaust gases must be cooled to approximately 33 °C in order to achieve water autonomy. The required condenser would have been too big for available space. For JuMOVe 2, an exhaust gas temperature of approx. 46 °C is sufficient. The purpose-built condenser in JuMOVe 2 achieves water autonomy for ambient temperatures up to 30 °C.

1.2 20 kW SOFC plant: set-up and functional test

The design work and prelimary tests for the design concept of the 'integrated module', which houses all the components of the system operating at temperatures between 500 °C and 950 °C, were concluded in 2004. In early 2005 all of the manufacturing drawings were produced and the production process for manufacturing four modules of this type was agreed with ZAT-MF.

Steam production was further optimized simultaneously to component manufacture. Using optimized jet nozzles to inject water into the natural gas enables a partial load of as little as 25 % the peak load to be achieved. Modifying the control concept for activating the water injection pump led to a considerable improvement in dynamic behaviour. Whereas with the previous setting a load change took one to two minutes, load jumps now take 18 to 25 seconds. The new configuration also made it possible to save money and space.

The various components of the module were soldered from the individual plates using vacuum solder. The application of the solder should be done via screen printing in order to keep material costs as low as possible and to increase the potential for future large-scale manufacture. The screen printing was performed in IEF-1 and the vacuum soldering in ZAT. The screen thicknesses required were ascertained in preliminary tests and the variation range of the layer thicknesses printed was determined. Various quality assurance measures ensured that a thickness variation of 150 µm ± 30 µm could be achieved, which produced good results in soldering during preliminary tests. For soldering larger components, such as the first air pre-heater, problems were encountered with the vacuum oven since the vacuum could not be maintained owing to the large quantity of binders that had to be outgassed from the soldering paste. Removing the binder in a different vacuum oven from the one used for the actual soldering provided the solution. This enabled the air pre-heater, after-burner and stack dummies to be soldered successfully. The helium leak test in ZAT showed external leak rates in the rate of 10^{-4} to 10^{-6} mbar·l·s^{-1}. Major problems with leak tightness were encountered when soldering the prereformer, due to the very poor bonding strength of the layer of solder. Either the prereformers became permeable immediately after soldering, or leaks appeared during subsequent processing or even during storage. A large number of tests were carried out in order to find the cause of this abnormal behaviour. The main difference between the prereformers and the other components is the additional internals (nickel mesh and anode substrate). Various possible interactions were tested and it as decided to reduce the nickel oxide substrates to nickel prior to soldering in order to avoid any oxygen outgassing, which thermodynamically could not be ruled out completely. This failed to produce any improvement, however. Tests of the gas evolution behaviour of the mesh and substrate carried out in IEF-1 showed small mass losses, although a mass spectrometer failed to detect any suspicious substances in gas extracted during soldering tests at ZAT. New, reconstructed prereformers are now soldered using metallic solder sheets and meshes and substrates that have previously been degassed under soldering conditions. The first step in commissioning the SOFC system can now make use of two improved reformers that have proved to be sufficiently tight in initial tests.

Measurements with the first gas tight air pre-heater in the heat exchange test rig showed a slightly better heat transfer (approx. 78 W/m²K) than calculated, although the pressure loss was some higher, so further improvements were made to the screen printing geometry. One important aspect is the branch pressure during loss while flowing from the distribution holes

into the individual layers, which was confirmed by CFD calculations. This is far more critical for the air pre-heater than for the stack, since the same quantity of gas has to flow through significantly fewer layers (5 instead of 36).

The system will be heated up using electrically heated metal plates, which will each have a power of 2 kW. A design was developed for this in which a meander-shaped heating filament of a specified length was soldered into a steel plate. The soldering was done by the manufacturers of the heating filament (Thermocoax). The position of the two heating plates in the integrated module was determined via CFD calculations. These positions (above and below the after-burner) enable the fuel cell stack to be heated up in a largely homogeneous manner in less than six hours, and simultaneously ensure that the after-burner rapidly becomes hot enough to ignite the residual fuel gases leaving the stack and burn them in a controlled manner. The temperature field after 3 and 5 h heating time is shown in Fig. 6. It can be seen that there is a temperature difference in the stack of less than 50 K in plane and approx. 100 K from top to bottom. Most critical are the gradients in the plane, which are lower during heating than during operation.

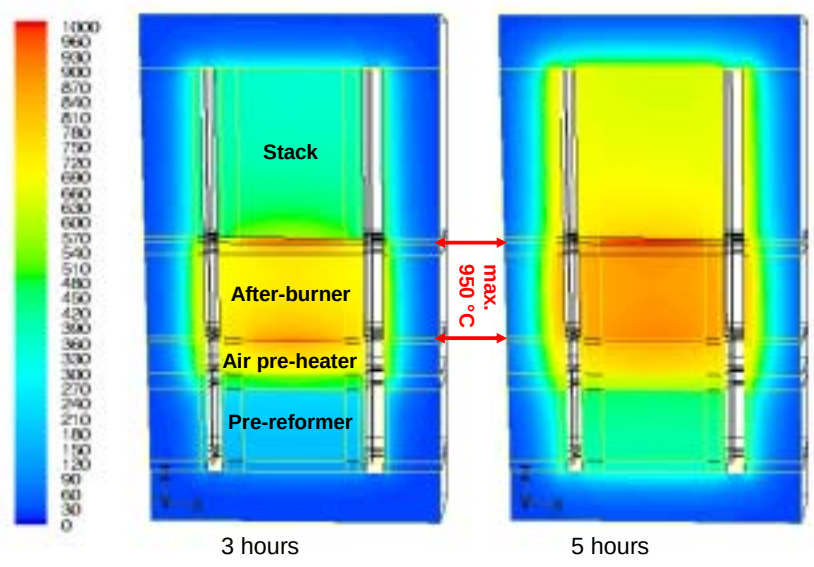


Fig. 6: **Calculated temperature profile inside the integrated module after 3 and 5 h of heating time, with heating plates above and below the after-burner**

Since the system is designed to be run on natural gas, the sulphur, and the odorizing agent THT (tetrahydrothiophene), which contains sulphur, must be removed before the fuel enters the stack. To this end, a special test facility was used to measure the effectiveness of coated activated carbon and a special granulate (from SulphCatch). The latter had a service life 4 times as long as activated carbon and is used in the 20 kW system. The tank size is such that the granulate must be changed after seven weeks' operation at full load. To change the granulate, there is no need to switch off the system – a solenoid valve coupling can be used to switch to a parallel tank.

A detailed 3D drawing was used in the construction of the system. Based on this drawing the process components were fitted into a frame measuring 2.25 x 1 x 2.1 m³, with conventional part of the system (operating temperature under 350 °C) being accommodated in the lower section up to a height of 80 cm. The four integrated modules are installed on an insulating plate 10 cm in thickness and a strain of 500 kg was applied via the insulation. The electrotechnical components, control and data acquisition are installed in a control cabinet measuring 1 x 0.6 x 2.15 m³. On the rear side of this control cabinet the gas supply (valves, controllers) and the desulphurization system are mounted at a depth of 40 cm. Fig. 7 shows the set-up.

Fig. 7: 20 kW SOFC system with two mounted integrated modules

After the set-up had been completely assembled and the whole system thermally insulated, dummy pilot operation with two integrated modules was launched. In the initial heating test the heating plates integrated into the module were heated from room temperature to 800 °C at a ramp of 4 K/min. The temperature evolution of the module components is shown in Fig. 8. After approx. 3.5 h the heating plates were heated to 800 °C, a temperature at which they were expected to continue operation for several hours. After approx. 15 minutes it was observed that the first heating plate had failed. Four additional heating plates proceeded to fail at intervals of a few minutes. The test was therefore abandoned.

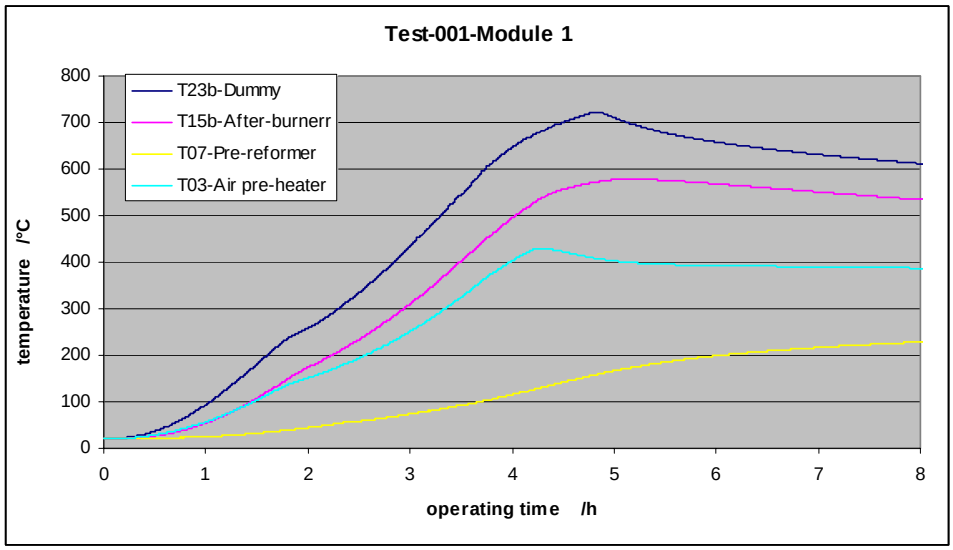


Fig. 8: Temperature evolution of the components in the integrated module during heating

Inspection of the failed heating plates relevated defective heating filaments on three of the plates and defective connections to the supply cable on two plates. A second testing was thus constructed on in which the heating plates were not installed in the system itself. This test confirmed the result of the system. The manufacturer of the heating filaments was contacted. To date the manufacturer has provided no conclusive explanation for the failure of the heating filaments. Possible causes of failure continue to be sought. Parallel to this search, a new design for heating plates involving the use of heating cartridges was developed and an order was placed, so that tests will continue in 2007.

1.3 Operating of an autothermal reformer with conventional fuels

If reforming technologies are to be used in the mobile sector, the long-term stability of the reforming process is a key issue that will determine whether the reforming technology can be developed into a commercial product. Research Centre Jülich's objective in developing the ATR 7 and ATR 8 reactors for autothermal reforming was to show that considerable progress had been made and that a reforming device was available to convert Ultimate Diesel and Jet A-1 into a hydrogen-rich gas mixture for use in mobile fuel cell applications. Such use generally requires a lifetime of approx. 5000 h.

1.3.1 Conventional fuels from middle distillates

In order to use fuel cell technology for APU applications, fuels are required that can be used in drive units on board of vehicles, ships and aircraft. In the transport sector, approx. 94 % of the liquid energy sources used are produced from crude oil i.e. gasoline, diesel and kerosene. The properties of gasoline and diesel fuels are laid down in the DIN EN 228 and DIN EN 590 standards. In the first stage of crude oil processing in refineries, the oil is broken down into separate distillation fractions via atmospheric distillation. Around 40 % of the refinery products sold in Germany are middle distillates, and include kerosene, diesel and EL heating oil (EL: Extra Light). To make the optimum use of crude oil as an energy source, the high-boiling fractions are broken down 'cracked' thermally and chemically, and added to the other fractions. Typical fractions are gasoline – raw gasoline is referred to as naphtha –, with a boiling point of 30 - 180 °C; kerosene, with a boiling point of 150 - 250 °C; and diesel, which boils at 250 - 360 °C. Not all of the properties of the fuels listed above are of relevance for fuel cell technology. Raw gasoline contains between 2 % (light naphtha) and 20 % (heavy naphtha) aromatics such as benzene, toluene and xylene. In order to raise the knocking properties of petrol to octane values higher than RON 95, the raw gasoline is converted chemically. Gasoline can contain up to 40 % aromatics and 15 % olefins. These substances are important intermediate products during soot formation and their poorer reforming characteristics have been widely documented. Commercial diesel fuels contain a maximum of 30 % aromatics, 25 % monoaromatics, 10 % diaromatics and 3 % polyaromatic compounds. They contain a maximum of 5 % olefins and an ash content of 100 ppm. The boiling behaviour and the so-called evaporation residues play an important role in determining the substance's evaporation properties in a reformer (see Fig. 9).

For diesel this residue amounts to a maxiumum of 5 % of total mass. Premium-quality diesels are already available from two manufacturers in the German infrastructure today. Dry residue is less than 1 % at 400 °C; the sulphur content is less than 10 ppm and the ash content is only 2 - 10 ppm. The aromatic content is also reduced to about 15 %, and the percentage of diaromatics in the fuel is also less than 0.5 %. With the exception of its boiling point and its high permissible sulphur content of 3000 ppm, kerosene displays similar chemical properties: approx. 15 - 20 % aromatics, an evaporation residue of 1 % and an olefin content of less then 1 %. Half of the remaining compounds – which form around 80 % of the total – are simply ring-shaped, and the other half are long-chain alkanes.

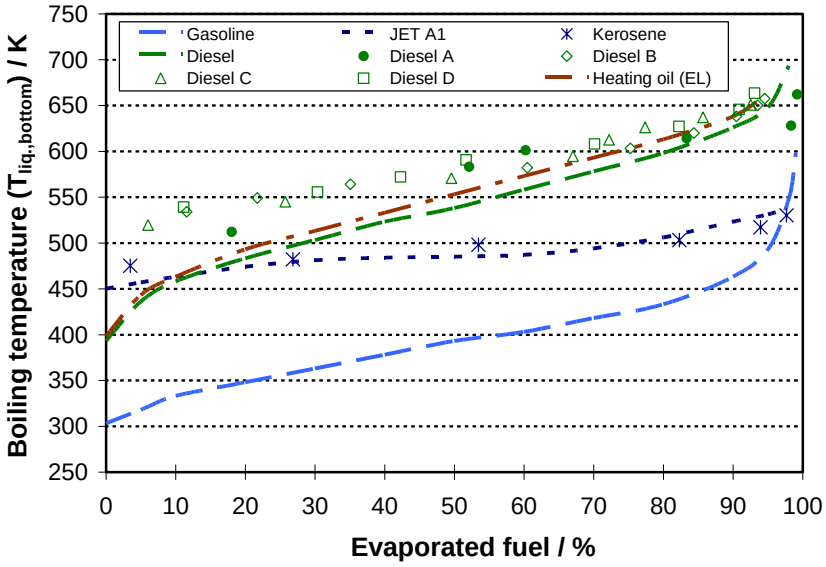


Fig. 9: **Boiling behaviour of conventional diesel fuels, gasoline and diesel. Lines represent information taken from the literature and dots our own measurements**

Many developers use diesel fuels for testing that do not contain aromatics. In IEF-3 the diesel fuels available in the infrastructure, such as ARAL Diesel Ultimate™, are used. The only way to obtain low-sulphur kerosene is direct from the refinery. This fuel is different from the JET A1 fuel used in aviation because it does not contain any of the additives the latter contains, such as antioxidants, anti-icing agents for metal deactivators. A mixture of additives is added to the kerosene used in IEF-3.

1.3.2 Long-term behaviour

Fig. 10 shows the hydrogen concentrations in the ATR 5, ATR 7 and ATR 8 reactors during the autothermal reforming of desulphurized Jet A-1 over time. In ATR 5 the H_2 concentration dropped from 36 vol.% to 32.5 vol.% within only 90 h. At the same time, the quantity of total organic compounds (TOC) in the unconverted, condensed water rose (cf. Tab. 1) from 13.6 to 64.4 mg/l. Furthermore, benzene and C_3-, C_4- and C_5-olefins were detected in the reformate. From the literature it is known that olefins are precursors to the formation of carbonaceous deposits. These results mean that conversion of Jet A-1 decreased during the 90 h of this experiment. ATR 7 was considerably more stable over the long term. Fig. 10 illustrates that the H_2 concentration remained constant at values of between 36.5 vol.% and 37 vol.% over a period of 500 h. The results in Tab. 1 support this observation. The TOC values are, with the exception of the value directly after the start of the experiment, below the value for demineralized water (2.0 mg/l). No other by-products were detected. This means that conversion of Jet A-1 was 100 % for the entire duration of the test. Unfortunately, the load of ATR 7 dropped during the test from 100 % at the start to 65 % at the end. This was a continuous, linear process. Since the nozzle in ATR 7 no longer functioned properly at loads

of less than 65 %, the test had to be ended after 500 h. The same experiment was then begun on ATR 8. For the first 1000 h of this experiment, the load was maintained at approximately 100 %. Following an emergency shutdown and also a planned shutdow, the nozzle of ATR 8 became completely blocked, so no further Jet A-1 could be supplied. Analytical measuring methods (scanning electron microscopy, energy dispersive X-ray analysis) showed that these deposits were primarily composed of carbon. The deposits probably formed via pyrolysis during the two shutdowns. In ATR 8, however, as opposed to ATR 7, the nozzle can be replaced. After the nozzle was replaced, the load returned to 100 % and remained fairly constant until the experiment was ended after approx. 2000 h.

Fig. 10 shows that the H_2 concentration in ATR 8 fell continuously and linearly during the first 1130 h from 36.9 vol.% to 32.7 vol.%. It then increased again to 33.8 vol. % and remained constant for approx. 400 h.This rise and the period where the H_2 concentration remained constant are attributable to the fact that 1130 h into the experiment, a different kerosene with a lower content of aromatics was used. Towards the end of the experiment, however, the H_2 concentration dropped once again. In addition to the H_2 concentration, Fig. 10 shows the total of the CO and H_2 concentration for ATR 8. This concentration remained approximately constant for the first 1500 h. This result suggests that the drop in the concentration of H_2 during the first 1130 h can be explained by a decreased level of catalyst activity for the water gas shift reaction. Unfortunately the total of the CO and H_2 concentration also fell towards the end of the experiment, which leads to the conclusion that catalyst activity for the actual reforming reaction also decreased.

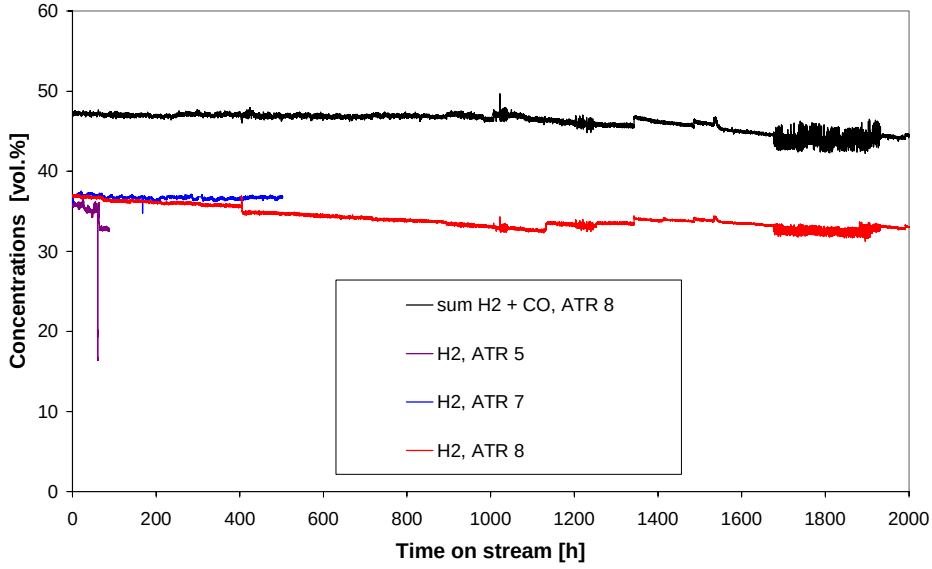


Fig. 10: **H_2 concentrations during the autothermal reforming of desulphurized Jet A-1 in the ATR 5, ATR 7 and ATR 8 reactors, total of the H_2 and CO concentration in ATR 8**

ATR 5		ATR 7		ATR 8	
Time [h]	TOC [mg/l]	Time [h]	TOC [mg/l]	Time [h]	TOC [mg/l]
23	13.6	6	5.5	2	1.7
44	14.0	95	0.7	138	0.6
65	20.5	239	0.6	211	0.7
81	40.5	311	0.5	304	0.9
86	64.4	407	0.5	401	1.7
		500	0.6	497	4.3
				640	22.6
				711	29.8
				804	41.2
				893	26.8
				1005	75.3
				1101	69.7
				1245	139
				1299	128
				1411	105
				1506	116
				1602	115
				1700	126
				1843	131
				1915	108
				2007	103

Tab. 1: **Quantity of total organic compounds (TOC) in the unconverted, condensed water from the autothermal reforming of Jet A-1**

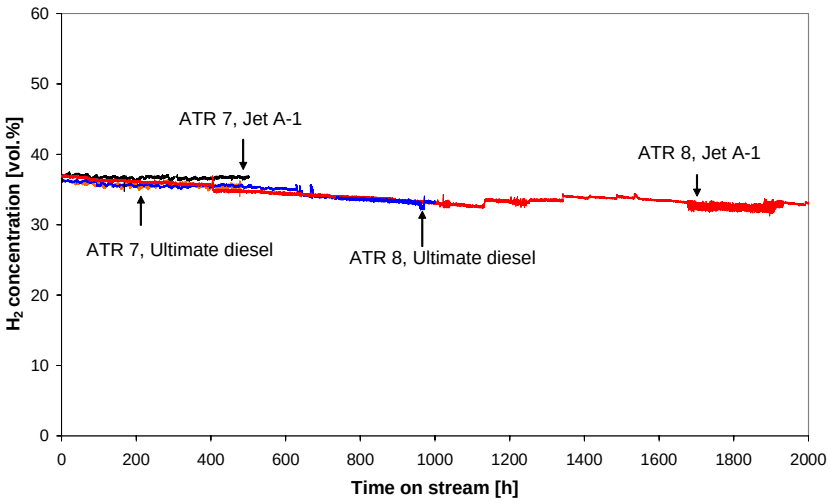


Fig. 11: **H_2 concentrations during the autothermic reforming of desulphurized Jet A-1 and Ultimate Diesel in the ATR 7 and ATR 8 reactors**

The TOC values from Tab. 1, which rose from approx. 1 mg/l at the beginning to approx. 100 mg/l – 130 mg/l, support this conclusion. Furthermore, an oil phase was observed on the condensed water during the second half of the experiment. This is evidence of unconverted Jet A-1. A gravimetric analysis of the mass of the oil phase revealed that the conversion of Jet A-1 still remained at 99 % after 2000 h. Fig. 11 shows a comparison of the H_2 concentrations for the autothermal reforming of Jet A-1 and Ultimate Diesel in the ATR 7 and ATR 8 reactors. This clearly demonstrates that comparable long-term stability was achieved for both fuels.

1.3.3 Determining conversion

As a general rule, the carbon budget is used to determine the conversion. On the product side, the carbon budget contains the desired products carbon monoxide (CO), carbon dioxide (CO_2) as well as unconverted or only partially converted hydrocarbons. The formation of methane (CH_4) is inevitable owing to the position of the chemical equilibrium and this is counted as a product. Calculating conversion on the basis of the products formed leads to significant errors at high conversions (>95 %), so residual hydrocarbons should be determined for the evaluation.

Carbon input = carbon in product gas + residual carbon

$$N \cdot \dot{n}_{C_N H_M}^{Ein} = \frac{\dot{V}_{tr}^{G} p}{R \cdot T} \cdot \left(y_{CO}^{MS} + y_{CO_2}^{MS} + y_{CH_4}^{MS} \right) + \frac{\dot{V}^{G} p}{R \cdot T} \sum_j \left(y_j \cdot N_j \right)$$

The residual hydrocarbons are analyzed in a maximum of three material flows. In the gas phase, highly volatile by-products – C_2 - C_7 hydrocarbons – are detected via GC/MS coupling. Once the product gas has cooled, a condensate forms, which at high conversions only consists of anaqueous phase and the liquid hydrocarbons dissaved in it. At lower conversions an organic liquid phase (OP) is formed, which first becomes visible as a separate phase by the formation of flowmarks during stirring and the appereance of spots of grease. Conversion is calculated using the measurement data according to

$$\xi_{OC} = 1 - \left(\sum_j \left(y_j^{GC-MS} \cdot N_j \right) \cdot \frac{\dot{V}_{tr}^{G} p}{R \cdot T} + N_{fOP} \frac{\dot{m}_{(C_N H_M)_{OP}}^{OP}}{M_{(C_N H_M)_{OP}}} + TOC \cdot \frac{\dot{V}_{WP}}{M_c} \right) \Bigg/ N \cdot \frac{\dot{m}_{C_N H_M}^{Ein}}{M_{C_N H_M}}$$

The distribution of residual hydrocarbons across the phases depends on the composition and thermodynamic properties of the individual components. Short-chain hydrocarbons like butadiene, propene and benzene remain in the gas phase after cooling to approx. 15 °C, whereas long-chain molecules liquefy. The solubility of these components will determine whether an organic phase occurs. An aromatic such as orthoxylene (C_8H_{10}) reaches a solubility of 173 mg/l at 25 °C (TOC: 1384 mg/l), whereas ring-shaped compounds like cyclooctane (C_8H_{16}) reach a solubility of 8 mg/l and iso- and n-octane (C_8H_{18}) 2 and 0.7 mg/l respectively.

In the product gas, only approx. 55 ppm of toluene, 25 ppm of butadiene and 13 ppm benzene have been found so far after 1000 h in older reactors (ATR 5) and in an ATR 8 reactor following a short operating stage running on ARAL Ultimate Diesel at extremely unfavourable mixing ratios. Together with a TOC value of 130 mg/l, this gives a conversion of

99.7 %, with hydrocarbons in the gas phase dominating conversion. Although mass signals from individual components can be detected at under 1 ppm, they can no longer be distinguished from the basic signal. Assuming a C concentration of 3 ppm in the gas phase, a conversion of 99.999 % can be calculated.

The precision of the TOC measurement, on the other hand, – 0.1 mg C/l –, is much greater. It must be noted that when taking measurements of pure water, the samples should be irradiated with ultraviolet light in order to prevent nucleation and the formation of organic carbon. Untreated water displays TOC values of approx. 2 mg/l. After type ATR 7 reactors were run on ARAL Ultimate Diesel and low-sulphur kerosene, 2 and 0.6 mg/l of organically bound carbon, respectively, were detected after 500 h. After 1100 h of running an ATR Type 8 on low-sulphur kerosene a TOC value of 70 mg/l was measured. Since no peaks were observed in the GC/MS and no organic phase in the condensate, a conversion of 99.99 % can be calculated.

After 1100 h of continuous stationary operation on low-sulphur kerosene, first organic phases appeared on the surface of the aqueous condensate. After 1800 h and 1900 h it was possible to measure the conversion gravimetrically. Conversions of between 98.2 % and 98.8 % were determined. The correction of the quotient of carbon atoms in the molecule to the molar mass between kerosene and the separated organic phase is negligible. The measurements can only be evaluated above an absorbed mass per hour of 5 g/h in the organic phase and 2000 g/h in the aqueous phase. This represents a maximum conversion of 99.5 % for this method of determination.

Tests during system operation will demonstrate which conversions are required to avoid of ageing phenomena in the gas treatment and the fuel cells.

1.3.4 Benchmark

To classify investigations into the degradation behaviour of ATR 5, ATR 7 and ATR 8 reformers during operation with conventional fuels, published data from peer-reviewed journals, conference proceedings and internet pages were collated and analyzed. A complete benchmark covers various performance characteristics, e.g. power class, lifetime, conversion behaviour, product range, power density, power-to-weight ratio, start-up time, response times during load changes etc. Fig. 12 shows an international comparison of the values for residual hydrocarbons, operating time, power and power density. For many developments only a few results exist, and the boundary conditions in terms of reforming behaviour and fuel used are sometimes different. In the diagram on the right, a micro steam reformer run on isooctane can achieve the power densities of an ATR 5/ ATR 8, i.e. between 1.5 - 2 kW_e/l. Reformers that work according to the partial oxidation principle are very compact. No performance data for these reformers are known.

The conversions published for POX reformers when run on kerosene and diesel are in the range of 95 - 90 %. Autothermal reformers achieve conversions of between 98 % for a fresh catalyst and 92 % after 300 h. Japanese researchers are developing systems for use in domestic heating technology based on steam reforming of kerosene that have operating times as long as 8000 h and impressive conversions of 98 %.

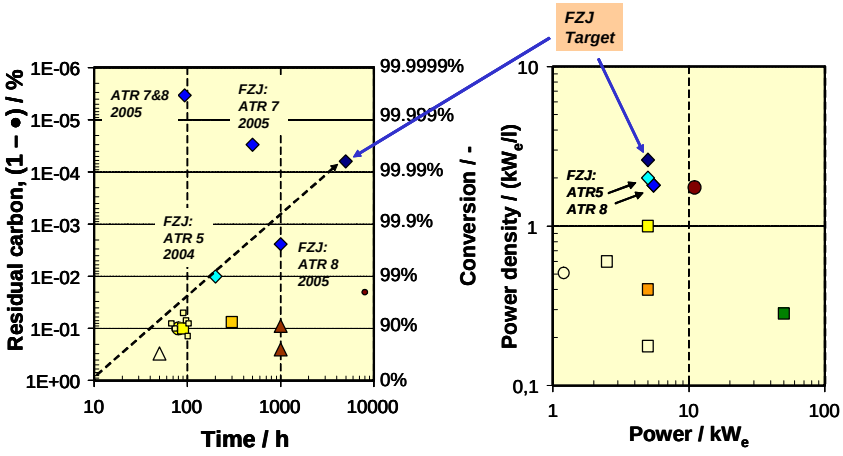


Fig. 12: **Benchmark for reformer development in IEF-3 with regard to conversion, or residual hydrocarbons in the product gas, operating time, power and power density**

The development carried out in IEF-3 is intended to achieve conversions of 99.99 % after 5000 h operating time. Current measurements on reactors of the ATR 8 type show conversions of greater than 99.7 % after 1000 h with ARAL Diesel Ultimate and 99 % for low-sulphur kerosene after 2000 h.

2

Education

Education and Training

- Fuel cell training and demonstration centre
- Staff teaching at universities

2.1 Fuel cell training and demonstration centre

After the conclusion of the 'Fuel Cell Training and Demonstration Centre' project promoted by the Federal Ministry of Economics and Technology (BMWi) and the North Rhine-Westphalia Federal Ministry of Education and Labour (MWA), a number of different training schemes were funded by IEF-3.

2.1.1 Organization of seminars, practical training, information events and guided tours

As part of our collaboration with local schools Aachen University of Applied Sciences - Jülich Section Vocatinal Training Centre and the Corporate Communications division of Research Centre Jülich, numerous events were organized for pupils, teachers, students and visitors to IEF-3. These events involved the following activities in particular

- IHK Aachen and KvK Maastricht: two fuel cell seminars, each attended by around 35 participants, were organized for members of the two regional chambers of industry and commerce, at which the potential for technology transfer from research to the industry was demonstrated.
- Schools in North Rhine-Westphalia: three fuel cell seminars for pupils and students were organized in 2005, attended by a total of 55 participants, as an introduction to the basic principles and applications of fuel cell technology.
- Research Centre Jülich Schools Laboratory: involvement in developing, setting up and carrying out experiments on fuel cell technology
- Seminar for pupils and teachers (20 people) from Immanuel Kant Grammar School, Dortmund, on the topic of 'Fuel Cell Systems and Hydrogen'
- Practical solar training for students of of Aachen University of Applied Sciences, Jülich Section, on the topic of 'Electrolysis and Fuel Cells' (7 students in 2005 and 11 students in 2006)
- Vocational orientation and job experience for pupils from local schools on the topic of 'specialist Careers in Fuel Cell Technology'
- Information events and guided tours open to all (over 40 events with a total of almost 1000 participants in 2006) on the topic of 'Fuel Cell Technology'
- Multimedia learning programme 'Fuel Cells' presented in Research Centre Jülich as part of Open Day 2006

Fig. 13: Visitors on Open Day view exhibits at IEF-3

2.1.2 Involvement in external events

IEF-3 employees were invited to several external events to give a general introduction to 'Fuel Cell Technology' or to speak about certain specialized fields in the subject:

- Specialist seminar for interested parties involved in the research, development and manufacture of fuel cell systems on the topic 'Simulation of Fuel Cell Systems' as part of events organized by the Fuel Cell Education and Training Centre Ulm (WBzU), with 16 participants
- Specialist seminar for interested parties involved in the research, development and manufacture of fuel cell systems on the topic of 'Small Fuel Cell Drives – An Attractive Market Niche?!' as part of events run by the WBzU with 16 participants
- Tutorial for newly qualified employees and other interested in the scientific and technical aspects on the topic of 'Fuel Cells' as part of the Lucerne Fuel Cell Forum 2006
- Lecture topic for experts and others interested on the topic of 'Fuel Cell Technology' as part of the Energy Congress during the Aachen Energy Days in 2006
- Multimedia learning programme 'Fuel Cells' presented as part of the 2006 annual meeting of the Solar Energy Association (FVS) and Aachen Energy Days 2006

2.1.3 Lectures on project status

During specialist industry conferences and information events designed to promote cooperation, the results of the 'Fuel Cell Training and Demonstration Centre for Fuel Cells' project were presented to a wide audience. Depending on the nature of the event, questions were answered on educational aspects of 'Fuel Cell Technology', and the future needs of various sectors were discussed.

- Presentation of the results of the FZJ and WBzU project 'Hydrogen and Fuel Cells: Education and Training Prior to Market Introduction“ as part of the 3rd International German Hydrogen Energy Congress 2006 in Essen
- Presentation of the project results as part of a start-up meeting and orientation discussions with representatives from RAG Bildung, the Confederation of German Trade Unions (DGB), the Town of Herten and the HyChain Minitrans project

2.1.4 Collaboration with other organizations

Designing, launching and running education and training measures and qualification programmes on the topic of 'Fuel Cell Technology' are increasingly becoming the focus of attention both of manufacturing industry and the relevant educational establishments. To meet this demand, special initiatives have been launched combining specialist technical knowledge and existing opportunities for collaboration.

- Fuel Cell Education and Training Centre Ulm (WBzU), Heinz Piest Trade Technology Institute (HPI) at the University of Hanover, Fraunhofer Institute for Systems and Innovation Research (FhG-ISI) and Ludwig-Bölkow-Systemtechnik GmbH (LBSt):
  a. Founded the Fuel Cell Education Network (BZ-BNW)
  b. Launched a 'Fuel Cell Education Network' internet portal
  c. Organized and ran a series of events

d. Drew up the master plan for 'Education and Training in Fuel Cells'

- Federal Technology Centre for Electrical Engineering and Information Technology e.V. (bfe), Chair of Electrical Power Supply at the University of Dortmund and the Aachen University of Applied Sciences, Jülich Section:
  a. Prepared and provided lectures for teaching modules as a subcontract
  b. Prepared and provided multimedia teaching units and classroom teaching units (bfe only) as a subcontract
- Practical Vocational Training Research Group (FPB) of the University of Bremen, Vaillant GmbH, Fuel Cells Initiative (IBZ) and the Federal Vocational Training Institute (BIBB):
  a. Determined the need for information and qualifications for applying fuel cells in domestic engineering and published the results in the form of a brochure
  b. Founded a 'Fuel Cells in Domestic Engineering' working group in order to promote cooperation
  c. Founded the Fuel Cells Qualification Initiative (IQ-BZ), an association handles the need for information and qualifications for fuel cell and hydrogen technologies
- The 'Fuel Cell Education and Training Centre Ulm (WBzU)' is involved as a partner and is pursuing similar objectives. This shared focus is marked by an extensive exchange of information and constructive cooperation between the partners
- The project 'New Demands Made on Technicians by Fuel Cell Innovations' was supported by demonstration materials, discussion papers and information in collaboration with BZ-BNW
- European Hydrogen and Fuel Cell Technology Platform – Group Education and Training Initiative: Involvement as the deputy leader of the 'Craft and Industry' working group in the formulation of a European action plan for the 7th EC Framework Programme for the 2007 - 2011 period
- Hydrogen and Fuel Cells Expert Council (HYBERT): Involvement in the HYBERT co-ordination group, advising and exchanging information with the government departments involved
- International Partnership for the Hydrogen Economy (IPHE): Education Task Force: Supported the IPHE's education and training activities concerning hydrogen and fuel cells
- Involvement in the Fuel Cells Qualification Initiative (IQ-BZ), working to implement information and qualification schemes for fuel cells and hydrogen technologies
- Preparation of a project proposal for support of the Fuel Cells Qualification Initiative (IQ-BZ) to implement the results of the activities carried out by the 'Education and Training/Public Relations' working group of the Hydrogen and Fuel Cells Strategic Council
- Presentation of the IQ-BZ project proposal to representatives of the BMBF in Bonn and discussion of possible funding. Due to the different nature of their responsibilities and objectives, non of the BMBF departments present, 'New Media in Education', 'Vocational Training' and 'Microsystems Technology', would be able to fund the project

- Prepared a proposal for a project outline on 'Training towards Qualifications in Fuel Cells' for the North Rhine-Westphalia Fuel Cells Measures and Hydrogen Network of Expertise
- Continuation of the activities of the 'Fuel Cells Training Network' consortium and continuing maintenance of the joint internet portal
- Promotion and sale of a 'Fuel Cells' CD-ROM produced by the Federal Technology Centre for Electrical Engineering and Information Technology e.V., Oldenburg, and Vogel Industrie Medien GmbH, Würzburg, designed to provide information, raise the profile of fuel cells and promote training. 285 copies of the CD-ROM have been sold since it came out in 2004
- Adapted existing teaching modules to customer requirements
- Supported and participated with Training and Competence Centres (WBzU and RAG Bildung) in the planning and execution of joint training events the respective Centres.
- Preparation for the nomination of and cooperation with actors from the planned Qualification and Information Centre for Fuel Cell Technology in Herten

2.2 Staff teaching at universities

As part of the IEF-3 approach, scientists at the Institute also teach at universities in parallel to their research and development work. In accordance with the Jülich model, IEF-3 has one joint appointment of a professor at RTWH Aachen University (Prof.Dr.-Ing. Detlef Stolten) and two professorships at Aachen University of Applied Sciences, Jülich Section (Prof. Dipl.-Ing. Ludger Blum and Prof. Dr.-Ing. Ralf Peters). In addition, other members have teaching commitments at the University of Bochum (Dr. Ing. Andreas Gubner and Prof. Dr.-Ing. Ralf Peters) and the University of Ulm (Prof. Dr. rer. nat. Eckhard Spohr).

Fig. 14: Universities where IEF-3 staff have teaching duties

2.2.1 Courses taught by professors

Name	Subject	Type/extent	semester	University
Prof. L. Blum	Fuel Cells – The Future for Dispersed Power Supply!?	V/2	WS	Aachen Univ. of Applied Sciences, Jülich Section
		V/2	SS	
Prof. Dr. R. Peters	Basics and applications of chemical reaction theory – simulation of dynamic processes in energy systems with MATLAB/Simulink	V/2 Ü/2	WS	Aachen Univ. of Applied Sciences, Jülich Section
Prof. Dr. D. Stolten	Basic principles and technology of fuel cells	V/2 Ü/2	WS	RWTH Aachen

2.2.1.1 Fuel Cells – The Future for Dispersed Power Supply?

Prof. Ludger Blum teaches the specialist subject Fuel Cell Technology at Aachen University of Applied Sciences, Jülich Section. The optional subject 'Fuel Cells for Stationary Applications' in the undergraduate course in Energy and Environmental Technology and the graduate course in Energy Systems covers the function, construction, behaviour, advantages and disadvantages of the different types of fuel cells and teaches the process engineering for designing fuel cell systems. The topics include: basic principles of fuel cells; fuel gas supply; efficiency; function and construction of different types of fuel cell; fuel cell system requirements; process engineering of various fuel cell systems for different applications; energy balance of a fuel cell system; and modern plant engineering. An average of 12 students take the course each semester.

2.2.1.2 Basics and applications of chemical reaction theory – simulation of dynamic processes in energy systems with MATLAB/Simulink

Prof. Dr.-Ing. Ralf Peters teaches the specialist subject Energy Process Engineering at Aachen University of Applied Sciences, Jülich Section. The subject of "Basics and Applications of Chemical Reaction Theory – Simulation of Dynamic Processes in Energy Systems with Matlab/Simulink“ links the basic principles of chemical process engineering and dynamic simulations of reaction apparatus. The lectures and practical exercises take fuel processing and fuel cell system technology for methanol fuel cell drive systems and for diesel-based on-board power supply as examples. The course is compulsory for the 30 - 40 students taking the Master of Science in Energy Systems course.

2.2.1.3 Basic principles and technology of fuel cells

Prof. Dr.-Ing. Detlef Stolten holds the Chair for Fuel Cells at RWTH Aachen. The courses offered deal with converting renewable and fossil energy carriers for use in fuel cells for portable, stationary and mobile applications. The process engineering and systems technology cover high-temperature and low-temperature fuel cells and fuel processing systems. These elements are accompanied by a discussion of the basic physical and chemical principles involved. Systems analysis of the process engineering including how to estimate costs, round off the coverage of the subject with a view to future market launch. During the 2006/2007 winter semester just under 50 students attended the lectures and exercises. As part of the existing cooperation with Research Centre Jülich, the Chair offers students the chance to work on term projects and diploma dissertations and also gives them the opportunity to work in projects as research assistants.

2.2.2 Courses taught by university lecturers

Name	Subject	Type/extent	semester	University
Dr. B. Emonts	Hydrogen and its Conversion via Electrolysis and the Fuel Cell Process	V/2	WS	Aachen Univ. of Applied Sciences, Jülich Section
	Elecrolysers and Fuel Cells	P/4	SS	Aachen Univ. of Applied Sciences, Jülich Section
Th. Grube	Basics and Applications of Chemical Reaction Theory – Simulation of Dynamic Processes in Energy Systems with Matlab/Simulink	Ü/2	WS	Aachen Univ. of Applied Sciences, Jülich Section
Dr. A. Gubner	Fuel Cell Applications: High-Temperature Fuel Cells	V1,5	WS	Ruhr-University, Bochum
Prof. Dr. R. Peters	Fuel Cell Applications: Low-Temperature Fuel Cells	V1,5	WS	Ruhr-University, Bochum
Prof. Dr. E. Spohr	Theoretical and Computer Chemistry	V/2	WS	University of Ulm
		V/2	SS	

2.2.2.1 Hydrogen and its Conversion via Electrolysis and the Fuel Cell Process

Dr.-Ing. Bernd Emonts lectures at Aachen University of Applied Sciences, Jülich Section, on the subject of Electrolysers and Fuel Cells and hold a four-hour practical for small groups (5 - 10 students). This course provides an introduction to the basic electrochemical principles of both conversion processes, explains the design of different types of device and presents examples of proven applications. The subsequent practical exercise is carried out under supervision at IEF-3 on an energy storage system with pressure electrolyser and PE fuel cell and pressure storages for hydrogen and oxygen. During this practical the main operating parameters are recorded by the students, who then determine the operating characteristics and the device efficiency.

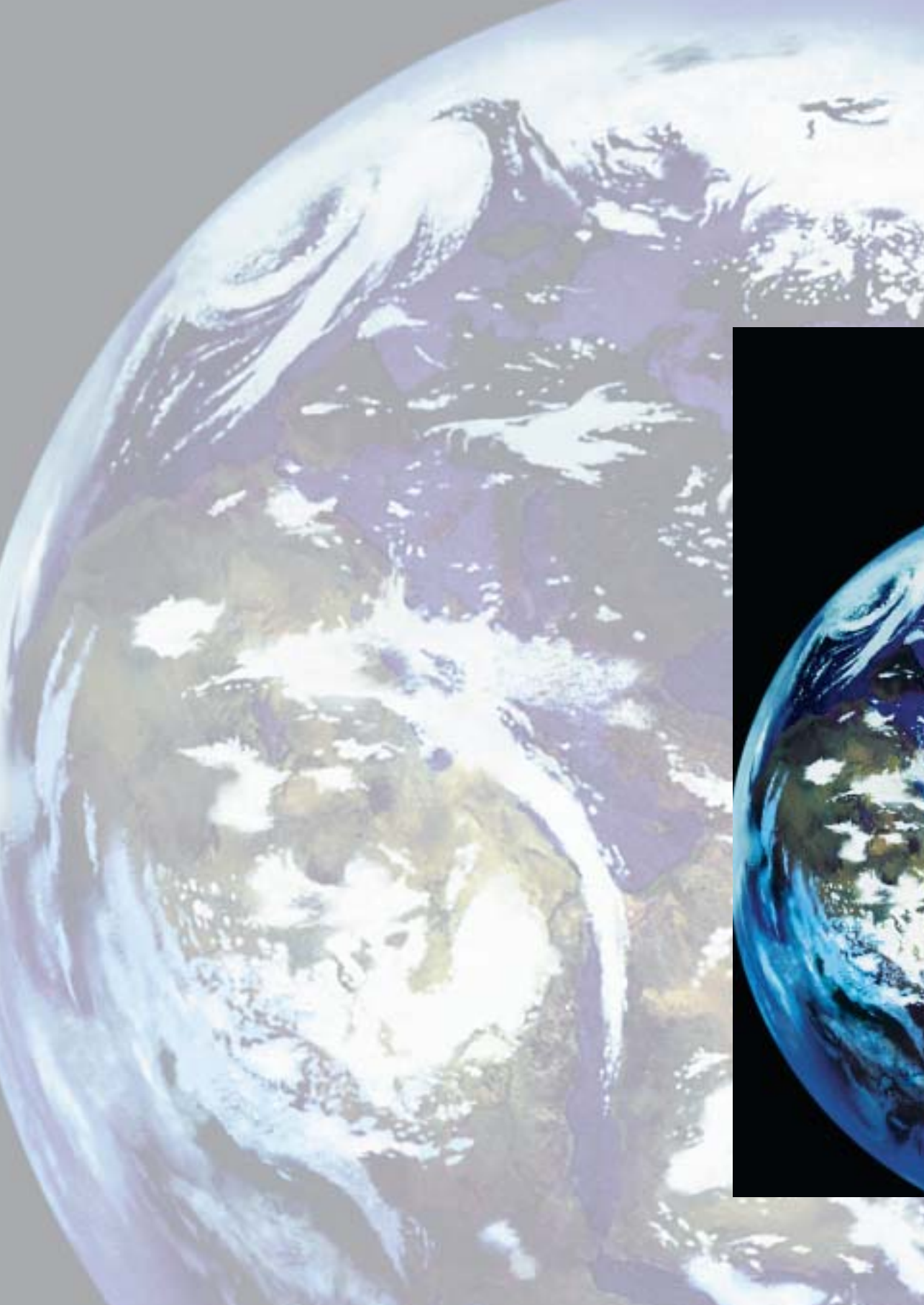
2.2.2.2 Fuel Cell Applications: High- and Low-Temperature Fuel Cells

Prof. Dr.-Ing. Ralf Peters und Dr. Andreas Gubner lecture at the Faculty of Mechanical Engineering at the University of Bochum on the subject of 'Fuel Cell Applications: High- and Low-Temperature Fuel Cells' as part of the course for advanced students (5 - 10 students). Following a comprehensive introduction to fuel cell technology and the global energy situation, the basic physical and chemical principles of electrochemistry are dealt with. The basic technical principles chiefly cover heat and mass transport in cells, stacks and peripherals. An examination of fuel cell systems then leads onto reforming techniques. These

basic sections are followed by a detailed discussion of special fuel cell types and their applications. The lecture ends by examining and analysing potential energy carriers for fuel cells and by discussing some of the economic aspects involved.

2.2.2.3 Theoretical and Computer Chemistry

Prof. Dr. Eckhard Spohr gives special lectures to small groups of 5 - 10 PhD students in theoretical and computer chemistry at the University of Ulm on the subjects of computer simulation, statistical mechanics and liquid state theory.



3

Reports

Scientific and Technical Reports

- Key topic: polymer electrolyte fuel cells
- Key topic: solid oxide fuel cells
- Key topic: fuel processing systems
- Cross-cutting topic: process and systems analysis
- Cross-cutting topic: analysis
- Cross-cutting topic: quality management

3.1 Key topic: polymer electrolyte fuel cells

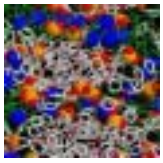
3.1.1 Objectives and fields of activity

In view of the fact the background that the use of direct methanol fuel cells (DMFCs) as a replacement for lead acid batteries in small, mobile applications, i.e. in the “material handling” sector appears interesting in the short to medium term, IEF-3 focus on researching and developing these fuel cells. Work ranges from investigating the electrochemical reactions on optimized electrode structures, characterizing new membranes, optimizing cell components such as the membrane electrode assembly (MEA) or the flow-field structure and the suitable manufacturing methods and characterizing MEAs up to and including cell and stack development. The work is supported by physical and chemical investigations into the underlying principles.

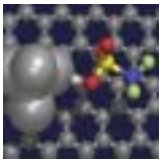
3.1.1.1 Underlying physical and chemical principles

In order to develop fuel cell components, fuel cells and stacks successfully, an extensive knowledge is required of the properties of the materials involved and of the complex interactions between these materials. Apart from experiments, theory and computer modelling also make major contributions towards obtaining a fundamental understanding of the structures and processes in fuel cells. The objective of these physical and physico-chemical studies is on the one hand to obtain a fundamental understanding of the structures and processes over various timescales and length scales, and also to apply this understanding to further developing PEFCs and DMFCs. To this end, atomic and continuum simulation procedures are being developed and implemented.

Our numerical and analytical models for cells and stacks are based on fluid dynamics equations. These models were created to shed light on the physics of cell and stack operation and as part of the search for new designs. Amongst other things, our DMFC model enabled us to identify the requirements for a new mode of operation in which one part of the cell reforms methanol to hydrogen and the other part uses the hydrogen as fuel. The stack model shows how a local increase in current density can result in the vicinity of non-conductive defects, which is one possible cause of stack ageing.



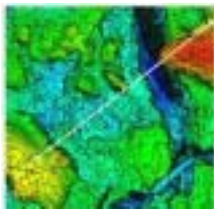
At a molecular level, models of proton transport in polymer electrolytes are being developed and simulated using the classical molecular dynamics method. Simulation calculations point to possible conclusions about the atomic mechanisms of proton transport. In addition, they enable proton mobility to be determined as a function of the operating parameters of the fuel cell or the chemical structure of the polymer.



Ab initio molecular dynamics simulations are performed in order to study the elementary steps in the anodic oxidation of methanol at precious metal catalysts and oxygen reduction on lanthanum strontium manganese oxide surfaces. A consideration of the environmental effect on the electronic structure of the reactants and products enables the mechanistic details of the electrocatalytical reactions in the fuel cell to be investigated.

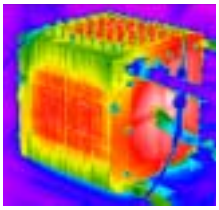
3.1.1.2 Electrochemistry and stack and system development

The R&D work on direct methanol fuel cells (DMFC) focuses on developing DMFC systems in the kW class for battery-driven material handling vehicles (forklift trucks). DMFC energy systems are required here to replace the existing lead acid batteries, both structurally and in terms of energy. During period under review work fucused on increasing the lifetime and the power density of the stack and also on the overall development or hybridization of the DMFC system to make it economical.



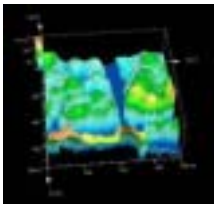
A prerequisite for the market introduction of direct methanol fuel cells for portable and small mobile applications is not only high power density and long-term stability but also a cost level that is comparable to existing technologies. The specific electric power of the membrane electrode assembly has a direct influence on the size of the fuel cell stack and thus on power density and cost. Work on the development of MEA components thus concentrates on electrochemical and structural investigations with the aim of optimizing media and current distribution in the electrodes and thus improving MEA performance while reducing the quantity of noble metal required.

Work on the further development of manufacturing processes for components of the membrane electrode assembly (MEA) concentrates on continous machine fabrications of gas diffusion electrodes (GDE), with the focus on developing suitable catalyst inks for applying the catalyst layer on diffusion layers via the knife over roll technique on a pilot plant scale.



In the priority area of stack development, work concentrates on further increasing density in DMFC and PEFC stacks whilst maintaining their functionality and reliability. An important consideration here is the choice of materials for bipolar and collector plates. Of particular importance are materials that do not interact with the cell components, that retain their functionality over an extended period of time and that can be produced cheaply.

With regard to system technology, potential applications for services in the kW sector are being sought and initial demonstration units are being realized in collaboration with industry partners. To this end, the system is analysed, the process engineering designed, components procured and characterized and finally the complete system is constructed and tested.



Particular attention is paid to the development, setup and application of special measuring techniques for the structural analysis of components of the membrane electrode assemblies and for the characterization of stacks and single cells. Highest priority is given here to the clarification of ageing effects of the membrane electrode assembly.

3.1.2 Important work results

3.1.2.1 Basic physicochemical principles

Bifunctional regime of a DMFC

In conjunction with the experimental work, modelling studies showed that in certain circumstances a DMFC operates in bifunctional mode. If airflow is reduced below a critical value, a part of the cell works as an electrolyser and produces hydrogen. If the airflow is increased, this domain reverts to working galvanically as a fuel cell, but at a *higher* power density than before. This effect can be exploited for the efficient electrochemical activation of the DMFC by periodically reducing the air flow. A patent application has already been filed.

Under certain conditions, the hydrogen generated in the electrolytic domain is consumed within the cell. This produces a new type of fuel cell – the direct methanol hydrogen fuel cell (DMHFC). This type of cell is a low-temperature fuel cell system with internal reforming (see Fig. 15). Hydrogen generated in the electrolytic domain permeates through to the galvanic domain on the anode side and thus forms part of the current generated.

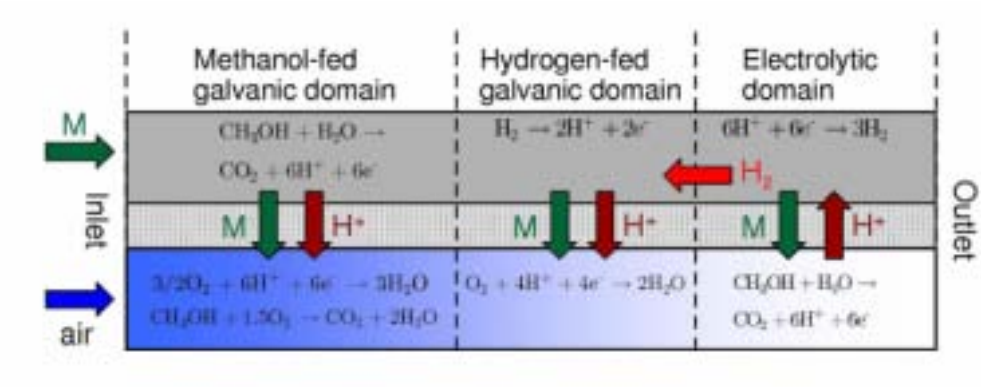


Fig. 15: Diagram of the processes in a DMFC

Heat transport in MEAs and stack

A model of the heat transport in the MEA of a PEFC or DMFC was developed. Analysis carried out on this model helped to develop a new procedure for measuring the thermal conductivity of the catalyst layer and membrane while the fuel cell is in operation.

A model was produced to show current transport through the stack and the fundamental relation for the voltage drop owing to cross-currents within the bipolar plates was established. On the basis of this equation a model was developed for a stack with high resistance defects. The calculations show how the local current density j increases around the defects (represented by white circles in Fig. 16) The closer two defects are to each other, the greater is this increase (see Fig. 16). The local increase in current density (excess current density = local current density minus average current density) leads to greater thermal and electrochemical disturbances (e.g. in the voltage in each half of the cell, for the electroosmotic water flow, etc.), which in the long term can cause the defects to increase in size.

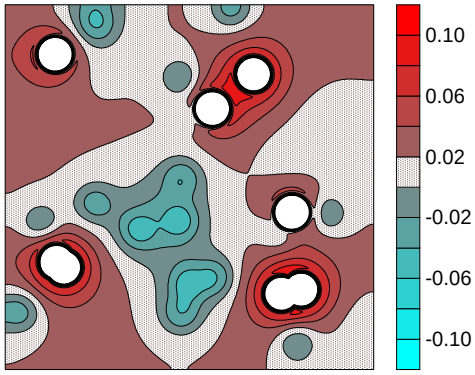


Fig. 16: Distribution of dimensionless local excess current densities across the surface of a cell with 8 high-resistance defects

Methanol oxidation at the Water-platinum interface

Since a dilute methanol solution is used as fuel in the liquid-operated DMFC, oxidation occurs on the catalyst surface *in the presence* of water. We created a simple model of the oxidation of methanol at the platinum/water interface and investigated its electronic properties using quantum mechanical density functional theory. The simulations show at positive potentials water is not only capable of influencing the mechanism of methanol oxidation in comparison to the gas phase process; but it even takes an active part in the reaction (see Fig. 17)

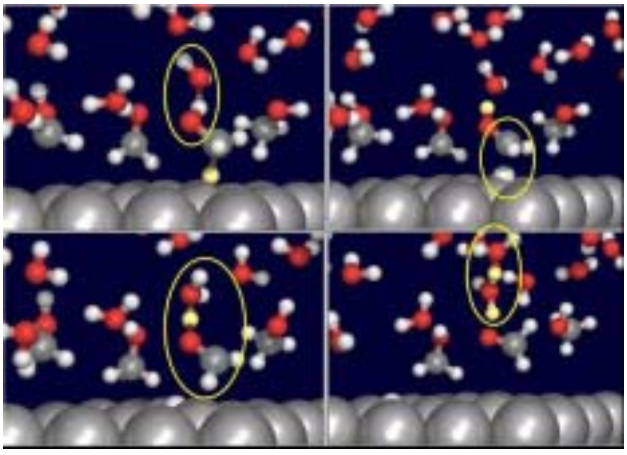


Fig. 17: Chemical reaction dynamics for the oxidation of methanol to formaldehyde at the water/platinum interface

If the potential is strongly positive a hydrogen bond then forms between a water molecule and the adsorbed methanol molecule (see Fig. 17, above left). The C-H bond activated via

surface interaction is then split (above right). The adsorbed hydroxymethyl group decays rapidly via proton transfer of the hydroxyl hydrogen atom to the water molecule bound to the hydrogen bond (below left) into adsorbed formaldehyde and an aqueous proton complex (Zundel ion $H_5O_2^+$) (below right). In the whole process, one proton is transferred directly into the aqueous phase and another adsorbed at the surface, without any adsorbed carbon monoxide being formed.

3.1.2.2 MEA development

The performance and durability of the membrane electrode assemblies were improved in the period under review via an improved structure for the diffusion layers and more efficient catalysts. For fuel cell technology to be marketed successfully, it must be possible to manufacture the components cost-effectively. This is possible only if manufacturing is automated. During the period unter review a system for the hot pressing of membrane electrode assemblies was developed and automated electrode manufacture was further developed.

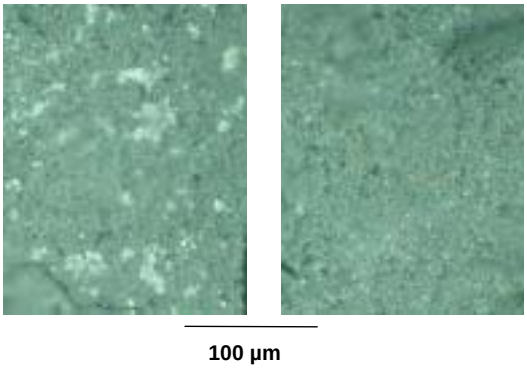


Fig. 18: Diffusion layer with coarse (left) and fine (right) PTFE distribution

The automated manufacturing of electrodes for fuel cells requires suitable inks that can be knife-coated and dried to form the appropriate contact layers. During the period under review improvements were made in particular to the inks for diffusion layers. PTFE is distributed considerably more finely in comparison to previous inks. Whereas, previously, PTFE particles could be seen under the microscope in the layers deposited (see, Fig. 18, left), this is no longer the case (see Fig. 18, right). This means that the diffusion layers are now uniformly hydrophobized. The stability of the inks was also improved. The inks can now still be used (after brief stirring) six months after they were manufactured. The inlets can now be manufactured in advance and components can thus be coated on a continuous basis. In the period under review an entire roll of carbon fabric 45 m in length was coated continuously to a constant level of quality.

The transition was made during the period under review to using solely carbon-supported platinum catalysts for the cathodes. Relative to the amount of expensive precious metal, these catalysts are more effecient and less affected by ageing than the unsupported platinum catalysts used previously, although they are more difficult to process. Using the experience acquired with the 'Deskcoater' coating machine, it was possible to manufacture inks during

the period under review that produced effective catalyst layers even when only a single coat was applied. A higher power density has already been achieved with cathodes using 2 mg/cm² than was previously achieved by using 4 mg/cm² on the unsupported platinum cathodes (see Fig. 19).

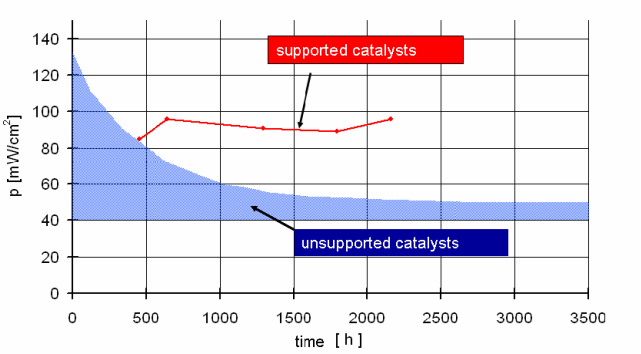


Fig. 19: Performance and stability over time of MEAs with different catalysts

A major step forward in terms of manufacturing fuel cell components cost-effectively was the automation of MEA manufacture. During fabrication of the last stack, manual pressing of the MEA proved to be very time-consuming and fault-prone. To reduce the risk of error and to accelerate the manufacturing process, a robot system was combined with pressing equipment. The system for the hot pressing of membrane electrode assemblies has the following characteristics:

- can be coupled to a 6-axis robot
- CNC programming of pressing processes
- choice of power control or path control
- documentation of the pressing process
- tact time of 7.5 min as opposed to 45 min for manual assembly

The picture on the left in Fig. 20 shows the fully assembled presses installed next to the robot's storage tables. Special receiving fixtures ensure that pressure is distributed evenly across the border area and across the active MEA surface, and also ensure that the MEA is positioned centrally. The 6-axis robot equipped with the universal grapple system is shown in the picture on the right in Fig. 20. The vacuum grippers grip the mounting plate bearing the presorted MEA. A lifting fork system is used to transport the MEAs for pressing and is coupled to the 6-axis robot, with which it communicates via handshake signals.

Using a robot system combined with two presses (one heated press and one cooled press) means that manufacturing time could be reduced by a factor of ten and assembly accuracy could be increased considerably at the same time.

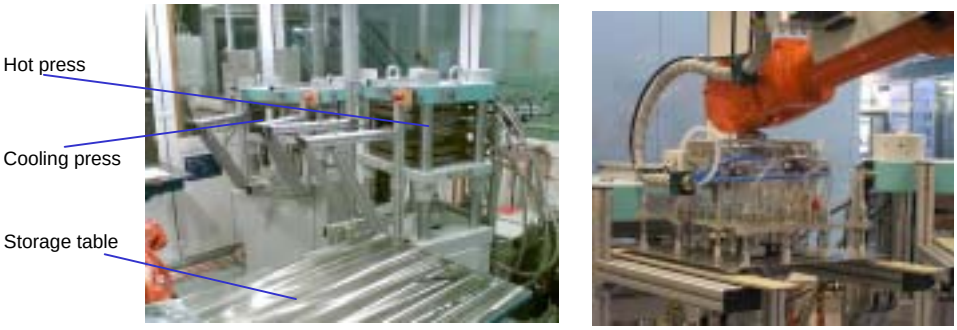


Fig. 20: Automated pressing system for MEAs (left); robot grapple system (right)

Using a robot system combined with two presses (one heated press and one cooled press) meants that manufacturing time could be reduced by a factor of ten and assembly accuracy could be increased considerably at the same time.

3.1.2.3 MEA characterization and ageing test

To study the effect of media distribution on the electrochemical processes in an MEA, a measuring device with segmented flow fields was developed in collaboration with the Institute for Power Electronics and Electrical Drives (ISEA Aachen). These fow fields enable the distribution of current, temperature and impedance across the active cell surface to be measured. The method can be adapted to fit the geometry of the MEA and the flow field structures.

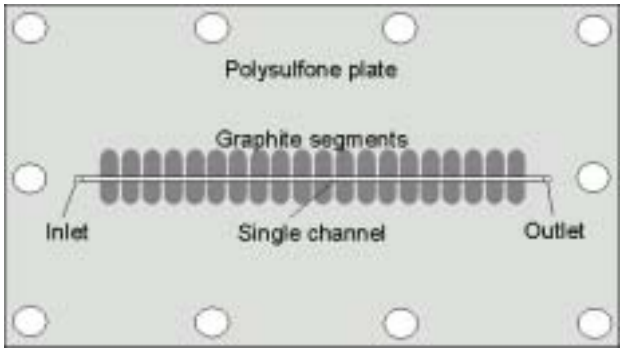


Fig. 21: Segmented flow field with single-channel geometry

The experiments were initially carried out using a single-channel geometry for the flow fields, since one-dimensional flow geometry is more accessible for modelling. In this configuration 20 graphite segments were electrically insulated from each other and embedded in a polymer plate (see Fig. 21) in which a channel approx. 10 cm in length was cut and holes were drilled to transport the media (methanol solution and air) to and from the site. Adapter and measuring circuit boards were used to measure the distribution of the above mentioned parameters along the channel.

The configuration of the test cells with the adapter circuit board on the anode side is shown in Fig. 22 When measuring current distribution in the segmented single channel for DMFCs, 'bifunctional operation' was directly demonstrated during oxygen depletion for the first time – an electrolytic current was observed in the area around the air outlet and galvanic current was observed in the area around the air inlet.

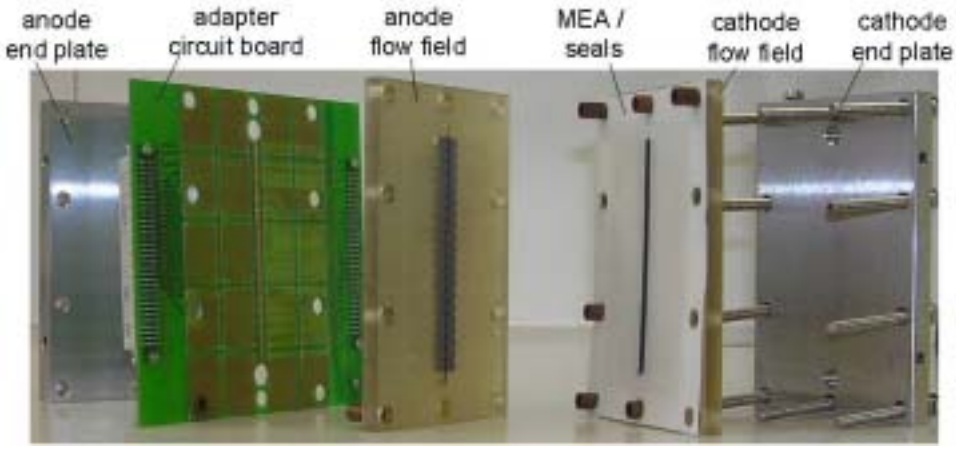


Fig. 22: Configuration of test cells with segmented channel – flow field

These results have already been published. The practical use of bifunctional operation lies in the fact that it activates the MEA, which progressively deactivates over the operating time. Periodic activation while the fuel cell is in operation means that DMFC performance can be increased by 30 % or more. A patent application has been filed for a setup exploiting this activation effect.

In addition to the measurements in the single channel, current distribution was also measured in a cell with a quadratic flow field composed of 25 electrically insulated segments with grid structure and an active surface of 18 cm^2. These measurements show that, depending on operating conditions, the current distribution can be very uneven, which could be critical for ageing in DMFCs. Previously, particularly critical operating conditions were identified as methanol depletion in the anode and air entry in the anode when the cell was only partially filled with methanol. These measurements showed that the MEA aged considerably more quickly in the area of the anode containing air than in the area containing methanol.

Test cells with new flow fields were developed in order to examine ageing during continuous operation. These flow fields contain a new manifold design based on simulation calculations. As shown in Fig. 23, the manifold has only one connection for inlet and outlet (whereas previously there were two) and its geometry ensures that media will flow to and away from the cell in a more homogenous manner. In particular, water removal from the cathode compartment is improved, thereby preventing oscillations in the cell voltage under load operation.

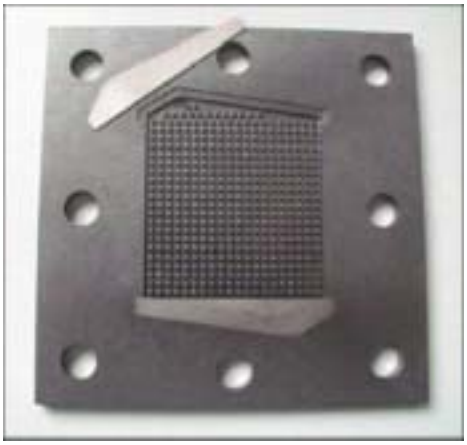


Fig. 23: Improved flow field design

Ageing effects on the integrated DMFC stack were observed when the scooter constructed in 2005 was operated. Analysis of the MEA specimens taken from the scooter produced two significant findings:

- ruthenium had dissolved in the DMFC anode and had been deposited in the channels of the bipolar plate (see Chapter 3.5)
- a chloride content of approx. 10^{-4} mol/litre was detected in the anode feed.

It is known from the literature that ruthenium corrosion is one of the main causes of ageing in DMFC MEAs. It is also known that chloride ions act as a complexing agent and promote the corrosion of ruthenium and platinum. Ageing tests were therefore carried out in which various concentrations of hydrochloric acid from 10^{-4} -10^{-2} mol/litre were added to the methanol solution.The content of platinum and ruthenium in the methanol solution escaping from the anode was determined using ICP-OES (ZCH/FZJ). This showed that the cell voltage already dropped considerably even at low concentrations of chloride ions. At higher concentrations of chloride ions the effect is more pronounced. If the anode is rinsed with pure methanol solution once more, the cell voltage increases but does not regain its initial value. This therefore shows that the MEA has aged irreversibly, which is confirmed by the results of the analysis. Both platinum and ruthenium were found in the anode effluent.

It is thus essential to prevent chloride ions from entering the anode liquid. In order to achieve long-term stability in cell operation, it is also necessary to prevent the electrochemically active areas of the fuel cell being contaminated by foreign Ions. Such ions could be transported by an insufficiently filtered air supply, for example, or via other media flowing into the stack. Another source of ions is the materials used. During operation, different ions, including chloride ions, can be leached out of the components of a stack and diminish the performance of the MEA. In order to avoid this, the materials used must be chosen very carefully, and the stack and system components must be carefully cleaned. A cleaning method based on an anode-model liquid was developed for this purpose and demonstrably reduced ion discharge during operation To this end, the components are processed in different cleaning fluids at a temperature of approx. 80 °C for a set period of time.

In addition to the presence of complexing agents like chloride, the anode overpotential also plays a significant role, since in general the catalyst corrodes more strongly at higher overpotential. High anode overpotentials must therefore also be avoided. Without chloride, 50-fold higher platinum and ruthenium contents are measured in the anode effluent during high anode overpotentials under moderate conditions with 1 mmol chloride (see Fig. 24).

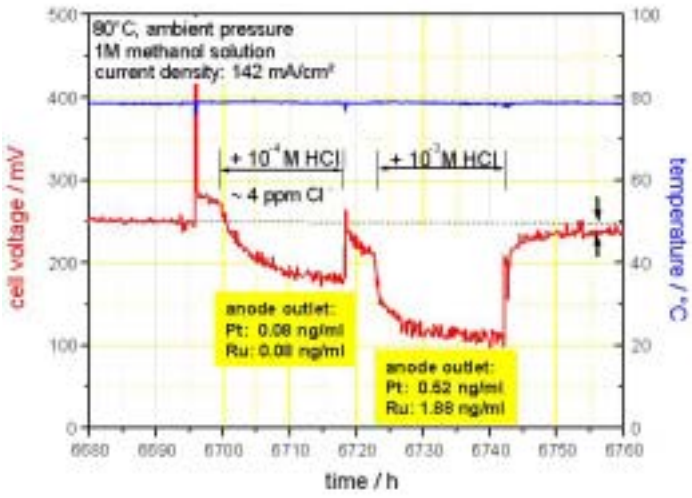


Fig. 24: Galvanostatic ageing experiment with addition of hydrochloric acid to the methanol solution; data highlighted in yellow: Pt and Ru content of the anode effluent

Critical operating conditions that can cause anode overvoltages in the stack are

- low operating temperatures and moderate current densities during start-up;
- high current densities during operation; and
- ingress of air into the anode compartment during shutdown times.

As a consequence of these results, anion exchangers were used in the 2006 scooter in order to reduce the concentration of chloride ions in the anode feed. The results were also used in designing the system control in order to optimize the regulatory algorithms.

3.1.2.4 Stack development

Further developments in DMFC stack design can be characterized by an improvement in the flow structures supplying methanol solution to the individual anodes. Previous work had always focused on uniform flow distribution across the cell surface and paid little attention to how the media were distributed in the cells themselves. Changing the anode supply channels meant that gas removal could be improved and thereby a uniform media distribution was obtained. This is proven by the fact that cells at the edge of the stack now receive an improved supply of methanol.

Research into new MEAs was also carried out as part of the development work. This research focused on measuring the electrochemical properties of new gas diffusion layers based on non-woven materials. The advantage of non-woven materials lies in their mechanical properties: since there are no warp and no weft threads, non-woven substances stretch very isotropically under mechanical load compared with woven substances. This lends better support to the membrane and prevents the MEA expanding into the flow channels of a cell. The surface of non-woven materials, which is very smooth compared with that of woven materials, enables channels to be cut to a more accurate size and therefore ensures that flow through the flow fields is more uniform. This in turn leads to improved gas or water removal from the cell. The measurements taken showed comparable performance to that achieved with conventional MEAs, although it was not possible to raise the specific power in the stack (as opposed to in single cells) using these materials.

The range of tests in short stacks was extended so that there is now a test stand for short stacks to be subjected to automatically determined load profiles and thus dynamic ageing tests can be carried out. Given the increasing volume of data involved, administering the information generated is a particular challenge. Further developments were introduced in performance data processing to ensure that the large volumes of data could be efficiently administered. A standardized data transfer from the automated test stands meant that evaluation routines could be run to filter out and evaluate the relevant data.

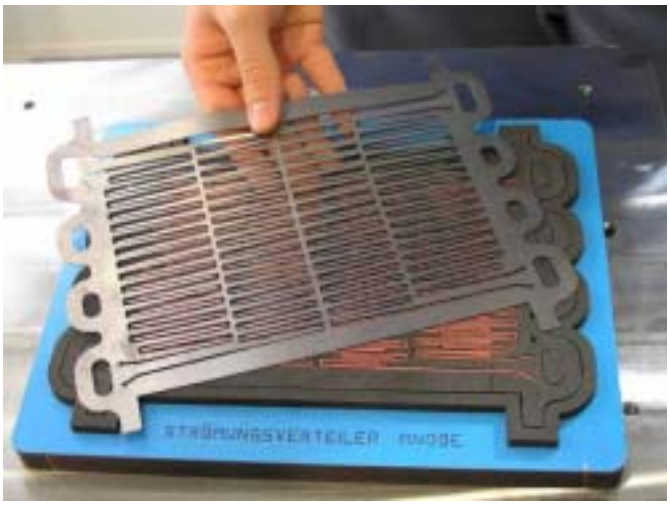


Fig. 25: Flow structure manufactured by die-cutting

In further developing stack components, particular emphasis was placed on the manufacturing technique. The time-intensive process of laser-cutting components was abandoned in favour of a die-cutting procedure, which has a much higher throughput rate. A high level of process reliability has already been achieved in die-cutting the bipolar plate, the anode frame, the cathode frame, the membrane and the GDE, although the process for manufacturing the delicate flow distributor (particularly the cathode flow distributor) has not yet been refined to a sufficient degree. Developing suitable tools for removing the structures from their moulds has proven to be a major challenge. As Fig. 25 shows, the anodic current

distributor, which poses challenges in terms of its shape, can be removed easily from the mould and manufactured with a high level of process reliability. However, the level of accuracy obtained in manufacturing does not yet equal that obtained with laser-cutting. The supporting frames for the MEA were previously manufactured using the water jet cutting procedure. This procedure is well-adapted to the manufacture of individual parts, although manufacturing a large number of parts remains very time-consuming. For this reason the frames composed of fibre-reinforced plastic were manufactured by die-cutting. The dimensional accuracy of the frame is good and the lifetime of the tools is also sufficient.

During the period under reviewe a DMFC stack (see Fig. 26) was built that achieved a maximum power of more than three kilowatts. The exact data can be taken from the characteristic curve in Fig. 26 and the technical data in Tab. 2.

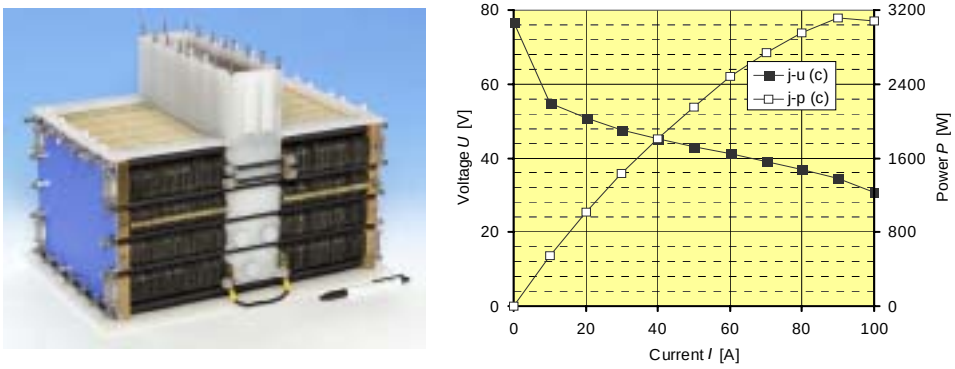


Fig. 26: DMFC stack from 2006 (left) and characteristic curve of the DMFC stacks at 80 °C

Nominal power	W	2000
Peak power	W	3100
Weight	kg	30
Volume	l	25
Maximum current	A	99
Number of cells		100
Precious metal loading	mg/cm²	5
Spec. nominal power	W/l W/kg	80 67
Spec. peak power	W/l W/kg	124 103
Active cell area	cm²	320

Tab. 2: Technical data for the DMFC stack

The design of this stack is similar to previous models. The model also has graphite flow distribution plates and an 'open' cathode in order to keep pressure loss on the air side to a

minimum. In comparison to previous models, however, this stack was built to have optimum durability. This involved not only making further developments to the MEAs but also modifying the power supply leads, in order to ensure that all cells were uniformly supplied with current. The modifications to the design and operation of the stack led to its durability being improved by more than a factor of ten as compared with the first stacks. This was partially due to the improved mechanics of the MEA and the stable interconnection of its function layers and partially to the fact that the stack components were subjected to a novel cleaning procedure that helped to reduce the load arising from ions being detached during operation. In terms of design the geometry of the flow distribution plates was adapted so that it could be manufactured at low cost, and also so that it could very reliably support the loads arising during operation.

Using an optimized clamping device (see Fig. 27) the end plates can now be braced as parallel to each other as possible, which improves the reliability of the stack. By aligning the end plates exactly it can now be ensured that the contact pressure is distributed evenly. This prevents any untightness and local deformations. A further advantage of the new assembly apparatus configuration is that the pressure is applied to the end plates by hydraulic cylinders, so the tie rods can be braced without torsion. This enables thinner tie rods to be used and reduces the cost and weight of the stack.

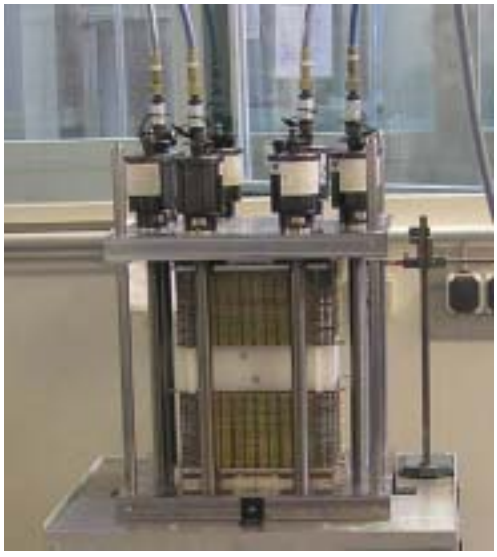


Fig. 27: Clamping device for plane parallel assembly of the stack

3.1.2.5 DMFC system development

R&D work in DMFC process engineering is part of the medium-term target of developing a hybridized DMFC system for use in industrial factory trucks (forklift trucks) until it is commercially viable. Two milestones on the way to achieving this objective were the two electric scooters (JuMOVe and JuMOVe 2, Chapter 1.1). The focus of the development was largely determined by practical experience of operating the previous systems.

The system design of the JuMOVe 2 scooter is shown in Fig. 28. The basic design of the DMFC peripherals was taken from the previous system. Air is supplied via a suction blower behind the stack, the stack is cooled by water evaporation on the cathode side and water is recovered in an air-cooled condenser. The methanol/water supply is provided by an anode loop comprising a circulation pump, a CO_2 separator and feed inlets for methanol and water.

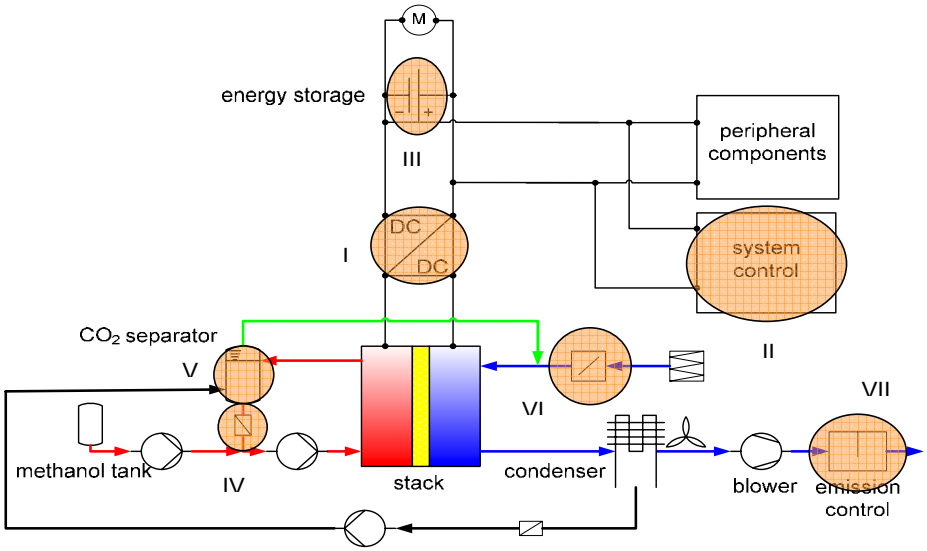


Fig. 28: Flow diagram JuMOVe 2; system components affected by R&D work are indicated by Roman numerals

The procedures and components that have been developed further or integrated in the design are discussed below.

I. The hybrid concept was modified through the integration of a DC/DC converter (see Chapter 1.1).

II. A system controller was implemented to protect the stack, as well as a limitation map controller that compensates for ageing-related deviations in stack performance from the nominal state (see Chapter 1.1).

III. The Li-ion battery was replaced by a new high-current spiral wound lead acid battery (manufactured by Hawker, 'Cyclone' model). The advantages of modern lead batteries are lower cost (approx. 10 % of the cost of Li-ion batteries) and the fact that no battery management system is required.

IV. A liquid filter/ion exchanger cartridge was integrated in the anode loop. Damage to the membrane due to foreign ions was identified as a possible cause of stack ageing. The source of this contamination was stack and system materials coming into contact with operating fluids (anode solution, methanol, condensate). Comprehensive analyses of various materials have shown that no material is completely inert. For the

JuMOVe 2 great care was taken in selecting the materials, and a mixed bed ion exchanger (to eliminate anions and cations) was incorporated into the anode loop.

V. The CO_2 separator in the anode system integrated into the stack was modified. In order to prevent the anodes coming into partial or temporary contact with the air (which can also lead to losses of power and long-term durability), the system was modified so that an anode liquid reservoir was permanently maintained above the upper stack surface. This reservoir is monitored by sensors.

VI. A cathode sealing system was developed. To minimize water loss during shutdown times, a lamellar sealing system was developed to seal off the cathode inlet on the upper surface of the stack.

VII. An oxidation catalyst was developed for cleaning the exhaust gas. A large quantity of pollutant emissions (particularly methanol) must be expected from the cathode exit. In collaboration with a catalyst manufacturer, a cleaning module was developed that will have a high conversion rate even at low temperatures (approx. 40 °C). Its effectiveness and durability are not yet known; this will be explored in future work.

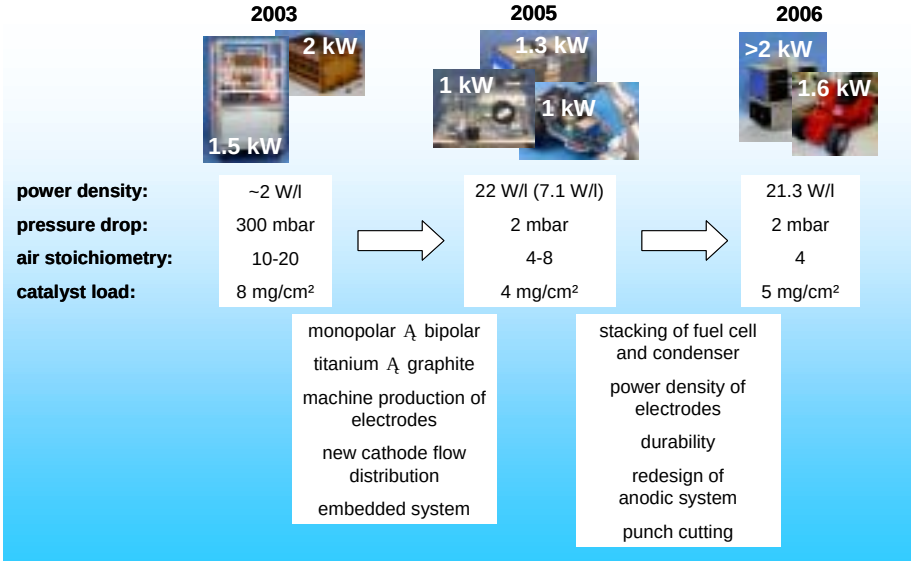


Fig. 29: DMFC system and stack development 2003 - 2006

Fig. 29 shows the advances made in system and stack development from 2003 – 2006 in figures. The major advances in stack development were a reduction in the quantity of catalyst required for the membrane electrode assembly and a minimization of pressure loss in combination with reduced excess air for the cathodes. In terms of system development, further advances continued to be made, including a considerable increase in power density in particular.

3.1.3 Staff members and fields of activity

Name	Telephone no. (+49-2461-61-) E-mail address	Field of activity
Prof. L. Blum	6709 l.blum@fz-juelich.de	Head of Fuel Cell Process Engineering (VBZ)
J. Mergel	5996 j.mergel@fz-juelich.de	Head of Electrochemistry/Low Temperature Fuel Cells (EWN)
Prof. E. Spohr	6687 e.spohr@fz-juelich.de	Head of the Basic Physical & Chemical Principles (PCG), research into electrocatalytic and transport processes using quantum mechanical and molecular dynamic simulation methods
A. Bendzulla	2177 a.bendzulla@fz-juelich.de	Development of stack concepts for HT-PEFC, benchmarking of stack materials
Dr. A. Glüsen	5171 a.gluesen@fz-juelich.de	Evaluation of new membranes and performance optimization of MEA components for the DMFC
Dr. H. Dohle	6884 h.dohle@fz-juelich.de	Head of the 'HT-PEFC Stack Development' Group, technical optimization of stacks, selection and testing of materials for cell parts
Dr. H. Janßen	5082 h.janssen@fz-juelich.de	Head of the 'DMFC System Development' Group, development of components, plant design, construction and testing of DMFC systems
D. Kalkreuth	2378 d.kalkreuth@fz-juelich.de	General quality management, quality assurance for 'Standardized electrochemical testing procedures', development of sample-preparation procedures for imaging test procedures
N. Kimiaie	6484 n.kimiaie@fz-juelich.de	Technical and organizational project coordination for DMFC system development, developing automated production and assembly procedures for fuel cells components and stacks
Prof. A. A. Kulikovsky	5396 a.kulikovsky@fz-juelich.de	Development of analytical and numerical models of PEFCs and DMFCs, interpretation of experimental data, designing new cell and stack set-ups
Dr. M. Müller	2775 mar.mueller@fz-juelich.de	Head of the 'DMFC Stack Development' Group, technical and economic optimization of DMFC stacks, selection and testing of materials for stack parts
H.-F. Oetjen	6029 h.-f.oetjen@fz-juelich.de	Development of test stand concepts for HT-PEFCs, testing of UI characteristics of HT-PEFCs

H. Schmitz	4113 hei.schmitz@fz-juelich.de	Development of continuous automatic manufacturing procedures for components of the membrane electrode assembly (MEA)
Dr. M. Stähler	2775 m.staehler@fz-juelich.de	Head of the 'MEA Characterization' Group, standardized characterization of DMFC membrane electrode assemblies and development of new characterization procedures.
C. Trappmann	5590 c.trappmann@fz-juelich.de	Development of metallic stack concepts for DMFCs Evaluation of metallic materials, investigation of ageing mechanisms
Dr. Chr. Wannek	4013 c.wannek@fz-juelich.de	Head of the 'MEA Development' Group, development of membrane electrode assemblies for DMFC and HT-PEFC
M. Wannert	5590 m.wannert@fz-juelich.de	Development of technical systems for and characterization of a mobile direct methanol fuel cell unit
J. Wilhelm	1573 j.wilhelm@fz-juelich.de	Development of technical systems for and characterization of a mobile direct methanol fuel cell unit
Dr. K. Wippermann	2572 k.wippermann@fz-juelich.de	Clarification of physical/chemical ageing mechanisms in direct methanol fuel cells
W. Zwaygardt	2109 w.zwaygardt@fz-juelich.de	Development and testing of DMFC system components

3.1.4 Important publications, PhD theses and patents

Important publications

Dohle, H.; Mergel, J.

DMFC stacks and systems in the kW range

Proceedings of the Professional Engineering Publishing. - Suffolk, UK, 2003. - 2005. - S. 43 - 60

This article discusses DMFC stack technology in the range from several hundred watts to kilowatts. Various stack concepts are compared and contrasted. The article also discusses the design of individual stack parts. Taking the example of the current collector, the influence of coatings on contact resistance is discussed with the aid of IR measurements. Operating conditions, particularly operating temperature and methanol concentration, are investigated as an example in terms of the power output. The necessary system components are described and evaluated with the example of a 2 kW system in terms of design and their influence on overall effectiveness.

Dohle, H.; Stolten, D.

DMFC systems: state of the art and future trends

Proceedings of the 2005 Fuel Cell Seminar, Palm Springs, USA. - S. 338 - 341

This article sheds light on the market situation for DMFC applications in the industrial sector. At first sight, several mass markets appear promising from the current perspective of European research owing to their volume, but these markets are not sustainable bases for DMFC by reason of the cost situation (automotive sector) or the lack of industrial users in Europe (consumer electronics sector). However, the field between these two – mid-power applications – shows potential. These applications are covered in greater detail in the article in terms of cost objectives and the technical potential for realization.
This article particularly focuses on a comparison between DMFC and PEFC systems in the kW class. It emerges that DMFC systems are equal, if not superior, to PEFC systems in terms of all their properties up to a power of approx. 2 - 5 kW. Below this threshold the simplicity and robustness of the DMFC system design have an ever-increasing effect, and above this threshold the higher power density of the PEFC stack determines how the systems compare. 'Light traction' applications in particular, such as golf carts, forklift trucks or special vehicles, benefit from the quick and easy procedure for refilling with methanol and the theoretically greater ease with which a nation-wide infrastructure can be set-up.

Fricke, B.*; Sanders, T.*; Mergel, J.; Schmitz, H.; Wippermann, K.; Kulikovsky, A. A.

Bifunctional regime of DMFC operation close to open circuit conditions

Spring Meeting of the Electrochemical Society 2006, Denver, Colo.: 07.05.2006 - 12.05.2006

We designed a DMFC with single straight channels and segmented electrodes. The length of the cell is 10 cm; on each side of the cell 20 electrodes (segments) of the size å0.5 × 1.5 cm are located equidistantly along the channel. The measuring system enables to record current from each pair of segments. The segments are connected to load resistor through the specially designed compensation system, which provides equipotentiality of all segments. Close to open circuit conditions the

cell exhibits bi-functional regime of operation. Part of the cell located close to the outlet of oxygen channel works as electrolysis cell and consumes current generated in the fuel cell (galvanic) domain to produce hydrogen.

Fricke, B.; Sanders, T.; Baumhöfer, T.; Sauer, D.U.; Wippermann, K.

Position-resolved impedance spectroscopy on fuel cells

Technische Mitteilungen : TM - Organ des Hauses der Technik Essen, 1-2 (2006), 231 – 234, ISSN 0040-1439

The requirements were presented for a measuring device to measure position-resolved impedance in DMFCs. A necessary condition is the position-resolved measurement of the current distribution, with the easiest place to measure it being between the end plate and the manifold plate. It is particularly important, especially when making measurements at a DMFC, to take the measurements as simultaneously as possible. In order to make precise measurements any possible feedback effects on current distribution caused by measuring that current must be avoided. Finally, the article presented a measuring system suitable for measuring impedance and fulfilling these requirements.

Hartnig, C.; Spohr, E.

The role of water in the initial steps of methanol oxidation on Pt(111)

Chem. Phys., 319 (2005) 185

We report the results of quantum-chemical and ab initio molecular dynamics studies within the framework of density functional theory for the oxidation of methanol on the (111) face of a platinum single crystal. In aqueous solution the oxidation of methanol starts by the formation of a hydrogen bond from the OH group of the methanol to a solvent molecule. The initial step of the reaction is the cleavage of a CH bond which points towards the platinum surface; this is followed by rapid dissociation of the methanol OH bond, which leads to formaldehyde as a stable intermediate on the time scale of the simulation. Charge delocalization is achieved by the formation of a Zundel ion ($H_5O_2^+$) in the aqueous phase. The further evolution provides hints for the following steps of methanol oxidation and proton conduction in the environment of a liquid-fed direct methanol fuel cell.

Janßen, H., Blum, L., Kimiaie, N., Maintz, A., Mergel, J., Müller, M., Stolten, D.

Performance Characterization of a 4-Wheel DMFC Scooter

Proceedings of the 3rd European PEFC Forum, Lucerne, 04-08 July 2005, File No. P302, p. 40

To demonstrate the feasibility of a small DMFC driven electric vehicle Forschungszentrum Jülich built as demonstrator a 4-wheel scooter, called JuMOVe (**Ju**elich **M**ethanol **O**perated **Ve**hicle). Using the platform of a commercially available battery powered electric wheeler the DMFC system is installed replacing lead acid batteries. Only for the purpose of start-up and for peak power, as steep inclines or high acceleration the system is hybridized by a Li-ion battery. This paper shows the results of performance measurements during scooter operation.

Kulikovsky, A. A.; Klafki, K.; Wippermann, K.

Experimental verification of the effect of bridge formation in a direct methanol fuel cell

Electrochemistry Communications, 7 (2005), 394 - 397

Recently the effect of formation of a narrow current-carrying domain (bridge) in a direct methanol fuel cell was predicted theoretically. The bridge supports finite local current density under vanishingly small total current in the cell. This effect explains a well-known phenomenon of mixed potential: the bridge short–circuits the electrodes, thus reducing cell open-circuit voltage. In this work we report the first experimental confirmation of the new effect.

Kulikovsky, A. A.; Schmitz, H.; Wippermann, K.; Mergel, J.; Fricke, B.; Sanders, T.; Sauer, D. U.

DMFC: Galvanic of Electrolytic Cell?

*Electrochem. Comm., **8** (2006) 754*

The work reports the first direct observation of a bifunctional regime of DMFC operation, when part of the cell located close to the oxygen channel works in a normal galvanic mode while the rest part operates in electrolytic mode and converts methanol to hydrogen. As predicted by our model of DMFC, this regime arises when air flow rate is below certain critical value. The predicted values of the critical air flow rate and the length of galvanic domain are in good agreement with the experiment.

Kulikovsky, A.A.

Electrostatic broadening of current—free spots in a fuel cell stack: The mechanism of stack aging?

*Electrochem. Comm, **8** (2006) 1225*

Fuel cell stacks usually contain numerous resistive spots, which arise due to local corrosion, delamination of the catalyst layer from membrane or simply due to uneven distribution of clamping pressure. Our calculations show that local cell current around the spot edges increases, thereby increasing the thermal and the electrochemical stress in the defective cell. Furthermore, if two spots are located nearby, the disturbance of current between the spots is even higher. The non-uniformity in local current induced by the spots can lead to their broadening with time, which is a possible scenario of stack ageing.

Kulikovsky, A.A.

Heat transport in a PEFC: Exact solutions and a novel method for measuring thermal conductivities of the catalyst layers and membrane

*Electrochem. Comm., **9** (2007) 6*

We report analytical solution to the problem of heat transport in the membrane-electrode assembly of a hydrogen cell. Rather cumbersome solutions lead to remarkably simple formulas for the temperature drop over the membrane. Measuring this drop one can determine thermal conductivities of the catalyst layers and membrane in a working fuel cell environment.
the spot edges increases, thereby increasing the thermal and the electrochemical stress in the defective cell. Furthermore, if two spots are located nearby, the

disturbance of current between the spots is even higher. The non-uniformity in local current induced by the spots can lead to their broadening with time, which is a possible scenario of stack ageing.

Müller, M.; Blum, L.; Dohle, H.; Kimiae, N.; Mergel, J.; Nölke, M.; Janßen, H.; Stolten, D.

Progress in DMFC stack development and system integration at Forschungszentrum Jülich

Proceedings of the 3rd European PEFC Forum, Luzern, Schweiz, 04.-08.07.2005.

In this paper we focus on the newest developments in DMFC stack and system set-up at Forschungszentrum Jülich. We decided to build up a mobile DMFC application to demonstrate the feasibility of small fuel cell driven vehicles. For these applications the required power demand is up to 1 kW and the installation space is less than 80 liters. The aim of the project is the improvement of the total cruising range of such vehicles in comparison with conventional battery technology.
To reach this aim stack and system must be in tune. For the cathode air supply a channel structure with a low pressure drop is suggested. The use of a blower instead of a compressor guaranties high system efficiency. The removal of the liquid water is independent of the gas flow because a wick structure is integrated into the flow field. Also the exhaust gas treatment is done in the stack so the waste heat can be directly recovered at the anode.
The anode supply system is realized in an embedded system in the middle of the stack so the supply channels are half length of a usual stack. This improves the homogeneity of flow distribution. In the system a circulation pump and two separators are integrated. The anode exhaust gas is cleaned inside the tack so that only low emissions of gaseous methanol leave the system.
The external components are a blower for the cathode air supply and a condenser with an additional blower for water recovering. This unit can be placed anywhere around the stack so the systems can effectively be adapted to the low installation space.

Wippermann, K.; Kulikovsky, A. A.; Schmitz, H.; Fricke, B.; Sanders, T.; Sauer, D. U.

Basic principles of impedance spectroscopy on fuel cells and position-resolved measurements

Technische Mitteilungen: TM - Organ des Hauses der Technik Essen, 1-2 (2006), 96 - 106

Impedance is a nondestructive in situ measuring method and as such permits a detailed investigation of the state and processes in fuel cells. It enables significant findings to be obtained about mechanisms of ageing, process optimization and the efficiency of new materials. In particular, position-resolved impedance spectroscopy provides a valuable contribution to research and development, and is also of benefit when monitoring fuel cell stacks in applications. The large number of publications on this subject shows the relevance (and also the complexity) of the method, which must be applied with great care if usable results are to be obtained.

Zhang, X.; Glüsen, A.; Garcia-Valls, R.*

Porous lignosulfonate membranes for direct methanol fuel cells

Journal of Membrane Science, 276 (2006), 301 - 307

Porous lignosulfonate membranes were prepared and their potential considered for application in direct methanol fuel cells (DMFC). Membranes were characterized by impedance spectrometry and water uptake measurement. Both their ion exchange capacity and water uptake capacity affected porous membrane conductivity.

Membrane conductivities were in the range of 5 - 12 mS/cm at 80 ºC. Membrane electrode assemblies based on these lignosulfonate membranes were also prepared and test in fuel cell operation in order to determine whether they can be used in a DMFC. A current density of 42 mA/cm^2 was reached at 300 mV and 80 ºC.

PhD Theses

Müller, M. J.

Development and optimization of direct methanol fuel cell stacks

Schriften des Forschungszentrums Jülich, Reihe Energietechnik, Band 51,
ISBN 3-89336-434-X, RWTH Aachen University, 2006

The present study considers various ways to increase the power of fuel cell stacks. On the one hand, the work concentrates on investigating the influence of operating parameters on cell performance and, on the other hand, on the influence of structural measures on the performance of the whole system and the fuel cell stack.
Major phenomena in DMFC stack design are investigated by measuring stacks and single cells. The investigation is divided into three sections: identifying characteristic properties, assessing operating parameters and studying stack components. On the basis of the results of these sections models are developed containing key parameters for stack design and cell operation. Various properties are used in assessing power density, as well as the linearized current/voltage characteristic curve, as a practical instrument for optimizing and designing stacks.
Understanding mass transport in the membrane electrode assembly (MEA) is crucial to the function of single fuel cells and stacks. Proceeding from the mass flows, calculations of the mass transport were carried out for the anode and the cathode. To describe the diffusive mass transport at the cathode, a diffusion model was developed and transferred to a channel. This procedure makes it possible to gather information about current density distribution along a channel. An additional finding from these measurements is that in particular the water discharge has a influence on possible current density. The functionality of different stack concepts is discussed using case studies. Reticulate structures were compared with meandering structures for flow distribution. The structures distribute media homogeneously under single-phase continuous flow. When operated with multiphase media flows, channel structures with integrated wick systems show the most uniform flow distribution with little loss of pressure. This comparison of different stack concepts shows that stacks with a monopolar design are well-suited for applications where little space is available, whereas stacks with a bipolar design are more suitable for more powerful systems.
The electrical properties of the cell were simulated using a resistance network. For large cell surfaces, ohmic losses in the collector plates can lead to an inhomogeneous potential distribution and to losses of power. A design program is presented in which boundary conditions can be entered to generate design data for stacks. Parameter variations can be used to show that there is an optimum number of cells for each stack power in with respect to a maximum specific power. Integrating system components in the stack is proposed in order to optimize the performance of the fuel cell system, thus enabling more compact systems to be built.

Markus Nölke

Development of a direct methanol fuel cell system for the power class below 5 kW

Schriften des Forschungszentrums Jülich, Reihe Energietechnik, Volume 64,
ISBN 978-3-89336-481-7, RWTH Aachen University, 2007

A DMFC system can only be made more compact and simplified to the necessary degree via an integrated concept that combines the peripheral system and the stack in an optimum way. An accurate analysis of the stack and the peripheral components is a necessary condition for an optimized design. The potential of a system concept can be identified via a modelling of the whole system. This work was carried out in parallel to a development project for constructing a DMFC scooter at FZJ. The models were validated via experiments and model parameters were fitted accordingly. The system components developed as part of the work (e. g. a stack-integrated anode system) were successfully applied in an initial prototype system.

Schlumbohm, C.

Stability and structural modifications in catalyst dispersions of direct methanol fuel cells

Schriften des Forschungszentrums Jülich, Reihe Energietechnik, Volume 48,
ISBN 3-89336-429-3; RWTH Aachen University, 2005

Fuel cell systems are characterized by the fact that they provide energy with a low level of emissions. In spite of previous advances in research and development for mass production, manufacturing costs for fuel cell systems are still relatively high compared with those for other energy generation or storage systems. Improving performance whilst keeping costs low is thus an essential condition for successfully developing a commercially mature fuel cell system and launching it on the market.
The goal of this work is thus to use systematic analyses to identify and optimize the main parameters for more cost-effective machine fabrication of electrodes with respect to power per surface area and quantity of precious metal required.
Adding suitable additives to adjust the rheostatic behaviour and mechanical stability of the electrode proved beneficial during coating tests in order to reduce the costs of machine processing of the dispersions. Additives lead to stable and repeatable dispersion for technical coating procedures. The best results in terms of viscosity and cell performance were achieved by adding polyacrylic acid. The mechanical properties of the electrodes were improved considerably through the addition of fibres to the dispersion.
In order to achieve optimum electrode performance, a systematic examination of the structure of the dispersion was carried out in order to determine and modify the interactions between the constituents of the dispersion. This analysis indicated that the principal parameters were the solvent used and the dispersion procedure. The addition of polymers had particularly beneficial effect on the stability over time and thus the reproducibility of the electrodes. Another possibility of obtaining stable dispersions was shown by measurements of particle sizes with different solvents: relatively small particle aggregates in the sub-nanometre range show greater durability than aggregates or agglomerates above one micrometre. The dispersion procedure is a further parameter that influences the range of aggregate size distribution as well as the structure of the dispersion and the electrodes manufactured from it. Dispersion methods using a magnetic agitator and ultrasound produced considerable improvements in performance.
Statistical test planning was used to examine the influence of the following key parameters: influence of the dispersion procedure, the percentage of precious metal in the catalyst and additives on the dispersion, and finally the performance of the finished fuel cell. At a constant percentage of precious metal in a supported catalyst it

was shown that in addition to the percentage of precious metal the dispersion procedure showed the greatest potential for increasing the efficiency of a fuel cell.

Stähler, M.

The normal hydrogen electrode as a reference electrode in direct methanol fuel cells

Schriften des Forschungszentrums Jülich, Reihe Energietechnik, Volume 47,
ISBN 3-89336-428-5, RWTH University, 2005

Direct methanol fuel cells (DMFCs) can convert chemical energy from the liquid energy carrier methanol directly into electrical energy. They are particularly suitable for a mobile energy supply for power classes of up to a few kilowatts. For DMFCs to be optimized, classical electrochemical examination methods must be developed further and tailored to the specific situation in DMFCs. The procedures described in this work focus on reference electrode measurement and impedance spectroscopic examinations of catalyst-coated membranes for DMFCs. In addition to a comprehensive discussion and experimental examination of potential measuring errors, a method is also presented for characterizing DMFC anodes.

Wüster, T.

Development and modelling of a polymer electrolyte fuel cell stack in the 5 kW class

Berichte des Forschungszentrums Jülich, Reihe Energietechnik, Volume 46,
ISBN 3-89336-422-6, RWTH Aachen University, 2005

The goal of this work is to develop a 5-kW PEFC stack based on graphite bipolar plates from the scientific aspect. Partially conflicting objectives in this respect are, on the one hand, achieving as compact a stack design as possible whilst, on the other hand, creating flow structures with as homogeneous a distribution and as low a pressure loss as possible. Various flow structures were developed and measured as part of this work. A structure consisting of 14 parallel meanders with an active surface of 244 cm^2 was found to be optimum. The related sealing methods were also developed and tested.
A major priority of the work was simulations and measurements for the flow distribution. Working from simulation calculations, simplified design criteria for creating flow profiles were determined based on characteristic numbers. These numbers in turn were determined from the geometry and operating conditions such as the flow and the air required, and were incorporated in the design.
Finally, it was shown that a technically appropriate design of the cooling flow field considering the lateral heat conduction of the graphite bipolar plates led to small losses of pressure in the water circuit, which enabled a compact design with low pump capacity to be achieved.
The last chapter contains an economic evaluation of selected components from the stack in terms of material required and production technology.

Important patents

Patent applications:

Principal inventor	PT	Title
Dr. H. Dohle	1.2130 PCT/EP	Process for determining current density distribution in fuel cells
M. Schonert	1.2143 EP	Low-temperature fuel cell
Dr. M. Stähler	1.2151 EP	Process for coating a membrane with catalyst

Dr. C. Schlumbohm	1.2152 PCT	Fibres for a textile fabric and their manufacture and use
Prof. L. Blum	1.2175 EP	Manifold plate for a solid oxide fuel cell
Dr. H. Janßen	1.2196	Process for operating a direct methanol fuel cell stack
Dr. H. Janßen	1.2196 EP	Process for operating a direct methanol fuel cell stack
Dr. H. Dohle	1.2201	Low-temperature fuel cell and process for operating the same
Dr. H. Dohle	1.2201 PCT	Low-temperature fuel cell and process for operating the same
Dr. H. Dohle	1.2202	Low-temperature fuel cell and process for operating the same
Dr. H. Dohle	1.2202 PCT	Low-temperature fuel cell and process for operating the same
Dr. H. Janßen	1.2209	Direct methanol fuel cell system and process for operating such as a system
Dr. H. Janßen	1.2209 PCT	Direct methanol fuel cell system and process for operating them
Dr. A. Glüsen	1.2233	Process for manufacturing a membrane electrode assembly
M. Schonert	1.2251	Procedure for operating a fuel cell
Dr. H. Dohle	1.2260	Membrane electrode assembly for a low-temperature fuel cell and process for manufacturing the same
Dr. M. Müller	1.2261	Flow distributor structure for a fuel cell
Dr. M. Müller	1.2262	Fuel cell system and process for operating the same
Prof. D. Stolten	1.2268 G A	Procedure for determining flow density distribution in a fuel cell stack
M. Schonert	1.2291	Structured electrodes for use in a fuel cell
Dr. M. Stähler	1.2295	Direct alcohol fuel cell stacks with a carbon dioxide separator

Patents granted

Principal inventor	**PT**	**Title**
Dr. H. Dohle	1.2129	Process for determining current density distribution in fuel cells
Dr. H. Dohle	1.2129 EP	Process for determining current density distribution in fuel cells

3.2 Key topic: solid oxide fuel cells

3.2.1 Objectives and fields of activity

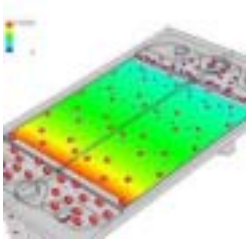
Due to its high operating temperatures and its ability to use carbon-containing fuel gases, the solid oxide fuel cell (SOFC) is envisaged for applications in the field of decentralized energy supply, including smaller systems, such as domestic energy supply, as well as use as an auxiliary power unit (APU) in vehicles, ships and aircraft. In the Electrochemical Converters / High Temperature department (EWH), at first sight work is being carried out in very different areas, which all serve the over-arching aim of using the findings obtained on SOFC performance to derive proposals for improving the performance under the conditions of the different applications.

The electrochemical operating test must be seen as a first very important indication of the functionality of SOFCs. At operating temperature (in the laboratory this means in a furnace) the SOFC is supplied with fuel gas and air, and the power output is measured as a function of load. First and foremost, the absolute value of the electric power is determined, but more important in terms of further development is the comparison of the results of different cells and the analysis of the influence of cell materials and/or manufacturing procedures.

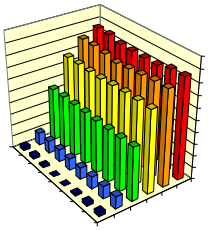
In order to accelerate the development process for stacks, special measurement setups and measuring techniques are used enabling a faster and simpler investigation of the interaction between various stack components. Thus, for example, the interaction between the metallic interconnect and the glass sealant can be investigated, in the usual fuel cell atmospheres (air, fuel gas) while at the same time the electrically insulating function of the glass sealant is verified by resistance measurements.

The different applications make different demands on the service life of a fuel cell. Thus, for example, a minimum service life of 40,000 h (5 years) is required for stationary applications. During this time, the fuel cell must not lose more than 10 % of its initial power. To this end long-term tests with a constant current are carried out on single cells and stacks (up to 10,000 h in the laboratory). Besides the current, the temperature and the fuel gas composition are also important parameters whose influence on performance is being investigated.

The SOFC is also exposed to so-called load changes, when the current consumed changes. Much more stringent are the demands made on the materials and their mechanical stability when the SOFC is switched off. In real systems, the temperature of the stack will decrease and the stack must be brought to operating temperature as rapidly as possible for a restart. These processes are also mainly performed on stacks to investigate technically interesting systems, since the stack must not lose more than 10 % of its power after, for example, 100 cycles.



In addition to experimental studies, SOFC operation is also simulated by mathematical modelling. The processes in the SOFC (charge, heat and mass transport) are described by partial differential equations. The models can describe the geometrical structures in a very detailed (three-dimensional coordinates) manner or it is possible to focus on only one coordinate along which the processes of interest mainly occur. Considering only one coordinate leads to a substantial reduction of model size, and therefore of computing time, so that these models can be used as components in higher level tools for systems simulation. Electric current, temperature and concentration distributions are determined by simultaneously solving all partial differential equations numerically (simulation calculations). The operating behaviour of an SOFC can thus be predicted, so that optimized approaches are provided for rating design concepts and accelerating development work.



Modelling work is complemented in special areas by experimental work, either to determine the input parameters for the calculations or to verify the calculation results. Thus, for example, mass transport parameters in porous bodies (anode substrate, cathode layer) are selectively determined by diffusion and permeation measurements, which are then incorporated in simulation calculations. Moreover, in special measurement setups, the flow distribution in the manifold is determined across the cell by pressure loss and pressure distribution measurements. The measurement setup permits a simple and rapid variation of the manifold, so that the geometry for homogeneous mass distribution can be optimized.

The Fuel Cell Process Engineering group is involved with the layout design, and construction of test set-ups and their components as well as process engineering analyses. Its chief responsibilities are:

Testing plant components - for the specific boundary conditions connected with operation at temperatures between 600 and 900 °C the essential components of an SOFC system, such as recuperative heat exchangers, pre-reformers or after-burners, are not commercially available. Test facilities are therefore constructed in which both external products and products developed in house are characterized and tested for their suitability.

Testing and optimizing the control and feedback control concepts for SOFC and DMFC facilities: Fuel cells require complex plant and process engineering. To ensure that this equipment can be operated reliably, the various operating states can be controlled safely and that damage to the equipment and the fuel cells can be avoided, special control and feedback control concepts are required. These procedures are developed on the basis of experience of tests with cells and stacks and are tested and refined in the existing facilities.

Development and construction of facilities: to demonstrate the feasibility of the technology and to test the interaction between all the components, an SOFC demonstration plant was developed and built that was designed to produce 20 kW of electrical power when run on natural gas. DMFC plant engineering is being tested and further optimized with the development and construction of DMFC systems up to 5 kW.

Calculation and evaluation of system concepts: experimental results form the basis for stationary and dynamic simulation calculations carried out with different tools that can be used to evaluate different plant concepts and to do the layout of individual system components. The plants are analysed in terms of both efficiency and cost.

3.2.2 Important work results

3.2.2.1 Modelling and simulation

At IEF-3 the simulation of fuel cells and fuel cell stacks is performed at two levels of abstraction: SOFC models embedded in the computational fluid dynamics (CFD) tool FLUENT permit the simulation of the flow distribution, temperature profiles, gas concentrations and the current density distribution in three spatial dimensions. Even with a sophisticated continuum-approach ("Integrated Volume"), which makes the simulation for example a 60-cell stack on a standard computer feasible, simulation times are in the order of several hours or even days. SOFC models embedded into applications of balance-of-plant analysis require computing times in the range of seconds or below. In this case, a higher level of abstraction is needed that leads to a 1D model. The "Integrated Volume" approach of the CFD model is verified by the simulation of a single pair of fuel and air channels, which is within the domain of both models. In the verification the simulated electrical power and the temperature distribution along the flow channels are compared. The electrical power computed with the 1D model only deviates by 0.5 % from the value obtained with the CFD model. The comparison of the temperature distribution is shown in Fig. 30. A very good agreement can be observed in case of the temperature of the fuel gas, whereas there is a deviation of about 2 K (0.25 % deviation) for the bipolar plate and about 4 K (0.5 % deviation) for the air. In view of the fact that a comparison of two independent methods, one in 1D and the other in 3D, is presented, the deviation can be regarded as very small. This validation shows that both models can be applied for the simulation of SOFC stacks.

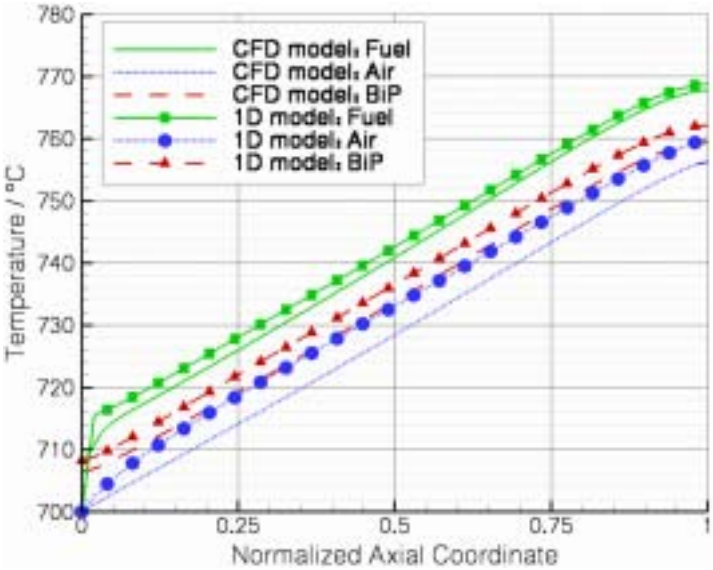


Fig. 30: **Comparison of the temperature distribution in the fuel channels, the air channels and the bipolar plates (BiP) obtained with the CFD model and the 1D model. Flow is shown from left to right**

For the validation of the 3D CFD model, measured temperature profiles of a 60-cell SOFC stack were compared with simulation results. At FZJ, temperature profiles from inside a running SOFC stack can be obtained by embedding special interconnector plates into the SOFC stacks. Fig. 31 shows the comparison of a measured temperature profile of an SOFC stack with both the simulated temperature profiles of the detailed CFD model and the 1D model. The resulting temperature profiles of the 1D model show a agreement both with the measured values as well as with the results of the CFD simulation in an adequate range. The current density distributions in Fig. 31 (right) also show the same trend for both models.

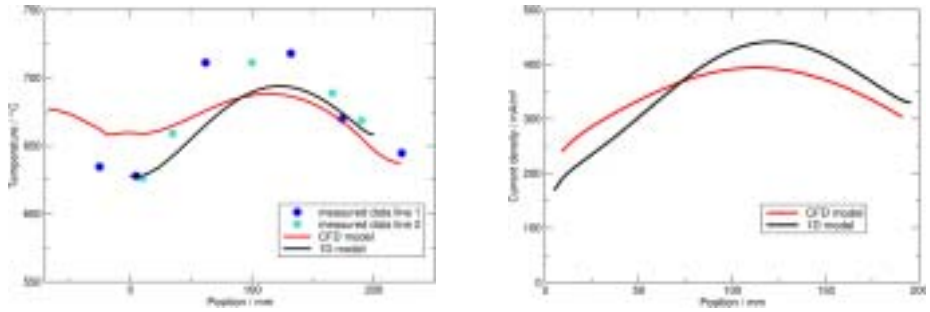


Fig. 31: **Measured and simulated temperature profiles (left) and simulated current density profiles (right) of a 60-cell SOFC stack**

In a more general framework, the modelling group is active as project coordinator of the project 'GenFC' (Generic Fuel Cell Modelling Environment, www.genfc.org) of the European Commission, which aims at an enhancement of the use of modelling techniques and fuel cell models in research and development. This will be achieved by providing fuel cell models and their data sets – preferably with a comfortable graphical user interface. The main tasks are the acquisition and the documentation of existing models, the development of new models in cases where no satisfactory solution exists and the development of a framework (with data base) for the respective models and their data. Until now, software specifications and usage scenarios for the specification of operating sequences supported by GenFC have been established in collaboration with the partners. Furthermore, an interface concept between fuel cell models, fuel cell systems and fuel cell applications was developed and a data model for representing the work flow in GenFC was specified.

3.2.2.2 Cell and stack development

One of the targets in the development of solid oxide fuel cells (SOFCs) is to increase the electrochemical performance. During the past ten years significant progress has been made (in co-operation with IEF-1). Starting from 1995 promising results were obtained, as illustrated in Fig. 32. Nowadays, with LSM-type SOFCs, a power output of about 1.0 W/cm² (800 °C / 700 mV) can be achieved, due to optimization of various processing and microstructural parameters. However, at operating temperatures lower than 750 °C other types of cathode materials are required with higher electrocatalytic activity. Consequently, research was also started on the application of LSCF cathodes. Here as well, significant progress has been made during the last six years. Fig. 32 shows the development status of LSCF-type SOFCs. Today, with this type of SOFC, a power output of about 1.8 W/cm² (800 °C / 700 mV) and about 1.2 W/cm² at 700 °C (700 mV) can be achieved.

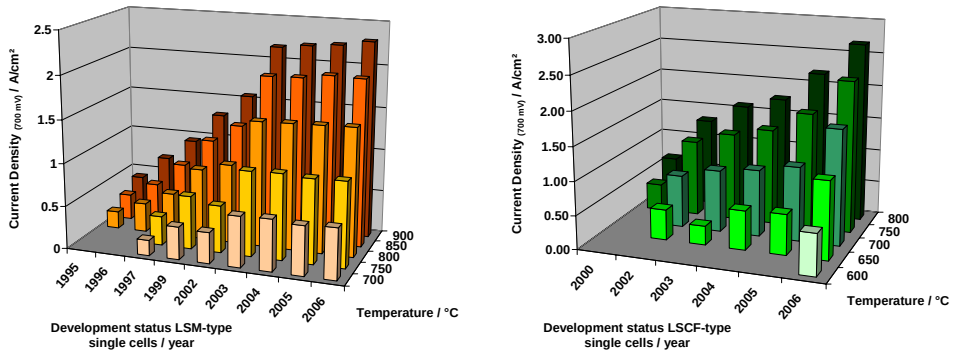


Fig. 32: Status of performance development of anode-supported SOFCs with (a) LSM (left) and (b) LSCF cathodes (right)

In addition to the improvement of the electrochemical performance, long-term durability is another main issue. FZJ is working on improved SOFCs with 40,000 h of stationary operation. At the moment, degradation rates of 1 % / kh can be achieved for an extended operation period of several thousands of hours (see Fig. 33). Longer periods with lower degradation rates are at present one of the main goals.

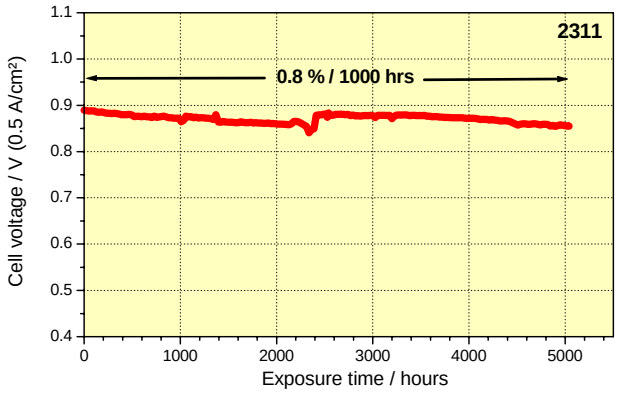


Fig. 33: Cell voltage versus operation time for an anode-supported SOFC with LSCF cathode (16 cm²) under constant current load (0.5 A/cm²) at 800 °C

As part of the Integrated Project 'Real-SOFC' of the European Commission, four identical SOFC stacks each with 2 planar anode-supported cells were operated for up to 3000 h under varying fuel and electrical load conditions. The variations comprised the externally applied current density (0.3 or 0.5 A/cm²) and the fuel gas (hydrogen or methane). All four stacks were operated galvanostatically at a temperature of 800 °C and a fuel utilization of 40 %. A pronounced difference in degradation behaviour was observed between the two stacks operated at 0.5 A/cm² and those operated at 0.3 A/cm² (see Fig. 34 and Fig. 35). The latter displayed linear behaviour of the voltage with time over the full duration of operation. The degradation rate was always below 1%/kh. From the start, the two stacks operated at 0.5 A/cm² showed an increased degradation rate (> 2 %/kh).

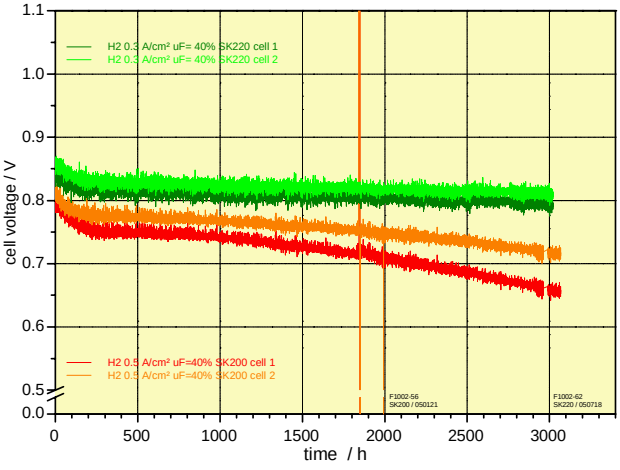


Fig. 34: Time dependence of the cell voltages during constant current operation (0.3 A/cm² for stack #2, 0.5 A/cm² for stack #1) at 800 °C with humidified hydrogen as fuel

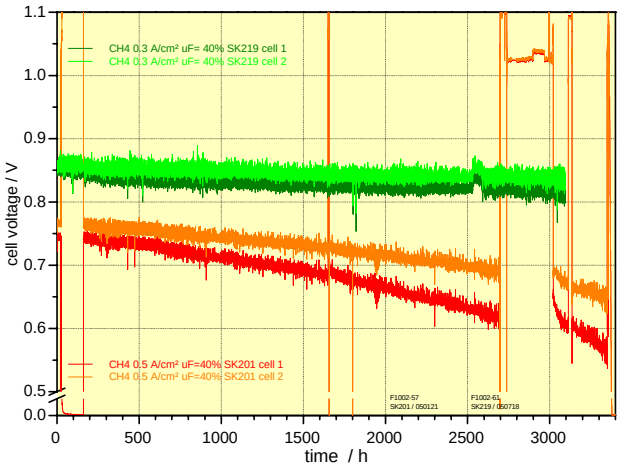


Fig. 35: Time dependence of the cell voltages during constant current operation (0.3 A/cm² for stack #4, 0.5 A/cm² for stack #3) at 800 °C with methane/steam (S/C=2) as fuel

Post-test characterization of the cathode with respect to chromium poisoning was performed on cells from all four stacks. However, no clear relationship between degradation rate of the stacks and amount of Cr incorporated in the cathode could be established. The major difference between the stacks operated at 0.3 and 0.5 A/cm² was the changed microstructure of the cathode in the region near the electrolyte interface; in the stacks operated at lower current densities the structurally changed zone was clearly thinner than in those stacks operated at higher values.

To compare degradation rates at different electrical loads, the transient behaviour of the SOFCs can be described by the area specific resistance (ASR). The ASR can be interpreted as the decisive parameter that characterizes the degradation, i.e. the performance loss over time, of the stack. With the aid of the computer code ORMS (Ohmic Resistance from Long Term Measurements of SOFC Stacks) the ASR can either be calculated on the basis of previously measured data or directly during a long-term experiment still in progess. For the determination of the ASR using the slope of an I-V curve, this curve should be measured with low fuel utilization $U_f < 14$ % and high water vapour content (about 50 vol % H_2O). ORMS was made available to the partners of the EU project RealSOFC for testing.

3.2.2.3 System development and verification

20 kW SOFC system:

The design of a 20 kW SOFC plant, developed in 2003 and 2004, is now to be constructed. The design work and preliminary investigations for the design concept of the 'integrated module', comprising all the components of the plant that operate at temperatures between 500 °C and 950 °C, were concluded in 2004. In early 2005 all of the technical drawings were ready and the components were then manufactured by ZAT-MF. Numerous problems arose during the subsequent metallic vacuum soldering, however, which meant that a good deal of post-production work and additional preliminary tests were required. Whilst the components of the integrated module were being qualified, the design of the plant was completed, the

process engineering components were designed and selected and the plant was assembled. When a sufficient number of the components of the integrated module had been manufactured, the start-up process was commenced. Unfortunately this process had to be interrupted, as problems emerged with the electrical heating plates used to heat the system, so a new heating plate design was required and the plates had to be remanufactured. Details of the 20 kW system are given in Chapter 1.2.

1 kW SOFC-System:

As part of the EU project CexiCell, a 1 kW domestic energy supply system is to be built in which stacks containing newly developed fuel cells are to be tested. After the company originally selected to build the system was unable to proceed with the work, IEF-3 took over the development and construction of this plant. The plant concept is based on the development work for the 20 kW plant. The construction of the integrated 1 kW module is based on that of the 5 kW module (see Chapter 1.2). It contains all the components of the 5 kW module (being based on cells measuring 20 x 20 cm²) and is based on the same measurements as the F10 stack design (cells measuring 10 x 10 cm²).

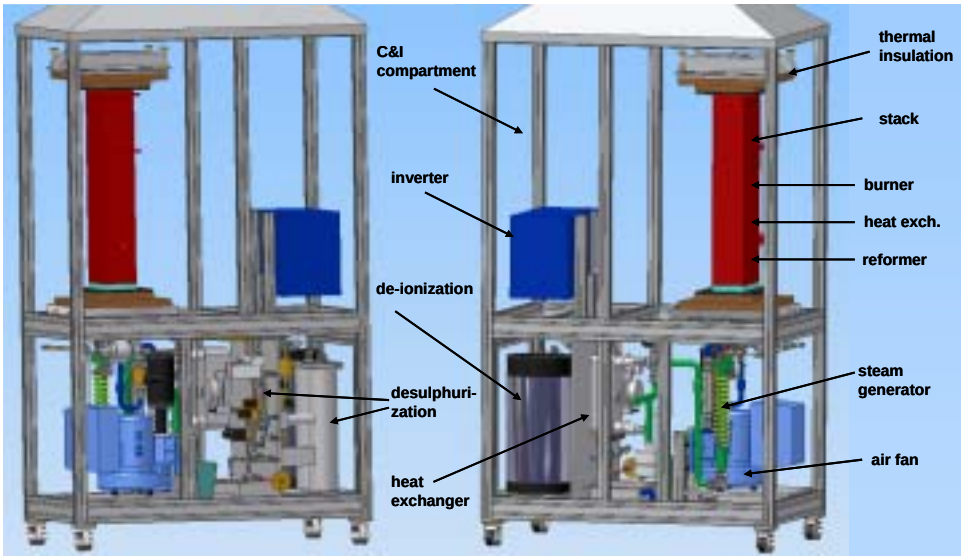


Fig. 36: Detailed design of 1 kW system

The fluidic layout design of the manifold in the stack, the pre-reformer, air pre-heater, stack dummy and after-burner was carried out on the basis of system calculations carried out in Cycle-Tempo. Only short stacks are currently manufactured according to the existing F10 stack design. To be able to manufacture larger stacks, the size of the gas distribution manifolds within the stack must be increased. The manifold dimensions were recalculated. It was possible to configure the geometry of the larger manifold in such a way that no changes had to be made to the external dimensions of the stack. The new stack design was designated F'10. The other components of the module were constructed analogously to

those of the 5 kW module. Manifold geometries that ensured a sufficient gas supply were established here as well. The heating plates for the integrated 1 kW module of the CexiCell plant were fitted with four heating cartridges, as a departure from the original concept for the 5 kW module, since this resulted in significantly lower manufacturing costs. Each cartridge is capable of generating a thermal output of 400 W. The design of the system is shown in detail in Fig. 36.

The system components were manufactured in ZAT-MF and then printed with metal solder in IEF-1 and vacuum soldered in ZAT. All components show high tightness; however, the results of the tests carried out on the reformer soldering for the 20 kW are not yet available. As part of CE certification the plant was analysed using the Safexpert software package. The assembly of the mechanical part of the system has been completed. The next step is to wire up the components and devise the system controls.

STEAG-SaarEnergie

As part of the project with the STEAG Saar Energie company to use mine gas in an SOFC, all preliminary tests for preparing mine gas in a reformer and subsequent usage in an SOFC have been completed successfully. The manufacturing documentation detailing the set-up of the test plant was also handed over to Saar Energie.

The next task was to provide a reformer for a 2 kW SOFC stack. The first reformer, which like the stack was joined together with glass sealant showed an excessive degree of leakage when operated in the test stand after delivery, and it therefore could not be used. Another test with glass sealant also failed to produce an acceptable level of cycling resistance, so the design was modified enabling metallic sealant to be used for the reformer. The first reformer with nickel oxide substrates and stainless steel meshes demonstrated unacceptably high leak rates. Annother reformer was therefore manufactured with reduced substrates (no risk of oxygen release in a vacuum at 1150 °C) and nickel meshes (no chromium evaporation during soldering). This reformer could be soldered successfully and has now been in operation at STEAG Saar Energie for over 1000 h and more than 15 temperature cycles without incident.

Although the stacks initially envisaged for STEAG Saar Energie demonstrated that they had the required performance, excessively high leakage occurred during glass soldering, and various measures were taken to remedy this problem. Finally, a 10-level stack with LSCF cells was supplied that showed the requisite tightness under operating conditions in the furnace at FZJ. The stack was installed in the test stand at Saar Energie by FZJ staff. The installation, bracing, connection of the measuring elements, insulation and connection to the power supply did not involve any problems. It was only when the mine gas was connected, after the system had been warmed up, that operation had to be interrupted after a short period. The pressure difference in the system between the anode and the cathode side, which were higher than during testing at FZJ, meant that leakage rose to an unacceptable extent and further operation of the stack became impossible. Work is continuing on further improving joining technology and on optimizing the plant in order to minimize pressure difference.

Heat exchanger test rig:

As part of the BMW project to develop an APU for use in cars, IEF-3 is carrying out fast temperature cycle tests for Behr, the company developing compact heat exchangers. In

order to do this an existing facility rig for high-temperature heat exchangers (T_{max} = 900 °C) had to be modified in such a way that two heat exchangers run in parallel could be heated from room temperature to 900 °C within 4 minutes and then cooled back down at different rates. To do this, SPS controls, a new measuring and operating system, two compressors and several valves were installed, and the test stand was CE certified. The temperature curve of the test programme designed in collaboration with Behr is shown in Fig. 37.

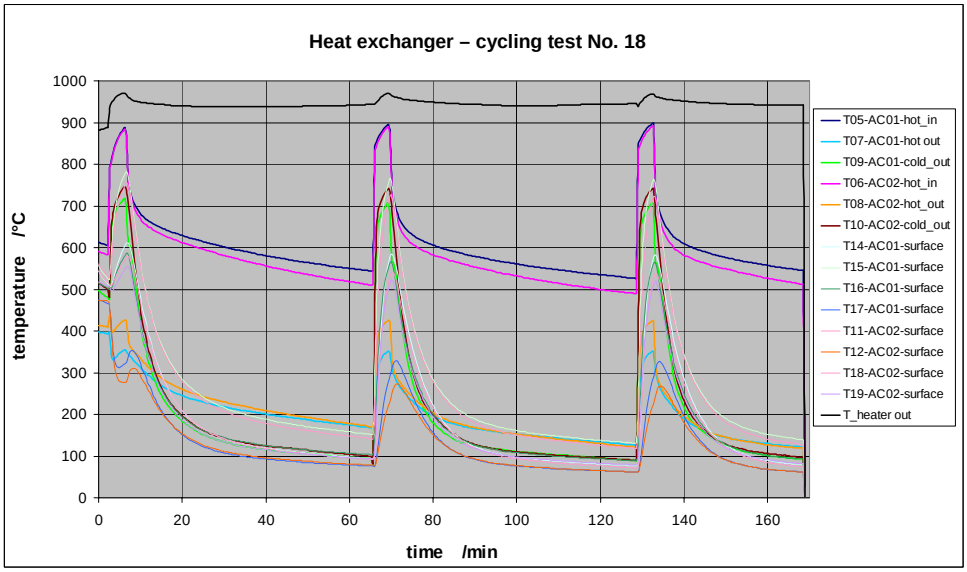


Fig. 37 Cycling programme to test heat exchangers for use in APUs

In the meantime 10 pairs of heat exchangers have been tested for 200 cycles each (4 mins heating up, 60 mins cooling down). After every 50 cycles the heat exchangers were removed and checked for tightness.

3.2.3 Scientific staff and fields of activity

Name	Telephone no (+49-2461-61-) E-mail address	Field of activity
Dr. L.G.J. de Haart	6699 l.g.j.de.haart@fz-juelich.de	Head of the Electrochemical Converters/High-Temperature department (EWH)
Prof. L. Blum	6709 l.blum@fz-juelich.de	Head of Fuel Cell Process Engineering department (VBZ)

P.H. David	4652 p.h.david@fz-juelich.de	Electrical engineering, measurement data recording and systems control
R. Deja	5291 r.deja@fz-juelich.de	Plant simulation, development and testing of SOFC plant components
D. Froning	6676 d.froning@fz-juelich.de	Computer science, software engineering, modelling
Dr. V.A.C. Haanappel	4656 v.haanappel@fz-juelich.de	Chemical technology, (high-temperature) corrosion
Ro. Peters	4664 ro.peters@fz-juelich.de	Head of the SOFC System Technology Group, component development, plant design, construction and testing
Dr. U. Reimer	3537 u.reimer@fz-juelich.de	Electrochemistry, software development, modelling
Dr. M. Spiller	1573 m.spiller@fz-juelich.de	Physics, software development, CFD modelling
Dr. I.C. Vinke	6688 i.c.vinke@fz-juelich.de	Electrochemistry, solid state ionics, high-temperature electrochemistry

3.2.4 Important publications, PhD theses and patents

Important publications

Blum L., Steinberger-Wilckens R., Meulenberg W. A., Nabielek H.

Worldwide SOFC Technology Overview and Benchmark

International Journal of Applied Ceramic Technology, Vol. 2, No. 6, 2005, 482–492

Solid Oxide Fuel Cells (SOFC) are generally considered to be 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. Nevertheless, their development still faces various problems with high temperature materials, design of cost-effective materials and manufacturing processes, and efficient plant design.
This paper summarizes world-wide efforts in the field of SOFC, presenting an overview of the main SOFC designs and the main developers active in this field. Based on data published in proceedings of international conferences during the last few years, a comparison is made of the results achieved in cell, stack and system development.

Blum, L.; Peters, Ro.; Deja, R.; Haart, de L.G.J.; Dengel, A.; Dörr, H.; Gross, B.

Reformer and Stack Development for Use with Coal Mine-Gas

7th European Solid Oxide Fuel Cell Forum, 03.-07.07.2006, Lucerne, Switzerland: CD-ROM proceedings SOFC orals, Durability and Integrity. - 2006. - P1103

Apart from natural gas there is another important natural source of methane. The so-called coal mine-gas is a by-product of the geo-chemical process of the carbonization of sediments from marsh woods of the Earth's Carboniferous Period. Methane evaporates from the coal and has to be removed out of the mines where it represents one of the main safety risks. Also in abandoned mines methane is produced. In the Saarland in Germany there exist more than 100 km of pipeline for the drained coal mine gas. The content of methane varies between 30 and 90 % while there can be oxygen content up to 10 % in the gas. This wide range of fuel content and the oxygen cause a lot of problems for the use in conventional machines. Therefore the utility STEAG SaarEnergie AG is interested in using SOFC with coal mine-gas as efficient as possible to produce electric power. For that purpose at Forschungszentrum Jülich the available SOFC technology was adapted to the use with coal mine-gas and a test bench was designed to operate a reformer together with a 2 kW stack. Within this contribution the design of test bench and reformer and test results are presented.

Gubner, A.

Non-Isothermal and dynamic SOFC Voltage-Current Behavior

Solid Oxide Fuel Cells (SOFC IX) / ed.: S. C. Singhal, J. Mizusaki. - Pennington, NJ, 2005. - (Proceedings of the Electrochemical Society; 2005-07). - 1-56677-465-9. - pp. 814 - 826

Mathematical modeling aiming at SOFC design and operating condition optimization is conducted at the Forschungszentrum Jülich. Computational Fluid Dynamics (CFD) software is used for three dimensional (3D) stack modeling and a rapidly executing self-written one dimensional (1D) code for calculating "along-the-channel" profiles of e.g. the gas temperature of the fuel and the air, of the solid body, which is mainly composed of the bipolar (interconnector) plate, of the gas compositions (e.g. in molar fractions) and other interesting quantities. This co-simulation approach using both the 3D and the 1D tool was presented at the SOFC VIII already. This contribution discusses results regarding the non-isothermal Voltage-Current (V-I) behavior including dynamic load changes produced by using the 1D tool.

Gubner, A.; Saarinen, J.*; Ylijoki, J.*; Froning, D.; Kind, A.; Halinen, M.*; Noponen, M.*; Kiviaho, J.*

Dynamic Co-Simulation of a Solid Oxide Fuel Cell (SOFC) and the Balance of Plant (BoP) by Combining an SOFC Model with the BoP-Modelling Tool APROS®

Proc. 7th European Solid Oxide Fuel Cell Forum (Ed., U. Bossel), Lucerne 2006, CD-ROM

An increasing demand for co-simulation capabilities for modelling a planar Solid Oxide Fuel Cell (SOFC) stack as a part of a fuel cell system is caused by the recent progress in developing planar SOFC stacks. These SOFC stacks need balancing with a support system composed of all process engineering components, e.g. heat exchangers, of the targeted power plant application, and modelling can assume an important role in the design process of that application's fuel cell system. However, any modelling effort will only be relevant if the SOFC behaviour can be simulated with a reasonable degree of detail so that the results can provide useful design information. In order to facilitate the SOFC power plant development efforts the VTT Technical Research Centre (VTT), the Forschungszentrum Jülich GmbH (FZJ) and

Wärtsilä Corporation have conducted a joint project in 2005 for redesigning the interfaces of a one dimensional (1D) SOFC modelling software tool provided by the FZJ to match the interfaces of a systems modelling software tool called Advanced Process Simulation environment APROS® developed by VTT and Fortum Nuclear Services. The 1D SOFC model can now be used as a plug-in software component for APROS®. This contribution introduces the resulting co-simulation environment and discusses some simulations of an example application.

Gubner, A.; Nguyen-Xuan, T.; Bram, M.; Remmel, J.; Haart, L.G.J. de

Lightweight Cassette Type SOFC Stacks for Automotive Applications

Proc. 7th European Solid Oxide Fuel Cell Forum (Ed., U. Bossel), Lucerne 2006, CD-ROM

In cooperation with the Forschungszentrum Juelich GmbH and a national consortium of component suppliers, the BMW Group is developing Solid Oxide Fuel Cell (SOFC) technology as Auxiliary Power Units (APU) in passenger cars. Considering key challenges of the automotive application like rapid start-up, large number of thermocycles, compact construction and low cost, a lightweight cassette design has emerged allowing the mounting of high power density anode supported SOFCs. The design is based on modeling results which were conducted to optimize gas flow distribution and reduce contact resistance. The flow distribution was investigated by using commercial Computational Fluid Dynamics (CFD) software and validated by experiments, whereas the contact resistance was analyzed by means of commercial Finite Element software. The results provide a design guideline to optimize the geometry of the interconnect. Cost-effective embossing and punching technologies were used to produce the metal components of the cassettes to be completed by laser beam welding. Up to now, stacks with 2, 5, 10 and 20 levels have been assembled using conventional glass ceramic seals. First results of stack operation are presented covering tests under stationary conditions for more than 3000 h as well as thermocycles with heating rates up to 25 K/min.

Haanappel, V.A.C.; Shemet, V.; Vinke, I.C.; Quadakkers, W.J.

A novel method to evaluate the suitability of glass sealant-alloy combinations under SOFC stack conditions

Journal of Power Sources, 141 (2005), 102 - 107

A novel test method has been developed to evaluate the suitability of combinations of sealing materials and alloys under simulated SOFC stack conditions. This method is based on test samples of two metallic sheets, joined together with a glass or glass–ceramic sealant. The outer side of the sample is exposed to ambient atmosphere, whereas the inner side can be exposed to different gas compositions. The whole set-up is placed in a furnace. Optionally, an external voltage can be applied across the sheets. The background of the experimental design was the development of a methodology to investigate interaction phenomena between steel alloys and glass ceramic sealants under controlled conditions. Although exposure and oxidation experiments under atmospheric conditions did not show any indication of detrimental effects, corrosion was observed in actual stack experiments. The experimental set-up was thus designed in order to study the influence of single parameters on the interaction of alloy and sealant. With this method the governing influences in simulated SOFC operation can be identified. Obviously, the presence of a dual hydrogen–air atmosphere induces enhanced corrosion in certain material combinations. The experiments presented here allow a systematic study of the materials in question and a rapid characterisation.

Haanappel, V.A.C.; Mertens, J.; Rutenbeck, D.; Herzhof, W.; Sebold, D.; Tietz, F.

Optimisation of Processing and Microstructural Parameters to Improve the Electrochemical Performance of LSM-Based Anode-Supported SOFCs

Journal of Power Sources 141 (2005) 216 – 226

To improve the electrochemical performance of LSM-based anode-supported single cells, a systematic approach was taken for optimising processing and materials parameters. Four parameters were investigated in more detail: 1) the LSM/YSZ mass ratio of the cathode functional layer, 2) the grain size of LSM powder for the cathode current collector layer, 3) the thickness of the cathode functional layer and the cathode current collector layer, and 4) the influence of calcination of YSZ powder used for the cathode functional layer.
Results from electrochemical measurements performed between 700 and 900 °C with H_2 (3 vol.% H_2O) as fuel gas and air as the oxidant showed that the performance was the highest using an LSM/YSZ mass ratio of 50/50. A further increase of the electrochemical performance was obtained by increasing the grain size of the outer cathode current collector layer: the highest performance was achieved with non-ground LSM powder. In addition, it was found that the thickness of the cathode functional layer and cathode current collector layer also affects the electrochemical performance, whereas no obvious detrimental effects occurred with the different qualities of YSZ powder for the cathode functional layer. The highest performance, i.e. 1.50 ∂ 0.05 A/cm² at 800 °C and 700 mV, was obtained with a cathode functional layer, characterised by an LSM/YSZ mass ratio of 50/50, a d_{90} of the LSM powder of 1.0 µm, non-calcined YSZ powder, and a thickness of about 30 µm, and a cathode current collector layer, characterised by d_{90} of the LSM powder of 26.0 µm (non-ground), and a thickness of 50–60 µm. Also interesting to note is that the use of non-ground LSM for the cathode current collector layer and non-calcined YSZ powder for the cathode functional layer obviously simplifies the production route of this type of fuel cell.

Haanappel, V.A.C.; Shemet, V.; Gross, S.M.; Koppitz, Th.; Menzler, N.H.; Zahid, M.; Quadakkers, W.J.

Interactions of Various Glass-Ceramic Sealants with Ferritic Steels under Simulated SOFC Stack Conditions

Journal of Power Sources 150 (2005) 86 - 100

The suitability of various combinations of glass-ceramic sealants with high-chromium ferritic steels under conditions simulating SOFC stacks has been evaluated, i.e. three glass-ceramic sealants and seven types of ferritic steels. The test method used is based on test samples consisting of two metallic sheets, joined together with a glass-ceramic sealant. The outer side of the specimens was exposed to air for 400 h at 800 °Cr, whereas the inner side was exposed to hydrogen saturated with 3 vol% water vapour. In particular, attention is paid to the influence of small amounts of additives to both the glass-ceramic sealant and the steel on the electrical and chemical behaviour of the specimens. In the case of the glass-ceramic sealants, in particular the effect of small amounts of PbO, and, in the case of the ferritic-type steels, the effect of the presence of small amounts of Si and Al are studied in more detail.
Under experimental conditions simulating SOFC stacks, it was obvious that excessive internal Cr oxidation of the ferritic steels, sometimes accompanied by external Fe-oxide formation, only occurred in the case of glass-ceramics containing minor amounts of PbO. This internal oxidation finally resulted in a volume change of the ferritic steel, which was manifested in bulging of the steel. As a consequence, the glass-ceramic was pushed away from the steel surface and crack formation at the glass-ceramic – steel interface occurred. The rate of corrosion attack strongly

depended on the detailed steel composition. Increasing Si content apparently increases the rate of the corrosion attack, and thus possibly decreasing the time for the occurrence of short-circuiting.

Haanappel, V.A.C.; Mertens, J.; Mai, A.

Performance Improvement of (La,Sr)MnO_3- and (La,Sr)(Co,Fe)O_3-Type Anode-Supported SOFCs

Proceedings of the 1st European Fuel Cell Technology & Applications Conference, 14-16 December 2005, Rome, Italy,
Journal of Fuel Cell Science and Technology 3(3) (2006) 263 - 270

Targets in the development of anode-supported or planar solid oxide fuel cells (SOFCs) are low operation temperatures, high durability, high reliability, high power density, and low production costs. During the past ten years steps have already been taken at Forschungszentrum Jülich to lower the operating temperatures while maintaining the power output. This was achieved by optimising processing and microstructural parameters of the electrodes. This paper will present the latest results concerning performance improvement through variations of the processing route and the microstructure of $La_{0.65}Sr_{0.3}MnO_3$ (LSM) and $La_{0.58}Sr_{0.40}Co_{0.2}Fe_{0.8}O_{3-ŭ}$ (LSCF) type SOFCs.
In the case of the LSM-type single cells, the following aspects relating to the electrochemical performance were investigated in more detail: 1) production of the anode substrate by tape casting versus warm pressing, 2) deposition of the anode functional layer (AFL) and electrolyte by screen printing versus vacuum slip casting, 3) use of non-calcined and non-ground YSZ for applying the cathode functional layer (CFL), and 4) sintering temperature of the CFL and cathode current collector layer (CCCL).
In the case of LSCF-type cells, a systematic approach was initiated for optimising the $Ce_{0.8}Gd_{0.2}O_{2-ŭ}$ (CGO) diffusion barrier layer: 1) deposition techniques of the CGO layer, and 2) sintering temperature of the screen-printed CGO layer.
Results have shown that certain modifications of the processing route led to a slightly lower electrochemical performance, whereas others did not affect the performance at all. Regarding LSCF-type SOFCs, a slight improvement of the performance was achieved by optimising the sintering temperature of the CGO layer.

Haanappel, V.A.C.; Mai, A.; Mertens, J.

Electrode Activation of Anode-Supported Solid Oxide Fuel Cells with LSM- or LSCF-Type Cathodes

Proceedings of the 15th International Conference on Solid State Ionics, 17-22 July 2005, Baden-Baden, Germany,
Solid State Ionics 177 (2006) 2033 - 2037

Electrode activation of SOFCs refers to one or more processes, which generally occurs during a first period of usually not longer than 100 h of applying a constant electrical load, and is associated with a decrease of the area specific resistance. To study this effect in relation to different starting-up and test conditions in more detail, the electrochemical performance between 650 and 900 °C of two types of anode-supported single cells, one with an $La_{0.65}Sr_{0.3}MnO_3$ (LSM) and the other with an $La_{0.58}Sr_{0.40}Co_{0.2}Fe_{0.8}O_{3-ŭ}$ (LSCF) cathode, was evaluated.
Both types of cells, when heated to and reduced at 800 °C, showed a decrease of the area specific resistance during the first 70 h at 800 °C of exposure under constant electrical load. The initial area specific resistance of cells heated to and reduced at 900 °C was comparable with that of the cells reduced at 800 °C and exposed for 70 h at 800 °C under constant electrical load. In other words, electrode activation is much faster at 900 °C than at 800 °C.

Lokurlu A., Blum L., Riensche E.

Two SOFC development strategies for pure CO_2 production and for partly syngas/H_2 production with very effective cooling by internal reforming

Proceedings of the Int. Hydrogen Energy Congress, 13-15 July 2005, Istanbul, Turkey

This paper discusses technical and economical aspects of using Solid Oxide Fuel Cells (SOFC) for CO_2 sequestration and for H_2 production, where the SOFC only partly produces power directly. Specifically a conceptual design study involves the combination of SOFC with an electrochemical afterburner (ECAB) to reach the high CO_2 concentration of about 90 mol-% in the exhaust gas that is required for efficient CO_2 sequestration.
The other SOFC development strategy is based on low fuel utilisation in the cells. When operating with nearly complete internal reforming and only partly electrochemical oxidation the cooling effect by internal reforming is essentially enlarged. This is very advantageous with respect to the demand of cooling air. In addition the Nernst potential is increased, because the SOFC now completely operates under high H_2 concentrations. Both effects will reduce the specific SOFC investment costs. In this case syngas leaves the SOFC system and can be processed in further steps to produce pure H_2 or to use the syngas for other processes.
A commercial simulation program (PRO/II) is used to quantify the influences of flow-sheet variations on plant efficiency and costs. In the first case particularly the process variants involve different types of ECABs (oxygen pump, mixed oxide conductor and second SOFC). By comparison with a conventional reference system without CO_2 capture CO_2 abatement costs are calculated and economic break-even points on a scale of increasing CO_2 tax values, expected for the future, are determined.

Vinke, I.C.; Röwekamp, B.; Tietz, F.; Zahid, M.; Quadakkers, W.J.

Influence of Stack Materials on the Electrochemical Performance of Anode Supported SOFC Single Cells

Solid Oxide Fuel Cells (SOFC IX) / ed.: S. C. Singhal, J. Mizusaki. - Pennington, NJ, 2005. - (Proceedings of the Electrochemical Society; 2005-07). - 1-56677-465-9. - S. 603 - 610

In planar SOFC stacks with ferritic interconnects and anode substrate-supported SOFC cells degradation is still too high for stationary applications. Determination of the sources of this degradation is difficult due to the complexity of the stacks. Methods for separating detrimental influences under conditions closely resembling stack operation and environment must be developed. Using an inert cell housing with exchangeable inserts at the electrodes provides an opportunity to selectively place stack materials close to the cathode. Results from these measurements show a clear poisoning of the cathode by chromia from the ferritic steel. Degradation rates above 20 % in 1000 h are observed for Chrofer22APU-1st (first commercial batch) despite the formation of chromium manganese spinel oxide layer on the surface of the steel that reduces the release of chromium species from the steel. Coating the steel with contact layer material on average reduces the degradation rates close to 0 % for the first 600 h. After prolonged operation for 1800 h a noticeable degradation can also be observed for cells exposed to coated steel. The mechanism of the chromia retention by the contact layer is not yet clear.

Vinke, I.C.; Erben, R.; Song, R.-H.*; Kiviaho, J.*

Installation and Operation of kW-Class Stacks from Jülich in External Laboratories

Proc. 7th European Solid Oxide Fuel Cell Forum (Ed., U. Bossel), Lucerne 2006, CD-ROM

The anode substrate cell based SOFC stack technology from the Forschungszentrum Jülich has reached a maturity that allows for first trials of operation in external laboratories. The trials were conducted with kW-class stacks at VTT in Espoo Finland and at KIER in Daejeon South Korea. In preparation of the actual experiments personal from both institutes received training on stack operation and handling in the laboratories of the Forschungszentrum. Information and pre-requisites on the operational details and test stands were made available to the partners well in advance of the actual experiments.
During the years 2004 and 2005 several tests were performed. At VTT a 1 kW-class and a 5 kW-class stack were installed and operated. At KIER two 0.5 kW-class stacks and two 5 kW-class stacks were installed and operated. Considering the difficulties of assembling and operating stacks in an unfamiliar environment the test results were quite satisfactory. All stacks reached their projected power or more. The lifetime of some of the stacks was however relatively short due to short circuits, leakage or problems with the test stands. One of the 5 kW-class stacks installed at VTT is successfully being operated in a system under varying conditions and fuel compositions. One of the 5 kW-class stacks at KIER was successfully operated at elevated pressure (3.5 bar) which resulted in a 20 % power increase compared to measurements at the same current density under ambient pressure.

PhD theses

Matthias Finkenrath

Simulation and analysis of the dynamic behaviour of power plants with solid oxide fuel cells (SOFC)

Berichte des Forschungszentrums Jülich, Reihe Energietechnik, Volume 44,
ISBN 3-89336-414-5, RWTH Aachen University, 2005

The objective of the work is to develop a dynamic system simulation for SOFC power plants that is as generalizable as possible, in order to analyse interactions between subsystems at a preliminary stage and then to develop strategies for optimum operation. The system simulation developed during the work is based on a common generic modelling approach for all process units. All component models developed were validated using measuring data available at FZJ. The focus of the work was on dynamic system calculations for all operating states, including heating-up, start-up, full load, partial load and shutdown.

Important patents

Patent applications:

Principal inventor	PT	Title
C. Wederhoven	1.2140 EP	Process and device for determining microstructure parameters of thin porous layers

3.3 Key topic: fuel processing systems

3.3.1 Objectives and fields of activity

The use of fuel cells in mobile and stationary applications is based on the availability of hydrogen. At the present time, however, there is still no infrastructure for hydrogen as a future energy source. Hydrogen therefore needs to be extracted from readily available energy carriers. These sources include natural gas and heating oil for stationary applications, and gasoline, kerosene and diesel for mobile applications. For leisure uses, hydrogen must be made available from LPG or propane in camping gas bottles. The energy carriers mentioned are currently primarily produced from the fossil primary energy carrier crude oil. Over the long term, part of the liquid energy carriers required today can be manufactured from biomass.

In IEF-3, work in the field of fuel processing focuses on reforming middle distillates such as kerosene, diesel and light fuel oil. For use in electricity or on-board power generation for portable or mobile applications (APU: auxiliary power unit), the same fuel must be used in the fuel cell APU system as is used to power the vehicle. For aeronautical applications kerosene and jet fuel for small aircraft are the only usable fuels, although they may contain significant amounts of sulphur-containing components. Theoretical and practical work is therefore being done on desulphurizing and vaporizing liquid fuels as part of fuel preparation.

For on-board power supplies in the 5 $kW_{el.}$ power range the emphasis of the work is on autothermal reforming of kerosene, diesel and fuels similar to diesel. During the reforming process, the input material is converted into a hydrogen-rich gas at high temperatures either with water alone, air alone or with a water/air mixture at a solid catalyst. Important elements of the R&D work are process engineering, analyses, component development and system design. All fields of work make appropriate use of modelling. Reformers are being developed both for high-temperature SOFCs and also for low-temperature PEFCs at 80 °C and high-temperature PEFCs (160 °C - 200 °C).

Since the gas produced during reforming (the so called reformate) generally has too high a carbon monoxide content to be used for continuous operation of a PEM fuel cell, the gas is subjected to secondary processing

The staff in the Fuel Processing department have been involved in the reforming of natural gas, methanol, diesel and kerosene since 1990. They have 200 man-years' experience in process engineering and industrial chemistry, particularly in the field of heterogeneous catalysis.

3.3.2 Important work results

3.3.2.1 Desulphurization

At IEF-3, work has been carried out on desulphurizing liquid fuels since 2005. The work focuses on desulphurizing kerosene and EL heating oil with maximum sulphur-containing hydrocarbon contents of 3000 ppm and 2000 ppm, respectively. Once a theoretical analysis had been carried out and procedures that are generally suitable for decentralized desulphurization in fuel cell systems had been selected, the individual procedures were examined on a laboratory scale.

particularly in aircraft. Tests carried out at IEF-3 for thermal separation and adsorptive desulphurization showed more promising results, however. In thermal separation, the fuel is divided via fractional distillation into a pre-desulphurized light fraction and a residue. During tests with kerosene Jet A-1, it was possible to reduce the sulphur content in the light fraction by up to 60 %. The sulphur-enriched residue is channelled back into the turbo-jet engine to be burnt. An adsorption process was developed to purify the light fraction further. After ten different adsorbents had been screened, a newly developed adsorbent was chosen. The chief selection criterion besides adsorption capacity was easy regenerability. The subsequent process optimization showed the great potential of adsorptive desulphurization. Optimizing the adsorption and regeneration conditions enabled the capacity of the selected adsorbent to be increased by a factor of 2.5 (see Fig. 38).

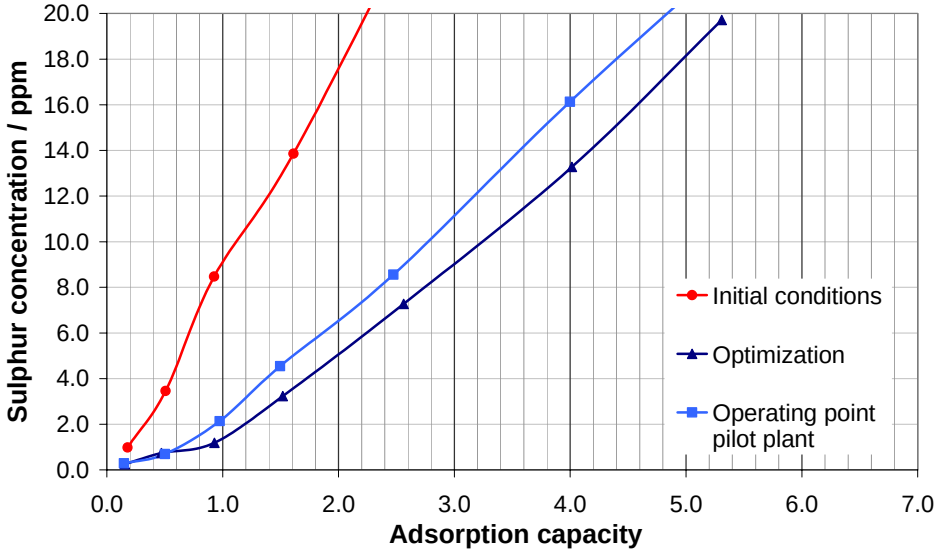


Fig. 38: Optimization and process design for the adsorptive desulphurization of kerosene Jet A-1 using the selected adsorbent

Using this combined process of thermal separation and adsorption, kerosene can be desulphurized to a level of 1650 ppm. An alternative to thermal separation is membrane separation. Five different pervaporation polymer membranes were examined on a laboratory

scale. Under laboratory conditions, this procedure permitted the sulphur content in the permeate to be reduced by over 50 % for kerosene.

3.3.2.2 Reforming

A reformer for middle distillates was developed in IEF-3 over the past few years and is shown in Fig. 39. This reformer is capable of using standard diesel fuel to produce a sufficient quantity of hydrogen for fuel cells with a power of between 3 und 5 kW_{el}. The preferred procedure is so-called here autothermal reforming. The reformer has a cold-start mechanism, an internal steam generator and a mechanism for extracting the heat generated during the reforming process.

In order to gain a better understanding of the flow and temperature conditions within the mixing chamber of the reformer and to be able to predict them more easily, **C**omputational **F**luid **D**ynamics modelling is carried out at Jülich. The objective of this modelling is to provide as precise data as possible as to the temperatures reached in the mixing chamber and the local zones in which the fuel is vaporized. This is based on given reaction conditions such as the temperatures and flow rates of air, steam and diesel fuel and the spray pattern of the nozzle. A detailed description of the modelling and the experimental results obtained, particularly for hydrocarbon conversion and degradation behaviour, is given in Chapters 1.3 and 4.3 below.

Fig. 39: Diesel reformer

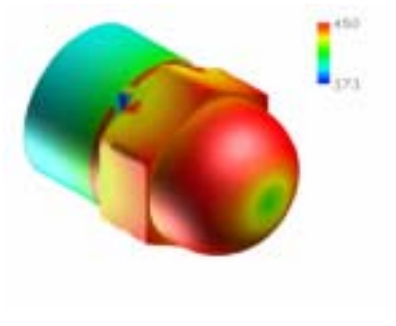


Fig. 40: Temperature distribution at an atomization nozzle as determined by CFD modelling

The upper section of the reformer contains a one-fluid nozzle, which uses a stream of steam to spray the liquid diesel fuel into the chamber. On the one hand, it is important to ensure that the temperature inside the nozzle does not exceed 120 °C in order to avoid pyrolysis and vaporization of the fuel. Pyrolysis causes the nozzle to become blocked. If the diesel begins to vaporize while still in the nozzle, fuel can no longer be added continuously. On the other hand, the temperature of the external surface of the nozzle must not fall below approx. 150 °C, as otherwise the fuel will condense and accumulate on the nozzle (see Fig. 40). This leads to an accumulation of carbonaceous deposits on the nozzle, which over the long term have a negative effect on the nozzle's performance. Fig. 41 shows views of various nozzle heads from 2005 - 2006. For comparison, the nozzle head of an ATR 5 is represented at the commencement of a test run and after approx. 50 h operation with a commercially available

diesel fuel. Comprehensive CFD calculations led to the development of reformers with new mixing chambers (ATR Type 7 and Type 8). The reactor types differ in the nozzle types used and in the design of the mixing chamber. There is almost complete conversion in the reactor types at the beginning of the test series. No carbonaceous deposits were found in either of the mixing chambers or on the external surface of the nozzle, in contrast to the Type 5 reactor. After 500 h the Type 7 reformer showed a dramatic increase in pressure loss through the nozzle. The cause of this pressure loss was found to be crystalline structures inside the nozzle, i.e. in the twist insert of the fuel nozzle of the ATR 7. Fig. 41 (c) shows an SEM image of the deposits. The image covers a width of 4 µm. EDX procedures were used to show that the structure was composed of the chemical elements K, Na, Ca, S and O. An XRD spectrum detected small quantities of potassium sulphate (K_2SO_4) – possibly also potassium sulphite (K_2SO_3) – and calcium sulphite ($CaSO_3$). The source of the deposits was the ash content in the fuel, specified as a maximum of 100 ppm for diesel fuel. The ATR 8 reformer is better adapted in terms of preventing potential deposits of inorganic ashes. Fig. 41 (d) shows the nozzle tip of an ATR 8 after operation for 1000 h with low-sulphur kerosene (Jet A-1). A change in the surface can be observed but there are no deposits which could affect nozzle function.

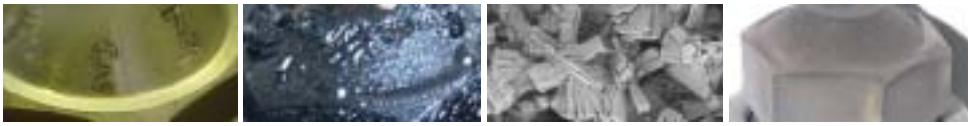


Fig. 41: Deposits occurring during diesel and kerosene reforming. From left to right: (a) New nozzle); (b) Carbonaceous deposit after 50 h operation with a diesel fuel in a Type 5 reformer in early 2005; (c) Ash deposit from diesel fuel in the interior of the nozzle from a Type 7 reformer in late 2005; (d) Nozzle head of a Type 8 reformer after 1000 h operation with low-sulphur kerosene Jet A-1 in late 2006

3.3.2.3 System development

One important tool for system development is the modelling of all basic components of fuel cell systems involving reforming. This modelling covers SOFC, PEFC and HT-PEFC fuel cells. The emphasis is on modelling all subcomponents, primarily components with integrated heat exchange mechanisms and phase transformations, and on static and dynamic system simulations. The aspects examined are the start-up process and behaviour during load change. The main findings are discussed below:

- The SOFC is the simplest system and needs the shortest start-up time for the reformer, but the longest start-up time for the fuel cell.
- During system start-up, steam must be supplied via additional components. An ignition boiler is twice as rapid as an electric heating cartridge but requires 25 % more energy.
- Owing to the temperature fluctuations during load change the shift reactor cannot always function at optimum operating temperatures, and the CO content at the outlet rises from 1 % to a maximum of 2.5 %. The PEFC system with preferential CO oxidation (PROX) requires a PROX reactor with advanced temperature regulation.

- The HT-PEFC provides a good compromise between SOFC and PEFC. The high CO tolerance of the cells in particular facilitates system development.

The coupling of autothermal reforming with catalytic combustion was examined in experiments carried out on the ATR 5C reactor. The ATR 5C consists of a reformer and a catalytic burner. These experiments showed that, under certain reaction conditions, it is possible to achieve appropriate temperatures within the reforming process by directly coupling the heat released during catalytic combustion of the anode exhaust gas from a polymer electrolyte fuel cell, and to do so in steady-state conditions irrespective of any additional external heat source. The reaction conditions required are:

- A ς value of 1.3 for catalytic combustion;
- A molar water/carbon ratio of 1.9 for autothermal reforming at a molar oxygen/carbon ratio of 0.47;
- The heat exchanger in the reformer and catalytic burner must be connected so that the water/air mixture to be vaporized flows first through the heat exchanger in the catalytic burner and then through the reformer section.

Under these reaction conditions a temperature of 475 °C was reached for the steam/air mixture and a temperature of 401 °C was reached for the reformate. A value of 475 °C for the steam/air mixture is enough to vaporize completely the cold hydrocarbon stream entering through the nozzle. 400 °C for the reformate is ideal in terms of obtaining sufficiently rapid kinetics at the moment of entry into the WGS reactor connected behind the reformer when the WGS reaction is in the appropriate state of equilibrium.

The successful coupling of autothermal reforming and water-gas shift reaction has already been described in the previous IEF-3 Report. The experimental connection of all three main components involved in fuel processing for a system based on an HT-PEFC – the autothermal reformer, the catalytic burner and the water-gas shift reactor – will be tested in a specially constructed test module during 2007.

3.3.3 Staff members and fields of activity

Name	Telephone no. (+49-2461-61-) E-mail adress	Field of activity
Prof. Dr. R. Peters	4260 ra.peters@fz-juelich.de	Head of Fuel Processing (BGS)
R. Menzer	6708 r.menzer@fz-juelich.de	Procedure design and analysis
Dr. J. Pasel	5140 j.pasel@fz-juelich.de	Head of the Industrial Chemistry Group in BGS
Dr. Z. Porš	5322 z.pors@fz-juelich.de	CFD modelling, diesel reforming
R.C. Samsun	4616 r.c.samsun@fz-juelich.de	Dynamic modelling (MATLAB/SIMULINK), kerosene reforming

J. Latz	2779 j.latz@fz-juelich.de	Desulphurization, fuels
A. Tschauder	5400 a.tschauder@fz-juelich.de	Reactor development, reforming, secondary gas treatment

3.3.4 Important publications, PdD theses and patents

Important publications

Meißner, J.; Pasel, J.; Peters, R.; Stolten, D.

Preferential oxidation of carbon monoxide in a hydrogen rich gas mixture in catalytically coated turbulence structures

3rd European Polymer Electrolyte Fuel Cell Forum 2005, Luzern, Schweiz, proceedings on CD, File No. B121

Polymer electrolyte fuel cells (PEFC) suffer from reduced performance at working temperatures around 80 °C due to the presence of even small quantities of carbon monoxide. For fuel cells of this type, therefore, carbon monoxide (CO) is therefore preferentially oxidized via an ultrapurification process. This process must reduce the initial concentration of approx. 1 vol.% CO to values in the range of 10 - 50 ppmv. Dissipating heat from this strongly exothermic reaction represents a particular challenge, in order to ensure that selectivity for the desired CO oxidation remains high at temperatures of approx. 150 °C. The publication presents the results of measurements taken at catalyst-coated turbulence structures. These structures show very good heat and mass transport properties.

Pasel, J.; Samsun, R.C.; Schmitt, D.; Peters, R.; Stolten, D.

Test of water-gas-shift reactor on a 3 kW_e-scale - design points for high- and low-temperature shift reaction

J. Power Sources, 152, pp. 189-195, 2005

The concentration of carbon monoxide (CO) in the product gas from autothermal reforming can be significantly diminished using the water gas shift (WGS) reaction. A two-stage WGS reactor with a high-temperature section operating at an inlet temperature of 400 °C and a space velocity of 42,200 h^{-1} and a low-temperature section (300 °C, 26,000 h^{-1}) can reduce the CO concentration from approx. 10 vol.% to under 1 vol.%. Injecting water into the reactor between the two stages improves the CO conversion. It also enables the gas to be brought to the correct inlet temperature for the low-temperature section of the WGS reactor.

Pasel, J.; Duisberg, M.; Samsun, R. C.; Dahl, R.; Peters, R.

Deaktivierung von Katalysatoren für die autotherme Reformierung von Dieselkraftstoff

XXXVIII. Jahrestreffen Deutscher Katalytiker, Weimar, proceedings, 2006, 257 – 258

The influence of the chemical properties of four different liquid fuels converted via autothermal reforming (ATR) on the activity of a monolithic catalyst coated with precious metals was investigated. X-ray photoelectron spectroscopy (XPS) of the surface of the catalyst used during the ATR of 'artic diesel' showed that silicon partly

covered the catalytically active sites and thus led to a deactivation. The XPS analyses also showed that the catalyst surfaces had a heavy coating of carbon after each experiment, although this had only a minor effect on catalytic activity.

Porš, Z.; Peters, R.; Dahl, R.; Pasel, J.; Tschauder, A.; Stolten, D.

Optimized mixture formation for diesel fuel processing

Fuel Cell Seminar 2005, Palm Springs, CA, U.S.A.: proceedings, 2005, 113 – 116

Diesel reforming involves many process engineering difficulties. One major problem is the long-term stability of the catalyst. At the present time, carbon formation is recognized as one of the main obstacles to diesel reforming. Apart from the development of suitable catalysts, the quality of the mixture formed by the educts in the inlet to the reaction zone is also of importance. Detailed numerical simulations of flow profiles in the reformer mixing chamber, supported by flow experiments, provided the basis for a new design of the autothermal reformer. This new design had a long-term stability several orders of magnitude better than that of its predecessor. The reformer was run for 1000 h on the commercial diesel fuel *ARAL Ultimate*. The conversion of the fuel was more than 99.999 % at the start of the experiment and was still above 99.7 % after 1000 h operation. The hydrogen concentration remained constant within measuring accuracy.

Samsun, R,C.; Pasel, J.; Tschauder, A.; Klüttgen, U.; Schmitt, D.; Meißner, J.; Porš, Z.; Peters, R.

Reforming of kerosene for APU application in aircraft

7th European Solide Oxide Fuel Cell Forum, Lucerne, Schweiz, proceeding on CD, 2006

Fuel cell systems can significantly contribute to reducing emissions in the aviation sector. One possible field of use for fuel cells is as auxiliary power units in aircraft. The publication presents the results of experiments carried out with the ATR 5B reformer. Results on characterization of the reformer, on starting up the reformer from the cold and hot state, on its behaviour during load change and the results from a trial of 100 h with desulphurized Jet-A1 kerosene are presented.

PhD theses

Porš, Z.

Educt preparation and mixture formation in a reactor for the autothermal reforming of diesel-type fuels

Schriften des Forschungszentrums Jülich, Reihe Energietechnik, Band 49,
ISBN 3-89336-432-2, RWTH Aachen, 2005

This work is concerned with optimizing the fuel mixture generation in an autothermal reformer within a diesel-based fuel processing system for a polymer membrane fuel cell to be used as the APU (auxiliary power unit) of a vehicle. It is imperative that the diesel fuel is completely vaporized if a homogeneous mixture is to be formed. The formation of carbon in the mixing chamber and the reaction zone of the reformer is recognized worldwide as one of the main obstacles to diesel reforming. This is primarily due to aromatic compounds and high-boiling components in the diesel fuel.
The starting point for this work is the fifth generation of the ATR-5A reformer for diesel fuel developed at Research Centre Jülich. The educts air, steam and fuel are mixed together in the mixing chamber of this reactor. The liquid fuel is sprayed into the

mixing chamber via an atomization nozzle, finely atomized and vaporized using superheated steam.
CFD (computational fluid dynamics) simulations were used to analyse and optimize the fuel mixture. In these CFD simulations, the turbulent flow and the mixing of the three gases – air, steam and evaporated fuel – were modelled, as were the spray pattern of the injected fuel and its vaporization properties. Experiments in model reactors made of quartz glass using optically labelled fluids to allow the complex turbulent flow to be visualized showed a good correlation with the CFD model. Additionally, the shape of the spray patterns of the injection nozzles was examined in glass reactors.
Using CFD simulations meant that the optimum operating conditions for the ATR 5A reactor could be identified and adjusted. The second step was to construct a performance-optimized variation of the reactor, known as ATR 5B. Two new designs for the mixing chamber were then devised that enabled the reactor to run on conventional diesel fuel. The seventh- and eighth-generation reactors based on these new chambers, ATR 7 and ATR 8, were constructed and tested.
This thesis establishes CFD modelling as an important tool for the optimization of fuel mixtures in reactors.

Important patents

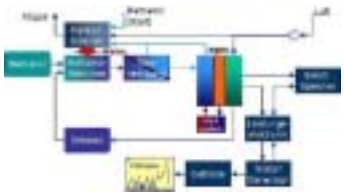
Patent applications:

Principal inventor	PT	Title
Prof. R. Peters	1.2168 PCT	Mixing chamber for a reformer and process for operating this chamber
Dr. Z. Porš	1.2169 PCT	Mixing chamber for a reformer and process for operating this chamber
Prof. R. Peters	1.2188	Process for fuel treatment
Prof. R. Peters	1.2188 EP	Process for fuel treatment
Prof. R. Peters	1.2221	Autothermal reformer
Prof. R. Peters	1.2221 PCT	Autothermal reformer
Dr. Z. Porš	1.2272	Process for vaporizing a liquid fuel and a mixing chamber for implementing this process
Dr. Z. Porš	1.2281	Apparatus for producing a fuel oxidant mixture

3.4 Cross-cutting topic: process and systems analysis

3.4.1 Objectives and fields of activity

An indispensable element in the development of energy conversion systems involving fuel cells for mobile and stationary applications is the development of process concepts and the analysis of systems. This makes it possible to analyse and evaluate drives for the vehicles of tomorrow and also for stationary power generation. Systems employing conventional technology as well as the fuel supply are also included in the analysis.



The results of the development using the PRO II and SIMSCI computer programs of process concepts for fuel cell systems to be used in both the mobile and stationary conversion systemsplants are reflected in process flow diagrams and net electricity balances. This is followed by a comparative process evaluation of competing energy conversion systems. This involves subjecting systems selected on the basis of the relevance of their operating conditions (e.g. strict dynamic requirements in cars) to a process evaluation via a detailed simulation of energy requirements and emissions (using SIMBA) (e.g.: APU for on-board power generation). The results provide information on energy balances and emissions.

The above technical analyses are expanded by application-related peripherial considerations such as process chain analyses (using KRAKE) and cost analyses (using KOSTEX). For the same or similar balancing range the entire process chain is evaluated. The results then provide indications on specific energy requirements (MJ/km, MJ/kWh), specific emissions (g/km, g/kWh) and specific costs (€ cent/km, € cent/kWh) for mobile/stationary applications.

The manufacturing processes of future energy carriers or fuels are essential steps in process chain analysis. The focus here is currently on synthetic fuels, such as synfuels (based on fossil sources of primary energy) or sunfuels (based on biomass), which are both manufactured from synthesis gas. The engineering is additionally illustrated and evaluated with the aid of analyses taken from the literature. Furthermore, steps of the process chains and entire competing processes are compared in terms of emissions, primary energy requirements, material consumption and costs (benchmarking). The results of these analyses can be used to derise recommendations for process development and the R&D investment required.

3.4.2 Important work results

3.4.2.1 Process engineering analysis

The focus of the work on process analysis was on establishing process concepts for fuel cell APUs for SOFC high-temperature fuel cells and HT-PEFC and PEFC low-temperature fuel cells. The evaluation covers fields of use in cars, trucks and aircraft. Fuel cell APUs provide different advantages for each application mentioned in Fig. 43. The efficiency achievable is very important here, as well as other properties such as low emissions and low-noise operation. One particular concern in aviation is reducing emissions at airports and being able to use the water generated by the fuel cells for the water balance in the cabin area. Reduced amounts of water mean lower fuel consumption on start-up.

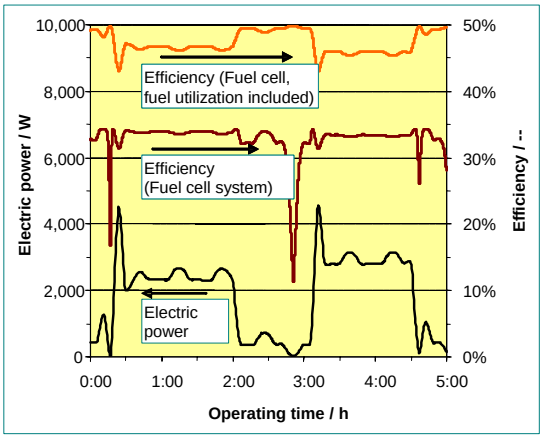


Fig. 42: Efficiency of an FC-APU in dynamic operation compared to the electrical power required for an average truck in the USA

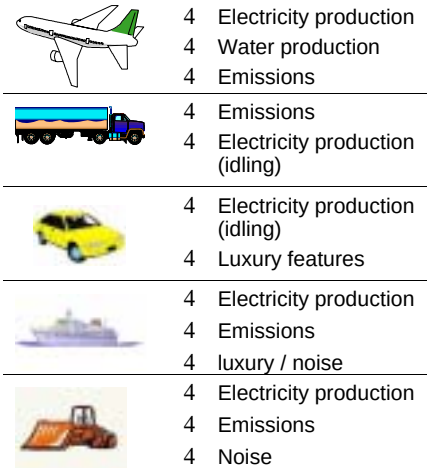


Fig. 43: Benefits of APUs in various vehicular applications

In vehicles such as cars and trucks, fuel cell APUs offer advantages over a combustion engine combined with a generator, particularly when running idle. The additional fuel required by modern diesel car engines in driving mode in order to generate power in the engine's fuel consumption map was analyzed and no advantages in terms of efficiency were found. In typical load profiles for American trucks, SOFC APUs and diesel reforming can be used to generate efficiency levels of 33 % and very low emissions, which is of major benefit to the environment. Efficiency is normally 11 % during power generation by the drive unit and a maximum of 20 – 30 % when small APU combustion engines are used (see Fig. 42).

3.4.2.2 Systems analysis

Of the work done on process and systems analysis, the contribution to the study entitled "Strategic evaluation of the prospects for synthetic fuels based on biomass in North Rhine-Westphalia" is worthy of particular mention. The analysis of the potential for manufacturing synthetic fuels from biomass (BTL – biomass to liquid) shows that for every ton of wood

used, the maximum yield that can be expected is approx. 200 kg of Fischer-Tropsch product or about 150 kg of middle distillate (see Fig. 44). The mass of the middle distillate is determined as the sum of the values for diesel and kerosene. The results for efficiency are calculated including the electric power assessed from primary energy.

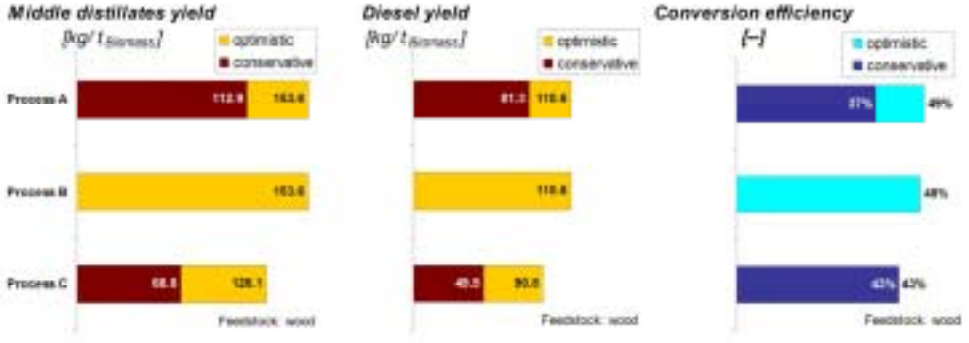


Fig. 44: Middle distillate and diesel yield in kg per ton of biomass and efficiencies with wood as input material

An estimation of fuel costs based on these results and for the respective boundary conditions gives a lowest filling station price of around 0.85 EUR/l including VAT (16 %) for BTL diesel. Energy-equivalent revenues for all products of the BTL process and the absence of further taxes are assumed.

Various ways of using biomass for energy mean that levels of reductions in greenhouse gas emissions at various additional costs compared with the conventional energy path. The study showed that coupled power and heat generation is advantageous solely when a large amount of heat is used. Of the biofuel alternatives, BTL and SNG (substitute natural gas: methane from biomass gasification and methanization) are of greater benefit than ethanol from wheat and biodiesel (RME). Heat generation from biomass produces lower additional costs but similar reductions in greenhouse gases compared to BTL and SNG. Using biomass for fuel production finally also contributes – although the potential volume is limited– to reducing dependency on imports of energy resources. This is of particular relevance to the transport sector, given its almost total dependency on products manufactured from crude oil.

3.4.3 Staff members and fields of activity

Name	Telephone no. (+49-2461-61-) E-mail adress	Field of acticity
Prof. D. Stolten	3076 d.stolten@fz-juelich.de	Head of the Process and Systems Analysis (VSA) department and Head of Institute
Th. Grube	5398 th.grube@fz-juelich.de	Drive simulation, process and systems analysis

3.4.4 Important publications

Biedermann, P.; Grube, Th.; Höhlein, B. (Hrsg.)

Methanol as an energy carrier

Schriften des Forschungszentrums Jülich, Reihe Energietechnik, 2006, Volume 55

Strategic challenges in terms of fuel supply and a continuing need to reduce the environmental effects of mobility mean that the future will see a need for considerably more efficient and 'cleaner' drive systems combined with new fuels available in sufficient quantities and at an economically viable cost. Methanol has been produced industrially for a long time now and can be generated from a wide range of fossil and renewable raw materials. Methanol is also easy to store and can be used as an environmentally friendly fuel additive or as pure fuel in highly efficient fuel cells. Methanol as an Energy Carrier" evaluates procedures for manufacturing and using methanol and discusses future markets as well as environmental and safety aspects.

Grube, Th.; Menzer, R.; Peters, R.; Arnold, K.; Ramesohl, S.

Process analysis of the manufacture of synthetic liquid fuels from biomass

VDI Berichte 1975, Düsseldorf, 2006, 379–403

Fuels capable of mitigating local and global environmental effects and reducing dependence on fuel imports are set to gain further in importance in years to come. Fuels from biomass in particular can contribute to the above processes, if they utilize the input material more efficiently and keep additional costs to a minimum, and therefore require only small adjustments to the infrastructure. Synthetic fuels from biomass are a very promising and innovative option in this regard. The present article concentrates on processes for manufacturing such fuels but also incorporates aspects of energy, climate and industric policy. The results presented were compiled on behalf of the State of North Rhine-Westphalia as part of the study entitled "Strategic evaluation of the prospects for synthetic fuels based on biomass in North Rhine-Westphalia".

Grube, Th.; Menzer, R.; Samsun, R.C.; Pasel, J., Peters, R.

Options and challenges in using auxiliary power units in mobile applications

VDI Reports 1975, Düsseldorf, 2006, 499 - 528

Electric power for various types of drives can be provided for different mobile applications at low emission levels and high degrees of efficiency via auxiliary power units (APUs) based on fuel cells. Demand for this is increasing in line with the growing need to provide electricity for luxury and safety features, as well as steering and control functions. Moreover, harbours and airports are increasingly being required to improve their local air quality, where the use of APUs as on-board power supplies for ships or aircraft can be of considerable assistance. APUs of this type furthermore promise to release a low level of emissions and are more efficient when installed in US trucks for on-board power supply. In this article the benefits for fuel-cell-based car and truck APUs set out above are examined and verified via a comparison with conventional technology.

3.5 Cross cutting topic: analysis

3.5.1 Objectives and fields of activity

Although great technical progress has been made in fuel cells in recent years, the technology is still not sufficiently advanced to bring them to market. The already successful research and development work needs to be continued, but a deeper, more fundamental understanding of physico-chemical processes is also needed. Recent years have seen an increase in the importance of analytical methods that allow structures and effects to be analysed either in situ or in a very precise spatial resolution Thus, in IEF-3, particular efforts are being invested in developing, formulating and using special measuring methods for the structural analysis of components of the membrane electrode assembly and for characterizing stacks and single cells. The top priority here is elucidating signs of ageing in the membrane electrode assembly.

3.5.2 Important work results

As already mentioned in Chapter 3.1.2.3, ruthenium corrosion is one of the chief causes of ageing in DMFC MEAs. The problem is not just loss of ruthenium from the anode but also the potential migration and deposition of ruthenium on other MEA and stack components, i.e. the membrane, the cathode, the flow fields and the bipolar plates. Analysing the ruthenium content of individual components therefore forms an important part of investigations into ageing. Dissolving the Ru in a controlled way had previously proved problematic. In cooperation with the Central Division of Analytical Chemistry (ZCH), a dissolution process was developed that enabled the ruthenium content in the electrodes to be determined quantitatively. This process was used to determine the ruthenium and platinum contents of aged and unaged MEA electrodes.The analysis of an MEA that had been aged during more than 50 h operation in a scooter under dynamic conditions revealed a significant drop in the percentage of Ru in the metal in the anode, from 33 % to 27 % by weight, and an increase in the corresponding Ru content in the cathode, from 0 % to 5 % by weight.

The presence of ruthenium was also detected on the bipolar graphite plates of the aged scooter stack using SEM/EDX equipment from IEF-2. Fig. 45 shows SEM images taken around the meander-shaped channel floor of the anode side of a bipolar plate. These images clearly show white deposits, which the EDX analysis revealed to be ruthenium. The EDX analysis revealed no ruthenium deposition outside the channel floor (Area 1) but significant ruthenium deposits on the channel floor, particularly on the channel edge (Area 2).

Significant structural changes can occur due to MEA ageing. Examining the morphology and the micro- and nanostructure of MEA layers is thus essential in order to detect and prevent ageing effects. This examination covers not only the surface of the layers but above all the cross-section of the composite layer. Appropriate specimen preparation is of crucial importance if microscopic and surface analysis methods are to be used. During the period under review, work was begun on developing a specimen preparation method with respect to embedding MEAs with epoxy resin, and then taking cross-sections using micrographs and microtome specimens.

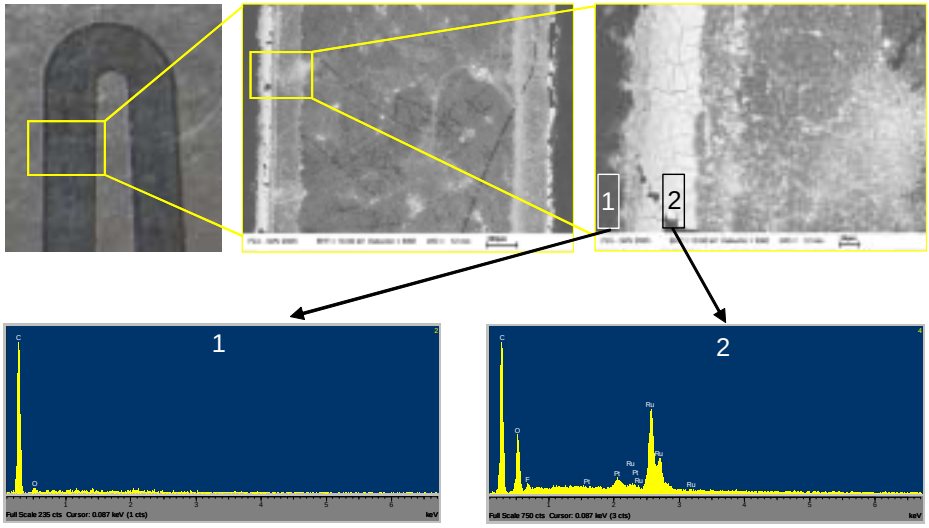


Fig. 45: SEM/EDX analysis of the anode side of a bipolar graphite plate following ageing due to operation in a scooter

Fig. 46 shows SEM images of a microtome section 1000 nm in thickness taken from an MEA aged for 2500 h after embedding in epoxy resin. The pictures shown in Fig. 46 were taken in backscatter mode (BSE).

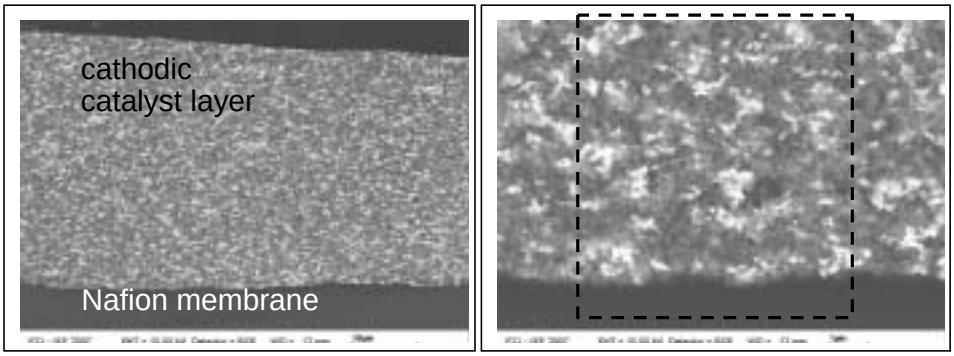


Fig. 46: SEM images of an MEA microtome specimen, left: magnified approx. 1000 times; right: magnified approx. 6000 times. Dotted lines: section for EDX analysis (Fig. 47)

The figure shows the cross-section of the approx. 35 σm-thick cathodic catalyst layer (top) and the adjoining Nafion membrane (bottom) at two different magnifications. The light areas correlate with elements of high atomic numbers, in this case platinum. It can be observed

that the distribution of platinum across the catalyst layer is relatively inhomogenous. The EDX element analysis in Fig. 47 confirms this. In the areas in which the platinum content is higher (light areas in left-hand picture), the amount of carbon is generally lower (dark areas in middle picture). Nafion distribution, which is represented by fluorine analysis (right-hand image), appears to be independent of Pt and C distribution.

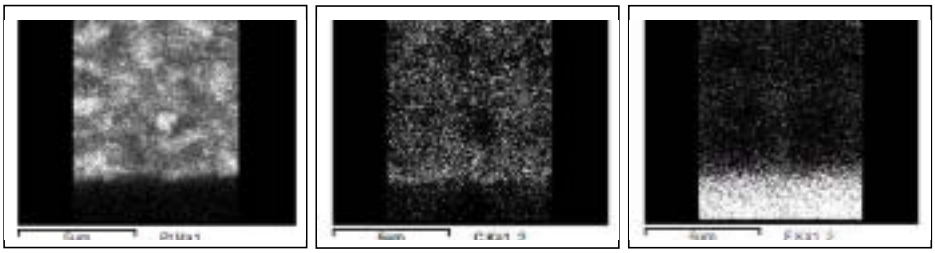


Fig. 47: EDX element distribution in the marked area in Fig. 46. Left: platinum, middle: carbon; right: fluorine

During the period under review a magnetic tomograph developed in collaboration with the Tomoscience company and the Central Technology Division (ZAT) was put into operation and tested (see Fig. 48).

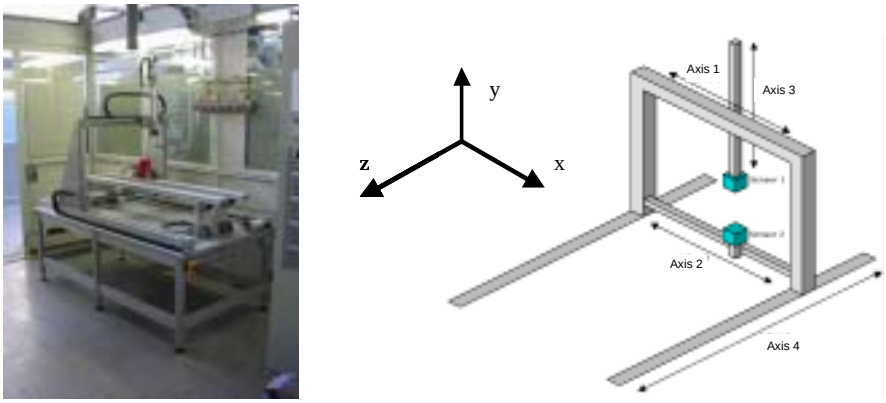


Fig. 48: Magnetotomograph based on a positioning table and two magnetic sensors (Honeywell HMC 1063)

Magnetic tomography is based on measuring the external magnetic field of fuel cells and the subsequent calculation of internal current density distribution. The functionality of the system and the mathematical models used wereconfirmed using various measurements:

- measurements at a defined 2D conductor loop
- measurements at hydrogen-driven single cells during variations in the air ratio.
- measurements at direct methanol fuel cells during variations in the operating conditions

The measurements at the individual cells were accompanied by reference measurements taken by segmenting the cells to be measured. An active area of 240 cm² was divided into 196 segments of equal size, which were then evaluated via a separate data logger.

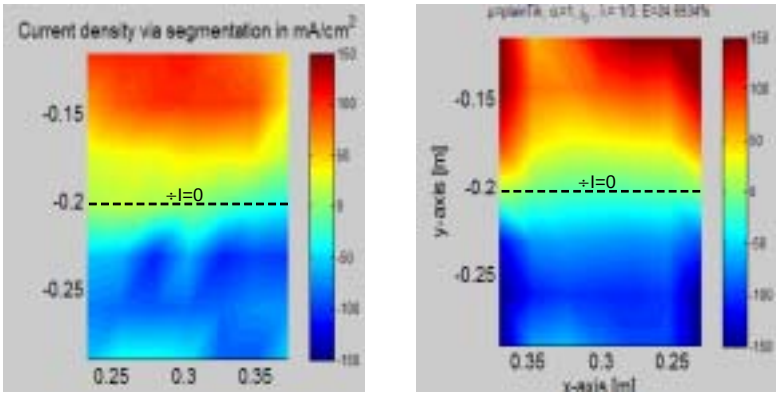


Fig. 49: Current density distribution along the cell surface of a DMFC during a change in the air ratio from 2.5 (left) to 1.25 (right):current 48.8 A; methanol concentration 0.5 M

A selected example is shown in Fig. 49. This shows a comparison between the different measurement procedures for changes in current density distribution in DMFC during variations in the air ratio. The first measuring procedure uses a segmented cell, and the second applies magnetic tomographyy. The trend in terms of changes in current density distribution based on an average current density of 200 mA/cm² can clearly be seen: when the air ratio changes from 2.5 to 1.25, the MEA areas at the cell outlet make only an insignificant contribution to the total current. Magnetic tomography and the segmented cell method both enabled the line for ÷I=0 to be determined. An increase in currrent density is shown above the ÷I=0 line and a decrease in current density above it.

3.5.3 Staff members and fields of activity

Name	Telephone no. (+49-2461-61-) E-mail adress	Field of activity
D. Kalkreuth	2378 d.kalkreuth@fz-juelich.de	General quality management, quality assurance for 'Standardized electrochemical testing procedures', development of sample-preparation procedures for imaging testing procedures
Dr. M. Stähler	2775 m.staehler@fz-juelich.de	Head of the 'MEA Characterization' Group, standardized characterization of DMFC membrane electrode assemblies and development of new characterization procedures

Dr. Chr. Wannek	4013 c.wannek@fz-juelich.de	Head of the 'MEA Development' Group, development of membrane electrode assemblies for DMFC and HT-PEFC
M. Wannert	5590 m.wannert@fz-juelich.de	Development of mathematical procedures for magnetic tomography, reconstruction of current density distributions using 2-D and 3-D models and single cells
Dr. K. Wippermann	2572 k.wippermann@fz-juelich.de	Clarification of physical/chemical ageing mechanisms in direct methanol fuel cells

3.5.4 Important publications

Ghosh, P. C.; Wüster, T.; Dohle, H.; Kimiaie, N.; Mergel, J.; Stolten, D.

Analysis of single PEFC fuel cell performances based on current density distribution measurement

Journal of Fuel Cell Science and Technology, 3 (2006), 351 - 357

This paper presents and evaluates measurements made with a segmented cell composed of expanded graphite that was developed at the Institute. In principle the test set-up can be used for both single cells and stacks, since the measuring cell is thin and its components mean that it has a conductive sealant and can therefore be integrated into existing set-ups without any problems.
The operating conditions were varied in such a way that spatially resolved information could be obtained on the influence of the air ratio, the humidification of the reactants and the temperature.
The measurements showed that the current density around the gas inletdepends largely on the extent of humidification of the reactants. On the other hand, around the gas outlet, it was noted that a strong influence was exerted by the absolute pressure and the air ratio, but that humidification had a lesser effect. Finally it was shown that when cell temperature rises, cell performance can fall as a whole if internal humidification from product water is insufficient if the reactants do not experient sufficient external humidification. The segmented cell showed that, for single cells in particular, achieving a homogeneous power output is dependent on an even contact pressure being applied to the active area. By assigning the segments to the respective position of the meander meant that the effects from inlet to outlet could be displayed clearly using a graphics application.

Ghosh, P. C.; Wüster, T.; Dohle, H.; Kimiaie, N.; Mergel, J.; Stolten, D.

In situ approach for current distribution measurement in fuel cells

Journal of Power Sources, 154 (2006), 184 - 191

This paper presents a new, simple method to determine current density distribution in fuel cells. Using a single cell based on a meander flow structure, the reaction rate was measured along the length of the meander channel. The hardware developed is simple to build, cost-effective and easy to integrate into the cell. A thin semi-segmented plate composed of expanded graphite serves as a passive resistance network. The construction is based on the special property of expanded graphite whereby resistance at right angles to the surface, i.e. in the direction of flow, is substantially higher than in the planar direction. When technically relevant current densities were applied, voltage differences of several mV were observed between the

upper and lower side. These differences were proportional to the current applied. The better conductivity in the planar direction also ensures that the potential in each segment remains constant. The first measurements taken on a 244 cm² PEFC single cell were performed in this study and show the changes in local current densities when operating conditions are varied.

Hauer, K.-H.*; Potthast, R.*; Wüster, T.; Stolten, D.

Magnetotomography – a new method for analysing fuel cell performance and quality

Journal of Power Sources, 143 (2005), 67 - 74

Magnetic tomography is a new method for the measurement and analysis of the current density distribution of fuel cells. The method is based on the measurement of the magnetic flux surrounding the fuel cell stack caused by the current inside the stack. As it is non-invasive, magnetotomography overcomes the shortcomings of traditional methods for the determination of current density in fuel cells. After several years of research a complete prototype system is now available for research on single cells and stacks. This paper describes the basic system (fundamentals, hardware and software) as well as the state of development until December 2003. Initial findings on a full-size single cell will be presented together with an outlook on the planned next steps.

3.6 Cross-cutting topic: quality management

3.6.1 Objectives and fields of activity

To guarantee a high standard of quality in development, manufacturing and characterization, quality assurance measures are treated as an integral part of the work process and are implemented with increasing frequency.

Within a fuel cell stack, the single fuel cells are connected in series. This serial arrangment means that the cell with the worst polarization characteristics can dominate the behaviour of the whole stack. When developing fuel cell components, therefore, it is crucial to ensure that the quality of the MEAs remains constant, i.e. that all components used in the stack have the same physical and chemical properties.

In practice this means the following:

- The materials purchased for use in constructing the stack MEAs must be subjected to an ongoing quality control as part of incoming goods inspection.
- The MEAs manufactured in IEF-3 must be tested regularly for the reproducibility of their polarization characteristics.

3.6.2 Important results

3.6.2.1 Incoming goods inspection:

All incoming materials were tested for quality as part of quality control. This examination determined the suitability of the materials for use in the stack. The results of the ageing tests carried out on MEAs (see the Chapter on LTFC MEA development) showed that chloride ions affect the performance of an MEA. This meant that a quality criterion could be established for a material intended for use in the construction of MEAs. This quality criterion is:

"If ion concentrations in a sample are below a certain threshold after the sample has been treated in a model liquid that simulates the chemical load in a fuel cell, that material can be used in the construction of MEAs."

The results of the ageing tests can then be used to define a threshold value in combination with the boundary conditions for the system, e.g. the volume of liquid to be circulated in the anode loop of the stack, which is relevant for the ion concentration formed. Before these thresholds can be used in the selection of materials, the various materials must first be measured and recorded.

Fig. 50 shows the measurement results of six graphite samples from different batches. The 'Graphite 4' sample was taken from an unsatisfactory delivery of inferior quality that was identified as such during the incoming goods inspection.

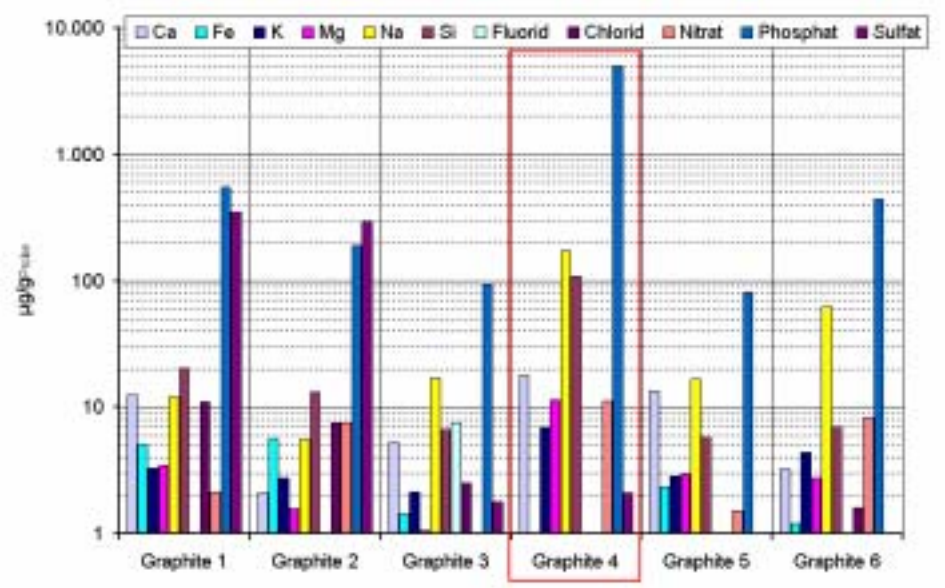


Fig. 50: Identification of an unsatisfactory graphite delivery

For the future extention of incoming goods inspections, further factors of influence on MEAs, such as the influence of chloride ions on cell performance, must be identified and appropriate quality criteria must be defined.

3.6.2.2 Reproducibility of polarization characteristics in MEAs:

The materials used in the construction of MEAs – polymers, graphite, catalysts etc. – exhibit very complex behaviour in certain respects. Physical effects such as hysteresis and bifurcation are not uncommon. These effects frequently make it difficult to reproduce polarization characteristics, because they make cell voltage dependent on previous stages of the measuring process.

New run-in protocols were thus developed to enable MEAs to be operated in completely automated test stands. It was shown that the polarization characteristic curves for the MEAs run in using these protocols were reproducible to within a few milivolts.

It emerged that quality assurance measures for use in the development of fuel cells are in need of further refinement. It also emerged, however, that ever greater volumes of data need to be collected, examined, administered and saved about the various properties of MEAs and the materials of which they consist. Concepts for qualifying and tracing all relevant data using auto ID techniques are currently being tested. The objective is to record all steps in the process of fuel cell manufacture by using a barcode and chip storage systems, in combination with a database.

3.6.3 Staff members and fields of activity

Name	Telephone no. (+49-2461-61-) E-mail adress	Field of activity
D. Kalkreuth	2378 d.kalkreuth@fz-juelich.de	General quality management, quality assurance for 'Standardized electrochemical testing procedures', development of sample preparation procedures for imaging testing processes

4

Projects

Selected R&D Projects

- Market study for DMFC applications in the kW class
- ZeuS – the SOFC for on-board power supply in cars
- Mixture formation in autothermal diesel reformers
- Process analysis of future CO_2-free membrane power plants

4.1 Market study for DMFC applications in the kW class

4.1.1 Motivation

The Institute of Energy Research - Fuel Cells (IEF-3) - carries out application-oriented research and development with an emphasis on fuel cells. In the field of direct methanol fuel cells (DMFC), the Institute has already created the first prototypes in the electric power class up to 2 kW (see Fig. 51).

Fig. 51: 2 kW DMFC module from 2003 (left) and DMFC fuel cell system in an electric vehicle (scooter) from 2005 (right)

In order to be able to develop marketable products, however, collaboration with industry partners is required. Between 2003 and 2005 IEF-3 was not sufficiently familiar with the market for systems in the 0.5 kW to 5 kW range. Management engineers (ME) were thus commissioned to carry out a market study in the field of low-temperature fuel cells with particular emphasis on DMFCs and thus to check that the work on DMFCs carried out by IEF-3 was focusing in the right direction. The market study was designed to include analyses of the applications and the market participants as well as an evaluation of market trends. The results obtained were then used as the basis for a subsequent feasibility study, also carried out by management engineers.

4.1.2 Results of the market study on DMFC systems

The market analysis comprised 4 steps:

- analysing and identifying possible applications/market segments and competing technologies
- analysing market participants and carrying out expert interviews, followed by a profiling of market participants
- evaluation of market trends: establishing the cause-effect relationships for the market segments and evaluating the trends. Identifying of opportunities and risks and, on the basis of these, determining the application-specific challenges for IEF-3
- determining the strategic consequences: evaluating strategic partners and devising recommendations for action

In the first analysis, nine out of an original 26 possible applications from the fields of portable systems, APU (auxiliary power units), stationary systems and small drives for light traction were judged sufficiently suitable to warrant further investigation (see Fig. 52). The criteria used were market and product attractiveness and economic and technical potential for success. A total of 14 interviews were carried out for the nine different applications with questions covering the subjects of companies; the market; trends; application; and development competences. If the product is to be developed to a stage of quantity production the potential cooperation partner should be able to contribute additional construction, production/joining, testing and system technical capabilities. The competence profile was also investigated. Nine of the 14 potential partner companies interviewed met the technical criteria involved; eight of these would make suitable cooperation partners. If the criterion of cost-effectiveness is included, this leaves only five possible partners or applications: forklift trucks, industrial vacuum cleaners, lawnmowers, yachts and special applications such as observation devices.

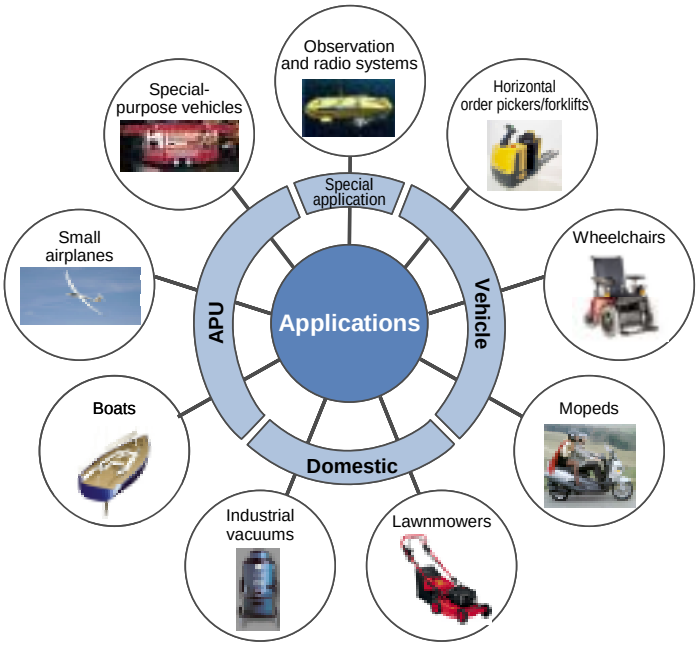


Fig. 52: Applications investigated for DMFC fuel cells

In terms of evaluating trends for fuel cells on the market, spheres of activity were determined from five given search fields (cost-effectiveness, legal regulations, constructions, performance parameters and social factors), and trends then determined. Fuel cells are not yet cost-effective, by virtue of the significantly higher investment costs as compared with competing technologies. On the other hand, legal regulations (exhaust gas standards) and social factors such as the modern increase in environmental awareness operate in favour of the development and use of fuel cells. With regard to performance parameters, the DMFC is compelling in performance and consumption, but there is still room for improvement in reliability, lifetime and behaviour during use. Moreover, the fuel cell still needs to be

optimized in terms of physical characteristics such as the space it takes up, its robustness and its weight. The most significant trend from the DMFC fields of application is that of falling manufacturing costs. This means that fuel cells must become more cost-effective and reliable. The current stricter exhaust gas standards and increased environmental awareness may help fuel cells to break into the market.

Out of the five potential applications and partners, forklift trucks were identified as an appropriate application for the use of advanced DMFC systems, after the size of the market accessible was included as a further selection criterion. Finally, in a comparison between DMFC and PEFC technology, tests were carried out to see how well DMFC systems would satisfy the demands for use in a forklift truck. In terms of its response to these demands, the PEFC is not superior to the DMFC. The advantages of the PEFC over the DMFC are manufacturing costs, performance dynamics and start-up time. The advantages of the DMFC over the PEFC are its running costs and its handling/fuel supply.

The following recommendations for action for DMFCs therefore resulted: manufacturing costs must be reduced and the lifetime must be extended. The required performance dynamic and start-up time can be achieved by hybridizing the fuel cell with a battery and by improving the fuel cell's control system. Finally, in a cost reduction workshop on system costs, it was shown that the cost objective for the DMFC system can be achieved via scaling effects (10,000 units p.a., automated processes, suitable construction) and a new design (double power density, recycling catalysts, optimizing layout etc.). The next step was then to redesign the DMFC system in a feasibility study and to initiate cooperation by identifying potential development partners.

4.1.3 Results of the feasibility study for DMFC systems

Redesigning and initiating cooperation were selected as the two top priorities for the feasibility study.

Redesigning involves:

- designing the DMFC system in accordance with the requirements of a forklift truck
- generating ideas to optimize DMFC technology from the perspective of costs and functions
- evaluating these ideas in terms of how practical they are to implement, their potential and the time it would take to realize them

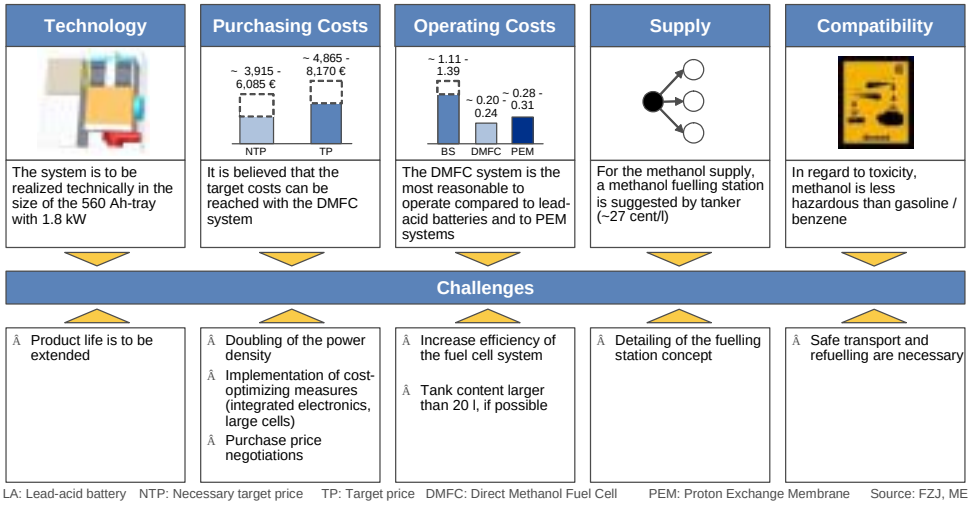
Then, building on the results of the redesign, steps were made to initiate cooperation involving the following work packages:

- developing a progressive cooperation with JUNGHEINRICH (from provision of samples to joint development)
- bridging competitive gaps via competent partners with experience of production and joining procedures for stack components
- analysing the procurement market and identifying potential suppliers and development partners

The first step of the redesign process was to establish the technical prerequisites for the ECE 220 forklift truck with JUNGHEINRICH. A construction space of 206 litres is available for the entire DMFC fuel cell system, corresponding to the construction space taken up by the

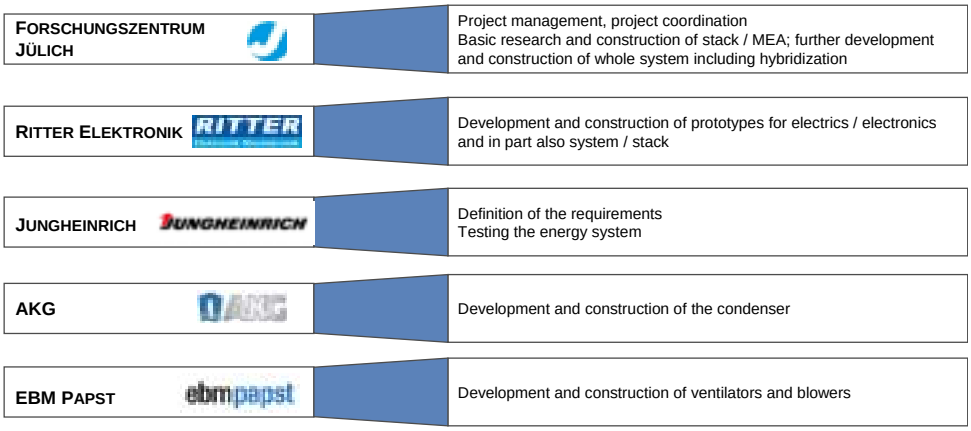
batteries. The ambient temperature of the fuel cells can be between 0 °C and +40 °C. The system voltage is 24 V DC and the peak current reaches ~300 A, whereas nominal current is ~100 A. The average energy requirement is 0.82 kWh, and the DMFC system must be able to power continuous operation for 20 minutes.

The key findings of the feasibility study are summarized in Tab. 3 along with the relevant requirements for operating forklift trucks using DMFCs. It was thus possible to show that fuel cell hybrid systems based on direct methanol fuel cells (DMFC) have the potential to achieve a comparable cost level to conventional battery systems when used in forklift trucks and can thus be used to bring fuel cell applications on the market Research and development should be carried out on the service life and power density of the stack and the overall development/hybridization of the DMFC energy system to ensure an economic application.



Tab. 3: Core findings and requirements in terms of forklift truck operation by DMFCs

In terms of initiating cooperation, the major cooperation partners in the value adding chain were identified and a rough timeline including the appropriate milestones defined up to the 2011/2012 series run. The key tasks of the partners Jungheinrich AG, Ritter Elektronik GmbH, Ebm-Papst Landshut GmbH and AKG Verwaltungsgesellschaft mbH, which formed a consortium with Research Centre Jülich, are represented in Tab. 4. The objective of the cooperation is to develop a marketable DMFC energy system for a forklift truck. In this regard the DMFC energy system should be at least as competitive in terms of costs and as functional as batteries, and it must be possible to handle the methanol required safely. Every participant in the cooperation contributes its skills as a partner in this regard. The requirements, in addition to reducing costs, in particular are service life over a period of at least 5000 operating hours and an increase in the specific power density of the fuel cell system to approximately 30 W/l at a total efficiency of Ó30 %.



Tab. 4: Core tasks of the cooperation partners

As part of a project commissioned by the Federal Ministry of Economics and Technology (BMWi), joint DMFC systems are now to be realized and should be used to power electrically driven forklift trucks as a pilot application (see Chapter 5.3).

4.2 ZeuS - the SOFC for on-board power supply in cars

4.2.1 Introduction

Solid oxide fuel cells (SOFC) are primarily developmed for use in decentralized energy supply. In addition, recently, attention has also been focused on use as an auxiliary power unit (APU) in vehicles, ships and aircraft. This mobile use makes high demands on the design of the SOFC in terms of the ratios between power and weight or volume. In the ZeuS project (German ZEllen Und Stacks (cells and stacks)), the first basic investigations into cells and stacks in terms of these requirements were carried out between 2001 and 2003. The project was financed by an industrial consortium led by BMW and supported by the Federal Ministry of Economics (BMWi). The German Aerospace Centre (DLR) and Forschungszentrum Jülich (FZJ) were involved as research partners. The project was a success and as a result a second phase, ZeuS-II, was started covering the period 2004-2006. The objective of this phase was to develop further the lightweight stack concept for use in an APU system. IEF-3 (formerly IWV-3) had the following tasks during this phase:

- modelling the stacks and their subunits (cassettes) so as to optimize gas and temperature distribution and the electrical contact area between the cassettes
- carrying out operating tests on individual cells and stacks with up to 20 cassettes

4.2.2 Modelling

The so-called 'CS design' for lightweight APU SOFC stacks is based on thin metal sheets, which can be processed using conventional forming techniques. Two sheets are combined to form a cassette, in which one planar anode substrate cell is integrated. These cassettes containing the cells form the building block from which the stacks are assembled (see Fig. 53).

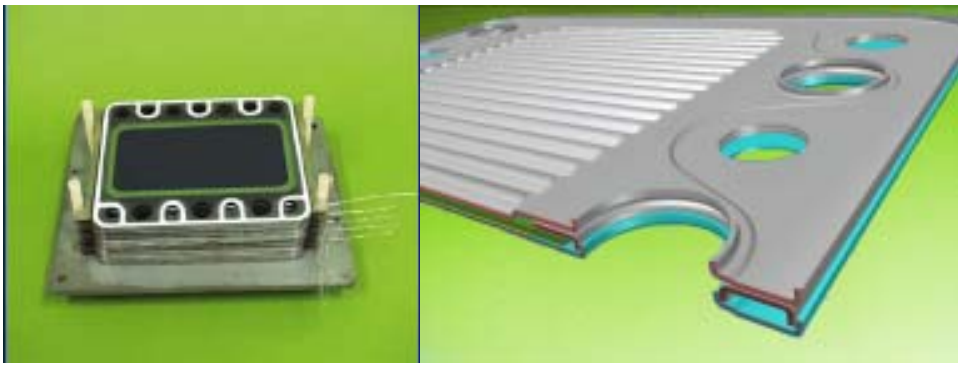


Fig. 53: Lightweight SOFC stack in the CS design for APU systems in mobile applications

The cassettes are joined together using a glass-ceramic seal. This seal ensures that the gas compartments remain separated and guarantees that the different layers are electrically insulated from each other. A wave profile in the lower section of the cassette distributes the operating gases homogeneously over the cell area. The waves simultaneously fulfil the

function of the interconnect, i.e. they connect the cathode of one cell electrically to the anode of the next cell.

One of the first aspects addressed via modelling and simulation calculations was the influence of the geometry of the wave profile on gas distribution and particularly on pressure loss within the cassette. The simulation calculations were carried out using the CFD software package Fluent. The wave profile forms for the area of through-flow a complex geometry (see Fig. 54). After the geometry was integrated into the model, parameter variations were carried out. The results of the simulation calculations were used to optimize the shape of the wave profile.

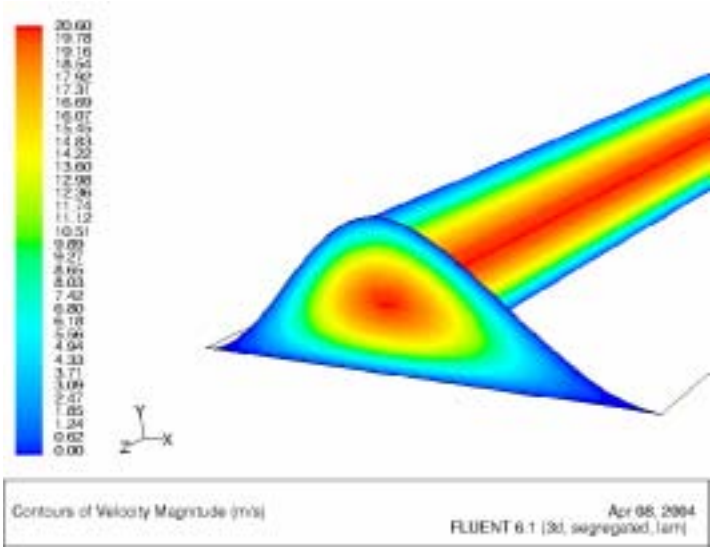


Fig. 54: Flow distribution of a wave in a channel

The simulation calculations of flow distribution in the cassette were verified by measurments of the pressure distribution. Pressure sampling points were set up at over 50 locations distributed over the wave profile and the inlet and outlet manifolds. The measurements showed a significant effect on homogeneous distribution in the cassette, caused by flow loss at the side of the cell. Closing these by-passes resulted in an improved homogeneity in gas flow over the cell areas.

A second aspect influenced by the geometry of the wave profile is the electrical contact resistance between neighbouring cells. For electrical current flow and potential distribution across a wave a model was constructed and simulation calculations performed (see Fig. 55). The narrow contact areas between the wave and the cathode layer of the cell, combined with the relatively long period of the waves, produce a significantly higher contact resistance compared with the standard F design of stacks for stationary applications. These ohmic losses can be reduced by the period of the waves; this causes an increase in pressure loss. Manufacturing restrictions for the sheet metal parts prevented the wave profile in the CS design cassettes from being optimized.

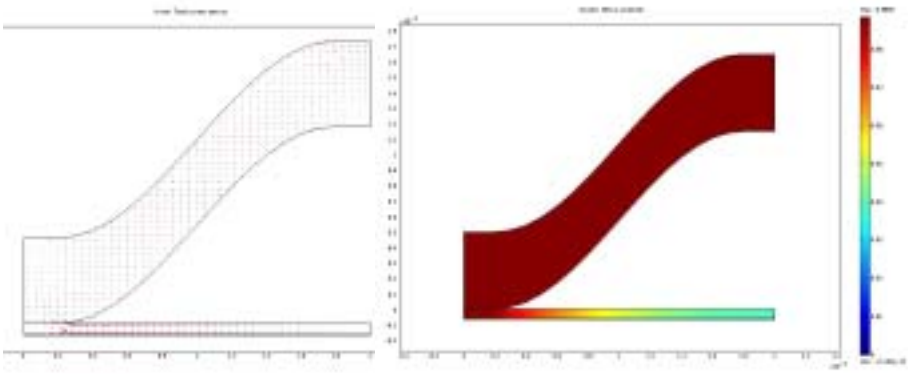


Fig. 55: Distribution of potential (left) and current (right) in parts of the cathode and the interconnect

A third important aspect investigated via modelling and simulation calculations was temperature distribution in the stack during operation (see Fig. 56). The results of the simulation show that air volume flow has a major effect on the homogeneity of temperature distribution. Insufficient air volume flow can result in temperatures in the stack locally rising well above 800 °C. The resulting high temperature gradients can jeopardize the mechanical integrity of the stack. Experimental conditions during the operating tests were adjusted appropriately.

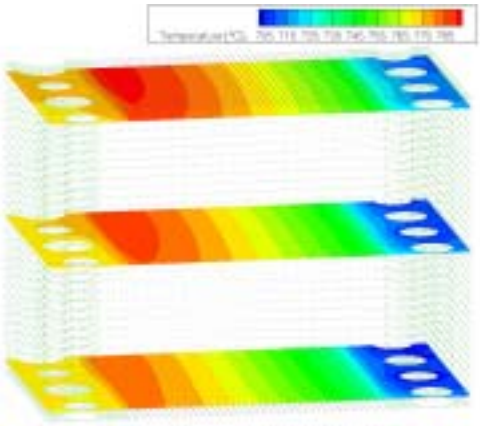


Fig. 56: Simulation of temperature distribution in a CS stack with 30 cassettes: only cassettes 3, 15 and 29 are shown

4.2.3 Operating test of cells and stacks

The operating tests on single cells were carried out to accompany the technology transfer of Jülich cell manufacturing technology to industrial partners. The measurements were performend in ceramic housings. A gold wire separated the air and fuel gas compartments Pt

mesh and Ni mesh were used as a current collector at the cathode and the anode. The measurements were all carried out using humidified hydrogen as a fuel gas. The results were used to optimize cell manufacture. Power densities for the anode substrate cells are typically up to 1 W/cm² at a working voltage of 0.7 V and an operating temperature of 800 °C (see Fig. 57).

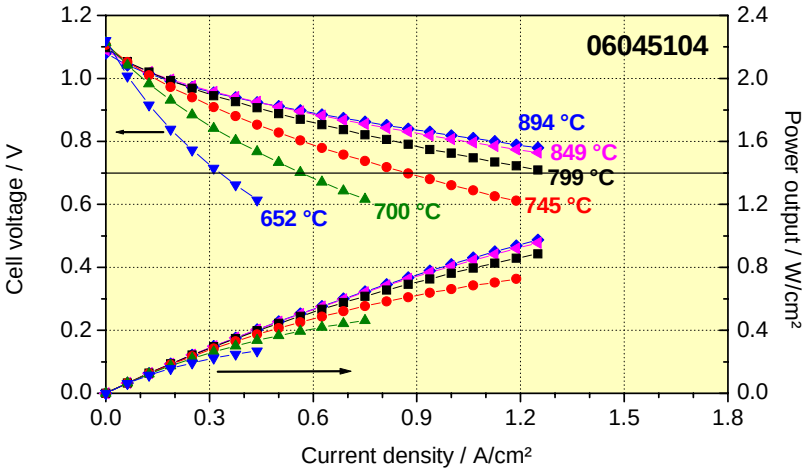


Fig. 57: **Current density/voltage characteristic curves (3% H_2O in H_2 vs air) typical of anode substrate cells**

The operating tests on stacks were carried out in order to increase power, improve long-term stability and resistance to thermal cycles.

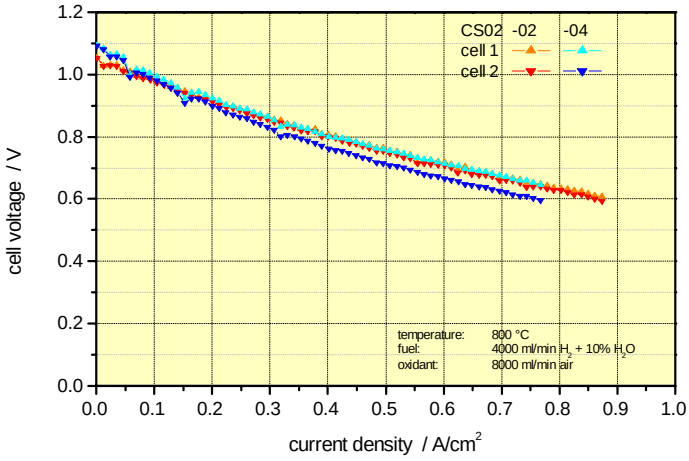


Fig. 58: **Current density/voltage characteristic curves (10% H_2O in H_2 vs air) typical of CS design stacks**

With a CS design stack at 0.7 V, 800 °C and 10 % H_2O in H_2, a current density of 0.63 A/cm² was achieved (see Fig. 58). The resulting power density of 0.44 W/cm² is typical of the CS design. The power density in the stack is considerably lower than the power densities in the single cell (see Fig. 57). The simulation calculations for contact resistance described above have already shown the main reason for this. The manufacturing limitations for the sheet metal parts have so far stood in the way of any improvement.

Long-term stability was tested on a CS stack for 5000 h (see Fig. 59). During an initial period of 2000 h the stack was run at 800 °C under constant current load (0.3 A/cm²) with 10 % H_2O in H_2 at a fuel gas utilization rate of 9 %.

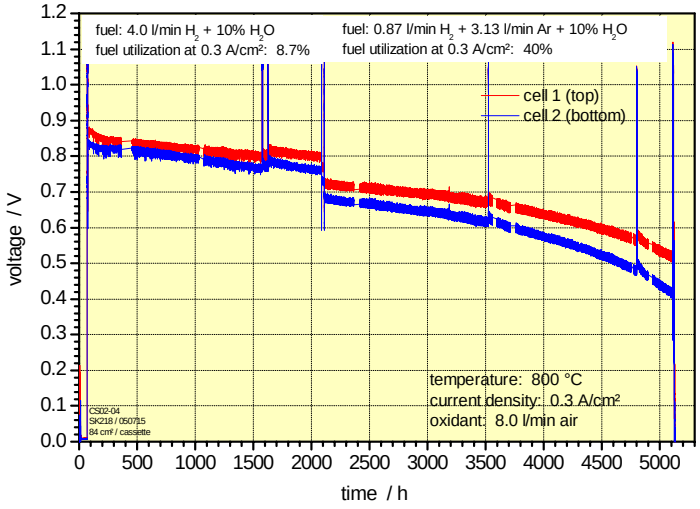


Fig. 59: Cell voltages over time during continious operation of a CS design stack at 800 °C with 10 % H_2O in H_2 at a variable fuel utilization

During a second period the fuel utilization was increased to 40 % by replacing part of the hydrogen with argon. A degradation that steadily increased over the entire operating time of 5000 h was observed. This progressive degradation has already been observed in other tests, including tests with standard F design stacks. For F design stacks, the timescale for the progressive behaviour is considerably longer and the degradation rate is considerably lower. The precise reasons for this degradation are not yet known, and research therefore continous.

The resistance of a CS design stack to thermal cycles was tested with a 10-cell stack in a new test stand. In this new test stand the stack was not heated up in a furnace but via two heating plates installed above and below the stack. The stack and the heating plates are thermally insulated. The stack can also be heated from inside using air brought up to temperature by a pre-heater. This concept was first used to show that a 10-cassette stack can be brought from room temperature to 600 °C within 40 minutes. The influence of these thermal cycles on the stack was investigated by operating the stack every time after it had been heated to 600 °C. The stack was operated potentiostatically for 15 minutes at a voltage

of 7 V, while the heating plates heated the stack up to a target temperature of 750 °C. It proved impossible to heat the stack to the target temperature within the 15-minute period. The current load of the stack was nevertheless switched off after every cycle and the stack was allowed to cool. It took approximately 4 h for the temperature of the stack to fall below 300 °C. The 10-cassette stack was put through a total of 115 thermal cycles. The measurements taken during individual cycles provide information on the status of the stack (see Fig. 60).

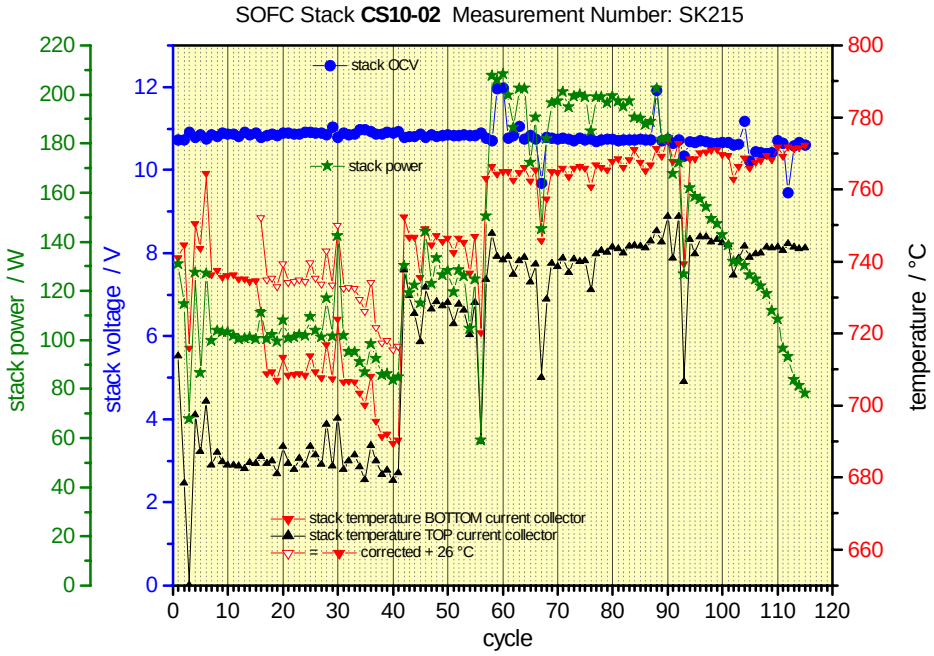


Fig. 60: **Stack temperatures and stack power after 15 minutes' current load during every cycle on the CS10-02 stack and open stack voltage 2 minutes had been after the current switched off in each case**

Open cell voltage is an indication of the mechanical integrity of the stack. No serious leakage was observed either during or after any cycles. Comparing stack power with stack temperatures shows that the values for power track the progress of the stack temperatures almost exactly. Although a moderate heating time was used in this first test, compared with the target time of 5 minutes, the results show that a CS design SOFC stack with a glass ceramic seal can be subjected to multiple thermic cycles.

4.3 Mixture formation in autothermal diesel reformers

4.3.1 Challenges in diesel reforming

One of the most important technical problems in diesel reforming is currently the lifetime of the system, which is very restricted due to the often unsatisfactory long-term stability of the catalyst. The objective of the most recent development work was to modify, improve and optimize the design and construction of the reformer in terms of flow technology and heat technology in such a manner that the reformer could be run on a standard European diesel fuel at full fuel conversion over the long term, and thereby to establish the practicality of diesel reforming for fuel cell systems.

As well as the development of suitable catalysts, the quality of the educt mixture at the inlet to the reaction zone also plays an important role. A homogeneous mixture of educts enables the maximum use to be made of the reformer capacity and ensures that the chemical reactions involved will proceed in an ideal manner. Even more important, however, is the fact that a poor-quality educt mixture can lead to the formation of carbonaceous deposits and so-called 'hot spots' in the reaction zone, which would damage the catalyst.

A mixing chamber of a reformer must atomize and vaporize the fuel, mix the educts and homogenize the flow. For middle distillates such as diesel or heating oil, above all complete vaporization presents the greatest challenge and is simultaneously a crucial requirement if a homogeneous mixture is to be obtained. High temperatures are required in order to apply sufficient heat to the fuel to vaporize it, which involves unwelcome risk that the fuel could ignite. During vaporization diesel shows a strong propensity for pyrolysis, which leads to carbon formation and carbonaceous deposits being accumulated. For this reason, optimizing the vaporization process is of great significance.

4.3.2 Methodology for optimizing the mixing chamber

Calculations and experiments were carried out in parallel at Research Centre Jülich to investigate and optimize the mixing process in the mixing chamber of the autothermal reformer.

To determine the quality of the educt mixture at the inlet of the catalytic reaction zone in the reformer, a computational fluid dynamics (CFD) model of the mixing chamber was created. In a first step, the CFD model simulates the mixing of steam and air. The injection, vaporization and mixing of the fuel were then modelled. The test began with an alkane used as a model fuel to represent diesel. More complex models were subsequently developed for diesel vaporization in which diesel was defined as a mixture of up to five hydrocarbons.

To verify and complement the CFD simulations, two types of flow visualization experiments were conducted in model reactors made of glass:

- experiments with optically labelled fluids to visualize and investigate complex turbulent flow in quartz glass models of the reactors on a 1:1 scale (see Fig. 61). This method is based on fluid dynamic similarity, which is defined by the Reynolds number.
- tests of the shape of the spray profiles of the injection nozzles used and the interaction of the intensive turbulent gas flows on the drop tracks (see Fig. 62).

Fig. 61: **Turbulent flow field in the glass model of an autothermal reformer: visualized using coloured water for the introduction of a steam flow (top) and an air flow (bottom)**

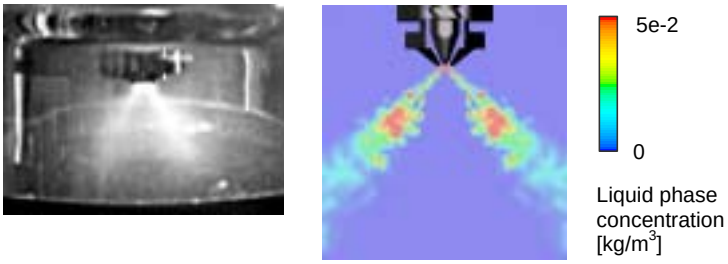


Fig. 62: **Comparison of the spray profile of an atomization nozzle in a mixing chamber: experiment (left); CFD simulation (right)**

These tests enabled weak spots to be identified for each of the designs in question. Understanding the vaporization and mixing process in this way made it possible to carry out modifications to the mixing chamber geometry that produced an optimized mixture.

4.3.2.1 Optimizing the original Type ATR-5 type reactor

The starting point for optimizing fuel mixture formation was the fifth-generation autothermal reformer (ATR-5A) for diesel fuel for a fuel cell system with a power of 3 kW_{el}. The educts air, steam and fuel are mixed together in the mixing chamber of this reactor. Air and the liquid fuel are injected into the mixing chamber of the reactor via a two-fluid nozzle. The dynamic effect of the expanding air means that the fuel is finely vaporized. Superheated steam is introduced into the mixing chamber via a curved steam pipe opposite the two-fluid nozzle. Using CFD simulations meant that the optimum operating conditions for the ATR-5A reactor could be identified and established. The drop in hydrogen concentration in the product gas during extended operation, which can be regarded as an indication of catalyst deactivation, was reduced by almost an order of magnitude as a result of the measures based on the CFD simulations.

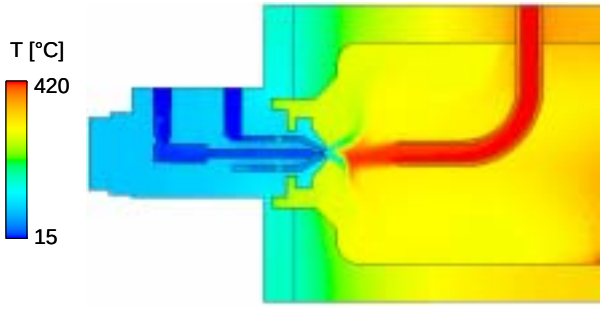


Fig. 63: Temperature curve along the mixing chamber with two-fluid nozzle

Fig. 63 shows the temperature profile along the mixing chamber as an example of numerically determined flow profiles. Another consequence was the construction of a performance-optimized variant of the reactor, known as ATR-5B. In this version, power was increased from 3 to 5 kW_{el}, for the same reactor volume and weight.

4.3.2.2 Concept development of the ATR-7 and ATR-8 reactors

The improvements made to the mixing process in the ATR-5 by optimizing the operating parameters proved insufficient for long-term operation of the reactor on conventional diesel, however. A progressive drop in catalytic activity in the region of 5 % was observed after 200 h operation. A new concept for the mixing chamber was thus developed for operation with conventional diesel fuel and optimized in terms of fluid mechanics.

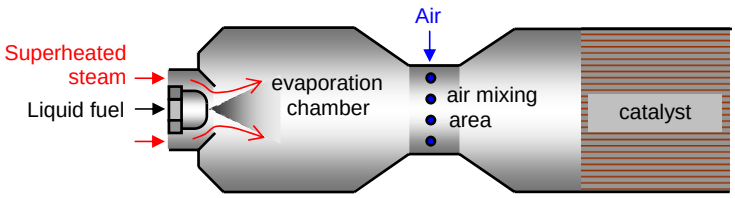


Fig. 64: Concept of fuel evaporation and mixture formation in the reactor ATR-7

The mixing chamber is divided into two zones. In the first zone, known as the vaporization chamber, superheated steam is introduced and liquid fuel is injected and atomized via a single-fluid nozzle. The absence of oxygen in the vaporizer prevents any ignition taking place. Another important aspect is the way in which the steam atmosphere in the vaporizer inhibits carbon formation. In the second zone, air is then added to the mixture of steam and diesel vapour. The autothermal reformer based on this concept is referred to as ATR-7 (see Fig. 64).

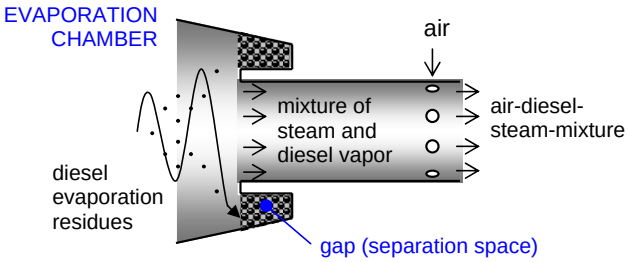


Fig. 65: Schematic of the separation of unvaporized residue from the gas flow in the ATR-8 reformer

In a modification to the mixing chamber concept, some unvaporized distillate residues were separated from the mixutre by the dynamic forces in the flow structures (see Fig. 65). This variant of the reformer is known as ATR-8.

4.3.2.3 Construction and operation of the new reactors

The seventh- and eight-generation reactors based on these new chambers, ATR-7 and ATR-8, were designed and tested. The catalyst used in these reactors was identical to that used in the ATR-5 reactor.

Fig. 66: Mixing chamber assembly in the ATR-7 reactor

The reactors were operated on *ARAL Ultimate* diesel for 500 or 1000 h, and no measurable decrease in catalytic activity was detected in either of the reactors during this time (see Chapter 1.3). These are the only reactors in the world to display such long-term stability in reforming conventional diesel fuel. The long-term stability achieved in the ATR-7 and ATR-8 diesel reformers means that the new concept can be successfully applied in fuel cell systems.

Subsequent analyses of the operation revealed a conversion for the fuel introduced into the ATR-8 of more than 99.999 % at the start of operation and 99.7 % after 1000 h of operation. The present state of the art currently achieves a conversion of approx. 98 %. The functionality of the new ATR-8 mixing chamber is several orders of magnitude greater than this.

4.3.2.4 Further development of reformer technology

Experience acquired during operation of the ATR-7 and ATR-8 diesel reformers was reflected in a further development of this concept. This knowledge included the fact that active cooling of the injection nozzle, particularly when the reactor is being heated, is very beneficial. In the ATR-7 reactor and the first variant of the ATR-8, the nozzle was cooled solely from the fuel flowing through it.

Fig. 67: Mixing chamber – upper section of the ATR-8 reactor with cooled nozzle fitting

A slightly modified version of the ATR-7 mixing chamber construction was used in an autothermal reformer for high-temperature solid oxide fuel cell systems (SOFC) run on kerosene, and was termed ATR-6B. The injection nozzle is easy to remove and replace, which is a great advantage for the maintenance of this critical component. The expected operating properties of this kerosene reformer may already make it the first step towards introducing the technology onto the market, in the form of a component in an SOFC fuel cell system, especially since the first major changes have been made at the design and production stage.

The ATR-9 variant has an improved fuel vaporization process compared with existing reformer concepts, obtained by modifying certain details of the mixing chamber design. In addition, production procedures have been optimized in terms of cost reduction or have been adapted for some assemblies.

Furthermore, a concept for a 50 kW_{el} kerosene reformer was developed based on the CFD calculations. Several new reactor-specific design features were evaluated as part of this process.

4.4 Process analysis of future CO_2-free membrane power plants

In the internal FZJ "CO_2 Separation" project, process analyses of future carbon-based membrane power plants were carried out. These analyses focused on developing 'post-combustion' and 'pre-combustion' concepts. In the BMWi project OXYMEM, which was implemented jointly with SIEMENS, new approaches for oxyfuel processes were identified.

4.4.1 Separation of CO_2 from the flue gas after combustion (post-combustion capture)

Fig. 68 shows the main boundary conditions for positioning a CO_2 membrane within its process environment at the exit of a coal-fired power plant. There is a law for porous membranes according to which the local permeate flow densities are proportional to the partial pressure differences of the permeating components (propulsion from the feed side to the permeate side of the membrane). With regard to the purity of the CO_2 product stream required for liquefaction, the following can currently be used as a working hypothesis (in IEF-3 and in various industrial projects work is currently being carried out towards exploring the question of CO_2 purity in greater detail):

- A CO_2 purity of 90 mol% is sufficient for problem-free CO_2 liquefaction by compression.
- For CO_2 purities under 90 mol% a preliminary purification process is required (partial separation of N_2 and O_2). For CO_2 purities of at least 80 mol% it should be possible to devise appropriate procedures whilst keeping efforts relatively low (efficiency loss in the range of 1 to 2 percentage points).
- For CO_2 purities below 80 mol% the efforts involved are likely to be unacceptable high, since low-temperature procedures would have to be employed.

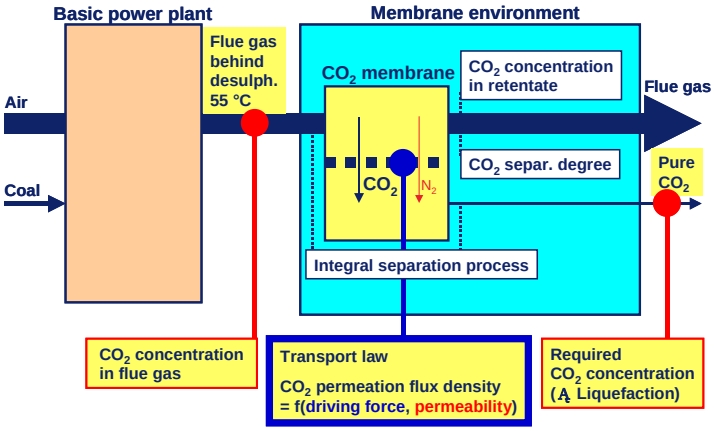


Fig. 68: Boundary conditions for use of a CO_2 membrane

The low partial pressures of the CO_2 in the flue gas (approx. 140 mbar) are problematic, since the pure CO_2 gas accumulates on the permeate side of the membrane and must then be liquefied. Added to this is the fact that not only does the propulsion for permeation (CO_2 partial pressure differential) drop over the course of the membrane process, due to the diminishing concentration on the flue gas side, but the selective CO_2/N_2 separation becomes increasingly difficult, as shown in Fig. 69.

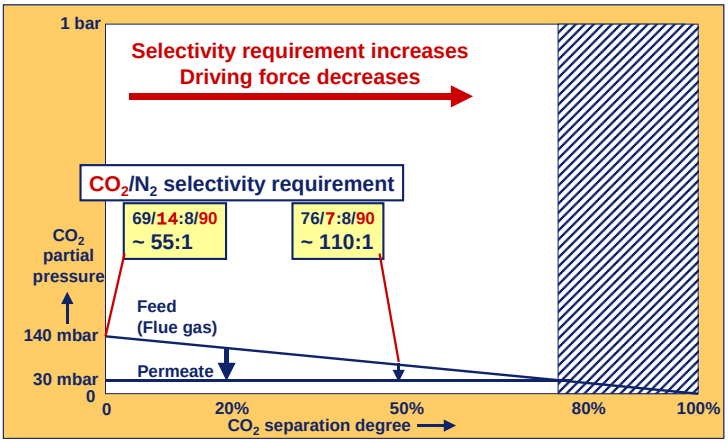


Fig. 69: Progressive separation process

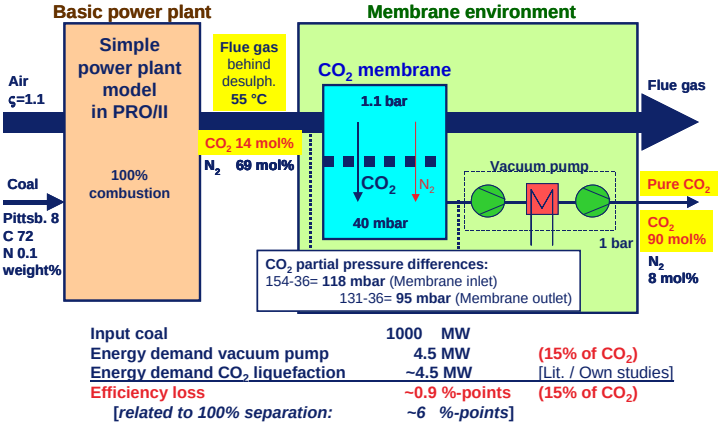


Fig. 70: CO_2 membrane with permeate vacuum in the flue gas of a coal-fired power plant, CO_2 separation level of 15%, CO_2/N_2 separation factor of the process 55:1 (=69/14:8/90)

Fig. 70 shows the results of a simulation calculation. A partial separation of approx. 15 % permits a compromise to be struck between the conflicting requirements of minimal efficiency loss on the one hand and providing suitable conditions so that the membrane can separate the gases on the other hand. A process engineering membrane environment with a vacuum pump (40 mbar) on the permeate side proved to be an attractive solution, with a loss of efficiency of approx. 1 percentage point including liquefaction. Extrapolated linearly, this corresponds to 6 percentage points for complete separation – around half of the loss encountered in conventional separation via amine washing, which typically results in efficiency losses in the 10 - 15% range.

Fig. 71 shows that increasing the pressure of the flue gas causes significant losses in efficiency compared with lowering the pressure on the permeate side, so this procedure should be employed only for high degrees of separation (which requires greater membrane selectivity). This also generates high levels of propulsion for CO_2 permeation (500 mbar in Fig. 71), so a membrane module in the power plant would become very compact and cost-effective relative to one ton of carbon dioxide separated. The calculations involve 4 principal factors:

- Ratio of the molar flows of the total flue gas/total CO_2 = approx. 7:1
- Ratio of the molecular weights for N_2/CO_2 = 28:44 = 0.64 = 1/1.57
- Approximately 2/3 of the compression work is recovered via an expansion turbine.
- No energy can be recovered from the vacuum pump.

In a cascaded arrangement with stayed membranes (required if membrane selectivity is insufficiently high), energy expenditure is high, since the relevant pressures must be created for each individual membrane. However, this means that the high degree of CO_2 purity required can be obtained in principle even if membrane development has not achieved the ambitious targets set in terms of CO_2/N_2 selectivities of almost 100:1.

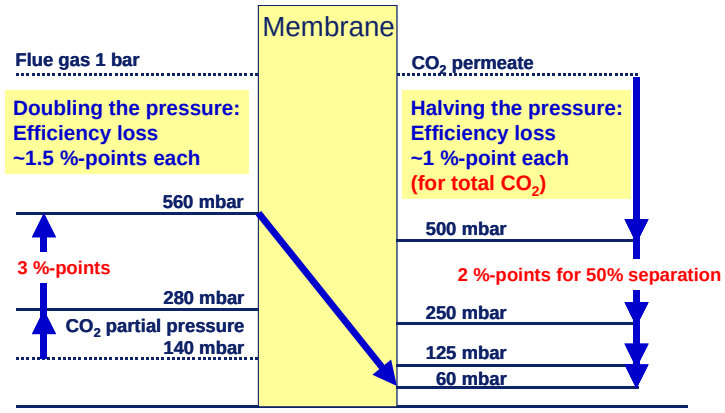


Fig. 71: Increasing the propulsion for CO_2 permeation by raising the pressure of the flue gas

4.4.2 Separation of CO_2 after coal gasification and combustion with air in front of (pre-combustion capture)

Fig. 72 shows an IGCC (integrated gasification combined cycle) with CO_2 separation. The coal gas, preferably produced using oxygen and under pressure (approx. 20 - 30 bar), shows high partial pressures of CO and H_2 on leaving the gasification process.

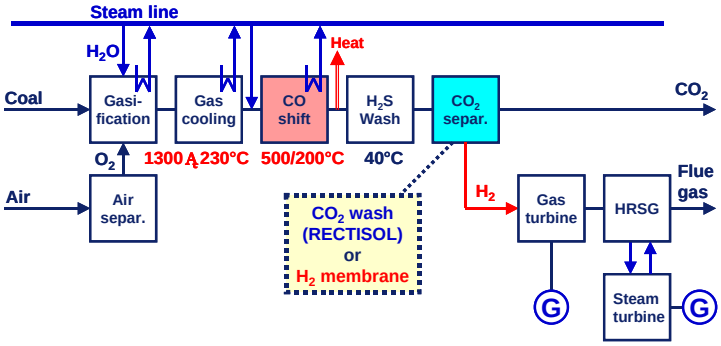


Fig. 72: IGCC power plant with CO_2 separation

A shift reactor to convert CO to CO_2 by adding steam and producing a CO residue of under 1 mol% was designed by SÜDCHEMIE (see Fig. 73).

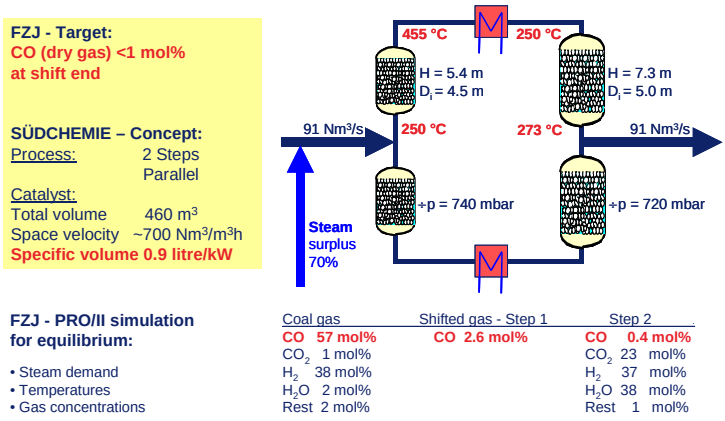


Fig. 73: CO shift in the IGCC power station (500 MW)

A porous H_2/CO_2 membrane positioned downstream with a H_2 separation level of min. 97 % produces the required pure CO_2 gas on the retentate side (see Fig. 74), but results in undesirable "CO_2 slippage" owing to its restricted selectivity. In view of the strong competition posed by the physical scrubbers (RECTISOL) available today with only approx. 7 - 10 %

points loss of efficiency with almost all CO_2 being captured, developing additional advanced protonic/electronic mixed-conducting membranes, promising excellent selectivities, seems to be particularly important.

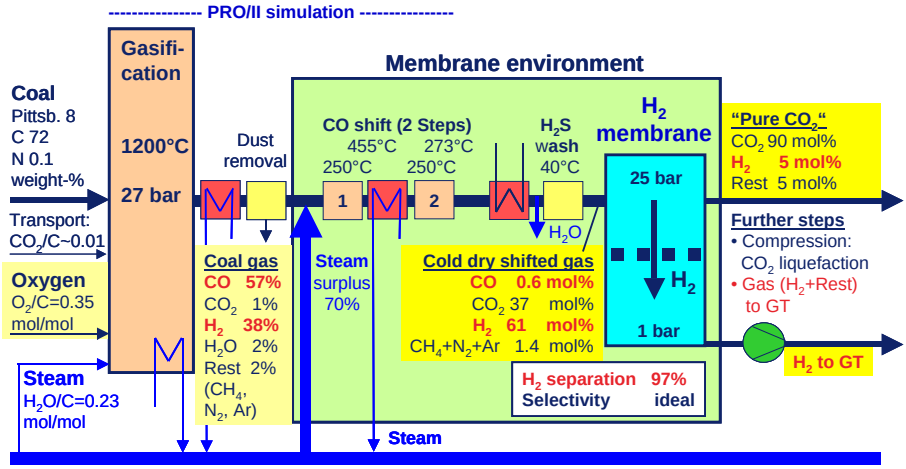


Fig. 74: IGCC power plant with H_2 membrane

4.4.3 Combustion with pure oxygen to CO_2 and H_2O and separation via condensation (oxyfuel processes or oxycoal)

In 2006, new and promising possibilities for combustion with pure oxygen using oxyfuel processes emerged for high-temperature O_2 membranes. These possibilities opened a gateway into power plant engineering via a (specifically lower quantity of) oxygen generation for coal gasification (approx. ¼ stoichiometric). A concept for incorporating a membrane into the IGCC process was proposed by AIR PRODUCTS. Variations of this concept were developed in collaboration with SIEMENS (OXYMEM Project) for burning coal with pure oxygen, and these variations resulted in an alternative to the OXYCOAL-AC process.This alternative is characterized by the fact that the permeate side of the O_2 membrane is no longer sweeped with recycled flue gas, and is therefore free from combustion products. It therefore now seems that a simple overall concept is possible.

The process analyses for an oxycoal power station, based on the more familiar OXYCOAL-AC process, show that greater difficulties are involved here. An initial design of the high-temperature section of the air pre-heater for heat recovery (air 20 bar, flue gas 1 bar) produced intolerably high specific weights and costs (high temperature, high differential pressure and large mass flow rates). A comprehensive optimization of the process is therefore required.

4.4.4 Staff members and fields of activity

Name	Telephone no. (+492461-61-) E-mail adresse	Field of activity
Prof. L. Blum	6709 l.blum@fz-juelich.de	Head of Fuel Cell Process Engineering (VBZ)
Dr. E. Riensche	6689 e.riensche@fz-juelich.de	Plant simulation, power plant engineering, reforming, CO_2 sequestration

4.4.5 Important publications and patents

Important publications

Linssen J., Blum L., Riensche E., Walbeck M.

CO_2 Capture and Storage: Possible Bridging Function for a Future Hydrogen Economy?

Proceedings of the 3rd Int. German Hydrogen Energy Congress (e-world), Essen, February 15-16, 2006, pp. 411 - 419

One of three possible routes for separating CO_2 in power plant processes involving fossil primary energy (carriers) is to capture CO_2 prior to combustion following coal gasification and the water-gas shift reaction (objective: to separate H_2/CO_2). The hydrogen is then burned with air. The coal gas, preferably produced using oxygen and under pressure (approx. 20-30 bar), shows high partial pressures of CO and H_2. A shift stage to convert CO to CO_2 by adding steam and producing a CO residue of under 1 mol% was designed by SÜDCHEMIE. A porous H_2/CO_2 membrane positioned downstream with an H_2 separation level of min. 97 % produces the requisite pure CO_2 gas on the retentate side, but results in undesirable "CO_2 slippage" owing to its restricted selectivity.
In view of the strong competition posed by the physical scrubbers (RECTISOL) available today with only around 7 % points' loss of efficiency with almost total CO_2 separation, developing additional advanced protonic/electronic mixed-conducting membranes, promising excellent selectivities, would seem to be particularly important.
The hydrogen separation taking place in this power plant leads to scenarios that could make a potential contribution towards creating an H_2 infrastructure in addition to merely generating electricity (bridging function of this CO_2 separation)

Meulenberg, W. A.; van der Donk, G. J. W.; Riensche, E.; Blum, L.

CO_2 separation with ceramic membranes and concurrent procedures

Carbon Capture and Sequestration Tagung - CO_2-Separation and Storage: A future option for German environmental protection strategy, 10.-11.11.2005. - Jülich, 2005. - (STE Arbeitsbericht; 4). - pp. 37 - 67

Fossil-fuel powered power stations in Europe are set to be modernized over the coming decades. The magnitude of replacement needed in Germany is 40,000 megawatts (200,000 megawatts for Europe), which corresponds to the output of 40 large power stations, or in other words 40 % of the current power station capacity.
The possibility of separating gas in order to separate the greenhouse gas CO_2 can be achieved using three possible power plant variations and various separation procedures. One potentially suitable method with considerably lower losses of efficiency compared with conventional separation procedures, such as chemical scrubbers, is gas separation using ceramic membranes. Ceramic membranes have high chemical and thermic stability and can be incorporated into all power station processes. The present article describes different membrane concepts for gas separation and gives an example of how membranes can be incorporated into a power station design with a calculation of the relevant losses of efficiency.

Important patents

Patent applications:

Principal Inventor	PT	Title
Dr. E. Riensche	1.2220	Power plant with CO_2 hot gas recycling and process for operating the same
Dr. E. Riensche	1.2220 PCT	Power plant with CO_2 hot gas recycling and process for operating the same
J.M. Serra Alfaro	1.2263	Proton-conducting layer system and process for manufacturing the same

5

Outlook

Outlook for Future R&D Projects

- Physicochemical fuel cell laboratory
- Systems with high-temperature polymer electrolyte fuel cells
- Commercialization of DMFC systems in the kW class

5.1 Physicochemical fuel cell laboratory

Although great technical progress has been made with fuel cells in recent years, the technology is still not sufficiently advanced to bring them to market. The already successful research and development work needs to be continued, but a deeper, more fundamental understanding of the physicochemical processes is also needed. It is thus vital to have detailed knowledge of the electrode processes in polymer fuel cells and analysing the vaporization process and catalyst layers during reforming. IEF-3 has begun the construction of a 'Physicochemical Fuel Cell Laboratory', funded by €3.6 m in start-up investment from the HGF, containing the equipment necessary to perform these analyses. The laboratory is scheduled to be completed in early 2008.

5.1.1 Background

IEF-3 is carrying out an integrated development of fuel cell technology in the fields of DMFC, HT-PEFC, SOFC and diesel reforming. To this end it has at its disposal essential tools such as systems analysis, modelling, cell and stack technology as well as systems engineering. Benchmarks show that the activities in these fields are at the forefront of current research. Recent years have seen an increase in the importance of analytical methods that allow structures and effects to be analysed either in situ or with very fine local resolution. It is precisely this approach that IEF-3 intends to pursue in the laboratory now under construction. The laboratory is designed to represent, at least in the medium term, a unique selling point for a fuel cell institute on the European level and beyond. The equipment is intended to further extend IEF-3's leading position in fuel cell development. The underlying research themes receive significant support from industry and facilitate close and noteworthy cooperation with industry. Interestingly, this orientation towards practical application leads to additional basic research, as themes such as reducing ageing and increasing power density and efficiency become important once again, but at advanced stages of development can be tackled solely by carrying out basic research. This is the reason why there is an increasing need for basic scientific research.

5.1.2 Description

A major theme to have emerged from R&D work over the past few years is the clarification of the structure-effect relationships of functional layers. Currently available facilities and methods for physicochemical analysis do not permit the necessary leap towards gaining a deeper fundamental understanding of the structure-effect relationships. Further extending the results obtained from basic research into electrode and catalyst development in terms of performance, stability over time and costs requires visualization methods that extend down to the nanometre scale, plus on-site chemical analysis and electrochemical microanalysis. This also includes methods for gently preparing samples of moist polymers; which are currently being borrowed and further developed from biology, medicine and surface engineering. The methodology and theme for all the equipment in the laboratory will focus on the following fields of analytical work (see Fig. 75):

A. Imaging

1. Microscopy
   Analysis of the morphology of membrane electrode assemblies (MEAs) and the distribution of components
2. Locally resolved electrochemical characterization
   Identifying weak spots of MEAs, cells and stacks under load in order to optimize operating conditions
3. Fluid dynamics analyses
   Tracing rapid processes in reformers using high-speed cameras with magnification optics
4. Spectroscopy
   Use of spectroscopic methods to analyse reaction intermediates

B. Dispersion development

5. Structural analysis of catalyst dispersions
   Production of reproducible processable dispersions for the manufacture of high-performance MEAs
6. Physical properties of MEAs and cell components
   Control and determination of mechanical and thermodynamic requirements of fitness for use in fuel cells
7. Reaction technology analyses
   Determination of optimal operating points at low degrees of catalyst ageing

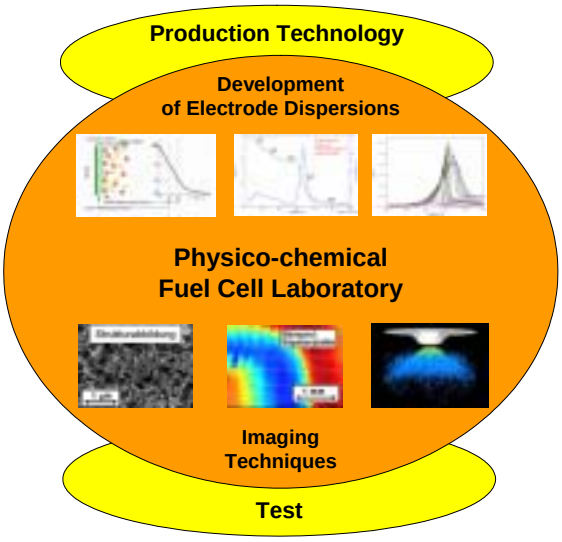


Fig. 75: Integration of the Physicochemical Fuel Cell Laboratory

This laboratory is a further step towards implementing the Senate's recommendation to build a high-temperature PEFC membrane centre. FZJ does not intend to synthesize membranes itself – its target is to investigate and optimize the properties of the membrane and the electrodes. The laboratory is furthermore of primary importance in terms of in-depth electrochemical research into implementing existing results, for example in order to commercialize the DMFC scooter. This investment measure is also supported in particular by the recommendation from the current review by of the German Energy Advisory Council that the work by FZJ on the behaviour and demonstration of complete fuel cell systems under realistic operating conditions should be pursued further.

5.2 Systems with high-temperature polymer electrolyte fuel cells

5.2.1 Motivation and objectives

HT-PEFC development, from the cell all the way up to a whole system, should be the new emphasis for work of the Institute. The significance of this technology as part of the EU's strategic research agenda (SRA) has recently been underlined by industry. Simultaneously, canvassing carried out by IEF-3 has shown that there is a considerable degree of interest on the part of industry for systems with a performance class in the 5 kW region based on liquid energy carriers. Application-orientated systems can be constructed in combination with the IEF-3 reformer. The reforming technology was actually designed for the conventional polymer electrolyte fuel cell (PEFC); it emerged during the course of the work, however, that preferential oxidation was extremely difficult to control and has evidently not been mastered anywhere in the world at the present time. We therefore intend to combine diesel reforming with the HT-PEFC in order to arrive at a system that tolerates approx. 1 % CO in the fuel gas at an operating temperature of 160 °C without major performance losses. This corresponds approximately to the fuel gas quality following the shift stage of fuel gas preparation.

Jülich possesses the major capabilities required to adopt this technology as part of the fuel cell technologies it already operates.

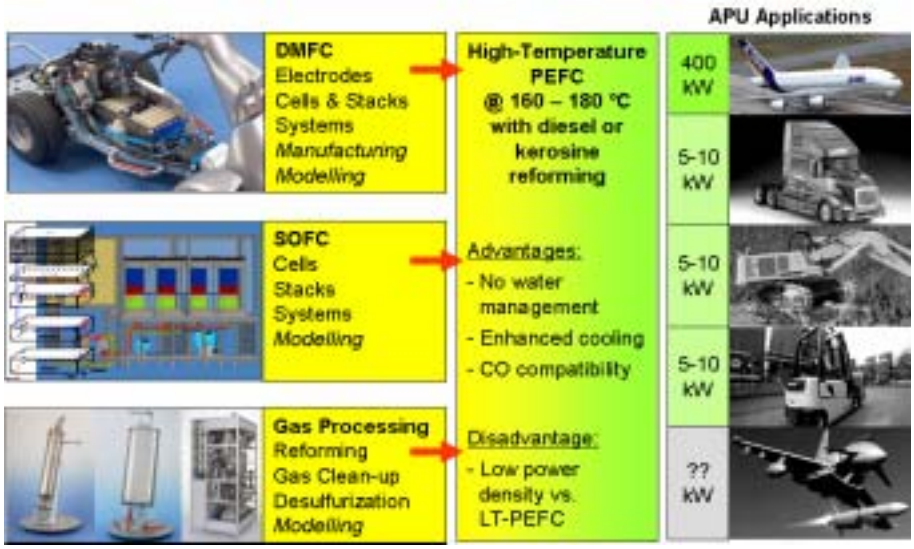


Fig. 76: Synergies from three development lines for HT-PEFC

Fig. 76 shows the main sources of technology on the left-hand side and their applications on the right-hand side. This means that electrode, cell and stack technology are all essentially covered by prior DMFC know-how in the Institute. Essential parts of the modelling can be transferred from the corresponding SOFC activities. The physics behind the processes can be transferred directly, although the material and temperature-related constants must be determined afresh. Gas process engineering can be transferred directly to the HT-PEFC,

both in terms of process control and in alsoof manufacturing components in their performance class.

The HT-PEFC is eminently suitable for a combination with reformers owing to its higher CO tolerance. It is additionally well-suited to smaller systems, since there is no need for any complicated water management. A further advantage is its improved cooling ability compared with the conventional PEFC at 80 °C. Its disadvantages, on the other hand, are its lower power density and the greater difficulty involved in"cold starts".

Most of the applications in the 5 kW performance class are as auxiliary power units for various civil applications – aircraft, trucks, construction machinery – and possibly also for various applications in the 'dual use' field. This technology also shows potential for light traction in forklift truck operation within the 10 - 20 kW power class. In the longer term, domestic CHP heating appliances run on fuel oil or liquid biofuels represent an additional field of application.

The plan is to develop this technology analogously to DMFC from cells up to complete systems in IEF-3 with the close involvement of industry partners, as yet to be acquired. We are currently working with the membrane manufacturer FuMA-Tech, for whose membranes IEF-3 is currently developing electrodes and MEAs. Furthermore, a complete system based on HT-PEFC stacks from Sartorius with an electrical power of 5 kW will be demonstrated in 2007. The details of this system are currently being designed, the components are being manufactured and the system will be constructed in early 2007 to include gas preparation from diesel. These activities are being actively supported with projects from Airbus Germany.

5.2.2 HT-PEFC stack development

As preliminary work for the construction of complete high-temperature PEFC systems since 2005, IEF-3 has been developing fuel cells capable of operating at a temperature of 160 – 180 °C and at ambient pressure, which can use either hydrogen or reformate (with a carbon monoxide content of up to 1 %) and ambient air as reaction gases. Additionally, in contrast to conventional PEM fuel cells, there is no need to laboriously charging these cells with steam.

This development line is based on the new range of membranes from a cooperation partner that can absorb large quantities of phosphoric acid. The anodic and cathodic catalyst layers are optimized at IEF-3 to accommodate this material and are combined with the membrane. These cells are then characterized at the Institute under the appropriate operating conditions for their intended uses.

In just 18 months, it was possible to develop, and manufacture reproducibly, membrane electrode assemblies (MEAs) whose electrochemical performance and long-term stability are only slightly below those of the high-temperature PEFC MEAs currently available from the sole commercial supplier. At 160 °C performance densities of more than 300 mW/cm^2 were obtained during operation with hydrogen and almost 200 mW/cm^2 with reformate. A long-term stability of over 1000 h in a single cell has been documented; the mean degradation rate depends on the load profile of the fuel cell and is currently extrapolated as just 3 % over 1000 h under favourable conditions (see Fig. 77).

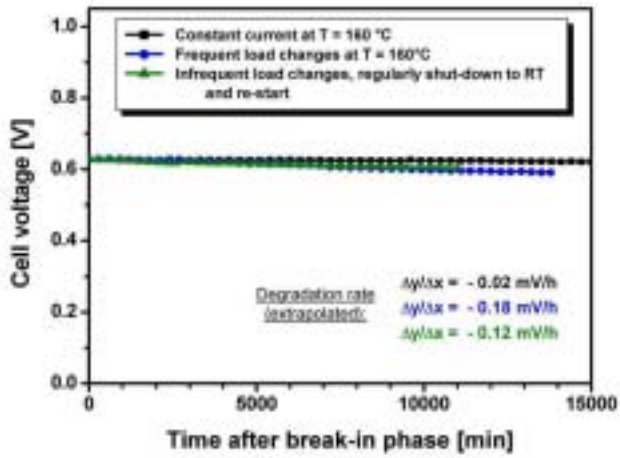


Fig. 77: Operational behaviour of an HT-PEFC (single cell)

Moreover, a first HT-PEFC stack in the 100 W class has already been developed (see Fig. 78). This stack consists of 10 single cells, each having an area of 50 cm². This stack is distinguished by the cartridge system used to separate the coolant circuit from the current conductors. This construction makes it possible to replace individual cells without any coolant flowing into the catalytically active areas. The modular stack design also basically enables an air cooling system to be used in place of the coolant circuit.

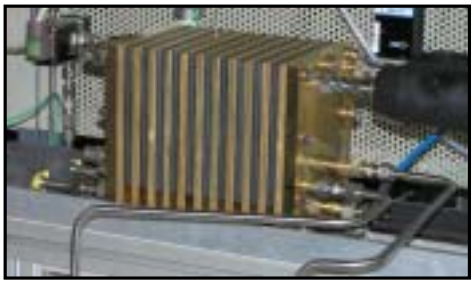


Fig. 78: HT-PEFC stack in the 100 W class

The most significant challenges for the future are to further increase the power density of the MEA (particularly by lowering the performance-draining cathode overpotential), to optimize bipolar/cooling cells and to further develop stack configuration, especially in terms of improving cold-start behaviour and durability during dynamic operation. The research work required to do this will be especially supported in future by the construction of the Physicochemical Fuel Cell Laboratory.

5.2.3 HT-PEFC system development

High-temperature polymer fuel cells are already characterized by simpler process engineering at an operating temperature of 120 °C. Their higher tolerance of carbon monoxide is, however, of technical benefit to the reforming process only at temperatures above 160 °C, since at these temperatures an otherwise critical gas purification stage can be dispensed with. This makes the process engineering simple, even if carbonaceous energy carriers are used.

Whereas using hydrogen as a fuel for cars is currently being considered, utility vehicles, construction vehicles, ships and aircraft will all continue to run on diesel or kerosene. Significant energy savings can be made in today's increasingly electrified vehicles by generating electricity more efficiently on board. Fuel cells enable efficient electricity generation to take place on board, especially when the drive train is idle. These so-called auxiliary power units (APUs) must be able to be run on the fuel already stored on board the vehicle in a way that is practical for the end user, which means that these so-called middle distillates have to be reformed.

With the above mentioned qualities, high-temperature polymer fuel cells are thus also particularly suitable for domestic energy systems. This is true for systems that are run on natural gas as well as for those run on fuel oil. Since fuel oil is also a middle distillate, in the same way as the energy carriers mentioned above, the development shares a lot of common features. This is true in terms of both reforming with subsequent gas purification and of the entire systems engineering itself, since the resulting reformates display very similar compositions. In Germany, the majority of homes are heated using natural gas. A not insignificant number of homes (approx. 25 %) still use fuel oil, however. Here too, we can expect that power-heat coupling will produce a considerable on the spot effect on primary energy consumption and specific CO_2 emissions. Primary energy savings of up to 25 % can be achieved in comparison with using a power plant mix (35 % efficiency) combined with a boiler (95 % efficiently) whilst simultaneously reducing specific CO_2 emissions by up to 35 %. In addition to efficiency-related aspects, the resulting reduction in other pollutants such as NO_x, is also a clear advantage. Facilities run on fuel oil are furthermore genuine decentralized systems, since they are not connected to the mains and can therefore be run autonomously depending on the capacity of their fuel tank. These aspects, and the fact that fuel oil systems represent a major potential market, are prompting heating system manufacturers to look for technologies that can offer these advantages. The HT-PEFC is an interesting technology by virtue of its relatively high resistance to CO and the favourable temperature level of the waste heat for of heat extraction.

Differences in process engineering are the result of the higher sulphur content of kerosene and fuel oil as compared with diesel. Aircraft also require particularly light systems.

Development of the HT-PEFC in the central cell and stack field has been in progress for several years now, principally in Germany. Various commercial suppliers are currently attempting to achieve different levels of integration for their productis (FuMA-Tech: membrane; PEMEAS; membrane electrode assembly; Sartorius: stack).

Since only one external supplier currently exists for each of these development stages and it would appear their reliability to deliver punctually over several years cannot be guaranteed, IEF-3 has set itself the objective of carrying out the entire development process in-house, from single cells to complete systems, although this process expressly excludes membrane

development. As part of this development, high-performance membrane electrode assemblies have been developed at the Institute since 2005. The first in-house stack was constructed in late 2006 (see Chapter 5.2.2).

Improving the multilayer MEA primarily concentrates on optimizing the mechanical properties of the membrane, immobilizing the electrolyte and improving the kinetics of the cathode reaction. Special electrochemical methods developed partially in collaboration with universities (e.g. investigating reaction kinetics using local electrochemical probes) are particularly targeted at gaining an understanding of high cathode overpotentials (phosphate adsorption at Pt catalyst, oxygen solubility and transport); other major investigations cover the development over time of gas permeation through the membrane and the influence of trace gases contained in the reformate on the function of the HT-PEFC.

In the newly-constructed IEF-3 Physicochemical Fuel Cell Laboratory, both standard technologies (DSC, TGA, BET, porosimetry) and analytical techniques specially tailored to HT-PEFCs will be used, such as investigating the as yet insufficient durability of the membrane under changing operating conditions, e.g. via stress/strain tests at 160 °C and controlled humidity. Preparing samples for high-resolution microscopy of MEA cross-sections still poses a challenge, and the first step in meeting this challenge is to design and implement suitable preparation techniques. Closely meshing analysis and materials development will ensure that research into MEA components will be particularly efficient.

Fuel cell stacks will be developed that operate at temperatures of between 160 and 180 °C and do not require any separate humidification. HT-PEFC cells of this type are based on plastic membranes treated with phosphoric acid (polybenzidimazole, PBI). These membranes are particularly mechanically sensitive and the load must be applied as uniformly as possible in the flow field, and in the sealing region. At the same time, the careful heat removal is needed for the components in order to dissipate product heat evenly and avoid any local overheating. Based on the materials used for bipolar plates, the following development lines can be differentiated:

- stacks based on metallic bipolar plates
- stacks based on graphitic bipolar plates

In principle, metallic bipolar plates can be used for compact designs that save both space and weight. On the other hand, graphitic bipolar plates have advantages in conventional PEFC stacks in terms of long-term stability and processability.

In addition, the existing components and stacks have not yet been tested to a sufficient degree. Practical and rapid systems with good market prospects can be developed only in a closed R&D circuit extending from individual components to whole systems and back again.

The intention is to pursue system development further in combination with the autothermal reformer technology developed at IEF-3 for three lines of application (APU; kerosene and diesel and domestic energy systems; fuel oil) by verifying one aspect of the system per year. Systems engineering development guidelines and results will be defined and evaluated using the criteria of power density and weight, device efficiency, ageing and dynamics. At least two of the specifications mentioned should be improved every year. The target is to create a compact basic system in which the fuel gas generation modules for kerosene, diesel and fuel oil are interchangeable. In addition, the most promising stack technology in terms of its system technical suitability should be identified within five years. This should provide the

technological basis for industrially relevant topics, such as adapting manufacturing techniques and quality assurance measures as well as product developments, verifications and field tests.

In parallel to increasing performance in fuel cell modules, a considerable weight and volume reduction should also be achieved, primarily via different approaches to incorporating system components and by dispensing with certain components. Thermal integration of the stack and reformer and simplifying safety technology are the first important approaches. Device efficiency should be improved by systematically reducing energy consumption and increasing the performance of the indispensable system components. Ageing behaviour during system operation is not comparable with the behaviour of individual components under test conditions in the laboratory. If ageing rates are to be lowered it is essential to verify and evaluate systems and to use the results in order to develop both components and systems further. In addition, particular attention should be paid to the dynamic behaviour of the system. Rapid heating and rapid load changes require modifications to be made to the hardware and an adaptation of the system controls.

5.3 Commercialization of DMFC systems in the kW class

5.3.1 Motivation

The development of direct methanol fuel cells (DMFC) has been at the forefront of research activities at the Institute of Energy Research – Fuel Cells (IEF-3) for the past 9 years. Around 25 people work on the project, more than half of whom are funded by the Research Centre's own resources. IEF-3 has refined DMFC technology from laboratory status to a state where it can be used in small mobile applications (scooters) over the past few years. A joint market study (see Chapter 4.1) carried out with the management consultancy firm Management Engineers (ME) showed that the best chance of commercialising of a DMFC energy system in the kW class was to use such a system to power a forklift truck (see Fig. 79).

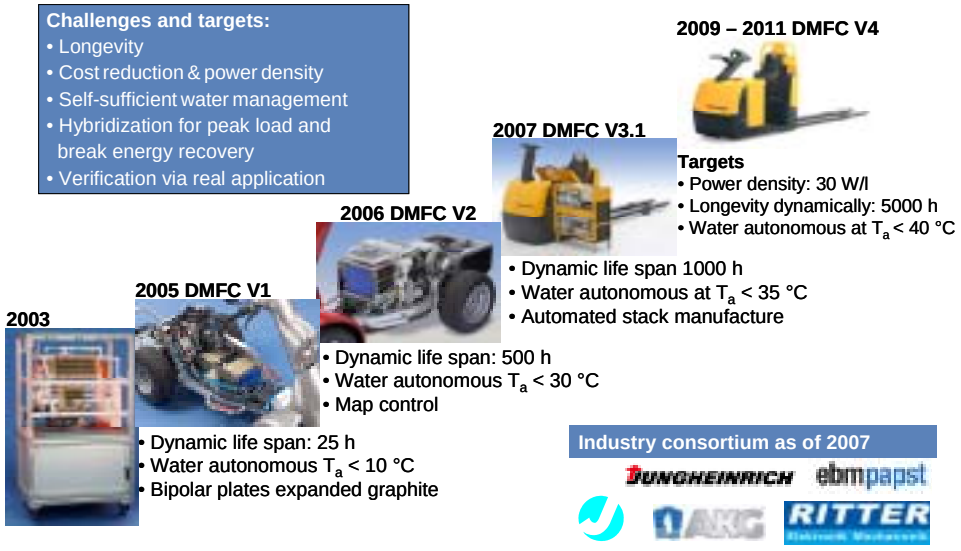


Fig. 79: Example of successful system development: from demonstrator to consortium

An advantage of such energy systems is that they dispense with the need for the comparatively laborious and time-intensive process of recharging batteries and the need for spare batteries in multi-shift operation. DMFC energy systems can be used here to replace the existing Pb accumulators, both structurally and in terms of energy. It was possible to show, as part of a feasibility study, also carried out jointly with Management Engineers, that fuel cell hybrid systems based on direct methanol fuel cells (DMFC) have the potential to achieve a comparable cost level to conventional battery systems when used in forklift trucks and can thus be used as an entry point for fuel cell applications onto the market. In order for this use of DMFC fuel cells to be cost-effective, research and development still needs to be carried out on the service life and power density of the stack and the overall development/hybridization of the DMFC energy system.

As part of a project, joint DMFC systems are now to be realized and applied in pilot operation to power electrically driven forklift trucks. The firms of Jungheinrich AG, Ritter Elektronik GmbH, Ebm-Papst Landshut GmbH and AKG Verwaltungsgesellschaft mbH have formed a consortium with Research Centre Jülich in order to contribute their skills to the joint development project. The requirements, in addition to reducing costs, are service life over a period of at least 5000 h of operation and an increase in the specific power density of the fuel cell system to approximately 30 W/l at a total efficiency of Ó30 %.

5.3.2 Planned work

A development plan based on milestones and extending to the start of series production of the DMFC energy system in 2011/2012 was laid down together with the partners (see Tab. 5).

	Phase 1		Phase 2
Dvlpmt. phase	**DMFC V2** 	**DMFC V3** 	**DMFC V4** 
Dvlpmt. targets	Â Service life 1,000 h at 10% Â 50 mW/cm² Â No water at 30°C Â Efficiency Ó20%	Â Service life 3,000 h at 25% Â 75 mW/cm² Â No water at 40°C Â Efficiency Ó25 % Â Cost competitiveness	Â Service life 5,000 h at 25% Â 100 mW/cm² Â No water at 45°C Â Efficiency Ó30% Â Cost competitiveness
Decision re.	Â Build 3 - 5 DMFC V3 Â Function sample Â Function test Â Cooperation agreement	Â Development production processes Â Build prototypes DMFC V4 Â Field tests Â Purchasing negotiations Â Service development	Â Set up production Â Service concept Â Sales training

Status: July 2006 Sources: FZJ,

Tab. 5: Development plan extending to start of series production for the DMFC energy system

During the first research and development phase the focus of the work on DMFC technology and hybridization is on aspects such as:

- long-term stability
- cost reduction, power density
- water autonomy
- prototype development

The work in Research Centre Jülich concentrates on refining the components of the membrane electrode assembly (MEA), investigations into ageing, stack development, systems development, energy storage systems and overall design. During this development phase a total of 3 energy systems will be built, and at the end of the project phase two functional samples (DMFC V3) should be available for field tests, which should be helpful in

terms of making decisions with regard to further development towards commercial viability and market launch.

In this second phase, the emphasis is on preparations for marketing and demonstration in field tests. The following activities are planned in this regard:

- refinement of fuel cell and system components
- development of production processes
- construction of prototypes
- field tests
- accompanying R&R for performance optimization and long-term stability
- recycling of stack components (catalysts)

5.3.3 Core tasks

The core tasks or work packages for the partners as part of their cooperation include the following activities during Development Phase 1:

Ritter Elektronik

Ritter Elektronik will be responsible for developing and building the prototypes for the power electronics within the project (e.g. DC/DC actuators) and for controlling the DMFC energy system. In its planned role as a future stack and system integrator, Ritter Elektronik will construct one of the three DMFC V3 prototypes.

Research Centre Jülich

Research Centre Jülich will be responsible for investigating in particular long-term stack stability under realistic operating conditions as part of the project, as well as stack and system development. A long-term stability with < 25% degradation after 3000 h is planned for the last stack of the DMFC V3 prototypes. An additional requirement is that the system be water autonomous at an ambient temperature of 40 °C. Research Centre Jülich will also carry out the complete development/hybridization and construction of 2 DMFC V3 prototypes and 3 stacks. The work on hybridization will encompass modelling and simulating various energy storage media as well as recording long-term profiles and operational cycles for the forklift truck under real operating conditions and investigating various energy storage media (batteries, SuperCaps) and connecting them in a test stand.

Jungheinrich AG

Jungheinrich will provide an ECE 220 forklift truck (see Fig. 80) for use in building the prototypes in the project and will provide support for the design of the DMFC energy system in the form of data and information on typical uses of the forklift truck in question. If it should become necessary to carry out adjustments to the forklift truck during the project, Jungheinrich will provide all information and practical assistance required in order to implement such adjustments. Furthermore, Jungheinrich will provide information in a timely manner regarding technical requirements, standards and special features that need to be taken into account as well as regarding DMFC-specific requirements and the expectations of final customers. Finally, Jungheinrich will also carry out the corresponding vehicle tests with the DMFC energy system.

Ebm-Papst

Ebm-Pabst will be responsible for developing and building prototypes for blowers and compressors for the DMFC system as part of the collaboration. Accommodating the blowers and compressors in the whole system and thereby positioning the cool air flow in the battery hutch as favourably as possible will be a core task in this regard.

AKG

As part of the collaboration, AKG will be responsible for developing and building prototypes for the condenser/heat exchanger, which enable the system to be water autonomous in the limited space available. The design of the condenser will be determined in close agreement with Research Centre Jülich and Ebm-Papst in order to ensure that the condenser is positioned as favourably as possible within the whole system.

Fig. 80 shows a hybridized battery/DMFC system (DMFC V3.1) in a battery hutch that is planned to be presented to the public for the first time at the Hanover Fair 2007.

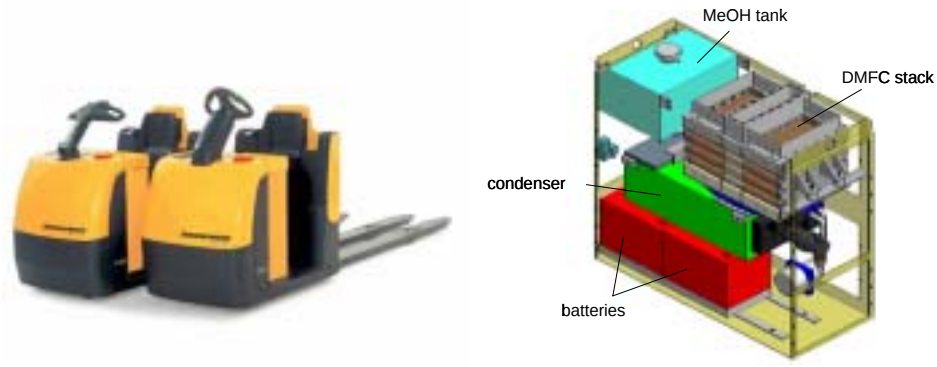


Fig. 80: ECE 220 forklift truck and DMFC energy system V3.1 design study

6

Data

Facts and Figures

- Institute of Energy Research – Fuel Cells (IEF-3)
- Overview of department competences
- Publications, technology transfer and resources
- Committee work
- Contributions to trade fairs and exhibitions
- How to get there
- List of abbreviations

6.1 Institute of Energy Research – Fuel Cells (IEF-3)

The Institute for Energy Process Engineering (IEF-3) concentrates on the research and development of fuel cells. It pursues three principal pillars in this regard – direct methanol fuel cells (DMFC), ceramic solid oxide fuel cells (SOFC) and reforming liquid energy sources for use in providing hydrogen for fuel cells. Another branch is currently being established in the form of high-temperature polymer fuel cells.

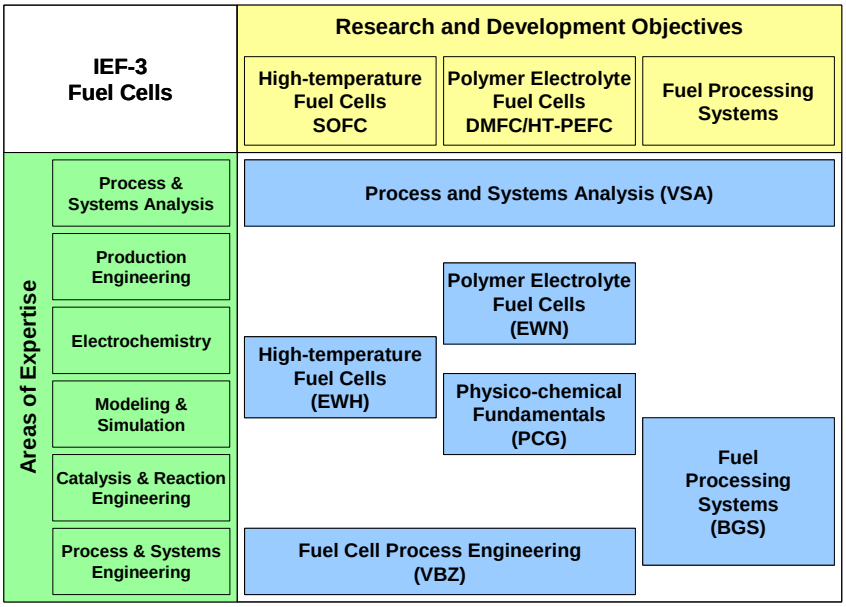
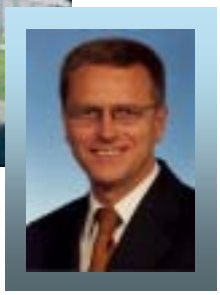


Fig. 81: Specialist departments vs. areas of expertise and R&D objectives of IEF-3

Focusing intensively on a small number of fuel cells and a restricted range of performance makes it possible to work on them from the electrochemical cells concerned up to the process engineering for the whole system with teams of supercritical size. The Institute's objectives and focal points are unlocking the synergies between the specialised disciplines and putting theory into practice in order to reach a pilot stage. Stationary, portable and mobile applications up to and including complete systems with fuel cells, together with cost aspects, are evaluated during procedural and system analysis in terms of their usability as well as from a technical, energetic and economic perspective in comparison with competing systems. This work is accompanied by fundamental physical-chemical investigations in selected fields.

The research work carried out by IEF-3 should yield groundbreaking results of social, ecological and economic relevance in comparison with other international research facilities. This aspect of the work should be achieved via fundamental research in close coordination with technical development work into relevant scientific-technical areas of expertise. International cooperation with partners from research and industry is of particular importance in this respect. The Institute is focused on implementing results from research in innovative products, procedures and processes in collaboration with industry, and is keen to make a

contribution towards bridging the gap between science and technology. Cooperation with universities, universities of applied sciences, training workshops and training centres is designed to promote opportunities for further education and training.



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With around 160 employees, Jülich represents the largest institutional fuel cell research team in Europe. Around half of its employees are involved with SOFCs, of which around 1/3 concentrate on the activities of IEF-3. For SOFCs, two further Institutes – IEF-1 and IEF-2 – are respectively responsible for manufacturing materials; and testing materials and steel research and the Central Technology Division (ZAT) for stack construction.

The other 75 or so of the 100 employees at IEF-3 divide their work equally between low-temperature fuel cells and high-temperature ceramic fuel cells. Each field has a coordinator for general themes, and these coordinators also act as initial contact persons for SOFC and with the Institutes involved.

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6.2 Overview of department competences

Polymer Electrolyte Fuel Cells

Fields of work, service offerings

Direct methanol fuel cells are developed within the department from membrane electrode assemblies to stacks and, in close cooperation with process engineering, up to systems. New membrane and catalyst materials are characterized and innovative membrane electrode assemblies are devised and manufactured from them. This work focuses on the structure-efficiency relationship of the electrode layers. Stack modelling and design combined with new analytical procedures in stack engineering and automated manufacturing methods at the pilot plant scale ensure that the gap between science and technology is bridged.

Facilities, processes, methods

Equipment

- Test stands for electrochemical investigations of DMFCs, PEFCs and stacks
- Apparatus for magnetotomographic investigations of current density distribution and single cells and stacks
- Apparatus for characterizing membranes for PEFC and DMFC
- Coating machine for continuous manufacture of MEA components
- Robot facilities for automated manufacture of cell components (die-cutting and pressing) and stacks (assembling)

High-Temperature Fuel Cells

Fields of work, service offerings

In Jülich, SOFCs are researched and developed from materials up to entire systems. This department works on electrochemistry and stack tests and modelling from cells to stacks. Cells and stacks are tested both under laboratory conditions, so that special information can be obtained, and under realistic operating conditions, also for extended periods. In-house models for simulation calculations that take account of the mass, charge and heat transport processes for cells and stacks are used and refined. The results are used to develop new concepts.

Facilities, processes, methods

Equipment

- Test stands for electrochemical investigations of solid oxide fuel cells (SOFC)
- Test stands for electrochemical investigations of solid oxide fuel cell stacks
- Apparatus for measuring permeation and diffusion

Models

- Computer models to simulate operating conditions in SOFCs (membrane, cell and stack) on the basis of the transport processes for heat, mass and charge

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Fuel Processing Systems **Process and Systems Analysis**

Fields of work, service offerings

Hydrogen for fuel cells manufactured from commercially available fuels will enable fuel cell technology to be introduced in a short space of time. The Institute is specialized in reforming middle distillates – diesel, kerosene and fuel oil. Work is carried out on the entire system, including desulphurization and CO purification. The focus is on reactors in the 3 - 50 kW power class incorporated into high-temperature PEFC or SOFC systems. Complex, realistic models, on which reactor design and construction are directly based, are supported by the use of glass models in which similarity theory is used to visualize flows.

Selecting meaningful energy systems for the future and guiding their development requires a systematic approach, used here for fuel cells. The investigations are based on observations of energy chains, benchmarks for other application and manufacturing technologies and economic comparisons. This broad experimental basis and the more extensive modelling in the Institute are used to mode entire systems and to compare them with competing technologies in realistic conditions. They are also used to identify development potential and shortcomings. Studies are also carried out for industry and confidentiality of information is assured.

Facilities, processes, methods

Equipment and models

- Test stands for investigating reactors for reforming, water-gas shift reaction, preferential oxidation and desulphurization
- Test stands for screening and investigating catalysts with regard to activity and selectivity
- Analytical devices to determine concentrations (GC, GC/MS, NDIR) of reaction gases
- Apparatus for fractional distillation of fuels
- CFD simulation program for visualizing educt mixture and reactor design

Methods

- Design engineering for fuel gas generation and energy systems with fuel cells
- Integrated evaluation methods (life cycle analyses) for energy systems

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Fuel Cells Process Engineering **Physicochemical Fundamentals**

Fields of work, service offerings

Designing and building efficient fuel cell systems requires detailed knowledge of every step of the process, from cells to complete systems. The intensive interactions between cells and the system as a whole represent a challenge in terms of development which is fully explored here. Three DMFC test plants (2 kW) and two SOFC test plants (1 kW, 20 kW) were constructed in close collaboration with Cell and Stack Engineering. The focus of the work is on developing new system concepts; testing and evaluating them; developing components in collaboration with industry; and testing and developing control and regulating concepts.

As technical development becomes more and more advanced, fundamental work on future generations of systems becomes more and more important. This department is therefore concerned with the fundamental theoretical aspects of the industry, with particular emphasis currently being placed on molecular dynamic and quantum chemical modelling of proton generation and transport in polymer fuel cells. Continuum simulations of PEFCs and DMFCs also contribute towards gaining an elementary understanding of processes at a cell level. These procedures require the use of Supercomputing facilities, available on campus in Jülich.

Facilities, processes, methods

Equipment and models

Test stands for

- high-temperature heat exchangers up to 850 °C and 200 m^3 air/h
- reformers and after-burners for SOFC stacks with a power of 5 kW
- air compressors up to 100 m^3/h and exhaust-gas-heated steam generators for SOFC plants
- process engineering investigations of DMFC stacks and system components
- CFD models to determine flow distribution in stacks
- simulation models for the design of fuel cell plants

Theory and methods

- programs and continuum models of PEFCs in 1- to 3-D
- 1-D analytical models of PEFCs
- programs and molecular dynamic simulations of H^+ transport in polymer electrolytes
- ab initio quantum mechanical and molecular dynamic investigation of electrocatalytic reactions
- simulation calculations on NIC'sJUMP supercomputer and the IEF-3 Linux cluster

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6.3 Publications, technology transfer and resources

The scientific and technical results of the work carried out at IEF-3 have been published in relevant journals and presented to interested specialist audiences at national and international conferences on the subject. Leading journals publishing peer-reviewed articles from IEF-3 are the Journal of Power Sources (2005: 6; 2006: 5), Electrochemistry Communications (2005: 3; 2006: 2), Electrochimica Acta (2005: 2, 2006: 2) and the Journal of the Electrochemical Society (2005: 2, 2006: 1). The major conferences atwhich IEF-3 was represented in 2005 were the Fuel Cell Seminar in Palm Springs, CA, USA, at which 2 articles were presented; the 3rd European Polymer Electrolyte Fuel Cell Forum in Lucerne, Switzerland (7 articles); and Solid Oxide Fuel Cells (SOFC IX) in Pennington, NJ, USA (3 articles). In 2006 IEF-3 presented 10 articles at the 7th European Solid Oxide Fuel Cell Forum in Lucerne, Switzerland, and 3 articles at the 3rd International German H_2 Energy Congress 2006 in Essen. IEF-3 also contributed individual articles by its specialized departments to numerous other specialist conferences both within Germany and abroad. In addition, 5 doctoral theses focusing on DMFC/PEFC research and development and 1 each on SOFC and feed gas generation were completed during the 2005 - 2006 report period.

Institute of Energy Research – Fuel Cells (IEF-3)			
		2005	**2006**
Publications	Peer-reviewed journals[1)]	18	20
	Books and journals	5	8
	Proceedings	26	33
	PhD theses	2	4
Technology Transfer	Ongoing projects third party funding	41	51
	Patent applications	18	16
	Patents granted	1	0
Resources[2)]	Staff (POF[3)]/third party funded)	75 (67/8)	77 (64/13)
Comments	[1)]According to ISI citation index [2)] Data in PY/a [3)]POF: Program-oriented funding		

Tab. 6: IEF-3 core data for 2005 and 2006

To ensure successful technology transfer, IEF-3 was involved in numerous R&D projects (41 in 2005 and 51 in 2006) that received funding from the European Commission, the Federal Ministries of Economics (BMWi) and of Education and Research (BMBF) and the corresponding Ministries of North Rhine-Westphalia or were financed by industry. IEF-3 was also responsible for managing and coordinating some of these projects. The numerous patent applications submitted during the report period (18 in 2005 and 16 in 2006) and patents granted (1 in 2005) were another step towards smooth technology transfer.

IEF-3 has a staff of just under 100 and is financed partially by programme-oriented funding (POF) from the Heimholtz Association of National Research Centres (HGF) and partially from third-party funds. Some of IEF-3's workforce are employed on a part-time basis, so the effective personnel capacity was 75 PY/a in 2005 and 77 PY/a in 2006.

6.4 Committee work

The competence of IEF-3 in fuel cells and hydrogen technology, which is renowned both throughout Germany and abroad, is reflected in the fact that several IEF-3 scientists are members of and collaborate with national and international committees. The work led and coordinated by Professor Stolten between 2004 and 2005 on drawing up the strategic research agenda for the Hydrogen and Fuel Cell Technology Platform (HFP) of the European Commission can be considered as an outstanding contribution made by the Institute as a whole. In addition, Professor Stolten chaired the Quality Assurance Panel that examined the implementation plan for the HFP in 2006. Another contribution of international significance is Professor Stolten's collaboration in the Global Roundtable on Climate Change (GROCC) and an expert for the current IPCC (Intergovernmental Panel on Climate Change) report. Additionally, Professor Stolten and Dr. Dohle occupy leading roles on the committees of the International Energy Agency (IEA) responsible for fuel cells. At a national level, Professor Stolten was a member of the coordination group of the Hydrogen and Fuel Cell Strategic Council, representing the German research sector, and collaborated on the creation of the National Development Plan for the 'Hydrogen and Fuel Cell Technology Development Programme'. The committee work performed by IEF-3 employees at a German and European level is set out in detail below.

Fuel Cells and Hydrogen Competence Network NRW
until 2005, Prof. Dr.-Ing. D. Stolten
Head of the Competence Network

Working Group of Electrochemical Research Facilities (AGEF e.V.)
since 2000, Prof. Dr.-Ing. D. Stolten
Member of the Executive Committee

IEA Executive Committee in the "Advanced Fuel Cells" Annex
since 2000, Prof. Dr.-Ing. D. Stolten
Head of the German Delegation

Fuel Cells and Hydrogen Working Party NRW
since 2002, Dr.-Ing. H. Dohle
Head of the Stack Design and Assembly Working Group
since 2006, Dr.-Ing. H. Dohle
Member of the Complete System Working Party

Advisory Council der Hydrogen and Fuel Cell Technology Plattform der EC
since 2003, Prof. Dr.-Ing. D. Stolten
Member

GVC Energy Process Engineering Expert Committee/VDI Society for Engineering and Chemical Engineering
since 2003, Prof. Dr.-Ing. D. Stolten
Member of the "Energy Process Engineering" Expert Committee
since 2006, Prof. Dr.-Ing. D. Stolten
Vice Chairman of the "Energy Process Engineering" Expert Committee

Masterflex AG Supervisory Board
since 2004, Prof. Dr.-Ing. D. Stolten
Vice Chairman of the Supervisory Board

BMWi 'PEFC' BERTA Working Party
since 2004, Dipl.-Ing. J. Mergel
Member of the Working Party

IEA ANNEX XVI: Collaborative Research on Polymer Electrolyte Fuel Cells
since 2004, Dipl.-Ing. J. Mergel
Member

IEA ANNEX XXI: Collaborative Research on Polymer Electrolyte Fuel Cells
since 2004, Dr.-Ing. H. Dohle
Member

EC "Strategic Research Agenda" Steering Panel
2004 - 2005, Prof. Dr.-Ing. D. Stolten
Chairman of the Panel

EC Joint Technology Initiative
since 2005, Dr.-Ing. B. Emonts
Member of the Working Party

Fuel Cell Qualification Initiative
since 2005, Dr.-Ing. B. Emonts
Member of the Executive Committee

BREZEL Expert Committee of the Association of German Engineers
since 2005, Prof. L. Blum
Member of the Expert Committee

Global Roundtable on Climate Change der Columbia University, New York
since 2006, Prof. Dr.-Ing. D. Stolten
Member

Intergovernmental Panel on Climate Change
since 2006, Prof. Dr.-Ing. D. Stolten
Expert for the current IPCC report

Strategic Council for Fuel Cells and Hydrogen
since 2006, Prof. Dr.-Ing. D. Stolten
Member and Representative of German Research Facilities in the Coordination Group

"High-Temperature PEFC" Working Party of the Strategic Council
2006, Prof. Dr.-Ing. D. Stolten
Chairman of the Working Party

DKE "Portable Fuel Cell Systems" Standardization Working Party
since 2006, Dipl.-Ing. J. Mergel
Member of the Working Party

6.5 Contributions to trade fairs and exhibitions

IEF-3 has set itself the objective of showing its capacity for innovation by demonstrating new technologies arising from the focal points of the Institute's R&D work. Alongside this, one major technology trade fair is selected per year at which to showcase the proven highlights of the year. In 2005 IEF-3 was able to present its first small vehicle, the compact DMFC-powered JuMOVe (see Fig. 82) to an interested specialist audience at E-world of Energy in Essen and at the annual HPF conference in Brussels. A fuel gas processing module and a diesel reformer and shift stages was also exhibited. Over the course of the year IEF-3 took part in seven further trade fairs and exhibitions both in Germany and beyond. Details of these fairs and exhibitions are listed below.

Fig. 82: DMFC Scooter at E-world of Energy

2005

E-world of Energy 15-17/03/2005, Essen *JuMOVe DMFC-Scooter and diesel reformer with shift stages*
HFP Annual Conference 16-17/03/2005, Brussels/Belgium *JuMOVe DMFC-scooter*
KNBW NRW Annual Meeting 8-9/04/2005, Düsseldorf *JuMOVe DMFC-scooter*
Hanover Trade Fair 11-15/04/2005, Hanover *JuMOVe DMFC-Scooter and diesel reformer with shift stages*
3rd European PEFC Forum 4-8/07/2005, Lucerne/Switzerland *JuMOVe DMFC-Scooter and diesel reformer with shif stages*
Int. Hydrogen Energy Congress & Exhibition 13-15/07/2005, Istanbul/Turkey *Portable DMFC system*
f-cell 2005 26-28/09/2005, Stuttgart *JuMOVe DMFC-Scooter and portable DMFC system*
Fuel Cell Seminar 2005 14-18/11/2005, Palm Springs/CA, USA *Machine-made membrane electrode assembly (MEA)*

One year later, IEF-3 presented a further development of the scooter, JuMOVe 2, at the Hanover Trade Fair 2006 (see Fig. 83). The reworked drive system, including a new stack, showed in the subsequent test phase that the device's lifetime had been improved twenty-fdd. as compared with the original model. The largest stand in the history of Jülich fuel cell exhibitions presented information about all aspects of fuel cell research carried out at Jülich.

Fig. 83: DMFC-Scooter JuMOVe 2 at the Hanover Trade Fair 2006: FZJ stand (left) and presentation at the joint Hydrogen & Fuel Cells stand (right)

Fig. 84: JuMOVe 2 DMFC scooter at the Energy Innovation Forum in Essen

On the occasion of the 50th anniversary of Research Centre Jülich, the Jülich Energy Sector held an anniversary event at the former Zollverein mine in Essen to which numerous VIPs from ministries, research and industry were invited. The JuMOVe 2 DMFC scooter (see Fig. 84) was presented as part of a driving event alongside exhibits from all energy-related research fields.

All this, plus IEF-3's involvement in ten further trade fairs and exhibitions, saw 2006 as a year characterized by an extensive amount of sales promotions and public relations. The exhibits presented at the different events are listed below.

2006

E-world of energy and water and 3rd Int. German Hydrogen Energy Congress 2006
14-16/02/2006, Essen
Machine-made gas diffusion electrode (GDE)

Hanover Trade Fair
24-28/04/2006, Hanover
JuMOVe 2 DMFC scooter, system components for converting diesel into hydrogen, SOFC plant model and manufacture of SOFC and DMFC components

limitless…the Aachen-Eifel region
3-5/05/2006, Düsseldorf
JuMOVe 2 DMFC scooter

Idea Park 2006 – Experience the technology of the future
20-28/05/2006, Hanover
JuMOVe DMFC scooter, SOFC components and stack model

Energy Innovation Forum
23/05/2006, Essen
JuMOVe DMFC scooter, SOFC components and stack model

Lucerne Fuel Cell Forum
3-7/07/2006, Lucerne, Switzerland
JuMOVe 2 DMFC scooter, SOFC system model and manufacture of SOFC and DMFC components

60 years of NRW
26-27/08/2006, Düsseldorf
JuMOVe and JuMOVe 2 DMFC scooters

Open Day, Research Centre Jülich
10/09/2006, Jülich
Work results and facilities for Jülich fuel cell research and technology development

Solar Energy Association Annual Conference 2006
21-22/09/2006, Berlin
"Fuel Cells" multimedia learning programme

3rd General Assembly and Exhibition
4-6/10/2006, Brussels, Belgium
JuMOVe 2 DMFC scooter, manufacture of SOFC and DMFC components

H_2Expo 2006
25-26/10/2006, Hamburg
System components for converting diesel into hydrogen

Aachen Energy Days
26-29/10/2006, Aachen
JuMOVe 2 DMFC scooter

6.6 How to get there

6.6.1 By car

Coming from Cologne take the A 4 motorway (Cologne-Aachen) and exit at Düren, then turn right towards Jülich (B 56). After approx. 10 km turn right onto the L 253 and you will see signs for the Research Centre (“Forschungszentrum”).

Coming from Aachen take the A 44 motorway (Aachen-Düsseldorf) and exit at Jülich-West. At the first roundabout turn left towards Jülich, and at the second roundabout turn right towards Düren (B 56). After approx. 5 km turn left onto the L 253, and you will see signs for the Research Centre (“Forschungszentrum”).

From Düsseldorf Airport take the A 52 motorway (towards Düsseldorf/Mönchengladbach), then the A 57 (towards Cologne) until Neuss-West, then continue on the a 46 (towards Jüchen/Grevenbroich), then the A 44 (towards Aachen). Thereafter the directions are the same as from Düsseldorf.

Coming from Düsseldorf take the A 44 motorway (Düsseldorf-Aachen). You then have two options:

1. (shorter distance but with local traffic) take the Jülich-Ost exit, turn right, then after approx. 500 m turn right onto the B55n towards Jülich. Turn left after 200 m, before the radio masts, and continue until the “Merscher Höhe” roundabout, where you turn left. Pass in front of the University of Applied Sciences’ Solar Campus, keep going straight down Brunnenstrasse, go straight over the crossroads with Römerstrasse, keep going down Wiesenstrasse, after the roundabout and the caravan dealer’s turn left and continue to the Research Centre (which will be signposted).

2. longer but faster) exit the motorway at Jülich-West. At the first roundabout turn left towards Jülich, and at the second roundabout turn right towards Düren (B 56). After approx. 5 km turn left onto the L 253, and you will see signs for the Research Centre (“Forschungszentrum”).

Fig. 85: Euregio Rheinland access map

Sat-nav systems: Please enter “Wilhelm-Johnen-Strasse” as your destination in your GPS system. This is only a few hundred metres away from the main entrance; please follow the signs. The Research Centre itself does not form part of the public road network and is therefore not recognized by sat-nav systems.

6.6.2 By plane

Cologne/Bonn airport: Either take the S13 train from the airport station to Cologne Central Station and then take the Regional Express to Düren station, or take the Regional Express to Cologne-Ehrenfeld and then take the S12 to Düren. For directions from Düren onwards, see "By train”.

Düsseldorf airport: Take the train from the airport station to Cologne Central Station and then on to Düren. Some trains are direct, although for some you will have to change at Cologne Central. For directions from Düren onwards, see "By train”.

6.6.3 By train

Take the Bundesbahn from Aachen or Cologne to Düren station, then take the local train (“Rurtalbahn”) to the “Forschungszentrum” stop. Turn right from the stop, and it is about a 20-minute walk to the main entrance of the Research Centre.

Fig. 86: Jülich Research Centre site map

Fig. 87: Institute of Energy Research – Fuel Cells (IEF-3) site map, address Helmholtz-Ring H8, Geb. 03.2

6.7 List of abbreviations

AFL	Anode Functional Layer
AK HT-PEFC	Working Party on High-Temperature Polymer Electrolyte Fuel Cells
APROS	Advanced Process Simulator
APU	Auxiliary Power Unit
ASR	Area-Specific Resistance
ATR	Autothermal Reforming
BET	Method for determining specific surface area developed by Stephan Brunauer and Emmett and Edward Teller
Bfe	Federal Technology Centre for Electrical Engineering and Information Engineering e.V.
BGS	Fuel processing systems
BIBB	Federal Institute for Vocational Education and Training
BiP	Bipolar Plate
BMBF	Federal Ministry of Education and Research
BMW	Bayrische Motorenwerke
BMWi	Federal Ministry of Economics and Technology
BSE	Back-Scattered Electron
BTL	Biomass-to-Liquid)
BZ-BNW	Fuel Cell Training Network
CE	Certification Europe
CFD	Computational Fluid Dynamics
CGO	Cerium Gadolinium Oxide: $Ce_{0.8}Gd_{0.2}O_{2-\delta}$
CH	Carbon-Hydrogen:
CNC	Computerized Numerical Control
CS	sintered cells with code letter C
DC	Direct Current
DGB	German Trade Unions Federation
DMFC	Direct Methanol Fuel Cell
DMHFC	Direct Methanol Hydrogen Fuel Cell
DSC	Differential Scanning Caloriemeter
ECAB	Electrochemical Afterburner
EDX	Energy-Dispersive X-Ray Analysis
ECE	name of a model of forklift truck manufactured by Jungheinrich
EL	Extra Light
EWH	Electrochemical converter high-temperature
EWN	Electrochemical converter low-temperature
FC	Fuel Cell
FC & H_2	Fuel Cells and Hydrogen
FH	University of Applied Sciences
FhG-ISI	Fraunhofer Institute for Systems and Innovation Research
FLUENT	company name and finite volume programme system for resolving problems in fluid mechanics
FPB	Practical Vocational Training Research Group
FUMATech	company name (Functional Membranes and System Technology

FVS	Solar Energy Association
FZJ	Research Centre Jülich
GC/MS	Gas Chromatograph/Mass Spectrometer
GDE	Gas Diffusion Electrodes
GenFC	Generic Fuel Cell Modelling Environment
GROCC	Global Roundtable on Climate Change
HFP	Hydrogen and Fuel Cell Technology Platform
HGF	Helmholtz Association of German Research Centres
HPI	Heinz Piest Institute for Trade Technology
HT-PEFC	High-Temperature Polymer Electrolyte Fuel Cells
HYBERT	Hydrogen and Fuel Cells Expert Council
IBZ	Fuel Cell Institute
ICP-OES	Inductively Coupled Plasma Optical Emission Spectrometry
IEA	International Energy Agency
IEF-1	Institute of Energy Research – Materials Synthesis and Processing
IEF-2	Institute of Energy Research – Materials Microstructure and Properties
IEF-3	Institute of Energy Research – Fuel Cells
IGCC	Integrated Gasification Combined Cycle
IHK	Chamber of Trade and Industry
IQ-BZ	Fuel Cell Qualification Initiative
IPCC	Intergovernmental Panel on Climate Change
IPHE	International Partnership for the Hydrogen Economy
i.PU	in Personal Union
IR	Infrared
ISEA	Institute for Power Electronics and Electrical Drives
ISI	Institute of Scientific Information – citation database
JET A1	light oil
JuMOVe	Juelich Methanol Operated Vehicle
KNBW-NRW	Fuel Cells and Hydrogen Competence Network North Rhine-Westphalia
KIER	Korea Institute of Energy Research
KOSTEX	Cost calculation programme
KRAKE	Evaluation programme for energy conversion chains
KvK	Maastricht Chamber of Commerce
LBSt	Ludwig-Bölkow-Systemtechnik
LPG	Liquefied Petrolium Gas
LSCF	Lanthan-Strontium-Cobalt-Eisenoxid: $La_{0.58}Sr_{0.40}Co_{0.2}Fe_{0.8}O_{3-ŭ}$
LSM	Lanthan-Strontium-Manganoxid: $La_{0.65}Sr_{0.3}MnO_3$
LuVo	Air pre-heater
MATLAB	programming language for technical calculations
MCFC	Molton Carbonate Fuel Cell
ME	Management Engineers
MEA	Membrane Electrolyte Assembly
MWA	Ministry of Economics and Labour
NTBZ	Low-temperature fuel cell
OH	Oxygen-Hydrogen

ORMS	Ohmic Resistance from Long Term Measurements
OP	Liquid Phase
OXYMEM	Project for the development of ceramic membranes for ultraclean gas generation and selective catalytic oxidation with membrane reactors
PBZ	Fuel Cell Project
PCG	Fundamental physicochemical principles
PE	Polymer Electrolyte
PEFC	Polymer Electrolyte Fuel Cell
PEMEAS	Company name
PID	Controller with proportional, integrating and differentiating transfer behaviour
POF	Programme-oriented Funding
POX	Partial Oxidation
PRO/II	Tool for stationary modelling
PROX	Preferential Oxidation
PTFE	Polytetrafluroethylene
RAG	Company name
R&D	Research and Development
RECTISOL	Linde AG process for separating CO_2 and sulphur compounds from gas streams
REM	Rape Methyl Ester
RME	Rape Methyl Ester
ROZ	Right Octane Number
SEM	Scanning Electron Microscope
SIMBA	Programme for evaluating procedures
Simulink	Software for system modelling
SNG	Substitute Natural Gas
SOFC	Solid Oxide Fuel Cell
SPS	Programmable logic controller
SRA	Strategic Research Agenda
STE	System Analysis and Technology Evaluation
TGA	Thermogravimetry Analyser
THT	Tetrahydrothiophene
TOC	Total Organic Carbon
VBZ	Fuel Cell Process Engineering
VDI	Association of German Engineers
VTT	Technical Research Centre of Finland
WBzU	Fuel Cell Education and Training Centre, Ulm
WGS	Water Gas Shift Reaction
WP	Aqueous Phase
XRD	X-Ray Defraction
XPS	X-Ray Photoelectron Spectroscopy
YSZ	Yttrium-Stabilized Zirconium
ZAT	Central Division of Technology
ZAT-MF	Central Department of Division - Mechanical Manufacturing
ZCH	Central Division of Analytical Chemistry

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1 kW DMFC fuel cell system for a forklift truck. The system has technical and economic advantages over conventional battery systems since there is no need to replace or recharge the batteries.

20 kW SOFC demonstration plant consisting of four 5 kW stacks connected in parallel. The plant is operated with natural gas and will achieve a net electrical efficiency of more than 40 %.

The fuel processing component of an HT-PE fuel cell system contains an autothermal reactor for producing hydrogen and a water-gas shift reactor to reduce the CO concentration on a 5 kW_{el} scale. The catalytic burner is simulated by a water vaporizer.

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