



IWV-3 Report 2005

Future as a challenge



Forschungszentrum Jülich GmbH
Institut für Werkstoffe und Verfahren der Energietechnik
Institut 3: Energieverfahrenstechnik

IWV-3 Report 2005

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Preface

The Institute for Energy Process Engineering (IWV-3) is one of three departments of the Institute for Materials and Processes in Energy Systems at Research Centre Jülich. The research tasks of IWV-3 are oriented, on the one hand, to the design and construction of polymer and high-temperature fuel cells and stacks for stationary, portable or mobile applications extending up to complete systems with fuel cells. On the other hand, process- and system-engineering developments comprise the provision of apparatus for fuel processing. These activities are accompanied by basic physico-chemical studies and systems analyses of energy process engineering.

The research work of IWV-3 is of social, ecological and economic relevance and aims at providing top-class results in an international comparison. This quality of work is to be achieved by basic research in close coordination with technical development in relevant scientific and technical competence fields. Special significance is attached here to international cooperations with partners from research and industry. The Institute is oriented to the transformation of research results into innovative products, methods and processes. It contributes to education and further training in cooperation with universities, universities of applied sciences, training workshops and training centres.



Jülich, August 2005

A handwritten signature in blue ink, appearing to read 'D. Stolten', with a stylized flourish at the end.

Prof. Dr.-Ing. Detlef Stolten
(Head of Institute)

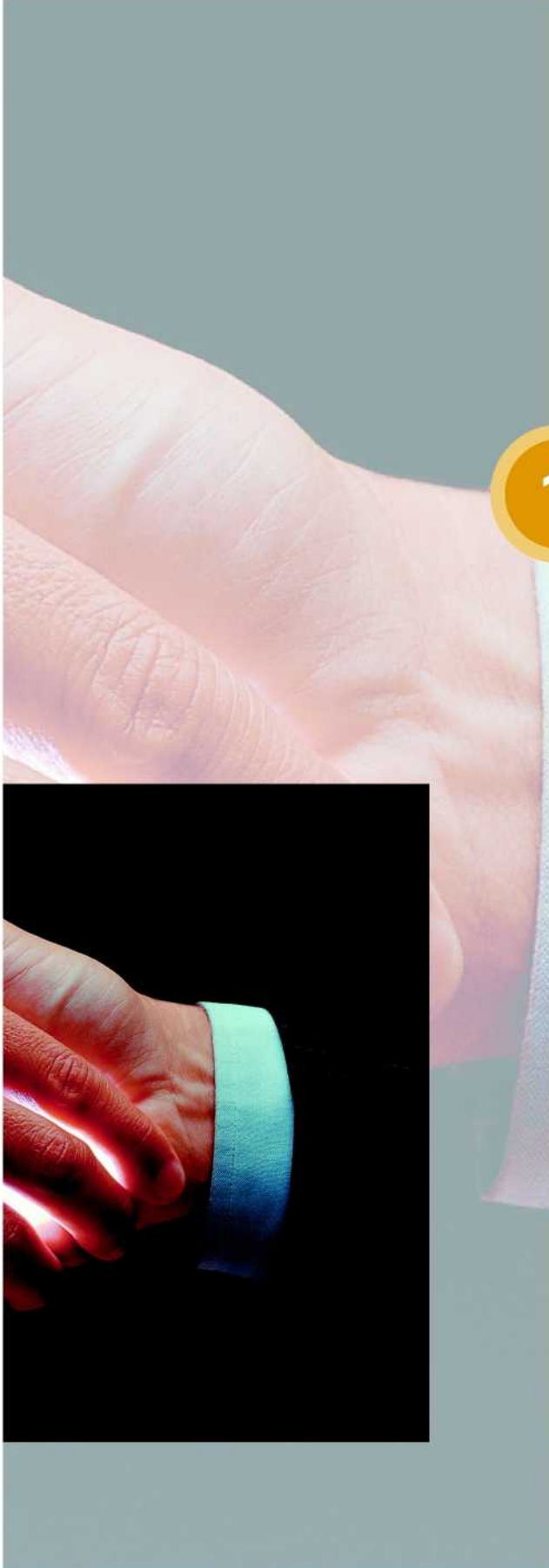
Introduction

Fuel cells can generate electric current in a wide power range with extremely low pollutant emissions and high efficiency. In conjunction with advantageous technical features such as modularity, low-noise operation, favourable behaviour at partial load and potentially long service life they are therefore an important element of our future energy economy. The close coupling to hydrogen as the energy carrier is of special economic and political relevance and, in addition to other driving boundary conditions, has internationally lead to intensive development activities in recent years.

Research and development for fuel cells are an interdisciplinary task, which in its entirety can only be accomplished with diversified expertise from the natural and engineering sciences and from application technologies in combination with an adequate infrastructure. It is therefore a typical topic of large-scale research that has been taken up by Research Centre Jülich in close cooperation with universities and other research institutes. The tasks range from the development of materials and single components through the assembly of complete fuel cell stacks up to complex application systems. Important task fields are also suitable manufacturing techniques for technical fabrication and the generation of gases suitable for fuel cells from relevant energy carriers. Research Centre Jülich concentrates its R&D activities on the solid oxide fuel cell (SOFC), the low-temperature polymer electrolyte fuel cell (PEFC) and the direct methanol fuel cell (DMFC). Solutions are to be found for a technically reliable and economically competitive use of fuel cells, which in that depth cannot be provided by industry, and contributions to next-generation technologies still far from market maturity are to be made. The innovation potential results from the development of new materials and new possibilities of basic modelling, technology-based systems analysis, experimental verification and demonstration in technical systems. The latter tools are being applied as an integrated work approach at the Institute for Materials and Processes in Energy Systems (IWV-3).

The present report wants to provide an insight into the diversified aspects of scientific and technical work at IWV-3. Examples of success demonstrate the implementation of the claim to provide top-class results of social, ecological and economic relevance in an international comparison. The Institute contributes to education and further training in cooperation with universities, universities of applied sciences and training workshops. The description of the fields of activity and of relevant work results illustrates the connection of basic research with technical development work in priority topics. It thus becomes apparent that the Institute's scientific and technical work is oriented to the transformation of research results into innovative products, methods and processes. The presentation of selected R&D projects documents the significance and the role of international cooperations with partners from research and industry. Explanations concerning priority activities and the work approach as well as the allocation of special departments, competence fields and R&D goals will round off the report.





1

Success

Examples of Success

- Development and construction of a DMFC system for installation in an electric vehicle
- Operation of the 5-kW SOFC stack

1.1 Development and construction of a DMFC system for installation in an electric vehicle

JuMOVE (**J**uelich **M**ethanol **O**perated **V**ehicle) is a demonstration vehicle with electric drive, which is supplied with electric energy by a direct methanol fuel cell (DMFC) and a buffer battery. A lithium-ion storage battery serves as the buffer battery and is only used when starting the system and at peak loads such as accelerating and driving the vehicle uphill.

With the aid of this project a possible mobile application of DMFC systems is presented. The aim of the project was, based on an already existing battery-powered electromobile (Fig. 1-1), to realize a DMFC demonstration vehicle enabling a clear increase in range.



Fig. 1-1: Electric vehicle "Arrow" from E.Mobile Technologies in the original state with fairing (left), without fairing (right)

The greatest challenge for this project was to accommodate all system components in the available space. Practical scooter application is shown in Fig. 1-2.



Fig. 1-2: Practical application of fuel cell technology in a scooter

A DMFC system consists of a DMFC stack (cell stack), the components that supply the stack with air and fuel and remove air and fuel from the stack (process engineering), and a control system which ensures, depending on the load, that the DMFC stack is sufficiently supplied

with air and fuel. Stack and process technology will be dealt with in more detail in the following.

1.1.1 Cell development and stack construction

A DMFC stack consists of series-connected single cells. A single cell contains, among other components, a membrane electrode assembly (MEA), in which electrodes are applied onto both sides of a proton-conducting membrane. These electrodes consist of porous, catalytically acting layers on which the respective electrochemical reactions take place. On both sides of the MEA there are manifold structures (flow fields) which, among other functions, have the task of homogeneously distributing the fed fuel and atmospheric oxygen across the entire MEA area. For the electrodes it has been possible to halve the quantity of expensive noble metal catalyst needed for the same power output. For the manifold structures, a low pressure loss in the range of a few mbar was realized, enabling an operation of the stack with an energy-saving blower instead of a compressor with high internal energy consumption.

Fig. 1-3 shows the 1.3-kW DMFC stack and its installation in the scooter. Apart from the 100 active cells, the stack of the dimensions 210 x 270 x 410 mm³ (HxWxL) contains an integrated system composed of methanol pump, CO₂ separator and water equalization tanks in its centre for the supply of the anode.



Fig. 1-3: 1.3-kW DMFC stack with integrated system (left), installation of the 1.3-kW stack in the scooter (right)

The CO₂ generated and collected during operation contains traces of methanol vapour. The treatment of the exhaust gas thus required has also been integrated into the stack. For exhaust gas treatment the gas mixture is recycled inside the stack and catalytically burned. This integrated design permits considerable compacting of the overall system.

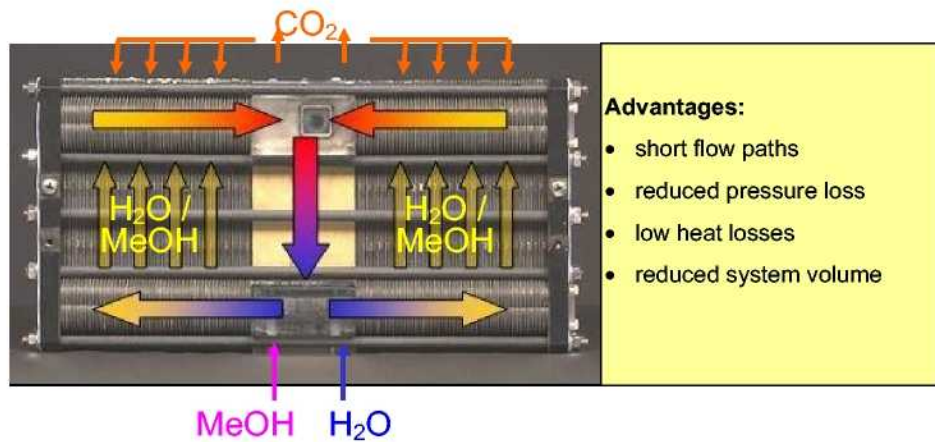


Fig. 1-4: Compact stack design by system integration

A further advantage of the system integrated into the two substacks (Fig. 1-4) is a shortening of the transport paths in the manifolds, since the methanol/water mixture is not fed in through the entire stack but from the centre to both sides.

1.1.2 Process engineering and system integration

A methanol/water solution and atmospheric oxygen are needed to operate a DMFC stack. Fig. 1-5 shows a simplified process flow diagram.

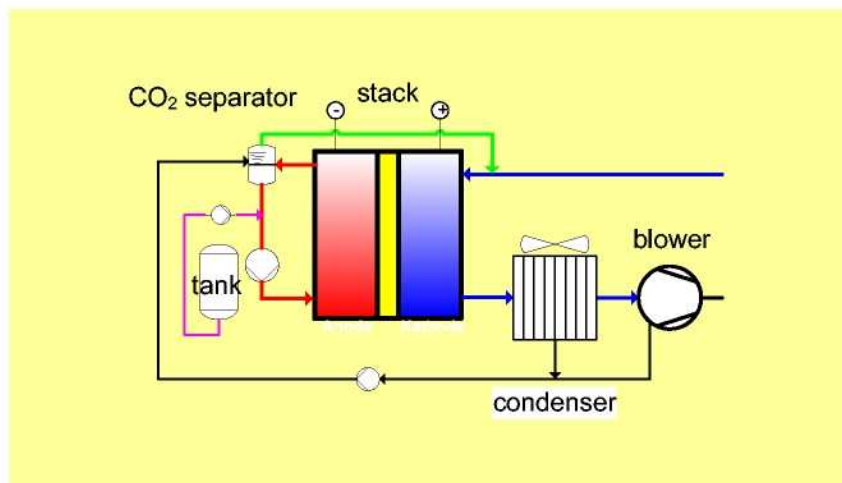


Fig. 1-5: Process flow diagram of the DMFC system of the scooter

In the anode loop (shown in red) the methanol/water solution is circulated by means of a recirculation pump. Pure methanol is stored in a tank and delivered to the anode loop by a proportioning pump as a function of fuel cell current. The water for the methanol/water solution is recovered from the cathode exhaust gas with the aid of a condenser developed for this application, interim-stored in a collecting tray underneath the stack and fed from there into the anode loop as required. The advantage of water recovery is that no extra tank has to be taken along in the system for the water required. The CO_2 produced during operation on the anode side is separated from the liquid phase in a separator integrated into the stack.

In the cathode system (shown in blue) the air is taken in by a blower through open gas channels (cathode flow fields) at the stack top (Fig. 1-6). The stack exhaust air and the water produced during chemical reaction correspondingly escape from the entire stack bottom, where a collecting tray is screwed gastight onto the stack, in which the water leaving the stack in the liquid state is collected and stored for delivery to the anode loop. The still wet exhaust air is diverted in the rear tray region and enters the specially developed, air-cooled condenser from below. The liquid water separated in the condenser also flows back into the tray.

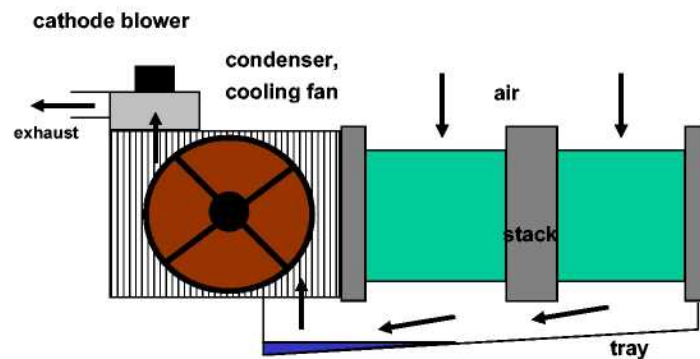


Fig. 1-6: Supply/exhaust air configuration of the scooter

The cathode blower is connected to the condenser on the intake side. The advantage of arranging the cathode blower on the intake side is better air distribution across the entire stack length and the omission of a flow distributor at the stack inlet. Heat extraction is effected by water evaporation inside the stack. The cathode air entering the stack under ambient conditions is simultaneously heated and humidified in the stack. In this way, no additional component for air humidification and no external cooler are needed.

1.1.3 Evaluation

A commercial four-wheel electric vehicle has been successfully modified in its energy supply section from lead-acid batteries to a hybrid system composed of direct methanol fuel cell and lithium-ion storage battery. The advantages of the new system are doubling the vehicle range and shortening the recharging time (refuelling and battery recharging). JuMOVE is the first and currently only mobile system worldwide based on a direct methanol fuel cell in the kW range. The greatest challenge for this project was to accommodate all system components in the available space while increasing the range.

The experience gained with direct methanol fuel cells at Research Centre Jülich in IWV-3 (Institute for Energy Process Engineering) since 1999 has been incorporated in the advancement of the electrodes for the membrane electrode assemblies and of stack and system technology. Due to close interdisciplinary cooperation of the working groups on stack construction (development of the fuel cell stack), process engineering (development of the peripherals needed for the stack, such as air and fuel supply) and MEA development (development and characterization of the electrodes), great progress was achieved in the fields of system integration, system simplification and increase in system efficiency, which decisively contributed towards realizing the JuMOVE.

1.2 Operation of a 5-kW SOFC stack

Within the framework of the European project "ProCon", an SOFC (solid oxide fuel cell) stack was constructed according to the F-design for planar cells developed at Research Centre Jülich. The interconnect plates with integrated manifold for parallel flow against the cell area by the reactants in the counterflow direction were manufactured from ferritic steel JS-3 (CroFer22APU). The 60 cells of the "anode substrate" type were produced in a size of 20 x 20 cm². The resulting effective electrode area is 361 cm². Glass ceramics was used to seal the cells from the metallic frame and the planes from each other.

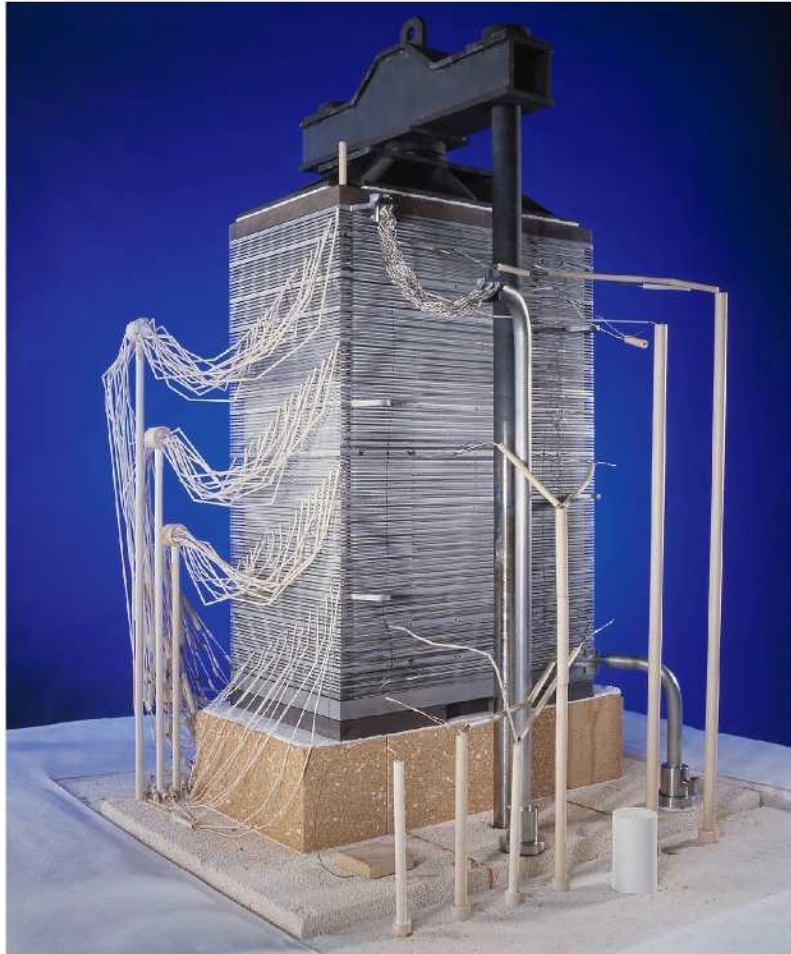


Fig. 1-7: The 5-kW ProCon SOFC stack with 60 anode substrate cells (20x20 cm²)

Fig. 1-7 shows the assembled stack prior to operation in the test rig. The stack was put into operation shortly after Easter 2004. After the reduction phase, all the 60 cells showed an

open circuit voltage (OCV) of $1,067 \pm 3$ mV/cell under a fuel gas mixture of 90 % H_2 and 10 % H_2O .

During the first current loading of the stack, the current was increased in steps every second minute. The stack was thus not allowed to reach a stable temperature distribution. It was determined beforehand that a maximum temperature of 800 °C must not be exceeded in the stack in order to be able to guarantee a service life of at least 2,000 hours. This also meant, however, that the maximum power of the stack could not be achieved. The limit temperature was already reached at a current intensity of 267 A (0.74 A/cm^2) and a stack voltage of 49.9 V (0.831 V/cell). The highest power run at this operating point with 90 % H_2 /10 % H_2O as the fuel gas was 13.3 kW (0.614 W/cm^2).

The fuel gas was changed in the course of operation into a mixture simulating 6 % partially prereformed methane (6 % H_2 , 31 % CH_4 , 63 % H_2O). With this fuel gas a highest power of 11.9 kW (0.55 W/cm^2) was reached at a current intensity of 266 A (0.73 A/cm^2) and a stack voltage of 44.6 V (0.743 V/cell).

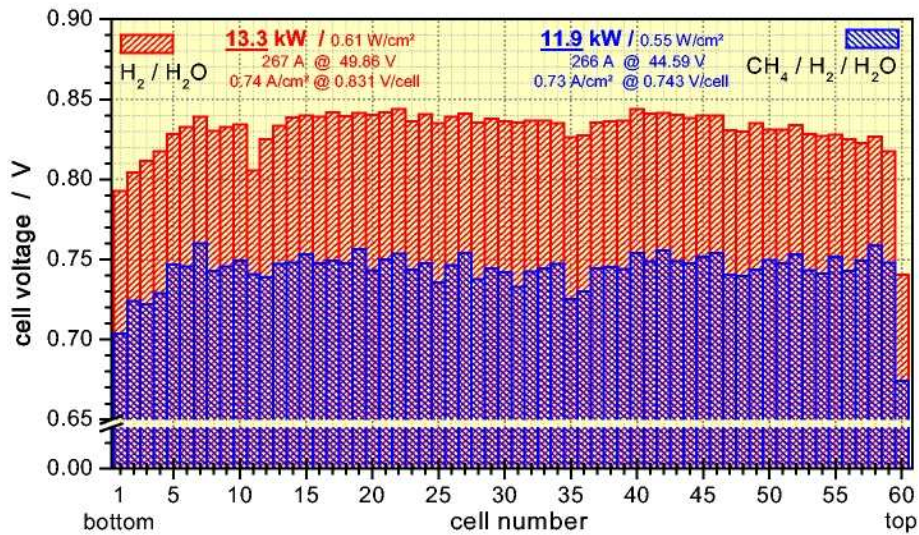


Fig. 1-8: Cell voltages during operation with hydrogen and prereformed methane at highest power point

Fig. 1-8 shows the voltages of the individual planes at the highest power point with hydrogen and prereformed methane as the fuel gas. The voltages are very uniform across the entire stack. Only plane 60 right at the top in the stack clearly deviates from the average value. This could be due to significant cooling by the top plate. The increase of the voltages in the first five planes down in the stack may also be explained by increasing temperatures.

The stack was then loaded for a prolonged time with the simulated, partially prereformed methane as the fuel gas at a constant current of 125 A and a current density of 0.35 A/cm^2 .

Fig. 1-9 shows the variation in stack voltage and the resulting stack power over time. The power was still about 6.0 kW (0.277 W/cm²; 48.1 V, 0.802 V/cell) after more than 500 hours of operation at constant current. The maximum temperature in the stack, monitored by a total of 36 thermocouples, was kept below 720 °C by controlling the furnace temperature and the air flow rate. The minimum temperature in the stack was about 614 °C.

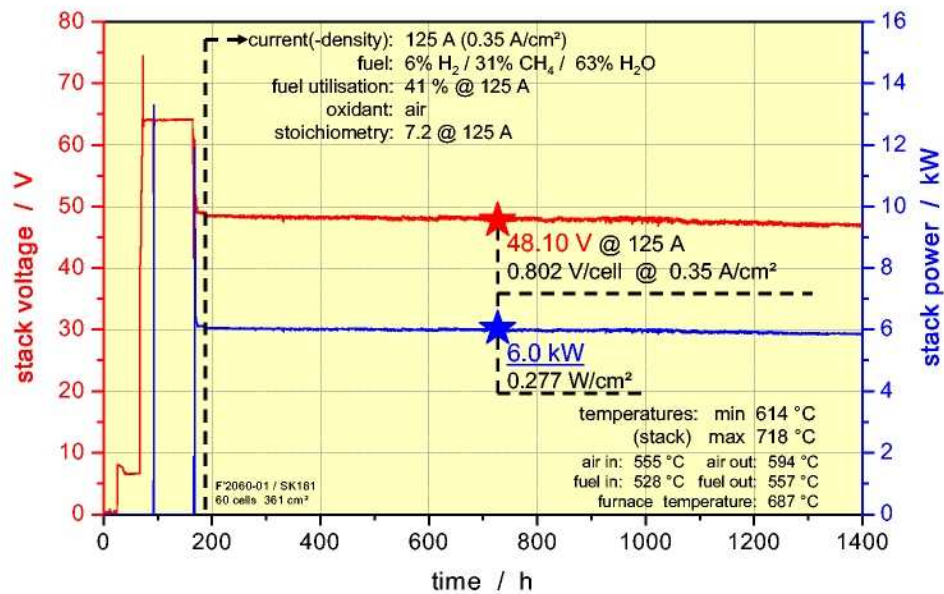


Fig. 1-9: Variation in stack voltage and stack power as a function of operating time at 125 A load with prereformed methane

The 36 thermocouples were distributed to five planes and placed in interconnect plates specifically provided for this purpose. It was thus possible to measure relevant temperatures in several planes both in the region of the active cell area and in the air and fuel gas manifolds. Fig. 1-10 shows the temperature distribution in plane 31 right at the centre of the stack during operation at the time marked in Fig. 1-9. The distribution was determined by interpolation between the 11 measured values. Effects at the edges were not taken into account in the interpolation. The cooling caused by endothermic methane/steam reforming can be clearly seen at the fuel gas inlet on the right.

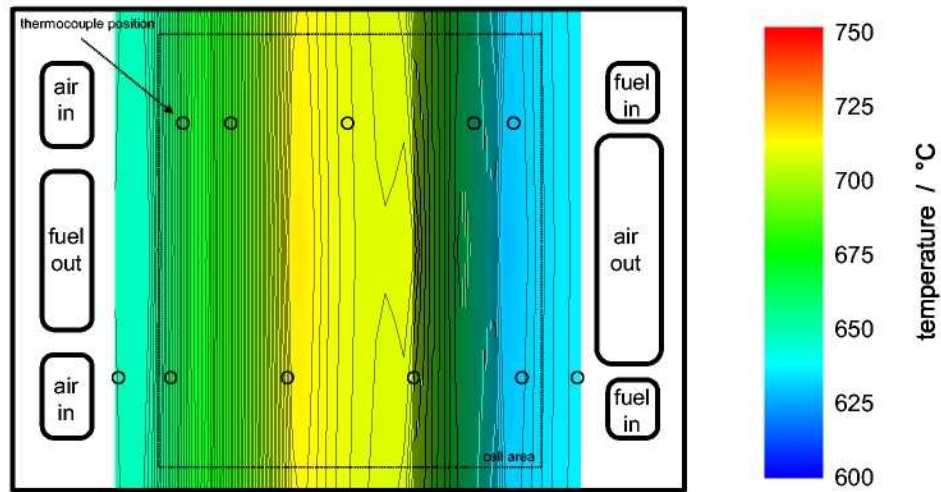


Fig. 1-10: Temperature distribution in plane 31 at 125 A with prereformed methane. Visualization of the interconnect plate contour, cell area and positions of the thermocouples

Fig. 1-11 shows the temperature distribution across the height of the stack at the same point in time. The cooling due to reforming is also clearly visible here. The cooling on the left is caused by the large volume of cold inflowing air.

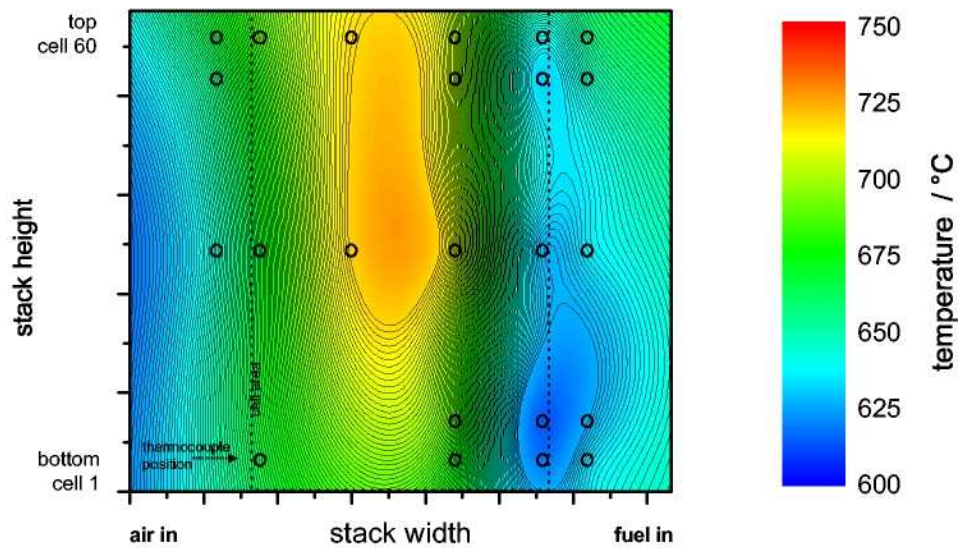


Fig. 1-11: Temperature distribution across the stack height at 125 A with prereformed methane. Visualization of the cell area and positions of the thermocouples

In the upper region of the stack, air and fuel gas are warmed on their way through the supply ducts of the stack. The "hottest" part is apparently in the middle of the upper half of the stack. The interpolation indicates that the maximum temperature will be higher than that measured by thermocouples. However, the difference between calculated and measured values is not more than ten kelvin.

It can be seen in Fig. 1-9 that the stack loses power after prolonged operating time. The ageing rate for the first 800 hours of operation is about 1 to 2 % in 1,000 hours. The ageing rate then increases to 5 to 6 % in 1,000 hours. This ageing is mainly caused by only a few "poorer" planes that were statistically distributed across the stack.

After inadvertent cooling caused by an interruption in electricity supply due to a stroke of lightning into the supply grid, the operation of the stack was resumed without apparent performance degradation. The stack apparently suffered damage which manifested itself after another 150 hours of operation. Due to a leakage, the temperature at the fuel gas outlet rose above 950 °C. For this reason, the experiment had to be discontinued prematurely but finally after a total of 1,450 hours of operation.

In spite of this premature discontinuation, a further success of Jülich SOFC development was recorded with the 60-plane stack. Even its predecessor, a 40-plane stack which had shown similarly good power values (9.2 kW / 0.63 W/cm² with hydrogen), aroused worldwide interest in Jülich stack technology. Both test results promoted the establishment of international cooperations with research institutions in Korea and Finland.

In November 2004, a 40-plane stack of the same design as the 60-plane stack was put into operation at the Korean Institute for Energy Research (KIER, Daejeon, Korea). This stack, too, confirmed the good performance efficiency of Jülich stack technology. A power output of 8.05 kW (32.2 V / 250 A) was achieved with hydrogen. A special feature was that the stack was assembled on site and put into operation by Jülich staff, which is globally unique for planar SOFC stacks of this power class. In preparation for the test with a 50-plane stack planned for 2005, a 10-plane stack was put into operation at VTT (Espoo, Finland). This stack was also assembled on site and put into operation by Jülich staff.





2

Education

Education and Training

- Fuel cell training and demonstration centre
- Members of staff teaching at universities

2.1 Fuel cell training and demonstration centre

The broad-based market introduction of fuel cell technology requires the creation of opportunities for further training and qualification in industry, the craft trades and education. For this purpose, tools such as training courses, training materials and training equipment are to be developed, tested and made available. A relevant project handled by the Institute for Materials and Processes in Energy Systems (IWV-3) of Research Centre Jülich was funded by the Federal Ministry of Economics and Labour and the Ministry of Economics and Labour of the federal state of North Rhine-Westphalia and supported by subcontractors with special competence. The following results were obtained during a project period of two and a half years.

2.1.1 Creation of an Internet portal "Fuel Cell Education Network"

On 29 May 2002, Fuel Cell Education and Training Centre Ulm (WBzU), Research Centre Jülich (FZJ), Heinz Piest Institute for Craft Technology (HPI) at the University of Hanover, Fraunhofer Institute for Systems and Innovation Research (FhG-ISI) and Ludwig Bölkow System Technology (LBSt) founded the Fuel Cell Education Network (BZ-BNW). In this way, an information platform in the field of fuel cell education was created providing basic information and training documents, presenting training courses, combining activities, advising and finding partners and supporting projects.



The platform is presented on the Internet at www.bz-bildung.de. The portal provides the following information:

- About us: presentation of BZ-BNW, of the operators and the advisory council; tasks and objectives of BZ-BNW; master plan for education and further training; form for contacting; imprint
- Fundamentals: function and design of a fuel cell; fuel cell types, fuel cell applications; glossary; literature references on fuel cells and hydrogen
- Who is Who: brief description of eleven project activities and working groups on the topic of "fuel cell education"
- Teaching material: pdf files of lectures by the WBzU and FZJ fuel cell education actors; pdf files of lectures by fuel-cell-related utilities and by equipment and vehicle manufacturers
- Events: offers by the operators of BZ-BNW

An envisaged area designated "marketplace" will in future enable contributors and interested parties to join forces concerning "fuel cell education".

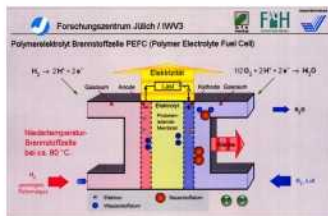
2.1.2 Development and provision of teaching modules

Based on the documents available at the Institute for Materials and Processes in Energy Systems (IWV-3) of FZJ on the topic of "fuel cells" from various lecture manuscripts (higher education level), papers, reports and publications, multimedia draft learning units and presenter training units have been developed.

2.1.2.1 Multimedia learning units

The multimedia learning units processed by the Federal Technology Centre for Electrical Engineering and Information Technology Oldenburg (bfe) consist of three thematic blocks, each comprising topic-related task blocks. The learning programme on fuel cells is part of a learning programme series of bfe-Oldenburg on the topic of electrical engineering. The learning programmes are suitable for both beginners and advanced learners wishing to learn and refresh the theoretical fundamentals of electrical engineering. The learning programmes are also suited for educational and initial training and for qualification in the electrical occupations. The learning units established are divided into the following sections:

1. Introduction and overview (16 thematic presentations, 17 tasks)
2. Physico-chemical fundamentals (24 thematic presentations, 36 tasks)
3. Fuel cell types (14 thematic presentations, 23 tasks)



A CD-ROM on "fuel cells" can be obtained from bfe Oldenburg by fax.: 0441-34092-129; e-mail: info@bfe.de or online shop: www.bfe.de at a price of € 45.50. An animated version is available at www.bfe.de/lernprogramme/v2/bsz/. All learning contents are communicated by spoken texts. Memory aids, important formulae, summaries and tasks are additionally shown as video text. Numerous animations, videos and interactions are intended to contribute towards increasing the learning effect. Knowledge queries are made during the communication of material, and the programme responds by corresponding feedback to any answers by the learner during a task.

2.1.2.2 Presenter training units

The presenter training units were jointly prepared by FZJ, Aachen University of Applied Sciences, Division Jülich (FHJ), and the University of Dortmund (UDO) as draft schemes and post-edited by bfe within three thematic and task blocks in the form of PowerPoint files suitable for presentation:

4. Fuel cell systems (105 thematic presentations)
5. Energy carriers for fuel cells (41 thematic presentations)
6. Fuel cell applications (79 thematic presentations)

Within the "Fuel cell systems" block FHJ contributed its expertise to the chapter "Integration into domestic installations" and UDO to the chapter "Power electronics". The systematically structured training units allow high variability in the adaptation to thematic and temporal presentation schemes.



Another important part of the training documents comprises procedural descriptions concerning the operation, maintenance and troubleshooting of the training equipment described in the next chapter. Participants in practical training courses are familiarized with the basic functions, special operating states, malfunctions and inspection measures by stepwise description. Written instructions and the equipment installed are designed such that it is possible to carry out measures for startup and shutdown, for the initiation of load cycling, for debugging and maintenance on one's own.

2.1.3 Setup of fuel cell systems of practical relevance in the demonstration centre

The demonstration centre is located in an experimental hall of FZJ on an area of more than hundred square metres. On this area, various facilities of practical relevance are provided that permit the operation of single cells, fuel cell stacks and systems as well as peripheral components. The membrane electrode assemblies (MEAs) needed for the DMFC and PEFC are manufactured on a desk coater. Within the framework of training courses, the manufacturing processes and special features of this core component can be explained with the aid of this desk coater.

2.1.3.1 2-kW direct methanol fuel cell system

The transportable module was set up using a DMFC stack from IWV-3 rated at 2.5 kW as well as purchased and in-house peripherals, tested for functioning and put into operation. A CE certification was successfully performed and the DMFC stack subjected to a long-term test. The open and well accessible module design is particularly suited for system tests and system optimizations. Apart from determining the operating characteristics of the DMFC stack, blower and humidifier/afterburner unit, different operating regimes can be implemented by varying the reaction and humidification water cycle, the purge rates and the methanol/water ratio, and inspection work under guidance can be carried out by training participants. The technical description of the system and core components can be seen from the detailed report on the fuel cell demonstrators of IWV-3.



2.1.3.2 Solid oxide fuel cell (SOFC) test rigs



Two stationary furnaces allow the operation of SOFC short stacks and single cells of the same type under typical solid oxide fuel cell conditions at temperatures between 700 and 1,000 °C with hydrogen or methane (internal reforming) as the fuel gas. Besides recording current/voltage characteristics of the single cell or short stack used, the facility permits fuel gas changes and temperature cycling as well as continuous operation. The operating instructions required for the

respective regime can be seen from procedural descriptions concerning operation, maintenance and troubleshooting.

2.1.3.3 Test facilities for components

The test facilities for fuel cell system components consist of three independent plant parts designed for the stand-alone operation of prereformers, afterburners and heat exchangers of an SOFC system of the 5-kW class. Extensive instrumentation and measuring techniques permit the implementation and evaluation of different operational situations as a function of fuel gas composition, operating temperature and operating pressure as well as air ratio. Applicable measurement programmes enable systematic training with respect to the operating performance of the core components necessary for an SOFC system. The operating instructions necessary for trouble-free startup, operation and shutdown can be seen from the pertinent operating manual.



2.1.3.4 5.6-kW polymer electrolyte fuel cell (PEFC) system



The stationary system has a PEFC stack from Siemens AG rated at 5.6 kW and is operated with hydrogen and oxygen from the supply facility of the experimental hall. The prototype system is integrated into a 10-inch rack and provides the necessary access for maintenance and troubleshooting due to the clear arrangement of the individual system components. Recording the current/voltage characteristics of the PEFC stack as a function of different operating parameters (stack

temperature, air ratio) is envisaged as a practical training course. In addition, different operational situations can be implemented and described under guidance. The description of possible test and operating procedures can be seen from the procedural descriptions concerning operation, maintenance and troubleshooting.

2.1.4 Organization and implementation of multiplier training events

During the past project period, training events for multipliers from industry, the craft trades and education with annually changing thematic priorities were organized and implemented together with the consortium of BZ-BNW. As part of the organization activities, more than hundred target institutions and groups from the HPI contact database were addressed and informed about the training events. Moreover, flyers announcing the training events were distributed at various exhibitions, trade fairs and conferences. Furthermore, the training events were announced via the Internet portals of BZ-BNW, FZJ and WBzU. In addition, relevant networks, associations, initiatives and editors of e-mail newsletters were supplied with the corresponding announcements. For the seminars held, accompanying documents were produced that could be handed over to each participant. The corresponding data were made available to the public as a "download" at the Internet portal of BZ-BNW.

**Praxisseminar:
Kraft-Wärme-Kopplung
mit Brennstoffzellen**

- eine Seminarreihe des
Bildungsnetzwerk Brennstoffzelle -

Termine:
Jülich: 05.-06. Mai 2004
Osnabrück: 15.-16. September 2004
Ulm: 24.-25. November 2004



Bz
Bildung
WBzU HPI

Eine Veranstaltung der Partnerinstitutionen:
Forschungszentrum Jülich GmbH (FZJ)
Heinz-Piast-Institut für Handwerkstechnik (HPI)
Weiterbildungszentrum Brennstoffzelle Ulm e.V. (WBzU)
Mitveranstaltet durch den
Deutschen Handwerkskammertag DHKT

- Training events performed in 2002:
10 – 11 June 2002: two-day fuel cell seminar at FZJ in Jülich with 24 participants from the education, guild and association sector;
30 September 2002: one-day fuel cell seminar at the Centre of Solar Energy and Hydrogen Research (ZSW) in Ulm with 22 participants from the education, guild and association sector
- Training events performed in 2003:
23 September 2003: one-day training on "Fuel cells – technology and applications" at WBzU in Ulm for 20 participants from the education, chamber and guild sector and from the commercial planning sector;
27 November 2003: one-day training on "Fuel cells – technology and applications" at FZJ in Jülich with 15 participants from the Hesse Professional Association for Electrical Engineering and Information Technology Crafts
- Training events performed in 2004:
15 – 16 September 2004: two-day practical seminar on "Combined heat and power generation with fuel cells" in the BHKW Training Centre at the Osnabrück Chamber of Handicrafts with 18 participants from the education, chamber and guild sector and from the commercial planning sector

Furthermore, IWV-3 sees its task in illustrating professions and fields of work in technical fuel cell development and introducing interested schoolchildren to the world of fuel cells. Topic- and profession-related information as well as four project activities were offered to pupils of appropriate age within the framework of a one-week job experience placement.

- Job experience for pupils organized in 2004:
11 – 15 October 2004: one-week job experience "Around the world of fuel cells" at FZJ in Jülich with 11 participants from secondary and grammar schools in the districts of Düren and Aachen

2.1.5 Setting up a project advisory council

A project advisory council was set up to accompany, evaluate, focus and comment on the Jülich project work. The advisory council is composed of the following representatives:

- Prof. Dr. Jürgen Garche, ZSW Ulm
- Dipl.-Ing. Wolfgang Koschorke, HPI Hannover
- Dipl.-Bw. Simon Boris Estermann, ZVEH Frankfurt
- Dr. Lothar Vahling, HWK Düsseldorf
- Dipl.-Kfm Dipl.-Ing. Peter Panzer, HWK Cologne
- Dipl.-Kfm. Guglielmo Menon, HWK Aachen

During the project period, two meetings were held at which the advisory council was informed about the project goals and project progress. The council's advice was fully taken into consideration in handling the project tasks.

2.1.6 Drawing up a master plan on "Fuel cell education and training"

The ten-page report (as of 24.02.2004) was elaborated by the BZ-BNW consortium as an education and training plan to support the market introduction of fuel cells, and it presents in its introduction the current projects and actors concerned with the above topic. An analysis then defines various target groups such as craft, industry, universities, schools and the public, the estimated spans of time as well as the education and training contents for the different phases on the road to market introduction. Furthermore, estimations are made concerning the number of persons employed in the fuel cell area in future. Concrete implementation, i.e. the transfer of knowledge from present knowledge providers through mediators (multipliers) to users is proposed to take place on two transfer levels. On level 1, knowledge is transferred from the provider, at present essentially research institutions, universities and manufacturers of fuel cell systems, to the mediator (education and training establishments) of the individual target groups (train the trainer). On level 2, knowledge is transferred from the mediator of a regional education and training establishment to the users of the respective target group. In order to estimate the arising demand for qualification, FhG-ISI and LBSt provided an excel-based computer model, with the aid of which the personnel requirements are determined as a function of predicted sales figures, times of market introduction, perceptibility and establishment as well as progress of technical maturity. Model calculations were generated, but the incorporation of the results into the master plan is still pending.

2.1.7 Support for and incorporation of training centres

As part of the training activities, cooperation with training centres and other institutions from the sector of craft trades, industry and education took place in many ways. Thus, in particular, the representatives of the regional Aachen, Cologne and Düsseldorf Chambers of Handicrafts were incorporated as members of the project advisory council into the project control process. Moreover, various coordinating and informative discussions as well as project presentations took place at the following institutions:

- Chambers of Handicrafts: Aachen, Düsseldorf, Cologne
- Chambers of Industry and Commerce: Aachen, Duisburg
- Training establishments: Aachen, Aalen, Bonn, Bottrop, Osnabrück, Potsdam, Ulm, Wuppertal
- Associations: Frankfurt a.M.
- Universities: Aachen, Bremen, Dortmund, Düsseldorf, Hanover, Stuttgart
- Universities of applied sciences: Aachen/Jülich, Cologne
- Technology centres: Kaisersesch, Oldenburg, Werl

The Technology and Founder Centre Region Kaisersesch (TGZ) requested FZJ to join in marketing and qualification measures on the topic of "Fuel Cells". Relevant training events are planned for 2005 and are being prepared by TGZ.

As part of the project activities, IWV-3 has also competently contributed to the acquisition of R&D results from other project consortia.

2.2 Members of staff teaching at universities

According to the IWV-3 guiding principle, scientists from the Institute also make contributions to teaching at universities in addition to their research and development work.

2.2.1 University teachers with an appointment

In fully exercising the "Jülich Model", Prof. Dr.-Ing. Stolten has accepted the appointment by RWTH Aachen University, where he heads the chair of fuel cells. His teaching activities comprise lectures and exercises for advanced students and the supervision of term papers, diploma dissertations and PhD theses.

Aachen University of Applied Sciences (FH Aachen), Division Jülich, has appointed Jülich scientists as lecturers in the field of knowledge transfer within the framework of a cooperation with Research Centre Jülich. For the engineering and Master courses offered by the department of Environmental Engineering and Energy Technology, Prof. Blum gives the lecture "Fuel Cells – The Future for Dispersed Power Supply!?" and Prof. Dr. Höhleln the lecture "Potentials of New Energy Carriers and Power Trains for Road Traffic". All lectures for the Master courses are given in English.

Name	Topic	Type/Scope Semester		University
Prof. L. Blum	Brennstoffzellen – Die Zukunft der dezentralen Energieversorgung!?	V/2	WS	FH Aachen Div. Jülich
	Fuel Cells – The Future for Dispersed Power Supply!?	V/2	SS	
Prof. Dr. B. Höhleln	Neue Energieträger und Antriebe für den Straßenverkehr	V/2	SS/WS	FH Aachen Div. Jülich
	Potentials of New Energy Carriers and Power Trains for Road Traffic	V/2	SS	
Prof. Dr. D. Stolten	Grundlagen und Technik der Brennstoffzellen	V/2	WS	RWTH Aachen

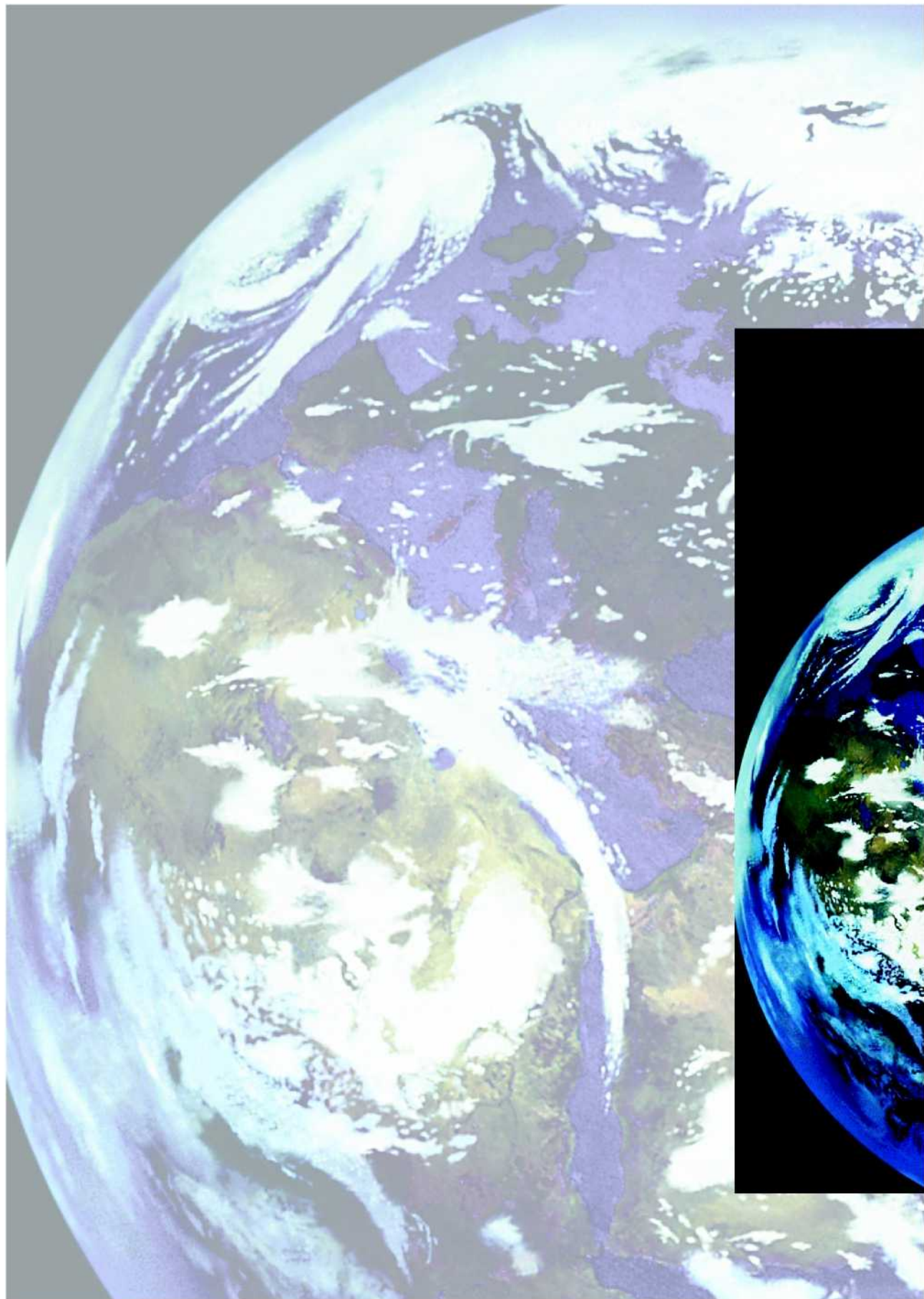
Tab. 2-1: Appointed university teachers and their teaching courses

2.2.2 University teachers with teaching assignment

In addition, there are contractual cooperation agreements with the Aachen University of Applied Sciences, Division Jülich, for cooperation in training young scientists and technicians. In this context, four staff members of IWV-3 are also active as assistant lecturers. Three further teaching courses are held by IWV-3 staff at RWTH, Ruhr University Bochum and the University of Ulm. These teaching activities also comprise the supervision of term papers, diploma dissertations and PhD theses in addition to lectures, exercises and practical training.

Name	Topic	Type/Scope Semester		University
Dr. H. Dohle	Grundlagen und Technik der Brennstoffzellen	Ü/2	WS	RWTH Aachen
Dr. B. Emonts	Der Wasserstoff und seine Umwandlung über den Elektrolyse- und Brennstoffzellenprozeß	V/2	WS	FH Aachen Div. Jülich
	Elektrolyseur und Brennstoffzelle	P/4	SS	FH Aachen Div. Jülich
Th. Grube	Basics and Applications of Chemical Reaction Theory – Simulation of Dynamic Processes in Energy Systems with Matlab/Simulink	Ü/2	WS	FH Aachen Div. Jülich
Dr. R. Peters	Basics and Applications of Chemical Reaction Theory – Simulation of Dynamic Processes in Energy Systems with Matlab/Simulink	V/2	WS	FH Aachen Div. Jülich
	Brennstoffzellen–Anwendungen	V/2	WS	Ruhr University Bochum
Prof. Dr. E. Spohr	Theoretische und Computerchemie	V/2	WS/SS	University Ulm
Dr. K. Wippermann	Brennstoffzellen – eine Zukunftstechnologie?	V/2	SS	FH Aachen Div. Jülich

Tab. 2-2: Assistant lecturers and their teaching courses



Reports

Scientific and Technical Reports

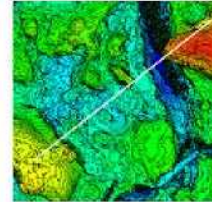
- Focus on polymer electrolyte fuel cells
- Focus on solid oxide fuel cells
- Fuel processing
- Fuel cell process engineering
- Physico-chemical fundamentals
- Process and systems analysis

3.1 Focus on polymer electrolyte fuel cells

3.1.1 Objectives and fields of activity

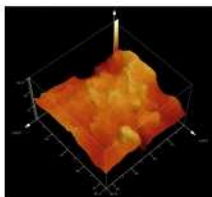
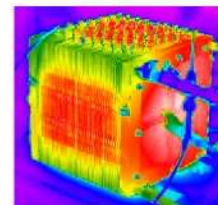
Against the background that the use of direct methanol fuel cells (DMFCs) for small applications appears interesting in the short to medium term, IWV-3 has placed clear emphasis on their exploration and development. Work ranges from modelling DMFC cells and stacks, investigating the electrochemical reactions on optimized electrode structures, characterizing new membranes, optimizing cell components, selecting and developing suitable manufacturing methods for composite structures of the membrane electrode assembly (MEA) and characterizing MEAs up to cell and stack development.

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 for 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 noble metal quantity required.



Work for the further development of manufacturing processes for components of the membrane electrode assembly concentrates on the machine fabrication of catalyst layers with the focus on developing suitable catalyst pastes for applying the catalyst layer on diffusion layers by the knife over roll technique. In this connection, it was possible to transfer the coating process from the laboratory to the pilot scale.

In the priority area of stack development, work concentrates on a further increase in power density while maintaining the functionality and reliability of PEFC and DMFC stacks. An important aspect is the selection of materials for bipolar and collector plates. Of particular importance are materials that do not interact with the cell components, maintain their functionality over a long time and can be cheaply produced.



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.

3.1.2 Important work results

A particular priority at the time of reporting is work on the direct methanol fuel cell (DMFC) and the polymer electrolyte fuel cell (PEFC). Both development lines have been successfully implemented under the project "Development of compact 5-kW fuel cell stacks with polymer electrolyte membranes (EKOPEC)" funded by the Ministry for Science and Research.

3.1.2.1 Development and construction of DMFC stacks

Based on a system of the 2-kW class (Fig. 3-1) realized successfully in 2002, work in the year under review related, in particular, to the miniaturization of the system and the further improvement of the stack. Two DMFC systems were designed and constructed in the period under review:



- Portable DMFC system as a further development of the 2-kW module created in 2002. Possible applications of the 1-kW system are in the field of back-up power and decentralized electricity production for the private sector and for fire brigades and THW (technical emergency services). In comparison to the 2002 system, especially the power density has been significantly increased.
- DMFC scooter as a demonstrator for a possible mobile application. A commercial four-wheel electric vehicle is being modified in its energy supply section from lead-acid batteries to a hybrid system composed of DMFC system and Li-ion storage battery. The advantages of the new system are the greater range and the shorter supply time (refuelling and battery recharging).

Fig. 3-1: DMFC-system

The core of the DMFC systems is the fuel cell stack. Like its predecessor from 2002, this advanced stack is also provided with exclusively planar components that can, in principle, be manufactured using mass production techniques such as punching or cutting. Instead of a monopolar design based on metallic current collector plates, a bipolar design with bipolar plates of expanded graphite was selected. Apart from its low density of 1 g/cm³, expanded graphite has the advantage over metallic materials that no additional coating for corrosion protection or contact improvement is required. Expanded graphite is an established industrial material and is normally used as flat packing. The stack is designed so that the sealing properties of the material are utilized and thus no additional seals are required.

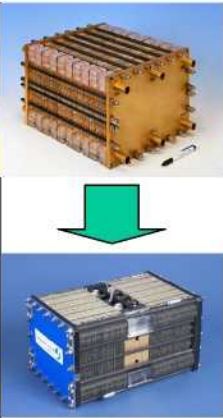
Fig. 3-2 shows the 1.3-kW DMFC stack and its installation in the scooter. The stack consists of a total of 100 single cells with a cell area of 310 cm² each. The dimensions of the stack with integrated subsystem are 210 x 270 x 410 mm³ (HxWxL). The stack length comprises

80 mm in total for the subsystem. The thickness of the end plates contributes 15 mm each to the total length.



Fig. 3-2: 1.3-kW DMFC stack with integrated system (left), installation of the 1.3-kW stack in the scooter (right)

Tab. 3-1 illustrates the progress in the development of stack technology by means of figures. An essential step is the reduction in catalyst requirements for the membrane electrode assembly, i.e. anode and cathode, from 8 mg/cm² to half that quantity. The values specified are the sum of anodic and cathodic loading.

Pt/PtRu- requirement mg/cm ² /cell	Pressure loss mbar	air ratio λ	power density W/kg	
8	300	10-20	28	
optimized electrodes	optimized flow distributors	optimized cathodes	material: expanded graphit	
4	2	4-8	90	

Tab. 3-1: Characteristics of the optimized stack with bipolar plates of graphite (bottom) in comparison to the 2002 predecessor stack with metallic current collectors (top)

Whereas the electrodes of the 2002 stack had been manufactured by hand, the electrodes for the present stack were completely manufactured using an automated desk coater. The resulting reproducibility of the layers is one reason for halving the catalyst requirements in addition to optimizing the layer composition. With the aid of a system for droplet removal based on capillary forces, the problem of droplet accumulation on the cathode has been solved. This prevents the clogging of channels due to droplet accumulation in the outlet

region, so that a better – because more homogeneous – supply of the cathode with air is achieved. By optimizing the flow distributors, especially by omitting tubular flow cross-sections in the stack involving pressure losses, it has been possible to reduce the pressure loss from 300 mbar to only 2 mbar.

The principle of air distribution can be seen from Fig. 3-3 and was successfully tested in advance using a 10-cell short stack. The necessary reaction air is fed in across the entire available width of the stack with the aid of a manifold, whereas internal flow distributors causing pressure losses were still needed for the previous stack. The total liquid water arising on the cathode is passed downwards inside the cathodic flow distributor and is collected in a tray positioned underneath the stack. This type of air supply is maintained in the portable system, whereas in the DMFC scooter a suction-type manifold is arranged below the stack for reasons of space. The collecting tray moreover performs the function of a flow collector for the product water. Since the pressure loss in the stack is only a few mbar, the associated decrease in oxygen partial pressure and thus also in power is negligible.

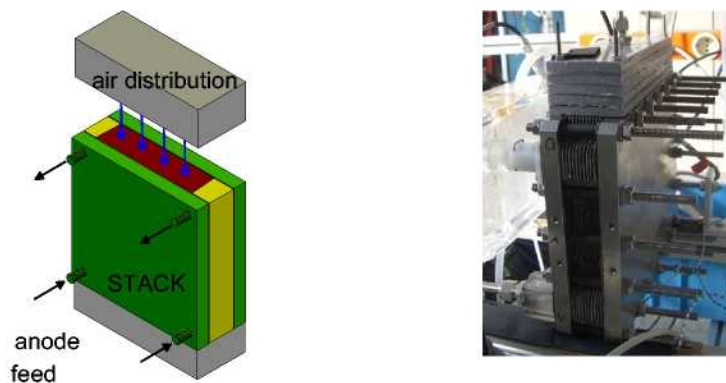


Fig. 3-3: Principle of air distribution (left), 10-cell short stack (right)

From a system engineering point of view, this is very decisive since a pressure loss of 300 mbar can only be overcome using a compressor, whereas an energy-saving blower can be used for the new stack. Since the flow distribution inside the cathodes is optimized and the catalyst layers exhibit an improved water discharge, it has moreover been possible to more than halve the air ratio required for operation.

The 2-kW system presented in 2002 was too large and too heavy (approx. 450 kg) for practical applications. A large portion of the construction volume was occupied by system components and piping. Due to the principle of system integration it has been possible to reduce the space requirements to such an extent that the complete system can be integrated into the scooter. System integration means that as many system components as possible are integrated into the stack. Fig. 3-4 on the left shows the chassis of the scooter as delivered with three lead-acid batteries, whereas on the right the stack together with the system can be seen after installation in the scooter. Apart from the actual 100 active cells, the stack for the

scooter also contains an integrated system composed of methanol pump, CO₂ separator and integrated water equalization tanks. This design permits a compacting of the overall system omitting, in particular, a large portion of the connecting pipes between the individual system components. Process engineering and system integration will be dealt with in detail in the chapter "1.1.2 Process engineering and system integration".



Fig. 3-4: Scooter in the as delivered state with lead-acid battery (left), after modification with DMFC system (right)

3.1.2.2 Development and construction of PEFC stacks

In cooperation with RWTH Aachen University, work on the development of a 5-kW PEFC stack was continued in terms of flow distribution, sealing, humidification concept and heat management and has contributed to the design and construction of the 5-kW stack.

At the centre of stack design in the period under review were the design and construction of a 5-kW PEFC stack with optimized bipolar plates of graphite. The fuel cell stack is provided with a total of 55 single cells with an active area of 240 cm² each. This corresponds to a specific power density of 380 mW/cm². The membrane electrode assemblies (MEAs) produced on the desk coater at IWV-3 are compared inside the stack with commercially available MEAs. The total of 55 cells are divided into 18 commercial MEAs used for reference purposes and 37 MEAs manufactured in house. The arrangement of the plates and connections can be seen from Fig. 3-5. Moreover, for assessing the influence of the material of the bipolar plates on electric power, plates of titanium are also used in addition to graphite plates. These titanium plates are gold plated to create an ideal contact resistance. The total of 55 single cells are subdivided into 45 graphite and 10 metal cells. The geometry of the metal cells is identical to that of the graphite cells and especially the flow structures correspond to those of the graphite plate. This is necessary in order to adjust uniform pressure losses and exclude influences, for example, due to different flow rates inside the cells.

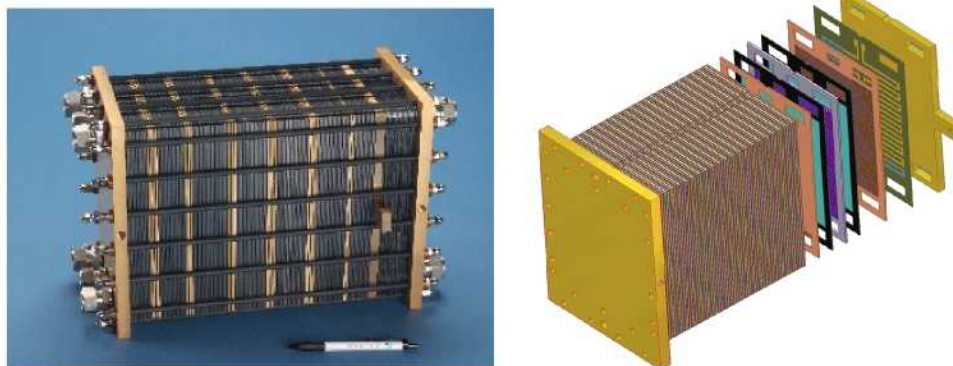


Fig. 3-5: 5-kW PEFC stack (left) and exploded view (right)

A structure with parallel meanders is used as the flow distributor. This structure has been simulated and visualized within the framework of a PhD thesis. The results are shown in Fig. 3-6. The individual meanders show a uniform flow so that a homogeneous distribution of the reactants with low pressure loss is ensured. In contrast, a trend to undersupply of the structures located at the centre is to be seen for the combined channel/grid structure.

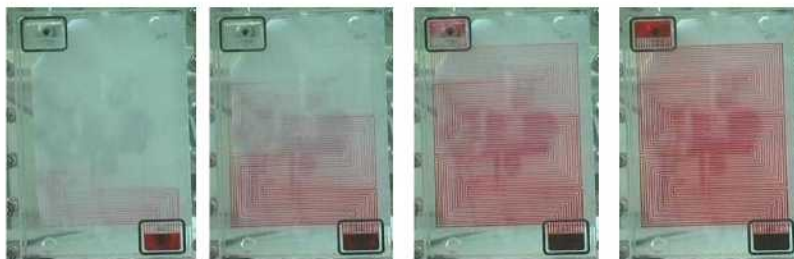


Fig. 3-6: Visualization experiments on flow distribution using coloured liquids in a meandering structure

Membrane humidifiers are used for humidifying the hydrogen and the air side. The H_2 humidifier is integrated into the stack and consists of 8 humidifier cells. The air humidifier is of membrane design in analogy to the H_2 humidifier and consists of 20 humidifier cells. In order to be able to adjust a large range of humidifier temperatures, the air humidifier is designed as a separate component and uncoupled from the cooling water circuit of the stack.

The waste heat from the stack must be dissipated to the environment through a cooling loop. This purpose is served by cooling cells, which should be as small as possible for reasons of constructed space and weight. The original cooling flow field, which at the start of the project was still milled into the bipolar plate involving much effort, has been replaced by a concept based on a distributor plate of expanded graphite. This distributor plate is positioned between the bipolar plates divided into halves.

Fig. 3-7 shows a comparison between the MEAs produced at FZJ and MEAs commercially available. All the 55 cells are plotted. Up to an electric current of 100 A no difference is observable between the two MEA types and among the cells. Only above approx. 100 A do the in-house and commercial MEAs exhibit perceivable differences in performance. With increasing current intensity the higher internal resistance of the FZJ MEAs, which is caused by the higher membrane thickness of 90 μm , becomes more apparent in comparison to the commercial MEAs with a membrane thickness of only 35 μm .

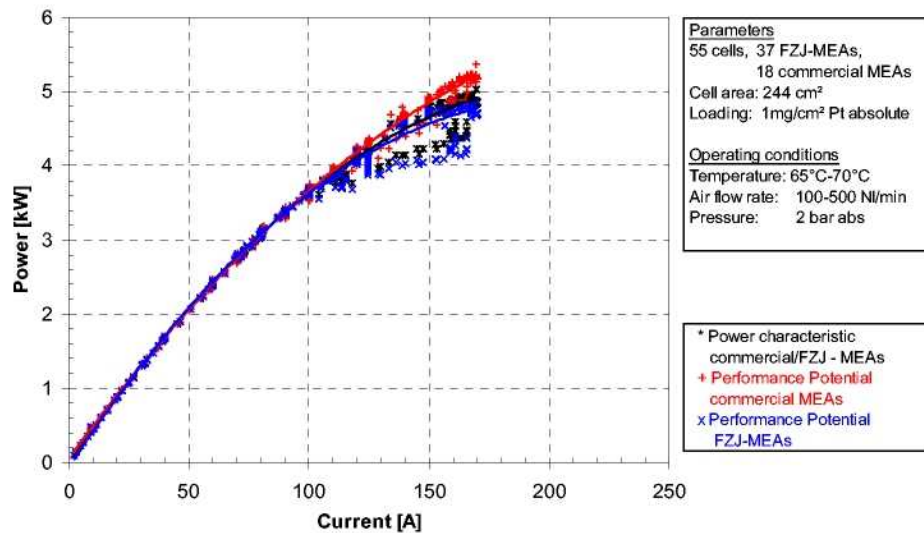


Fig. 3-7: Performance data of the 5-kW stack

3.1.3 Scientific staff and fields of activity

Name	Tel. no. (+49 2461-61-) e-mail address	Field of activity
J. Mergel	5996 j.mergel@fz-juelich.de	Head of the Electrochemical Converters/Low Temperature department (EWN)
Dr. H. Dohle	6884 h.dohle@fz-juelich.de	Head of the "PEFC/DMFC Stack Development" group, technical and economical optimization of stacks, selection and testing of materials for cell components
D. Kalkreuth	2378 d.kalkreuth@fz-juelich.de	Development of continuous machine fabrication processes for components of the membrane electrode assembly (MEA), quality management
N. Kimiaie	6484 n.kimiaie@fz-juelich.de	Technical and organizational project coordination for DMFC systems, analysis of current density and flow distribution in fuel cell stacks
M. Müller	2775 mar.mueller@fz-juelich.de	Volume- and performance-optimized design of DMFC stacks, integration of peripherals, technical and economic evaluation of innovative approaches
C. Schlumbohm	2177 c.schlumbohm@fz-juelich.de	Development of highly disperse, stable catalyst pastes for the continuous machine fabrication of optimized catalyst layers
H. Schmitz	4113 hei.schmitz@fz-juelich.de	Degradation and ageing mechanisms in direct methanol fuel cells
M. Stähler	2775 m.staehler@fz-juelich.de	Development of new characterization methods for the electrodes of DMFCs
Dr. Chr. Wannek	4013 c.wannek@fz-juelich.de	Development of membrane electrode assemblies without humidification requirement for the operation of PEM fuel cells at elevated temperature
Dr. K. Wippermann	2572 k.wippermann@fz-juelich.de	Electrochemical characterization of single electrodes and MEAs, performance optimization of MEA components by elucidating the structure/effect relationship

3.1.4 Important publications, PhD theses and patents

Important publications:

Dohle, H.; Jung, R.; Kimiaie, N.; Mergel, J.; Müller, M.

Interaction between the diffusion layer and the flow field of polymer electrolyte fuel cells - experiments and simulation studies

Journal of Power Sources, 124 (2003), 371-384

The flow distribution in a fuel cell has a decisive influence on fuel cell performance. The flow distribution structures inside the bipolar plate may be channels or also meanders. If regions in a cell are insufficiently supplied with reactants, the desired performance is no longer achieved there. In this paper, the interactions between the distribution structure and diffusion layer of the fuel cell are studied. Due to the fact that the diffusion layer is porous, it also provides the reactants with transport paths in parallel to the plane, which are also used if a pressure difference occurs. The resulting flow in a fuel cell is thus generally composed of a flow fraction in the diffusion layer and a flow fraction inside the distribution structures of the bipolar plate. Within the framework of this publication, the characteristic parameters have been determined, with the aid of which the homogeneity of the flow can be calculated for different meander geometries and different porosities of the diffusion layer. It was found that meanders do not always distribute the flow homogeneously, but can also cause inhomogeneous distribution in the case of practically relevant geometries.

Bewer, T.; Beckmann, T.*; Dohle, H.; Mergel, J.; Stolten, D.

Novel method for investigation of two-phase flow in liquid feed direct methanol fuel cells using an aqueous H₂O₂ solution

Journal of Power Sources, 125 (2004), 1-9

One major issue in the development of direct methanol fuel cells (DMFCs) is the design of flow structures enabling an easy detachment of the CO₂ gas bubbles produced. These gas bubbles influence the flow distribution and can thus have a negative effect on the electric power output of the cell. In this publication, a new method is described for the in-situ observation of gas bubbles in a special perspex cell. The method is based on the catalytic decomposition of hydrogen peroxide to oxygen and water. With the aid of a suitable concentration of the hydrogen peroxide solution, the gas release rate can be controlled and brought to the same level as in a real DMFC. This new approach allows the technical simulation of the flow distribution in a DMFC without expensive hardware. In particular, no current-conducting parts are needed, so that the whole cell can be made of transparent components. With the aid of this novel measuring method, flow structures based on channels and based on grids were investigated.

Dohle, H.; Wippermann, K.

Experimental evaluation and semi-empirical modeling of *U/I* characteristics and methanol permeation of a direct methanol fuel cell

Journal of Power Sources, 135 (2004), 152-164

The operating parameters in a DMFC have a great influence on the power output and efficiency. Especially the methanol concentration influences the overpotentials on

both the anode and the cathode. Furthermore, the efficiency of the cell is affected by methanol permeation from the anode to the cathode and subsequent methanol oxidation. In this publication, based on measurements of the individual effects, a model is generated which predicts the current-voltage characteristics and methanol permeation as a function of the operating parameters. Thus, the generation of performance charts for different operating parameters is possible, which can be used for system design.

Havránek, A.; Wippermann, K.

Determination of the proton conductivity in anode catalyst layers of the direct methanol fuel cell (DMFC)

J. Electroanal. Chem. 567 (2004) 305–315

A new method has been developed, with which the specific ion conductivity in electrochemically active composite layers consisting of an ion- and an electron-conducting phase can be determined. As an example, the proton conductivity of DMFC anode catalyst layers was determined for different Nafion contents (6 – 12 vol.%). The ion or here proton conductivity is, in principle, determined from a combination of two measuring methods: a. impedance spectroscopy, determination of the ratio of proton conductivity and double layer capacitance from the impedances of high measurement frequencies b. cyclic voltammetry with determination of the double layer capacitance. As a result, the specific proton conductivities in the range of some mS/cm were obtained. This is more than one order of magnitude lower than the specific proton conductivity of Nafion. This permits the conclusion that the proton conductivity of the catalyst layer is very much influenced by its microstructure and tortuosity.

Kulikovskiy, A.A.; Scharmann, H.; Wippermann, K.

Dynamics of fuel cell performance degradation

Electrochemistry Communications 6 (2004) 75–82

A theory on the dynamics of fuel cell degradation has been developed, with which the course of galvanostatic ageing experiments can be explained. This concerns, in particular, a degradation behaviour in which the ageing rate continuously increases with operating time and the cell voltage finally collapses. The theory presented does not presuppose a specific ageing mechanism for explaining this ageing behaviour. It rather follows the assumption that a universal scenario is involved, in which, upon exceeding a critical current density at the gas inlet of the cathode, a degradation wave is generated, which propagates along the channel towards the gas outlet. Assuming that local degradation means complete inactivity, i.e. that the front of the degradation wave interrupts the locally flowing current, the activity of the channel section behind the wave front equals zero. To this end, the channel section in front of the wave front must take up the total, constant current. The further the degradation wave propagates, the smaller is the remaining, active channel section and the higher the average current density across this channel section. The model presented here is supported by the fact that it explains the rapid drop in cell voltage at the end of the ageing test. This theory also explains why the oscillations of cell voltage increase with small stoichiometry factors on the air side (λ near 1).

Kulikovsky, A.A.; Scharmann, H.; Wippermann, K.

On the origin of voltage oscillations of a polymer electrolyte fuel cell in galvanostatic regime

Electrochemistry Communications 6 (2004) 729–736

The paper compares experimental and theoretical investigations of the voltage oscillations that occur in a PEFC under galvanostatic conditions below a critical value of cathode air supply. The physical reason for this behaviour is the fact that two possible polarization voltages exist for the same average current density at low air supply. The model allows the determination of the critical values and, moreover, explains the time dependence of the average voltage and the oscillation amplitudes.

Bewer, T.; Beckmann, T.*; Dohle, H.; Mergel, J.; Stolten, D.

Novel method for investigation of two-phase flow in liquid feed direct methanol fuel cells using an aqueous H₂O₂ solution

Journal of Power Sources, 125 (2004), 1-9

One major issue in the development of direct methanol fuel cells (DMFCs) is the design of flow structures enabling an easy detachment of the CO₂ gas bubbles produced. These gas bubbles influence the flow distribution and can thus have a negative effect on the electric power output of the cell. In this publication, a new method is described for the in-situ observation of gas bubbles in a special perspex cell. The method is based on the catalytic decomposition of hydrogen peroxide to oxygen and water. With the aid of a suitable concentration of the hydrogen peroxide solution, the gas release rate can be controlled and brought to the same level as in a real DMFC. This new approach allows the technical simulation of the flow distribution in a DMFC without expensive hardware. In particular, no current-conducting parts are needed, so that the whole cell can be made of transparent components.

Dohle, H.; Wippermann, K.

Experimental evaluation and semi-empirical modeling of *U/I* characteristics and methanol permeation of a direct methanol fuel cell

Journal of Power Sources, In Press

The operating parameters in a DMFC have a great influence on the power output and efficiency. Especially the methanol concentration influences the overpotentials on both the anode and the cathode. Furthermore, the efficiency of the cell is affected by methanol permeation from the anode to the cathode and subsequent methanol oxidation. In this publication, based on measurements of the individual effects, a model is generated which predicts the current-voltage characteristics and methanol permeation as a function of the operating parameters. Thus, the generation of performance charts for different operating parameters is possible, which can be used for system design.

Stähler, M.; Wippermann, K.; Stolten, D.

Instabilities of the reversible hydrogen reference electrode in direct methanol fuel cells

The Electrochemical Society, Proceedings 2004 Joint International Meeting, Honolulu, Hawaii October 3-8, 2004

Electrode characterization plays a central role in optimizing the performance of direct methanol fuel cells (DMFCs). The usual single cell measurements only provide information about the potential difference between cathode and anode. The introduction of an electrochemical reference point, a so-called reference electrode, in the membrane electrode assembly is necessary for the investigation of single potentials. The dynamic hydrogen electrode (DHE) and the reversible hydrogen electrode (RHE) are suitable electrodes for this purpose. It has been shown that in the case of RHE the reference potential is instable and the measurements may thus become unusable. It is necessary to investigate the stability of measurements versus the reference electrode as a function of current density j and cathodic air ratio λ_{air} in the DMFC. In this way, it is possible to identify value ranges for j and λ_{air} within which the reference electrode provides a stable reference point and the single electrodes can be measured.

PhD theses

Bewer, T.

Mass flows and current density distribution in liquid-fed direct methanol fuel cells

Reports of Research Centre Jülich, 4029, RWTH Aachen, 2003 (in German)

The study is concerned with the fluid mechanics and process engineering investigation of liquid-fed direct methanol fuel cells (DMFCs). The influence of operating and design parameters on the performance and the mass and heat balance is considered, combining experimental investigations and model calculations. The results are used to derive clues for optimized cell and stack design.

examined as a function of different flow structures and manifold designs. With the aid of a newly developed method based on the catalytic decomposition of H_2O_2 , the investigation is extended to the two-phase flow situation on the anode. A model concept developed for the flow distribution in the cell very well describes the experimentally observed behaviour. In order to reproduce a cell in real operation, apart from the flow, the heat distribution and the electrochemical potentials of the anode and cathode must also be determined. Measurements of the heat distribution by an infrared camera confirm the model assumptions made for thermal management. The electrochemical potentials and methanol permeation are determined as a function of the operating parameters and subsequently integrated into the existing model. A verified model is thus available which describes the processes in a DMFC in a three-dimensional manner. This modelling tool is used to analyse the influence of operating and design parameters on flow, temperature and current density distribution. A single cell with an active area of 20 cm^2 and a five-cell short stack with the same active area are investigated. The analysis shows that the current density distribution is strongly coupled to the temperature distribution. For a homogeneous flow and current density distribution, it is moreover necessary to carefully match the manifold and flow structure. Splayed manifolds show a homogeneous flow distribution on the anode, since they are very suitable for removing the CO_2 bubbles.

Important patents:

Patent applications:

Principal inventor	PT	Title
Dohle, Dr., H.	1.1989	Niedertemperatur-Brennstoffzellensystem sowie Verfahren zum Betreiben eines solchen
Dohle, Dr., H.	1.1998	Kühlzelle für eine Brennstoffzelle
Bewer, Dr., Th.	1.1999	Niedertemperatur-Brennstoffzellenstapel
Egmen, A.	1.2000	Korrosionsstabiler, gasdichter Medienanschluß für Niedertemperatur-Brennstoffzelle
Dohle, Dr., H.	1.2001	Bipolare Platte für eine Brennstoffzelle
Dohle, Dr., H.	1.2048	Verfahren zur Abtrennung von Katalysatorpartikeln
Müller, M.	1.2060	Fluidverteilerstruktur für eine Brennstoffzelle
Bringmann, Dr., J.	1.2062	Niedertemperatur-Brennstoffzelle sowie Verfahren zum Betreiben derselben
Dohle, Dr., H.	1.2067	Kathode für eine Direkt-Methanol-Brennstoffzelle sowie Verfahren zum Betreiben derselben
Jakoby, K.	1.2072	Katalysatorschicht, geeignete Katalysatorpaste sowie Herstellungsverfahren derselben
Müller, M.	1.2077	Polymerelektrolytmembran-Brennstoffzellensystem sowie Verfahren zum Betreiben desselben
Schlumbohm, C.	1.2105	Verfahren zur Behandlung poröser, Kohle umfassender Substrate
Ghosh, Dr., P.	1.2129	Verfahren zur Bestimmung der Stromdichteverteilung in Brennstoffzellen
Ghosh, Dr., P.	1.2130	Vorrichtung zur Bestimmung der Stromdichteverteilung in Brennstoffzellen
Müller, M.	1.2142	Membran-Elektroden-Einheit für eine Brennstoffzelle
Schonert, M.	1.2143	Niedertemperatur-Brennstoffzelle
Müller, M.	1.2147	Brennstoffzellensystem
Stähler, M.	1.2151	Verfahren zur Beschichtung einer Membran mit Katalysator
Schlumbohm, C.	1.2152	Fasern für ein textiles Gewebe sowie deren Herstellung und Verwendung
Nölke, M.	1.2174	Kathodendiffusionsschicht einer Brennstoffzelle

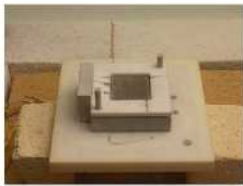
Patents granted:

Principal inventor	PT	Title
Meissner, Dr., D.	1.1595	Einkristallpulver- und Monokornmembranherstellung
Meissner, Dr., D.	1.1624	Verfahren zur Regelung der Brennstoffkonzentration in einem Alkohol oder Ether als Brennstoff und Wasser enthaltenden Brennstoffgemisch einer Brennstoffzelle und Brennstoffzellensystem
Kulikovskiy, Prof. Dr., A.	1.1661	Brennstoffzellen-Elektrode
Gupta, A.K.	1.1748	Herstellung von Katalysatorschichten auf Membranen für Niedertemperatur-Brennstoffzellen
Divisek, Dr., J.	1.1770	Brennstoffzelle
Divisek, Dr., J.	1.1771	Brennstoffzelle
Schmitz, H.	1.1815	Verfahren zur Beschichtung einer Membran-Elektroden-Einheit mit Katalysator und Vorrichtung dafür
Dohle, Dr., H.	1.1823	Bipolare Platte
Dohle, Dr., H.	1.1826	Zweiphasenströmungen
Divisek, Dr., J.	1.1864	Verfahren zur doppelseitigen Beschichtung einer Membran-Elektroden-Einheit mit Katalysator
Stolten, Prof. Dr., D.	1.1931	Beschichtungsvorrichtung
Bewer, Dr., Th.	1.1999	Niedertemperatur-Brennstoffzellenstapel und Verfahren zum Betreiben
Dohle, Dr., H.	1.2067	Kathode für eine Direkt-Methanol-Brennstoffzelle sowie Verfahren zum Betreiben derselben

3.2 Focus on solid oxide fuel cells

3.2.1 Objectives and fields of activity

Due to the high operating temperatures, the solid oxide fuel cell (SOFC) is mostly assigned to applications in the field of decentralized energy supply. After initial experience with SOFC stacks, the application field was extended to smaller systems for e.g. domestic energy supply, probably also involving steady-state base-load operation. Due to the applicability of carbon-containing fuel gases, it has recently also been envisaged to use the SOFC as an auxiliary power unit (APU) in vehicles (or ships and aircraft). In this case, however, more dynamic SOFC performance is required. The SOFC will only be in operation, when the vehicle is used. At the Electrochemical Converters/High Temperature department (EWH), work is being carried out in at first sight very different areas, which all serve the over-arching aim of using the findings on SOFC performance obtained to derive proposals that improve the performance under application conditions.



The electrochemical operating test is regarded as a first very important indication of the functionality of SOFCs. At constant operating temperature (in the laboratory this means in a furnace) the SOFC is supplied with fuel gas and air, and the power output (voltage) is measured as a function of load (current). 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 measurement results of different cells. In close cooperation with material developers it is being analysed whether and how a change in composition or a change in the manufacturing process causes an improvement in power output.

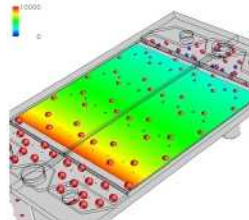
In order to accelerate the development process for stacks, special measurement setups and measuring techniques are used enabling a faster and simpler investigation of the interaction between various stack components. Thus, for example, the interaction between the metallic interconnect and the glass sealant in usual fuel cell atmospheres (air, fuel gas) can be investigated, verifying at the same time the electrically insulating function of the glass sealant 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 hours (5 years) is required for stationary applications. During this time, the fuel cell must not lose more than 10 % of its initial power. Long-term tests are carried out on both single cells and stacks to fulfil this requirement. The cells and stacks are loaded with a constant current over a prolonged time (up to 10,000 hours in

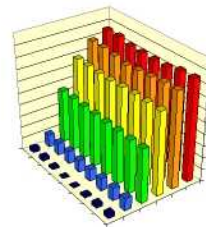
the laboratory). Besides the electric current level, the operating temperature and the fuel gas composition are also important parameters whose influence on cell performance is being investigated.

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 e.g. 100 cycles.



SOFC operation is simulated by modelling in a completely different area. The processes in the SOFC (charge, heat and mass transport) are described by mathematical equations. The models can describe the structures and processes in a very detailed (three-dimensional) or very simplified (one-dimensional) manner. Current, temperature and concentration distributions are determined by simultaneously solving all equations (simulation calculations). The operating behaviour of an SOFC can thus be predicted, providing optimized approaches for rating and design and accelerating the development.

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 determined.



3.2.2 Important work results

The development of planar SOFC technology, as described by the example of the 60-cell stack (see Chapter 1.2), has made considerable progress in recent years. This progress has caused an increased demand for modelling such large stacks, in order to be able to more efficiently derive operational design improvements and optimizations for SOFC power plants from the simulation of stack performance. At IWV-3 the computational fluid dynamics (CFD) tool FLUENT has been used since 1999 for SOFC modelling. Electrochemistry and methane reforming are modelled by user defined functions (UDF).

The CFD model was first used to simulate the flow distributions of the reactants for designing the interconnect plates of the new stack with integrated manifold structures. After validating the calculation results by measurements performed in parallel, the geometries for the E- and the later F-design interconnect plates were determined.

In this first CFD model, the computational grids for the calculation of larger stacks were built up by repeatedly stacking individual planes. Since the models thus became very complex and the computing times very long, it was only possible to model stacks with a maximum of 5 planes in the F-design with large cells ($20 \times 20 \text{ cm}^2$). This restriction was overcome by a new approach. The recent CFD model ("Integrated volume") also enables the simulation of the 60-cell stack on a standard PC.

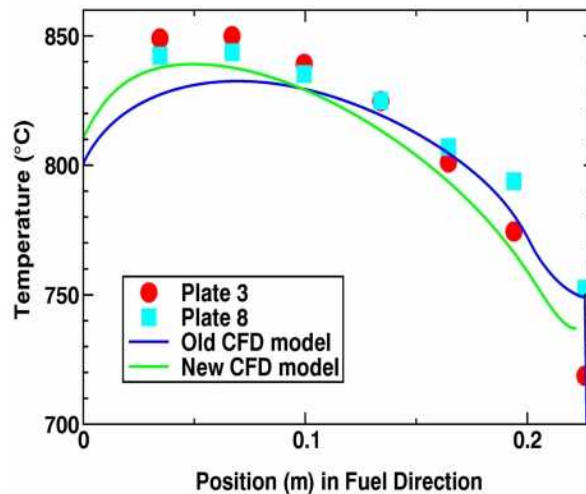


Fig. 3-8: Temperature profiles in one plane: CFD model (lines) and measurement (points)

After the first larger stacks in the F-design had been tested, the CFD models were compared with measured data. In a stack with 10 planes, several thermocouples were arranged on two planes in the interconnect plates. Fig. 3-8 shows a comparison between measured temperatures in the stack and calculated temperature profiles. The CFD simulation calculations very well reproduce the measured temperature profiles, apart from minor deviations in the absolute values; the new model for large stacks is even better than the old one.

Fig. 3-9 shows the simulation of hydrogen distribution in a 60-cell stack during operation with simulated prereformed natural gas, performed with the new CFD model ("integrated volume"). The fresh fuel gas (methane/steam mixture) enters the stack from the bottom left. In the stack, the methane is reformed to hydrogen on nickel in the anode substrate, and rather high hydrogen concentrations rapidly arise. This accelerates the current-supplying electrochemical oxidation of the hydrogen, so that these high concentration values decrease again in the further course (to the right).

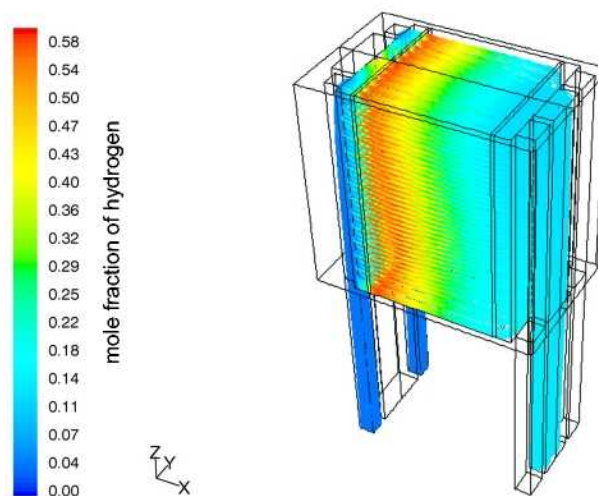


Fig. 3-9: Variation of simulated hydrogen concentration in the SOFC stack with 60 planes

Similar simulation calculations were also performed for other gas components. The distribution of the current density in each plane and the distribution of the temperature in the stack were calculated as well. The calculated temperature profile was later confirmed by measurements. The simulations had already predicted the maximum temperature in the upper half of the stack, just to the left of the centre of the cell area on the fuel gas inflow side.

However, modelling does not only comprise stacks, but also plants in which the SOFC stack is only one component among many others. For such a plant simulation, a 3-D model is much too computing-time-intensive. For plant simulations, a spun-off one-dimensional model is therefore used, which enables a much shorter computing time using the same physico-chemical equations. Whereas the three-dimensional CFD model provides detailed information on all structures (structure model), the one-dimensional spin-off describes the performance of an SOFC considering an individual gas channel (performance model). The one-dimensional simplification is possible in co- and counter-flow operation. Cross-flow operation would merely permit a simplification to two dimensions.

The efficiency of the spun-off one-dimensional performance model is demonstrated in Fig. 3-10. The operating conditions are typical of an SOFC plant with internal methane reforming. A cell voltage of 0.71 V is obtained for an average current density of 300 mA/cm² and a fuel gas utilization of 80 %. The gas composition (Fig. 3-11) along the flow direction of the gas channels is determined in the same way as in the CFD simulation.

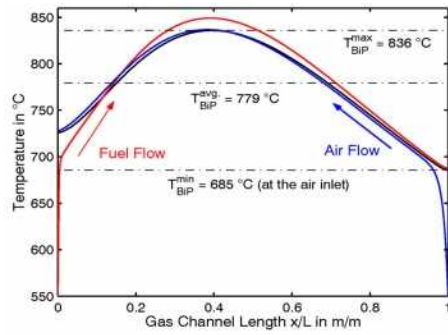


Fig. 3-10: Temperature in the gas channel

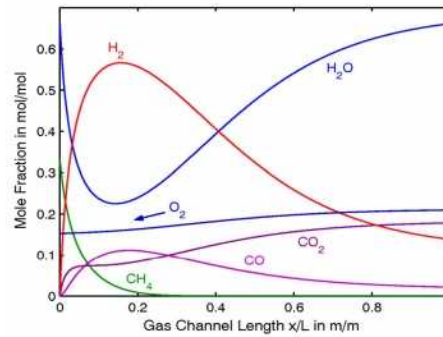


Fig. 3-11: Concentrations in the gas channel

The very fast 1-D performance model allows and therefore also enables the simulation of transient processes such as load changes. Fig. 3-12 shows as an example how the temperature profile changes over time in bipolar plates, if the load is reduced from 300 mA/cm² (as in Fig. 3-10) to 240 mA/cm². After slightly more than one hour, a new stable operating situation is already reached again. Since fuel gas utilization and air volume were kept constant during simulation, the maximum temperature decreases to a clearly lower value.

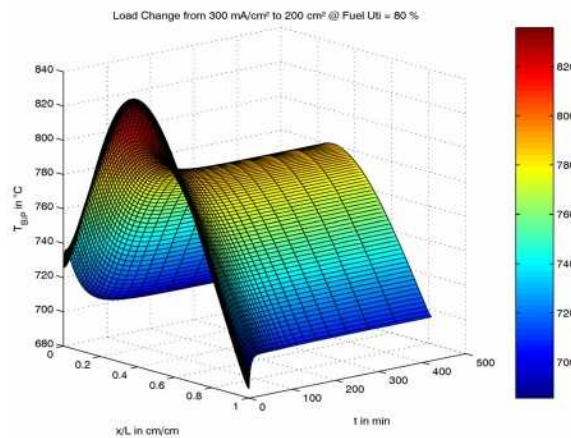


Fig. 3-12: Simulation of a load change by the 1-D performance model

3.2.3 Scientific staff and fields of activity

Name	Tel. no. (+49 2461-61-) e-mail address	Field of activity
Dr. L.G.J. de Haart	6699 l.g.i.de.haart@fz-juelich.de	Head of the Electrochemical Converters/High Temperature department (EWH)
D. Froning	6676 d.froning@fz-juelich.de	Informatics, programming, modelling
Dr. A. Gubner	3018 a.gubner@fz-juelich.de	Head of the "SOFC Modelling" group, physics, programming, modelling
Dr. V.A.C. Haanappel	4656 v.haanappel@fz-juelich.de	Chemical technology, (high-temperature) corrosion
Dr. I.C. Vinke	6688 i.c.vinke@fz-juelich.de	Electrochemistry, 'solid state ionics', high-temperature electrochemistry
C. Wedershoven	4124 c.wedershoven@fz-juelich.de	Mass transport in porous bodies; experiment and modelling

3.2.4 Important publications, PhD theses and patents

Important publications

Ackmann, Th.; Haart, L.G.J. de; Stolten, D.

Modelling of mass transport in planar substrate type SOFCs

Proc. 5th European Solid Oxide Fuel Cell Forum (Ed., J. Huijsmans), Lucerne 2002, Vol. 1, 1011-1016

A mathematical model has been developed, which incorporates the mass transport in the porous structures of planar substrate type solid oxide fuel cells (SOFCs). Two dimensional simulations can be performed for both anode as well as cathode substrate type cells. Results of a systematic analysis of the transport processes in anode substrate type cells show that molecular diffusion has the largest contribution, but both permeation and Knudsen diffusion cannot be neglected. As an example of identifying critical parameters influencing gas concentration and current density, results of simulations with different cathode layer geometries are presented.

Blum, L.; Haart, L.G.J. de; Vinke, I.C.; Stolten, D.; Buchkremer, H.-P.; Tietz, F.; Blaß, G.; Stöver, D.; Remmel, J.; Cramer, A.; Sievering, R.

Planar anode substrate type SOFC kW-class stack development

Proc. 5th European Solid Oxide Fuel Cell Forum (Ed., J. Huijsmans), Lucerne 2002, Vol. 1, 784-790

Within the technology development programme on anode supported SOFCs in Jülich, short-stack (2 cells 10 cm x 10 cm; 81 cm²) tests were performed in order to improve, amongst others, the durability. First results with interconnect plates machined from a newly developed ferritic steel JS-3 show degradation rates of less than 1%/1000 h

during the first thousand hours of continuous operation. In order to improve, amongst others, the reproducibility, several kW-class stacks (10-40 cells 20 cm x 20 cm; 361 cm²) were assembled and operated. The 40-cell stack produced a maximum power of 9.2 kW (300 A @ 30.2 V) operating on hydrogen (10 % humidified, 76 % fuel utilization). Operating on simulated partially pre-reformed methane the stack produced a maximum power of 5.4 kW (180 A @ 30.2 V).

Gubner, A.; Froning, D.; Haart, L.G.J. de; Stolten, D.

Complete modelling of kW-range SOFC stacks

Solid Oxide Fuel Cells VIII (SOFC VIII), S.C. Singhal and M. Dokiya, Editors, PV 2003-07, 1436-41, *The Electrochemical Society Proceedings Series*, Pennington, NJ (2003)

As planar solid oxide fuel cell (SOFC) stack development progresses and moves on from watt-range single cells to commercially more interesting kW-range SOFC stacks, a need for adequate modelling capabilities arises to simulate these SOFC stacks properly. Commercially available tools for 3-D modelling such as computational fluid dynamics (CFD) programs nowadays allow unstructured meshing for coding even complex design topologies. At Research Centre Jülich, the transition to the CFD tool Fluent was made. Initial stack models were composed of repeating single cell units. This contribution also proposes a strategy for speeding up simulations of complete power plant cycles involving the CFD model and a spun-off 1-D simplification.

Haanappel, V.A.C.; Vinke, I.C.; Wesemeyer, H.

A Novel Method to Evaluate the Suitability of Sealing Materials for SOFC Stacks

Proc. 6th European Solid Oxide Fuel Cell Forum (ed. M. Mogensen), Lucerne 2004, Vol. 2, 784-791

A novel method has been developed to evaluate the suitability of sealing materials under more realistic conditions for SOFC stacks. This method is based on two metallic sheets, joined together with a 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 sealant. Experiments reveal that the chemical and electrical behaviour of the sample strongly depends on the experimental conditions. Under oxidative conditions (exposure to air) no undesired interactions take place detrimentally affecting the electrical properties, indicating a satisfactory suitability of the sealant – alloy combination. However, under more realistic conditions, in this case the use of a dual environment with air on the outer part and hydrogen on the inner part of the sample, a different behaviour occurred. Under these test conditions, a strong reduction of the specific resistance of some samples (between the two metallic sheets and based on 4-point DC resistivity measurements) was observed, due to undesired chemical reactions between the sealant, the alloy, and the surrounding gas atmosphere.

Steinberger-Wilckens, R.; Haart, L.G.J. de; Vinke, I.C.; Blum, L.; Cramer, A.; Rimmel, J.; Blaß, G.; Tietz, F.; Quadackers, W.J.

Recent results of stack development at Forschungszentrum Jülich

Solid Oxide Fuel Cells VIII (SOFC VIII), S.C. Singhal and M. Dokiya, Editors, PV 2003-07, 98-104, *The Electrochemical Society Proceedings Series*, Pennington, NJ (2003)

Test results from the 5th generation SOFC stacks developed at Forschungszentrum Jülich, the so-called E-design stacks, are presented. Reported are 5 kW_{el} gross

power operating on methane, long-term testing up to 4,000 hours and thermal cycling capabilities. The 6th generation F-design is introduced with basically the same configuration, but reduced manufacturing effort and costs and improved reproducibility.

Steinberger-Wilckens, R.; Vinke, I.C.; Blum, L.; Haart, L.G.J. de; Remmel, J.; Tietz, F.; Quadakkers, W.J.

Progress in SOFC Stack Development at Forschungszentrum Jülich

Proc. 6th European Solid Oxide Fuel Cell Forum (ed. M. Mogensen), Lucerne 2002, Vol. 1, 11-19

Forschungszentrum Jülich (FZJ) is currently testing the 6th and 7th generations of stack design (F- and G-design, respectively) for solid oxide fuel cells (SOFCs). These stack designs are based on the planar, anode substrate type cells and metallic interconnect plates made of the ferritic steel JS-3 developed in Jülich. The focus of current research is on the one hand directed at achieving low degradation rates and a high reproducibility of results. In the new G-design on the other hand the principal design of the massive F-design has been transformed into a concept where the cell frames and interconnects are made of sheet metal. The potential for weight reduction is in the order of 90 %. The cathode and cathode contact layer materials applied today allow the operation of stacks at temperatures between 700 and 750 °C whilst maintaining power densities similar to those obtained in earlier tests at 800 °C and above. This results in reduced degradation due to lower corrosion rates of the metallic interconnect and reduced interaction with the other stack materials. Setting up of the 20 kW SOFC system has further progressed and resulted in a compact design which integrates pre-reformer, stack and afterburner into a stack module within a single thermal encapsulation.

Vinke, I.C.; Haart, L.G.J. de; Blum, L.; Stolten, D.

Influence of SOFC stack materials on the time behaviour of the performance of anode substrate cells

Proc. 5th European Solid Oxide Fuel Cell Forum (Ed., J. Huijsmans), Lucerne 2002, Vol. 1, 162-169

The influence of the commercial ferritic steel 1.4742 on the ageing of a planar anode substrate SOFC is studied. Steel plates are placed near the electrodes or are used as current collector. Over a maximum period of 650 hours, little or no ageing is observed during these single cell experiments when 1.4742 is present at the anode side. A slight degradation may occur due to alumina scale growth effectively increasing the contact resistance between the nickel mesh and the steel current collector. On the cathode side, significant ageing due to the presence of the steel is observed. This effect is probably due to chromium poisoning of the cathode. A layer of lanthanum cobaltite on the steel significantly reduced the ageing effect. This is most likely caused by the reaction of the chromium oxide evaporated from the steel and the lanthanum cobaltite contact layer. Due to the oxide scale formation the steel 1.4742 is considered inappropriate for use in SOFC stacks.

Wedershoven, C.; Rommerskirchen, R.; Gubner, A.; Stolten, D.

Determination of Transport Parameters on Thin Cathodes of Planar Anode Substrate Type Solid Oxide Fuel Cells

Proc. 6th European Solid Oxide Fuel Cell Forum (ed. M. Mogensen), Lucerne 2004, Vol. 3, 1330–1340

Products of the transport parameters of the mean transport pore model were determined at cathode layers (and anode substrates) of ready-made planar anode substrate type solid oxide fuel cells (SOFC) by means of permeation experiments. For this purpose the experimental set-up and the description of the fluxes of the permeation experiment, originally developed by Fott and Petrini, were modified.

In order to realize a flow entering and leaving the cathode layer through the same surface a sample holder was developed which extends the area of the lower compartment to the surface of the cathode. Due to the new direction of flow the latter has to be described in a 3-dimensional manner. In order to be able to analyse the experiment according to the standard procedure the description of the fluxes had to be simplified. These simplifications and first results of the permeation experiments are presented.

PhD theses:

Ackmann, T.

Micromodelling of a ceramic high-temperature fuel cell

Reports of Research Centre Jülich ; 4119, RWTH Aachen, 2003 (in German)

The planar solid oxide fuel cell, SOFC, provides a great application potential for stationary energy supply. Apart from experimental work on fuel cell stacks and single cells, modelling is a suitable tool for the optimization of operating conditions, material characteristics and design parameters. In the theoretical part of this thesis, a three-dimensional model for the description of the mass and heat transport in the channels and solid materials of a planar SOFC has been developed. This model was implemented in a commercially available computational fluid dynamics software and takes the mass transport of a multi-component mixture in the porous electrodes into account. In contrast to other models, permeation is integrated as a function of essential structural parameters, which are characteristic of porous solids and provide information about the transport behaviour of gases in porous solid material. The experimental part of the study comprises the determination of these structural parameters needed as input data for simulation. The parameters are determined by diffusion experiments in a modified Wicke-Kallenbach cell and from permeation experiments.

Apart from the mass transport of the multi-component mixture, the chemical and electrochemical reactions, the heat transport and the geometry of the cell are taken into account in modelling. The analysis of the mass transport in the anode shows that the concentrations of the electrochemically active gases hydrogen and carbon monoxide are sufficiently high at the electrolyte. In contrast, the oxygen transport in the cathode as a function of cathode thickness exhibits too low concentrations underneath the protuberances. This leads to concentration overpotentials which have a negative effect on the cell output. The results provide minimum values for the structural parameters of the cathode and a limit value for the cathode thickness, which in combination enable high cell voltages.

The temperature distributions also calculated from the simulations allow conclusions to be drawn concerning the thermal load of the materials. The three-dimensional

temperature distributions show a comparatively homogeneous temperature field inside the porous solid (1.7 to 3.9 K/mm) despite the chemical and electrochemical reactions with heat influence. At the surface of the solid, however, significant temperature differences between gas and solid are observed. They can cause cracks in the porous electrodes due to resulting temperature stresses. The model developed is an extensive tool for SOFC modelling, which can be used, in particular, in future work with different cell designs for optimizing the material properties.

Important patents:

Patent applications:

Principal inventor	PT	Title
Froning, D.	1.2071	Verfahren zur Modellierung von Stoff- und/oder Wärmetransportvorgängen in einer Vorrichtung sowie Vorrichtung zur Durchführung des Verfahrens
Wedershoven, Ch.	1.2140	Verfahren und Vorrichtung zur Ermittlung von Mikrostrukturparametern von dünnen, porösen Schichten

Patents granted:

Principal inventor	PT	Title
Haart, de, Dr., L.G.J.	1.1576	Bipolare Platte mit poröser Wand
Thom, Dr., F.	1.1715	Kühlsystem für Brennstoffzellen

3.3 Fuel processing

3.3.1 Objectives and fields of activity

The availability of hydrogen forms the basis for the use of fuel cells in mobile and stationary applications. At present, however, there does not yet exist an infrastructure for hydrogen as a future energy carrier. It is therefore necessary to produce hydrogen from natural gas, gasoline or diesel as adequately available energy carriers. Under the heading of fuel processing, the working group on fuel processing systems is concerned with three different aspects of this topic: fuel processing, reforming and fuel gas conditioning. In the reforming process, the feedstock is converted into a hydrogen-rich gas either with water only or with air only or with a water-air mixture on a solid catalyst at high temperatures. Important parts of the R&D activities are process engineering analysis, component development and system configuration. All areas of work are supported by adequate modelling.

In the field of fuel processing, process development for the desulphurization of liquid fuels and their evaporation is theoretically and experimentally studied.

Concerning electricity and on-board power supply for portable and mobile applications in the 5-kW_{el} power range, the focus of activities is on the autothermal reforming of diesel and diesel-like fuels. Reformer development is carried out both for the SOFC high-temperature fuel cell and the PEFC low-temperature fuel cell.



Since the fuel gas produced by reforming generally contains too much carbon monoxide for the permanent operation of a PEM fuel cell, it must be subjected to gas conditioning.

The Fuel Processing and Systems department has been concerned on a priority basis since 1990 with the reforming of fuels such as natural gas, methanol and diesel. Experience is available in the fields of process engineering and technical chemistry, specifically in the field of heterogeneous catalysis, in the form of 200 staff-years, broken down into 50 % for methanol, 30 % for natural gas and 20 % for diesel as the energy carriers. Work on natural gas reforming is specifically geared to the concept of internal reforming for the SOFC. At the end of 2003, the research and development results on prereforming were transferred to the SOFC systems development after successful completion. Since the beginning of work on diesel reforming in 1999, methanol as an energy carrier for reforming has been more and more cut back, because there is no infrastructure for methanol for vehicle applications. Since 2004 all research and development projects have been subsumed under the term "middle distillates". Analysed are the refinery products diesel and kerosine and in future mixtures of diesel, biodiesel and synthetic Fischer-Tropsch diesel.

3.3.2 Important work results

3.3.2.1 Reforming of liquid hydrocarbons

At IWV-3, a reformer for diesel fuel has been developed in recent years, which can be seen in Fig. 3-13. This reformer is capable of producing an amount of hydrogen which is sufficient for a power of 3 – 5 kW_{el} in the fuel cell. The preferential process is so-called autothermal reforming, involving a cold-starting device, internal steam generation and a device for extracting the process heat arising during reforming. If the catalyst is already preheated to 350 °C, the starting time is 1 min, otherwise approx. 7 min.



Fig. 3-13: Diesel reformer

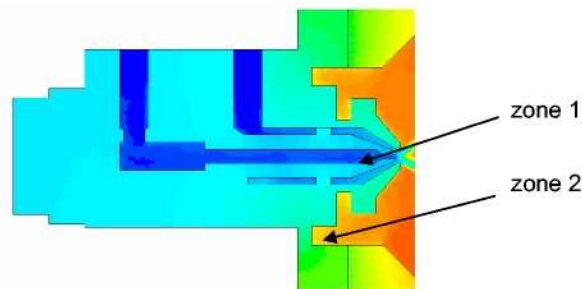


Fig. 3-14: CFD modelling of the two-fluid nozzle

In the upper region of the reformer there is a two-fluid nozzle, with the aid of which the liquid diesel fuel is atomized as aerosol together with an air stream. It is important here, on the one hand, that the temperature in the interior of the nozzle (cf. zone 1 in Fig. 3-14) does not exceed 120 °C to avoid pyrolysis and evaporation of the fuel. Pyrolysis leads to clogging of the nozzle. If diesel evaporation already sets in inside the nozzle, continuous proportioning is no longer possible. On the other hand, the temperature on the outer surface of the nozzle must not fall below approx. 150 °C, since otherwise the fuel condenses and accumulates there (cf. zone 2 in Fig. 3-14). This leads to the formation of carbon-containing depositions on the nozzle impairing the function of the nozzle in the long run. Fig. 3-15 illustrates such a process of continuously growing depositions on the nozzle under unfavourable reaction conditions.



Fig. 3-15: Carbon-containing depositions in diesel reforming

In order to better understand and predict the described demands made on the temperature conditions in the interior of the two-fluid nozzle, computational fluid dynamics (CFD) modellings are carried out. They are aimed at providing as precise information as possible about the temperatures arising in the nozzle as a function of given reaction conditions such as the temperatures and flow velocities of air and diesel fuel and the geometry of the two-fluid nozzle. Fig. 3-14 shows the result of such modelling.

With the diesel reformer from Fig. 3-13 it is possible to convert commercially available diesel fuel, Ultimate Diesel from Aral, into a hydrogen-rich gas over a period of 200 h using autothermal reforming. Fig. 3-16 illustrates this experiment.

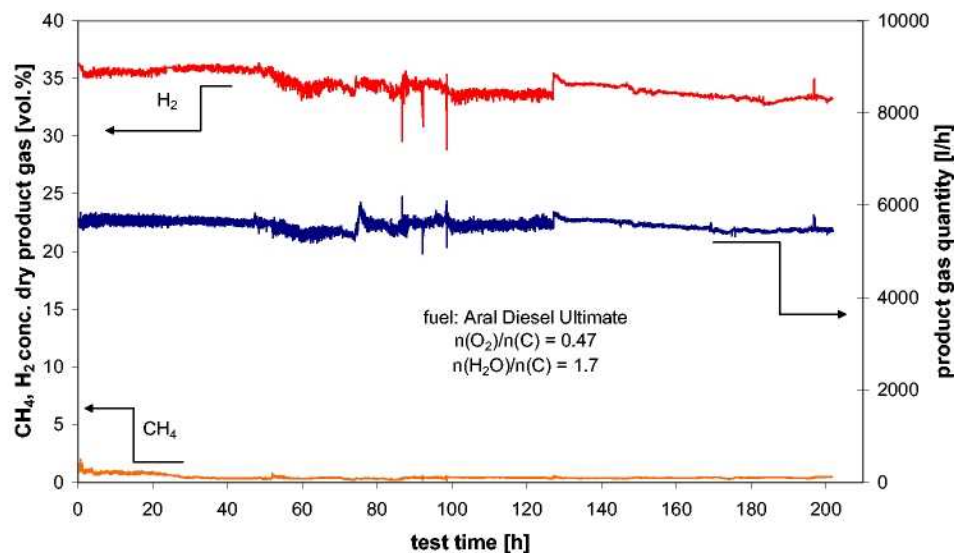


Fig. 3-16: CH₄, H₂ concentration and product gas quantity in autothermal reforming

Fig. 3-16 shows that the concentration of hydrogen during the experiment decreased from approx. 35 % at the start to approx. 33 % after 200 h. A comparable trend was found for the product gas quantity. The concentration of methane arising as a byproduct for thermodynamic reasons, as do carbon monoxide and carbon dioxide, was very low during the entire experiment. Methane has a much greater potential for causing the so-called greenhouse effect than carbon dioxide, so that its formation must be avoided as far as possible. In spite of the slight deterioration in hydrogen yield, it was possible to carry out reforming with high efficiencies. At the beginning of the experiment, the efficiency was more than 80 % and after 190 h still more than 75 %. Diesel conversion was also greater than 99 % after 190 h.

3.3.2.2 Gas conditioning by water-gas shift reaction

In order to be able to feed the gas mixture produced during reforming (reformat) into the fuel cell, it is necessary to greatly reduce the concentration of carbon monoxide in the reformat. It amounts to approx. 10 vol.%. According to the present state of the art, low-temperature fuel cells can only operate without power losses if the concentration of carbon monoxide in the supplied reformat is less than 50 ppm. Carbon monoxide causes the active centres of the electrocatalysts of the fuel cells to be blocked by adsorption, so that they are no longer available for catalysis of the electrochemical oxidation of the hydrogen. A very simple reaction, from the chemical point of view, for reducing this carbon monoxide concentration in the reformat is the so-called water-gas shift (WGS) reaction, in which the carbon monoxide is converted with water into carbon dioxide and the desired target component during hydrogen reforming. A disadvantage of this reaction is that it proceeds with poor kinetics, i.e. at low reaction rates, and that the catalysts available at present are too inactive. In order to nevertheless identify the most suitable catalyst for the development of a water-gas shift reactor, extensive screening of catalysts available on the market was performed. The result is shown in Fig. 3-17. It can be seen that catalyst C clearly showed the best catalytic activity at a space velocity (GHSV) of $12,200 \text{ h}^{-1}$ and already displayed a conversion of approx. 90 % at temperatures of 280°C . Carbon monoxide conversion was still about 40 % at 220°C .

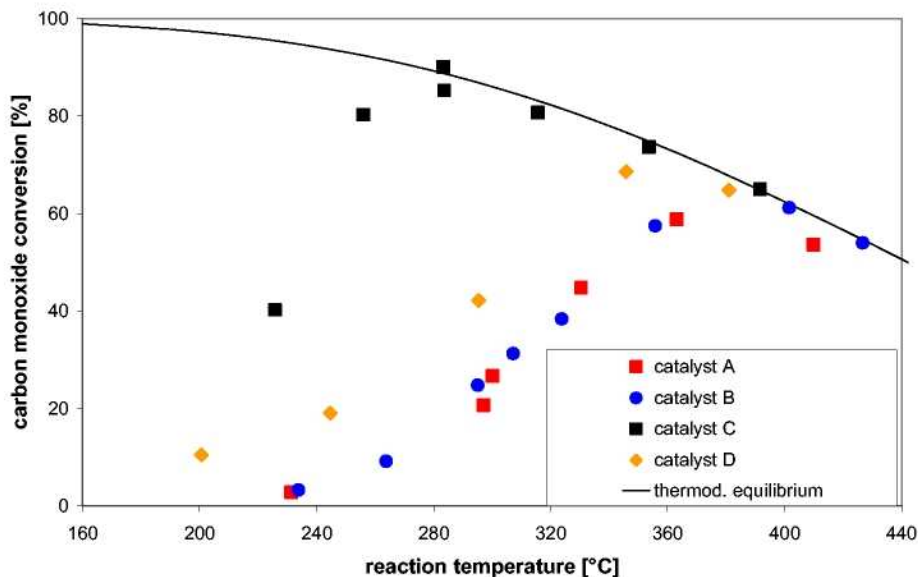


Fig. 3-17: CO conversion as a function of reaction temperature using four different, commercial, monolithic catalysts, GHSV = $12,200 \text{ h}^{-1}$

For this reason, this catalyst was incorporated in the reactor for water-gas shift reaction shown in Fig. 3-18. This reactor is of two-stage design and contains a high-temperature section operated at approx. 400°C and a low-temperature section with an inlet temperature

of approx. 300 °C. Water can be added in front of the low-temperature region. The high- and low-temperature sections are operated at different space velocities. This constellation makes it possible to reduce a carbon monoxide concentration of approx. 10 vol.% in the reformat to less than 1 vol.%. A further reduction using the WGS reaction is not possible at present due to the insufficient kinetics when applying the catalysts available. This requires an additional process step, the so-called preferential oxidation (PROX) of carbon monoxide, in which carbon monoxide is oxidized with atmospheric oxygen to carbon dioxide. Problems are encountered in this process step by the strong exothermy of the reaction along with considerable control efforts and the fact that due to the atmospheric oxygen the target component of fuel processing, hydrogen, is also catalytically burned. Nevertheless, the concentration of carbon monoxide can be reduced below 50 ppm by this reaction.

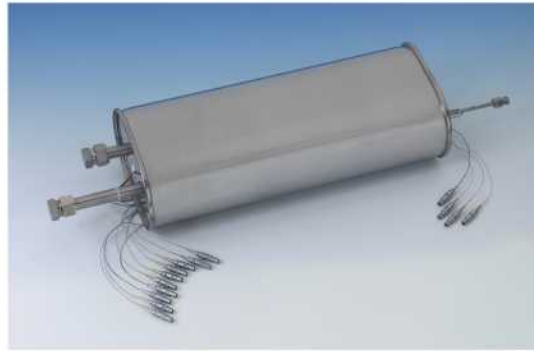


Fig. 3-18: WGS reactor of FZJ

3.3.2.3 Outlook on development goals in fuel processing

If the gas produced by reforming is to be fed into a low-temperature fuel cell, it must be cleaned of carbon monoxide as described above, which also arises during reforming for thermodynamic reasons. Cleaning is performed in a so-called water-gas shift reactor and in the reactor for preferential oxidation. The aim of future work at Jülich will be to interconnect these two reactors already developed and tested at Jülich with the actual reactor for reforming and to feed the then clean product gas into the fuel cell. At the Fuel Cell Seminar in San Antonio, USA, in 2004 these components were presented in the unconnected state. Fig. 3-19 shows the interconnected components.



Fig. 3-19: Interconnected components

3.3.3 Staff members and fields of activity

Name	Tel. (+49 2461-61-) e-mail address	Field of activity
Dr. R. Peters	4260 ra.peters@fz-juelich.de	Head of the Fuel Processing and Systems department (BGS)
Dr. J. Pasel	5140 j.pasel@fz-juelich.de	Heterogeneous catalysis, reforming, gas conditioning
Z. Pors	5322 z.pors@fz-juelich.de	Modelling, diesel reforming
R.C. Samsun	4616 r.c.samsun@fz-juelich.de	Kerosine reforming, fuels
A. Tschauder	5400 a.tschauder@fz-juelich.de	Reactor development, reforming, gas conditioning

3.3.4 Important publications, PhD theses and patents

Important publications:

Pasel, J.; Cremer, P.; Wegner, B.; Peters, R.; Stolten, D.

Combination of autothermal reforming with water-gas-shift reaction – small-scale testing of different water-gas-shift catalysts

Journal of Power Sources, 126 (2004), 1/2, 112 – 118

The concentration of carbon monoxide (CO) in the product gas of autothermal reforming (ATR) can be significantly reduced by means of the water-gas shift (WGS) reaction. It is possible to directly feed the reformat from ATR into the WGS reactor without a deactivation of the catalyst. For this purpose, it must be ensured that the reformat does not contain any constituents that are detrimental to the catalyst of the WGS reactor, such as higher aliphatic or aromatic hydrocarbons.

Pasel, J.; Meissner, J.; Cremer, P.; Pors, Z.; Peters, R.; Stolten, D.

Steady-state and dynamic tests of an autothermal diesel reformer with a power of 3 kW_{el}

XXXVII. Jahrestreffen Deutscher Katalytiker, Weimar: proceedings. 2004, 327 – 328 (in German)

The basis for the successful application of the process of autothermal reforming (ATR) is the selection of a suitable catalyst. At IWV-3, extensive tests have been carried out leading to the design, manufacture and experimental investigation of a reformer that provides a molar hydrogen flow corresponding to a power of 3 kW_{el} in a fuel cell. This paper presents the experiments on the steady-state and dynamic behaviour of this reformer and deals with the results on modelling the mixing chamber of the reformer.

Pasel, J.; Meißner, J.; Pors, Z.; Palm, C.; Cremer, P.; Peters, R.; Stolten, D.

Hydrogen production via autothermal reforming of diesel fuel

Fuel Cells (2004), in press

Hydrogen for the operation of a polymer electrolyte fuel cell (PEFC) can be produced with the aid of autothermal reforming (ATR) of liquid hydrocarbons. The reformer should preferentially be operated in the temperature range between 700 °C and 850 °C and at $n(\text{O}_2)/n(\text{C})$ ratios of 0.43 – 0.47 and $n(\text{H}_2\text{O})/n(\text{C})$ ratios of 1.7 – 1.9. Under these conditions, the conversion of hydrocarbons is complete. The reformer can be brought to its starting temperature of 340 °C within 7 min.

Peters, R.; Meißner, R.

Gas Conditioning for Fuel Cells

Chemie Ingenieur Technik, (2004), in press

The article gives a compact overview of the processes of gas conditioning and their state of the art. The focus of the brief report is on CO removal from the product gas of the reformer.

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

Autothermal Reforming of Diesel

Der Internationale Deutsche Wasserstoff Energietag 2004, Essen: proceedings 2004, CD-ROM

A possible application for fuel cells are compact power generating units – so-called auxiliary power units (APUs) – in the power range of 1 -10 kW_{el}. APUs can be found in all means of transport ranging from passenger cars through trucks, trains, aircraft up to ships and in stationary applications in domestic engineering and the leisure sector. The autothermal reformer exhibits a high power density, almost complete conversion for operation with diesel and a short start-up time. For the gas conditioning units, microstructured components coated with catalyst have been investigated and evaluated. The article describes a comparison of current developments with the target data of the Department of Energy for Fuel processing.

PhD theses:

Stalling, J.

Evaluation of the application of microstructures in mobile fuel processing units

Reports of Research Centre Jülich, 4120, RWTH Aachen, 2004 (in German)

In this thesis, the application of microstructures for heat exchangers and reactors in the gas treatment system of a mobile fuel processing unit was experimentally and theoretically investigated. Fuel processing is based here on the catalytic, autothermal reforming of diesel-like fuels. The experimental work involved simplified experimental setups and initial prototypes of microchannel heat exchangers. The theoretical part dealt with the fluid dynamic modelling of the transport phenomena that are relevant in microstructured apparatus. The thermohydraulic characterization shows a sharp increase in pressure loss if the length of the structure is reduced. The transition from laminar to turbulent flow in small structures is observed below the value commonly specified for the determining dimensionless number. Optimum structural dimensions are in the range between 0.4 and 1 mm. Application-oriented, catalytic investigations

were performed for all steps in the gas treatment process. The catalysts for the shift reaction generally show a moderate activity. The precious metal catalyst used for selective CO oxidation shows a very narrow temperature window with moderate activity for optimum reactor operation. H₂ oxidation is possible at high space velocities, low reaction temperatures or low loadings with catalytically active component. For CH₄ oxidation clearly longer residence times, higher reaction temperatures and higher loadings with active component are needed to accomplish the complete conversion required. For all reactions, an improved conversion temperature profile can be observed in smaller structures. Simulations using a model that includes the diffusive mass transport in microstructured apparatus as well as calculations based on experimentally determined maximum catalyst loadings illustrate that a direct combination of heat transfer with chemical reaction in an apparatus is only meaningful and possible with clearly more active catalysts.

Patents:

Patent applications:

Principal inventor	PT	Title
Peters, Dr., R.	1.2168	Mischkammer für einen Reformer sowie Verfahren zum Betreiben derselben
Pors, Z.	1.2169	Mischkammer für einen Reformer sowie Verfahren zum Betreiben derselben

3.4 Fuel cell process engineering

3.4.1 Objectives and fields of activity

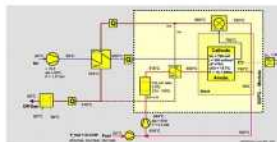
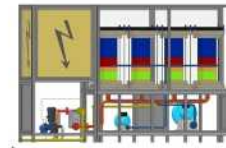
The working group for Fuel Cell Process Engineering is concerned with the design and construction of test facilities and their components and with the process-engineering systems analysis for high-temperature fuel cells of the SOFC type and low-temperature fuel cells of the DMFC type. The main fields of work are:

Testing of plant components: For the specific boundary conditions in conjunction with the operation at temperatures between 600 and 900 °C, the essential components of an SOFC plant such a recuperative heat exchanger, prereformer or afterburner are not commercially available. For this reason, test facilities are set up, in which products from outside and internal developments are characterized and examined for their suitability.



Testing and optimizing the control and regulating concepts of SOFC and DMFC plants: Fuel cells require complex plant and process engineering. In order to be able to reliably operate the plants, safely control the different operating regimes and prevent damage to the plant and fuel cell, special control and regulating concepts are required. These are evolved on the basis of experience acquired in the testing of cells and stacks and tested and further developed in the existing plants.

Development and construction of plants: In order to demonstrate the feasibility of the technology and test the interplay of all components, an SOFC demonstration plant is being developed and built, which will have an electric power output of 20 kW in natural gas operation. This plant technology is examined and further optimized by the development and construction of DMFC plants of up to 5 kW.



Calculation and evaluation of plant concepts: Based on experimental experience, steady-state and dynamic simulation calculations are carried out using different tools, with which different plant concepts can be evaluated and individual plant components designed. The plants are analysed both with

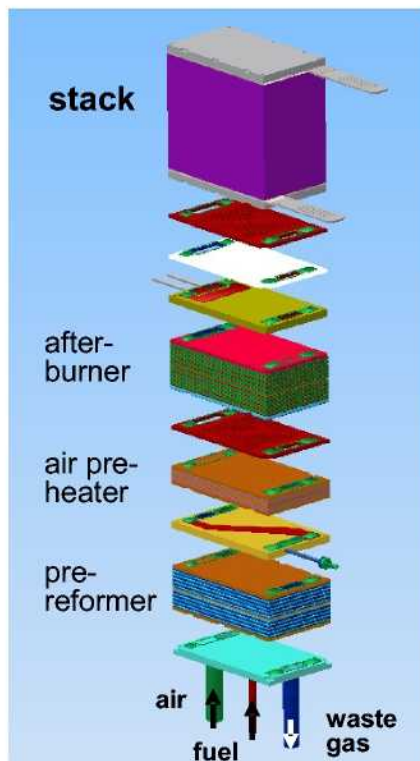
respect to their efficiency and concerning their costs.

CO₂ sequestration: In addition to fuel cell plants, other power plant types are also investigated specifically with respect to the possibilities of CO₂ separation.

3.4.2 Important work results

3.4.2.1 Process engineering for high-temperature fuel cells

In the field of SOFC plant technology, the essential goal in 2003 and 2004 was to develop a design concept for the 20-kW plant and to design and qualify the associated components. Based on process-engineering systems analyses, a plant concept as simple as possible was derived. The aim for this first plant is an electrical net efficiency of approx. 40 % and a thermal efficiency of at least 30 %.



In order to reduce the construction volume and the complexity of the plant and to minimize piping and heat losses, an "integrated modular concept" has been developed, in which the high-temperature plant components, separated by a flat gasket, are stacked on top of each other (see Fig. 3-20). For this purpose, a special planar design was developed for the prereformer, recuperator and afterburner (see Fig. 3-21) based on the external dimensions of the stack and its manifold geometry. For the development and qualification of these components, test rigs were designed and set up, in which components can be tested for an electrical stack power of 5 kW. Various measurements with the prereformer show that the reforming performance is completely sufficient for the planned application. Based on the measurement results and on plant simulation calculations, a required fuel gas outlet temperature from the reformer of approx. 500 °C was determined, at which the desired reforming rate of 20 % will then be achieved.

Fig. 3-20: Integrated modular concept

A 1000-hour long-term test did not show any perceivable ageing. These tests confirmed the suitability of the design. The same applies to the planar afterburner, which was also characterized and qualified in a corresponding test rig. Measurements of the exhaust gas composition revealed that no NO_x and only very small quantities of CO are contained, which are well below the limit values for heating plants with less than 70 kW thermal power.

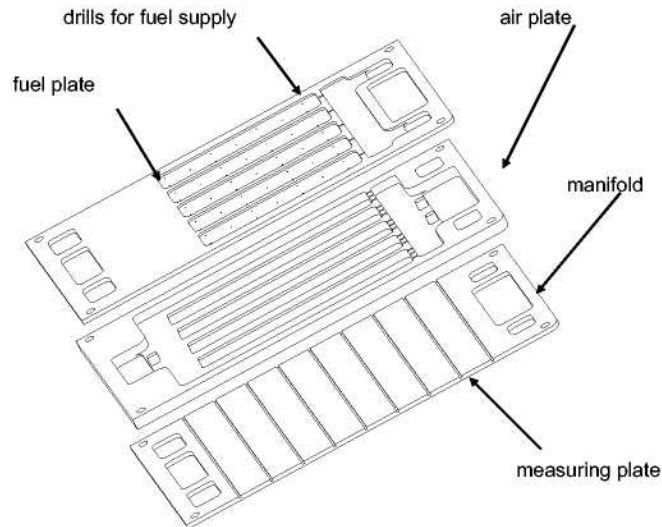


Fig. 3-21: Planar afterburner with measurement plate for determining the temperature profile

For the module constructed from these components, a concept has been developed which permits the removal and transport of the joined stack with mechanical bracing without having to relieve the critical solder glass joining. In final assembly, tied rods and springs can be applied at room temperature, so that creep is no longer a problem and low-cost materials can be used.

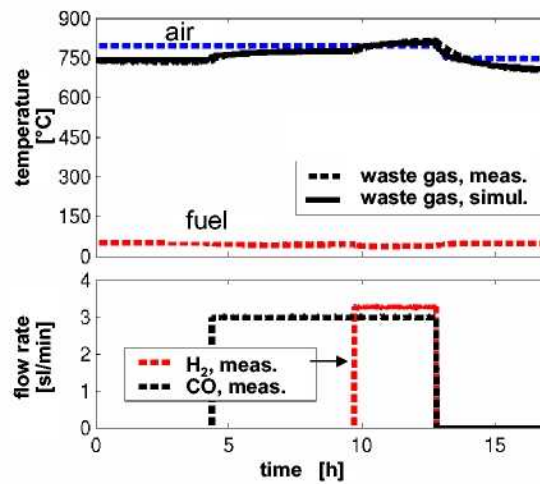


Fig. 3-22: Dynamic simulation of the afterburner – variation of exhaust gas temperature with sudden addition of CO and H₂

In parallel to this experimental work, computer models based on Matlab/Simulink are being developed for these plant components, with the aid of which the dynamic plant performance can be simulated. Dynamic plant modelling was verified using the measured data available, so that the performance of the essential components can be calculated within less than 5 % (see Fig. 3-22). The heating, cooling and load change performance was simulated for the planned plant. The results serve to optimize plant design and plant control.

3.4.2.2 Process engineering for low-temperature fuel cells

In the field of DMFC system technology, the essential aim in 2002 was the realization of a first functional 2-kW system and the development of an optimized system concept on this basis. An autonomous 2-kW DMFC system composed of a 2.5-kW DMFC stack and associated peripherals was successfully completed and presented at the Fuel Cell Seminar in Palm Springs and at Hanover Trade Fair to the general public. No external water supply was needed due to an integrated water management. The system was CE-certified as one of the first FZJ in-house facilities.

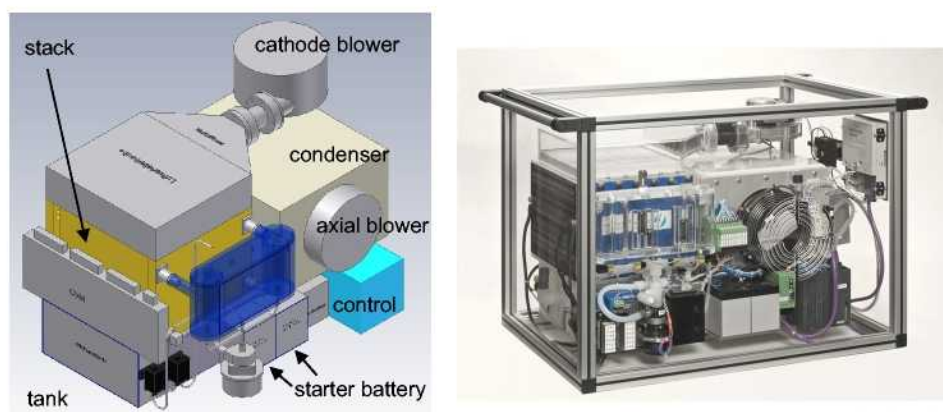


Fig. 3-23: Portable 1-kW DMFC module : schematic (left) and photograph (right)

The system was further developed, in particular, with a view to clearly increased power density. The construction volume of the new 1-kW system (Fig. 3-23) is essentially determined by the components stack, condenser and methanol/condensate tank. The greatest difference in comparison to the DMFC system from 2002 is the configuration of the cathodic system part. In the new system, air supply takes place via open cathode flow fields at the top of the stack. The cathode blower is thus connected to an air distributor, which encompasses the entire stack top. The exhaust air correspondingly escapes from the entire stack bottom. The stack is tightly screwed onto a tray in which the water leaving the stack in the liquid state is collected. The exhaust air is diverted in the upper tray region and enters the condenser also tightly screwed onto the tray from below. The liquid water produced in the condenser also flows into the tray. The dehumidified exhaust gas leaves the condenser together with the cooling air flow.

The anodic system has been considerably improved and simplified in comparison to the 2002 system:

- the inlets and outlets of the stack are now located in a single end plate
- there is no heat exchanger in the anode system
- the pipe length and number of fittings are clearly reduced

The total weight is approx. 60 kg. At a net power of 1000 W, the weight-related power density is approx. 17 W/kg and the volume-related power density about 9 W/l in comparison to 3.8 W/kg and 1.3 W/l, respectively, in the system from 2002.

3.4.3 Staff members and fields of activity

Name	Tel. no. (+49 2461-61-) e-mail adress	Field of activity
Prof. L. Blum	6709 l.blum@fz-juelich.de	Head of the Fuel Cell Process Engineering department (VBZ)
Dr. S.F. Au	5291 s.f.au@fz-juelich.de	SOFC plant simulation, component development and testing
M. Finkenrath	2902 m.finkenrath@fz-juelich.de	Simulation and analysis of the dynamic performance of SOFC power plants
Dr. H. Janßen	5082 h.janssen@fz-juelich.de	Head of the DMFC system development and testing group, DMFC component development, system design, system development and testing, electrolyser development and testing
Ro. Peters	4664 ro.peters@fz-juelich.de	SOFC component development, plant design, plant construction and testing
Dr. E. Riensche	6689 e.riensche@fz-juelich.de	Plant simulation, power plant technology, reforming, CO ₂ sequestration

3.4.4 Important publications, PhD theses and patents

Important publications:

Blum, L.; Lokurlu, A.; Riensche, E.; Vitale, A.; Bakke, K.*; Heidug, W.*; Li, K.J.*; Huijsmans, J.P.P.*

Conceptual Design Study of a 20 MW CO₂-Separating SOFC/ECAB/GT Plant

Proceedings of the 6th European SOFC Forum, Luzern/Schweiz, 28 Juni - 2 Juli 2004, 218-228

CO₂ sequestration projects have been implemented in Norway and Canada and are starting in Germany and Japan. This opens up opportunities for new power generation technologies like the CO₂-separating solid oxide fuel cell (SOFC), for instance of the tubular cell type with integrated methane reformer. One challenge is the development of integrated pressurized hybrid SOFC-gas turbine (GT) systems with high electrical efficiency. For carbon capture the additional challenge is the development of a modified SOFC system including an electrochemical afterburner (ECAB) to reach high CO₂ concentrations in the exhaust gas, which are a prerequisite for subsequent CO₂ liquefaction. The potential of such a technology – not expected to be available in the very near future – is investigated in this paper. A conceptual design study of a 20 MW SOFC/ECAB/GT plant is carried out. The special features of the process are discussed. Variants with different pressure levels and different ECABs (second SOFC, mixed oxide conductor, oxygen pump) are energetically analysed. Finally, a cost analysis and ranking of the variants is carried out in order to identify the most important cost driving components.

Blum, L.; Peters, R.; David, P.; Au, S.F.; Deja, R.; Tiedemann, W.

Integrated Stack Module Development for a 20 kW System

Proceedings of the 6th European SOFC Forum, Luzern/Schweiz, 28 Juni - 2 Juli 2004, 173-182

FZJ is currently executing a 20 kW SOFC pilot plant project. In this context, a new 'hot' parts packaging concept has been designed. 'Hot' here denotes all BOP components with an operating temperature above 400 °C. Pre-reformer, air pre-heater, afterburner and stack are integrated into a single package that is thermally insulated and can be operated in self-sustaining mode. The package is held together by spring-loaded bolts located outside the insulation. The spring loads ensure sealing and contacting pressure throughout the stack. The unique packaging concept reduces total volume and weight by avoiding unnecessary piping. Thermal mismatch is reduced by manufacturing all components from the same ferritic chromium steel as used for the interconnect plates of the SOFC stack. The high level of component integration results in a highly efficient heat transfer but also a complex interaction of the processes in the packed system. All components are scaled to an SOFC stack with a nominal power of 5 kW, based on a cell size of 20 x 20 cm². In this contribution, the development and testing of the components as well as the layout and design of the system for a 20 kW pilot plant is presented.

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

SOFC Worldwide – Technology Development Status and Early Applications

Proceedings of the NATO ARW – “Fuel Cell Technologies: State and Perspectives” June 06. – 10. 2004, Kyiv

Solid oxide fuel cells (SOFCs) of various types and designs have been developed worldwide through the last two decades. They offer interesting advantages over other fuel cell types, but also have inherent materials problems that have caused a slower development pace as compared, for instance, to the low-temperature polymer electrolyte fuel cell (PEFC). Due to their high operating temperature in the range of 700 to 1,000 °C, SOFCs can be used with a variety of fuels ranging from hydrogen to hydrocarbons with a minimum of fuel processing, can be coupled with gas turbines for the highest electrical system efficiency known in power generation, deliver process heat in industrial applications or supply on-board electricity for vehicles, to name but a few typical applications. This report summarizes the more prominent SOFC development strands and gives an overview of the achievements of the various R&D groups. The analysis includes a benchmark that attempts to compare cell and stack characteristics on a standardized basis.

Finkenrath, M.; Lokurlu, A.; Blum, L.; Stolten, D.

Modelling the Dynamic Behaviour of a Planar SOFC CHP System

Proceedings of the 6th European SOFC Forum, Luzern/Schweiz, 28 Juni - 2 Juli 2004, 569-578

For the analysis of transient operation of a solid oxide fuel cell (SOFC) system, such as start-up, load change or shut-down, a system-level dynamic simulation is required. This paper focuses on the development of dynamic models for all SOFC system components using a cascade of continuous stirred tank reactors as a basic modelling element. Modelling details concerning heat transfer or reaction kinetics are considered for individual components. All component models were validated with experimental data. Validation results presented for an air preheater and a reformer show reasonable agreement between measurements and simulations. To demonstrate system-level dynamic simulation, a heat-up simulation of a complete SOFC system is shown.

Janßen, H.; Bringmann, C.J.; Emonts, B.; Schröder, V.

Safety-related studies on hydrogen production in high-pressure electrolyzers

International Journal of Hydrogen Energy, 29 (2004), 759-770

High-pressure electrolysis makes special safety demands on pressure vessel design and operation management. This paper presents design approaches that comply with the safety requirements despite weight minimization. Furthermore, investigation results are presented, which describe the determination of the pressure- and temperature-dependent explosion limits of H₂/O₂ mixtures and of product impurities in electrolyser operation at high pressures of 100 and 200 bar. The results show that due to the high diffusion coefficient of hydrogen its concentration in product oxygen increases at most up to 40 % of the lower explosion limit at 200 bar.

Janßen, H.; Bringmann, J.C.; Emonts, B.; Schroeder, V.

Safety-Related Studies on Hydrogen Production in High-Pressure Electrolysers

International Journal of Hydrogen Energy, Volume 29, Issue 7, July 2004, 759-770

At Research Centre Juelich the prototype of an alkaline 120-bar electrolyser has been developed and built. Constructive and process-engineering measures must be taken to ensure the safe operation of such facilities. Potential hazards occur due to the high operating pressure in conjunction with the reactivity of the product gases and the electrolyte. First of all, the operating mode and technical features of the Juelich high-pressure electrolyser will be dealt with. Within the framework of a parametric study, the potential for weight reduction of the prototype while observing the rules for pressure vessel design will be shown. The Federal Institute for Materials Research and Testing in Berlin has performed measurements concerning the explosion limits of H_2/O_2 mixtures at different temperatures and pressures up to 200 bars. At an electrolysis test rig of IWV-3, which can also be operated up to 200 bars, investigations were carried out concerning the gas composition on the H_2 and O_2 path under different operating conditions. These measurement series were compared to the explosion limits determined and evaluated to derive safety measures required for the operation of high-pressure electrolysers.

Janßen, H.; Mergel, J.; Nölke, M.; Schmitz, H.

Water Management in DMFC and its Impact on Performance and System Design

Proceedings of the 9th Ulm ElectroChemical Talks, Ulm, 17.-18.05.2004

In this paper we describe and discuss the impact of the DMFC water management on the stack and system performance as well as on the system configuration and sizing. In an advanced DMFC stack model we consider the power limitation caused by water flooding. It can be shown that the maximum current density of the stack is correlated to the maximum water capacity of the cathode air flow. We will also show that the heat and water balance of a DMFC stack plays a drastic role for the overall performance of the system. The water permeation in single cells was studied as a function of temperature, air stoichiometry and composition of the backing layer. Hydrophobization improves the transport of water and air diffusion in the cathode. The model is validated by experimental data. To achieve a water-autonomous DMFC system a big effort must be made for water recovery. Water permeation through the membrane and evaporation at the cathode side leads, on the one hand, to internal heat extraction, so that there is no additional need for a cooling system. On the other hand, a large amount of water will be discharged via flue gas. To recover the water which is lacking at the anode side a condenser must be integrated. The paper will contain aspects for the sizing of compact DMFC-specific condensers.

Lokurlu, A.; Riensche, E.; Thom, F.; Stolten, D.

CO₂ Separation in Modified SOFC Systems and Possible CO₂ Reduction Potentials by Biological Fixation in Algae

VDI Reports No. 1714, 2002, 217-227

technologies to reduce the amount of CO₂ that is released to the atmosphere. One possibility is the fixation of CO₂ involving CO₂ sequestration, underground or in the ocean. Sequestration requires CO₂ liquefaction. It is shown that this is only feasible, if the CO₂ is concentrated to about 90 %. In common SOFC plant design several % of CO₂ are present in the exhaust gas depending on the air ratio in SOFC and afterburner. The high CO₂ concentrations can be reached with modified tubular SOFC bundles including an electrochemical afterburner. An SOFC power plant can also be combined with a biotechnological reactor for algae mass production. Mass and

energy balances show that the advantages of the high-temperature fuel cell match with the demands of the algae production unit. All products of the SOFC can be used: electric power for driving the pumps recirculating the culture medium, CO₂ and H₂O for the growth of the algae during daylight and high-temperature heat of about 200 °C for the drying apparatus.

PhD theses:

Prakash Chandra Ghosh

Cost optimisation of a self-sufficient hydrogen based energy supply system

Reports of Research Centre Jülich, 4049, RWTH Aachen

The overall goal of this thesis has been to develop an optimisation method for a hydrogen based renewable energy supply system. The cost-effective application of such a system is also analysed. The main cost intensive components, used in the system are optimised on the basis of the lifetime and the investment cost of the different components. The main cost intensive components are the renewable energy converter, the battery, the electrolyser, the gas tank and the fuel cell. The sizes of the first four components are interdependent. A comparative analysis of hydrogen and diesel generator based renewable energy supply system is performed.

Important Patents:

Patent applications:

Principal inventor	PT	Title
Thom, F.	1.1990	Kompakte Bioreaktor-Brennstoffzellen-Anlage
Blum, Prof., L.	1.2053	Hochtemperatur-Brennstoffzellensystem
Au, Dr., S.F.	1.2094	Verfahren zur Erzeugung elektrischer Energie mit Hilfe einer Festelektrolyt-Brennstoffzelle
Lokurlu, Dr., A.	1.2122	Verfahren zur Erzeugung von Strom und Wärme
Nölke, M.	1.2174	Kathodendiffusionsschicht einer Brennstoffzelle
Blum, Prof., L.	1.2175	Gasverteilerplatte für eine Hochtemperatur-Brennstoffzelle
Janßen, Dr., H.	1.2179	Brennstoffzellenstapel sowie Verfahren zum Betreiben eines solchen

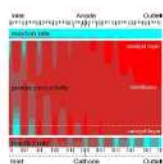
Patents granted:

Principal inventor	PT	Title
Thom, F.	1.1663	Brennstoffzelle mit modularen, flexiblen Gasverteilerstrukturen
Thom, F.	1.1715	Kühlsystem für Brennstoffzellen
Peters, Ro.	1.2041	Verspannung eines Hochtemperatur-Brennstoffzellenstapels

3.5 Physico-chemical fundamentals

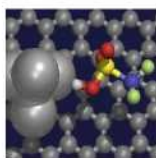
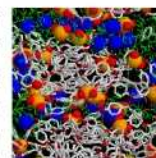
3.5.1 Objectives and fields of activity

For a successful development of fuel cell components, cells and stacks comprehensive knowledge of the material properties and of the complex interactions between materials is required. Apart from experiments, theory and computer modelling also essentially contribute to the basic understanding of structures and processes in fuel cells. The focus of institute activities is currently on PEFCs and DMFCs in the low-temperature range. The aim of physical and physico-chemical investigations is the basic understanding of structures and processes on different time and length scales, on the one hand, and the use of these findings for the further development of PEFCs and DMFCs, on the other. For this purpose, atomistic and continuum simulation methods are being developed and used.



Physical continuum simulation methods are used to develop numerical and approximative analytical PEFC and DMFC models. Emphasis is placed here on the interpretation of properties observed in fuel cell operation, the characterization of cells on the basis of experimental power curves and the prediction of the optimum working conditions of a single cell.

On the molecular level, models for proton transport in polymer electrolytes are developed and simulated using the classical molecular dynamics method. The simulation calculations permit conclusions to be drawn concerning the atomic mechanisms of proton transport. They additionally allow the determination of proton mobility as a function of fuel cell operating parameters or the chemical structure of the polymer.



Ab-initio molecular dynamics simulations are used to investigate the elementary steps of the anodic oxidation of methanol on noble metal catalysts. By taking the ambient effect on the electronic structure of reactants and products into account, mechanistic details of the electrocatalytic reactions in liquid-operated direct methanol fuel cells can thus be investigated.

Essential aspects of work will also be in future to overcome the gap between the time and length scales of continuum simulations and an even closer interlinkage with experiments.

3.5.2 Important work results

3.5.2.1 Continuum simulations of PEFC and DMFC

Hydrodynamic modelling is an indispensable tool for interpreting the functioning of fuel cells and improving cell designs. The models describe the fuel cell on length scales from micrometres (the thickness of the cathode layer) up to a metre (the length of a gas channel); the processes on the smaller time scales are integrated into effective kinetic and transport coefficients. Instead of comprehensive modelling with a complex three-dimensional model, the approach of hierarchical modelling is being pursued, in which three-dimensional numerical models are combined with mostly one-dimensional analytical models. In many situations, simple analytical models lead to a basic understanding of the processes. They

often also provide more reliable results than extensive three-dimensional models with many, in part only poorly determinable, parameters and a considerable demand for computer resources. On the other hand, the three-dimensional models are indispensable for a detailed picture of the cell function; moreover, they validate the simpler models.

Fig. 3-24 shows a function card of a PEFC as an example of three-dimensional modelling. The strong inhomogeneity in the direction perpendicular to the cell surface (x-axis) is caused by inhibitions of the proton and reactant transport. The inhomogeneity in the other direction is determined by gas consumption and by the meandering structure of the flow field. In the case of high currents and low stoichiometric gas flow, these inhomogeneities have a considerable influence on cell performance.

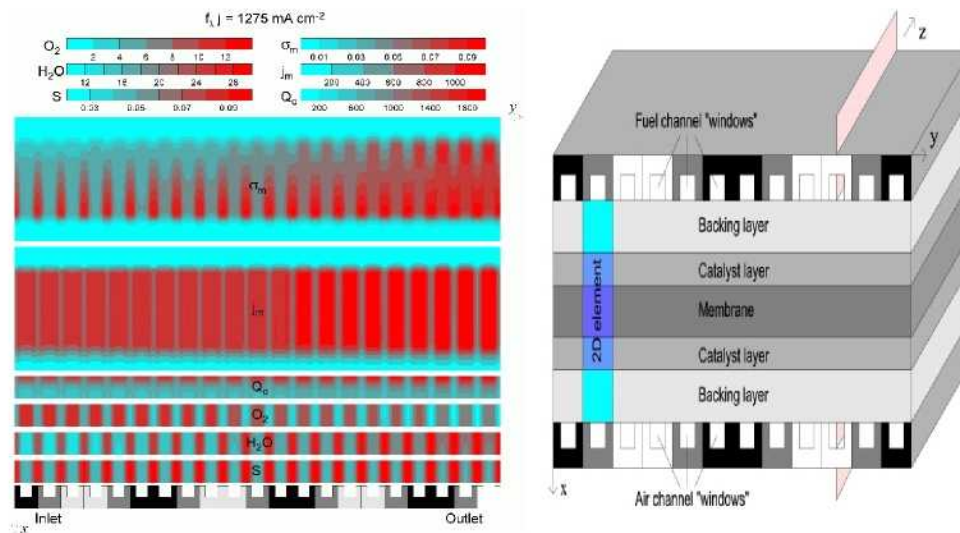


Fig. 3-24: Distribution of proton conductivity (σ_m), proton current density (j_m), O₂ reduction rate (Q_c) and concentration (O₂), water concentration (H₂O) as well as saturation (S) in a PEFC limited on both sides by three parallel meanders

3.5.2.2 Analytical modelling of PEFC and DMFC

The value of three-dimensional simulations is apparent, above all, if the parameters of significance for the performance of the fuel cell can be varied in them. For the identification of these key parameters and for the basic understanding of cell performance, analytical modelling is an indispensable tool, which will be demonstrated by two examples:

PEFC: In lifetime tests and also under real operating conditions, a PEFC frequently runs in the galvanostatic regime under constant load. Occasionally, fluctuations in cell voltage occur.

A simple analytical PEFC model, which contains the impact of finite oxygen stoichiometry on cell performance, can explain this effect. The model predicts below a critical threshold of oxygen supply two different values for cell polarization corresponding to two different oxygen distributions along the gas channel. The system oscillates between these voltage values with a frequency determined by the residence time of the air in the cell and ranging between 1 and 10 Hz. The cell voltages measured at the Institute and shown in Fig. 3-25 confirm the predicted occurrence (regime 2a) and disappearance (regimes 2b and 2c) of the oscillations.

In the course of time, the oscillation amplitude increases, whereas the mean cell voltage decreases. The model explains this behaviour by a progressing deterioration of the transport properties of the diffusion layer. It is the first model which relates the voltage fluctuations in normal operation to cell ageing and is thus a starting point for systematic theoretical ageing studies.

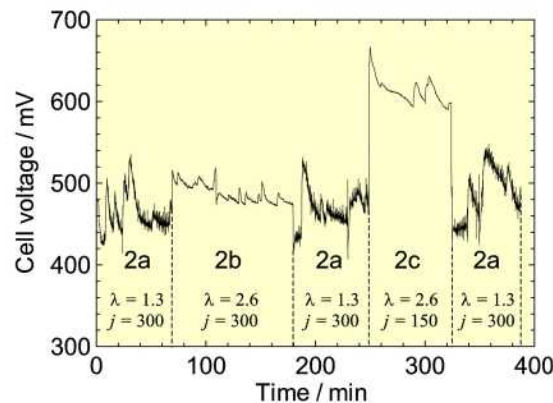


Fig. 3-25: Influence of critical oxygen flow velocity

DMFC The cell voltage of a DMFC decreases significantly even at a very low load. In the range of low current intensities, this decrease of about 500 mV is the greatest contribution to total loss. The causes for this phenomenon are largely not understood despite extensive studies. An analytical DMFC model explains this phenomenon as a consequence of methanol permeation through the membrane. The model predicts the existence of a narrow "bridge", which "short-circuits" the electrodes even at very low current and which does not exist in the absence of crossover. The model permits the calculation of the voltage loss due to that bridge, and numerical estimates show that the order of magnitude of this value is in agreement with experiments.

Fig. 3-26 compares the modelled polarization curves with those measured at the Institute. The rhombi indicate the beginning of bridge formation. The bridge manifests itself in the model as a plateau at low current densities, whose width decreases systematically with increasing oxygen stoichiometry. The experiment confirms this effect: it shows an even more drastic change in ascent for the current-voltage curves compared to the model at a characteristic current density whose value decreases with increasing stoichiometry in the way predicted by the model (rhombi).

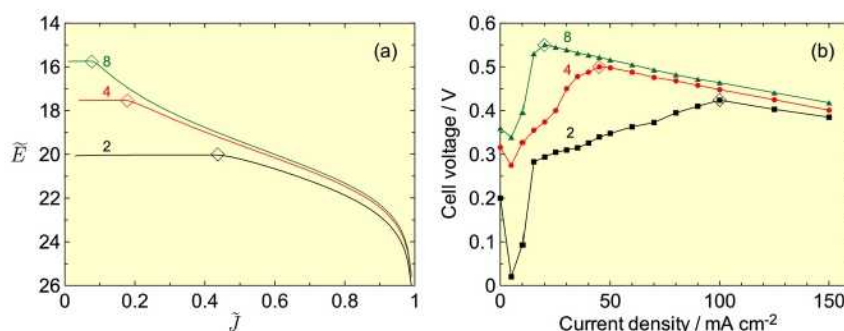


Fig. 3-26. (a) Model polarization curves (in dimensionless units) for the specified values of oxygen stoichiometry and (b) experimental polarization curves of a DMFC

3.5.2.3 Molecular dynamics of proton transfer in polymer electrolytes

The protonic conductivity in polymer electrolyte membranes (PEMs) essentially contributes to the performance characteristics of a low-temperature fuel cell. The largely accepted interpretation of the molecular proton transport mechanism in sulphonated polymer electrolyte membranes (PEMs) starts out from the idea that a bicontinuously nanophase-separated material is produced while taking up water from the polymer. The coherent polymer network serves to obtain mechanical stability, whereas the equally continuous aqueous pore network is responsible for the proton conductivity observed. It is furthermore assumed that proton transport in an individual pore results from the interaction of liquid-like (bulk-like) transport, in which the protons jump from one water molecule to the next, and surface transport, in which the protons migrate from the environment of one sulphonic acid group of the polymer to the environment of the next. Using this picture, the experimentally observed increase in proton mobility with increasing water content and decreasing equivalent weight can thus be explained or else the change in activation energy from a low liquid-like value in wet membranes to a higher value in dry membranes due to an elevated fraction of surface transport.

On the basis of these considerations, molecular models for proton transfer in porous PEMs have been developed. An essential result of the molecular dynamics simulations of individual pores performed with these models, which with the exception of the increased activation energy in dry membranes describe most experimental properties qualitatively correctly, was that surface and bulk mechanism cannot be regarded independent of each other. It was concluded that the change of the proton transfer mechanism is not attributable to a change of transport in an individual pore, but is the result of changes in the pore network. It was speculated that the aqueous phase in dry membranes is only intermediately continuous due to fluctuatively formed liquid bridges which rapidly disappear again.

Further simulations of polymer networks qualitatively confirm this hypothesis. Fig. 3-27 shows snapshots of a Nafion membrane at low humidity, in which the visualization of the aqueous regions (blue: proton-rich and yellow: sulphonate-group-rich regions) and of the

polymeric regions (red) always hides a multitude of atoms under the surfaces. At the locations indicated by the arrow an aqueous bridge is formed and disappears twice. Proton transport in these locations is only possible during the existence of the bridge. The simulations permit the identification of further modes of polymer motion favouring proton transport. The insights into desirable local properties of the polymer thus obtained by refining the motion model are incorporated into the development of new polymer materials.

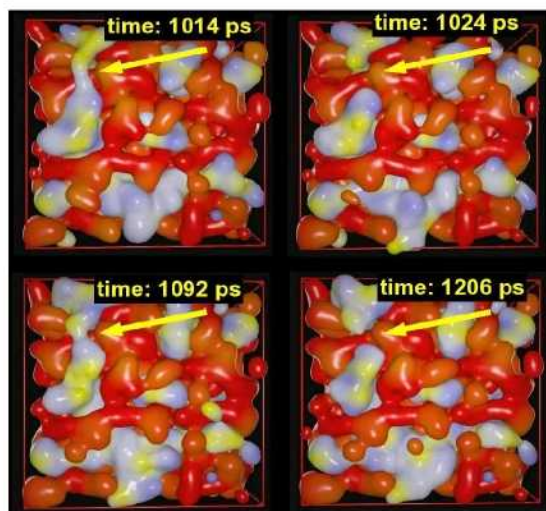


Fig. 3-27: Snapshots of the simulation of a Nafion membrane with a low water content of $\lambda=5$

3.5.3 Staff members and fields of activity

Name	Tel. no. (+49 2461-61-) e-mail address	Field of activity
Prof. Dr. E. Spohr	6687 e.spohr@fz-juelich.de	Head of the Physico-Chemical Fundamentals department (PCG) Investigation of the elementary steps of electrocatalytic and transport processes using quantum mechanics and molecular dynamics simulation methods. Understanding of the correlations between atomic and material properties.
Prof. Dr. A. Kulikovsky	5396 a.kulikovsky@fz-juelich.de	Analytical and numerical models of PEFCs and DMFCs. Interpretation of experimental data. Search for novel cell designs and for improved operating conditions of current designs.

3.5.4 Important publications, PhD theses and patents

Important publications:

Kulikovsky, A.A.; Scharmann, H.; Wippermann, K.

On the origin of voltage oscillations of a polymer electrolyte fuel cell in galvanostatic regime.

Electrochem. Comm., 6 (2004) 729

The paper compares experimental and theoretical investigations of the voltage oscillations that occur in a PEFC under galvanostatic conditions below a critical value of cathode air supply. The physical reason for this behaviour is the fact that two possible polarization voltages exist for the same average current density at low air supply. The model allows the determination of the critical values and, moreover, explains the time dependence of the average voltage and the oscillation amplitudes.

Kulikovsky, A.A.

Semi-analytical 1D+1D model of a polymer electrolyte fuel cell

Electrochem. Comm., 6 (2004) 969; *The effect of cathode water on performance of a polymer electrolyte fuel cell. Electrochim. Acta* 49 (2004) 5187

The first quasi-two-dimensional semi-analytical PEFC model with water management is described. The model explains the recently published along-the-channel profiles of local current density. It predicts the formation of a parasitic proton current in the cell plane in the case of insufficient humidification of the air or oxygen flow. The model furthermore permits the estimation of an optimum air composition or, if this is not possible, of an optimum value of oxygen stoichiometry.

Kulikovsky, A.A.

1D+1D model of DMFC: localized solutions and mixed potential. *Electrochem. Comm.*, 6 (2004) 1259; A.A.Kulikovsky. On the nature of mixed potential in a DMFC

J. Electrochem. Soc. (2004, submitted). A.A.Kulikovsky, K.Klafki, K.Wippermann.
Experimental confirmation of the effect of bridge formation in a direct methanol fuel cell. Electrochem. and Solid—State Lett. (2004, submitted)

A novel bridging effect is described, which is based on the formation of a local "current bridge" that short-circuits the DMFC electrodes. The model shows that the bridge is generated by methanol crossover; it already causes a high overvoltage at very low average current densities. Numerical estimates provide values between 500 and 700 mV for the voltage loss, which is in agreement with many experiments. The bridge disappears with increasing oxygen and methanol stoichiometry. The model predictions were confirmed by experiments.

Commer, P.; Hartnig, C.; Seeliger, D.; Spohr, E.

Modeling of Proton Transfer in Polymer Electrolyte Membranes on Different Time and Length Scales

Mol. Simulation, 30, 755-763 (2004)

Recent atomistic simulations of the proton transport in polymer electrolyte membranes are analysed with a view to multiscale understanding. The relation between the elementary steps of proton transport and macroscopic conductivity is discussed together with the different interaction mechanisms used, which cover time scales ranging from individual proton jumps up to the diffusion processes coupled to the polymer motion.

Kreuer, K.D.*; Paddison, S.J.*; E. Spohr, E.; Schuster, M.

Transport in Proton Conductors for Fuel Cell Applications: Simulations, Elementary Reactions and Phenomenology

Chem. Rev. 104, 4637-4678 (2004)

The study refers to the transport properties in different proton-conducting materials such as acids, oxides, polymer electrolytes and inorganic/organic compounds. It contains a critical review of the existing literature on proton transport (conductivity) and parasitic transport (electroosmotic drag of water and methanol crossover) from the aspect of the applicability of these materials in fuel cells. It moreover gives an overview of the theoretical tools and atomistic simulation tools that are suited for modelling these materials.

PdD thesis:

Commer, P.

Computer simulation and theoretical modelling of the proton dynamics of model pores of a polymer electrolyte membrane*Heinrich Heine University Düsseldorf, 2003 (in German)*

The influence of local molecular properties of the polymer on the proton transport in model pores of a polymer electrolyte membrane was investigated with the aid of molecular dynamics simulation and continuum theory. The aim was to derive structural proposals for an improved design of fuel cell membranes. For this purpose, the proton transport in idealized layer and cylinder pores of perfluorinated sulphonate-acid-functionalized ionomers was simulated. The polymer phase was modelled as a rigid pore-limiting surface with embedded functional sulphonate groups and side chains. Proton transfer between two water molecules was described within the framework of an empirical valence bond model (EVB model) developed at the Institute.

The simulations showed that the factor that influences proton mobility most strongly is the acid strength of the sulphonate group. Besides that, the mobility of the sulphonate group and, to a lesser extent, the mobility of the side chains also have a significant influence on conductivity. The simple model for molecular dynamics enables systematic investigations to be carried out as a function of operating parameters such as water content and temperature and of chemical properties of the polymer such as the equivalent weight of the polymer, i.e. on the proton density in the membrane, were reproduced by simulation. In contrast, the temperature dependence of proton conductivity and the activation energy thus obtained were independent of the water content, which is contradictory to the experiment. By comparison with a continuum-theoretical description it was shown that this is not an artefact of the EVB model used in simulation. Instead, indications were found for the fact that the change in pore shape and pore volume changes the activation energy. On this basis, the hypothesis was put forward that the dynamics of the polymer network is responsible for these observations.

Besides proton transport, some investigations were also carried out concerning the molecular properties that influence methanol crossover. The effect of the side chain length in the pores of a Nafion-like polymer on the rate of proton, water and methanol transport in an aqueous methanolic solution was investigated. It was found that longer side chains both favour proton transport and slow down methanol transport. Reduced crossover can therefore be expected for membrane polymers with longer side chains.

Important patents:

Patents granted:

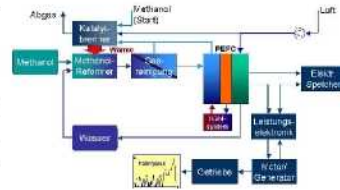
Principal inventor	PT	Title
Kulikovsky, Prof. Dr., A.	1.1661	Brennstoffzellen-Elektrode

3.6 Process and systems analysis

3.6.1 Objectives and fields of activity

An indispensable contribution to the development of energy conversion systems with fuel cells for mobile and stationary applications is the development of process concepts and the analysis of systems. In this context, the power trains for the traffic of tomorrow and those for stationary electricity production are analysed and evaluated. Systems involving conventional technology as well as fuel supply (fuel pathways) are also included in this analysis.

The results of the development of process concepts with the PRO II and SIMSCI computer programs for fuel cell systems for application in stationary and mobile energy conversion plants are reflected in process flow diagrams and net electricity balances. This is followed by a comparative process evaluation of competing energy conversion systems. Due to the relevance of the application conditions (e.g. high dynamic requirements for passenger cars), selected systems are subjected to a process evaluation in a detailed simulation concerning energy demand and emissions (tool: SIMBA) (example: APU for on-board power supply). The results that are available provide information about energy balances and emissions.

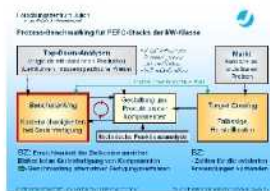


The above-mentioned technical analyses are extended by application-oriented peripheral considerations such as process chain analysis (tool: KRAKE) and cost analysis (tool: KOSTEX). For the same or a similar balancing frame, the entire process chain is evaluated. The results for mobile/

stationary applications are then statements on specific energy demand (MJ/km, MJ/kWh), specific emissions (g/km, g/kWh) and specific costs (€ cents/km, € cents/kWh).



Essential building blocks of process chain analysis are the production processes of future energy carriers or fuels. In the foreground of discussions are currently synthetic fuels such as synfuels (basis: fossil primary energy sources) or sunfuels (basis: biomass or biogas), which are each produced from synthesis gases. Process engineering is, moreover, described and evaluated with the aid of literature evaluations. As a supplement, building blocks of the process chains and entire competing processes are compared with a view to the criteria of emissions, primary energy demand, materials input and costs (benchmarking). Recommendations for process development and necessary R&D application can be derived from the results of these analyses.



3.6.2 Important work results

The focus of current work on process and systems analysis has been on the determination of process concepts for a fuel cell APU with a solid oxide fuel cell and on technology-based benchmarking for the determination of area-related material costs for PEFC stack components.

3.6.2.1 Process concepts and their evaluation

The procedure for establishing a process concept with subsequent evaluation will be described in the following by means of a system comparison of fuel cell APU (auxiliary power unit) and starter generator. Motor vehicles with internal combustion engine are to consume less fuel along with rising engine power and increasing convenience equipment. A relevant solution approach is targeted energy management for uncoupling electric units necessary for drive and convenience. This can be achieved by independent, powerful electricity supply, e.g. by an APU. Fig. 3-28 shows a process concept for such a fuel cell APU.

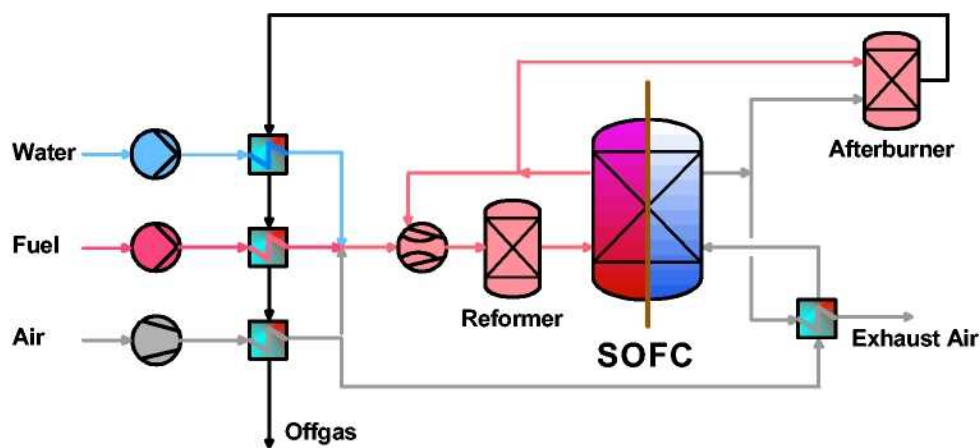


Fig. 3-28: Example of a simplified process concept for a fuel cell APU with a solid oxide fuel cell (SOFC)

The analysis is based on a system comparison of the present status of on-board power supply (generator/battery) with variants based on the crankshaft starter-generator and the fuel cell APU (solid oxide fuel cell, SOFC).

The results of process analysis define the efficiency potential of electricity generation with SOFC fuel cell systems. Fig. 3-29 shows the energy balance for the process concept presented using diesel fuel.

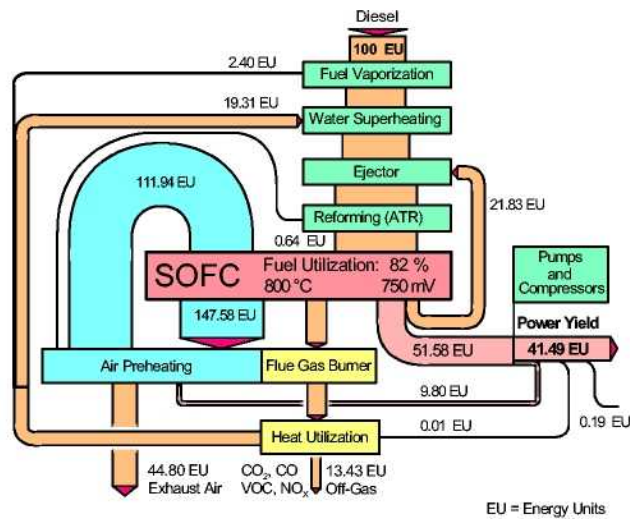


Fig. 3-29: Energy balance for the process concept presented using diesel fuel

Within the framework of model-based dynamic drive simulation (tool: SIMBA), load profiles are also defined for ancillary consumers in addition to the drive cycle, unless they result from vehicle operation proper. This ultimately gives the fuel consumption per kilometre covered in dynamic operation and the associated greenhouse gas emissions as shown by the example in Fig. 3-30 including fuel supply (tool: KRAKE).

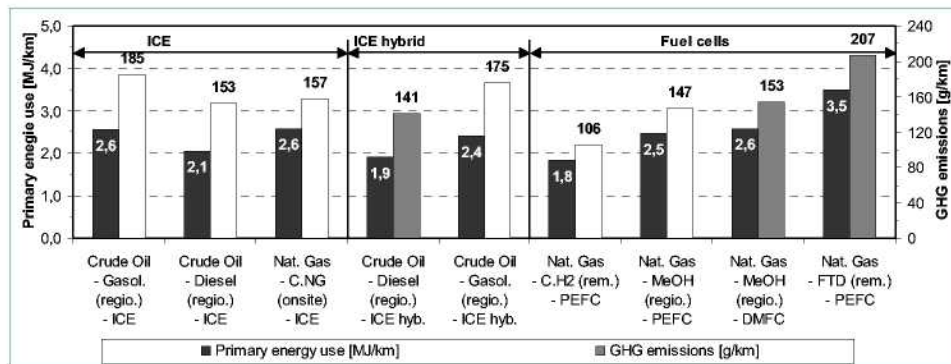


Fig. 3-30: Energy demand and greenhouse gas emissions of competing vehicle and fuel systems

The configuration status for a combustion engine drive with APU is shown in Fig. 3-31. Initial results show that at a cost that cannot yet be specified today APU fuel cell systems in competition with starter generators will in future provide the possibility of a more efficient power supply of the on-board electrical system in certain operating cases of combustion engine drives.

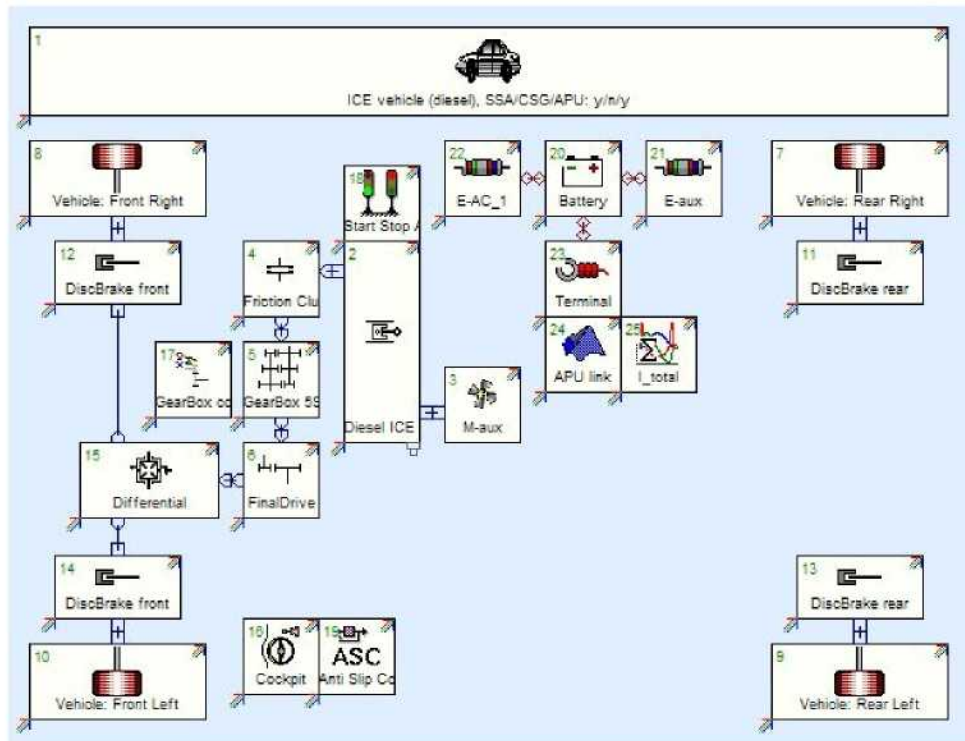


Fig. 3-31: Simulation model for a vehicle drive with internal combustion engine and APU

3.6.2.2 Technology-based benchmarking

Besides aspects of lifetime, efficiency and environmental compatibility, the question of the attainability of given cost targets is of paramount interest in fuel cell technology. Therefore, a techno-economic cost model was developed for the components contained in PEFC stacks with the aim of comparing different technical solutions with respect to their production costs – based on the concept of benchmarking management. Determining the mass-specific production costs takes on a key role. For this purpose, amongst others, a Monte Carlo approach is used, which provides expectation values and confidence intervals for the material costs of the stack components. The consideration of production costs incorporates fabrication conditions of individual components. This method of analysis, in conjunction with detailed cost analyses for common ionomers, is applied to the injection moulding of composite plates as one of several technologies for producing bipolar plates.

The result of a benchmarking of different polymer variants with the aid of a Monte Carlo simulation is, for example – on certain assumptions – probability distributions for area-related material costs as shown in Fig. 3-32.

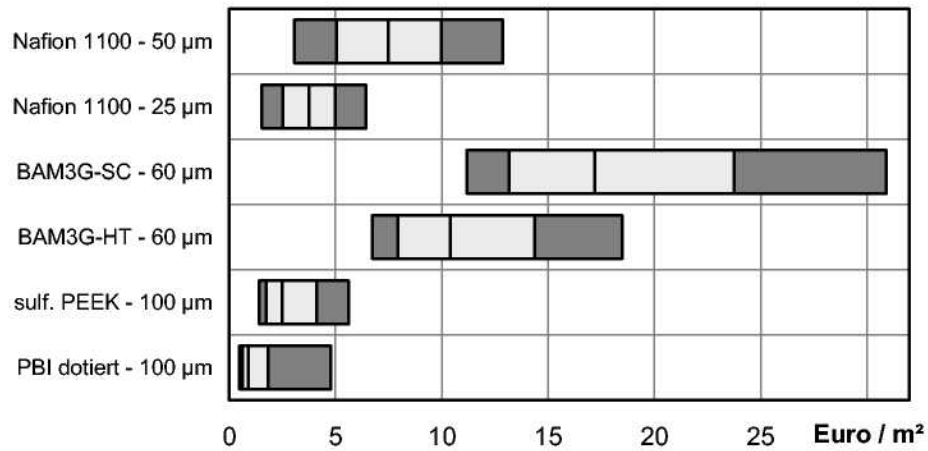


Fig. 3-32: Probability distributions of area-related material costs

Such material and process cost calculations accompanying development can contribute towards faster recognizing the cost potential of promising technologies for fuel cell materials and components. Existing resources for research and development can thus be geared more efficiently to the commercial success of the fuel cell.

3.6.3 Staff members and fields of activity

Name	Tel. no. (+49 2461-61-) e-mail Adresse	Field of activity
Prof. Dr. B. Höhlein	3235 b.höhlein@fz-juelich.de	Head of the Process and Systems Analysis department (VSA) Comparative systems evaluation
Th. Grube	5398 th.grube@fz-juelich.de	Power train simulation, process and systems analysis
R. Menzer	6708 r.menzer@fz-juelich.de	Process design and analysis

3.6.4 Important publications, PhD theses and patents

Important publications:

Emonts, B.; Höhle, B.; Stolten, D.

European, German and North Rhine-Westphalian hydrogen and fuel cell activities

Proc. Hydrogen and Fuel Cells 2004 Conference and Trade Show; 25.-28.09.2004, Toronto, Canada

Fuel cells are becoming established in many areas as an essential technology of the future. Of great significance is the energy carrier issue, since fuel cells predominantly need high-purity hydrogen. This requires a simultaneous discussion of hydrogen and fuel cell technology. On the part of the political institutions with responsibility at the European, German federal government or German federal state level it is indispensable to take relevant approaches into consideration in the strategic programmes for research, development and demonstration. In the first part, this paper will outline the essentials of the strategic orientation of hydrogen and fuel cell technology. In the second part, the respective initiatives in the field of hydrogen and fuel cells undertaken in Europe, Germany and North Rhine-Westphalia will be presented and their goals compared. The third and last part will give a survey of the different activities in the field of hydrogen and fuel cells.

Grube, Th.; Höhle, B.; Menzer, R.

Comparing Assessment of Future Fuel Supply and Use

International German Hydrogen Energy Congress 2004, Essen, Februar 2004

Highly efficient energy conversion systems with fuel cells for vehicles as well as for stationary and portable applications are currently being discussed all over the world. Fuel cell technology is expected to help reduce primary energy demand and emissions of limited and climate-relevant pollutants. The high flexibility of fuel cell systems with respect to energy carriers opens up possibilities of modifying the energy sector in the long term. Introducing new fuels based on low-carbon energy carriers can contribute to reducing greenhouse gas emissions as well as locally and regionally active atmospheric pollutants. The use of hydrogen on the basis of non-fossil, renewable energy carriers as fuel gas for fuel cells leads to special benefits with respect to conserving resources and climate protection, but at present still represents a medium- to long-term prospect. A major milestone on the road to market success for all energy conversion systems with fuel cells is the reduction of costs. The definition of the "appropriate" fuel represents a serious obstacle to the market introduction of fuel-cell-powered vehicles. Presenting data from a well-to-wheel analysis of various vehicle fuel systems at FZJ this article aims to discuss the potential benefits of future vehicle concepts with fuel cells in terms of primary energy use and greenhouse gas emissions. Results from a comparison of international studies on this subject will be used to identify relevant assumptions that lead to different answers in the evaluation process.

Höhlein, B.; Grube, Th.

Comparative Assessment of Future Drive Systems

Management Circle Conference "Hybridfahrzeuge auf dem Vormarsch", 14 and 15 June 2004, Bad Homburg

The advantages of hybrid drives in terms of energy demand and emissions are determined by design and driving strategy. Simulation calculations also show clear energy demand and emission advantages for hybrid drives, especially for diesel and C.NG combustion engines, in comparison to improved conventional drives. Hydrogen-operated hybrid drives are particularly efficient if operated with low-temperature fuel cells. This involves a higher price. On the whole, this analysis must be superimposed on fuel supply, especially if hydrogen is included in the comparison and the costs, energy demand and emissions per kilometre are determined.

Höhlein, B.; Grube, Th.; Biedermann, P.; Bielawa, H.*; Erdmann, G.*; Schlecht, L.*; Isenberg, G.*; Edinger, R.*

Methanol as an Energy Carrier

*Publications by Research Centre Jülich, Series Energy Technology, Volume 28
Research Centre Jülich, Jülich, 2003*

In the medium- to long-term perspective, new final energy carriers for mobile and stationary applications will be produced on a fossil and increasingly on a renewable basis. Methanol, large amounts of which are currently produced for the chemicals market, will be able to play a significant part in this field. Methanol can be used in both internal combustion engines and also in fuel cells: in mobile aggregates for vehicle drives, in stationary plants for power and heat generation, in portable fuel cell assemblies for electric power generators. Furthermore, industrial downstream products of methanol are also conceivable. Today, methanol is mainly produced from natural gas, but it can also be obtained from fossil energy sources and biomass. The present book – with contributions from research and industry – presents a comprehensive picture of the production methods and applications of methanol and its downstream products, and also an overview of market and cost developments worldwide, as well as issues of safety and environmental impacts. In the applications, major emphasis is focused on road traffic.

Höhlein, B.; Grube, Th.; Stolten, D.

Fuel Cell Systems and Hydrogen as an Energy Carrier

BWK, January 2004

Europe's future situation with respect to energy and the environment requires changes in the type of energy conversion and in energy carrier selection. Fuel cells and new energy carriers such as hydrogen will increasingly contribute towards converting and using energy efficiently and in a clean manner. The European High Level Group (HLG) and the International Partnership for the Hydrogen Economy (IPHE) enable new optimum prerequisites to be created for a medium- to long-term successful implementation with the aim of pooling the resources necessary for successful development and market launch.

Lokurlu, A.; Grube, Th.; Höhle, B.; Stolten, D.

Fuel Cells for mobile and stationary power generation – Environmental aspects and cost analysis

Proc. Hyforum, 24.-28. May 2004, Peking, China

Highly efficient energy conversion systems with fuel cells for vehicles and stationary applications are currently being discussed all over the world as a technology which will be able to reduce primary energy demand and emissions of limited and climate-relevant pollutants. The high flexibility of fuel cell systems with respect to energy carriers opens up possibilities of modifying the energy market in the long term. New environmental legislation, above all in the USA, stipulating the introduction of emission-free cars from 2003, has led in the transport sector to an intensified search for alternatives to conventional drive concepts. In stationary applications, numerous demonstration plants and some field tests already implemented reflect the developmental stage of fuel cell systems. In Germany, a new CHP (combined heat and power) modernization law has been enacted. This act is of special significance for the market launch of fuel cells. A major milestone on the road to market success for all the above-mentioned systems – in order to compete with conventional technologies – is the reduction of costs. In this contribution, system analyses for mobile and stationary applications of fuel cell systems are presented as well as economic analyses for different fuel cell systems for stationary applications, especially combined heat and power generation based on natural gas as the energy carrier.

PhD theses:

Gebert, M.

Benchmarking methodology for components of polymer electrolyte fuel cells

Publications by Research Centre Jülich, Series Energy Technology, Volume 30, 2004 (in German)

Besides aspects of lifetime, efficiency and environmental compatibility, the question of the attainability of given cost targets is of paramount interest in fuel cell technology. Therefore, a major aim is to develop a universally valid techno-economic cost model for components of PEFC stacks in the kilowatt range. Different technical solutions will be comparable with regard to their corresponding production costs – based on the concept of benchmarking management. Determining the mass-specific production costs takes on a key role. The utilization of a Monte Carlo approach is a novel procedure providing expectation values as well as confidence intervals for the material costs of the stack components. The consideration of production costs incorporates the fabrication conditions of individual components. The new methodology is first applied to the injection moulding of composite plates as one of several technologies for producing bipolar plates. Furthermore, detailed cost analyses of five common ionomers constitute the main technological focus of the thesis in view of the complexity of ion-conducting polymer membrane production and the difficulties customers would have in assessing this process. The material cost of platinum-based catalysts outweighs their production aspects in a cost analysis, at least under present conditions. From a strategic point of view, such accompanying process cost calculations can undoubtedly contribute to a faster recognition of the cost potential of promising fuel-cell materials and component technologies. Limited R & D resources can thus be focused more efficiently on the commercial success of the innovative fuel cell technology.

Important patents:

Patents granted:

Principal inventor	PT	Title
Andrian, von, S.	1.1747	Optimierung der Betriebsparameter eines Direkt-Methanol-Brennstoffzellensystems





4

Projects

Selected R&D Projects

- Durable PEFCs as a prerequisite for a hydrogen economy
- Pilot operation of a solid oxide fuel cell (SOFC) with mine gas
- APAWAGS – System for on-board power and water production based on fuel cell technology

4.1 Durable PEFCs as a prerequisite for a hydrogen economy

4.1.1 Objectives and tasks

The project goal is the identification and verification of ageing mechanisms of the PEFC. The project is subdivided into two phases: in the first phase (01.10.2000 - 31.12.2002) basic investigations were carried out, whereas in the second phase (topping-up project, 01.01.2003 - 31.12.2005) the focus is on application-oriented experiments with commercial MEAs. The priority tasks of IWV-3 in the first project phase were the modelling of ageing mechanisms. In the second project phase, the influence of dynamic changes of operating parameters on ageing is investigated.

The project is run for a total period of 63 months (01.10.2000 - 31.12.2005) and has been funded by the Federal Ministry of Education and Research (BMBF).

4.1.2 Status and results

Within the framework of the first project phase with the work packages "Selected standard tests" and "Investigation of interactions" it was examined in a galvanostatic ageing test on an MEA from the DLR project partner (German Aerospace Centre) whether metallic materials in the peripherals of the test rig corrode under PEFC operating conditions and whether corrosion products accumulate in the membrane or the electrodes. The cell voltage already dropped after 400 hours of operation and collapsed after 930 hours. After the end of the ageing test, various regions of the MEA were subjected to an elemental analysis. Within the detection limit of ICP-MS no iron, chromium, nickel or titanium serving as indicators for possible corrosion products were found. The same result was obtained for product water samples taken during the ageing test. This means that corrosion of the stainless-steel peripherals and the titanium plates is of no importance under the chosen operating conditions and that materials containing stainless steel can be used in the peripherals.

A quasi-3-D model of PEM fuel cells (work package on model improvement) was developed at IWV-3. For the project, in particular, the influence of inhomogeneous distributions along the gas channels of total length L was simulated with the aid of this model. These inhomogeneous properties relate both to the local concentrations of the substances involved and to the local current densities j ($\bar{j}$ is the mean current density in the cell). They may explain the voltage fluctuations observed in galvanostatic ageing tests. According to theory, high ageing rates are to be expected, in particular, at low air ratios λ , since under these conditions the current density at the gas inlet ($z = z/L = 0$) is high and may lead to local exsiccation and warming, see Fig. 4-1. A theory on the dynamics of fuel cell degradation has been additionally developed, with which the course of galvanostatic ageing experiments can be explained. This concerns, in particular, a degradation behaviour in which the ageing rate continuously increases with operating time and the cell voltage finally collapses. The theory presented does not presuppose a specific ageing mechanism for explaining this ageing behaviour. It rather follows the assumption that a universal scenario is involved, in which, upon exceeding a critical current density at the gas inlet of the cathode, a degradation wave is generated, which propagates along the channel towards the gas outlet ($z = L$; $z/L = 1$).

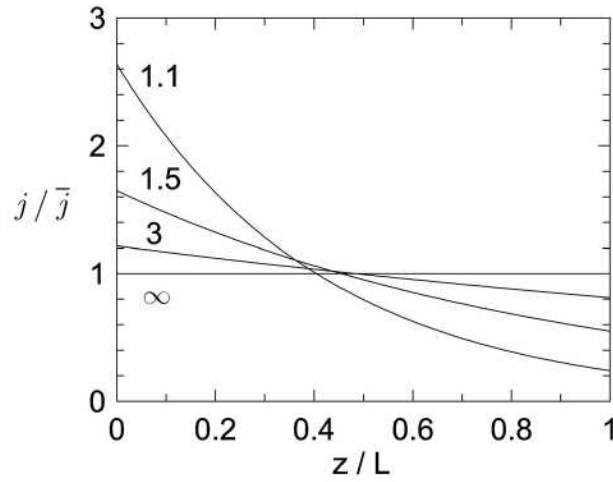


Fig. 4-1: Profiles of dimensionless current density at different air ratios

Assuming that local degradation means complete inactivity, i.e. that the front of the degradation wave interrupts the locally flowing current, the activity of the channel section behind the wave front equals zero. To this end, the channel section in front of the wave front must take up the total, constant current. In the shaded region at t_0 , where the local current density j initially exceeds the critical value j_{crit} , degradation takes place and no current is produced there any more after a time Δt . The front of the degradation wave thus shifts along the channel z towards a new position (e.g. at $t_1 = t_0 + \Delta t$). The wave continues to propagate until $z = L$ is reached. The further the degradation wave propagates, the smaller is the remaining, active channel section and the higher the average current density across this channel section, see Fig. 4-2.

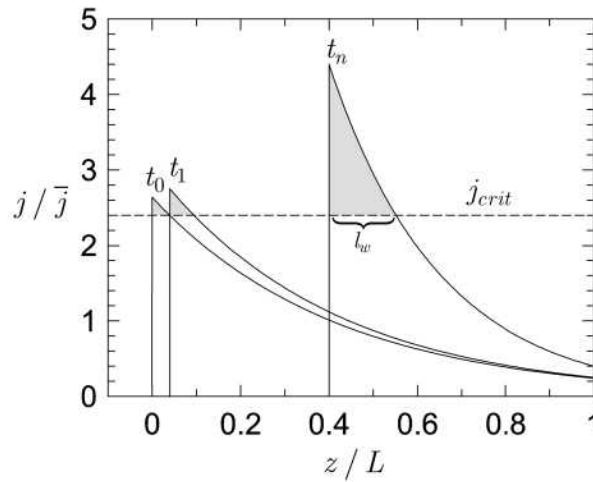


Fig. 4-2: Schematic representation of the formation of a degradation wave

The model presented here is supported by the fact that it explains the rapid drop in cell voltage at the end of the ageing test. This theory also explains why the oscillations of cell voltage increase with small stoichiometry factors on the air side (λ near 1).

In the 2nd project phase (topping-up project) with the work package "clocked mode of operation", preliminary experiments were carried out concerning the influence of a dynamic change of the CO_2 concentration in the fuel gas and of gas humidification on the cell voltage of a commercial MEA of type B2. The aim of the preliminary tests is to investigate the effect of a dynamic change of operating parameters on the temporal change in cell voltage and to derive suitable measuring programmes for the ageing experiments over 1000 hours. The following results were obtained:

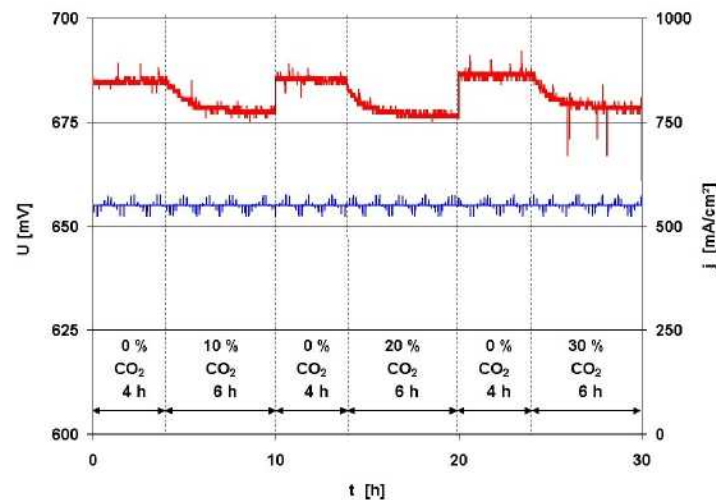


Fig. 4-3: Influence of the increase in CO_2 concentration on the cell voltage U (red) and the current density j (blue)

The change in cell voltage is only in the range of about 10 mV even for the greatest variation of the CO_2 content (0 - 30 %) and the greatest change of the relative humidity of the fuel gas (20 - 100 %), see Fig. 4-3. It is therefore not meaningful for the planned ageing experiments to further graduate the dynamic change of these operating parameters. The voltage change only depends slightly on CO_2 concentration and is about one order of magnitude lower than in the case of adding a corresponding amount of CO, as would be expected after the retro-shift reaction. This is indicative of the fact that the formation of CO from CO_2 rather takes place on the surface of the platinum catalyst.

The changes in cell voltage are reversible over the measuring period of a few weeks, i.e. the MEA B2 seems to be stable to CO_2 addition and exsiccation. The voltage change due to CO poisoning/CO desorption or exsiccation/humidification of the anode takes place in a period of hours or days. The planned ageing experiments should also be performed at corresponding intervals.

4.2 Pilot operation of a solid oxide fuel cell (SOFC) with mine gas

4.2.1 Objectives and tasks

Saar Energie AG (SE) is implementing the research project "Pilot operation of a solid oxide fuel cell (SOFC) with mine gas" jointly with Research Centre Jülich (FZJ) and the Institute for Future Energy Systems (IZES). The project enables Saar Energie to enter into decentralized electricity generation with the near-term aim of efficiently converting the existing climate-relevant mine gas into electricity and heat using the fuel cell. The findings obtained can be easily transferred to other lean gases such as biogas, sewage gas or landfill gas and their use for energy generation. The project involves in its first phase the further development of a suitable prereformer for operating a solid oxide fuel cell (SOFC) with mine gas as well as the construction of a corresponding pilot plant for testing an SOFC stack in the kilowatt range.

The project is run for a total period of 26 months and has been funded by the Federal Ministry of Economics and Labour (BMWA).

4.2.2 Status and results

In the Saarland, mine gas has already been used for economically and ecologically meaningful electricity and heat production since the 1940s. Today, mine gas is part of STEAG's energy concept, which also relies on decentralized energy supply in combined heat and power generation. The mine gas arising in the Saarland is subject to considerable variations concerning quantity and methane concentration. For better handling, therefore, an approx. 110 km long mine gas grid has been constructed in the past decades, which connects Saarland extraction points with users. The infrastructure conditions for connecting a fuel cell operated with mine gas are therefore ideal at Luisenthal, Saar Energie's former power plant site.

Major component	Concentration
CH ₄	30 - 90 Vol-%
N ₂	3 - 55 Vol-%
O ₂	0.1 - 10 Vol-%
CO ₂	2 - 8 Vol-%
C _x H _x	0.3 - 1 Vol-%
CO	2 - 300 ppm

Tab. 4-1: Mine gas composition

The composition of Saarland mine gas, which is virtually free from sulphur and halogen, varies as shown in Tab. 4-1 for the six main components. Mine gas can thus contain up to 10 % oxygen. The methane content is then in the range of the minimum value of 30 %. It

therefore had to be verified beforehand whether mine gas can be heated without risk in a fuel cell plant. The lower ignition limit for methane/air mixtures at RT is a methane content of 4.8 %, the upper limit is 16 %, which corresponds to a methane/oxygen ratio of approx. 1:1. The methane content for the upper ignition limit increases with rising temperature. The methane/oxygen ratio in the mine gas is 3:1 and thus clearly higher than the ratio in the range of the ignition temperature at the upper ignition limit (about 27 % methane, which corresponds to a methane/oxygen ratio of approx. 1.8:1). Thus, there can never be an ignition below the ignition temperature in the pipe even if a corresponding ignition energy is applied.

In the reformer test rig, the mine gas is mixed with steam with an S/C ratio of at least 2 and heated to 150 °C to avoid a condensing out of the water. This does not change the ratio of methane to oxygen, so that the mixture always remains above the upper ignition limit. Only in the reformer, which is heated by hot exhaust gas, is the gas heated to higher temperatures. On the basis of measurements in the test rig of IWV-3, it reaches a temperature of 350 to 500 °C in the manifold (i.e. before contact with the catalyst) (see Fig. 4-4). This means that the temperature remains well below the ignition temperature of methane of 595 °C (this applies to the composition with 8.5 vol.% CH₄ having the highest ignition ability; in the case of deviations from this composition the ignition temperature will increase). This means that neither the ignition temperature nor the mixture composition required for an ignition is reached, so that an ignition before reaching the catalyst can be excluded.

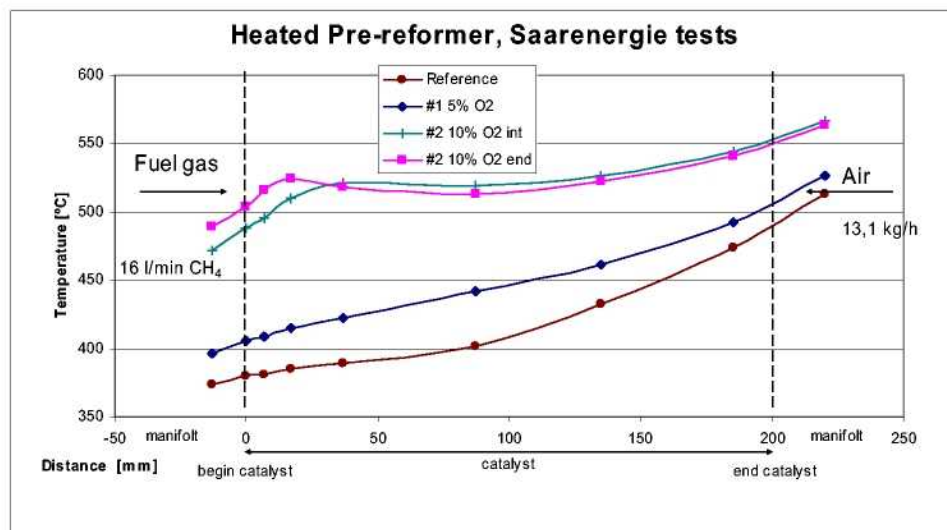


Fig. 4-4: Temperature variation in the reformer during operation with different mine gas containing 10 % O₂ at the start of measurement and in the steady state

Measurements in the reformer test rig at IWV-3 show that the oxygen on the nickel catalyst completely reacts with the fuel gas. The gas composition in front of and behind the reformer is shown in Fig. 4-5 in comparison to pure methane. Neither with 5 % nor with 10 % oxygen in the mine gas can oxygen be detected by gas chromatography at the outlet of the reformer.

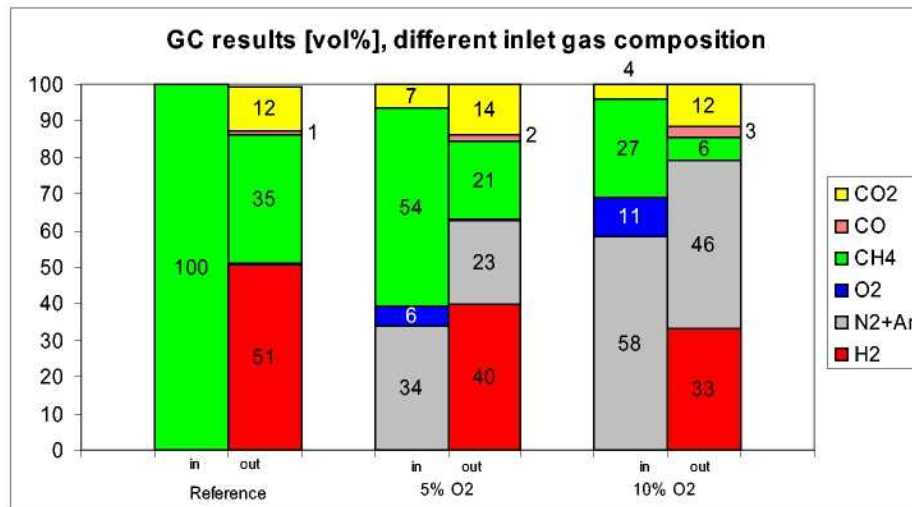


Fig. 4-5: Gas composition in front of and behind the reformer in comparison to pure methane operation (reference) – measuring results of gas chromatograph (GC)

Continuous operation over three weeks showed no change in reforming performance. Fig. 4-6 shows the temperature profile at the beginning and end of the experiment. The lines are virtually congruent.

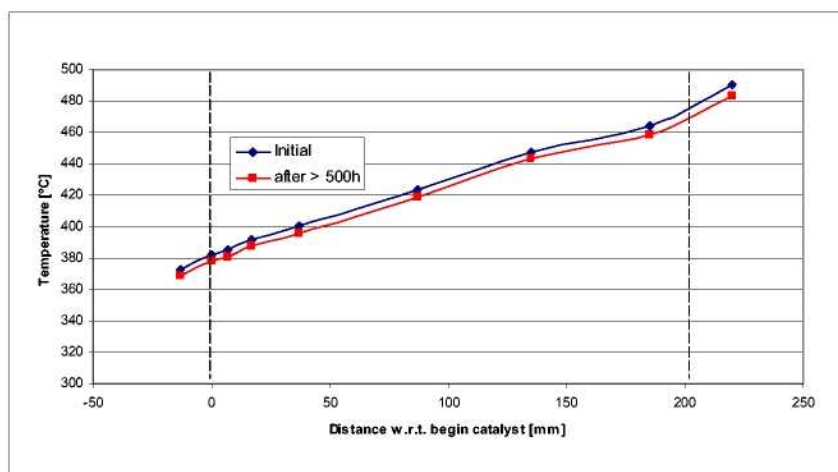


Fig. 4-6: Reformer temperature profiles at the beginning of the long-time test and after three weeks of continuous operation

Following the measurements at the prereformer with mine gas and the measurements at SOFC single cells with simulated reformed mine gas, an SOFC short stack was also operated with simulated reformed mine gas.

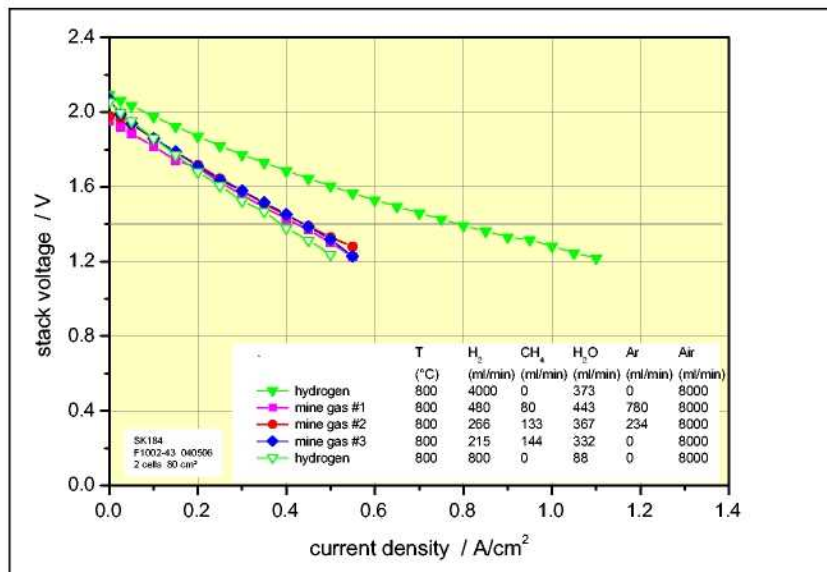


Fig. 4-7: Characteristics of an SOFC short stack during operation with mine gas of different compositions and with hydrogen

Curves with three selected mine gas compositions were first recorded at the stack. The curves (see Fig. 4-7) were not very different from each other. At 0.7 volt per cell, a current density of about 0.44 A/cm² was reached, i.e. a power density of 0.31 W/cm². The short stack was then run under constant current (0.3 A/cm²) for periods of 24 hours with the selected mine gas compositions setting the fuel gas utilization at 50 % and then at 70 %. Minor differences were found here concerning the power of the short stack. Higher methane fractions in the simulated mine gas resulted in a higher power density (0.24 instead of 0.23 W/cm²). The increase in fuel gas utilization caused a slight lowering of the power from 0.23 to 0.22 W/cm². On this basis a 2-kW stack was designed for operation together with the prereformer.

Various concepts were examined for the test rig to be set up at the Luisenthal site and, together with Saar Energie, that concept was selected which offers greatest flexibility along with minimum expenditure. The concept selected provides the possibility of testing the reformer together with the fuel cell or the reformer alone while bridging the fuel cell. Fig. 4-8 shows the flow chart of the test rig.

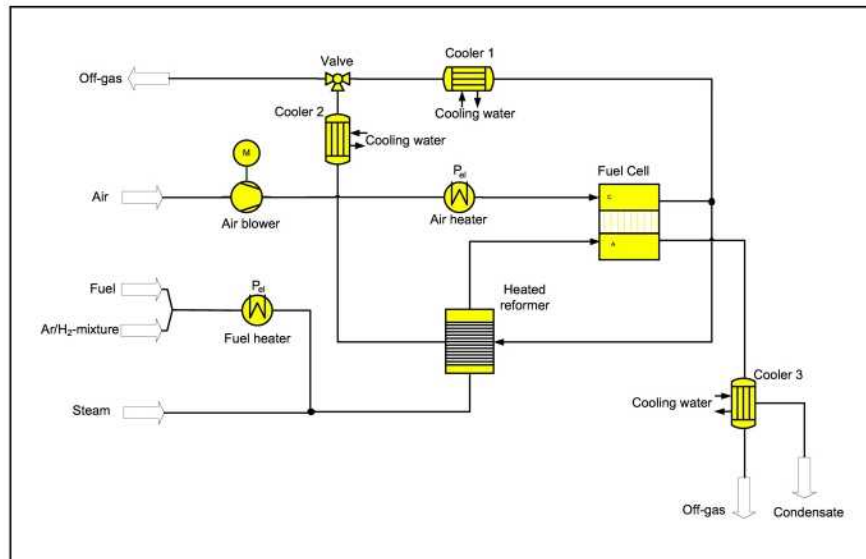


Fig. 4-8: Flow chart of the test rig for operating the reformer and stack with mine gas

Both the mechanical setup and the measuring and control technology were planned. All components were designed and selected. The relevant documents were made available to Saar Energie. The setup of the rig is shown in Fig. 4-9:

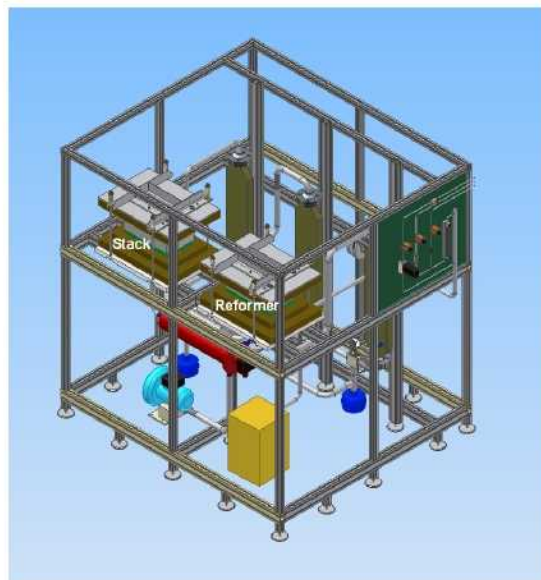


Fig. 4-9: Setup of the test rig for reformer and stack

The next steps were the assembly of the rig at the Saar Energie site and the construction and acceptance test of the reformer and stack by FZJ.

4.3 APAWAGS – System for on-board power and water production based on fuel cell technology

4.3.1 Objectives and tasks

Present-day gas turbines applied as so-called auxiliary power units (APUs) for on-board power supply in the aircraft sector are characterized by excellent performance values. However, they cause flue gas and noise emissions, the level of which will further rise in future due to the increasing traffic volume in the aviation sector and which will be subject to more stringent legal standards. Fuel cell systems can contribute towards considerably lowering the emissions in the aviation sector. On the one hand, the gas-turbine-based APU is to be relieved of electrical energy conversion and, on the other hand, fuel cell systems are to replace the main generators in aircraft engines. Furthermore, aeroplanes still carry a lot of drinking water for the passengers and the crew. The concrete quantity depends on the length of the flight, the number of passengers and the type of aircraft. The volume may be up to 2,000 l. Fuel cell systems make it possible to produce these water reserves on board, so that a uniform and high quality of the drinking water can be guaranteed, which no longer depends on supply at the point of departure.

The concrete task of the BGS department (Fuel Processing and Systems department) is to design and construct an autonomous reformer module for operation with kerosine. Apart from the reformer, this module also has components for educt supply (kerosine, water, air) and a regulating and control unit. It only has to be connected to the supply periphery for educts. The reformat can either be directly fed to an SOFC or passed into the flue gas system.

The project is run for a total period of 24 months (01.09.2003 – 31.08.2005) and is funded by the Federal Ministry of Economics and Labour (BMWA).

4.3.2 Status and results

The mechanical and electrical work for the module, which is shown as a photograph in Fig. 4-10 and can be seen in a perspective sectional drawing, is completed. After successful testing of all components, the module was delivered to the Airbus company early in March 2005. The first catalytic tests of the reformer have already been carried out. Their results will be described in the following. Moreover, general information on the energy carrier kerosine and its desulphurization will be given.



Fig. 4-10: Module for reforming kerosine, left: photograph, right: perspective sectional drawing

4.3.2.1 General information on kerosine fuel

Tab. 4-2 lists the different types of kerosine, explains their special features and indicates their applications. The information given is quoted from the following publication: M. Rachner, "Die Stoffeigenschaften von Kerosin Jet A-1", DLR communication 98-01, 1998.

Designation	Applications
Jet A-1	Commercial kerosine generally used for aircraft engines outside the USA.
Jet A	Commercial kerosine generally used for aircraft engines within the USA. Jet A-1 and Jet A mainly differ by their freezing point.
Avtur	Corresponds to the British military-specification, which approximately has the same limit values as the American specification for Jet A-1 (NATO code: F-35).
JP-1	Original American fuel for aircraft turbine propulsion. Very low freezing point (max. – 60 °C). No longer in use.
JP-4	Former usual fuel for aircraft engines in military applications in North America and Europe. Replaced by JP-8 (NATO code: F-40).
JP-5	Kerosine with high flash point (NATO code: F-44).
JP-7	Comparable to Jet A-1. Kerosine of low volatility and high heating value (at least 43.5 MJ/kg). Used by the US Air Force for special missions.
JP-8	Mostly used kerosine in North America and Europe for military applications (NATO code: F-34).
JP-B	Civil equivalent of JP-4.

Tab. 4-2: Various types of kerosine and their applications

Tab. 4-3 gives an overview of the exact specification of kerosine type Jet A-1, which is clear, light-coloured and optically free from solid particles and undissolved water at ambient temperature. The information given is quoted from the following publication: DEF STAN 91-91/4: Turbine Fuel, Aviation Kerosine Type, Jet A-1. Ministry of Defence, Defence Standard 91-91, Issue 4, Publication Date 14.06.2002.

Properties	Units	Upper and lower limits
total basicity	mg KOH/g	max. 0.015
aromatics content	Vol.-%	25.0
sulphur, total	Gew.-%	max. 0.30
sulphur, mercaptans	Gew.-%	max. 0.0030
distillation: 10 % condensate	°C	max. 205.0
end of distillation	°C	max. 300.0
distillation residue	Vol.-%	max. 1.5
distillation loss	Vol.-%	max. 1.5
flash point	°C	min. 38.0
density at 15 °C	kg/m ³	min. 775.0; max. 840.0
freezing point	°C	max. –47.0
viscosity at 20 °C	mm ² /s	max. 8.00
spec. combustion energy	MJ/kg	min. 42.80

Tab. 4-3: Detailed specification of Jet A-1 used for reforming

4.3.2.2 Desulphurization process

Tab. 4-3 specifies a value of 3000 ppm as the maximum upper limit for the sulphur content. This amount of sulphur is clearly too large for the operation of a fuel cell system based on kerosine and would soon lead to a deactivation of the catalysts of the reformer and the fuel cells. The sulphur content must therefore be reduced well below 10 ppm. Tab. 4-4 lists the candidate processes and briefly explains them with their advantages and drawbacks.

Process	Description	Experience	Remarks
hydrogenation	H ₂ breaks up S-compounds (catalyst)	commercial process used worldwide	high pressures and H ₂ required
adsorption	catalyst adsorbs S-compounds which are set free again in the regeneration step	commercial process, few plants	
extraction	extractant which binds S-compounds and is not soluble in hydrocarbon	stage of research and development	no H ₂ required, ambient pressure
oxidation	chemicals that oxidize S-compounds	stage of research and development	no H ₂ required, ambient pressure

Tab. 4-4: Overview of various desulphurization processes

The long-term stability in reforming kerosine suffers from the already mentioned sulphur components as well as from the residues in kerosine which cannot be evaporated in the mixing chamber of the reformer. This residue consists of very complex and highly condensed aromatic substances, inorganic silicon compounds such as SiO₂ and inorganic ashes. It should be removed prior to reforming, since it can greatly deteriorate the functioning of the reformer. During reforming there is a possibility of separation, if hydrogenation from Tab. 4-4 is applied, in which the residue can be removed by distillation.

In order to determine the amount of residue in kerosine Jet A-1 used for reforming in the APAWAGS project, the fuel was therefore subjected to a fractionated distillation. Fig. 4-11 illustrates that the kerosine contains a residue of approx. 1.2 vol.%, which cannot be evaporated at sump temperatures higher than 310 °C. It is also possible that the carbon-containing substances forming this residue do not evaporate in the mixing chamber of the reformer, in which similar temperatures prevail. These substances will then decompose and, on the one hand, deposit on parts of the reformer, such as the two-fluid nozzle, impairing their function. On the other hand, decomposition will lead to coking of the catalyst, so that its active centres for the chemical reforming reaction are blocked. Moreover, this experiment shows that the final boiling point is not reached at a temperature of approx. 250 °C. The greatest fraction of more than 50 % has a dew point of approx. 220 °C. It may be concluded from these figures that, apart from the mentioned residue, all the components of the kerosine investigated can be transferred into the gas phase in the mixing chamber of the reformer, in which temperatures above 300 °C prevail.

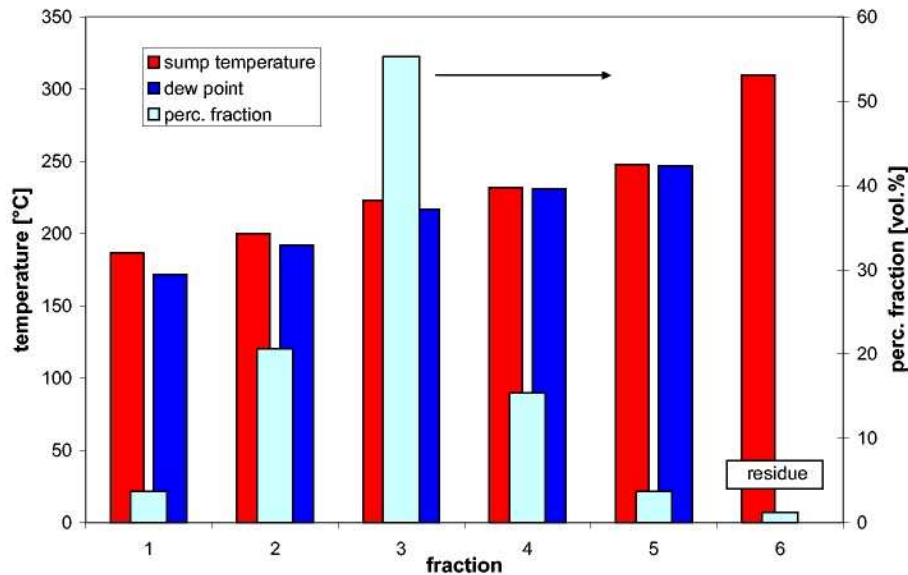


Fig. 4-11: Result of the fractionated distillation of kerosine Jet A-1

4.3.2.3 Experiments on reforming

Fig. 4-12 shows the temperature profiles in the axial direction of the reformer for kerosine at three different loads. In all three cases, the two-fluid nozzle, through which the kerosine and part of the reaction air were injected, displayed a temperature of 170 °C. In the 4-kW_{el} and 5-kW_{el} loading case the temperature in the upper and lower region of the mixing chamber ranged between 310 °C and 360 °C. These temperatures are high enough – as shown in Fig. 4-11 – to evaporate the individual kerosine fractions with the exception of the residue. At a load of 3 kW_{el} the temperature in the mixing chamber ranged between 350 °C and 400 °C and was thus approx. 50 K higher. This may be explained by a homogeneous gas phase reaction between air and kerosine, which is favoured by higher residence times. After being evaporated and mixed in the mixing chamber the reactants air, kerosine and water are passed into the monolithic catalyst, where the mixture ignites, and in all three cases a significant temperature increase to 790 °C – 830 °C takes place after 5 mm in the axial direction of the monolith. After 10 mm the temperature is already slightly lower. Behind the monolith it is only approx. 700 °C at each load. This temperature drop is explained by the fact that on the bottom part of the monolith seen in the axial direction the endothermic steam reforming between kerosine and water takes place, whereas in the upper region the strongly exothermic oxidation of the kerosine by the air proceeds at high reaction velocities.

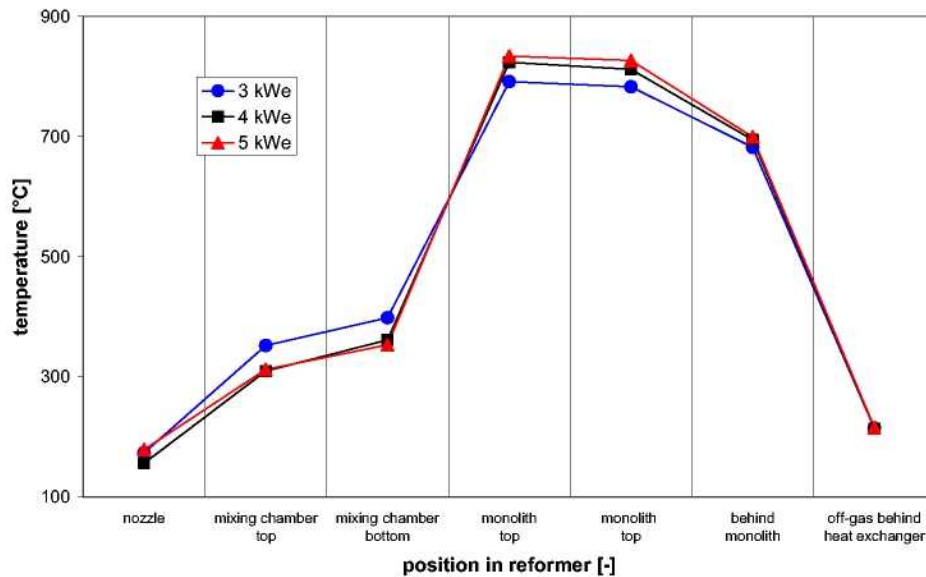


Fig. 4-12: Temperature variation in the autothermal reforming of kerosine Jet A-1 at three different loads

Fig. 4-13 shows the CO and H₂ concentrations after reforming and the efficiencies of reforming for the three load cases. The hydrogen concentrations were very similar for all three load cases and amounted to 36 - 37 vol.%. The same applies to the CO concentrations, which varied between 10 and 11 vol.%. The efficiencies of reforming were more than 80 % and also very similar in each load case.

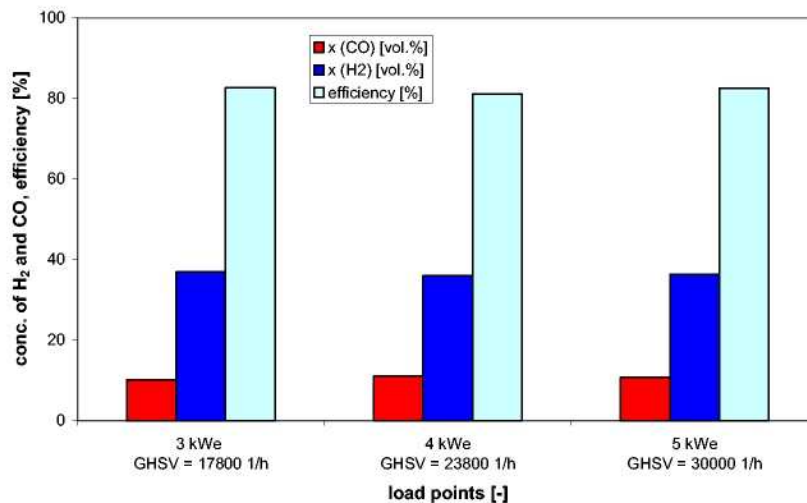


Fig. 4-13: H₂ and CO concentrations and efficiencies in the autothermal reforming of kerosine Jet A-1 at three different loads





5

Organization

Organization

- The Institute for Energy Process Engineering (IWV-3)
- Overview of department competences
- Site
- List of abbreviations

5.1 The Institute for Energy Process Engineering (IWV-3)

With a staff of just under 100 IWV-3 is oriented to the basic topics of electrochemistry and process engineering for energy converters. The focus of work for the electrochemical converters is on the two fuel cell types SOFC and PEFC/DMFC; the activities in the field of water electrolysis at high pressure are currently modest due to the lack of application interest. The focus of work for the chemical converters is on compact units for fuel processing from diesel fuel and diesel-like energy carriers.

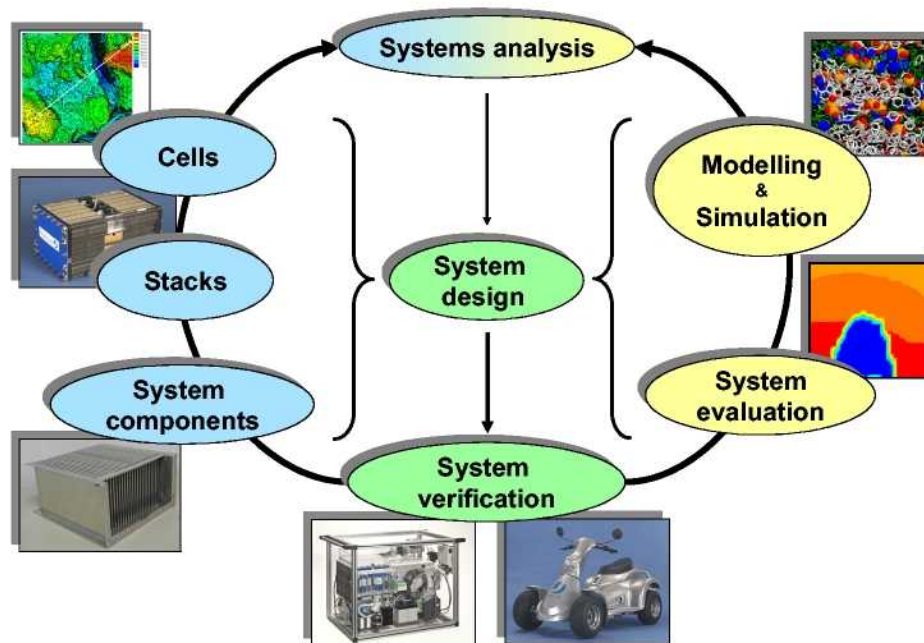


Fig. 5-1: Integrated work approach

In the sense of an integrated approach (Fig. 5-1) the three main activities, i.e. polymer fuel cells, solid oxide fuel cells and fuel processing, are accompanied by systematic studies, fundamental modelling and simulations as well as experimental and theoretical system evaluations. The findings obtained in these areas are used for the design of functional systems and their verification. In addition, particular attention is given to the development, setup and application of special methods of measurement for the structural analysis of membrane electrode assemblies (MEAs), for flow simulation and visualization and for the characterization of stacks.

Content-related R&D work is performed by six special departments, whose core competence is concentrated on the following main topics:

- focus on polymer electrolyte fuel cells,
- focus on solid oxide fuel cells,
- fuel processing,
- fuel cell process engineering,
- physico-chemical fundamentals and
- process and systems analysis.

An allocation of the competence fields at IWV-3, the R&D goals pursued and the special departments is given in Fig. 5-2. At present, on the whole, no other institute in Germany is active in the field of fuel cell technology to such an extent and with such intensity.

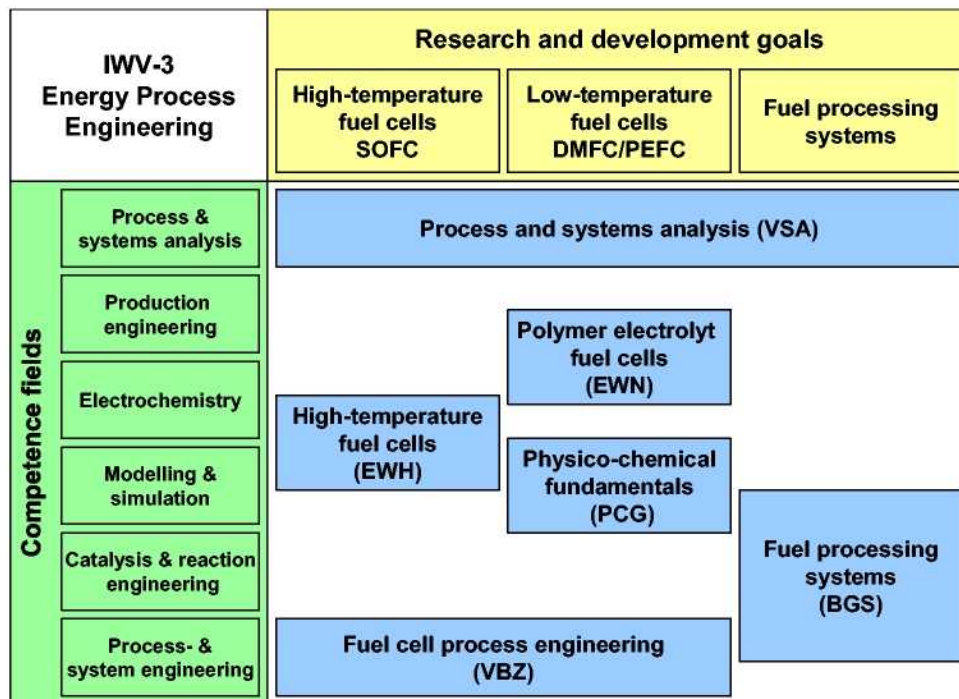


Fig. 5-2: Special departments vs. competence fields and R&D goals

5.2 Overview of department competences

Fields of work, scope of activities	Polymer electrolyte fuel cells The focus of work in the field of polymer fuel cells is on the development and optimization of the direct methanol fuel cell (DMFC) and the hydrogen-operated polymer electrolyte fuel cell (PEFC). Apart from the characterization of new membrane and catalyst materials, emphasis is placed on the optimization of components of the membrane electrode assembly (MEA), the selection and development of suitable, automated methods of manufacturing MEAs and the development of DMFC/PEFC stacks.	High-temperature fuel cells Work on the topic of solid oxide fuel cells concentrates on experimental investigations (electrochemical operating tests) and simulation calculations (modelling) to improve the efficiency. Cells and stacks are tested under practice-relevant operating conditions. In order to perform simulation calculations, models for cells and stacks are being developed taking the mass, charge and heat transport processes into account. Models, simulations and tests provide knowledge and approaches for advanced cell, stack and module concepts.	Fuel processing systems The most important fields of work for fuel processing are the reforming of natural gas and liquid fuels (diesel, kerosine) and gas conditioning. The focus in gas conditioning is on the water-gas shift reaction and the preferential oxidation of carbon monoxide. Work for the development of the reformers for diesel and kerosine is accompanied by modelling and visualization experiments. The results thus obtained are used for new designs with e.g. improved educt supply and educt mixing. Operational systems are constructed using the components thus developed.
	Plants, processes, methods Test facilities for - electrochemical investigation of direct methanol fuel cells and stacks - electrochemical investigation of polymer electrolyte fuel cells and stacks - characterization of membranes for the PEFC and DMFC - investigations on the current density distribution in single cells and stacks - desk coater for the continuous fabrication of MEA components	Test facilities for - electrochemical investigation of solid oxide fuel cells - electrochemical investigation of solid oxide fuel cell stacks - diffusion and permeation measurements - computer models for the simulation of operating states in solid oxide fuel cells (membrane, cell, stack) on the basis of the transport processes for heat, mass and charge	- process development for fuel processing - test rigs for reforming, for the water-gas shift reaction and for preferential oxidation - methods for catalyst screening and for activity and selectivity testing - gas analysis techniques (GC, GC/MS, NDIR) for concentration determination - modelling and visualization of educt mixing - fractionated distillation of fuels
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Fuel cell process engineering

Fuel cell process engineering is concerned with the design and construction of test facilities and their components and with the process-engineering systems analysis for high-temperature fuel cells of the SOFC type and polymer fuel cells of the DMFC and PEFC type. The focus of work is on the testing of plant components, the testing and optimization of control and regulating concepts for SOFC and DMFC systems and on the development and construction of plants as well as the calculation and evaluation of plant concepts.

Physico-chemical fundamentals

The physico-chemical fundamentals department is concerned with the theory and modelling of components, single cells and fuel cell stacks. The focus is currently on modelling polymer fuel cells. The two main fields of work are the molecular simulation of proton transport in polymer electrolyte membranes (PEMs) and continuum simulations of PEFCs and DMFCs. All the methods used require supercomputing systems.

Process and systems analysis

In process and systems analysis, energy conversion systems with fuel cells for power trains for the traffic of tomorrow and for stationary electricity production are designed, analysed, evaluated and compared. The investigations relate to the development, description and analysis of process concepts for mobile and stationary energy conversion systems with fuel cells including different fuel gas preparations (fuel gas paths). Benchmarking finally relates to the selection of low-cost materials and adequate manufacturing techniques.

Fields of work, scope of activities

Test facilities for

- high-temperature heat exchangers up to 850°C and 200 m³/h air
- reformers for SOFC stacks of a power of 5 kW
- afterburners for SOFC stacks of a power of 5 kW
- air compressors up to 100 m³/h
- Fuel gas heated steam generator for SOFC plants
- DMFC-Stacks (< 2 kW) and system components

Models for design

- of the flow distribution in stacks
- of PEFC, MCFC und SOFC plants

Theory and methods

- programs and continuum models of LTFCs in 1-D to 3-D
- 1-D analytical models of LTFCs
- programs and molecular dynamics simulations on H⁺ transport in PEs
- ab-initio quantum mechanics and molecular dynamics methods for the investigation of electrocatalytic reactions
- simulation calculations on the Jump supercomputer system of NIC and on the IWW-3 Linux cluster

Overall assessments (life-cycle analyses)

- energy and emission analyses
- FC propulsion simulations for fuel cell power trains
- process concepts for stationary and mobile systems
- process analyses for on-board electricity producing systems
- strategies for future energy supply systems
- benchmarking analysis
- evaluation of fuel concepts

Plans, processes, method



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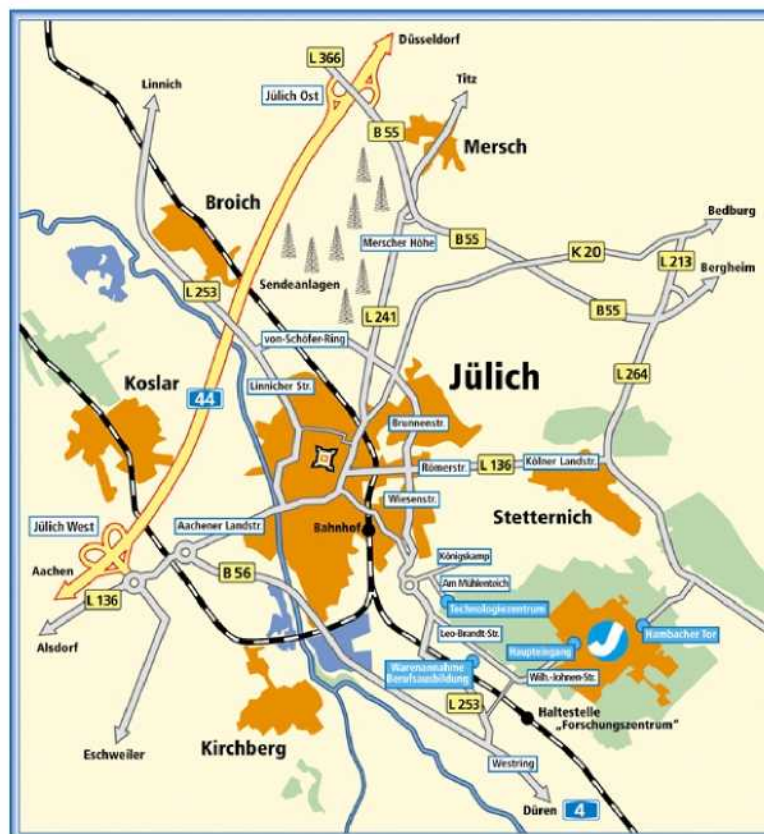
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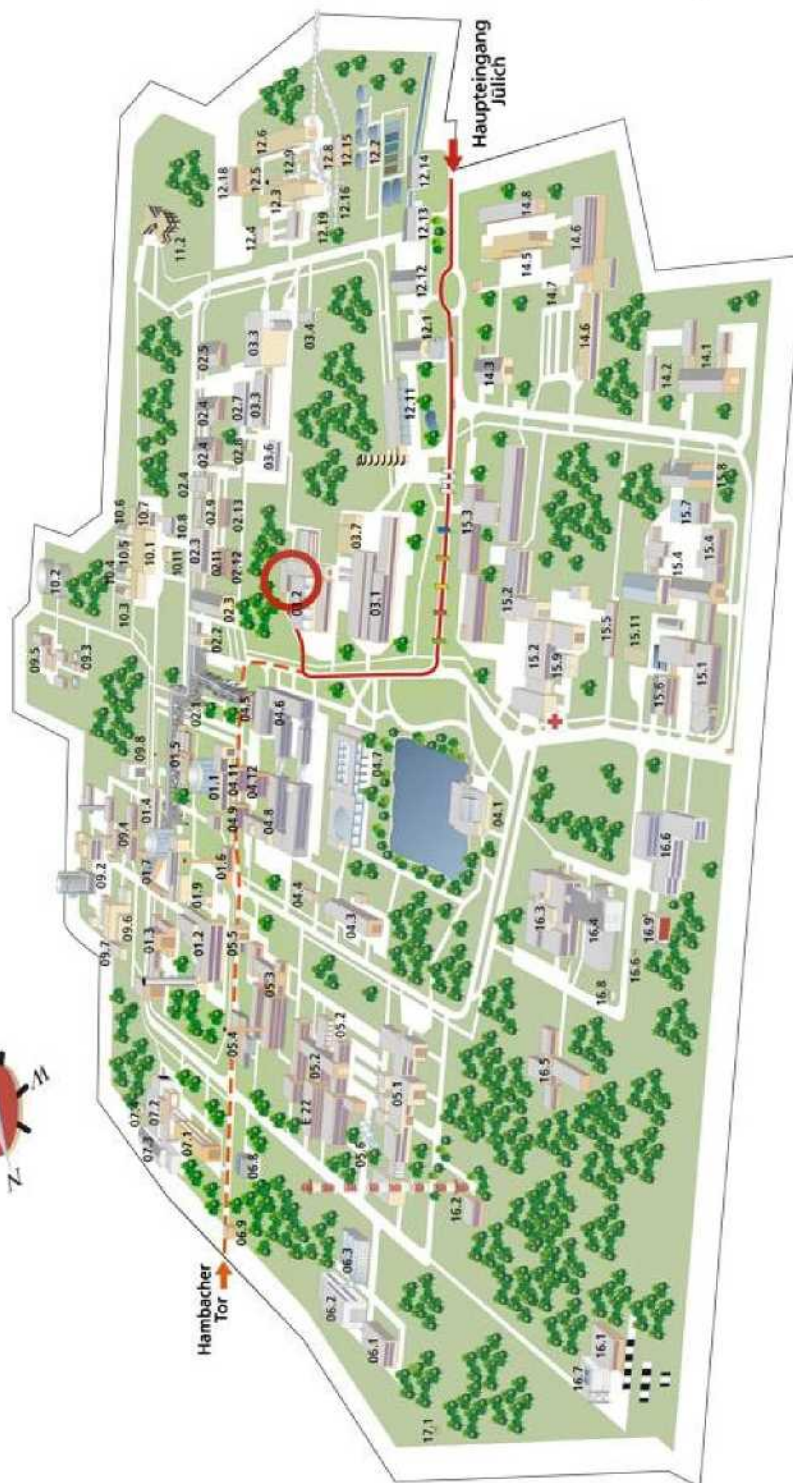
5.3 Site



How to reach us
Regional map:
Euregio and Rhineland



Regional map:
Jülich area





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5.4 List of abbreviations

APU	Auxiliary power unit
ATR	Autothermal reforming
BGS	Fuel processing systems
BMWA	Federal Ministry of Economics and Labour
CE	European Conformity
CFD	Computational fluid dynamics
C.NG	Compressed natural gas
DC	Direct current
DHE	Dynamic hydrogen electrode
DMFC	Direct methanol fuel cell
ECAB	Electrochemical afterburner
EKOPEC	Development of compact 5-kW fuel cell stacks with polymer electrolyte membranes
EWB	Electrochemical converter high temperature – solid oxide fuel cells
EWN	Electrochemical converter low temperature – polymer electrolyte fuel cells
EVB	Empiric valence fixation
FZJ	Research Centre Jülich
GC	Gas chromatograph
GC/MS	Gas chromatograph / mass spectrometer
GHSV	Gas hourly space velocity
ICP-MS	Ion coupled plasma mass spectrometer
GT	Gas turbine
IWV-3	Institute for Energy Process Engineering
JUMOVE	Juelich Methanol Operated Vehicle
MCFC	Molton carbonate fuel cell
MEA	Membrane electrode assembly
MWA	Ministry of Economics and Labour
NDIR	Non dispersive infrared
NTBZ	Low-temperature fuel cell
OCV	Open circuit voltage
PCG	Physico-chemical fundamentals
PEM	Polymer-electrolyte membrane
PEFC	Polymer electrolyte fuel cell
ProCon	Proof of Concept: "Decentralized Power Generation Plants based on Planar SOFC Technology"
PROX	Preferential oxidation
RHE	Reversible hydrogen electrode
SOFC	Solid oxide fuel cell
UDF	User defined functions
VBZ	Fuel cell process engineering
VSA	Process and systems analysis
WGS	Water-gas shift reaction

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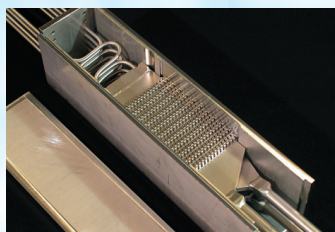
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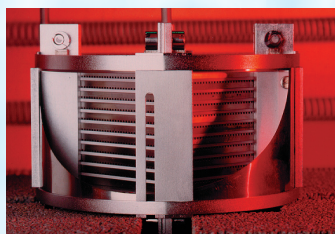
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1.3-kWe fuel cell stack with graphite bipolar plates for direct methanol use (DMFC) and use in small electric vehicles. The advantages of DMFC systems over conventional battery systems in mobile applications are the larger range and shorter supply time.



Open 1.0-kWe reactor for CO ultrapurification by preferential oxidation (PROX) of the carbon monoxide (CO) in the feed gas for low-temperature fuel cells. A special feature is the alternately packed metal pockets filled with turbulence structures for cooling and catalysed reaction.



Display model of a solid oxide fuel cell (SOFC) stack with planar anode substrate cells made of ceramics in B-design with external manifold for air and feed gas supply in cross flow. The advantage of SOFC systems over other fuel cell systems is the direct use of natural gas without extensive prior reforming and gas cleaning.

