

Bundesministerium für Verkehr, Innovation und Technologie



# IEA - Advanced Fuel Cells

## Austrian Participation in the IEA Implementing Agreement

on Advanced Fuel Cells 2004-2008

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Berichte aus Energie- und Umweltforschung



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## Austrian Participation in the IEA Implementing Agreement

on Advanced Fuel Cells 2004-2008

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#### Implementing Agreement on Advanced Fuel Cells

**Synopsis (Deutsch):** Das Hauptziel dieses Implementing Agreement ist die Entwicklung und Marktimplementierung von Brennstoffzellen-Systemen. Mittels eines international ausgerichteten Netzwerkes werden: (i) gemeinsame Forschungsziele formuliert und bearbeitet, (ii) Ergebnisse der Forschungsarbeiten diskutiert, disseminiert und (iii) System- und Marktanalysen durchgeführt.

**Synopsis (English):** The aim of the IEA Advanced Fuel Cells programme is to enhance the state of understanding of all contracting parties in the field of advanced fuel cells, through a co-ordinated programme of research, technology development and system analysis on Molten Carbonate (MCFC), Solid Oxide (SOFC) and Polymer Electrolyte Fuel Cell (PEFC) systems. There is a strong emphasis on information exchange through task meetings, workshops and reports. The work is undertaken on a task-sharing basis with each participating country providing an agreed level of effort over the period of the task.

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### Vorbemerkung

Der vorliegende Bericht dokumentiert die Ergebnisse eines Projekts aus dem Programm FORSCHUNGSKOOPERATION INTERNATIONALE ENERGIEAGENTUR. Es wurde vom Bundesministerium für Verkehr, Innovation und Technologie initiiert, um Österreichische Forschungsbeiträge zu den Projekten der Internationalen Energieagentur (IEA) zu finanzieren.

Seit dem Beitritt Österreichs zur IEA im Jahre 1975 beteiligt sich Österreich aktiv mit Forschungsbeiträgen zu verschiedenen Themen in den Bereichen erneuerbare Energieträger, Endverbrauchstechnologien und fossile Energieträger. Für die Österreichische Energieforschung ergeben sich durch die Beteiligung an den Forschungsaktivitäten der IEA viele Vorteile: Viele Entwicklungen können durch internationale Kooperationen effizienter bearbeitet werden, neue Arbeitsbereiche können mit internationaler Unterstützung aufgebaut sowie internationale Entwicklungen rascher und besser wahrgenommen werden.

Dank des überdurchschnittlichen Engagements der beteiligten Forschungseinrichtungen ist Österreich erfolgreich in der IEA verankert. Durch viele IEA Projekte entstanden bereits wertvolle Inputs für europäische und nationale Energieinnovationen und auch in der Marktumsetzung konnten bereits richtungsweisende Ergebnisse erzielt werden.

Ein wichtiges Anliegen des Programms ist es, die Projektergebnisse einer interessierten Fachöffentlichkeit zugänglich zu machen, was durch die Publikationsreihe und die entsprechende Homepage www.nachhaltigwirtschaften.at gewährleistet wird.

Dipl. Ing. Michael Paula Leiter der Abt. Energie- und Umwelttechnologien Bundesministerium für Verkehr, Innovation und Technologie

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# **1 Abstract**

The aim of the IEA Advanced Fuel Cells programme is to enhance the state of understanding of all contracting parties in the field of advanced fuel cells, through a co-ordinated programme of research, technology development and system analysis on Molten Carbonate (MCFC), Solid Oxide (SOFC) and Polymer Electrolyte Fuel Cell (PEFC) systems. There is a strong emphasis on information exchange through task meetings, workshops and reports. The work is undertaken on a task-sharing basis, with each participating country providing an agreed level of effort over the period of the task.

The current participants are: Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Italy, Japan, Korea, Mexico, Netherlands, Norway, Sweden, Switzerland, Turkey, UK and USA.

The five-year programme from 2004 till 2008 was structured in three technology oriented annexes and three application/market oriented annexes. The annexes will continue in the new period of the Implementing Agreement from 2009 till 2013.



Figure 1.1 Annexes of the five-year work programm from 2004 till 2008 (Source: IEA AFC)

Austria has been officially participating in this Implementing Agreement on Advanced Fuel Cells since autumn 2004. Work was carried out within Annexes XVI, XIX and XXI.

The specific Austrian goals of this participation were as follows:

- Integration of Austrian cutting-edge RTD<sup>1</sup> activities in international RTD emphasis
- Transfer of international know-how to (and in) Austria
- Promotion and mobilisation of Austrian applied and basic RTD (by gaining IEA knowhow)
- Strengthening the competitiveness and internationalisation of the Austrian economy by increased RTD contributions, in general.

The participation of Austria has been performed by a consortium consisting of the Austrian Energy Agency and the Fuel Cells Laboratory (TU Graz). This consortium guarantees both

<sup>&</sup>lt;sup>1</sup> RTD is the abbreviation for research and technological development.

technical-scientific and socio-economic contributions on a high level. Furthermore, there was a partnership with Energie AG Oberösterreich who delivered application oriented contributions within Annex XIX. Furthermore, the Austrian Agency for Alternative Propulsion Systems (A3PS) contributed in Annex XX during the past two years.

## **2** Introduction

The Austrian Energy Agency – on behalf of the BMVIT – started already in 1998 to launch information initiatives focussing on fuel cell technologies. The aim was to support Austrian companies and institutions with their RTD activities. Many analyses were published concerning the state of the art of fuel cells as well as ongoing European/international initiatives and research programmes. As a result of these efforts, several Austrian research institutes and companies could successfully be motivated to participate in European RTD programmes.

In the years 2001/2002 the Austrian Energy Agency – again on behalf of BMVIT – developed and published a strategic direction for fuel cells in the context of European, international and national RTD activities. Essentially an intensified integration of the Austrian RTD efforts in the European and international activities in consideration of the available national strengths was proposed. Overall, this strategy is still followed, especially with stationary and portable applications. The current Austrian participation on the Implementing Agreement (IA) on Advanced Fuel Cells (AFC) is also based on this strategy. For mobile applications a separate programme – called A3 technology programme "Austrian Advanced AutoMotive Technology" – was established by BMVIT. A series of fuel cell projects received funding by this programme.

The motivations to participate in the Implementing Agreement on Advanced Fuel Cells by Austrian institutes/companies are seen as follows:

- (i) Through the participation of the leading technology countries such as the US, Japan, Canada and others, Austrian RTD institutions achieve a significant access to RTD know-how. Usually, this information is hardly exchanged by other international activities. In this connection it should be noted that in an international ranking the European research institutions take only the third place after the US and Japan. By the participation in the Implementing Agreement on Advanced Fuel Cells, a cost-efficient access to know-how can be achieved by the Austrian participants.
- (ii) The Austrian fuel cell and hydrogen activities experienced a peak in the beginning of this century. Never before in the Austrian fuel cell research history so many projects were realized. This also applies for the number of involved scientists, institutions, companies and invested budgets. The Austrian participation in the Implementing Agreement on Advanced Fuel Cells could lead to a further strengthening of the scientific activities.
- (iii) Currently, worldwide significant fuel cell programmes and initiatives are in progress, such as e.g. the Fuel Cells and Hydrogen Joint Technology Initiative (JTI). The founding members of the Joint Undertaking are the European Commission and an industry grouping. The JU has been established as an international non-profit association representing the interests of the European industry. The Commission will fund 470 million Euro over six years till 2013, and at least the same amount will come from private industry. The results of the JTI are to deliver robust hydrogen supply and fuel cell technologies developed to the point of commercial take-off. For the automotive sector, the objective is to reach breakthroughs in bottleneck technologies and to permit the industry to take the large-scale commercialisation decisions required to achieve mass market growth in the time-frame 2015 till 2020. For stationary fuel cells (domestic and commercial) and portable applications, the JTI will offer the technology base to initiate market growth from 2010 till 2015 [Lit 1]. Austria

had (and in some parts still has) a low profile in European and international acitivities concerning hydrogen and fuel cells. Due to the participation in the Implementing Agreement, improved international networking (IEA, etc) and intensified Austrian research can be achieved.

# **3** Overview of the Implementing Agreement

The aim of the IEA Advanced Fuel Cells programme is to enhance the state of understanding of all contracting parties in the field of advanced fuel cells, through a co-ordinated programme of research, technology development and system analysis on Molten Carbonate (MCFC), Solid Oxide (SOFC) and Polymer Electrolyte Fuel Cell (PEFC) systems. There is a strong emphasis on information exchange through task meetings, workshops and reports. The work is undertaken on a task-sharing basis with each participating country providing an agreed level of effort over the period of the task.

The current participants are: Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Italy, Japan, Korea, Mexico, Netherlands, Norway, Sweden, Switzerland, Turkey, UK and USA.

The five-year programme from 2004 till 2008 was structured in three technology oriented annexes and three application/market oriented annexes. The annexes will continue in the new period of the Implementing Agreement from 2009 till 2013.

In Table 3.1 the annexes of the five-year work programme from 2004 till 2008 including objectives and duration are presented.

No.	Annex	Objectives	Duration
XVI	Collaborative Research on Polymer Electrolyte Fuel Cells (PEFCs)	To reduce the cost and improve the perform- ance of PEFCs, DMFCs and corresponding fuel cell systems	2004–2008
XVII	Molten Carbonate Fuel Cells towards Demonstra- tion	To assist the commercialisation of MCFC systems through collaborative research and development	2004–2008
XVIII	Solid Oxide Fuel Cells	To assist, through international co-operation, the development of natural gas-fuelled SOFC technologies	2004–2008
XIX	Fuel Cells for Stationary Applications	To understand better how stationary fuel cell systems may be deployed in energy systems	2004–2008
XX	Fuel cells for Transporta- tion	To understand better how fuel cells may be deployed in transportation applications	2004–2008
XXI	Fuel cells for Portable Applications	System analysis for portable fuel cells – optimisation by modelling; portable fuel cell activities world-wide; rules and regulations	2004–2008
		System, stack and cell development for portable fuel cells: operation concepts, design goals, evaluating fuel storage con- cepts, design of systems, design of stacks	
		Materials under operating condi- tions/materials innovation: life-cycle oriented stack and system testing, power out- put/efficiency/life-time testing, cost aspects	

Table 3.1: Annexes of the IEA Advanced Fuel Cells, objectives and duration (Source: [Lit 2])

Austria has been officially participating in this Implementing Agreement on Advanced Fuel Cells since autumn 2004. The specific Austrian aims of this participation were as follows:

Integration of Austrian cutting-edge RTD activities in international RTD emphasis

- Transfer of international know-how to (and in) Austria
- Promotion and mobilisation of Austrian applied and basic RTD (by gaining IEA knowhow)
- Strengthening the competitiveness and internationalisation of the Austrian economy by increased RTD contributions, in general.

Most of the countries in the Implementing Agreement – like Austria – participate in more than one annex, as shown in Table 3.2. The majority of the participants in the technology annexes come from universities and research laboratories. However, in the application annexes there is a much higher involvement of industry, including utility companies and vehicle manufacturers.

The participation of Austria has been performed by a consortium consisting of the Austrian Energy Agency and the Fuel Cell Laboratory (TU Graz). This consortium guaranteed both technical-scientific and socio-economic contributions on a high level. Furthermore, there existed a partnership with Energie AG Oberösterreich who delivered application-oriented contributions within Task XIX.

Austria participated in Annexes XVI, XIX, and XXI. The Fuel Cell Laboratory (TU Graz) contributed to Annexes XVI, "Polymer Electrolyte Fuel Cells", and XXI, "Fuel Cells for Portable Applications". Furthermore, the Austrian Agency for Alternative Propulsion Systems (A3PS) joined Annex XX during the past two years. There exists an obvious connection to Annex XIII, "Fuel cells for vehicles" of the Implementing Agreement on Hybrid and Electric Vehicles. The Austrian Energy Agency contributed to Annex XIX, "Fuel Cells for Stationary Applications".

Countries A	nnex XVI	Annex XVII	Annex XVIII	Annex XIX	Annex XX	Annex XXI
Australia			Х	Х		
Austria	Х			Х	Х	Х
Belgium	Х			Х		
Canada	Х		Х			Х
Denmark	Х		Х	Х	Х	
Finland	Х		Х	Х	Х	Х
France	Х		Х	Х		
Germany	Х	Х	Х	Х	Х	Х
Italy	Х	Х	Х	Х	Х	Х
Japan	Х	Х	Х	Х		Х
Korea	Х	Х	Х	Х	Х	Х
Mexico	Х					
Netherlands	Х		Х	Х	Х	
Norway	Х					
Sweden	Х		Х	Х	Х	
Switzerland			Х	Х		
Turkey	Х	Х				
UK	Х		Х			
USA	Х	Х	Х	Х	Х	

Table 3.2: Participation of the countries in the annexes (Source: [Lit 2])

Austria, Denmark, Mexico and Turkey joined during the phase of 2004 till 2008. There are contacts with other new countries interested in being involved in the work, for example China and Russia. Brazil and South Africa have also shown interest in participation.

The Implementing Agreement on Advanced Fuel Cells is sharing information with other implementing agreements, such as hydrogen and hybrid & electric vehicles. This leads to spill-over effects to other energy technology areas.

On the website (<u>http://www.ieafuelcell.com/</u>) all important information about the Implementing Agreement on Advanced Fuel Cells is presented, such as annual reports, the end-ofterm report, publications, dates for meetings and contact details of the members of the Executive Committee.

# 4 Achievements of the project (according to the tasks)

### 4.1 Task XVI: "Polymer Electrolyte Fuel Cells"

#### 4.1.1 Aim of the task

The aim of the task was to reduce the costs and improve the performance of PEFCs, DMFCs and corresponding fuel cell systems.

#### 4.1.2 Metho dology

This task consisted of three subtasks:

#### Subtask 1: New stack materials

Research in this subtask aimed to develop improved, lower cost membranes, electrode catalysts and structures, membrane-electrode assemblies (MEAs), bipolar plates and other stack materials and designs. The effort included:

- Composite and high-temperature membranes
- Membranes that conduct protons without external humidification
- Reduced precious metal loadings in electrodes
- Non-precious metal cathode and anode catalysts
- Anode catalysts and electrode layer configurations with enhanced tolerance to carbon monoxide
- Higher-activity cathodes
- Lower-cost bipolar plates and other stack materials
- Lower-cost, continuous fabrication techniques for MEAs
- Stack materials for stacks operating at higher temperatures (>100 °C)

#### Subtask 2: System and balance-of-plant issues

This subtask addressed system-level and balance-of-plant issues in PEFC systems. This subtask involved development, engineering, modelling, testing, and standardization of test procedures involving:

- Fuel processors, fuel processing catalysts, and supports
- Gas purification membranes
- Compact fuel reformers and micro-structured reactors
- The effect of contaminants, operating environments, duty cycles, and operating temperatures including temperatures below 0 °C
- System designs offering efficiency and dynamic response while maintaining costs, weights, and volumes within target values
- The reliability, durability, rapid-start, and dynamic behaviour of PEFC systems

#### Subtask 3: Direct fuel polymer electrolyte fuel cells

The objective of this subtask was to improve the performance and lifetime of direct fuel polymer electrolyte fuel cells, including direct methanol and direct sodium borohydride fuel cells. This subtask involved identification and development of improved:

- Anode and cathode catalysts
- Electrode/electrolyte structures
- Fuel impermeable membrane electrolytes
- Anion-conducting membranes
- Concepts in stack materials and designs

Agencies from fifteen countries were involved in Annex XVI:

- Graz University of Technology (Austria)
- Flemish Institute for Technological Research, Vito (Belgium)
- The Government of Canada (Canada)
- IRD Fuel Cells A/S (Denmark)
- VTT Processes (Finland)
- CEA (France)
- Forschungszentrum-Jülich GmbH ICT Fraunhofer (Germany)
- Ente per le Nuove Technologie, l'Energia e l'Ambient, ENEA (Italy)
- New Energy and Industrial Technology Development Organisation, NEDO (Japan)
- Korea Institute of Energy Research (Korea)
- Instituto de Electricas (Mexico)
- Netherlands Energy Research Foundation ECN (Netherlands)
- Norwegian Technical University, NTNU (Norway)
- Swedish National Energy Administration (STEM) (Sweden)
- TUBITAK (Turkey)
- Secretary of State for Industry (UK)
- The Department of Energy of the US Government (United States)

The task leader was the US Department of Energy, Argonne National Laboratory. Nancy Garland (DOE) was acting as operating agent.

#### 4.1.3 Activ ities

#### 4.1.3.1 Subtask 1: New stack materials

*Italy, ENEA:* ENEA developed an electrodeposition technique for fabricating high utilization Pt/carbon powder and Pt/carbon nanotube electrocatalysts. The Pt particles show high surface area and are well dispersed on the carbon material. Due to the lower platinum loading fuel cell costs can be lowered.



Figure 4.1 Pt nanoparticles formed by electrodeposition under various deposition conditions (Source: ENEA, Italy)

**United Kingdom, Newcastle University:** The Newcastle University has developed palladium alloy catalysts for the oxygen reduction reaction. Figure 4.2 shows that PdCoAu/C and PdCoPt/C (7:2:3) have acceptable activity compared with commercial Pt. Therefore catalyst costs can be reduced significantly by using alloys instead of pure platinum. However, the researchers face stability issues of the platinum alloy catalysts which have to be solved in future.



Figure 4.2 Oxygen reduction activity of Pd alloy electrocatalysts compared with commercial Pt (Source: Newcastle University, United Kingdom)

#### 4.1.3.2 Subtask 2: System and balance-of-plant issues

**Belgium, VITO:** VITO has completed an ammonia-PEMFC project (ACCEPT). A stack voltage-current monitoring system was developed which is now in use for testing several PEMFC stacks (see Figure 4.3).



Figure 4.3 Stack fitted with VITO's Cell Voltage Monitoring System (Source: VITO, Belgium)

**Denmark, IRD:** IRD determined that the performance degradation of reformate-fuelled cells is due to membrane pinholes, PTFE erosion of the cathode GDLs, catalyst particle growth, and Pt/Ru migration from the anode. Future work concentrates on the analysis of the remaining operational MEAs to understand how the operational conditions accelerate or decrease the degradation.



Figure 4.4 Cross-section of aged MEA (2,000 h) showing catalyst component deposition in membrane (Source: IRD, Denmark)

*Finland, VTT:* VTT developed fuel cell stacks, stack components, and auxiliary devices for stationary CHP (0.5 to 2 kW), back-up power, and industrial specialty vehicles.



Figure 4.5 1.4 kW PEM Power Pack (Source: VTT, Finland)

*France, CEA:* CEA has developed a 80 kW<sub>el</sub> system for transportation applications and determining durability (30,000 h). Electron microscopy techniques are used to understand the link between micro and nanostructure and MEA performance.





*Mexico, Electricity Research Institute:* The Electricity Research Institute studied the water transport in MEAs and developing a stack simulator.



Figure 4.7 Water uptake of a MEA with time upon changing relative humidity conditions (Source: Electricity Research Institute, Mexico)

**Netherlands, ECN:** ECN studied the causes of performance degradation with freeze/thaw and cool down/start-up cycling. Freeze/thaw cycling increases ohmic and mass transport resistances.



Figure 4.8 Loss of MEA performance with 52 freeze/thaw cycles (Source: ECN, Netherlands)

*Turkey, TÜBİTAK MRC:* Integrating systems and developing components for PEFCs. Figure 4.9 presents bipolar plates with various flow field patterns.



Figure 4.9 Bipolar plates with various flow field patterns (Source: Tübitak Marmara Research Center, Turkey)

**United States, ANL:** ANL determined the effect of air impurities on the performance of MEA components. Chloride poisons the oxygen reduction reaction on Pt<sub>3</sub>Co, whereas methylene chloride does not.



Figure 4.10 Voltammetry showing chloride poisoning of oxygen reduction reaction (by 100 mV), (Source: Argonne National Laboratory, United States)

**United Kingdom, Newcastle Univ.:** Newcastle University modelled the performance of high-temperature membrane PEFCs. The researchers found that GDL convective transport and thin ionomer film around catalyst agglomerate are important.



Figure 4.11 Representation of agglomerate thin film model of electrode layer (Source: Newcastle University, United Kingdom)

#### 4.1.3.3 Subtask 3: Direct fuel polymer electrolyte fuel cells

*Germany, Jülich:* Jülich investigated aging effects in the DMFC stacks used in their fuel cell-powered scooter (JuMOVe). In Figure 4.12 the fuel cell-powered scooter (JuMOVe2) can be seen.



Figure 4.12 JuMOVe2 which showed stack degradation of 1,800 W to 600 W over 500 operating hours (Source: Forschungszentrum Jülich GmbH, Germany)

**United Kingdom, Newcastle University:** Newcastle University developed anode materials for microbial fuel cells running on wastewater fuel.



Figure 4.13 Power output of microbial fuel cell utilizing various anode catalysts (Source: Newcastle University, United Kingdom)

#### 4.1.3.4 Activities in Austria for PEM fuel cells

The work presented at the task meetings was carried out by the Fuel Cell Laboratory, Graz University of Technology. The two research topics described below were intensely discussed and showed good feed back within the workshops.

#### High temperature proton exchange membrane fuel cells – The impact of fuel contaminants and temperature on fuel cell performance

The HT-PEM fuel cells examined in this work utilize a PBI-H<sub>3</sub>PO<sub>4</sub> membrane-electrodeassembly commercialized under the brand name Celtec, which is operating best at temperatures ranging between 120 °C and 180 °C. Due to these high operating temperature levels, rates of electrochemical kinetics are enhanced, water management and cooling are simplified and this type of fuel cell can be implanted into fuel reformer systems. The fuel gas impurities CO and CO<sub>2</sub> can be tolerated up to 5% respectively 40%.

The impact of fuel contaminants and temperature on HT-PEM performance and current distribution is examined. In order to gain insight, EIS measurements, recording of polarization curves, segmented current measurements using a segmented flow-field and current collector design and on-line gas analysis on the cathode and anode exhaust are used as characterization techniques.

The following experiments regarding fuel cell performance and current distribution are conducted in this work:

- Influence of temperature
- Influence of stoichiometry
- Influence of catalyst contaminant and other feed impurities
- Analysis of exhaust gas streams

In order to obtain reliable data sets and perform measurements under optimum operational conditions, two fuel cells of different active areas and design are employed:

A commercially available (BASF), high-performance HT-PEM fuel cell with an active area of  $50 \text{ cm}^2$  and a maximum power output of approximately 70 W (H<sub>2</sub>/O<sub>2</sub> operation) is used to conduct all experiments over a wide range of temperature sets, different stoichiometry parameters and various concentrations of feed impurities. This is also the cell where the exhaust gas analysis is realized on.

The attending current distribution experiments are realized on a prototype HT-PEM fuel cell with an active area of 10 cm<sup>2</sup> and a segmented cathode flow-field-design with a segmented current collector design, self-developed by workgroups of the Laboratory for fuel cell systems (Graz University of Technology).

#### BASF 50 cm<sup>2</sup> HT-PEM

An exploded view of the 50  $\text{cm}^2$  HT-PEM and an actual photo are illustrated in Figure 4.14 The operating temperature is maintained by a heating pad on either end plate.



Figure 4.14 BASF 50 cm<sup>2</sup> Fuel cell with PBI MEA (Source: [Lit 3])

#### CDL Segmented 10 cm<sup>2</sup> HT-PEM

Construction drawings and an actual photo of the segmented 10 cm2 HT-PEM fuel cell are presented in Figure 4.15 and Figure 4.16. An equal pressure distribution along the end plates is realized by the spring deflection of the disc springs. The operating temperature is maintained by two heating cartridges.



Figure 4.15 Construction drawings of the CDL segmented 10 cm<sup>2</sup> HT-PEM cell (Source: Fuel Cell Lab TUG, Austria)



Figure 4.16 Photo of the CDL segmented 10 cm2 HT-PEM cell (Source: Fuel Cell Lab TUG, Austria)

Current distribution measurement data are obtained through the segmented current collector measurement module. The different voltage drops along the precisely defined resistors are in correlation with the prevailing current flow via Ohm's Law and thus can be attributed to the eight different segments. The flow field of the segmented cell consists of a segmented meander with two parallel channels with an active area of 10 cm<sup>2</sup>. The current collector rods are contacted with the channel segments by the application of pressure via the end plates.

A detailed scheme for the location of segments 1 to 8 and the flow directions of incoming reactant and outgoing exhaust gases is given in Figure 4.17.



Figure 4.17 Location of segments 1 to 8 and flow scheme for reactant and exhaust gases (Source: Fuel Cell Lab TUG, Austria)

#### **PBI** membrane

Both, the 10 cm<sup>2</sup> and the 50 cm<sup>2</sup> HT-PEM fuel cells are operated with PBI membranes (Figure 4.18). Celtec®-P 1,000 MEAs are produced at BASF Fuel Cell GmbH (Frankfurt, Germany) using Celtec®-P membranes and electrodes produced at BASF Fuel Cell Inc. (Somerset, NJ, USA). The cathode contains a Vulcan XC 72 supported Pt-alloy with 0.75 mgPt cm-2. The anode contains a Vulcan XC 72 supported Pt catalyst with 1 mgPt-cm-2. The thickness of the membrane in the MEA is approximately 50–75 µm [Lit 4].



Figure 4.18 Photo of the 10 cm<sup>2</sup> Celtec<sup>®</sup>-P 1,000 PBI-H<sub>3</sub>PO<sub>4</sub> membrane electrode assembly (MEA) (Source: Fuel Cell Lab TUG, Austria)

These MEAs are operating best at temperatures between 120 °C and 180 °C. Therefore they are especially suitable in reformed-hydrogen-based PEM fuel cell applications. Due to these high operating temperatures, CO tolerances up to 3–5% can be achieved.

Table 4.1 briefly summarizes the inherent properties of the Celtec®-P 1,000 MEA.

Membrane material	Polymerous Polybenzimidazole (PBI) doped with phosphoric acid (H <sub>3</sub> PO <sub>4</sub> )		
Full temperature range	120 °C–200 °C		
Operating temperature	160 °C–180 °C		
CO-tolerance	3–5% at 180 °C [Lit 3]		
Significant advantage	no humidification necessary		

Table 4.1: Summarized properties of the Celtec®-P1,000 MEA (Source: BASF)

#### Test rig

The test rig is basically a set up of four different systems: gas supply, the fuel cell, the electrical measuring units and the gas analysis work stations.



Figure 4.19 Flow-Sheet of the CDL test rig (Source: Fuel Cell Lab TUG, Austria)

Four mass flow controllers (MFC) are installed on the anode side for hydrogen (H<sub>2</sub>), carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>) and nitrogen (N<sub>2</sub>) gas flow control. On the cathode side two MFCs are implemented for the regulation of air and N<sub>2</sub> gas flow. After being mixed these gases by-pass or pass the humidifier and reach the anode resp. cathode side in a dry resp. wet state and react according to the reaction pathways in a fuel cell. The current produced by the electrochemical reactions is withdrawn by the electrical measuring system. Exhaust anode and cathode gases as well as unreacted educt gases will leave the fuel cell and are detected and analyzed in the on-line gas analysis system.



Figure 4.20 Photo of the test rig (Source: Fuel Cell Lab TUG, Austria)

The measurement, controlling and data acquisition of the test rig is realized by the controller-PC running the LabView<sup>TM</sup> package. On the graphical user interface (GUI), various process parameters for the corresponding experiments can be adjusted (Figure 4.21). The gas analysis data sets are evaluated on the GA-PC running the program package DaisyLAB<sup>TM</sup>.



Figure 4.21 Screenshots of the LabView<sup>™</sup> GUI used for the HT-PEM measurements (Source: Fuel Cell Lab TUG, Austria)

#### Results – CO exposition step profiles at 10A and 170 °C

The results of the current distribution measurements during the exposition to carbon monoxide are presented in the following "called" figure-tables.

Figure-Table 4.1 illustrates the CO concentration step profiles of up to 4.5% and the corresponding voltage and current responses of the segmented 10 cm<sup>2</sup> cell at 170 °C cell temperature and a current load of 10 A.

Figure-Table 4.1: CO exposition step profiles at 10A, 170 °C (Source: Fuel Cell Lab TUG, Austria)



The expected voltage drop with increasing CO concentration in the anode gas feed is observed in Fig. 1. The measured overall current is slightly dropping as well, due to increasing membrane and kinetic resistances.

The resolved current distribution for each segment is presented in Figure-Table 4.2.



Figure-Table 4.2: Current distribution during the CO step profiles presented in Fig. 1 (Source: Fuel Cell Lab TUG, Austria)

Positive current shifts of up to +10% are observed in segments S1 to S5. The segments S6 to S8 are the locations of negative current shifts of up to -25%. As hydrogen is consumed alongside the flowfield-segment setup a smaller fraction of hydrogen is supplied to the last segments. As a consequence, the hydrogen/carbon monoxide equilibrium is shifted towards higher CO coverage onto the Pt-catalyst sites. These segments are therefore impinged with higher resistances resulting in a current shift towards the segments with lower resistances.

This linear CO concentration – current shift behaviour – is plotted in detail in Figure-Table 4.3 for the average current shift values of segments S1-S2 and S7-S8, and for the current shift values of the middle segments S5-S7.

Figure-Table 4.3: CO concentration – current shift behavior for CO exposition step profiles at 10A, 170 °C (Source: Fuel Cell Lab TUG, Austria)



#### Carbon nanofiber based electrodes for polymer electrolyte membrane fuel cells

Due to the high electric conductivity and the corrosion resistance of carbon nanofibers (CNFs) compared to spherical, carbon black material they are regarded as an ideal alternative for electrode materials in PEMFCs. Carbon nanofibers (CNFs) with different structures have already been used or considered for use as alternative electrode material in polymer electrolyte membrane fuel cells. CNF based membrane-electrode-assemblies (MEAs) with membranes based on PFSA-ionomers have already been produced and characterized [Lit 5 – Lit 15]. The increase of the power density especially for portable fuel cells enables the production of small dimension fuel cell systems.

Tubular CNFs are carbon nanofibers with an average diameter of about 200 nm. The characteristic of tubular CNFs cannot be generalized but an estimation of their physical, chemical and mechanical properties between those of graphite and carbon nanotubes is reasonable. The use of CNFs for electrode material increases the free path length for the electrons compared to the use of carbon black, due to less resistance at particle or crystal edges. Another advantage of the use of CNFs in fuel cell electrodes is that lower binder is demanded because no single spherically particles have to be held together. A further advantage of tubular CNFs is their almost closed outside graphite layer that might cause a higher oxidation resistance. Corrosion studies of catalysts in an acid environment at high voltage demonstrate that CNFs based catalysts have a better corrosion resistance compared to carbon black based catalysts. The reasons are the different behaviour of the carbonylformation at the carbon surface as well as the stronger interaction between platinum and the  $\pi$ -binding of the graphite-coating. A smaller surface area reduces the corrosion rate as well [Lit 16 – Lit 20].

#### **Carbon nanofibers**

Tubular CNFs with different structures, diameters, production methods and further treatments were characterized for the use as electrode material. The criteria for the selection of the most applicable fiber were processability, oxidation resistance and the quality of the catalyst metal deposition. The CNFs HTF150FF-LHT produced by Electrovac AG, Klosterneuburg, turned out to be highly applicable. The product has an average diameter of 150 nm and a narrow diameter distribution, although some fibers with a diameter less than 20  $\mu$ m or more than 500  $\mu$ m are included. The diameter-length varies from 1:150 up to 1:10, whereas these short lengths can be caused by fragmentation (see Figure 4.22). The CNFs are straight-line to a high degree, they have a constant diameter and a high inner diameter. Moreover, twisted and other irregular CNFs and spherical, carbon black exist. TEMinvestigations show a graphite surface with a parallel arrangement of the layers to the fiber axis (see Figure 4.23). The surface is between 17 and 25 m<sup>2</sup>·g<sup>-1</sup> relating to the datasheet.



Figure 4.22 SEM image of HTF150FF-LHT in the overview. (Source: Fuel Cell Lab TUG, Austria)



Figure 4.23 TEM image of HTF150FF-LHT with platinum particle on the surface (bottom left). (Source: Fuel Cell Lab TUG, Austria)

#### Catalyst deposition

The platinum deposition on the CNFs HTF150FF-LHT for their use as platinum catalyst support in PEMFC-electrodes has been optimised. An optimal particle size and particle distribution was obtained (see Figure 4.24 and Table 4.2). The electroless platinum deposition was carried out after a fictionalisation and purification step with concentrated nitric and sulphuric acid under reflux conditions. Platinum salt was reduced on the CNF surface with NaBH<sub>4</sub> in aqueous, alkaline media.



	Profile widths broadened* [° 2Θ]	Peak position [° 2O]	Crystallite size [nm]
Graphite	1.809	26.027	4.5
Platinum	1.761	39.955	4.8
Graphite	1.989	43.724	4.3
Platinum	1.885	46.431	4.6
Platinum	2.074	67.806	4.6
Platinum	2.896	81.540	3.6

\* Peak widths standard: 0.100 ° 20

Figure 4.24 TEM image of CNFs loaded with 25 wt.% platinum, 4.6 nm crystallite size. (Source: Fuel Cell Lab TUG, Austria)

Table 4.2 X-ray diffraction data (Source: Fuel Cell Lab TUG, Austria)

#### **Electrode preparation**

For the preparation of CNF-based electrodes a sedimentation process and a spraying process was used. Both processes enabled the production of optimally structured gas diffusion layers and active layers (see Figure 4.25 and Figure 4.26). The electrodes produced by spraying had a high quality, whereas the electrode parameters in lab scale could only be varied in a small range. Further disadvantages of the spaying process are the utilisation of solvent with a high vapour pressure, the formation of desirable dust, more time-consuming process steps and the dependency of the result on the operator skills. An advantage is the possibility for electrode preparation at a large scale. The fast and simple sedimentation process allowed the electrode preparation without the use of organic solvents. Due to the constant electrode surfaces, this process allowed an exact setting of the electrode parameters, and the results were therefore highly reproducible. The disadvantages of this process are that it is not possible to include the electrolyte directly into the active layer, and that it is a batch process that is not optimal for mass production [Lit 21 – Lit 23]. The electrolyte was applied on the active layer by a spraying process afterwards.





Figure 4.25 SEM image of a carbon CNF based gas diffusion layer with PTFE as binder. The CNFs build up a porous, hydrophobic structure. (Source: Fuel Cell Lab TUG, Austria)

Figure 4.26 SEM image of a sprayed active layer with 0.4 mg⋅cm<sup>-2</sup> platinum and 33 wt.% Nafion. The CNFs are completely coated with the ionomer. (Source: Fuel Cell Lab TUG, Austria)

#### Electrode characterisation and optimisation

By optimising the structure, the binder content, the thickness of the gas diffusion layer, and the active layer of the electrode, it was possible to reduce the platinum content by more than 40% with only small performance losses.

Especially the thickness of the gas diffusion layer affected the performance of the electrodes, especially at air operation mode (see Figure 4.27). A higher power density was obtained by using the sedimentation process, due to the exactly defined electrode parameters (see Figure 4.28). A MEA with the same membrane, the same amount of electrolyte in the active layer, and the same amount of platinum catalyst, but with carbon black as catalyst support (Quintech C-20-PT, 20 wt.% platinum on Vulcan Carbon) and gas diffusion layer material (Vulcan Carbon, VC) was produced. This MEA was compared with MEAs with sprayed or sedimented CNF electrodes with the same or less platinum loading. The Vulcan Carbon based electrodes with the same platinum loading performed considerably worse compared

to the CNF based electrodes, especially at higher current densities (see Figure 4.29). Better gas diffusion properties and a better catalyst utilisation are achieved with CNF based electrodes.



Figure 4.28 Potential curves and power density curves of MEAs with sedimented CNF based electrodes with different platinum loadings (A = anode, K = cathode, catalyst loading specified in mg·cm<sup>-2</sup>). <u>left</u>: sedimented electrodes, <u>right</u>: sprayed electrodes (Source: Fuel Cell Lab TUG, Austria)



Figure 4.29 Comparison of potential curves and power density curves of MEAs with CNF based electrodes and Vulcan Carbon based electrodes with different platinum loadings(A = anode, K = cathode catalyst loading specified in mg·cm<sup>-2</sup>).

left: air operating mode, right: oxygen operating mode (Source: Fuel Cell Lab TUG, Austria)

#### 4.1.4 Outc ome

Technical achievements in Annex XVI included

- new methods for making lower-cost,
- higher durability platinum electrodes,
- the development of an ammonia-fuelled PEFC,
- the development of an 80 kW system for fuel cell locomotives,
- improved understanding of the degradation mechanisms involved when cells are started up and shut down, and when they are exposed to sub zero temperatures,
- the development of a PEFC stack simulator for system studies,
- studies on the effect of air impurities on performance of cell components, and
- the performance modelling of high temperature PEFCs.
# 4.2 Task XIX: "Fuel Cells for Stationary Applications"

# 4.2.1 Aim of the task

The main objective of the work in Annex XIX was to achieve a better understanding of the possibilities for all kinds of stationary fuel cells.

The work in the Annex focused on

- (i) describing the market conditions from all aspects for stationary fuel cells and to analyse the present situation.
- (ii) identifying the commercial niche applications for the early introduction of stationary fuel cells and the market applications for the broad use of stationary fuel cells.
- (iii) analysing opportunities and obstacles for stationary fuel cells to reach the market.

All fuel cell technologies and sizes under development suitable for use as stationary fuel cells were considered for the analysis in Annex XIX.

# 4.2.2 Metho dology

The work was divided in five subtasks, together fulfilling the objective outlined above. It has to be emphasised that the work to a large extent concentrated on system aspects and issues related to the use of fuel cells.

The following institutions and businesses participated in Annex XIX:

- The Swedish Energy Agency (STEM) acting through E.ON Sverige AB (Sweden)
- Ente per le Nuove tecnologie, l'Energia e l'Ambiente, ENEA (Italy)
- The New Energy and Industrial Technology Development Organisation, NEDO (Japan)
- Forschungszentrum Jülich GmbH (Germany)
- MTU Motoren- und Turbinen-Union GmbH (Germany)
- Austrian Energy Agency & Energie AG Oberösterreich (Austria)
- ECN (the Netherlands)
- Department of Energy (USA)
- EPRI (USA)
- VITO, (Belgium)
- Risø National Laboratory (Denmark)
- Ceramic Fuel Cells Ltd (Australia)
- Electricité de France (France)
- Office Féderale de l'Energie (Switzerland)
- VTT Kemiantekniikka (Finland)

The task leader was the Swedish Energy Agency (STEM) acting through E.ON Sverige AB. Bengt Ridell, Grontmij AB was acting on behalf of E.ON Sverige AB as operating agent.

# 4.2.3 Activ ities

The following pages summarise the activities and achievements of each subtask during the period 2004 till 2008. The five subtasks will continue in the new Implementing Agreement from 2009 till 2013 (see chapter 5).

# 4.2.3.1 Subtask 1: Market outlook for stationary fuel cells

Subtask leader: Thoma & Renz, Switzerland

In subtask 1 the different markets for stationary fuel cells were identified and successful business concepts were examined. Therefore the market situation in different countries was analyzed and competing technologies for fuel cells were identified.

The report from the subtask leader was mainly based on several questionnaires answered by the participants in the Annex. One obvious finding was that the conditions for a fuel cell market in the participating countries vary significantly. A SWOT (Strength, Weaknesses, Opportunities and Threats) analysis was done with the input of the participants in Annex XIX.

The Austrian Energy Agency was involved in the SWOT analysis and answered the questionnaires for Austria.

Questionnaire 1 - Monitoring stationary fuel cell applications

Questionnaire 2 - Potential and characteristics of the market for fuel cells

Questionnaire 3 - Customers (buyer) and seller for stationary power (and heat) systems

Questionnaire 4 - Competing technologies to stationary fuel cells

The results from the four questionnaires are described in the following:

#### Questionnaire 1 – Monitoring stationary fuel cell applications

In questionnaire 1 information about applications of fuel cells was collected and analysed, for example technical data and market- and business-orientated material. The questionnaires cover 56 installations in Europe and 1,250 installed fuel cells in Japan.

The survey about small fuel cells (from 1 to 50 kW) included analyses from 5 countries. Energy and gas suppliers are the typical customers for this kind of fuel cell applications. Their interests are delivering usable energy to the end user, getting experience with new technologies, and implementing new business concepts, for example contracting. The advantages of fuel cells are high electrical efficiency and low emissions. In the mid size range (50 kW to 1,000 kW), both utility companies and companies from the trade and commerce sector like hospitals, hotels, office buildings and other industries show interest in the implementation of fuel cell applications. The estimated benefits for the customers are high electrical efficiency, low emissions, low noise, high availability and grid independency. For most customers the combination of heat and power supply is important. The high investment and maintenance costs were mentioned as major problems. There were no responses taking into account large fuel cells. Fuel cells over 1,000 kW are only installed in the United States.

#### Questionnaire 2 - Potential and characteristics of the market for fuel cells

Fuel cells produce electricity and heat. The markets for stationary fuel cells are therefore the electricity and heat market. In a lot of countries those two target markets are very different

concerning the structure, the participants, the customers, the size of applications, the size of companies and the power relationships.

In the heat market, a lot of small applications are typically deployed. The market is characterised by many customers (house-installations) and various suppliers. The electricity market is dominated by a couple of big power companies and large power plants like hydroelectric power, coal-fired power plants nuclear, etc.

In most countries the primary energy used in heat production is different from the one used to produce electricity. Figure 4.30 shows the primary energy used to produce electricity and heat in Austria and exemplarily in Finland. In Austria, the majority of electricity is generated with water (hydroelectric power). For heating, biomass/wood and gas is used mostly.



Comparison: Kinds and shares of primary energy to generate electricity and heat

#### Fig. 2: Austria

Fig.3: Finland

Figure 4.30 Primary energy to generate electricity and heat in Austria and Finland. (Source: [Lit 24])

It was concluded by this subtask that the analysis of energy markets should be extended including the present and future needs and characteristics in order to implement decentralised CHP systems:

A barrier for a broad market introduction of fuel cells is the fact that in most countries the electricity and the heat market are two different markets with different structures and participants. Entering niche markets was identified as a transition solution for the broad market implementation. In niche markets the advantages of fuel cells are more competitive to state of the art systems than competing with large scale power plants in established markets. The advantages are: high electrical and total efficiency, low emissions and low noise.

Niche and early markets for stationary fuel cells can be remote areas or areas where the electricity grid is weak. Other promising niche applications for fuel cells are back-up systems, CHP-systems in the industry, and small scale applications which need high electrical efficiency. The possibility of using renewable fuels can also be an advantage.

# Questionnaire 3 – Custo mers (buyer and seller) for stationa ry power (and heat) systems

In questionnaire 3 information about present and future customers of fuel cells and realistic business concepts were analysed.

At the moment, customers for small fuel cell systems are mostly utilities and/or energy service companies. These companies are interested in testing new productions, business concepts and energy-production strategies, for example contracting models, consumer retention or virtual power generation.

The customers for mid-size systems are energy and gas suppliers and end users. The reasons for power companies to use fuel cells are enlarging market services, fostering the customer relationship and providing additional power generation. Typical end users are hospitals, hotels (in resorts), industry or apartment buildings. The advantages appreciated by end users are: higher security of supply, CHP or tri-generation, cheaper energy supply costs, high efficiency, low emissions, and the ability to use decentralised power-generation. In the future, the mid-size range contracting models will be very important for the further market deployment.

The following business concepts were characterised as promising:

- Contracting models (small-, mid-size fuel cells)
- Consumer requirement of outsourcing energy supplies
- Direct sales to the end users with long term guarantee and maintenance contracts
- Integration of other players in the heating market such as heating contractors and engineers
- Products for energy supply in remote areas

#### Questionnaire 4 – Competing technologies to stationary fuel cells

In questionnaire 4 the features of several electricity producing technologies were quantified and rated by the Annex XIX experts. The scale for the rating was 0 to 5 points. The results were depicted in a "spider graph" (see below). The more points the individual features receive, the better the technology is in this field.

Figure 4.31 shows the evaluation of answers for small sized (1–50 kW) technologies in Austria. According to the experts analysis, the advantages of small sized fuel cells are low emissions and low noise in comparison to the other technologies (piston engine, micro-gas turbines, stirling, free piston). The fuel cells reach the fewest points for field experience, risk of new or established technologies, number of manufactures and maintenance costs.





Figure 4.32 presents the evaluation of answers for medium sized (50–1000 kW) technologies in Austria. The benefits of medium sized fuel cells are low emissions, low noise and complexity of fuel treatment, according to the opinion expressed by the IEA experts. The disadvantages of fuel cells are little experiences and the fact that only few manufactures exist.



Figure 4.32 Evaluation of answers for medium sized (50–1000 kW) technologies in Austria (Source: [Lit 24])

According to the IEA experts, the most important competing technology for fuel cells are piston engines. In Figure 4.33 it can be seen that in comparison to piston engines the main-tenance costs for fuel cells are too high. Till now, there are not many manufactures for fuel cells and only little experiences compared to piston engines. The advantages of fuel cells are the low emissions and the low noise. Furthermore, the use of heat and the variety of applications are advantages of fuel cells. The other upcoming technologies such as stirling, micro gas turbines and free piston engines are strong competitors for fuel cells (see Figure 4.31 and Figure 4.32).



Figure 4.33 Comparison fuel cell versus piston engine (Source: [Lit 24])

# SWOT-Analysis

A SWOT (strength, weaknesses, opportunities and treats) analysis is a planning tool and often a summary of experts opinions. The SWOT analysis was prepared within Annex XIX. In Figure 4.34 the SWOT-Matrix is presented. The number in the brackets means the number of votes for each attribute.

		Strengths									Weaknesses												
Aggregation from 10 votes. The figure in brackets shows the number of votes for the attribute.		ow emissions (10)	high electr. efficiency (9)	uel flexibility (8/10)	modularity (4)	enewable H2-fuel, blofuels, waste to energy (4)	CHP, Tri-Gen, high eff. in small scale (3)	silent, not very noisy (4)	good particad eff. + characteristics (3)	high efficiency in small units, below MW-scale (2)	to paricles (1)	eliable (1)	tigh investment/ cost (10)	short lifetime; degradation (7)	ow availability, difficult to buy (4)	complex unit, high safty requierments (4)	poor experience, development stage (3)	sensitive to contaminents, clean fuel required (3)	ong start up time (1)	ow temperatur for heating (1)	stow development, over promis-ing, undercritical protect teams (3)	high parasitic loads	no low cost technology, cost arges will not be achieved
	increasing need of distributed generation (6)	-	Ŧ	-	+		+	41	+	+		+	+/-	-	-	+/-	-	+/-	+/-	+	-	+/-	-
	importance of renewable energy (bio fuels,			T		+										+/-	+/-			+/-			+/-
Opportunities	waste etc.) (7) storage of energy (from wind, solar, biomass) (3)		+	+						+			+/-		+/-	+/-	+/-	+		+		-	+/-
	Increasing costs of fossile fuel (2)		+	+		+	+			+			-	-				-		-			
	Imortance of low emission energy systems (climate change) (3)	+									+		+/-			+/-	+/-			-		T	+/-
	problem of grid stabilisation (3)								+			+		-	1				-				
	hugh market potential, mass market (5)						+						-		-	-					-		
do	incresing electricity demand in remote areas (3)		+	+	+	+	+	+	+	+		+	+/-		+/-		+/-	-			+/-		
	new products with 'green' image = new businesses (2)	+				+	+						+/-	+/-	+/-	+/-	+/-				+/-		+/-
	demand for other applications (APU, mobil) (2)							+	+			-	+/-	+/-		+/-	+			+/-			
	high electr. efficiency needed (2)		+				+		+	+			+/-	-		+/-			+/-	+/-			
	Competing technologies are better or will improve at lower costs (9)		-/+	-/+				-/+		÷.,		-	-	-	-	-			-	-	-	-	-
	multi MW-Technologies dominate electr. market (2)		-/+	X			-							-					0		•		-
Threats	revival of nuclear, coal (2)		-/+	1		-	•		1	-		*		-		0							0
	low electricity prices (4)	-	-/+										-	-	1	1				-		4	
	high gas prices (3)		+	+		+	+						-	-						-		-	-
	natural gas is not renewable (2)			+		+	+													-			
	Depending on subsidies/ less or stop of funding (7)												-	-	-						-		
	increasing cost of specific raw material (1)									+	E		-	-		-							-
	decreasing energy demand of new building (2)				+		+		+	+				11						+			
	Increasing demand of electricity not heat is favorable for solar-pv, wind mills (2)		+		+		+	(+)	+	+												-	
	increasing demand of heat pumps (2)		-/+	1		-/+	+						-		-	-	-			-			1

#### Figure 4.34 SWOT-Matrix (Source: [Lit 24])

Several conclusions are that fuel cells for CHP can have an advantage if they are introduced in new buildings with modern energy systems where low heating temperatures can be used. For large buildings, tri-generation production of electricity, heat and cooling is a growing demand, and fuel cells have the possibility to fulfil these needs with a high efficiency.

In the past years, the fuel cell technology has improved significantly. However, the support of R&D and demonstration projects is still very important to reduce the investment and maintenance costs and to increase the life-time of fuel cells. The advantages are high electrical efficiency, low emissions, low noise, high availability and grid independency. Therefore the experts of the IEA AFC Annex XIX see the following market opportunities for fuel cells:

- in applications with mid-size fuel cells and multi energy use (CHP, tri-generation)
- in applications with bio-fuels
- in remote areas
- in stand-alone applications with hydrogen, methanol or propane as fuel
- as assured power supply-systems

There are interesting efficient product solutions to combine fuel cells with absorption chillers and heat pumps. A future mass market, however, is rather seen in the heating market than in the electricity market [Lit 24].

# 4.2.3.2 Subtask 2: The effect of a large number of fuel cells connected to the power grid

#### Subtask leader: NEDO, Japan

This subtask examines the effect of a large number of fuel cells connected to the power grid on the existing infrastructure. The influence of several aspects was analysed, for example, on the electrical grid, fuel distribution, safety issues and economic considerations. Different alternatives for the control of the operation of the fuel cells were discussed. Existing codes and standards for the connection to the distribution network were also studied.

In the year 2005 an extensive program called "Large-Scale Stationary Fuel Cell Demonstration project" started in Japan [Lit 25]. In this programme a lot of small stationary fuel cells were installed. This demonstration project handles most of the problems and was the basis of activities within subtask 2. Energy suppliers like gas or oil companies received subsidies for the installation and operation of PEFC at various homes throughout Japan. The operating data were studied and utilized for commercialization.

From 2005 till 2007 2,187 fuel cell systems each at 1 kW<sub>el</sub> were installed in single-family houses in Japan (Figure 4.35). The suppliers were Panasonic, Ebara-Ballard, Eneos-Celltech, Toshiba FCP and Toyota. Several frequent fuels like natural gas, LPG and kerosene were used to run the fuel cell systems. A vision is to have a fuel cell capacity of 10,000 MW installed in the year 2020 in Japan.



Figure 4.35 Number of installed fuel cell systems in Japan (Source: NEDO, Japan)

Figure 4.36 shows the number of fuel cell units and the subsidies for the installations. According to the progress of cost reduction, the subsidy for the installation of one fuel cell unit declined significantly every year.



Figure 4.36 Outline of the Project (Source: NEDO, Japan)

Figure 4.37 presents the average of the monthly energy and heat demand of each house and the energy supplied by the fuel cell systems. The electrical demand has two peaks (W-shaped). The heat demand is shaped like a V, depending on the temperature change. The fuel cell systems are controlled to produce both electricity and heat with minimum losses, therefore electricity supply is limited in summer in Japan. Typically, in a year about 1/3 of the electricity demand and about 3/4 for the heat demand are produced by the fuel cell systems.



Figure 4.37 Energy and heat demand in relation to energy and heat supplied by fuel cell systems of each home (Source: NEDO, Japan)

In Figure 4.38 the electrical efficiency and heat recovery efficiency of each house is shown. The electrical efficiency and heat recovery efficiency are the average figures during all producing conditions including partial load operation. For this reason the efficiency levels are slightly lower than those at rated operation. In general it can be said that the longer the time of partial load operation is, the lower the efficiency values are. The average of electrical efficiency is 30%. The average of the heat recovery efficiency is 42.2%.





Figure 4.39 presents the development of the systems costs for fuel cells from the years 2005 till 2008. The development of the subsidies is also shown. The increasing number of fuel cells resulted in an economy of scale factor and are the major reason for the decreasing costs. The target costs for 10,000 produced systems a year were 1.2 million Yens per unit.<sup>2</sup>



Figure 4.39 System costs (Source: NEDO, Japan)

Due to the broad field tests, fuel cell manufactures and energy companies could improve the performance and operation software of fuel cells. The project results show that residential fuel cells are very efficient for energy savings and  $CO_2$  emission reductions. As a result of manufacturing many fuel cell systems the cost dropped [Lit 25].

The stationary project will continue in a new program 2009–2013. This project will continue in a new 5 year period, and the activities will be followed by the Stationary Annex.

<sup>&</sup>lt;sup>2</sup> Conversion factor: 1 € corresponds to around ¥ 130; 1,2 Mio ¥ correspond to around 4.000 € (January 2010).

# 4.2.3.3 Subtask 3: Fuels for stationary fuel cells

Subtask leader: MTU Onsite Energy, Germany

A lot of different fuels can be used in stationary fuel cells. In general, and due to the existing infrastructure, natural gas is used as fuel. Hydrogen can be important in the future as it is possible to produce it locally in many ways including the use of renewable energy. In stationary high temperature fuel cells, fuels that count as waste can be used, such as gases from chemical industry plants like purge gas, synthesis gas or other process gases. The gasification of biomass or sewage gas is also possible.

In this subtask the availability of different gases and the possibilities and consequences to use them as fuels for stationary fuel cells were analyzed. A couple of aspects concerning the use of the different kinds of fuels like infrastructure, technical issues and environmental impacts were studied.

Fuel cell demonstration plants using anaerobic digester gases are already in operation in some places in the US, Japan and Europe. The potential to use anaerobic digester gases and biomass gasification in fuel cells is very high. Till now, there are no fuel cell plants known that are using syngas from biogas gasification as fuel, as this technology is not yet fully developed.

Fuel cell systems using bio-fuels have many environmental advantages by combining the high efficiency of fuel cells with renewable energy sources. High temperature fuel cells, such as MCFCs and SOFCs have been identified as more appropriate for the integration with bio-fuels because of their higher tolerance to contaminants and their ability to internally reform bio-fuel gases, which is leading to lower operational costs.

As an example for the use of fuel cells in existing plants, the biogas plant in Leonberg Germany is presented in Figure 4.40. In the biogas plant the MCFC "HotModule®" from the MTU Onsite Energy is used to produce electricity and heat from biogas. The fuel cell system was installed in the year 2006. The electric power is up to 250 kW and the thermal output around 120 kW. The biogas supply amounts around 120 m<sup>3</sup>/h. It is possible to produce 1.4 MW electricity per year. The electric efficiency is around 47% and the thermal efficiency is about 23%. The overall efficiency is about 70%. The biogas is gained from bio-waste from renewable raw materials and therefore  $CO_2$  neutral [Lit 26].



Figure 4.40 Biogas plant in Leonberg, Germany and Fuel Cell, HotModule, MTU Onsite Energy (Source: MTU Onsite Energy, Germany)

Decentralized fuel cell systems use biogas and gasification gas as fuel. The production of biogas is already a commercial technology. Gasification is still under demonstration. Liquid bio-fuels, such as methanol, ethanol, and dimethyl ether (DME) and gaseous substitute natural gas (SNG) may be produced in large centralized facilities. Natural gas and biogas are the best existing fuels for high temperature fuel cells and therefore the feeding of SNG produced from biomass to existing pipeline network would be technically relevant.

Figure 4.41 shows the production costs of several bio-fuels in Nordic conditions. The presented costs include commercial technologies and technologies that are still under development. Based on the cost estimates, methanol and DME have the lowest production costs. In Central Europe, by far larger (5- to 10-fold) bio-fuel production concepts have been suggested and estimated. If raw materials would be available for those capacities, the production costs could be decreased further.



Figure 4.41: Production cost levels of some biofuels for transportation (Source: VTT, Finland)

Ethanol production from sugar and starch including raw materials is a commercial technology. Bioethanol exported from Brazil is the lowest cost alternative. Currently these amounts are rather small, but they are expected to grow. Bio diesel production via esterification route is a commercial technology, too.

Higher hydrocarbons, like bio-diesel, are more prone to coke formation and therefore more research on reforming of these bio-fuels is required before proceeding to demonstration scale. In the case of liquid bio-fuels, competition with transportation sector would be important.

The results concerning the costs of bio-fuels from different sources are only comparable to a limited extent due to the fact that the cost estimates are highly variable. Production costs depend on different factors, like time of estimate, scale of production, location, process concept assessed, financial parameters applied, and even source of the estimate. Furthermore, the feedstock costs influence the costs of the product. When agricultural by-products

are used as feedstock, potential subsidies have to be considered. A lot of the concepts for the production of bio-fuels are still in development and therefore it is difficult to compare them to industrial alternatives [see Lit 27].

However, one finding of the work was that waste is an important fuel for fuel cells. There exists a high potential for the use of renewable waste fuels in fuel cells. Bio-fuels that compete with food production are regarded as non ideal fuels. In the new Annex waste for fuel cells will be again an important subject.

## 4.2.3.4 Subtask 4: Balance of plant for stationary fuel cells

#### Subtask leader: VTT, Finland

In subtask 4, ways to facilitate the commercialisation of stationary fuel cells by reducing the costs and increasing the reliability of the balance of plant were investigated. Two thirds of the costs for a complete fuel cell system occur from the balance of plant. Most failures in the demonstration plants for stationary fuel cells appear in connection to the balance of plant. System auxiliary components are very often selected from existing suppliers standard components. As the market for the near future is rather limited only a few suppliers develop components specially for fuel cells.

One aim of the subtask was to identify the components in the balance of plant (BoP), such as equipment for desulphurisation, reformers, and inverters. The requirements for the main components in the balance of plant are analyzed and harmonized.

All important developers of high temperature fuel cells were asked about their BoP components and suppliers. Because of the strong competition between the developers, however, it was very difficult to get feedback about their preferred specifications for the components. In the upcoming Implementing Agreement, therefore, the subtask will start a new approach.

In the next task period, the subtask will focus on specifying the main components in the BoP. The objective will be to investigate the important requirements, such as temperatures, pressures, costs for the components and predictions for future costs. The data will be verified with component suppliers. In the meantime, some contacts have been established and more progress has been made [Lit 28].

#### 4.2.3.5 Subtask 5: Market and technology status for stationary fuel cells

Subtask leader: EPRI/DOE, USA and E.ON Sverige AB, Sweden

Subtask 5 focused on the technology status and the development of stationary fuel cells. In this subtask the information concerning the development of the technology and the market conditions for stationary fuel cells was collected, presented and analysed. The experts presented the important news or achievements from their countries during the meetings.

The Task XIX experts used this subtask mainly for information transfer. A lot of interesting and valuable facts and information were presented during each meeting. In the following, one important contribution from Australia (Dr. Karl Föger) concerning the technology status of SOFC systems is described as an example. The BlueGen<sup>™</sup> from Ceramic Fuel Cells Limited – CFCL (see Figure 4.42) is the latest development in relation to SOFC technology to produce very efficient electricity. The 2 kW BlueGen uses the Gennex fuel cell module. The fuel cell system reaches 55–60% net electrical efficiency and a heat recovery of ca. 85%. It is possible to install the BlueGen as power generator or co-generation system (power

and heat). Following CFCL, the BlueGen can produce enough power and hot water for an average home in a year. It can generate up to 17,000 kWh electricity per year and offers enough heat for 200 litres of domestic hot water per day. The  $CO_2$  savings add up to 11 tonnes per year compared to black coal. [Lit 29]



Figure 4.42 2 kW BlueGen<sup>™</sup> Modular Generator (Source: CFCL, Australia)

Figure 4.43 shows that the net electrical efficiency of the BlueGen goes up to 60%. Due to the high level of electrical efficiency less heat compared to other electric generators occurs with the BlueGen. Therefore – following CFCL – the unit will be able to operate longer, and more-efficient electricity is produced that helps to reduce  $CO_2$  emissions.





# 4.2.3.6 Activ ities in Austria for stationary applications

The Austrian Energy Agency organized the activities in Annex XIX (stationary applications). The Energie AG took part in the meetings since 2005 and presented their project knowledge and experience.

Additional to the work performed within Annex XIX subtasks, fuel cell demonstration projects executed by Austrian utility companies were analyzed (sector: micro CHPs with an electrical power output between 1 and 5 kW<sub>el</sub>). In total, five fuel cell projects were carried-out in the years 2000 to 2006, as shown in Figure 4.44. In Austria, up to now four units from the company Vaillant and one unit from the company Sulzer Hexis<sup>3</sup> were tested. The Energie AG, ESTAG/Steir. Ferngas, Wiengas and Salzburg AG participated in the demonstration projects [Lit 30, Lit 31]. In this figure also a biogas MCFC prototype testing initiated by PROFACTOR in cooperation with Linz AG is shown.



**4.6 kWel/11kWth PEFC System** Figure 4.44 Project activities related to "stationary applications" from Austrian utility companies (Source: Austrian Energy Agency) kWel/2,5 kWth SOFC System

In Figure 4.45 and Figure 4.46, the units **Biogas MGRS Reatotype** nt GmbH are presented. These units were used in the Au**Tresting** postration projects.

Salzburg

<sup>&</sup>lt;sup>3</sup> The company Sulzer Hexis AG is now called Hexis AG.



Figure 4.45 <u>left:</u> 1 kW<sub>el</sub>/3 kW<sub>th</sub> HXS 1000 PREMIERE unit, <u>right:</u> coupling of the individual components (Source: Sulzer Hexis AG)

Legend: 1. Fuel cell and waste-gas heat exchanger, 2. heat store 200 litre, 3. flow regulation, 4. heating device.



Figure 4.46 The 4.6  $kW_{el}/7kW_{th}$  Vaillant fuel cell heating system with additional heating device and hot water tank (Source: Vaillant GmbH)

The demonstration cases show that, up to now, it has not been possible to realize the high electrical and thermal efficiency values achieved in laboratory testing and to develop commercial products for competitive markets. For the first time in history, the recent generation of fuel cells – realised in Japan and shown in subtask 2 – reach electrical efficiencies of more than 30% in real applications.<sup>4</sup> Some units achieve efficiencies of 35%. All other problems, such as high investment costs, life-time, etc., require further research efforts from the manufactures.

For further efforts in R&D programmes, the following lessons could be derived from the Austrian demonstration cases:

<sup>&</sup>lt;sup>4</sup> Typical electrical efficiencies of ICE CHP systems in this size category are below these values.

- Decrease investment-, O&M-costs and generally the complexity of the system and components
- Improvement of life-time, reliability and efficiency (decrease own consumption of electricity)
- Optimisation of controlling software and of conventional systems (incl. hydraulic connection)

Based on the present results achieved by the demonstration projects, an implementation of fuel cells in competitive markets still requires major efforts. However, over the past years the number of companies developing fuel cells has increased significantly, in particular the development of micro-systems. The realization of demonstration projects is currently promoted by different companies, though the time schedules concerning commercial product developments could not be kept so far.

Table 4.3 presents the development status of fuel cells. The most important parameters are shown, whereas for comparative purposes also phosphoric acid fuel cells (PAFC) and molten carbonate fuel cells (MCFC) units are considered. In recent years, these units have either not been used anymore or no projects could be implemented in Austria. They are mainly used in the industrial sector in the power range of about 200 kW<sub>el</sub>.

Parameter	PE	FC P	A	FC	MC	FC	SOFC		
raiametei	2008 A	im	2008	Aim	2008	Aim	2008	Aim	
Life-time stack [1,000 h]	> 5	40	40	-	30	40	12	40	
Electric efficiency [ %]	20–31	> 35	40	-	47	50	20–25	> 30	
Investment costs [€/kW]	n/a	2,500	4,000	-	7,000	1,250	11,000	2,500	

Table 4.3: Stage of development of fuel cells (limited number of critiera) (Source: Austrian Energy Agency)

Other activities within Task XIX included the following points:

- Potential analysis of the installation of micro-CHP systems
- Benchmark analysis taking into account the investment and O&M costs of conventional state-of-the-art CHP systems (based on ICE technology) as main competing technology of fuel cells, and
- Economic analysis of micro-CHP systems in real installations taking into account country specific state-of-the-art frame conditions.

For the successful implementing of CHP-units in the connex of typical Austrian business conditions the following three requirements are the most important: <sup>5</sup>

- 5,000 till 6,000 full load hours
- Demand of heat and electricity at the same time
- High electricity costs (bad energy rate for the end customer)

<sup>&</sup>lt;sup>5</sup> Detailed planning will require in-depth analysis following: "VDI – Richtlinie 3985, Grundsätze für Planung, Ausführung und Abnahme von Kraft-Wärme-Kopplungsanlagen mit Verbrennungskraftmaschinen", Düsseldorf 2004

Such conditions can be found especially in the sectors of tourism and leisure. Theoretically, about 1,500 units might be substitued by micro-CHP systems in these sectors (see Figure 4.47).



Figure 4.47 Theoretical implementation potential of micro-CHP systems in the Austrian tourism and leisure sector (Source: Austrian Energy Agency)

With substitution rates between 1 and 4% about 8 to 30 CHP-units can be realised every year in Austria (see Figure 4.48). Niche markets like ecological sensitive regions or local objects qualify in particular for the further implementation (supported by the Austrian "Schutzhüttenprogramm"). Through successful marketing activities or improved economic frame conditions higher substitution rates could be achieved in these sectors.



Figure 4.48 Expected annually CHP-Installations in Austria (based on three scenarios) (Source: Austrian Energy Agency)

In the private sector (households) no substitution rates were identified due to highcompetitive economic frame conditions. However, the theoretical implementation potential would be very high. For further conclusions a bottom-up analysis specialized on the specific buildings characteristics (incl. heat and electricity demand) would have to be performed. Present subsidies in the building sector focus next to energy efficiency mainly on solid biomass, thermal solar energy, district heating, heat pumps and so on. Till now, CHP-units are not regarded enough by these schemes. Possible adoptions would have to be done country-specific.

In the consumer markets, food trade and shopping centres, the interest in alternative energy sources has increased in the last years. On the one hand it is part of the marketing concept however on the other hand energy is a high cost factor. Currently first studies and feasibility projects are in progress. The positive results of these studies are required for the potential analyses (in particular air conditioning is important for these consumers).

Through the new EU directives (Building-directive, CHP-directive, Energy efficiencydirective) no significant changes of this tendency are expected due to missing incentives.

#### Investment and maintenance costs

Within the project, it was planned to identify investment and maintenance costs of newly installed fuel cells in Austria. However, this plan was not carried out, mainly due to the following reasons:

- too high investment- and maintenance costs compared to Motor-CHPs (since fuel cells are still in the demonstration phase)
- several technical problems slowing down the commercialisation of the fuel cell technology and limiting the availability of new fuel cell installations (only demonstration projects at a very early commercialisation stage were realised)
- economic problems forcing companies in some cases even to discontinue their fuel cell business activities

Instead of analysing the cost efficiency of fuel cells, the benchmarks of ICE CHP-systems were identified. These data sets will serve as benchmarks to be exceeded by future fuel cell projects. Furthermore, two test cases involving state-of-the-art micro-CHP units were studied.

In Figure 4.49 the average standard prices of CHP-units are presented. In the literature price indications based on surveys with spot samples from several hundred offers can be found. These surveys include the CHP-module with all components considering delivery, compilation, integration, and approval of the system.

Two significant things can be seen. On the one hand the specific costs differ depending on the kind of used fuel. The specific costs are the prices related to the electric power. On the other hand a degression of the specific costs with increasing electric power is noticed with all CHPs.



Figure 4.49 Average standard prices for CHP-units (Source: Austrian Energy Agency based on ASUE 2006 data sets)

In most cases a full maintenance contract with the producer is arranged. This contract provides comprehensive maintenance at a fixed cost rate per produced electric kilowatt hour. In general this means the services necessary for a failure-free operation of a CHP, including inspection, all maintenance and repairing works, spare parts and supplies (except the fuel). Due to the long duration of the contracts, a general overhaul is also included.

Such contracts have a lot of advantages, for example they are easy to calculate and there are no technical risks. In the case of a defect, the supplier is responsible for the reparation.

In the course of research the costs for a full maintenance based on standardised price surveys of motor driven CHPs were identified.



Figure 4.50 Costs for operation and maintenance of CHP-units (Source: Austrian Energy Agency based on ASUE 2005 data sets)

In Figure 4.50 it can be seen that the service and maintenance costs for natural gas and biogas CHPs are almost the same. Of course, when using biogas, the quality of the gas is

very important. For oil CHPs the maintenance costs of the natural gas CHPs can be assumed. With the increasing size of the CHPs a cost degression is given.

In the following, planning aspects and the cost-effectiveness of new installations are presented.

#### Case-Study: "Schneider-Gössl"

The "Schneider-Gössl" is located on the outskirts of Vienna. The building is splitted into a hotel and a "Heurigen"-restaurant. The hotel has 9 guest rooms and 20 beds and is open during the whole year.

A micro CHP was installed for heating, domestic hot water production and electricity generation. The micro CHP from Senertec company has an electric output of 5 kW and a thermal output of 12.3 kW. For covering the thermal peak loads, a Buderus low-temperature gas boiler with a thermal output of 44 kW was additionally installed. The micro CHP and the peak load boiler are fuelled with natural gas. For hot water storage a 1,000 litre storage tank is used.



Figure 4.51 Partial view of the lodge "Schneider-Gössl" (Source: Austrian Energy Agency)

Figure 4.52 shows the annual heat demand of the lodge, a picture of the installed micro CHP system, and the amount of heat generated by the micro CHP unit. The area in which the heat generation of the CHP is above the heat demand curve corresponds to the charging time of the storage tank. The operation time can be significantly extended by the installed hot water storage tank (with a volume of 1,000 I). Around 22% of the maximum thermal demand is covered by the micro CHP system.



Figure 4.52 Annual heat demand curve (blue line) and heat generation of the micro CHP system (purple line), photo: installed micro CHP system from Senertec company (Source: Austrian Energy Agency)

The analysis concerning the pay-back period is based on a dynamic calculation.

Figure 4.53 shows the development of the accumulated discounted cash flow. The payback period for the investment in the installed micro CHP system is calculated to 10.2 years.



Figure 4.53 Accumulated discounted cash flow showing a pay-back period of 10,2 years (Source: Austrian Energy Agency)

The analysis shows that the "existing" heating system consisting of a micro CHP unit in combination with a peak load boiler is a good technical solution. The installed micro CHP system achieves an appropriate operation time in a heat driven operation mode and supplies almost 38% of the electricity demand of the building.

Due to the frame conditions in Austria and due to the energy situation at "Schneider Gössl", the deployment of the micro CHP system shows an economic efficiency which leads to a pay-back period of 10.2 years. The main reasons for this result are:

- High tariff for the electrical energy supply
- Significant amount of avoided electric energy supply from the grid
- Subsidy of 30% for the investment and installation costs of the micro CHP system
- Reimbursement of tax on fuel used for the micro CHP unit

## Case-Study: "Marianneum"

The "Marianneum" is a religious centre located on the outskirts of Vienna. The lodge is open during the whole year. The building was renovated in 1987 and has 34 guest rooms and 66 beds. The following energy systems have been installed in the "Marianneum". For heating, domestic hot water production and electricity generation a micro CHP was integrated. The installed Buderus micro CHP is a modulating unit (9–18 kW<sub>el</sub>/ 17–34 kW<sub>th</sub>) fuelled with natural gas. Additionally an oil boiler with a thermal output of 116 kW is used to cover the thermal peak loads; furthermore two storage tanks with a volume of 1,000 litre (each) are used for the same purposes. Two heat pumps provide extra heat for the winter period. Each heat pump has a thermal output of 4.5 kW. For additional hot water production a thermal solar system with a collector area of 20 m<sup>2</sup> was installed (thermal output appr. 9,000 kWh/a).



Figure 4.54 "Marianneum" (Source: Austrian Energy Agency)

Figure 4.55 shows the annual heat demand which has to be supplied by the micro CHP and the peak load boiler, a picture of the installed micro CHP system and the amount of heat generated by the micro CHP unit. The area in which the heat generation of the micro CHP is above the heat demand curve corresponds to the charging time of the storage tanks. The operation time can be extended by the installed hot water storage tanks (with a volume of 2,000 l).

Around 53% of the maximum thermal demand is covered by the micro CHP system.



Figure 4.55 Annual heat demand curve (blue line) and heat generation of the micro CHP system (purple line), photo: installed micro CHP system from Buderus company (Source: Austrian Energy Agency)

Information concerning the pay-back period is based on a dynamic calculation. Figure 4.56 shows the development of the accumulated discounted cash flow. The payback period for the investment in the installed micro CHP system is 7.2 years.



Figure 4.56 Accumulated discounted cash flow showing a pay-back period of 7.2 years (Source: Austrian Energy Agency)

The analysis of the energy situation shows that the "existing" micro CHP unit represents a good technical and economic solution. The micro CHP supplies almost 40% of the electricity demand of the building. Due to the frame conditions in Austria and due to the energy situation at the "Marianneum", the deployment of the micro CHP system shows an economic efficiency which leads to a pay-back period of 7.2 years.

The reasons for this good pay-back period are:

- High tariff for the electric energy supply
- Significant amount of avoided electric energy supply from the grid
- Subsidy of 30% for investment and installation costs of the micro CHP system
- Reimbursement of tax on fuel used for the micro CHP unit

#### 4.2.4 Outc ome

The main objective of the work in Annex XIX was to receive a better understanding of the possibilities for all kinds of stationary fuel cells.

In recent years a lot of RTD activities concerning fuel cells were performed and therefore the fuel cell technology has improved significantly. However, the support of RTD and demonstration projects is still very important. Main technical and economic issues to be overcome in order to achieve commercial fuel cell products for competitive markets are:

- Decrease investment-, O&M costs, and generally the complexity of the system
- Improvement of life-time, reliability and efficiency (decrease of own consumption of electricity)
- Optimisation of controlling software and of conventional systems (incl. hydraulic connection)

The advantages of fuel cells are seen in the high electrical efficiency, low emissions, low noise, high availability and grid independency.

All subtasks will continue in the new period from 2009 till 2013. In the new period there will be 6 subtasks. New ideas, approaches and issues will also be considered in the new period (Annex 25). The new subtasks are:

Subtask 1: Small stationary fuel cells - subtask leader: FZJ Germany

Subtask 2: Fuel for fuel cells - subtask leader: MTU Onsite Power Germany

Subtask 3: Fuel cell plants components - subtask leader: VTT Finland

Subtask 4: Analyzing design, operating and control strategies for stationary fuel cell systems in globally divers installation contexts – subtask leader: Sandia USA

Subtask 5: Demonstration project, follow up of fuel cells technology status by examining demonstration projects and their influence on market development – subtask leader: ENEA Italy and Thoma & Renz Switzerland.

Subtask 6: Market status for stationary fuel cells - subtask leader: E.ON Sverige AB Sweden

# 4.3 Task XXI: "Fuel Cells for Portable Applications"

## 4.3.1 Aim of the task

The main objective of this application oriented task was to assist – through international cooperation – the development of portable fuel cells towards commercialisation through:

- the exchange of information to tackle complex problems in stack and systems design and operation,
- the consideration of end-user requirements on portable fuel cell operation with the goal to minimize size and costs of cells and systems,
- the study of alternative materials in case traditional concepts are too expensive or are too short-lived.

# 4.3.2 Metho dology

The subtasks of the Annex XXI covered the following issues:

#### Subtask 1: System analysis for portable fuel cells

The aim of the task was to tackle complex problems in stacks and systems by modelling of mass and energy flows and also to perform system analysis for portable applications, e.g. analysis of energy demand of the auxiliary components, water- and heat management, etc. Information about rules and regulations concerning operation, storage and transport of portable fuel cells were collected.

#### Subtask 2: System, stack and cell development for portable fuel cells

Test of operation concepts for portable systems, e.g. cold start concepts, dynamic load following, etc. were conducted. The design goals for portable systems, such as comparison with existing techniques as Li-Ion accumulators or batteries, deriving of specifications for stacks and systems were evaluated. Also fuel storage concepts suitable for the specific demands of portable applications were investigated. Systems for portable applications, e.g. DMFC systems with a liquid anode feed or micro-reformer systems with small PEFC systems were designed.

#### Subtask 3: Materials under operating conditions / materials innovation

Stack and system tests with regard to the power output, efficiency and lifetime were carried out. Cost aspects, e.g. use of cheap and easy to manufacture materials were taken into account.

Agencies from nine countries participated in the Annex XXI:

- TU Graz (Austria)
- The Government of Canada (Canada)
- Risø National Laboratory (Denmark)
- VTT Technical Research Center of Finland (Finland)
- Forschungszentrum-Jülich GmbH (Germany)
- Ente per le Nuove Tecnologie, l'Energia e l'Ambiente, ENEA (Italy)

- Korea Institute of Science and Technology (Korea)
- Netherlands Energy Research Foundation, ECN (Netherlands)

The task leader was the Forschungszentrum Jülich GmbH, Germany, and Jürgen Mergel was acting as operating agent.

#### 4.3.3 Activ ities

#### 4.3.3.1 Subtask 1: System analysis for portable fuel cells

*Germany (FZ Jülich):* Portable DMFC systems as a replacement for lead-acid batteries in the 100 W to kW class were presented. A special focus was put on testing the stacks at varying operating conditions. Figure 4.57 shows a warehouse truck with DMFC system.



Figure 4.57 Warehouse truck with DMFC system; 7 kW peak; 0.8 kW average; fuel: pure methanol (Source: Forschungszentrum Jülich, Germany)

*Italy, ITAE, CNR:* A PEFC bicycle with assisted pedalling was presented, see Figure 4.58. Also a PEFC 500 W (CNR-ITAE; Italy) stack was developed. Emphasis was laid on the design, realization and test of the 500 W stack for medium temperature (120°C) by using composite MEAs and a cell design totally developed and realized in CNR-ITAE labs.



Figure 4.58 250 W PEFC bicycle; hydrogen storage pressure: 0.7 - 0.8 bar; PEFC with 22 cells (31 cm<sup>2</sup> active area) (Source: ITAE, Italy)

*Japan, YAMAHA:* A DMFC scooter with a driving range of 125 km and a speed of 30 km/h was introduced, see Figure 4.59. As fuel a methanol water solution (54 mass-% methanol) was used. To overcome issues with rapid load change, a battery for hybridization was used. The system showed a durability of 1,500 h.



Figure 4.59 DMFC scooter with a system efficiency of more than 32% (Source: YAMAHA, Japan)

*Korea, KIER:* A DMFC 800 W stack for scooters including 2 sub-stacks (45 cells/sub-stack) with a power density of 175 W/I was developed.



Figure 4.60 800 W DMFC stack for scooters (Source: KIER, Korea)

# 4.3.3.2 Subtask 2: System, stack and cell development for portable fuel cells

*Sweden, myfc:* A fuel cell system for recharging personal handhelds fuelled by hydrogen was developed. In Figure 4.61 a prototype of a charger is shown.



Figure 4.61 Prototype of charger 2.5 W / 2.4 V (Source: myfc, Sweden)

**Japan, NEC:** NEC developed a fuel cell technology to realize cordless personal devices. The targets were the development of a compact thin and small stack with a power density of 70 W/L and a higher fuel efficiency by new fuel control method. A prototype fuel cell pack of 30 W/L for personal device was introduced.



Figure 4.62 Prototype FC pack with a power output of 3 W (max. 5 W); external size: 110 mm x 45 mm x 20 mm (Source: NEC, Japan)

# 4.3.3.3 Subtask 3: Materials under operating conditions / materials innovation

*Canada, NRC:* NRC developed DMFC-MEAs on basis of dry powders, for example see Figure 4.63. The method showed high reproducibility due to the use of powders instead of inks. The powders were easy to store and cause little waste. In experiments same performance as MEAs produced with inks were shown. A mud-crack free structure of the electrodes was observed. The MEAs were tested up to 3,000 h of DMFC operation at 40 °C and 100 mA cm<sup>-2</sup>. No failure was identified.

Actual status of the developed DMFC MEAs:

- Power density: 115 mW/cm<sup>2</sup> @ 70 °C; 1M CH3OH; air
- Catalyst: Pt (cathode) + PtRu (anode) = 4 mg/cm<sup>2</sup>
- Flexible line coater



Figure 4.63 Method for producing DMFC-MEAs with dry powders (Source: NRC, Canada)

*Germany, Forschungszentrum Jülich:* The Forschungszentrum Jülich developed DMFC-MEAs and new coating techniques for large scale producing of MEAs were installed:

Knife-coating (Flexible line coater)

- Slot-coating (vertical coater for slot coating)
- Screen-printing
- Robot assisted stacking of MEA components
- Robot assisted hot pressing of MEA

Actual status of the developed DMFC MEAs:

- Power density: 115 mW/cm<sup>2</sup> @ 70 °C; 1M CH3OH; air
- Catalyst: Pt (cathode) + PtRu (anode) = 4 mg/cm<sup>2</sup>



Figure 4.64 Flexible line coater (Source: Forschungszentrum Jülich, Germany)

#### 4.3.3.4 Activities in Austria for portable fuel cells

The Austrian activities were presented by the Fuel Cell Laboratory, TU Graz. The work concentrated on fuel processing for portable fuel cells.

#### Ethanol reforming for hydrogen production for fuel cells

Presently almost all energy is derived directly or indirectly from non-renewable sources as fossil fuels, thus causing the release of large quantities of greenhouse gases. The depletion of fossil fuel reserves as well as their location in countries different from the consumers leads to concerns in terms of security of energy supplies. Together with the aim of reducing greenhouse gas emissions considerable interests in alternative energy technologies have been generated in terms of increasing energy efficiency and renewable fuel resources, such as bio-alcohols, methane from biogas, bio-diesel and hydrogen. Hydrogen fuel cells are the most promising systems for energy production, since they are more efficient and environmentally clean than conventional thermal machines. Natural gas is up till now the main source of hydrogen production; however, there is great interest in the development of alternative hydrogen production by employing renewable resources. Among various feedstocks, ethanol is very attractive because of its high hydrogen content, availability, non-toxicity and storage and handling safety [Lit 34]. More important, the so-called 2<sup>nd</sup> generation bio-ethanol can be produced renewably from lingo-cellulosic biomass, such as energy plants, agricultural and forestry residues or organic waste materials. It is a cleaner process than methane steam reforming, because of the almost fossil CO<sub>2</sub> "free" cycle, formed by photolysis of lingocellulosic material, ethanol production and reforming of ethanol.

The need for hydrogen sources from reforming renewable fuels, such as ethanol, in the range of 1-100 kW makes it necessary

- a) to develop new reactor strategies to obtain high power density fuel reformers, as it is not possible to downscale the existing technology of large-scale methane reforming (in the MW-range) to small fuel processor units achieving compact and low cost units,
- b) to develop new catalysts for ethanol reforming, as there are no commercial catalysts available. The necessary new reactor configurations for high power density reformers make it necessary to use highly active catalysts with a high life-cycle, as a catalyst's change during the reformer life-cycle is no option.

This research project deals with the development of a micro-structured fuel processor based for ethanol reforming.

The topics covered were:

- Establishment of an experimental facility to investigate the ethanol reforming process on catalyst powders as well as in micro-structured reactors.
- Process gas analysis based on a gas-chromatography and mass-spectrometry
- Investigation of the thermodynamics of the reforming process
- Catalysts development as there are no commercial catalysts available for ethanol steam reforming
- Coating of micro-structures
- Reaction tests of the micro-structured reactor

#### **Experimental facility:**

The catalyst test bed consisted of a catalytic fixed bed reactor set-up from the company PID Engineering & Technology in Spain. The unit allowed a fully automatic operation with pressures up to 100 bar and temperatures up to 900 °C. The gas flows were maintained via Bronkhorst mass-flow controllers, and the liquid flow was dosed via a Gilson HPLC-pump. The whole gas supply and vapour mixing system as well as the catalytic fixed bed reactor was placed in a hot-box, which can be heated up to 200 °C to avoid condensation in the lines.





Figure 4.65 Experimental facility: Catalytic test bed (left), Micro-Gaschromatograph (right) and Mass Spectrometer (middle) (Source: Fuel Cell Lab TUG, Austria) Figure 4.66 Hot-box with reactor heater and stainless steel reactor (Source: Fuel Cell Lab TUG, Austria)

The unit had to be optimized as there were problems with evaporator pulsations and coke formation inside the reactor configuration. A quartz glass reactor was used to avoid coke formation in atmospheric pressure applications.

## **Process gas analytics**

In order to investigate the catalytic activity of ethanol steam reforming, a complete analysis of the product gases of the reforming process is needed. Possible components in the product gas are:

- Non condensable gases:
   H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>, O<sub>2</sub>, CO<sub>2</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>
- Condensable vapors:
   Ethanol, H<sub>2</sub>O, Acetaldehyde, Acetone

The difficulty was to establish quantitative product-gas analysis. Condensation of the vapors was not possible due to acetaldehyde ( $T_{Boil}$ =20 °C). So a combination of gaschromatography for non-condensable gases and mass spectrometry for condensable vapors was developed, with nitrogen and argon as inert gas for quantification of the molar flux of the product gases.



Figure 4.67: Product gas analysis (Source: Fuel Cell Lab TUG, Austria)

The setup allowed a product gas analysis to be done within 3 minutes, which is especially important to investigate catalyst deactivation processes.

#### Thermodynamics of the ethanol reforming process [Lit 35]

The possible operational modes are:

- Steam reforming
- Partial oxidation
- Auto-thermal reforming

Steam Reforming is an endothermic process:

$$CH_{3}CH_{2}OH + H_{2}O \Leftrightarrow 2CO + 4H_{2} \qquad \Delta H_{r} = +298kJ / mol \tag{1}$$

Complete steam reforming operation is given by reaction

$$CH_{3}CH_{2}OH + 3H_{2}O \Leftrightarrow 2CO_{2} + 6H_{2} \qquad \Delta H_{r} = +347kJ / mol$$
<sup>(2)</sup>

whereas the maximum hydrogen-yield of ethanol steam-reforming can be fixed by 6 mole  $\rm H_2$  / mole ethanol.

Partial oxidation is, as indicated by the name, the incomplete oxidation of the feed. The oxidizing products deliver water, and therefore also steam reforming (1) takes place to produce hydrogen.

$$CH_3CH_2OH + 3O_2 \Leftrightarrow 2CO_2 + 3H_2O \qquad \Delta H_r = -1368kJ / mol \tag{3}$$

As the oxygen content in the feed stream is increased, the feed is completely oxidized and the maximum heat output is achieved.

Auto-thermal reforming is the combination of these two processes of steam reforming and partial oxidation, whereas a small amount of oxygen is added to the educts, in order to achieve a minimum energy input necessary to maintain the required reformer temperature.

The space of operation for a fuel processor for hydrogen production is depicted in Figure 4.68.



Figure 4.68 Different operating conditions for ethanol reforming (Source:[Lit 35])

The aim of the analysis was the study for the best mode of operation in terms of hydrogen yield per energy input, CO-content in the reformate and coke free operation.

Ethanol is thermodynamically fully converted at already low temperatures. The problematic point is the methane formation at low temperatures, see Figure 4.69.

From a thermodynamic perspective, ethanol at low temperatures is reformed to methane according:

$$CH_3CH_2OH \Leftrightarrow 0.5CO_2 + 1.5CH_4$$
 (4)

At increasing temperatures the methane reforming together with the water gas shift reaction takes place:

$$CH_4 + H_2 O \Leftrightarrow CO + H_2 \tag{5}$$





Figure 4.69 Reformate product composition in mole-fraction on dry basis for steam-reforming of ethanol at steam-to-carbon ratio S/E = 3 and 6, oxygen-to-ethanol ratio O/E = 0 and atmospheric pressure (Source:[Lit 35])

Figure 4.70 Coke formation boundary for ethanol steam-reforming and partial-oxidation as function of steam-to-ethanol ratio S/E (mole H<sub>2</sub>O/mole EtOH) and oxygen-to-ethanol ratio O/E (mole  $O_2$ /mole EtOH) (Source:[Lit 35])

From thermodynamic perspective, high temperature is necessary for the activation of methane. At higher temperature the water gas shift reaction produces a high CO content. Another problem to be avoided is the coke formation. The coke formation boundary for steam and auto-reforming as well as partial oxidation is shown in Figure 4.70. Partial oxidation (S/E=0) is most prone to coke formation. The steam-reforming (O/E=0) shows decreasing coke formation with increasing steam content. The auto-thermal operation (S/E>0 and O/E>0) shows a positive effect on reduced coke formation.

One problem to keep in mind when adding oxygen, is the dilution of the product stream from the nitrogen content in the air. The nitrogen content in the reformate stream for partial oxidation is shown in Figure 4.71.



Figure 4.71 Nitrogen-dilution of hydrogen effluent for partial oxidation. Nitrogen content on dry basis for different oxygen-toethanol ratios O/E = 0.00-2.50 at a steam-to-ethanol ratio of S/E =0.0 (Source:[Lit 35])

Figure 4.72 Plots of the thermodynamic equilibrium product mole fractions of hydrogen and carbon monoxide on a dry-nitrogen basis and hydrogen-yield [moleH2/moleEtOH] and coke formation [moleC/moleEtOH] for pure partial-oxidation with steam-to-ethanol ratio S/E = 0.0 (Source:[Lit 35])

Using an oxygen to ethanol ratio of O/E=1.5, which is necessary to lower the coke formation boundary to 500 °C, produces a nitrogen content of almost 60mol% on dry basis in the reformate stream. The partial oxidation reaction is therefore limited by the acceptable nitrogen dilution.

Additionally, the partial oxidation shows a maximum of hydrogen production and CO formation in the same parameter region, so that partial oxidation leads always to high CO levels in the reformate stream, which can be seen in Figure 4.72.

Additionally, the auto-thermal reforming operation has been explored. As discussed before, the auto-thermal operation can lower the coke formation boundary. A second effect expected was a reduced CO content in the reformate stream. This effect can be seen in Figure 4.73 and Figure 4.74.


Figure 4.73 Hydrogen content for autothermal operation at T = 500 °C (Source: Fuel Cell Lab TUG, Austria)

Figure 4.74 Hydrogen/CO ratio for autothermal operation at T = 500 °C (Source: Fuel Cell Lab TUG, Austria)

The introduction of small amounts of oxygen in auto-thermal operation can elevate the hydrogen content in the reformate stream. The  $H_2/CO$  ratio is only slightly increased by increasing the oxygen content, so that a strong effect on the reduction of the CO-content in the reformate stream is not seen be auto-thermal operation.

Additionally, the energy demand of the different reforming options has been investigated. The reforming system is shown in Figure 4.75. The evaporation energy, pre-heating energy, reforming energy are considered as input and the cooling energy to 120 °C is considered as output.



Figure 4.75 Reformer energy-balance (Source:[Lit 35])

$$Q_{\text{Process}} = Q_{\text{Evaporation}} + Q_{\text{Heating}} + Q_{\text{Reformer}} - Q_{\text{Cooling}}$$
(6)

The process energy needed is related to the moles of hydrogen in the reformer effluent. So the amount of energy is specified, which is needed to produce a mol of hydrogen in the reformate stream.

Figure 4.76 shows a comparison of the energy demands for different operating conditions. Looking at the process energy, which is the sum of evaporation, pre-heating, reforming and cooling, steam reforming is most energy intensive compared to auto-thermal reforming. Partial oxidation has a negative process energy, which means there is more heat produced in the reformer, than needed. A comparison on basis of the process energy does not show the whole picture. A better comparison is the total energy. This is the process energy plus the energy from combustion of ethanol for the supply of heating energy. This shows a com-

pletely different picture, whereas the partial oxidation demands the most energy per produced hydrogen. Steam reforming and auto-thermal reforming are comparable from perspective of energy demand.



Figure 4.76 a.) Total process energy required to produce one mol hydrogen in reformer effluent; b.) Total energy demand, process energy plus lower heating value of ethanol in the feed to produce one mol hydrogen in the reformer effluent. POX ... partial oxidation, SR ... steam reforming, ATR ...auto-thermal reforming. Process Energy is the sum of energy necessary for evaporation, heating, reforming and cooling. (Source:[Lit 35])

#### Catalyst development

The ethanol reforming reaction mechanism is not thoroughly understood till now. There are two main reaction pathways in the beginning: the dehydrogenation to acetaldehyde and the dehydration to ethylene, which are further reacted to hydrogen and CO.





Figure 4.77 Reaction network for ethanol reforming (Source: [Lit 36]

Figure 4.78 bond energy for ethanol (Source: [Lit 37]

The catalysts have to be active for C-C-bond breaking and further reaction of the methylgroups. A problem, as it is for all hydrocarbon processing, is the coke formation. A literature review showed that Ni-based catalysts, which are standard for reforming catalysts, show a strong coking. Due to decomposition of ethanol into  $CH_4$  and CO, these catalysts showed a low hydrogen selectivity. The reaction temperatures have to be increased in order to activate methane steam reforming for a better hydrogen yield. Rh-based catalysts show a very good steam reforming capability, but methane selectivity demands higher reactions temperatures. Coking is strongly reduced in comparison to the Ni-catalyst, but is still an issue. Another problem of Rh is the high price for the metal. Co-based catalysts are an interesting option, as the Co metal show a strong C-C bond breaking activity already at low temperatures. The reaction pathway is of either dehydrogenation to acetaldehyde, and the dehydration to ethylene is strongly dependent from the used support material. On acid surfaces as  $AI_2O_3$  dehydration occurs and the intermediate ethylene is supposed to be a major coke precursor [Lit 38] due to polymerisation of ethylene. On basic surfaces as MgO dehydrogenation to acetaldehyde takes place, which is further reformed to synthesis-gas.

From literature research,  $CeO_2$ - supported Co and Ru catalysts were chosen.  $CeO_2$  as a support has the advantage of an oxygen storage property. The CeO2 can be reduced and re-oxidised under reaction conditions, which is supposed to be helpful in the reduction of the coke formation. Ru is cheaper than Rhodium and has noble metal catalytic properties.

Different catalyst preparation methods have been developed:

- Impregnation-method
- Solution-combustion-method
- Co-precipitation-method

Figure 4.79 to Figure 4.82 show the conversion, hydrogen yield, coke formation as well as carbon product selectivity for a 10% Co/CeO<sub>2</sub> and a 10% Ru/CeO<sub>2</sub> catalyst.



Figure 4.79 Carbon products of ethanol reforming  $10\%Co/CeO_2$ , S/E = 6, W/F = 3,135 mlEtOH/gKath (Source: Fuel Cell Lab TUG, Austria)

Figure 4.80 Conversion, hydrogen-yield ethanol reforming Co/CeO<sub>2</sub>, S/E = 6, W/F = 3,135mIEtOH/gKath (Source: Fuel Cell Lab TUG, Austria)





Figure 4.81 Carbon products of ethanol reforming 10%Ru/CeO<sub>2</sub> , S/E = 6, W/F = 3,135 mlEtOH/gKath (Source: Fuel Cell Lab TUG, Austria)

Figure 4.82 Conversion, hydrogen-yield ethanol reforming Ru/CeO<sub>2</sub>, S/E = 6, W/F = 3,135mIEtOH/gKath (Source: Fuel Cell Lab TUG, Austria)

Both catalysts show a good steam reforming activity, with full conversion around 450 °C. The product selectivity is similar for both, whereas the Co-catalysts produce more acetone and methane side products. At high temperatures both catalysts reach the thermodynamic hydrogen yield. The major difference can be seen in the coke formation, where the Co-catalysts produce far more coke in comparison to the Ru-catalysts. The coke formation occurs at lower temperatures, and by elevating the reaction temperature coke gasification takes place.

In the Figure 4.83 the coke build-up on  $Co/CeO_2$  catalyst is visible in form of a size extension of the catalytic bed.



Figure 4.83 Coke-formation at Co-/CeO<sub>2</sub> catalyst (Source: Fuel Cell Lab TUG, Austria)

The influence of different catalyst preparation methods on the coke formation on  $Co/CeO_2$  catalyst was determined and coke removal strategies were investigated. The results of this research is to be published in future.

# Water-gas-shift reaction in micro-structured reactors for gas purification of hydrogen for portable PEM-fuel cells

High- and low-temperature water gas shift reactions were examined for the ability to decrease the CO fraction of the synthesis gas down to a level tolerable for proton exchange membrane fuel cells. These gas purification methods had the advantage of a higher hydrogen yield due to the conversion of CO into  $H_2$  and thus leading to a higher overall efficiency of the fuel cell system. First, the performance of micro-structured reactors coated with different commercial catalysts was investigated under different temperatures, gas flow rates, and gas compositions. In the first part of this experimental work the preparation and optimization of catalysts for the water-gas shift reaction in a micro-channel reactor were analyzed with the aim of generating hydrogen for mobile fuel cell systems. The catalysts had to demonstrate high catalytic activity and strong adhesion to support foils and prove long-term stability, especially during start-up/shut-down procedures. In the first step the coating of microchannel metallic plates was performed with nanopowders/support materials by spin-coating methods. The dispersion stability and the detailed interaction between particles were determined by zeta-potential analysis. In the second step the catalytic active materials were deposited during the impregnation method and accomplished the forming of the two catalyst systems. Well-adhering wash coats were obtained on micro-structured foils. A detailed investigation of the influence of different variables on the coating uniformity has led to a coating of catalysts inside stainless steel micro-channels with good adhesion. The adhesion was highly dependent on the nature of binder, initial particle sizes, pH and also surfactant addition. Afterwards the catalysts were tested at reaction temperatures between 200 °C and 450 °C in one-stage water-gas shift reactors. The catalyst loading, the surface area and the platinum amount had a significant effect on the activity of both Pt/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> and Pt/CeO<sub>2</sub> catalysts.

A fuel processor is a complex device which converts conventional fuels such as gasoline or a renewable fuel like ethanol into hydrogen. Gas purification devices need to be installed downstream the reformer to reduce the CO amount to a tolerable level for HTPEM. Inexpensive fabrication techniques and lower catalyst cost may play a significant role in the overall fuel processor cost. Here the micro-structured devices offer many possibilities for cost reduction due to the improved mass transfer in small channel systems, which allow a highly efficient usage of the catalyst. Micro-structure devices allow rapid system stabilization due to the fast dynamic response because of the low residence times applied. Micro-structured reactors not only enable improved catalyst effectiveness but also work at a high efficiency/volume ratio, thus leading to a material and size reduction potential.

In this project, the alumina and cerium dioxide wash coats were obtained by spin coating. The coating parameters such as suspension-pH, binder (PVA) molecular weight and solid loading were varied to optimize the homogeneous, well adhered layer with good thermal stability and high surface area for water-gas shift reaction on micro-structures.

Furthermore, this project investigated the possibilities of using micro-structured reactors after adapting Pt-catalysts such as  $Pt/CeO_2/Al_2O_3$  and  $Pt/CeO_2$  on micro-channel foils for the one step water-gas shift reaction at low reaction temperature. Micro-structured reactors allow the creating of more compact and efficient systems due to new micro-channel devices coated with high surface area catalysts. Therefore, the weight of expensive precious metal catalysts can be reduced.

### Zeta potential

The charge interactions between particles were described by the zeta potential. The results from zeta potential measurements allowed controlling the dispersion process, especially the agglomerates rising, which influenced the defects appearance in the support layer.

Figure 4.84 illustrates the zeta-potential of CeO<sub>2</sub>- and Al<sub>2</sub>O<sub>3</sub>-suspensions as a function of pH. For this type of measurements, the powder particles were suspended in 10 mM KCL. Afterwards the pH was adjusted with HCl and NaOH solutions. The effect of the pH value on the zeta potential was significant for the Al<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> powders. The zeta potential of alumina was increased from 0 mV (isoelectric point) at pH = 8 to 38 mV at pH = 2. The CeO<sub>2</sub>-powder showed an elevated stability with rising pH-values. The isoelectric point was found at pH 3.5 and the highest particle dispersion of - 22 mV at pH 8. For the next steps of this experimental work the pH–values 8 and 4 were chosen for CeO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>-suspensions optimization.



Figure 4.84 Zeta potential as a function of pH for  $CeO_2$ - and  $Al_2O_3$ -powders. (Source: [Lit 39])

#### Effect of surfactant and binder addition

Before the suspension preparation, the particle diameters were measured in natural pH and without any addition of binder. The differences of particle diameters between the two powders were significant in this surrounding environment. Furthermore the dependence of effective particle size on the surfactant/suspension weight-ratio and on the flocculation level was measured. The important influence of surfactant concentration on the particle diameter changes was observed for CeO<sub>2</sub>. The agglomerates diminished, when the surfactant weight was extended. The addition of a small amount of surfactant to the suspension with 0.1-ratio was sufficient to achieve the smallest particle diameter of 8.7 nm.

To improve adhesion between the layers and the substrate metal made of stainless steel during drying and calcination procedure, and, moreover, to achieve crack-free catalyst layers, poly-vinyl alcohol (PVA) was chosen as a binder. The influence of PVA-molecular weight, the concentration at stable pH-value (3 for  $Al_2O_3$  and 8 for  $CeO_2$ ) versus effective particle diameter was analyzed. The highest dispersion of solid medium was observed for  $CeO_2$ -suspension with PVA-molecular weight of 18,000 g/mol and concentration of 20 wt.% in  $H_2O$ . For the  $Al_2O_3$  dispersed powders in suspension, the two smallest comparable parti-

cles were achieved after addition of PVA with molecular weight of 13,000 g/mol and 31,000 g/mol and the concentration of 10 wt.% in H<sub>2</sub>O. The largest agglomerates in the range of 117–166  $\mu$ m appeared in CeO<sub>2</sub>-suspension after addition of the highest PVA-molecular weight of 124,000 g/mol.

The CeO<sub>2</sub>-coatings showed the highest adhesion and long-term stability during the thermal shock tests compared to the  $Al_2O_3$ -layers. No coating losses were observed for the Ce1(I-coat) and-Ce2(II-coat)-samples. However, complete stabilization was found for all CeO<sub>2</sub>-layers after different shock times. The Ce2(I-coat)-sample showed the maximum of weight loss of 7.5 wt.% after the third shock (Figure 4.85 a, b). In case of  $Al_2O_3$ -layers, the adhesion was still increasing with PVA-molecular weight for all Al-1-samples. The first layer stabilization was noted for the Al-2(I-coat) and then for the Al-3(I,III-coat)-samples with the minimum of loss in range of 2 wt.% after fifth shock (Figure 4.85 c).



Figure 4.85 Mass loss of  $AI_2O_3$  and  $CeO_2$ -washcoats for three different layer thicknesses and PVA addition (a. Mw=13,000; b. Mw=31,000; c. Mw=124,000) vs. frequencies in the thermal shock test. (Source: [Lit 39])

The CeO<sub>2</sub>-coating showed the perfect heat resistance during the thermal shocks with no weight loss. Bulk densities of 1.31-1.42 g/cm<sup>3</sup> were achieved for these coatings due to the PVA-molecular weight increase.

Significant differences in the surface texture of both  $Al_2O_3$  and  $CeO_2$ -supports were found. Based on the SEM-micrographs, it can be seen that the  $Al_2O_3$ -layers had an uneven, porous structure with some nm-cracks but the highest BET-surface area (216 m<sup>2</sup>/g). On the other hand, the denser packed particles of the CeO<sub>2</sub>-layer formed an even and well adhesive layer with the highest total specific surface area.

# Effect of impregnation number on catalyst loading

For the first impregnation, using the slurry with 0.7 wt.% PVA (124,000 g/mol), the loading of coating was 1.7 wt.%. When the PVA<sub>1</sub> (13,000 g/mol) or PVA<sub>2</sub> (31,000 g/mol) with a slurry concentration of 7 wt.% was used, the loadings of the coatings could reach 8.9 wt.% or 15. wt.% respectively.

The loading of the catalyst by impregnation method depends on the support density and the material loading in the micro-channels. The different slurry viscosities were achieved by the addition of three PVA's (13,000, 31,000 and 124,000 g/mol), which influenced the bulk structure (densities) and also changed the layer loading. Therefore multiimpregnation was required, because generally, a suitable catalyst loading can not be achieved by a single impregnation. For the second application by an impregnation process, the loading amount on the support  $Al_2O_3$  was much lower than the first loading. The higher coating loading during first impregnation was caused by the increased capillary attraction of the support and by the particle absorption on the surface of  $Al_2O_3$ .

The results showed that the higher support loading with its lower bulk density was advantageous to enhance the catalyst loading. In this experimental work the Pt-impregnation was carried out in the channels under one run on both CeO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>-supports. It has been found out that if the loading of aqueous solution with catalyst precursors was too high, the coating deposited on the supporting Al<sub>2</sub>O<sub>3</sub>- or CeO<sub>2</sub>-coat peeled off easily from the micro-channel platelets during the calcination process. This was also the reason of choosing the multiimpregnation to achieve 1 wt.% of Pt on the CeO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>- layers.

# Water gas shift (WGS) - reaction tests

The WGS reaction is reversible and exothermic and due to its moderate exothermicity the WGS reaction is thermodynamically unfavourable at elevated temperatures. This is illustrated by the continuous decline and eventual change in the Gibbs free energy as a function of temperature, and by the corresponding decreasing equilibrium constant as temperature increases. Of course, the kinetics of the catalytic reaction are more favoured at higher temperatures. However, the higher operating temperature is favourable for smaller reactor size due to fast kinetics. The water content has a strong influence on the CO-conversion. The steam entering into the WGS reactor can be varied by controlling the water amount added into the reformer and by injecting water. In addition, CO, CO<sub>2</sub> and H<sub>2</sub> concentrations are mainly dependent on the reformer operating temperature, which in turn determines the thermodynamic limitations.

When the molar steam to dry gas (S/G) ratio in the reactor was rised from 0.24 to 0.72 the COconversion increased significantly for all catalysts. The highest conversion levels of 70% and 63.5% were found for the catalysts with the weights of 168 mg and 82 mg at 300 C and 350 °C under 70 ml n/min of total gas flow and 0.24 steam to gas (S/G) ratio.

It is important for fuel cell systems to determine the effect of WHSV (weight hourly space velocity) to prove its performance as it shows operating conditions after the application of the present water gas

shift catalysts. Due to the lower WHSV, the reactant gases had more time to be adsorbed by the catalyst thus leading to a higher CO conversion at a specific temperature.

To achieve 94.4% CO-conversion, a temperature of 250 °C and a WHSV of 1.3 N I/(h  $g_{cat}$ ) were necessary. At 300 °C and a WHSV of 25 N I /(h  $g_{cat}$ .), the conversion value was 4.4% lower. The CO-conversion value of 93% was the second best result and was achieved at 300 °C with a WHSV of 29.3 and 25.1 N I /(h  $g_{cat}$ ). The following conversions: 92%, 90% and 89% were resulted with the WHSVs of 41.2 (23.8) N I /(h  $g_{cat}$ ), 14.3 (51) N I /(h  $g_{cat}$ ) and 43.9 (72) N I /(h  $g_{cat}$ ) respectively (Figure 4.86)



Figure 4.87 CO-conversion over Pt/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>-catalyst samples; S/G=0.72; total feed flow rate: 40 ml n/min (dashed line), 70 ml n/min (continuous line). (Source: [Lit 40])

The well-adhered coats of CeO<sub>2</sub>-and Al<sub>2</sub>O<sub>3</sub> were prepared by stabilizing nm-powders such as Al<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> in suspensions and by depositing them on the metallic micro-channel plates by the spin coating method. The dispersion stability and the detailed interaction between alumina and cerium dioxide particles were demonstrated by zeta-potential measurements, which allowed controlling the flocculation and dispersion stages for solid medium in the suspension. The zeta-potential values enabled to find the maximum stability at pH of 4 and 8 for the CeO<sub>2</sub>-and Al<sub>2</sub>O<sub>3</sub>-suspensions respectively. The addition of PVA-binders with different molecular weight and concentration into the suspension influenced the particle diameter changes of the solid material. A small amount of surfactant with a 1/10-ratio was added in to the slurry and significantly increased particle dispersion. The highest resulting Al<sub>2</sub>O<sub>3</sub> surface reached 216.4 m<sup>2</sup>/g. The catalyst layers showed strong adhesion to support foils and proved long-term stability, especially during several start-up/shut-down procedures.

The Pt/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> catalyst has been coated by spin-coating method into micro-channels of stainless steel platelets. An optimum of platinum and ceria was resulted by their solutions impregnation on alumina and ceria carriers. The high thermal catalyst stability with high activity was proved under the condition of water-gas shift reaction. The 1 wt.% Pt/24 wt.% CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> catalyst was the best performing catalyst for the WGS-reaction compared to the 1 wt.% Pt/CeO<sub>2</sub>-catalyst and the industrial 2 wt.% Pt/CeO<sub>2</sub>-catalyst, especially in case of a low reaction temperature (250 °C). The 94.4% of CO-conversion was close to the thermodynamic equilibrium of the water-gas shift reaction at a WHSV of 14.3 N I/(h  $g_{cat}$ ). The 12 vol.% carbon monoxide content in the outlet reformat gas was decreased to 0.6% at 250 °C in one stage WGS-reactor.

# 4.3.4 Outc ome

During the last years valuable development work was done in the field of portable fuel cells. Especially the design of new systems with high power density lead to a wide range of possible applications. In first concepts small units in portable devices and powerful stacks for propulsion of wheel chairs, bikes and scooters were proofed. The results were most auspicious. The achieved high durability, excellent reliability and decrease of costs of portable fuel cell systems are motivation for further development. Now the phase of testing the systems under real operating conditions has started. The aims for the further development can be summarized to:

FC Stack / MEA:

- Improvement of power density
- Improvement of MEA performances and durability, and better quality control to minimize performance variation among cells
- Reduction of the platinum-loading and improvement of bipolar plate manufacturing.

Power generation system including BoP, secondary batteries and controls:

- Maximization of the system efficiency
- Maintaining of the water balance in the system
- Detoxification of the emission gas
- Assurance of reliability

Product development:

- Better product concepts and quality assurance to exceed customers expectations / requirement
- Assurance of fuel quality and establishment of fuel-supply network
- Cost reduction

# 5 Outlook for the new period 2009 - 2013

During the last period (2004 - 2008) six annexes were in progress. There were three technologybased annexes (MCFC, SOFC and PEFC) and three application-based annexes (stationary, portable and transportation applications). The application-oriented activities are becoming more important due to the fact that fuel cells reach into the demonstration phase prior to market deployment.

The structure of the programme is maintained. The application annexes will focus on implementation and application issues and the technology annexes will concentrate on research and technology development.

Table 5.1 shows the new annexes of the five-year work programme from 2009 till 2013 including objectives and duration. The new annexes are numbered from 22 to 27 (with the annex numbers no longer shown in Roman characters).

No.	Annex	Objectives	Duration
22	Polymer Electrolyte Fuel Cells	To reduce the cost and improve the perform- ance of PEFCs, DMFCs and corresponding fuel cell systems.	2009–2013
23	Molten Carbonate	To assist the commercialisation of MCFC systems through collaborative research and development.	2009–2013
24	Solid Oxide Fuel Cells	To assist, through international co-operation, the development of SOFC technologies.	2009–2013
25	Fuel Cells for Stationary Applications	To understand better how stationary fuel cell systems may be deployed in energy systems.	2009–2013
26	Fuel cells for Transporta- tion	To understand better how fuel cells may be deployed in transportation applications.	2009–2013
27	Fuel cells for Portable Applications	To assist, through international co-operation, with the development of portable fuel cells towards commercialisation.	2009–2013

Table 5.1 New annexes of the IEA Advanced Fuel Cells, objectives and duration (Source: IEA AFC)

Even though the Annex structure is planned to remain the same, it is proposed also to focus on issues that will promote the further development of the different technologies as well as topics that will facilitate realistic market introduction of the technologies. Therefore it is appropriate that the programme expands its work on application- and market-orientated issues, whilst continuing to address technology development and information management.

# Follow-on activities of existing annexes

In the follow-on activities based around the existing annexes the following issues will be considered [Lit 2]:

# Annex 22: Polymer Electrolyte Fuel Cells

 Identification and development of improved stack materials: membranes, electrode catalysts, bipolar plates, cells and stack assemblies.

- Resolution of stack and system issues such as contaminants, humidification and thermal management, operating environments and duty cycles, rapid-start, durability, freeze-thaw cycling, and characterisation of materials and components.
- R&D on direct fuel polymer electrolyte fuel cells, e.g. Direct Methanol Fuel Cells, including cell
  materials research, investigation of effects of operating conditions and stack/system modelling.

### Annex 23: Molten Carbonate Fuel Cells

- Improvement of performance, endurance, and cost effectiveness, for cells and stacks.
- Development and optimisation of MCFC system for various applications by evaluating performance of previous demonstrations or early market products.
- Identification of present and envisaged problems to be solved for rapid and further market penetration.
- Identification of possible opportunities for collaboration.

# Annex 24: Solid Oxide Fuel Cells

 Continuation and intensification of information exchange on SOFC through annual workshops and topic meetings, focusing on durability and costs of SOFC stacks and systems.

# Annex 25: Fuel Cells for Stationary Applications

- Evaluation of major demonstration projects.
- Identification of new early commercial applications for stationary fuel cells.
- Fuels for fuel cells, including locally produced fuels.
- Economic factors for market introduction.

#### Annex 26: Fuel Cells for Transportation

- Advanced fuel cell systems for transportation.
- On-board hydrogen storage.
- Hydrogen infrastructure.
- Technology validation and economics.

# 6 Summary

The aim of the IEA Advanced Fuel Cells programme is to enhance the state of understanding of all contracting parties in the field of advanced fuel cells, through a co-ordinated programme of research, technology development and system analysis on Molten Carbonate (MCFC), Solid Oxide (SOFC) and Polymer Electrolyte Fuel Cell (PEFC) systems. There is a strong emphasis on information exchange through task meetings, workshops and reports. The work is undertaken on a task-sharing basis with each participating country providing an agreed level of effort over the period of the task.

The current participants are: Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Italy, Japan, Korea, Mexico, Netherlands, Norway, Sweden, Switzerland, Turkey, UK and USA.

The five-year programme from 2004 till 2008 was structured in three technology oriented annexes and three application/market oriented annexes. The annexes will continue in the new period of the Implementing Agreement from 2009 till 2013.

The specific Austrian goals of this participation are as follows:

- Integration of Austrian cutting-edge RTD activities in International RTD emphasis
- Transfer of International know-how to (and in) Austria
- Promotion and mobilisation of Austrian applied and basic RTD (by gaining IEA know-how)
- Strengthening the competitiveness and internationalisation of the Austrian economy by increased RTD contributions, in general.

The participation of Austria has been performed by a consortium consisting of the Austrian Energy Agency and the Fuel Cell Laboratory (TU Graz) for the Annexes XVI, XIX, XXI. This consortium guarantees both technical-scientific and socio-economic contributions on a high level. Furthermore, there was a partnership with Energie AG Oberösterreich, that delivers application oriented contributions within Task XIX. Furthermore, the Austrian Agency for Alternative Propulsion Systems (A3PS) contributed in Annex XX during the last two years.

The Fuel Cell Laboratory (TU Graz) participated in **Annex XVI** with the R&D tasks characterisation of high temperature PEM fuel cells and development of carbon nano fiber based electrode material. The impacts of fuel contaminants and operating temperature on the performance of a high temperature fuel cell were investigated to determine optimum operating conditions.

Due to their high electric conductivity and the corrosion resistance, carbon nanofibers (CNFs), are regarded as an ideal alternative for electrode materials in PEMFCs. Catalysts were deposited on the carbon nanofibers and electrodes for PEM fuel cells were prepared by a new method. The electrodes showed high performance and durability.

Work performed within **Annex XIX (Stationary Applications)** focussed on the analysis of fuel cell demonstration projects executed by Austrian utility companies (sector: micro CHPs with an electrical power output between 1 and 5  $kW_{el}$ ). In total five fuel cell projects were carried-out in the years 2000–2006. The results of these projects are summarised below; these results may be also regarded as input for future R&D activities:

- Decrease of investment-, O&M-costs, and generally the complexity of system
- Improvement of life time, reliability and efficiencies (decrease own consumption of electricity)
- Optimisation of controlling software and of conventional systems (incl. hydraulic connection)

Based on the present results achieved by the demonstration projects, an implementation of fuel cells in competitive markets still requires major efforts. Other activities included analysis of the economics of conventional state-of-the-art CHP systems (based on ICE technology) serving as benchmark for future development requirements for fuel cells (considering comparable power outputs).

Within **Annex XXI**, contributions have been made by the Fuel Cell Laboratory (TU Graz). Experimental and simulation results regarding hydrogen production from ethanol in microreactors were introduced to the expert group. The hydrogen is used as fuel in portable fuel cells. Ethanol was found to be an excellent energy source for portable systems due to its high energy density. Ethanol is not toxic if compared to methanol and therefore much easier to handle.

Dissemination and networking activities included a project specific website (<u>http://www.energyagency.at/fileadmin/aea/pdf/energietechnologien/projekt-AFC.pdf</u>), furthermore a series of articles/posters, presentations and participations at workshops were performed. The Fuel Cell Laboratory initiated an international summer school, that received international good response.

In November 2008 the workshop "Fuel Cells: Deployment status and current product development" was held on the TU Graz. Speakers from active Austrian companies (such as AVL List GmbH, Fronius International GmbH) and international speakers from Germany, Switzerland and Denmark presented their work. Interested Austrian companies and institutions used this free workshop for information exchange about RTD activities [Lit 41].

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