

**FINAL REPORT OF THE IEA ADVANCED FUEL CELLS
IMPLEMENTING AGREEMENT (1996 – 1999)**

**Draft B
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Abstract

This report summarises the work and achievements of the IEA Advanced Fuel Cells Implementing Agreement over the period 1996-98. It comprises three sections:

- An introduction to fuel cells and the IEA Advanced Fuel Cells Implementing Agreement
- A summary of the key achievements of the Implementing Agreement over the period 1996-98
- Summary reports from each of the five Annexes in force during the period:
 - Annex VI - Molten Carbonate Fuel Cells under real operating conditions
 - Annex VII - Solid Oxide Fuel Cells under real operating conditions
 - Annex VIII- Collaborative Research on Polymer Electrolyte Fuel Cells
 - Annex IX - Fuel Cell Systems for Stationary Applications
 - Annex X - Fuel Cell Systems for Transportation

The Implementing Agreement has been extended to 2003 and thus continues to contribute to the development and application of fuel cell systems. Further information on the ongoing work of the Agreement can be found on the programme's web site <http://www.ieafuelcell.com>.

Introduction

Fuel cell based energy conversion systems offer potential energy, environmental and economic benefits compared to alternative existing systems. This is the reason that intensive development efforts are being made throughout the world in order to determine the viability of the technology. In order to accelerate developments and make best use of resources, intensive co-operation has been initiated on national and international levels.

Fuel cell technology is a competitor to several technologies used today, with much better performance in terms of efficiency and emissions. However, the technology is not yet mature and needs to be further developed. Fuel cells are already today introduced in small on-site applications as well as in the transport sector. The applications for fuel cells are broad and range from small scale portable applications through transportation and various combined heat and power (CHP) applications to medium and large scale power production, where the best of technologies can be utilised together, i.e. a fuel cell in combination with a gas turbine.

The Implementing Agreement (IA) for a programme of research, development and demonstration on advanced fuel cells was signed by seven countries in Paris on April 2nd 1990. Since then a further eight countries have signed the Implementing Agreement and the programme has grown accordingly. The participants come from Governments, R&D institutions, academia and industry in the following countries: Australia, Canada, France, Germany, Italy, Japan, Korea, Netherlands, New Zealand, Norway, Sweden, Switzerland, UK and USA. During 1998, over 70 experts participated in the work, of whom roughly half came from different market parties (utilities, vehicle manufacturers, fuel suppliers etc).

The aim of the IEA Advanced Fuel Cells programme is to advance the state of understanding of all Contracting Parties in the field of advanced fuel cells. It achieves this through a co-ordinated programme of research, technology development and system analysis on Molten Carbonate (MCFC), Solid Oxide (SOFC) and Polymer Electrolyte (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. Each Task is described in an Annex to the IA.

The major objective of the Implementing Agreement is strengthening the co-operation on the development and implementation of fuel cell systems. This is accomplished through the international network by activities like co-operative research, the exchange of experience and data, and by market and systems studies.

The first term of the Implementing Agreement originally ran from 1990 until December 1995 and was then extended until the end of 1998. During the latter period the following Tasks (Annexes) were in force.

Annex	Name
VI	MCFC under real Operation Conditions
VII	SOFC under real Operation Conditions
VIII	Collaborative Research on PEFC
IX	Fuel Cell Systems for Stationary Applications
X	Fuel Cell Systems for Transportation

These Annexes have now all been completed and their results are summarised in this report.

Achievements

The IEA Advanced Fuel Cells Implementing Agreement has contributed significantly to technology development in participating countries through the provision of expert networks. These networks have allowed expert groups to:

- Share R, D&D results,
- Define measurement and monitoring techniques,
- Exchange information on cell, stack and system performance,
- Collaborate on the development of new procedures and models,
- Share information on application requirements.

Information exchange in expert groups has led to the achievement of significant technical objectives. These include the development of standard test procedures for MCFC material, cells and stacks, the identification of degradation mechanisms for MCFC, SOFC and PEFC stacks under real operating conditions and the initial assessment of fuel cell systems against user requirements for stationary and transport applications.

Specific examples from the work during 1996-98 include the compilation of an inventory of MCFC stack and systems testing procedures (Annex VI), the exchange of new information on in-situ monitoring of SOFC performance (Annex VII) and the collaborative development of an improved reformer model for the steam reforming of methanol (Annex VIII). The work on system issues in relation to stationary applications (Annex IX) has significantly increased the knowledge of how to implement stationary fuel cells into the energy infrastructure. Though it is difficult to compare the importance of the achievements yet, Annex X's extensive network of experts from academia, research institutes and the car industry is seen as a particularly important achievement.

Through the task-sharing mechanism and by sharing resources, participants maximise the cost effectiveness of their work. In this way the Agreement has contributed to national fuel cell programmes and, indirectly, to the further collaboration through bilateral projects as well as EU-sponsored research. The IEA Advanced Fuel Cells Programme is regarded highly by the international fuel cells research community and is increasingly recognised by potential users, as evidenced by, for example, the participation of car manufacturers in Annex X. There are no comparable international collaboration organisations in the field.

Annex VI MCFC under Real Operation Conditions

Duration

Original period: November 1, 1996 to December 31, 1998
Extended Period: November 1, 1996 to December 31, 1999

Operating Agent

The Netherlands Agency for Energy and the Environment (NOVEM) acting through the Netherlands Energy Research Foundation (ECN) (until April 1997)

The New Energy and Industrial Development Organization (NEDO) (after April 1997)

Participants

Original Participants:

Germany	Forschungszentrum Jülich GmbH (KFA) through Motoren und Turbinen Union Friedrichshafen GmbH (MTU)
Italy	Ente Nazionale per le Nuove Tecnologie l'Energia e l'Ambiente (ENEA)
Japan	New Energy and Industrial Technology Development Organization (NEDO)
The Netherlands	Netherlands Agency for Energy and Environment (NOVEM) through Netherlands Energy Research Foundation (ECN)

New Participants since September, 1997:

Korea	Korea Institute of Science and Technology (KIST) through Korea Electric Power Research Institute (KEPRI)
The United States of America	Department of Energy (DOE) through M-C Power.

Objective

The objective is to provide for international collaboration in the research and development of certain aspects of MCFC technology, in order to improve its economics and effectiveness for power generation. These aspects include:

- (a) Improvement of performance and endurance, and reduction of costs, for stacks and BOP;
- (b) Identify present and envisaged manufacturers capabilities in this respect;
- (c) Development and standardization of effective test-procedures for materials, cells and stacks.

Task Description

Participants achieved the foregoing objectives by undertaking co-operative studies resulting in reports, according to the following Subtasks.

Subtask A: Stack Performance and Endurance

Subtask-leader: ECN (The Netherlands)

This subtask started with an inventory of stack performance decay contributions, comprising estimated decay-rates for the separate contributions. This led on to an overall analysis of MCFC stack performance decay. Subsequently, groups of contributions linked by their decay-mechanism were analyzed in more detail and methods for improvement were identified. Finally, decay-rates were estimated which should be accessible in future R&D.

Subtask B: Test Procedures and Standardization

Subtask-leader: MTU (Germany)

This subtask started with an inventory of all methods and procedures presently used by the participants for testing MCFC materials, cells, and stacks. This led to a catalogue of briefly described MCFC test methods. Subsequently, groups of test methods related to the specific items covered in Subtask A were evaluated in more detail, regarding cost-effectiveness and the meaning of the data obtained. More detailed descriptions as well as recommendations for modification were added to the catalogue. Finally, recommendations for standard test procedures were developed and included in the catalogue.

Subtask C: Development of BOP Technology

Subtask-leader: NEDO (Japan).

In this Subtask reviews were made on the reliability and cost of state-of-the-art BOP components. The reviews were made for each BOP component, and comprised of specifications, operational data, pending problems, targets, and possibilities for improvements. The three main MCFC system families were covered, i.e. External Reforming, Internal Reforming and Sensible-Heat type of systems. Stack-operation conditions were defined for each family.

Progress Summary

Background

The attractions of the Molten Carbonate Fuel Cell (MCFC) as a power source have been understood for quite some time. However, it was also realised that a number of problems, mainly related to endurance and cost, had to be overcome or overridden before commercialisation of MCFC technology can come within sight. By the end of 1991, initiatives were taken for collaborative work in this respect, within the IEA Programme on Advanced Fuel Cells. After establishing interest during a workshop in

June 1992, the Annex III "MCFC Materials and Electrochemistry" started in May 1993. The Annex remained active to the end of 1995, dealing with the endurance problems connected to corrosion of the bi-polar plate, dissolution of the cathode, and the electrolyte inventory of MCFC stacks. Apart from extensive data-exchange and fruitful expert discussions, the main result of the Annex was consensus on the relative importance of the endurance limitations mentioned. In addition, life-time estimations were made, related to the eventual malfunctioning of cells and stacks caused by the phenomena studied.

At the finalisation of Annex III it was recognised that for further progress in endurance improvement and cost reduction better quantitative studies would be necessary. Such studies should, in addition to estimates for endurance limitations by malfunctioning, analyse the rate of gradual degradation of stack performance and assess its contributions. Subsequently, ways to reduce the various degradation contributions should be identified.

In the course of the work performed in Annex III, frequently data was encountered without proper description of the methods or procedures used, or obtained with methods not allowing easy comparison. The demand was felt for the development and standardisation of effective test-procedures for MCFC materials, cells and stacks.

Another Annex related to MCFC, phase 1 of Annex I "MCFC BOP Analysis" started in April 1990, followed by phase 2 which was active between January 1, 1994. and December 31, 1996. The objectives of this Annex were to systemize and optimize technical factors, design conditions, operating conditions and background of BOP as well as large scale stacks, and identify R&D items and study the best strategy to resolve them for the purpose of developing the most cost-effective MCFC systems. Many of the data were collected and conclusions have been recorded in reports available for contributing participants. It became clear that further work would be necessary to reveal possibilities for Balance-Of-Plant (BOP) technology with improved reliability and reduced cost. Also, the study of BOP provides for an interface between system-user requirements and stack operational windows, and the resulting consequences for performance and endurance.

In this manner, Annex VI "MCFC under Real Operating Conditions" concentrated on the manufacturer's capabilities to improve MCFC technology.

Activities

Following the kick-off meeting, Annex VI activities started with four participants; Germany (MTU), Italy (ENEA), Japan (NEDO) and the Netherlands (ECN).

From the 14th ExCo Meeting in April 1997 onward, Japan (NEDO) took over Operating Agent from the Netherlands(ECN).

Korea and the U.S. expressed their intention to join the Annex VI activity. Following the decision at the 14th ExCo, enquiries on Annex VI subtask C were sent to Korea by ExCo Secretary to see if they could make contributions to the Annex. Enquiries comprised condensed questionnaires on Annex I, III and inventories on Annex VI subtask A, B. The

answers from both countries were received by the Operating Agent and circulated to ExCo Secretary and the Annex participants. Their participation to the Annex VI was approved by all participants at the beginning of the second Annex meeting in September, and then approved at 15th ExCo in September 1997.

To accomplish the Annex activity, one year extension of the activity period was proposed and approved at the 17th ExCo in March 1998.

The meetings held were as follows.

- Kick-Off Meeting was held in Roskilde, Denmark in September 16, 1996, hosted by NEDO.
- The first Meeting was held in Genoa, Italy in February 21, 1997, hosted by Ansaldo.
- The second Meeting was held in Petten, the Netherlands in September 17, 1997, hosted by ECN.
- The third Meeting was held in Munich, Germany in May 28-29, 1998, hosted by MTU.
- The fourth Meeting was held in San Diego, the U.S.A. in November 23-24, 1998, hosted by M-C Power.
- The fifth Meeting was held in Petten, the Netherlands, April 15 - 16, 1999, hosted by ECN.

Technical Accomplishments

Subtask A:

Stacks play a major role in determining the performance and cost of MCFC systems, and it is essential to obtain a low decay rate and long life time for the stack, to provide an effective system. This study was carried out in two stages.

Firstly, stack performances at participating fuel cell developers were surveyed in terms of their operating history and performance (i.e. decay rate). Then each factor which contributes to the decay was analysed.

In the second stage, detailed information was exchanged with regard to the improvement of each item which determines stack performance i.e. cathode dissolution, electrolyte and cathode materials, bipolar plate corrosion, electrodes, matrix, etc. Latest R&D data from each country's developers were provided and discussed.

To conclude the activity, prospects of stack lifetime up to 40,000 hours were shown by most of the participants as the outcome of the various development factors. Yet, more confirmation will be needed by carrying out long time operation tests.

Subtask B:

In test methods and procedures for MCFC elements, we have large differences between the 6 participants. We could not compare test results because of unreliable characterisation of the test data, according to the individual development of evaluation test methods.

Therefore we planned to establish the standardisation of test and characterisation methods in order to have common understanding. For this project, we prepared pre-questionnaires and held meetings about important test items and made comparison of test procedures and

detailed test methods. Consequently, we achieved standardisation of detailed test methods and got reliable R&D data from each other. This can save unnecessary test time and labour, and contribute to focusing the R&D activities.

We made pre-questionnaires and examined 50 characteristic features of 3 MCFC elements (cathodes, electrolytes and bipolar plates) for each of 3 stages (materials, cells and used stacks). Among them, we selected 37 essential items, consisting of 17 on cathodes, 4 on electrolytes and 16 on bipolar plates (for example, pore size distribution of cathode, carbonate ratio of electrolyte, corrosion layer thickness of bipolar plates and so on). For each characteristic feature, detailed test procedures were surveyed and compared among the participants. Furthermore, the same procedure was conducted for the other 2 MCFC elements (anodes, matrix), and 14 essential items for anodes and 12 for matrices were selected and surveyed.

Subtask C:

There are three types of MCFC systems under development: External Reforming, Internal Reforming, and Sensible Heat Reforming. In Subtask C, major specifications of BOP (balance of plant) were surveyed as to the systems which are under development in the participating countries. Each participant made a presentation and exchanged data on the overall plant system and major BOP (reformer, heat recovery steam generator, high temperature blower, and control system).

Furthermore, stack and system operation test methods and test experiences were reported. These data provide important information to other participants, although the specifications of each development program are different in system type, plant size, working pressure, etc.

The first stage of market entry period of MCFC is expected to be around 2000 - 2001, although it is still too early to determine the likely cost of a stack and components of BOP.

Further Work

This work will be continued under Annex XIV.

Appendix A History of Annex VI

	Kick-off Meeting	1st Meeting	2nd Meeting	3rd Meeting	4th Meeting	5th meeting
Date	Sep. 16, '96	Feb. 21, '97	Sep. 17, '97	May 28-29, '98	Nov. 23-24, '98	Apr. 15-16, '99
Place	Roskilde	Genoa	Petten	Munich	San Diego	Petten
Host Country	NEDO	Ansaldo	ECN	MTU	M-C Power	ECN
Number of Participants	8	11	12	18	13	13
Germany	1	1	1	6	1	1
Italy	1	4	2	2	2	2
Japan	4	4	3	4	4	4
Korea	-	-	2	2	2	1
the Netherlands	2	2	4	2	2	5
U.S.A.	-	-	0	2	2	0

Annex VII SOFC under Real Operation Conditions

Duration

January 1996 - December 1998

Operating Agent

Heinz Nabielek, Forschungszentrum Jülich, Germany

Participants

Country	Signatory Party
Australia	Ceramic Fuel Cells Limited (CFCL)
Denmark	Danish Energy Agency
France	L'Agence de l'Environnement et de la Maîtrise de l'Energie (ADEME)
Germany	Forschungszentrum Jülich
Japan	New Energy and Industrial Technology Development Organisation (NEDO)
Netherlands	Netherlands Agency for Energy and the Environment (NOVEM)
New Zealand	University of Waikato
Norway	Research Council for Norway
Switzerland	Office Fédérale de l'Energie (OFEN)
United Kingdom	Department of Trade and Industry
United States	Department of Energy

Experts

Each organisation is categorised as government or government agency (G), research institution (R), industry (I) or academic (A).

Dr Sukhvinder Badwal	CFCL (I)	Australia
Dr Karl Föger	CFCL (I)	Australia
Prof Finn-Willy Poulsen	Risø (R)	Denmark
Dr Alain Comte	EdF (I)	France
Dr Etienne Gehain	GdF (I)	France
Dr Philippe Stevens	EdF (I)	France
Dr Bert de Haart	FZJ (R)	Germany
Dr Aristides Naoumidis	FZJ (R)	Germany
Dr Ernst Riensche	FZJ (R)	Germany
Dr Frank Tietz	FZJ (R)	Germany
Prof Masayuki Dokiya	Univ Yokohama (A)	Japan
Dr Hirokazu Sasaki	NEDO (G)	Japan
Prof Harumi Yokokawa	NIMC (R)	Japan

Dr Joep Huijsmans	ECN (R)	Netherlands
Dr Reinder Boersma	Univ Waikato (A)	New Zealand
Prof Nigel Sammes	Univ Waikato (A)	New Zealand
Prof Kemal Nisancioglu	NTNU (A)	Norway
Dr Augustin McEvoy	EPFL (R)	Switzerland
Dr Steve Barnett	BNFL (I)	UK
Dr Andrew Dicks	British Gas (I)	UK
Dr Kevin Kendall	Keele University (A)	UK
Prof Rowland Travis	Imperial College (A)	UK
Dr Michael Krumpelt	ANL (R)	USA

Objective

The main objective of Annex VII was to assist, through international co-operation, the development of natural gas-fuelled Solid Oxide Fuel Cells (SOFC) technologies through:

- the exchange of information to tackle complex problems in stack and systems design and operation,
- the consideration of end-user requirements for stack operation with the goal to optimise lifetime of cells and stacks,
- a better understanding of interfacial and bulk charge and mass transport mechanisms to optimise cell components under actual SOFC operating conditions,
- the study of alternative materials in case traditional concepts are too expensive or are too short-lived.

Task Description

The Participants have shared the co-ordinated work necessary to carry out this Task in Annex VII. The work was divided into the following four Subtasks:

Subtask 1: “Balance-of-Plant Technology”

Activity 1.1 “Balance-of-Plant Optimisation by Modelling”

Tackle complex problems in stack and systems modelling by exchange of information. Workshops September 1996 in Paris and October 1998 in Arnhem.

Activity 1.2 “SOFC Activities World-Wide”

Establish an information directory to encourage information exchange on critical aspects of SOFC development (first directory 1996, second directory 1998).

Subtask 2: “Stack and Cell Operating Conditions”

Activity 2.1 “Stack Operating Experience and Evaluation”

Enable comparison of stack performances on a common basis. Activity meeting November 1996, annex meeting June 1998, final report at the 11th SOFC workshop October 1998 in Arnhem, NL.

Activity 2.2 “Lifetime of Cells”

Evaluate effects of operating conditions typical for end-user applications with the goal to improve and demonstrate SOFC lifetime. Questionnaire 1998.

Subtask 3: “Materials under Operating Conditions”

Develop a better understanding of interfacial/ bulk charge and mass transport mechanisms to optimise cells under actual SOFC operating conditions. The emphasis was on novel designs and performance of components based on relatively well known materials. Successful series of workshops established:

Lausanne	August 1992	4 th IEA SOFC
Wadahl	January 1995	7 th IEA SOFC
Les Diablerets	January 1997	10 th IEA SOFC
Wadahl	January 1999	12 th IEA SOFC

Subtask 4: “Materials Innovation”

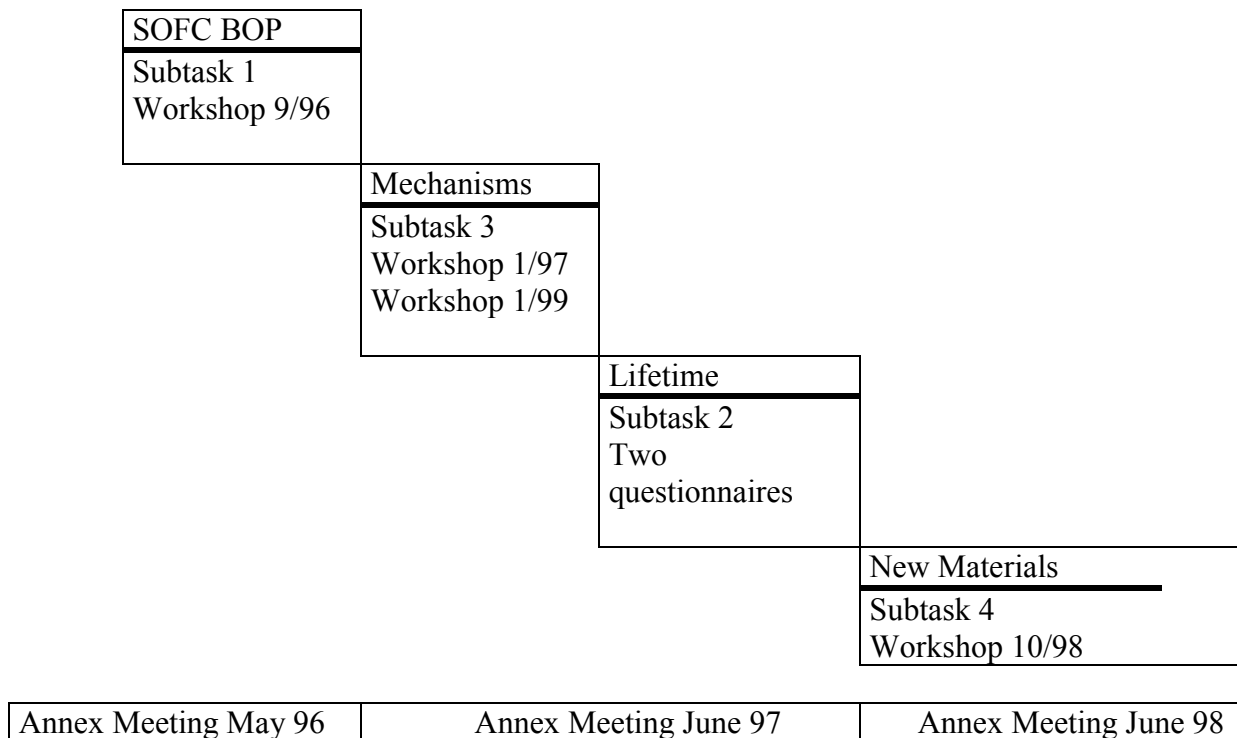
Develop and assess new SOFC materials for long-term stability and reduced manufacturing costs. Investigate materials in low-temperature operation <800°C: task meeting June 1997, Workshop October 1998.

Background

This task followed on from Annex II: Modelling and Evaluation of Advanced Solid Oxide Fuel Cells, performed during 1990 through to 1995.

Activities

The co-operation in Annex VII progressed along a carefully arranged series of Workshops and Annex Meetings, as shown below.



Technical Accomplishments

The main accomplishments in Annex VII were as follows.

Subtask 1: Balance of Plant (BOP) Technology

Activity 1.1: BOP Optimisation by Modelling (Philippe Stevens & Alain Comte, EdF)

Discussion summary from the 9th IEA SOFC Workshop, Paris, September 1996: Cooperation with Annex VI MCFC and Annex IX Stationary Applications proved to be useful. Many of the discussions were centred around the Forschungszentrum Jülich 200 kW_e reference design. For large-scale electricity production, it was thought that small SOFC plants of ≤ 200 kW_e will never be economical. The components of a SOFC plant can only be manufactured at a reasonable cost for plant sizes > 1 MW_e. A minimum stack lifetime of 40,000 hours is a must.

Some felt that a working temperature of 1000°C is preferable to the lower temperatures for optimum BOP conditions. Furthermore, these temperatures no longer present any technical or economical barriers for the BOP.

Activity 1.2: SOFC Activities World-Wide (Hirokazu Sasaki, NEDO- now at Osaka Gas)

An information directory was developed to encourage information exchange on critical aspects of SOFC development. The directory contains programme organisation/objectives/budget/schedule/contact person/cell or stack materials, dimensions, fabrication processes/cell or stack performance data/information on system studies or other work. The original directory was compiled in 1996 and distributed early 1997. An extensive update was performed in 1998 and the directory was distributed to the Activity 1.2 participants.

SOFC stacks can be classified as follows:

Tubular		Planar					
1 cell/tube	Several cells per tube	1 cell per layer				Several cells per layer	
		Metallic interconnect		Ceramic interconnect		Metallic intercon.	Ceramic intercon.
		<u>Thick electrolyte</u>	<u>Thin El.</u>	<u>Thick El.</u>	<u>Thin El.</u>	<u>Thick El.</u>	
Siemens-Westinghouse	MHI Nagasaki	Sulzer ECN TMI Ztek	Research Centre Juelich	SOFCo Dornier (term.) Tokyo Gas MHI MOLB Risø	--	Sanyo (term.) Siemens (term.)	Rolls-Royce
Toto			Allied Signal			CFCL	

For tubular cells, the market leader is Siemens-Westinghouse in Pittsburgh. The 100-kWe plant in Arnhem is the latest in a series of test sites since 1986. Arnhem was

operating in 1998 with natural gas for 3700 hours. After shutdown June 1998 and subsequent repair, the plant has been operational again since March 1999.

Toto in Japan is also developing SOFC tubes of similar design.

MHI, in co-operation with the Electric Power Development Co., works on stacking smaller tubes to achieve 0.5 to 0.7m length. They were tested in a 21 kW-pressurised system in March 1988.

To achieve significant cell areas in the planar SOFC design, some companies are working with an array of cells in one layer. Siemens in Erlangen used a 16-cell array to demonstrate an 80-layer stack producing 10.7 kW. The 50 kW project was terminated, when Siemens took over Westinghouse and it was decided to concentrate on the commercialisation of the tubular SOFC system.

CFCL in Australia uses a rectangular array concept: a 5.5 kW stack was demonstrated with a 2x4 matrix in 50 layers. The plan is to operate 25 kW this year and prepare for the 100 kW system in 2001.

The “classical” planar SOFC concept has one round or square cell per stack layer. Ceramic interconnects are used in the 900–1000°C temperature range. At lower operating temperatures, metallic interconnects can be used.

Tokyo Gas uses a thin $\text{Sc}_2\text{O}_3\text{-ZrO}_2$ film on the 3YSZ electrolyte for better conductivity. Plans are the demonstration of 3 kW operation with natural gas. MHI, in co-operation with Chubu Electric Power, continues the MOLB-design with a corrugated cell structure. 5 kW have been demonstrated and 25 kW in a 3-stack system are in preparation.

SOFCo, in an all-ceramic design, has demonstrated degradation rates below 0.5%/1000h, measured on several stacks over a period of 10,000h.

Risø has developed methods to reduce the overpotential at the electrodes during cell operation. This will be required for the use of cost-effective stacks in future SOFC systems.

Metallic interconnects are used by Sulzer, ECN, Allied Signal and the Research Centre Jülich. Sulzer puts round cells in their special HEXIS design that largely avoids the difficult sealing problems of planar SOFC systems. For Sulzer and their projected customers, reliable operation together with flexibility in the use of fuels and low investment is more important than high efficiency and high power densities. Small 1.5 kW units have been successfully operated in field tests in Germany, Switzerland and Japan.

ECN in Petten has produced cells for Siemens in Erlangen and Sulzer in Winterthur. This way, they have shown experience in reliable cell manufacture and have demonstrated good results in terms of power and long-term operation.

Traditional planar SOFC design called for 100–200 μm electrolyte thickness for reasons of mechanical strength and safe separation between fuel and air. However, it

is possible to transfer the support function to an electrode to enable much thinner (3–20 µm) electrolytes thereby reducing ohmic resistance. This way, 700-800°C operation is possible without using exotic electrolyte materials.

With their very thin electrolyte, Allied Signal has demonstrated a world-record current density of 2 A/cm² at 800°C.

Subtask 2: Stack and Cell Operating Conditions (Joep Huijsmans, ECN Petten)

Two activities were carried out, Activity 2.1 “Stack operating experience and evaluation” and Activity 2.2 “Lifetime of cells”. For both activities, questionnaires were prepared and circulated within the international IEA community. The results of the questionnaires were presented at the 11th IEA SOFC workshop in Arnhem, the Netherlands.

Activity 2.1: Stack operating experience and evaluation (Andrew Dicks, British Gas)

A stack-operating questionnaire was distributed as a first step in contacting stack developers to encourage their participation. Distribution of proceedings from activity meeting in November 1996 at which the participants discussed protocols and testing regimes for SOFC stack testing. Reports to the June 1998 Annex meeting in Nantes and the October 11th IEA SOFC Workshop in Arnhem.

Activity 2.2: Lifetime of cells (Joep Huijsmans, ECN Petten)

In a first approach it was tried to organise a workshop in Petten in the autumn of 1997. The title of the workshop would have been “Stack and Cell operating conditions”. However, due to limited interest from the participants of subtask 2 it was decided, after consultation of the Subtask leader with the Operating Agent, to cancel the workshop.

A questionnaire was prepared by ECN in which the participants of subtask 2 were asked to generate application-specific operating conditions and degradation data for cells, stacks and systems. On the basis of these data it was hoped to carry out an evaluation in order to identify deleterious effects of operating conditions on cell lifetime. Responses were obtained from NEDO (2 Japanese companies, i.e. TOTO and Tokyo Gas), Siemens in Erlangen, the Research Centre Jülich and ECN.

The results for the “single cell data” show that in principle state-of-the-art cell materials are being used. For the cell testing in general rather mild or ideal testing conditions are applied, i.e. the use of hydrogen and rather low fuel utilisation in general. The temperature range for cell testing is between 850 and 1000°C. A relatively low degradation is observed in most cases for the above mentioned testing conditions, i.e. below 1% of $V_{\text{cell}} / 1000$ hrs.

For the “SOFC stack data”, it can be concluded that both ceramic and metallic interconnect materials are used by the respondents. Stack test data for both hydrogen and methane (not natural gas) are available. With methane, internal reforming is

being demonstrated. Fuel utilisation varies between the different respondents between very low (6 %) to more realistic (70%). The temperature range of operation is between 800 and 1000°C. Different stack concepts are compared, i.e. tubular, electrolyte supported planar cells and anode supported planar cells. The longest testing times demonstrated for planar stacks do not exceed 7000 hrs. At acceptable current densities (0.3 A/cm²) the observed degradations are still relatively high compared to the required performance of < 0.5%/ 1000 hrs.

Due to the limited, though valuable, response of the IEA community to the questionnaire of subtask 2, it is difficult to draw firm conclusions from the data acquired. In general it can be stated that testing conditions for single cells are in general fairly ideal, whereas more realistic conditions are observed for stack tests. In general, the observed degradation rates are already low, but still too high for economic application of fuel cells.

Subtask 3: Materials under Operating Conditions (Kemal Nisancioglu, NTNU Trondheim)
and

Subtask 4: Materials Innovation (Aristides Naoumidis, Forschungszentrum Juelich)

These two subtasks focused on the theory and measurement of interfacial processes and recent advances in the fabrication and characterisation of materials applicable to modern SOFC technology. Ceramic gas separation membranes were included as a major spin-off subject. Particular topics included: Electrode Materials Science, Reaction Mechanisms, Electrolyte Processes, Interconnects, Oxygen Exchange and Transport, Gas Separation Membranes, Cell Components and Processes.

As far as materials science aspects are concerned, original contributions were given on electrode degradation under real operating conditions, including chromium poisoning from chromite interconnects and performance of interconnect materials under real operating conditions. Novel ceramic anode materials were presented, e.g. the performance of mixed conducting $Zr_{0.62}Y_{0.2}Ti_{0.18}O_{2-x}$. The current status of nonstoichiometric oxides considered for oxygen separation and membrane reactor technology was summarised. The exchanges contained one of the most comprehensive critical reviews of the electrode reaction mechanisms available to date. Discussion of this subject also emphasised questions related to methodology and modelling.

Serious problems associated with determining the significance of the probed potential in single cell measurements, first highlighted in the Les Diablerets meeting, were subjected to in-depth analyses and discussion. The popular issue regarding the significance of the electrochemical impedance technique in studying electrode processes, merits and pitfalls of using intuitive equivalent circuits as opposed to deterministic and statistical modelling was continued. Similarly, the measurement and interpretation of oxygen diffusion data in mixed conducting oxides was a popular subject, with noteworthy advances in methodology and understanding of mechanisms. A related area of progress was visualisation of oxygen concentration profiles at the submicron level by use of SIMS, in particular at grain boundaries and near three-phase contact regions at the cathode-electrolyte interface. This microstructural composite cathode investigation showed that oxygen exchange proceeds not only in

the three-phase boundary region, but also at LSM/ YSZ phase boundaries. Methodological studies of electrochemical measurements have resolved the classical problem of determining electrochemical parameters with reference cells.

Experimental work was started on the optimisation of cell components by the development of a better understanding of interfacial and bulk charge/mass transport under actual SOFC operating conditions. This work focused on micro- and nano-scale processes and three-phase boundary regions with emphasis on novel designs and performance of components based on relatively well known materials.

Results were obtained on the kinetics and understanding of the mechanisms of surface and bulk processes e.g. electrode processes, oxygen ion exchange and oxygen transport processes in mixed and in ionic conductors. Investigations of cells and stacks under realistic operation conditions and interpretation of degradation mechanisms like the effect of Cr evaporation from steel onto cathode performance (both LSM and LSFC) were carried out and showed significant deterioration rates using steel housings. Investigations of LSM compositions with emphasis on crystallographic phase changes depending on strontium content, oxygen partial pressure and temperature were performed.

In the case of cathode materials, analytical surface measurements with low energy ion spectroscopy surface properties like segregation, topography and corrosion effects on the first few atomic layers can be determined.

Electrochemistry

It has been pointed out that the electrochemical data for SOFC electrodes are in considerable disagreement. The reasons for this are not well understood, but the explanations can be related to both inappropriate experimental methods and real variations between the systems. It is well known that the electrocatalytic activity of electrode materials vary with time. There is also a general agreement that the electrode activity is influenced by long term polarisation. Such effects have been observed both for anode and cathode materials. The state of presumably identical electrodes may thus be different, giving rise to diverging kinetic characteristics. The main subjects for the discussions on cell components and processes were therefore: "Why is it so difficult to establish a set of consistent kinetic data for the SOFC electrode processes?" and "How should the measurements be carried out to obtain consistent data?"

Part of the change in electrode activity is clearly due to morphological variations in the range of micrometers and sub-micrometers, as observed for various anode and cathode materials. The importance of morphological changes on the nanometer range were also briefly discussed, in particular with respect to electron tunnelling processes and the effect on electrochemical reaction kinetics.

Other electrocatalytic effects are also important, e.g., changes in the work function of the active surfaces due to accumulation of impurities from the gas phase or the solid materials. Changes in surface defects ("active sites") and surface stoichiometry may also take place, possibly caused by polarisation or by alterations in the atmosphere composition.

The question, "What is really an active site?" was raised. Although a definition of the term was not found, a consensus was reached that solid surfaces are dynamic at the temperatures of interest, which can be as high as 1000°C. An active site, whatever its physical property, must therefore have a limited lifetime. All the above-mentioned effects were considered with respect to both anode and cathode materials. On the anode side, there is a certain solubility of NiO in YSZ, and depending on the preparation of the electrode, variations in the interactions between the Ni and YSZ can exist. Variations in the anode properties may also be due to different fabrication methods. Dissimilar interfacial and material properties of the cermet which result from this can affect the electrochemical properties.

For the cathode, it has been observed that small variations in the perovskite stoichiometry may have significant effects on the electrocatalytic properties of the material. Hence, electrochemical measurements on slightly varying material compositions are not necessarily comparable. The effect of mixed conduction in cathode materials was briefly discussed, and mentioned as a possible requirement for high electrode activity.

It was concluded that electrochemical measurements are insufficient for a full characterisation of the electrode processes in SOFCs. Even if the measurements are performed on a short time scale to avoid long term changes, one is left with the question of how representative the measurements are for an electrode operating in an SOFC stack for several thousand hours. In situ, real time observations by alternative methods, e.g., X-ray or synchrotron diffraction, were proposed as steps towards a better understanding of the electrochemical properties of SOFC electrodes.

Nevertheless, investigations of the electrode processes have to proceed, despite the complicated system properties and experimental difficulties, in particular because reliable electrochemical data are needed in order to establish operational parameters for SOFC stacks.

Three-electrode impedance measurements

The problems in three-electrode impedance measurements arise in the case of cells with a thin electrolyte. This takes place because of the absence of geometrical and electrochemical symmetry in the cell resulting from placement of the electrodes during manufacturing and differences in the polarisation resistance of the cathode/electrolyte and anode/electrolyte interfaces. Small distortions of symmetry in the placement of electrodes could lead to large errors in the measured impedance, such as distortion of the arcs in Nyquist diagrams and changes in the absolute values of the polarisation resistances.

Errors due to geometrical mismatch can be corrected by fitting the data to theoretical models of the cell impedance. In this case, the exact geometry of the electrode/electrolyte edge should be taken into consideration, especially if the electrode has a limited conductivity. However, the optimal position of electrodes with respect to one another is difficult to determine in cases where the electrode edges are tapered. Such edge geometry is often observed on the screen-printed electrodes.

Because of these complexities with the three-electrode approach, such measurements should be carried out on well defined geometries, amenable to correction by use of

theoretical models, or two-electrode measurements should be performed on symmetric cells without the use of a reference electrode.

Another possibility is to measure the impedance of a symmetrical cell by the two-electrode technique under conditions where the reaction kinetics on the counter electrode are very fast relative to the working electrode, or it appear in a different frequency region.

Modelling of impedance

The cell or electrode impedance is usually modelled by using a deterministic or a statistical approach. The deterministic approach is based on the solution of a mathematical model describing charge transport in the bulk and interfaces of cell components for the actual cell geometry. If it is possible to obtain an analytic solution, which may require simplifying assumptions, the derived impedance can be expressed in the form of an equivalent circuit. Possible problems are that the construction of a unique equivalent circuit may not be possible, and certain components of the circuit may not have a physical significance. The choice of best circuit would be based on intuition and how the experimental data fit the model.

In the statistical approach, an equivalent circuit is constructed by statistical fitting of data to a random or intuitive combination of RCL elements defining an equivalent circuit. The application of the equivalent circuit to the analysis of the impedance spectrum leads often to a good agreement between measured and simulated data set but the parameters extracted from the circuit as a rule cannot be physically interpreted. However, the approach may provide an easy way of obtaining an idea about the number of time constants (rate determining electrochemical processes) which determine the cell response.

Interpretation of impedance measurements may be further complicated by geometrical factors. The number of the arcs in a Nyquist plot do not always correspond to the number of rate limiting processes in the reaction kinetics. The additional arcs could be caused by distortions caused in the electric field by special geometrical effects, such as discrete contact points between the electrode and electrolyte.

Measurement of diffusion and surface exchange coefficients

Diffusion data for oxygen conductors obtained by various experimental techniques can be analysed, by using the solutions to Fick's second law, to obtain the diffusion and surface exchange coefficients. Depending on the technique used, a "self diffusion coefficient" or a "chemical diffusion coefficient" is measured.

The self diffusion coefficient is measured by use of a tracer technique and determination of the tracer oxygen profile, e.g., by the SIMS technique. Since a tracer is used, the self diffusion coefficient describes diffusion of oxygen in the absence of a concentration gradient.

The chemical diffusion coefficient describes diffusion of species in the presence of a true concentration gradient of oxygen in the material. The chemical diffusion coefficient is measured by using relaxation techniques such as conductivity relaxation or mass change (TG) in response to a sudden change of oxygen partial pressure in ambient atmosphere or by using the Belzner cell (the potential step technique).

The two parameters are related to one another according to the relationship

$$\frac{D}{a_i} = a_i \frac{\partial \tilde{a}_i}{\partial \tilde{A}_i} \quad (1)$$

where the activity term is the “enhancement factor” originally introduced by Wagner. The enhancement factor can be obtained from thermodynamic titration measurements relating the nonstoichiometry of the material to the equilibrium oxygen partial pressure.

The self diffusion coefficient obtained from chemical diffusion coefficients by use of equation 1 does not always give a good agreement with the self diffusion coefficients obtained from SIMS measurements. A possible reason may be the use of large initial steps in the oxygen partial pressure to start the diffusion process. If short-time data are used in data analysis, a large concentration gradient close to the surface may influence the measured diffusion coefficient, since this coefficient is a function of concentration. However, large time relaxation data should not be affected by this since the concentration gradients are relaxed.

The surface exchange coefficient is often determined simultaneously with the self diffusion or chemical diffusion coefficient. The difference in the surface exchange coefficients obtained from the different methods can vary by orders of magnitude. No simple relations exist between the surface exchange coefficients measured using different methods. A good overview on this topic was published recently.

The SIMS technique is considered as the most reliable method available for the measurement of diffusion and surface exchange coefficient of oxygen. However, the practical application of mixed conductors as cathode materials in SOFC is associated with concentration gradients in the material created by an overpotential at the mixed conductor/electrolyte boundary. In this connection, the following questions need to be answered:

- Are the transport data measured by SIMS applicable in the real situation?
- What takes place at the surface of the mixed conductor in the presence of a concentration gradient of the oxygen in the material?
- Is the concentration gradient of oxygen in the mixed conductor induced by the over-potential equivalent to the concentration gradient induced by change of the oxygen partial pressure in the environment?

We hope to get more insight in these topics in the future.

Annex VII Workshops and Task Meetings

- BOP Planning, Feb 1996, NEDO, Tokyo, Japan
- Start-up Annex Meeting, May 1996, ECN, Petten, The Netherlands
- 9th IEA SOFC Workshop BOP “SOFC Balance-of-Plant Modelling”, Sep 1996, EdF, Paris, together with Annex VI MCFC and Annex IX Stationary Applications
- Task Stack Testing, Nov 1996, Orlando, USA
- 10th IEA SOFC Workshop “Materials and Processes”, Jan 1997, Les Diablerets, Switzerland
- Mid Annex Meeting, June 1997, Jülich, Germany
- Annex Meeting, June 1998, Nantes, France

- 11th IEA SOFC Workshop, October 1998, Arnhem, The Netherlands
- 12th IEA SOFC Workshop, January 1999, Wadahl, Norway

Annex VII Reports

J P P Huijsmans, Proceedings Spring 1996 IEA SOFC Workshop
ECN Petten, 12-14 May 1996, Level 2

J Drennan et al. "Characterisation, conductivity and mechanical properties of the oxygen-ion conductor $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-x}$ " CFCL Australia, July 1996, Level 1

H Nabielek, The Operating Agent's Annex II SOFC Final Report, Forschungszentrum Jülich, August 1996, Level 3a

P Stevens "SOFC Balance-of-Plant Modelling: with participation of MCFC and Stationary Applications", EdF Paris, 19-20 September 1996, Level 2

H Nabielek "Annex VII SOFC under Real Operating Conditions, Implementing Agreement for a Programme of Research, Development and Demonstration on Advanced Fuel Cells" Research Centre Jülich, December 1996, Level 3

SPS Badwal et al "Interaction between chromia forming alloy interconnects and air electrode of SOFC", CFCL Australia, December 1996, Level 1

F W Poulsen "Danish Contributions to IEA Annex VII: SOFC under Real Operating Conditions", 28 January 1997, Level 1

A J McEvoy, K Nisancioglu "Materials and Processes", Proc 10th Workshop Les Diablerets, 28-31 January 1997, Level 2

H Sasaki "SOFC Activities Worldwide", 1996 Report of Activity 1.2 March 1997, NEDO Japan, Level 1a

N Sammes "Mechanical Properties of Ceria Gadolinia for Application as an Oxygen Ion Conductor in SOFC", Waikato, April 1997, Level 3

H Nabielek "SOFC Research in International Co-operation: The IEA SOFC Annex", Proc Vol 97-40 of the Fifth International Symposium on Solid Oxide Fuel Cells, ed. W. Lehnert, The Electrochemical Society, 1997, Level 3

H Nabielek, A Naoumidis, F Tietz "Mid-Annex-Meeting", Proc. Subtask 4 Meeting, Research Centre Jülich, 6 June 1997, Level 2

H. Nabielek, Annex Meeting Report Nantes June 1998, Level 2

H. Nabielek, Proceedings 11th IEA SOFC Workshop, Arnhem October 1998, Level 2

H Sasaki "SOFC Activities Worldwide", 1998 Report of Activity 1.2 December 1998, NEDO Japan, Level 1a

J P P Huijsmans, F H van Heuveln, J P de Jong "SOFC Stack and Cell Operating Conditions", Final Report Annex VII Subtask 2, January 1999, ECN Petten, Level 2

K. Nisancioglu, A J McEvoy "Proceedings 12th IEA SOFC Workshop" January 1999, Wadahl, Level 2

H Nabielek, The Operating Agent's Annex VII Final Report Forschungszentrum Jülich, April 1999, Level 3a

Future Plans

The work of Annex VII will be continued under the new Annex XIII Solid Oxide Fuel Cells. This new activity will comprise annual workshops on aspects of SOFC science and technology, starting with a workshop on "Cost-effective and upscaled manufacturing technology for SOFC ceramics" in July 2000.

Annex VIII Collaborative Research on PEFC

Summary

Agencies from eight countries participated in this Annex to conduct collaborative research on polymer electrolyte fuel cells (PEFCs). The activities of the Annex were directed towards improvements in performance along with reduction in cost of these fuel cells and systems. Over the three-year duration of the Annex, several significant technical advances were achieved, some by different countries collaborating in their research. For example, Simon Fraser University in Canada conducted basic research in oxygen transport in polymer membranes from commercial sources as well as new membranes from Cranfield University in the U. K. Another collaborative research activity between the Royal Military College of Canada and Wellman CJB of the U.K. resulted in the development of a validated dynamic model of a methanol steam reformer. Advances have been made in understanding and improving the CO-tolerance of the PEFC, long-term testing of cells, manufacturability of membrane-electrode assemblies, catalyst loading and dispersion, evaporative cooling in PEFC stacks, effects of cation poisoning of the membrane, metal bipolar plates, alternative polymer electrolyte membranes, effects of ammonia and sulfur dioxide in the cathode air, methanol steam reforming, system modelling, and direct methanol fuel cells and stacks. A majority of the participants agreed that these research activities should be continued under the auspices of the IEA, and made a formal request for a new Annex. The Executive Committee has approved a new Annex XI for three years to continue the work of Annex VIII.

Introduction

This Annex entered into force on January 1, 1996, and remained in force for a period of three years, until December 31, 1998. Argonne National Laboratory, a contractor for the United States Department of Energy, acted as the Operating Agent for the Annex. Agencies from eight countries participated in this Annex: Canada, Germany, Italy, Japan, the Netherlands, Switzerland, United Kingdom, and the United States.

Objectives

The objectives of the Annex were to contribute to the identification and development of techniques to reduce the cost and improve the performance of polymer electrolyte fuel cells (PEFCs) as well as PEFC systems. These included (1) development of improved electrode catalysts and structures with reduced catalyst loadings; (2) development of improved, lower cost polymer electrolyte materials; (3) investigation of the effects of contaminants in the ambient air, (4) development of catalysts and systems for fuel reforming; (5) design, simulation, and analysis of system configurations; and (6) development of improved direct methanol fuel cells and systems.

Description of the Work

The work carried out by the Annex was divided into five subtasks:

Subtask 1. Improved Single Cells

Aims of this subtask were to improve the understanding of the PEFC electrode catalysts and structures, and the factors affecting their performance, with the objectives of reducing catalyst loading and cost, reducing performance degradation due to poisoning by CO and other contaminants in the fuel gas, reducing ohmic losses within the cells, and improving performance on low pressure reactants. The specific effort included improvements in catalyst distribution and utilisation, gas access to catalyst sites, and cell testing and characterisation.

Subtask 2. Improved Stack Materials and Concepts

Under this subtask, work was carried out to define and develop new PEFC electrolyte membranes and other stack materials and designs to reduce manufacturing cost, simplify assembly, and improve performance. The effort included investigation of new materials for bipolar plates and coatings for these plates, new flow field designs, and improved seals and stacking concepts.

Subtask 3. Operating Parameters: Effects on Performance and Lifetime

This subtask investigated the effects of operating conditions typical for end-user applications on the performance and lifetime of the PEFC. The effort included developing an inventory of application-specific operating conditions, identifying potentially damaging substances and operating conditions, and developing standardised single cell and stack test procedures.

Subtask 4. Balance of Plant Components and Systems

This subtask addressed the balance-of-plant in PEFC systems. The work was divided into fuel processor development, and component and systems modelling. The fuel processor work developed new catalysts and supports, dynamic models, and compact reformers. The modelling activity had the objective of developing steady state (design-point and off-design) and dynamic models of systems and components, including systems for stationary electric power, combined heat and power, and transportation applications. Various fuels (hydrogen, natural gas, alcohols, and hydrocarbons) and fuel reforming processes (steam, partial-oxidation, and autothermal) were modelled.

Subtask 5. Direct Methanol Fuel Cell

The work in this subtask was directed toward improving the performance and lifetime of direct methanol fuel cells (DMFCs). This involved the identification and development of new anode and cathode catalysts, design and testing of new electrode/electrolyte structures, as well as developing and analysing new concepts in DMFC stacks and systems.

Technical Achievements

The work of this Annex has resulted in many technical accomplishments. Some of the highlights from this work are summarised below.

Subtask 1. Improved Single Cells

- CO tolerance to 100–1000 ppm CO was demonstrated (Forschungszentrum-Jülich, Germany) for over 600 h with hydrogen peroxide injection into the anode gas. The CO tolerance of pure Pt anodes was better than that of Pt/Ru anodes.
- A spray-on technique has been developed for depositing well-controlled gas diffusion and catalyst layers in three-layer electrodes (ENEA, Italy). Effects of heat treatment:
 - treatment at 70°C creates voids and leads to the lack of continuity between the diffusive and catalytic layers.
 - treatment at 110°C provides better adhesion between the diffusion and catalyst layers, decreases ohmic and polarization resistance, and improves the kinetics of the oxygen reduction reaction.
 - treatment at 190°C leads to collapse of the structure, intermixing of the layers, and degradation of the ionomer.
- A hot-roller machine has been developed for manufacturing MEAs (Toshiba, Japan).
- Endurance testing of single cells (Mitsubishi Electric, Japan) has shown a degradation rate of 4 mV/1000 h; in a 3-cell stack, the degradation rate was 40 mV/1000 h. Particle size increased from 40 Å before test to 60 Å after test, presumably due to the migration of Pt to the surface of adjacent particles.
- Very fine Pt particles (1.0–1.7 nm) on carbon support have been obtained (Osaka National Research Institute, Japan) by oxidation of the carbon, followed by ion exchange with a Pt cation complex. Electrode performance was much better than for electrodes prepared by the conventional impregnation method.
- A “divided cell” technique showed that CO poisoning is most significant in the fuel inlet region of the cell (Mitsubishi Electric, Japan). This can lead to voltage oscillations, during which CO can be oxidized with water.
- Improved electrodes with reduced diffusion losses have been made by using a single gas diffusion layer rather than the double gas diffusion layers used in the standard E-Tek electrodes (ECN, Netherlands).
- Effective CO oxidation is obtained by alloying Pt/Ru (Southampton University, U. K.). Optimum promotion is achieved after oxidative cycling of the alloyed Pt/Ru.
- Passive ambient air button cells have achieved 230 mA/cm² and 40 W/kg at 0.5 V, with projected improvement to 400 mA/cm² at 0.5 V, and 118 W/kg (Loughborough University, U. K.).

Subtask 2. Improved Stack Materials and Concepts

- Measurements of oxygen transport (Simon Fraser University, Canada) in Nafion 117, BAM3G, DAIS 585, radiation-grafted polymers from Cranfield University (U. K.), and poly(styrene/sulfonated styrene) copolymers have shown

that high diffusion coefficients but low solubilities of oxygen in high-water-content polymers lead to permeabilities that are similar for many membranes (except for low water content membranes and the lowest equivalent weight DAIS membranes).

- A 2-kW, 12-cell, 625-cm² active area stack (for stationary application) was operated for 500 h with little degradation in performance (Toshiba, Japan). The next generation 5 kW stack (at 2 bar hydrogen/oxygen) will provide a power density of 500–700 W/L.
- 5-kW and 8.5-kW stacks have been developed and tested (Toshiba Corporation, Japan).
- Evaporative cooling in the fuel cell stack has been used to (1) remove one-third of total waste heat generated, and (2) contribute to a uniform temperature profile in the stack (Toshiba Corporation, Japan).
- The influence of metal impurities (Na⁺, Ca²⁺, Fe³⁺) has been measured in Aciplex-S 1004 membrane (Asahi Chemical Company, Japan). Cell performance degrades with increasing concentrations of these cations in the membrane.
- The effects of freezing cells to –20°C have been measured (Asahi Chemical Company, Japan); cell performance degraded with increasing cycle number with Aciplex-S 1004 but remained essentially constant with Aciplex-S-BH.
- Life test of a Flemion-R MEA at 0.5 A/cm² showed that the open circuit voltage (0.91 V), internal resistance (0.11 Ω-cm²), and iR-free voltage (0.67 V) were unchanged over 2300 h (Asahi Glass Co., Japan).
- Membranes reinforced with 100 denier PTFE woven cloth showed that (Asahi Glass Co., Japan):
 - dimensional stability can be increased by an order of magnitude
 - tensile strength, ductility, and creep characteristics are all improved
 - hydrogen gas permeability is almost the same as for the plain membrane
- Measurements of the cathode contact resistance on a stainless steel bipolar plate show an essentially constant value of ~30 mΩ cm² over 350 h, with the cell tested at 60°C, H₂/air, at 0.5 V (ECN, Netherlands).
- Lifetimes of PSI membranes up to 10,000 h have been demonstrated in 30-cm² single cells (Paul Scherrer Institut, Switzerland). Maximum power densities of >500 mW/cm² have been obtained.
- Work with multiple-sandwich Nafion membranes showed that the resistance increase at higher current densities is confined to the membrane adjacent to the anode (Paul Scherrer Institut, Switzerland).
- Several 100-W, 10-cell, 100-cm², H₂/O₂ stacks (with no active cooling or humidification) have been provided to various engineering colleges in Switzerland (Paul Scherrer Institut).

- In the development of alternative polymer membranes (Cranfield University, U. K.), it was shown that for radiation-grafted fluoropolymer membranes, the ion exchange capacity (IEC) could be directly related to the membrane's equilibrium water content. The maximum power output of the cell is also related to the IEC, so that the maximum power density can be related to the equilibrium water content of the membrane in the cell.

Subtask 3. Operating Parameters: Effects on Performance and Lifetime

- Exposure to 10-ppm ammonia in the cathode air resulted in a small, partially recoverable loss in cell performance. Repeated exposures did not degrade the cell performance further. Conductivity of the membrane was not reduced by the reaction of ammonia with the acidic ionomer (ECN, Netherlands).
- Exposure to 5 ppm sulfur dioxide caused severe loss of cell performance, such that at 0.8 A/cm², the cell voltage decreased from 0.5 V to 0 V in less than 10 h (ECN, Netherlands). Exposure to 0.5-ppm sulfur dioxide led to a voltage drop of 100 mV. Repeated exposures to this level of SO₂ further degraded the cell performance.

Subtask 4. Balance-of-Plant Components and Systems

- For methanol steam reforming, reducing the copper-zinc oxide catalyst in methanol and water rather than in hydrogen, yields higher activity that is maintained over a longer period of operation (Royal Military College, Canada). With the improved catalysts, the required temperature for reforming was reduced from 210°C to 170–180°C. A “hydrotalcite-like” structure is important for catalyst activity; conventional calcining tends to destroy this structure, resulting in catalyst degradation.
- An improved kinetic model of methanol steam reforming on Cu/ZnO/Al₂O₃ catalysts has been developed (Royal Military College, Canada). This model, which has been validated for pressures to >30 bar, is able to predict variation in CO production with methanol conversion.
- A membrane reactor has been devised for the generation of high-purity hydrogen at 4 bara by permeation through a supported palladium-alloy membrane with the reformer operating at 12 bara (Royal Military College, Canada).
- Detailed simulation of a methanol-based power train for light duty vehicles, with on-board fuel processing and an energy storage system, shows a system efficiency of 28–34% over the New European Driving Cycle for the vehicle, and 18–20% for the energy conversion chain (Forschungszentrum-Jülich, Germany).
- A 2-kW-class plate type methanol steam reformer has been developed (Mitsubishi Electric, Japan), as well as a 2-kW-class natural gas reformer (Sanyo Electric, Japan).

- A dynamic model of a methanol steam reformer has been developed for modelling reformer start-up and transient and steady state operation (Wellman CJB, U. K., and Royal Military College, Canada).
- System design, integration, and trade-off analyses have been conducted for automotive gasoline-fuelled PEFC systems (Argonne National Laboratory, U. S.). The operating cell voltages need to be in the range of 0.75 to 0.85 V to achieve ~ 45% system efficiency. System start-up analysis shows that with present-day catalyst amounts in the various fuel processing steps, the fuel processor requires 200–400 s for start-up. The corresponding fuel consumption is in excess of 0.5 kg. Reducing the catalyst amounts by 50% or more decreases the start-up time to a little over one minute and the start-up fuel consumption to 0.25 kg.

Subtask 5. Direct Methanol Fuel Cell

- Methanol permeation rates at 110°C and open circuit are equivalent to 90, 130, 250, and 520 mA/cm² for methanol concentrations of 0.5 M, 1 M, 2 M, and 4 M, respectively (Forschungszentrum-Jülich, Germany). Methanol permeation rates increase with increasing current density for 4 M methanol, but decrease with increasing current density for lower methanol concentrations.
- DMFC single cells of 125 cm² active area, 0.5 mg/cm² for the cathode and 0.25 mg/cm² for the anode, achieved a current density of 500 mA/cm² at 700 mV at 80°C, 4 bar H₂, and 4.5 bar O₂ (Forschungszentrum-Jülich). No decrease in cell performance was observed at a constant current density of 300 mA/cm² over 1000 h.
- Improved DMFC performance has been obtained by (ECN, Netherlands):
 - reducing cross-over by using a solution-cast membrane (180 µm) instead of Nafion 117
 - unsupported PtRu catalyst instead of PtRu/C
- Good stability has been achieved for up to 8 h for a 2-cell, 25-cm² stack at 90°C, producing 2.5 W on 1 bar oxygen and 2 W on ambient air (University of Newcastle, U. K.).
- Transition metal sulfides were found to be inactive to methanol oxidation, making them good candidate materials for use in the DMFC (University of Newcastle, U. K.). M^a(RuS)_x prepared on Ketjen EC-600JD carbon black showed the highest activity to oxygen reduction. The best composition was Rh, x=5. DMFCs using this catalyst gave cell voltages within 80–100 mV of those obtained with Pt.
- A prototype 500-W, 25-cell DMFC stack has been built with a total active area of 6800 cm² (University of Newcastle, U. K.).

Conclusions

The Executive Committee is invited to note the satisfactory progress achieved in the various subtasks during the 1996–1998 period of performance of Annex VIII. The

participants in the new Annex XI look forward to continuing collaborative R&D on polymer electrolyte fuel cells during 1999–2001.

Recommendations

The development of PEFCs has reached a stage where several commercial organizations are conducting sub-scale and full-scale demonstrations of the fuel cells in stationary, mobile, and transportation applications. Much of this hardware and systems development is likely to be proprietary and competitive, and not appropriate for an IEA R&D program. However, there is still a need for collaborative research in advanced materials (electrolyte polymers and membranes, catalysts, bipolar plates), balance-of-plant issues (fuel processing, system modelling, end-use aspects), and direct methanol fuel cells (improved performance and lifetimes, reduced crossover, etc.). It is recommended that these activities be pursued under the new Annex XI.

Annex VIII Publications

- “Analysis of energy and water management in terms of fuel cell electricity generation,” *J. Power Sources*, **71**, (1998) 294–301.
- “Methanol Tolerant Oxygen Reduction Catalysts Based on Transition Metal Sulfides,” R. W. Reeve, P. A. Christensen, A. Hamnett, S. A. Haydock, and S. C. Roy, *J. Electrochem. Soc.*, **145**, 3463 (1998).
- “A liquid-feed solid polymer electrolyte direct methanol fuel cell operating at near-ambient conditions,” A. K. Shukla, P. A. Christensen, A. J. Dickinson, and A. Hamnett, *J Power Sources*, **76**, 54 (1998).
- “Methanol Tolerant Oxygen Reduction Catalysts for Use in the Direct Methanol Fuel Cell,” R. W. Reeve, P. A. Christensen, A. J. Dickinson, A. Hamnett, and S. A. Haydock, 49th Annual meeting of the International Society of Electrochemistry, Kitakyushu, Japan, 1998, Extended Abstracts, p. 798.
- “Simulation of Fuel Cell Powered Vehicles,” R. Peters, H. G. Düsterwald, B. Höhle, Proceedings 31st ISATA, June 2–5, 1998, Düsseldorf, Germany.
- “Design, Integration, and Trade-Off Analyses of Gasoline-Fueled Polymer Electrolyte Fuel Cell Systems for Transportation,” R. Kumar, R. Ahluwalia, E. D. Doss, H. K. Geyer, and M. Krumpelt, *1998 Fuel Cell Seminar Abstracts*, November 16–19, 1998, Palm Springs, California, pp. 226–229.
- “Development of BAM membrane for fuel cell applications,” A. E. Steck and C. Stone, 2nd International Symposium on New Materials for Fuel Cell and Modern Battery Systems II, Eds. O. Savadogo and P. R. Roberge, Level 3b, July 6-10, 1997, Montreal, PQ, Canada, 792-807.
- “The development of a mathematical model of a methanol reformer,” Wellman CJB, ETSU Report F/02/00060/REP/2.
- “Membrane electrolyte technology for solid polymer fuel cells,” Cranfield University, ETSU Report F/02/00110/REP.
- “Initial assessment of the environmental characteristics of fuel cells and competing technologies,” WS Atkins Ltd., ETSU Report F/02/00111/REP/1.
- “The effect of contaminants in the fuel and air streams on the performance of a solid polymer fuel cell,” Wellman CJB, ETSU Report F/02/00126/REP.

- “The Ti(IV) salt of N,N- (diphosphonomethyl) -glycine: synthesis, characterization, porosity and proton conduction,” E. Jaimez, G. B. Hix and R. C. T. Slade, *J. Mat. Chem.*, **7**, 475–479 (1997).
- “Titanium(IV) dihydroxyphosphate: characterization and studies of ion exchange properties, n-alkylamine intercalation and proton conduction,” E. Jaimez and R. C. T. Slade, *J. Chem. Soc., Dalton Transactions*, 1435-1440 (1997).
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- “Investigation of radiation-grafted PVDF-g-polystyrene-sulfonic-acid ion exchange membranes for use in hydrogen-oxygen fuel cells,” S. D. Flint and R. C. T. Slade, *Solid State Ionics*, **97**, 299–307 (1997).
- “Structure and enhanced proton conductivity in composites of antimony(III) oxide and mordenite,” G. B. Hix, R. C. T. Slade and B. Ducourant, *Solid State Ionics*, **99**, 233–239 (1997).
- “Development of improved membranes for solid polymer fuel cells,” J. A. Crisp, K. V. Lovell, L. Lim and D. J. Highgate, presented at the Fifth Grove Fuel Cell Symposium, Commonwealth Institute, London, UK 22–25th September, 1997.
- “Dynamic model of solid polymer fuel cell water management,” H. P. L. H. van Bussel, F. G. H. Koene and R. K. A. M. Mallant, poster presentation at the 1997 Grove Fuel Cell Seminar.

Annex IX Fuel Cell Systems for Stationary Applications

Background

A fuel cell power plant consists of many components of which the fuel cell stack is the most important one. Furthermore, the complete plant is only one of several units in an electricity producing system or in a combined heat and power system.

Until the present, fuel cell development has been concentrated to achieve an applicable technology without much consideration of the total system where the fuel cell plant is to work. However, there are a number of reasons to establish, at an early stage, a better understanding of how the fuel cell works in the total system from fuel to electricity/heat. One of the most important issues here is the economy of the system.

In Annex V (first phase of the work) a number of these items were addressed. The intention of this task (Annex IX) became then to further develop what was outlined in Annex V, with a clear focus on technical, economical and environmental aspects of the implementation of fuel cells for stationary applications.

It seems to be of utmost interest for all the actors in the fuel cell field to further consider these aspects, especially in a longer perspective. As the experience from operation of fuel cell plants is still very limited, the further work has to concentrate on how the fuel cell technology can compete with other technical alternatives, what technical obstacles still remain to be overcome and what environmental and safety issues that should be further addressed.

The following report is the result of a collaborative work in Annex IX within the framework of IEA Advanced Fuel Cells. All participating countries have gained from the work and have made progress in the understanding of implementing fuel cell technology in the market. The parties recognise that the fuel cell technology may be one of the important technologies for the future, though it is not yet fully matured.

Objective

The main objective of the work has been to gain a better overall understanding of how stationary fuel cell installations correspond with outer sources and connections and, thus how they should be constructed in the most optimal way to meet the user's requirements from a technical, economic and environmental point of view. The main emphasis has been:

- to analyse the economics for fuel cells in different applications, i.e. to identify the most important parameters for the economics of fuel cells,

- to identify the present level of the technology, i.e. how far from commercialisation is the technology from a technical point of view, and

- to recognize how well fuel cells perform (manufacturing, operation etc.) from an environmental point of view.

The co-operation is thus to analyse technical, economic and environmental issues of common interest and to exchange such information between the Participants.

In the study the following fuel cells for production of electricity in all applications have been considered: Molten Carbonate Fuel Cell (MCFC), Solid Oxide Fuel Cell (SOFC) and Polymer Electrolyte Fuel Cell (PEFC) technologies. Phosphoric Acid Fuel Cell (PAFC) technology has been used as a reference technology. The power plants studied have been in the size range from 50 kW to 50 MW.

Participants and Organization of the Work

The Contracting Parties have been

- The Swedish National Energy Administration, STEM
- 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
- The Netherlands Agency for Energy and the Environment, NOVEM
- The Research Council of Norway
- ADEME, France

The operating agent has been Sydkraft AB, Sweden, acting through The Swedish National Energy Administration (STEM).

The following persons have participated in the work and thus given substantial contribution to the work.

Sweden Lars Sjunnesson, Sydkraft AB (Operating Agent)
 Bengt Ridell, Sycon AB
 Per Carlsson, Sycon AB
 Per Holmberg, Vattenfall Utveckling AB

Italy Agostino Iacobazzi, ENEA

Japan Kenji Kono, NEDO
 Masao Enami, NEDO

Germany Ulf Birnbaum, Forschungszentrum, Jülich
Petra Zapp, Forschungszentrum, Jülich
Armin Kraft, Forschungszentrum, Jülich

Netherlands Jan van der Veer, Kema

Norway Oystein Ulleberg, IFE
Fritjof Unnander, IFE

France André Marquet, EDF
Philippe Stevens, EDF

Each participating country has acted through their participants. However, their interest in the different subtasks have varied due to previous experience, business activities etc. The organisations/persons responsible for the subtasks have been;

Operating Agent: Sydkraft AB, Sweden (Lars Sjunnesson)
Subtask I: ENEA, Italy (Agostino Iacobazzi)
Subtask II: NEDO, Japan (Masao Enami/Kenji Kono)
Subtask III: Forschungszentrum Jülich, Germany (Ulf Birnbaum)

Task Meetings

January 29-30, 1996 at NEDO, Tokyo, Japan
April 22-23, 1996 at KFA, Jülich, Germany
October 8-9, 1996 at ENEA, Rome, Italy
May 26-27, 1997 at EDF, Paris, France
November 5-6, 1997 at Statoil, Trondheim, Norway
March 30-31, 1998 at KEMA, Arnhem, the Netherlands
June 24-26, 1998 at NEDO, Tokyo, Japan
September 17-18, 1998 at FZJ, Jülich, Germany

Task Description and Task Results

Task Description

In the work, the technologies Molten Carbonate Fuel Cell (MCFC) and Solid Oxide Fuel Cell (SOFC) technologies have been included. The sizes studied have been 100-1000 kW, 5-25 MW and 30-100 MW. The Phosphoric Acid Fuel Cell (PAFC) technology has, in each case, been used as a reference technology.

The work has been divided into three subtasks as outlined below.

Subtask I: Parameters affecting the economy and competitiveness of fuel cells
(Subtask leader: ENEA, Italy)

This subtask was basically a parametric study of the factors influencing the cost of power from fuel cells and from competing technologies.

In Annex V it was established that the competitiveness of fuel cells vary widely between different countries, due to differences in key parameters like electricity price, gas price, rates of interest and inflation as well as ratios between some of these parameters. It was also shown that environmental factors, such as restriction or taxation of CO₂ and NO_x emissions, have considerable influence on the competitiveness of fuel cells and have to be studied further.

Subtask II: Current technology level to commercialisation

(Subtask leader: NEDO, Japan)

This subtask aimed to study existing fuel cell technologies, evaluating how far the technology has reached in terms of the technical levels they must achieve in order to be commercialised.

PAFC technology today may be improved and the improvements are sometimes easy to predict. For the MCFC and SOFC technologies, calculations exist for the system they are to be working within. However, in most cases the detailed specification may show that the current technology is far from commercialisation. It is of importance to study how far from commercialisation the technologies are and the parameters that are most interesting to investigate.

Subtask III: Environmental analysis

(Subtask leader: Forschungszentrum Jülich, Germany)

This subtask aimed to investigate the environmental impacts of high temperature fuel cell technologies on a life cycle basis, thus performing a Life Cycle Assessment (LCA). Power stations based on SOFC, MCFC and conventional gas turbine combined cycle technology were investigated using PAFC data as a baseline. In Annex V, work was carried out to study the environmental impact 'costs' for the production of SOFC material. In this Subtask the work also included manufacturing of the entire fuel cell plant as well as the operation of the fuel cell and its dismantling. In this respect it is important to compare the results obtained also with the results when evaluating other technologies which perform the same service.

Task results

The factors or parameters that affect the introduction of fuel cell plants at the energy market can be classified into three main groups.

Utility factors:	Load factors, type of fuel, heat integration, grid connection etc.
Economic factors:	Fuel price, value of electricity and heat, interest rate, inflation etc
Technical factors:	Configuration of system, balance of plant, operation and maintenance schemes, efficiencies, lifetime etc.

In subtask I, the economic factors were in focus and sensitivity analyses were performed with respect to such important factors, e.g. the fuel price. To verify the results a special report on market opportunities of fuel cells in Germany based on the MARKAL model has been performed.

An important result from the work is that fuel cell plants seem to have a good possibility to enter the stationary energy market for different applications if the cost is 1000 USD/kWe. If in a market the value of energy, electricity and heat is high and the reliability of the service is important, fuel cell plants may be cost effective at costs as high as 2000 USD/kWe. Each market has its own conditions mostly depending on the ratio between the cost for electricity and the fuel used.

Another important factor is the O&M cost. One example shows that a decrease of the O&M cost from 1.3 to 0.65 US cents/kWh (50%) gave an increase in the acceptable investment cost of about 28%. The operational reliability of the new fuel cell plants subsystems, stack and fuel treatment systems has lately been demonstrated to be very high. Thus, the inherent potential of a low O&M cost is one of the very attractive characteristics that should give fuel cell plants an advantage in the energy market.

The advantage associated with high electrical efficiency of a fuel cell plant is becoming less and less important since competing technologies such as gas turbine plants, are also beginning to achieve very high efficiencies. However, the fuel cell technology can be the best option with respect to the environment and operational reliability. Furthermore, in addition to the excellent energy performance, very low maintenance costs associated with fuel cell systems are foreseen.

The analysis of subtask II included the following steps:

1. Data from the R&D schedules of fuel cell plants in the participating countries was collected, including the specifications of their current manufacturing costs and technology level in order to clarify the status of the technology.
2. Evaluation of the essential factors for commercialisation of fuel cells by focusing on the economical aspects, in order to identify the breakeven cost for commercialisation. Fuel cell plants were compared with conventional technologies such as gas turbines, diesel and gas engines. The current costs were compared with the target costs in terms of manufacture and investment in each application.
3. Discussion of the measures to cope with the gap between the current costs (1) and the break-even costs (2), and to give the feasible point of time and the conditions to initiate commercialisation.

Subtask III gave the following results:

The energy demand during the manufacturing phase of a gas turbine is significantly lower than that of a fuel cell. The difference can be explained by the fact that the fuel cell is a new technology with non-optimised manufacturing processes, especially in the case of evaluating laboratory data.

The results of the calculations show that the operation stage is the most energy intensive part of the life cycles of the two technologies. Due to the higher electrical efficiency of the fuel cell, its overall energy consumption is significantly lower than that of the gas turbine. The total energy demand along the entire chain of the studied 10 MW plant was calculated by summing the energy needed for each part of the life cycle.

There is no available information about emissions from solid oxide fuel cells because of very limited operation of such fuel cells. However, emissions are expected to be lower than those for gas turbines due to the characteristics of the fuel cell. The calculations of carbon-dioxide emissions showed that the figures for fuel cells should be lower than those of gas turbine because of the higher electrical efficiency of the fuel cell.

In conclusion, the introduction of fuel cell plants into the market is possible. This is particularly true for the increasingly mature fuel cell technologies. However, fuel cell plants still require a reduction in the system cost and increase of the reliability of the BOP components. Furthermore, the economics of fuel cell plants depends heavily on local factors, such as fuel price, energy prices, regulations, environmental requirements, inflation and interest rates. In the huge energy market that exists in the world of today, there is clearly a need to individualise the technologies and to find areas where fuel cells can be introduced at a relatively high investment cost in order to meet the still relatively high cost of fuel cells.

In order to reduce the costs, much effort should be made by the manufacturers as well as by the users of the technology. The manufacturers are to simplify their production systems without losing qualities, such as reliability and life time, and to find alternative materials and methods. The users' task is to explore the market, and promote introduction of fuel cells as an attractive technology. At the start it may be necessary to establish some niche markets by financial support from the government.

Conclusions & Recommendations

Fuel cell technology has been developed for a long time. Large efforts have been put in to various fuel cell technologies and on many levels. Reviewing the number of resources working in the fuel cell community it is still the fact that a large majority is within the basic research area and consequently a small minority in the very important market entry area.

There are many reasons that only a limited number of fuel cell plants are in operation and that still only one company (ONSI Corp. in the United States) is producing power plants for the market. This is even more remarkable as their first market entry took place in the early 1980's.

The main reason that fuel cells are not introduced more rapidly than could be expected seems to be the weak competition with other technologies that already exist and are also under rapid development. In only a few countries and in limited applications will the fuel cell fit in instantly. The market for fuel cells in different applications has been analysed from time to time but there does not seem to be a general understanding about the market entry applications.

The costs for fuel cells are, undoubtedly, too high today. Large efforts are put in to reduce all kinds of costs but it might take a long time to establish a lower cost structure as long as there is a limited market. However, if only one can accept this fact and that the market entry may take long time for all technologies, including fuel cells, it may be appropriate to expect a slow but steady market entry.

There is no doubt that the fuel cell technology in comparison with other technologies, in its size, and in combination with the very low environmental impact makes fuel cell technology very promising in the future.

With this general background the findings in the total work (subtask I-III) more specifically can be outlined as follows.

Technical Case Studies

Illustrative case studies of fuel cells plants located in favourable energy markets should be made. These case studies should be specific and should ideally be detailed investigations of actual plants. It is recommended that the key issues addressed in these specific studies include detailed investigations of:

- Balance of Plant (BoP)
- Reliability of system

In addition, to this there should be made an effort to define a set of realistic technical target goals for the various fuel cell technologies. Specifically, a target system efficiency (one for each type of fuel cell technology) should be found.

Non-technical Studies

Based on actual fuel cell plant field experience in various countries, a set of international standards and regulations as well as emission rules should be made.

The implementation of the fuel cell technology – as a new technology – should be focused and important parameters analysed.

Economic Studies

Economic studies similar to the one performed in this work should be further performed. However, first an exhaustive and systematic study of the *niche* markets for stationary fuel cell plants should be made. The most favourable fuel cell applications should then be identified. Finally, microeconomic studies, similar to the one presented in this report, could be repeated for each specific application.

It is important to emphasise again that only the most promising technologies and markets should be considered in such an economic study. In other words, the study should be based on specific applications. The advantage of this approach would be more realistic and less uncertain numerical results. Hence, the uncertainties associated with some of the most important economic parameters, such as the payback time, could be minimised.

Recommendations

With the background described in this report, but also in the previous Annex V, it is suggested that the work should be continued, concentrating on applications where fuel cells seem to be attractive in the short and medium term. The applications that should be focused on are suggested to be:

- PEFC for small scale applications
- SOFC alone or in combination with a gas turbine for application in industry or in a district heating system.

Work undertaken by several groups, in industry as well as at universities, has shown obvious advantages for the two above applications in the market.

Even so, there are several areas of importance to be analysed before introducing fuel cell technology to a large extent in the market. Areas to be focused on include obstacles/possibilities such as part load conditions, electricity quality, temperature levels, balance of plant, maintenance, life cycle assessment, and implementation of the technology over a wide range of applications.

Annex IX Reports

1. Final report including subtask reports
2. Parameters affecting the economy and competitiveness of fuel cells
3. Current technology level to commercialisation
4. Environmental analysis

Annex X Fuel Cell Systems for Transportation

Duration

January 1997 - December 1999

Operating Agent

Forschungszentrum Jülich GmbH, Jülich Germany

Participants

Forschungszentrum Jülich GmbH (Germany)
The Netherlands Agency for Energy and the Environment (NOVEM)
Department of Energy (United States)
Paul Scherer Institute (Switzerland)
Volvo (Sweden)

Task Experts

This section lists the Operating Agents and the other experts who have participated in those tasks which were active during the year. Each organisation is categorised as government or government agency (G), research institution (R), industry (I) or academic (A).

Experts:

E. Noreikat, J. Kinder	Daimler-Chrysler (Industry – I)	Germany
A. Lamm	"	"
P. Schmitz	Ford Forschungszentrum (I)	"
A. Seibertz	"	"
G. Lepperhoff,	FEV Motorentechnik (R)	"
A. Wiartalla	RWTH Aachen (A)	"
St. Pischinger	RWTH Aachen (A)	"
B. Höhle	Forschungszentrum Jülich (R)	"
P. Biedermann	"	"
M. Walbeck	"	"
M. Altmann	Ludwig Bölkow Systemtechnik GmbH (R)	"
R. Matthe	Adam Opel AG (I)	"
D. Hasenauer	"	"
F. Gossen	RWTH Aachen (A)	"
K. Wittek	"	"
J.-W. Biermann	"	"
R. Busch	Siemens AG (I)	"
J. Große	"	"

G. Erdmann	TU Berlin (A)	"
M. Grahl	"	"
R. Kolke	UBA Berlin (G)	"
A. Schütz, M. Sattler	Mannesmann Pilotentw. (I)	"
A. Edel	"	"
A. König	Volkswagen AG (I)	"
D. Dübel	"	"
R.T.M. Smokers	TNO Road Vehicle Research Institute (R)	The Netherlands
D. Schmal	"	"
Bradford Bates	Ford Research Lab. (I)	USA
R.I.Sims	"	"
Th. Schucan,	Paul Scherrer Institute (R)	Switzerland
A. Röder	"	"
A. Wokaun	"	"
E. Newson	"	"
P. Ekdunge	Volvo (I)	Sweden
St. Unnasch	Arcadis (R)	"

Objectives

This Task concerned fuel cell systems for transportation and competing systems. The objectives were to undertake economic analyses, system analyses, life cycle analyses and the analysis of fuel supply systems based on the needs of users and authorities.

Task Description

The Participants shared the co-ordinated work necessary to carry out this Task. The work in the Task was divided into the following three Subtasks:

Subtask A: "Economics and System Analysis" (G. Erdmann, D-TU Berlin)

- Goal 1: Overall system analysis and economics for fuel cells
- Goal 2: Definitions and identifications with respect to mobile systems
- Goal 3: Acceptance criteria and target markets for new transportation systems
- Goal 4: Selection, comparison and evaluation of different new transportation systems
- Goal 5: Summary and identification of key improvements of fuel cell systems

Subtask B: "Balances of Life Cycles" (St. Unnasch, USA-Arcadis)

- Goal 1: Review and definition of life cycles
- Goal 2: Determine energy consumption and emission impact
- Goal 3: Comparison of emission impacts

Subtask C: " Fuel Supply Systems" (M. Altmann, D-Ludwig-Bölkow Systemtechnik GmbH)

- Goal 1: Analysis of technologies
- Goal 2: Analysis of infrastructure
- Goal 3: Analysis of fuel supply systems

Background

The Task started in February 1997 as a new activity of the IEA programme on Advanced Fuel Cells. A number of both national and international efforts are being undertaken with the aim of developing solutions for improved or new energy conversion processes in the field of transportation. Specific programs for research and technological development as well as demonstration programs have been started on a worldwide basis. Priority is given to the development of more efficient, safer and environmentally acceptable systems for both passengers and freight road-transportation. These efforts also include new power trains with fuel cell systems (Polymer Electrolyte Fuel Cells - PEFC) serving as energy conversion units and the use of hydrogen, methanol, ethanol or gasoline as energy carriers.

Medium-term options for vehicle fuel-supply based on a PEFC-system and an electric motor are methanol, ethanol or gasoline. All of these provide high-energy density for the storage systems and include proved handling and infrastructure. A long-term option for PEFC-powered vehicles of the future could be hydrogen produced on a non-fossil basis. Methanol, ethanol and gasoline as energy carriers have to be converted to hydrogen or a hydrogen-rich synthesis gas in front of the fuel-cell systems by means of a reformer before chemically-bound energy can be converted to electricity and mechanical energy required for driving the vehicle. Methanol could also be directly converted in the fuel cell.

Passenger cars with fuel-cell power trains may show, in comparison with a 5-litre-gasoline / 100 km passenger car of the next generation, advantages in terms of energy consumption and carbon-dioxide emissions, especially for hydrogen-powered fuel cell vehicles (hydrogen stored in the vehicle). These advantages are lessened as soon as fuel production and fuel transport are considered in the calculation of full fuel-cycle balances. Fuel-cell powered vehicles, however, present considerable advantages in terms of regulated specific emissions and ozone formation potentials in comparison with ICE-powered vehicles.

Annex X (Fuel Cell Systems for Transportation) as part of the *International Energy Agency - Implementing Agreement 026*, started in spring 1997 and was intended for three years to deal with the above mentioned topics (see Task description). Five countries (The Netherlands, USA, Germany, Sweden, Switzerland) were official members of Annex X with 17 participating companies or research groups; they contributed to 3 Subtasks with different goals. The objectives of Annex X focused on economics and systems analysis (Subtask A), the balances of life cycles (Subtask B) and the analysis of fuel supply systems (Subtask C) based on the needs of both users and authorities as well as the results of IEA-Annex VIII and other information sources.

The following activities were undertaken in 1995 and 1996 to prepare the work program for the Annex, which was formally approved in 1997.

May 1995	1st discussion: ExCo meeting in Oslo (N)
September 1995	2nd discussion: ExCo meeting in Loughborough (UK)
February 1996	3rd discussion: ExCo meeting in Tokyo
May 1996	1st workshop: Annex X in Jülich
September 1996	4th discussion: ExCo meeting in Risoe (Dk)

October 1996 2nd workshop: Annex X in Berlin

Activities

The following activities were undertaken in 1997.

February	Approval of Annex X by ExCo - Start of work
February	1st Technical Advisory Committee in Cologne
May	Subtask C meeting in München
May	Subtask A meeting in Berlin
June	3rd workshop: Annex X at TNO in Delft
August	1st progress report to the ExCo
September	Application by Switzerland to join Annex

The following activities were undertaken in 1998.

January 4	10/11th circular: Announcement of the 4th workshop in Vienna (USA)
February 15	13/14th circular: Annual report 1997 to ExCo + experts
February 16	Report of Annex VIII to Annex X from R. KUMAR
March 5	Advisory Committee Meeting in Washington D.C.
March 6-7	4th workshop: Annex X in Vienna (Virginia, USA)
March 16	16th circular: Report Annual Meeting Vienna (USA)
April 22	Meeting of LDV experts (Annex X only) in Aachen
July/August	17/18th circular: Report 1/98 to ExCo + experts
September	Paul Scherrer Instituts (CH) and VOLVO (S) new experts for Annex X
October	19th circular: Program for Annex X meeting in Munich
November 2	5th workshop: Annex X / Subtask C LBSt-D-München
December 1	20th circular: Minutes of the Munich meeting
December 22	21st circular: Draft of the Annex X annual report 1998

The following activities were undertaken in 1999.

January	Annual report 1998 to ExCO
January	22 nd circular: Minutes of the VKA/FEV power-train meeting Aachen (Germany)12.1.1999
September 22/23	6th workshop: Final Annual Meeting in Switzerland (PSI)
October 25	Minutes of the final meeting in Switzerland (PSI); Sent to ExCo and all experts; Including a draft of final report Annex X and a proposal for a new Annex
December	Final report Annex X to all experts and to the ExCo

Technical Accomplishments

The following information was exchanged in addition to the subtask-specific accomplishments listed in the following paragraphs.

- List of all international projects dealing with the Task (Höhlein, FZ Jülich)

- List of all literature available dealing with the Task (Grahl, TU Berlin)
- Minutes of 3rd workshop (Delft) including Subtask reports (7th circular)
- Annex X- Status Report 1997
- Fuel Cells in Water Transportation (Schmal, TNO, NL-Delft)
- Questions to Annex VIII concerning specific data of PEFC and fuel processing for powertrain simulation calculation (Kumar, ANL)
- Partnership for a New Generation of Vehicles (PNGV), 4th report 1998
- Minutes of 4rd workshop (Vienna, 16th circular)
- Minutes of 5rd workshop (Munich, 19/20th circular)
- Fuel Chain Evaluation – Efficiencies and Costs.
(G. Erdmann, M. Grahl, TU Berlin, D-Berlin, see Munich 1998, Subtask A)
- Life Cycle Emissions and Emission Considerations for Partial Oxidation Fuel Processors; Evaluation of Fuel Cycle Emissions on a Reactivity Basis Vol. ½ (1996) - available at (St. Unnasch, ARCADIS Geraghty and Miller, Inc., Mountain View, CA, see Munich 1998, Subtask B)
- Product Structure and Marginal CO² Emissions of the Refinery Process (D. Martinsen, M. Walbeck, FZ Jülich – STE, D-Jülich, see Munich 1998)
- Strategy for the Introduction of a Common Worldwide Transitional Alternative Transport Systems (H. Quadflieg, TÜV Rheinland, D-Cologne, see Munich 1998)
- Comparison of Passenger Cars with Internal Combustion Engines, Hybrid Drive Lines and Fuel Cells from an Environmental Perspective (R. Kolke, A. Friedrich, Umweltbundesamt, D-Berlin, see Munich 1998)
- Comparative Analysis of Passenger Cars with Internal Combustion Engines or Fuel-Cell Systems Including Fuel Cycles (B. Höhle, P. Biedermann, FZ Jülich, D-Jülich, see Munich 1998)
- 2nd updated “Draft Proposal for Fuel Cell Vehicle Specifications”
- (R. Smokers, et al., TNO, NL-Delft, see Munich 1998)
- Critical Remarks on the Comparability of Database; Different Examples of Fuel Cost Evaluations (M. Altmann, Ludwig Bölkow Systemtechnik GmbH, D-Ottobrunn, see Munich 1998, Subtask C)
- Fuel Cells for Traction Application (P. Ekdunge, Volvo, S-Sweden, see Munich 1998)
- Properties of German and American Gasolines (Opel, D-Mainz, see Munich 1998)
- Status and Prospects of Fuel Cells as Automobile Engines (Kalhammer et al., prepared for ARB, CA, 1998, see Munich)
- Analyse von Kraftstoffketten (Schurig, LBSt, D-Ottobrunn)
- Wechselwirkungen der Brennstoffzelle mit der Infrastruktur
(Sattler, MPE, D-Munich, see Munich 1998)
- Fuel Cells for Transportation 98 (DOE, Fuel Cell Seminar, Palm Springs USA, 1998)
- Polymer Fuel Cell Testing for Naval Ship Applications, available at (D. Schmal et. Al, TNO, NL-Delft, 1998)
- Draft of Annex X- Status Report 1998
- Contribution by F. Gossen - power-train meeting in Aachen (12.1.1999):
- “Power-train analysis”, extract from a report of the Bmb+f-project (Germany 0329769)

- Contribution by the experts - final meeting in Wislikofen, Sept. 1999, see final report
- Literature review: Prof. G. Erdmann / Dipl.-Ing. M. Grahl - TU Berlin.

Subtask A:

In the first progress report of Subtask A (July 1997) we gave a description of the boundary conditions involved in this work. These included time horizon, seasonal and national focus as well as social, economic, ecological and political boundary conditions. The definition of these boundary conditions contributed to goal 2.1 of Subtask A.

As stated in that first progress report, the Subtask group had already begun to refine the overall boundary conditions by working on goal 2.3 (Definition of conventional transportation systems) and 2.4 (Definition of transportation systems with fuel cell systems) as well as on 4.1 (Selection of transportation systems for comparison). Consequently, the period under concern was used to accomplish the definitions and selection. The selection of the relevant powertrains is outlined in the table below.

After considering drive train options, energy chains were chosen to be examined further. The table below shows those combinations of primary energy, fuel and powertrain which were further examined, divided into two parts. The ‘realistic’ options, marked in bold, which were the main focus of further work in Annex X and the ‘additional’ options which have considerable potential for the years beyond 2010. For the common understanding of all subtasks, the table shows only the considered energy chains (Status: Erdmann, Grahl, Munich 1998).

Definition of Full Fuel Cycles

<i>Primary Energy</i>	<i>Fuel Cycle</i>	<i>Powertrain</i>
<i>Before 2010:</i>		
1	Crude Oil: Refinery - Gasoline	ICE
2	- Diesel	ICE
3	- Middle Distillate	ICE
4	Natural Gas: Gas Compression - CNG	ICE
5	Natural Gas: H ₂ -Production/Compression - CH ₂	PEFC - E-Motor
6	Liquefaction - LH ₂	PEFC - E-Motor
7	Natural Gas: MeOH-Production - MeOH	H ₂ -Prod. - PEFC - E-Motor
8	Crude Oil: Refinery - Gasoline/Diesel	H ₂ -Prod. - PEFC - E-Motor
9	- Middle Distillate (MD)	H ₂ -Prod. - PEFC - E-Motor
10	Mix Electricity	Battery - E-Motor
11	Mix of 1/2 + 10 Ref. - Gasoline/Diesel + Electricity	ICE - E-Motor
<i>Additional options after 2010:</i>		
12	Natural Gas: MeOH-Production - MeOH	DMFC - E-Motor
13	Biomass: Alcohol Production - MeOH/EtOH	
	H ₂ -Prod. -	PEFC - E-Motor

Based on contributions from the experts, at the meetings in Vienna and Munich as well as from other sources in 1998 "Fuel Chain Evaluation" in terms of energy consumption, stage efficiencies and costs could be summarized by Erdmann/Grahl (Subtask A, see technical accomplishments).

Proposals concerning the definitions of the different power trains were provided in 1998 by FEV, FZJ, TNO, ika and the automobile industry and were finally discussed and concluded at a meeting in January 1999 in Aachen (FEV) and at the final meeting in Wislikofen (September 1999).

Subtask B:

This Subtask combined the infrastructure scenarios developed in Subtask C with emission data for fuel production processes to determine emissions on a g/unit fuel basis. Vehicle fuel economy in Subtask A and fuel cycle emissions were combined to determine life cycle emissions on a g/km basis. Input was provided to the Annex meeting in Delft in May 1997. The (Delft) meeting focused on project definition, boundary condition, and assumption issues. Key issues that affect Subtask B include the following:

- Selection of infrastructure options (defined in Subtask C)
- Marginal versus average Emissions
- Macro system data analysis versus individual system analysis.

Emissions were divided into feedstock extraction, fuel production, marketing, and distribution. These categories helped to provide a comparison between different fuel technologies. In addition, dividing the fuel processing steps into categories helped facilitate analysis of marginal emission effects. Feedstock extraction emissions will be evaluated later. Many of the raw materials for fuel production are extracted outside the boundary of the study areas. The feedstock extraction emissions have a minor effect on air quality within the study areas. Fuel production emissions will be evaluated later. Fuel production emissions depend on the analysis of marginal

emissions. In California, most fuel production facilities will be regulated and permitted stationary sources which will be operating under various types of emission caps. The analysis of emissions needs to take into account the demand for fuel and the effect of fuel cell vehicles on existing facilities.

In 1998/1999 special inputs concerning “Life Cycle Emissions and Emission Considerations for Partial Oxidation Fuel Processors” from ARCADIS, other contributions from different experts as well as results from the workshops (see technical accomplishments) supplemented the information with respect to Subtask B.

Subtask C:

Work in the first half of 1997 concentrated on goal 3.1 *Definition of fuel supply chains* in cooperation with Subtask B. In the second half of 1997 detailed work was executed on the methanol supply path by Mannesmann Pilotentwicklung (goals 1 and 3). Extensive studies on various hydrogen supply paths were made available to the Subtask by Ford Motor Company and by Ludwig-Bölkow-Systemtechnik. These studies cover the following supply paths: biomass conversion to hydrogen, natural gas to hydrogen in a decentralised scenario, natural gas to hydrogen in a centralised scenario, renewable electricity (hydro power, wind power, solar thermal power, photovoltaics) in a centralised scenario and grid electricity in a decentralised scenario. The results of the studies cannot be compared directly as they have different objectives, different levels of detail and are based on different technical and economical boundary conditions. Also, the Ford studies relying on US boundary conditions such as natural gas, electricity and gasoline prices cannot easily be transferred to the situation in Europe.

The fuel supply paths investigated in detail within the Subtask are shown in the annual report to the experts (restriction level 1b). In addition to the hydrogen, methanol, natural gas and gasoline/ diesel supply paths the supply of a dedicated middle distillate was also considered. The supply paths have been chosen according to the following criteria (in order of importance for Subtask C selection):

- paths where information is available from the IEA Annex X partners
- paths where publications are available
- paths that are considered by industry (car industry, mineral oil industry, natural gas suppliers, industrial gases industry, electric utilities)
- ”sensible“ paths: sensible with respect to economics as well as with respect to ecology (resources problem, reduction of greenhouse gas emissions, energy efficiency etc.).

It was agreed to look into graphite nano fiber (GNF) hydrogen storage in spite of the fact that information is very scarce. The prospects of this storage technology are so promising and the infrastructural possibilities it opens are so different from existing concepts that GNF simply had to be considered even if it might turn out not to be feasible at all.

In 1998, contributions for Subtask C supplemented the information with respect to the goals (see technical accomplishments). The Subtask leader Mr. Altmann initiated a discussion on the availability, quality and comparability of data. For reasons of boundary conditions it was decided that the on-board fuel processor should remain part of the car. To solve the problems of Subtask C databases a *Diplomarbeit* on “system analysis for fuel supply options including comparison of literature data” was initiated. In 1999 other contributions from different experts as well as results from the workshops (see technical accomplishments) supplemented the information with respect to Subtask C .

Reporting

A full final report was prepared, comprising the following contributions:

FUEL CELL ELECTRICAL VEHICLES

F. Gossen (ika)	Comparison of fuel cell systems and other future power trains concerning energy efficiency
R. Smokers (TNO)	Assessment of the energy consumption of conventional and fuel cell passenger cars
R. Matthé (Opel)	Electric vehicles powered by fuel cell at GM/Opel – a global project
W. Schmitz (FORD)	Ford’s fuel cell activities
A. König (VW)	CAPRI Project
K. E. Noreikat (DChR)	NECAR Project
W. Schütz (MPE)	Hydrogen storage in carbon nanofibers
A. Röder (PSI)	Fuel cell cars and platinum-group metals (PGM)
R. Smokers (TNO)	Simulation of energy consumption for urban buses
R. Smokers/ A. Schmal (TNO)	Fuel cell ships

FUEL SUPPLY SYSTEMS

A. Huber/ M. Altmann (LBSt)	Hydrogen production costs for fuel cell vehicle applications
M. Sattler/ W. Schütz (MPE)	Infrastructure for Methanol
St. Unnasch (Arcadis)	Local fuel cycle emissions from fuel cell powered vehicles

INTEGRATED SYSTEMS ANALYSIS

R. Kolke (UBA)	Correlation between mobile and stationary fuel cell applications
M. Grahl/ G. Erdmann (TUB)	Competitiveness of fuel cell electric vehicles
P. Ekdunge (Volvo)	Analysis of the environmental impact of fuel cell vehicles

M. Walbeck (FZJ)	An alternative view on competitiveness and market introduction of fuel cell powered vehicles
G. Erdmann/ M. Grahl (TUB)	Macroeconomics of fuel cell electrical vehicles
PERSPECTIVES	
K. Noreikat (DCHr)	Future fuel cell development
B. Höhlelein (FZJ)	Comparison of studies - fuel cell powered vehicle propulsion systems

Workshops and Task Meetings

May 1996, 1st workshop: Annex X in Jülich
 October 1996, 2nd workshop: Annex X in Berlin
 February 1997, 1st Technical Advisory Committee in Cologne
 May 1997, Subtask C meeting in München
 May 1997, Subtask A meeting in Berlin
 June 1997, 3rd workshop: Annex X at TNO in Delft
 July-Dec 1997, Various expert meetings (Subtask A)
 March 1998, 4th workshop: Annex X in Vienna (USA)
 November 1998, 5th workshop: Annex X in Munich (D) at LBSt
 January 1999, Subtask A/B meeting in Aachen at FEV/VKA
 September 22nd/23rd, 1999, 6th workshop: Annex X in CH-Wislikofen.

Task Reports and Publications

- Annex X- Status Report 1997/1 (8th circular - August 15, 1997)
- Schmal, D. "Fuel Cells in Water Transportation" in: Annex X Status report 1997/1, TNO-ME.
- Höhlelein, B, Brennstoffzellensysteme für den Transport - Ziele des entsprechenden IEA-Annex X (IA026) Fuel cell systems for transportation - Goals of IEA - Annex X (IA026)
 In proceedings: 6. Aachener Kolloquium Fahrzeug- und Motoren-Technik, 22.10.1997, Aachen.
- Höhlelein, B, Fuel-Cell Systems for Transportation, in proceedings: Conference Commercialising Fuel Cell Vehicles, 20. - 22.10.1997, Frankfurt/Main
- Annex X- Status Report 1997
- Annex X- Status Report 1998
- Höhlelein, B., Critical assessment of power trains with fuel-cell systems, Global Power-Train Congress, October 1999 in Stuttgart
- Annex X Final Status Report Annex X 1999.

