ADVANCED DIR-MCFC DEVELOPMENT

A SMARTER CONCEPT FOR COGENERATION APPLICATIONS

P.J. Kortbeek, R.G. Ottervanger (editors)

BRANDSTOFCEL NEDERLAND BV (BCN)
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1 ABSTRACT

The high capital cost of fuel cell power plants is recognised as one of the major obstacles that prevent penetration on the market. Careful selection of suitable markets, identification of legislative, environmental and end-user requirements and cost reduction by the integral development of system and stack. These are the key elements in a three-year (1996 - 1999), joint European 'Advanced DIR-MCFC Development' project, in which the Dutch Fuel Cell Corporation (BCN), Netherlands Energy Research Foundation (ECN), Stork and Royal Schelde, BG Technology plc, Gaz de France and Sydkraft have co-operated. The project covered three major fields: 1/ Market analysis; 2/ System design and 3/ Stack development. The scopes and results are summarised below:

Market analysis
From a variety of sources, such as literature surveys, dedicated databases and interviews, a massive amount of data was collected on the global and European energy market. Special attention has been paid to the market trends, since it may take five to ten years before fuel cell systems may enter the market. Based on the market data, the knowledge of the MCFC technology and of the competing technologies, the most suitable and achievable markets were selected applying a SWOT technique. A natural gas based co-generation DIR-MCFC power plant with a rated power output of 400 kWe and delivery of steam, hot water and/or cold will have a wide range of applications in the commercial and industrial sector. Hospitals, offices, leisure centres and small industries are good examples where these plants could be installed. Competitors are gas-engines, gas-turbines and central power/steam boiler systems. Critical success factors are the cost of the produced power and thermal energy, as is the system reliability. A capital cost between 500 - 1000 EURO/kWe installed is acceptable for the European market. An extensive set of functional and technical specifications has been defined for the selected application.

System design
A highly innovative system and stack concept (coded the SMARTER™ system) has been developed in an interactive process. It was optimised to achieve the lowest cost of energy (cost per electric and thermal kWh), a high reliability and availability, and a wide operational window necessary to allow part load operation and anticipate on stack degradation. The ambient pressure basic system module contains three DIR stacks mounted on a multi-functional header box. The process equipment consists of an air compressor, an anode exhaust gas burner, an anode gas recycle blower and a desulphuriser. This compact system can be packaged in a 30 feet standard container. The output of the module is 420 kWe DC power and cathode off gas with a temperature of 670°C. The DC/AC converter and the heat recovery unit are mounted in an additional skid. Special attention has been paid to the controllability[1] of the system. Two dynamic simulators were built to study the system dynamics. The system shows remarkable stability and no hot valves are required for control functions.

Stack development
A full scale, system dedicated, internal manifolded, co-flow Direct Internal Reforming (DIR) stack with a targeted service life of 25,000 hours has been designed that meets the different requirements for each of the three stacks in the system. A 3-D integrated stack model (electrochemical, hydraulic and thermal) and endurance models for porous and metal components were used as design tools. Each stack contains two types of cells: the fuel (anode)

off-gas is either fed to the anode recycle blower (recycle anode) or to the burner (exhaust anode). This improves the overall performance and temperature management within the stack. The fuel supplies to the anodes of the three stacks are connected in parallel and anode gas recycling is applied, the oxidant supplies to the cathodes of the three stacks are connected in series. One of the most critical issues in the stack design is the cell sealing. The separator plate has been adapted to the mechanical brittleness of the matrix. Furthermore the separator plate accommodates the reforming catalyst, developed by BG. The porous components comprise a γ -$LiALO_2$ fibre reinforced matrix, an in-situ oxidised Ni cathode and a Ni-Cr anode. A mixture of Li/Na/K carbonates has been selected, that fulfils lifetime and performance criteria. All porous components were produced at ECN with environmentally friendly methods. The solutions for the critical stack issues have been verified in a large number of lab and bench cell tests, in 12 stack tests on 0.1 m² scale and in various dedicated out of cell tests.

An extensive cost analysis for the system, including the stacks has been carried out at the end of the project. Based on vendor information and on a assumed production volume of 50 systems per year and a stack production volume of 200 MW/year, a cost level of 3000 EURO was obtained.

Co-operation between the project partners

The project was characterised by an intensive collaboration between the partners. Many tasks were carried out in task forces, thereby making optimal use of the combined expertise of the partners. Good examples are the market analysis, which was carried out in a joint effort by BG, GDF, SYD, Stork and BCN and the concurrent engineering process of system and stack design, where BG, GDF, ECN, Stork and Royal Schelde co-operated closely together.

Conclusions:

1/ The major objectives of the project, namely selection of suitable markets and cost reduction by an integral development of system and stack, have been achieved.

2/ As most attractive initial markets for DIR-MCFC systems were identified the commercial sector, especially hospitals, hotels and leisure centres, but also opportunities exist in the light process industry. The optimal system size was determined to be around 400 kWe with delivery of steam, hot water and/or cold.

3/. Competing technologies are the grid, combined with boiler systems, gas engines and gas turbines.

3/ Liberalisation of the energy market gives large uncertainties for the near future. A consequence of the increased competition in power generation is lower market prices for electricity and less willingness of the energy sector to invest in new technologies.

4/ A highly innovative system and stack concept (coded the SMARTER™ system) combines maximum simplicity, a high reliability and availability, and a wide operational window in order to obtain the lowest cost of energy (electric and thermal).

5/ Dynamic analysis of the SMARTER™ system showed a remarkable stability.

6/ A full scale, system dedicated, internal manifolded, co-flow Direct Internal Reforming (DIR) stack with a predicted service life of 25,000 hours has been designed, making use of advanced design tools and models.

7/ The solutions for the critical stack issues have been verified in a large number of lab and bench cell tests, in 12 stack tests on 0.1 m² scale and in various dedicated out of cell tests. In the final 20 cells stack test, the integration of all solutions was demonstrated successfully under representative (system) conditions.

8/ A detailed cost analysis, based on a production volume of 50 systems per year and a stack production volume of 200 MW/year, resulted in a total system cost level of 3000 EURO/kWe.
9/ Technical breakthroughs, significant changes in emission regulations, or higher fuel prices are a prerequisite for further commercialisation of the technology.

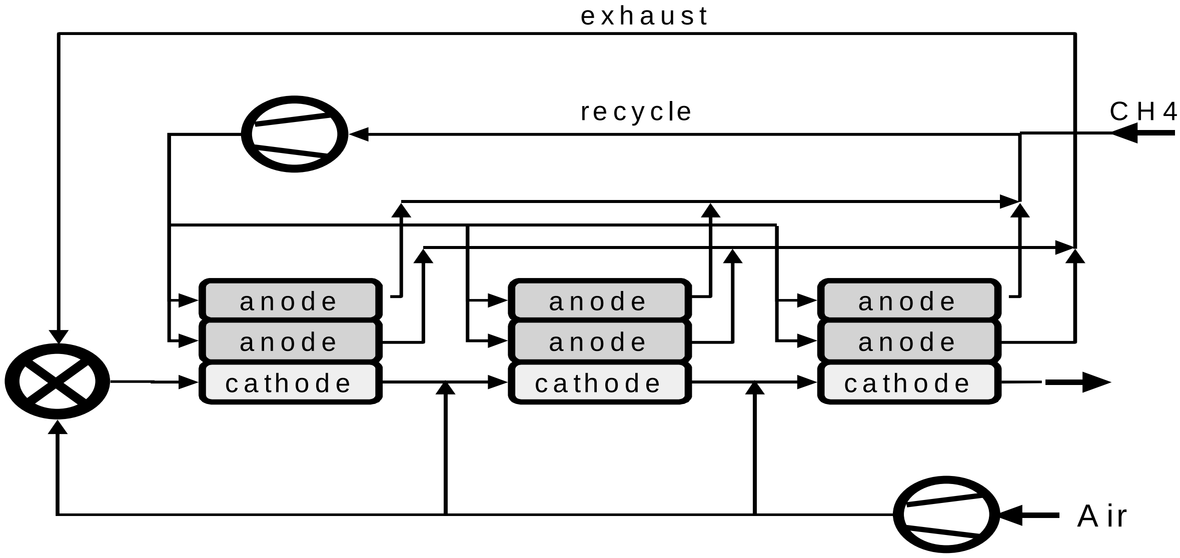


Figure 1. A schematic representation of the SMARTER™ system

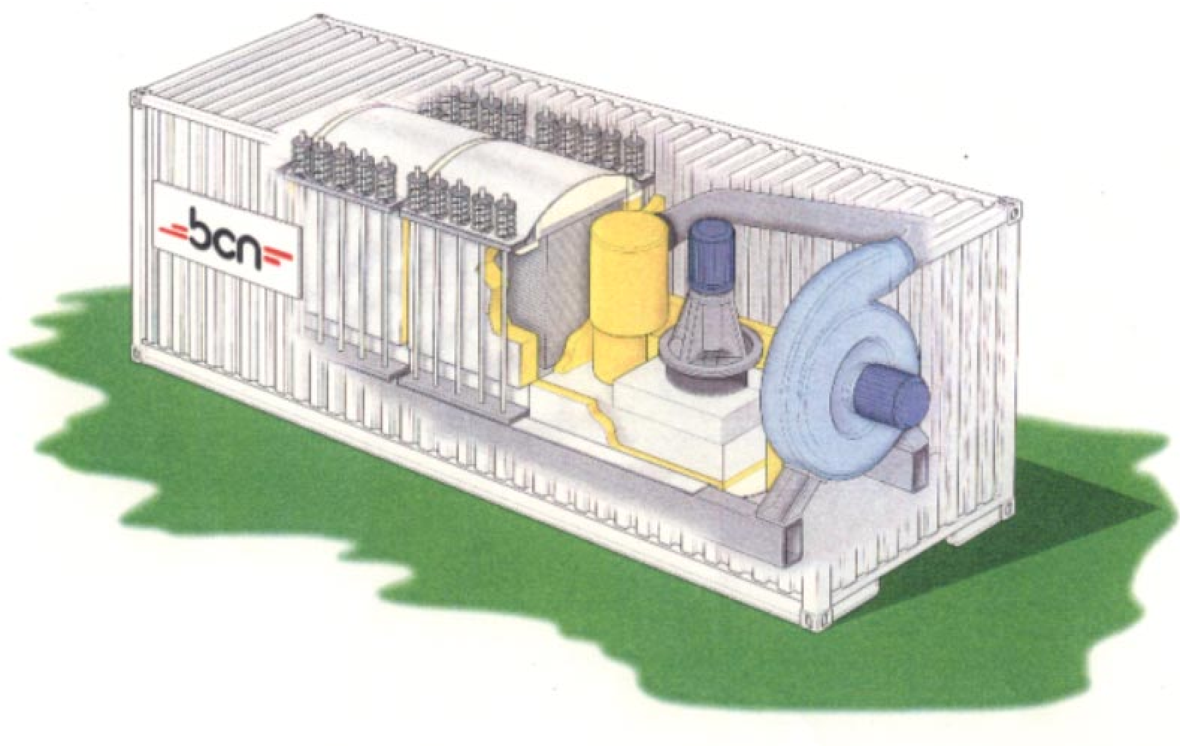


Figure 2. Conceptual design of the 400 kW DIR-MCFC power plant

2 PARTNERSHIP

Co-ordinator: Brandstofcel Nederland BV (BCN)
Contact: Dr. P.J. Kortbeek
Address PO Box 1093
1940 EB Beverwijk
the Netherlands
Phone +31 251 279 254
Fax +31 251 223 887
E-mail sae.bcn@wxs.nl
or: p.kortbeek@spe.storkgroup.com

Partner: BG Technology Plc. (BG)
Contact: Dr. A.L. Dicks
Address Ashby Road, Loughborough
Leicestershire LE 11, 3GR
United Kingdom
Phone +44 1509 282 818
Fax +44 1509 283 144
E-mail andrew.dicks@bgtech.co.uk

Partner: Gaz de France (GDF)
Contact: Dr. E. Gehain
Address B.P. 33
93211 La Plaine Saint Denis Cedex
La France
Phone +33 149 225 965
Fax +33 149 224 967
E-mail etienne.gehain@edfgdf.fr

Partner: Energieonderzoek Centrum Nederland (ECN)
Contact: Mr. G. Rietveld
Address PO Box 1
1755 ZG Petten
the Netherlands
Phone +31 224 564 452
Fax +31 224 563 489
E-mail g.rietveld@ecn.nl

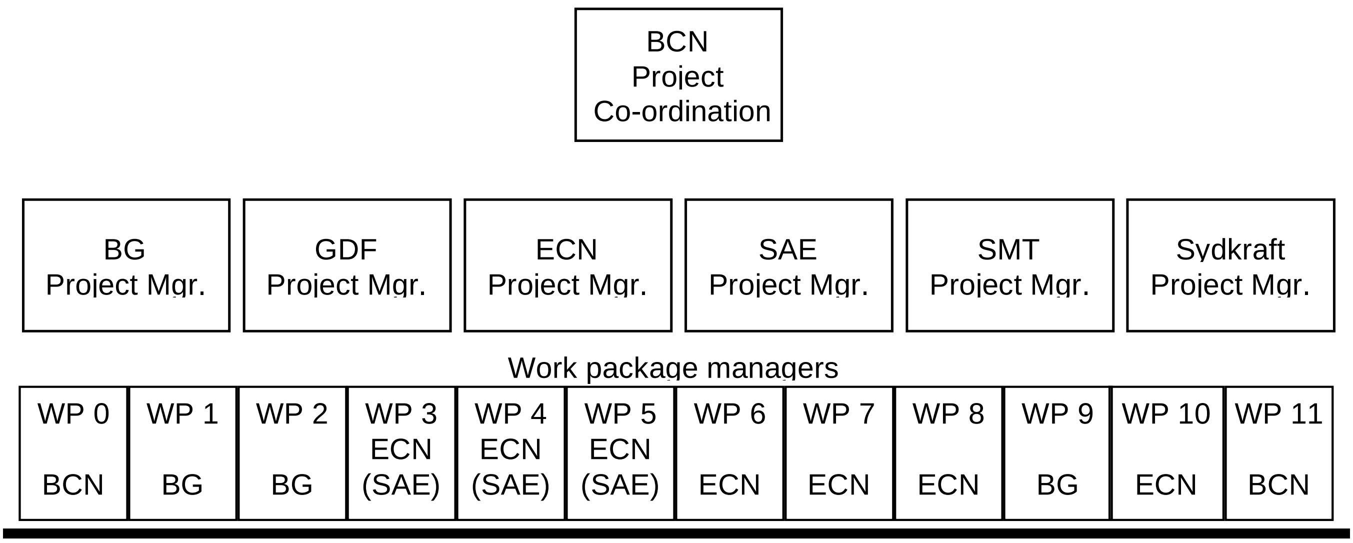
Partner: Stork (SAE)
Contact: Mr. E.W. Sloetjes (Stork Product Engineering)
Address PO Box 379
1000 AJ Amsterdam
the Netherlands
Phone +31 205 563 444
Fax +31 205 563 556
E-mail w.sloetjes@spe.storkgroup.com

Partner:	Schelde Technology Services BV (SMT)
Contact:	Mr. J.J. van der Goes
Address	PO Box 575 4380 AN Vlissingen the Netherlands
Phone	+31 118 485 700
Fax	+31 118 485 710
E-mail	-

Partner:	Sydgas AB (SYD)
Contact:	Mr. A. Molin
Address	Malmövägen 7 S-26590 Åstorp Sweden
Phone	+46 42 56 572
Fax	+46 42 54 690
E-mail	Anders.Molin@Sydgas.Sydkraft.se

2.1 Project organisation

BCN is the co-ordinator of the project. BG, GDF, ECN and Sydkraft are the partners. SAE and SMT are major sub-contractors of ECN.
BCN is responsible for the overall project management and for the liaison with the European Commission. Each partner appointed a project manager, who is responsible for all the work carried out by that partner. The co-ordinator and the project managers form the Project Team. Work Package Managers are responsible for the activities and the daily progress of their work package. The Work Package Managers report to the project manager and to BCN. A schematic presentation of the organisation is shown in the figure below.



3 OBJECTIVES OF THE PROJECT

The objectives of the ‘Advanced DIR-MCFC Development project are:

1. The selection of a specific combined heat and power (CHP) application of a DIR-MCFC system in the range of 200 - 2000 kWe and fuelled with natural gas or other methane containing fuels.
2. The development of a conceptual system design for this application with optimal reduction of the system complexity and investment costs.
3. The development of a second generation DIR-MCFC technology based on the system requirements, with an innovative approach on life-time extension and with highly efficient thermal, hydraulic and electrolyte management.

The objectives are described in more details below:

1. An achievable application of a DIR-MCFC in the range of 200-2.000 kWe will be selected, based on the specific characteristics of the DIR-MCFC technology and the expected advantages of the system relative to competing technologies, such as gas engines, gas turbines, the grid, etc.
2. Functional and technical system specifications will be established for this application
3. A conceptual system design will be developed according to the functional specifications and optimised for lowest cost of energy, high overall efficiency and high flexibility in heat-to-power ratio.
4. The availability of system components and sub-systems will be assessed. Specifications for non-standard components will be listed, based on own expertise or in co-operation with possible co-developers or vendors.
5. A full-scale second generation DIR-MCFC stack will be designed, tailored to the system specifications.
6. The stack (cell) internal sealing will allow pressure differences between fuel and oxidant flow up to 200 mbar. The same value is specified for the external sealing i.e. between stack interior and the environment.
7. The targeted stack service life is 25,000 hrs.
8. The ‘state of the art’ separator plate corrosion protection and porous component lifetime will be substantially improved.
9. A 3 dimensional integrated stack model will be further developed and will form the base for the stack design.
10. Design solutions for critical aspects in the full scale design will be verified in dedicated out-of-cell tests, in lab and bench scale tests and in stack tests up to 0.1 m^2 and 20 cells.
11. The experimental verification of the stack service life will be carried out in a 3000 hours stack test under representative conditions. The estimate of the service life will be based on post test analysis results and extrapolations based on models that describe the most critical stack service life limiting processes, such as cathode dissolution, separator plate corrosion built-up, electrolyte losses, etc.
12. The cost of the system, including the stack(s) will be assessed.

4 TECHNICAL DESCRIPTION

The technical description of the work carried out under the contract is summarised per work package.

Work Package 1: System Definition

This work package was has been led by BG, whereas GDF, SYD, SAE and BCN contributed. Many activities were carried out by task forces. The work package was divided into six tasks.

<u>1.10 Market analysis</u>
The first goal of the present project is to identify achievable markets for natural gas fueled co-generation systems using DIR-MCFC technology. A market study has been carried out which brought together global market information and individual country data by market sector, including information on energy consumption and energy cost.

<u>1.20 Customer requirements</u>
Customer requirements were obtained through a series of interviews with typical hospitals in several European countries. Essential requirements are low cost/kWe, high reliability and availability, flexible heat/power ratio, high electrical efficiency, steam production, low emissions, low noise and good part-load characteristics.

<u>1.30 External requirement</u>
Under this task external requirements, such as standards and legislation, were collated for seventeen European countries. Recommendations were formulated with respect to critical issues, namely electrical grid connection and safety, pressure vessels, fuel processing, gas grid connection, fire safety, noise and water discharge. Difficulties observed are is the facts that no specific regulations apply to fuel cell systems and that the legislation concerns three levels, namely a local, a national and a EU level.. This may lead to uncertainties in the external requirements, and to long procedures before the required permits are obtained. Moreover, standards and legislation are subjected to changes due to the process of liberalisation going on in several EU countries and elsewhere.

<u>1.40 Relevant market position</u>
The aim of this task was to estimate the relative market position of DIR-MCFC systems compared to competing technologies (e.g. gas engines, gas turbines etc.). The relative market position was analysed using a classical SWOT analysis (Strengths, Weaknesses, Opportunities and Threats) to firm up on the application selection. The SWOT analysis confirmed that in Europe, hospitals are the preferred market entry application at a size range of 300-400kWe.

<u>1.50 Competing technologies</u>
Competing technologies in the targeted size range are typically gas engines, gas turbines and the grid/steam boilers. The allowable investment for this size of co-generation plants varies throughout Europe but is typically between 800 and 1200 EURO/kWe.

1.60 Selection of suitable application

The type of application and the optimal size of DIR-MCFC power plants is mainly determined by the economic environment for power and heat generation.. Opportunities for DIR-MCFC plant will exist in many commercial and industrial market sectors (in the range up to a few MW of installed capacity). However, the commercial sector and especially applications in hospitals and hotels are the preferred initial market. Hospitals are in particular attractive since they are a ‘universal’ market and have a suitable heat-to-power ratio, continuous operation and steam demand.

Work Package 2: Foundation

This work package was led by BG, whereas GDF, SYD, SAE and BCN contributed. The nine tasks formed the foundation for further system development in work packages 3 and 5.

2.10 Select base systems

Five very different DIR-MCFC system configurations were chosen to provide an indication of the sizes and operating conditions for the balance of plant equipment to be assessed in the project. Another aim was to establish a base for investigations on issues such as safety, reliability and start-up.

2.20 Tools development

Originally it was considered worthwhile to build a large database for the system components as a resource for design in later stages of the project. As the work progressed, the need reduced due to the strongly reduced number of components in the system. Several flow-sheeting tools were available at the start of the project. Through an assessment, including benchmarks, a selection was made. Very useful were a spreadsheet model developed by BG and ASPEN, extended with dedicated routines. ECN' s stack model, which is based on first principles and extensively checked with experimental stack data, served as the master stack model. Dedicated techno-economic models were developed for the cost optimisation during the design and for the assessment.

2.30 Sensitivity analysis - System 3BG5

System 3BG5 had been developed within a previous EU-supported project. Further analysis showed that it has many advantageous features. Additional flow-sheeting calculations were carried out to examine the sensitivity of stack operating conditions for changes in the various system parameters. The information formed important input for the initial interface specification under WP 4,

2.40 Standards and regulations

GDF investigated the important issues concerning standards and legislation for DIR-MCFC power plant, such as pressure vessel regulations, electrical grid connection and safety, air quality standards, gas grid connection, fire safety, noise level and water discharge. The work provides a general guidance but local/country regulations need also need to be taken into account

2.50 Carbon deposition studies

Carbon deposition is considered to be a critical issue in the DIR-MCFC. A literature survey on carbon deposition in MCFC systems, stacks and reforming catalyst, has identified that different types of carbon result from gas phase pyrolysis and catalysed decomposition of hydrocarbons and CO. Formation of carbon or coke depends mainly on the prevailing gas composition and temperature. The carbon deposition boundary over a BG catalyst has been determined experimentally using thermo-gravimetry for equilibrium gas mixtures. Further experimental work will be carried out, focusing also on characterising carbon deposition within the system and stack hardware.

2.60 HAZID

A hazard identification analysis (HAZID) has been carried out. The 3BG5 system served as base-case. Safety issues were explored by a multi-disciplinary team of scientist and engineers on the base of a series of keywords (e.g. pressure, temperature, emission, containment), hazard category codes (e.g. personnel injury, equipment damage) and severity codes (ranging from minor to catastrophic). The HAZID report will played an important role in the preliminary and detailed design phase.

2.70 Component characteristics

Critical components in the balance of plant have been identified and pursued. These are principally the anode exhaust combustion, the anode hot blower and the gas clean-up (desulphurisation) system. Suppliers of hot blowers have been assessed and preferred manufacturers have been identified.

2.80 Development plan, critical components

A program of tests was drawn up for a burner developed by BG. These were completed and demonstrate that it is feasible to combust exhaust gas from a stack operating at up to about 88% utilisation, using a non-catalytic combustion. Combustion modelling has also been carried out and plans for further burner development are under consideration.

Two methods of desulphurisation of natural gas were selected: (a) a low temperature process and (b) high temperature absorption.

2.90 Functional and technical specifications

The functional and technical specifications (FTS) are set by the customer and external requirements, and by technical boundaries. A reference hospital was devised, based on analyses of typical hospitals in the Netherlands, France and the UK. Typical electricity and heat demand profiles were selected. The functional and technical specifications for the reference hospital forms the basis for the conceptual system design in WP 3 and the preliminary system design in WP 5.

Work Package 3: Conceptual system design

This work package was divided into six tasks (3.10 - 3.60). SAE had the lead, whereas BG, GDF, and SMT contributed in task forces.

3.10 Define selection and optimisation criteria

Under activity 3.10 two selection stages, each with its own set of selection criteria, have been defined: one for the pre-selection of basic configurations, and one for the final selection in activity 3.40.

3.20 Select Systems

Twelve basic configurations have been pre-selected and ranked, based on the first set of selection criteria, as mentioned under 3.10. Four Systems have been regarded to be promising and have been investigated in more detail in activity 3.30.

3.30 Analysis of the systems

A task force involving BG, GDF, ECN, SMT and SAE carried out the activities. The nominal load, part load and several degradation strategies have been calculated for the pre-selected systems. The controllability, pressure management, cost analysis and modularity were also important aspects of the analysis.

3.40 Ranking and selection of the systems

A trade-off has been made between four systems. Two systems were clearly preferred over the other: the 3BG5 and SMARTER™ system. The SMARTER™ system has been selected, due to its improved temperature control (reduced risk of carbon deposition) and higher power density, and therefor lower cost of electricity. Uncertainties were identified regarding the controllability of both systems.

3.50 Outline design of selected system

A base case 400 kW AC natural gas SMARTER™ system has been worked out. The start-up and other operational modes have been thoroughly investigated. Dynamic system simulations showed that the system is stable and can be controlled which simple equipment. Finally, stack model calculations and stack tests showed that the selected SMARTER™ system has a high power density and an improved temperature control, whereas the risk of carbon deposition is reduced.

3.60 Subsystem concepts

The stack module concept has been found suitable for the SMARTER™ system. The flow control and start-up equipment has been considered feasible. A study of the availability a hot anode recycle blower and a lean gas burner has been started.

Critical items that require extra attention in Work package 5 are the burner and carbon deposition. Less critical, but still of considerable importance are control, hot blower and desulphurisation.

Work Package 4: Interface management

This work package was led by SAE and contained two tasks.

4.10 To define and specify the interface boundaries

Under this task the initial Interface Document was issued, that contains a description of the interfaces between major process functions in Advanced DIR-MCFC systems. Boundary values for critical phenomena that may occur in each of the process functions have been specified and checked.

4.20 To guard and up-date the interface specifications

The initial Interface Document was regularly up-dated, according to the progress in system and in stack design and testing. Also the uncertainties in critical boundary values reduced in the course of the project. Other activities in this task were related to the identification of overall optimisation parameters that simultaneously govern cost, performance and lifetime aspects in the different process sections of the fuel cell system. Furthermore, test conditions for stack tests at ECN were specified in order to reduce uncertainties in boundary conditions.

Work Package 5: Preliminary Design

This work package contained four major tasks and was led by SAE, whereas BG, GDF, SYD, SAE and BCN contributed in task forces. The SMARTER™ system configuration and a modular system layout have been elaborated into a Preliminary Design of sufficient detail to meet the project objectives.

5.10. Modelling, design and engineering

Based on the results of WP 3 and 4, a process description and control philosophy was issued. The net electrical efficiency at nominal load ranges from 48,5% (begin of stack life) to 41,5% (end of stack life). At 30% part load these figures are 57,4% and 55,1% respectively. The thermal efficiency (averaged over lifetime) is 40% at nominal load and 24% at 30% part load.

Dynamic simulations were carried out to verify the control systems and strategy. Load variations from 100% to 30% and vice versa were simulated. The system showed fast responses, in the range of seconds, to load and flow control actions. Thermal stability is reached after several hours due to the large thermal mass of the stacks.
The simulations also pointed out, that the anode flow distribution to the stacks can be realised by trim valves. If a flow distribution for the recycle anodes and the exhaust anodes is set at nominal load, the setting will be maintained at part load within close range.

5.20 HAZOP

The HAZID carried out as part of Work Package 2 was re-examined The most important hazards are gas leakage, burner malfunctioning, flow control problems and electrical failures. The recommendations are to a large extend implemented in the design, such as ventilation, thermal insulation and accessibility for servicing. Other recommendations, e.g. concerning control and instrumentation and DC - AC conversion, will be implemented in the detail design.

5.30 Sub-system design

A design philosophy of the stack module has been formulated. Hot parts of a MCFC system need overhaul at regular time intervals. A "large overhaul" includes the exchange of stacks. Various components will be subjected to regular overhaul, whereas other parts of the system have long life and/or need little maintenance. Reducing the downtime required for overhaul can increase the availability of the system. The quickest way is to concentrate all parts that require maintenance into exchangeable modules. In order to minimise the heat loss of the system an aim of the design is to concentrate all hot parts (600-700°C) within a single thermal insulation shell. The stack module elaborated in WP5 can be considered as a complete system using purified natural gas as fuel and delivering DC power and hot exhaust gas.

The module has a sound proof enclosure with external dimensions of 12 x 2,5 x 2,6 m.

The gas distribution system conducts the anode and cathode gas flows. The main part of the gas distribution system consists of a header box, which also carries the stacks. The box provides a compact and (relatively) cheap solution for the gas distribution. Mixing of gases in the header box, especially recycle gas in the header box is extensively examined, since inadequate mixing could possibly lead to carbon formation. Computational fluid dynamics (CFD) and flow visualisation may yield more information if that is required for a detailed proof-of-concept design.

5.40 Preliminary design

1. Header box: The header box is made of stainless steel. Welds separate the anode and cathode compartments from each other. The box has to resist a high compression force due to the weight of the stacks. Electric insulation is realised between the stacks and the header box.
2. The anode recycle hot blower for an atmospheric MCFC plant can be manufactured with standard technology, provided that special attention is given to the impeller, the bearing configuration and the shaft.
3. The burner in the SMARTER™ system preheats the cathode gas to the stack inlet temperature and supplies CO_2 to the cathode. The burner has to operate under ultra lean gas conditions. Catalytic burners can cope with such lean mixtures but they are expensive and vulnerable. A pilot flame is considered necessary in order to cope with adverse transient conditions occurring at sudden load changes. Burner tests have been carried out in BG test facilities Based on the experimental results and on additional simulation data, the conclusion was drawn that a conventional can be used for a 400 kW system. The estimated burner volume is about 132 litres. The upper limit for combustion is determined to be a fuel utilisation” of 88%. No commercial manufacturer could be identified to deliver a suitable conventional burner. Therefor also manufacturers of catalytic burners were approached. In a follow-on program up-scaling and full size testing of the present burner design may become unavoidable.
4. Carbon deposition was further explored making use of the results from Work Package 2. A new thermal balance was commissioned to study carbon deposition on stack and system hardware as well as on the steam reforming catalyst in order to study the decomposition of methane over catalyst materials and C deposition via the Boudouard reaction. These experiments confirmed the boundaries for carbon deposition via the two reactions, as established earlier in Work Package 2.
5. Start-up heating: The start-up heating system is used during cold start-up to preheat the stack module from ambient temperature to operating temperature (>620ºC). The system can also be used to compensate heat losses during hot standby. This system is based on an electrical heating element placed in the anode inlet header. During start-up, the stack is fed with nitrogen, which is circulated by the anode recycle blower. The heater is of a radiant tube type, which is integrated in the header box. The heater has an estimated capacity of about 120kW, resulting in a start-up time of about 24 hours.
6. Flow control. The general control philosophy has been verified by dynamic simulations. The most critical issues are the anode flow control and the burner operation. The stacks are identical and have to be fed with equal fuel flows. A proper distribution is obtained by means of trim valves in series with the stack 1 and 2 recycle cells. Flow adjustment is achieved by balancing the valves in such a way that the stack exhaust temperatures are equal. Temperatures are measured by sensors in the recycle outlet header of each stack.

For the exhaust cells, the control approach is very similar. Here three trim valves are required

7. Burner. For good combustion and flame stability in the burner, the fuel/air ratio must be kept within certain limits. Common burners use 5 - 20% excess air. The control system responds slowly to load changes, since the temperature sensors have long response times. Moreover, the primary airflow is mainly balanced by the stack inlet temperature and cannot be accurately controlled to ensure a correct primary airflow to the burner. Therefor, the fuel-air ratio is be determined by O_2-sensor at the burner outlet. The primary air flow will be controlled by a fast responding valve
8. Stack module ventilation system: Due to leakage of a small portion of the process gases in the MCFC stack into the environment, an explosive gas mixtures may develop in the space around the stacks if no adequate venting is applied Several methods have been investigated. The chosen solution is based on recycling of cathode off gas through the module. Also a set of safety provisions have been worked out, such as monitoring of gas concentration in the purging space around the stacks and automatic valves in the fuel flow.
9. Mechanical design: The base frame of the system is connected to the bottom part of the container. The roof and sidewalls are placed after assembly of the system. Sound reducing materials are applied to the inside of the container. At both ends, the container is provided with access doors. The stack compression mechanism supplies the holding force to the stacks. During the service life, the height of the stacks will reduce significantly due to creep and sintering of the porous components.
10. Stacks will be assembled and conditioned in factory and delivered with interconnected top and bottom flanges to provide integrity during transport. During assembly the flange interconnections are removed after the compression force is applied. For this reason, the insulation panels at both sides have to be applied afterwards. They are designed as sliding doors. The gap above the side panels is filled with insulation materials

The balance of plant consists of long-life components, which are not subjected to regular major overhauls. They remain on the site. The main components of the balance of plant are discussed below.

11. The fuel processor is a gas purifier, which absorbs sulphur-containing components in the natural gas. Two processes are considered suitable: 1/ activated carbon operates at room temperature but requires a relatively large volume (about $2.5m^3$ for 1 year of operation), whereas 2/ Puraspec, operating at elevated temperatures (200-400 C), requires a smaller volume than activated carbon (about $1m^3$ for 4-10 years of operation). Additional testing is required to verify the lifetime and economics of the purifying agents. It has no advantage to integrate the fuel processor in the stack module
12. The DC-AC converter converts the (250 V) DC power from the stacks into (400 V) AC grid voltage. Several manufacturers have been contacted and suitable off-the-shelf equipment is available. In order to reduce costs and increase efficiency, the primary voltage must be as high as possible. This urges to electrically connect the stacks in series
13. The system controller may be a PC or microprocessor control unit. Usually it will be located in a control room. The controller itself will be a standard component
14. The heat recovery system extracts the heat of the cathode off gas to produce steam, hot water of cold, depending on the customers’ requirements. Standard technology will be applied.
15. Maintenance schedules and instructions are established for the system components, such as the hot blower, the header box, burner, sensors and start-up heater. High

reliability/availability and short down times for overhaul are achieved by completely exchanging the stack module at the customer's site and overhaul in the manufacturing workshop.

The cost of the system has been assessed, based on the preliminary design and assumptions regarding the production volumes, which are set to 50 systems per year and a stack production volume of 200 MW per year. A comparison between of the cost of electricity, calculated for the MCFC system and competing technologies shows that - within the present boundary conditions - the MCFC system cannot compete with the gas engine in CHP applications for hot water and low pressure steam. For CHP systems producing high-pressure steam, the MCFC system has a slight advantage over a gas engine system. This is due to the fact that the heat of the cooling water of the gas engine cannot be utilised. For power applications, the MCFC system can generate electricity 25% cheaper than a gas engine, but in this case, the electricity price from the grid (central power) is significantly lower

Work Package 6: Stack technology development

This work package has been carried out by ECN and contains nine tasks.

6.10 Co-ordinating and reporting

This work-package controls the stack development as carried out in the work-packages 6 to 10 and provides the interaction with the system development in the work-packages 1 to 5, in particular through work-package 4 Interface Management.. On a weekly base the WP manager discussed the progress with the managers of the WP's 4, 7, 8 and 10 and frequent meetings took place with the system developers.

6.20 Development of strategy & guidelines

The Cost of Energy (COE) of an MCFC power plant is mainly determined by the Cost of Investment (COI). A COI model for the stack module has been is developed. This model is used for system selection and parameter studies. Overall system and stack optimisations were performed. The current density and the module capacity have the largest influence on the investment costs per kWe.

6.30 Reference design and development specifications

Specifications for the full size (1m² active area, 120 cells) stack have been developed for the 3BG5 (reference) system. This system consists of three identical stacks of which the fuel flows are connected in parallel and oxidant flows in series. 3-D stack model calculations have been performed with the current density, system size and gas conditions as specified in the system interface document. Calculations showed that the maximum allowed temperature in the stack has to be exceeded in order to obtain the required power output of 440 kWe, due to flow variations over the height of the stack. For this reason the SMARTER™ system was selected. A critical item in the design is the margin for carbon deposition. Specifications for the stack components were derived and formed the base of activities of WP 7-10.

6.40 Inventory & selection of options for improved thermal-hydraulic management

The SMARTER™ concept enables a better temperature distribution within the stack. The increased recycling rate of the fuel flow lowers the fuel utilisation and has a positive effect on the cell performance of the recycle cells and increases the Nernst voltage. This allows a higher power density compared to the 3BG5 system. Also, the outlet temperature is lowered, which lowers the electrolyte evaporation and extends the stack lifetime of the stack. A further reduction of the temperature dip can be obtained by grading of the catalyst. Comparisons were made between experimental stack temperature distribution data and model calculations to verify the SMARTER™ concept. As a result of the comparisons, the methane reform rate equation was improved, based on out of cell data generated under WP 9.
Flow distribution tests were specified to verify a homogeneous flow distribution in the stack.

6.50 Advanced design & development specifications

The SMARTER™ stack design is based on the system interface conditions, a DIRSEP™ separator plate with pressure drops and manifold channel configurations optimised for the required power output, and state of the art porous components using $(Li/K/Na)_2CO_3$ as the electrolyte.
Calculations with the 3-D MCFC SMARTER™ stack model on this stack design show that the predicted conditions of the stacks in the system stay within the interface boundaries specified for the stacks at full-load (nom BOL), part-load (30% and 40 % BOL) and end-of-life load (EOL). These interface boundaries concern gas composition, minimum and maximum temperature in the stack and performance.

6.60 Specifications for stack tests

Specifications are determined for the stack verification test program. Special attention is given to the 20 cell stack tests for verification of the 3-D MCFC SMARTER™ stack model. The manifold channel distribution, the manifold diameter and the location of the thermocouples are addressed. The gas composition and flow at the stack, as provided by the system design, are translated to experimental conditions for the stack tests. Due to the endothermic water-gas shift reaction ($H_2 + CO_2 \leftrightarrow H_2O + CO$) , and the high fuel flows at SMARTER™ conditions, the stack test installation is extended with a CO supply in order to discriminate between cooling provided by the reform reaction at the inlet, and the cooling provided by the water-gas shift reaction.

6.70 System support and additional interface verifications

This activity aims at contributing to the task force for system configuration selection, co-ordinated by work-package 3. Subjects that are covered comprise the stack interface boundaries, capital cost, performance and lifetime for the different configurations. Specifications have been established for system operation modes, including start-up, full and part load operation, hot stand-by and shutdown. These specifications include stack flow and temperature control and boundaries for carbon deposition. Also much attention has been paid to stack degradation control strategies. Input is provided for the dynamic system modelling.

Dedicated single cell testing has been carried out to determine or verify essential parameters, such as the local resistance expression of ternary $(Li_{0.6}/Na_{0.2}/K_{0.2})_2CO_3$ electrolyte mixtures as a function of the gas compositions and of temperature. Carbon deposition and degradation behaviour has been examined in bench scale test on desulphurised natural gas. The test also showed that higher hydrocarbons are reformed for more than 99%. Endurance bench scale testing during 2,500 hours under low CO_2 and O_2 concentrations in the oxidant flow showed no degradation, and therefor proved that the specified minimum values are safe. In a similar way, the minimum value for the H_2/H_2O ratio in the fuel flow has been determined.

A 41-cells hydraulic test has been executed for determination and verification of hydraulic parameters used in the 3-D stack model. This test also showed that the rule of thumb calculation for the noise produced by the gas flow in the manifold channels seems applicable. This rule of thumb predicts a 95 dB noise level for the full size stack. The thermally isolated 20 cell stack test T7 is practically silent (< 50 dB), although the rule of thumb predicted 70 dB in the 4-16 kHz frequency range. The difference can possibly be explained by the thermal insulation which is very effectively absorbs sounds within this frequency range.

6.80 Technology development evaluation

Several major decisions in the stack development are taken. For the cathode in situ oxidised NiO is selected in stead of $LiCoO_2$. The lifetime of NiO is well beyond the target of 25.000 hrs due to favourable conditions. NiO cathodes in sub-scale stack tests perform better than $LiCoO_2$ cathodes. The latter stack tests were carried out by ECN in previous projects.

The $(Li_{0.6}/Na_{0.2}/K_{0.2})_2CO_3$, ternary mixture is chosen as electrolyte. The earlier chosen $(Li_{0.6}/Na_{0.2})_2CO_3$ mixture showed unstable behaviour at low temperatures and SMARTER™ stack 1 oxidant gas conditions.

The chosen separator plate design is an adapted version of the ER FLEXSEP™ separator plate, as developed earlier by ECN. A Failure Mode Effect and Criticality Analysis (FMECA) has been conducted in co-operation with Stork Product Engineering to assure the gas tightness. It was concluded that the selected set of improvements would lead to the targeted gas tightness. The stack holding force and the compressibility of components needed further investigation during the different phases of start-up and operation.

The milestone M2 'Verification stack test high pressure seals' is reached in the stack test T5. The targeted gas tightness implied a gas loss < 1% of nominal flow for fuel and oxidant to the surrounding at a pressure difference of 300 mbar for fuel and 200 mbar for oxidant. The targeted cross-over gas loss is < 1% of the nominal fuel flow at a pressure difference between fuel and oxidant of 200 mbar.

The milestones M4:'2nd generation porous component verification stack test' and the milestone M5:'2 kWe demonstration 2nd generation DIR stack, including improved temperature gradients' are achieved after the project contractual end date.

6.90 Cost estimate full-size advanced DIR-MCFC design

The Cost of Investment (COI) for the SMARTER™ stacks are calculated using the COI model developed in task 1. A value of 1023 Euro/kWe$_{ac}$ was obtained and assuming a stack lifetime of 3 years, the stack contributes 0.042 Euro/kWh$_{ac}$ to the Cost of Electricity (COE). The COI of the separator plate can be further reduced to 528 Euro/kWe$_{ac}$ by eliminating some expensive parts. If a stack service life of 5 years could be reached, the contribution to the COE would drop to 0.013 Euro/kWh$_{ac}$.

Work package 7: Separated plate developement

This work package contains 10 tasks and has been led by ECN, with contributions of Stork and Schelde.

7.00. Project co-ordination and reporting

The separator plate development was directed by a task force consisting of the WP manager and experts of Schelde and Stork. The progress of and interfacial aspects between this WP and the other WP's were frequently discussed.

The actually developed separator plate differed significantly from the concept that was foreseen before the actual start of the project, as is explained under task 10. The actual costs for the fabrication of sub-size separator plates and supporting research on gas tightness issues exceeded the budget. These issues were addressed in parallel projects funded by the Netherlands Agency for Energy and the Environment (Novem).

7.10 Development of basic design specifications (T1)

A first step was a trade-off study of the separator plate concept, based on an inventory of concepts in use by other fuel cell developers and of own concepts. Internal manifolding was one of the few prerequisites. Selection criteria comprised gas tightness, reliability, lifetime, costs and scalability. From this procedure the DIRSEP™ was selected.

Next an inventory was made of specific constructional solutions for all critical design issues. The best guess solutions were selected and integrated in the reference design DIRSEP™-v1.

Based on the full-scale (1m²) design, a down scaled (0.1m²) DIRSEP™-v1 was specified, pressing and assembly tools (modular for adaptability) were designed and manufactured, fabrication procedures developed and tested.. These separator plates were tested in stack test T1 as well as in out of cell tests. The requirements for the gas tightness of sub-size stacks were derived from the system-dictated requirements for full-size stacks. The internal gas tightness of T1 was 80%, the external gas tightness was only 2% of the targeted values. Post test inspection showed that the low internal and external gas tightness was due to matrix cracks.

Stack test T4 has been carried out with the same separator plate design. Modifications comprised the stack holding force and the application of foils in certain zones. The external gas tightness of the T4 stack was significantly improved with respect to the T1 stack test but still below the targeted value. The internal gas tightness was even lower than in T1. Post test inspection revealed that, also in this stack, the gas leakage was caused by bridging cracks.

7.20. Adjustment of the T1 separator plate design to $LiCoO_2$ (T4)

Deformation restrictions are considered necessary to adapt the separator plate to $LiCoO_2$ cathodes, since this type of cathodes has a low strength at a certain stage of the start-up procedure. The decision (see WP 8) to use NiO in stead of $LiCoO_2$ made modifications unnecessary..

7.30 Development of specifications for the basic design

Based on the experience with the DIRSEP™-v1.0 design improvements were defined to obtain a better mechanical matching of the separator plate to the matrix in order to prevent matrix cracks. One of the critical areas is the transition zone between the active area and the outer seal. Design solutions tested in out-of-cell and bench cell tests showed good results. However, it must be kept in mind that the stiff bench cell construction is only partly representative for the flexible separator plate.
Therefor, a large effort was put in finite element modelling, which proved to be a valuable tool. The modelling elucidated important properties of the separator plate concept and allowed quantification of parameters, such as the optimal thickness of the metal sheet of which the separator plate is built.
Numerous out-of-cell tests have been carried out to study the shrinkage and the related presence of a holding force turned out to be crucial. However, crack formation can never be completely eliminated and counter measures are required to limit the consequences. The investigations also focused on the ways to control crack formation and to prevent cracks to grow further (crack arrestors).

Gas distribution elements are applied on the anode and the cathode side. The dimensions and orientation are determined by the pressure drops and, for the anode side, by the geometry of the catalyst pellets, whereas on the cathode side the anticipated corrosion resistance and strength plays a role. Hydraulic calculations indicated that the flow distribution for the 20-cells stack tests T7 and T9 would be unacceptably skewed. Therefor it was decided to increase the manifold channel diameter in the DIRSEP™-v2.0 design. Other modifications concerned the seals at the edge and around the manifold channels, the by-pass and manifold springs and the gas distribution elements.

The DIRSEP™-v2.0 separator plate has been tested in stack test T2. The test comprises two separate tests on gas tightness. In the first test T2a a relatively high stack clamping force was applied. In the second test, T2b a lower clamping force was applied.
The targeted external gas tightness was met in both tests. The internal gas tightness of T2a complied with the target that of T2b was 50% below. Post test inspection of the stacks revealed that the seals functioned as anticipated. The tests showed that the design was significantly improved. Nevertheless improvement of the reliability is still necessary.

7.40 Adjustment of the T2 separator plate design to NiO-Li/Na (T6)

The electrolyte choice turned out to have no influence on the separator plate design.

7.50 Adjustment of the T2-separator plate design to catalyst loading (T7)

The DIRSEP™-v2 separator plate has been adapted for the SMARTER™ configuration, which has two separate anode manifold systems. The location of the gas distribution holes over the seven available manifold channels was modified. The DIRSEP™-v2.2 has been tested in stack test T7. The external gas tightness was according to the specs. The internal gas tightness was 25% below the targeted value due to a not proper assembly.

7.60 Development of specs for the basic design, high pressure seals and interiors (T3)

DIRSEP™-v3 has optimised of the springs to guarantee the sealing around the manifolds and edge of the separator plate. To improve the internal gas tightness the transition zone was modified. The separator plate was tested in stack test T3. The external gas tightness met the requirements. The internal gas tightness was very poor. A dramatic failure during stack operation prevented a proper post test inspection, so that the reason for the large crossover could not be determined. The stack test was repeated under code T3b. with separator plates DIRSEP™-v3.1, having a slightly modified transition zone. The external gas tightness of T3b again met the targets, the internal gas tightness was 30 % off.

7.80 Development of specs for the basic design, high pressure seals and interiors (T5)

Because of the results so far, it was decided to adapt the DIRSEP™-v3.1 to the SMARTER™ configuration. This DIRSEP™-v3.2 is the final sub-size design and has been applied in stack test T5, the milestone demonstrating gas tightness in a 3 cells stack Both, the internal and the external gas tightness of T5 met the targets and the milestone was achieved.

During the start-up procedure the stack was erroneously short-circuited. This did not affect the gas tightness result but interfered with the second objective (demonstration of performance) of this test. The stack was cooled down to room temperature, dismantled, the cause for short-circuit removed and started-up again. Even after this full thermal cycle the internal and external gas tightness complied to the requirements.

Because of the cell performance could not be demonstrated in T5, a second identical test T5b, was carried out. Since not enough DIRSEP™-v3.1 separator plates were available, the top and bottom plate were DIRSEP™-v2.1 plates. Again the test showed excellent gas tightness and demonstrated the performance.

7.90 Adjustment of the T3 and T5 separator plate design to improved temperature gradients (T9)

The temperature distribution over the stack can be improved by a non-homogeneous distribution of the catalyst in the cells. The DIRSEP™ v3.3 anode side needed no modification to allow this. All separator plates were manufactured, finished and characterised within the contract period. The stack test has been carried out after the contractual end date of the Advanced DIR-MCFC project.

The full-size DIRSEP™ design has been specified and incorporates all design solutions as verified with the sub-scale DIRSEP™-v3.3 design. Additionally an inventory of consequences and options for the fabrication of the full-size separator plate was made.

Work Package 8: Porous component development

The work package contains 5 tasks and was led by ECN with contributions of Schelde.

8.10 Adaption of anode and matrix for Li/Na-electrolyte

The physical properties of several Li/Na-carbonate compositions have been evaluated with respect to the porous components functional requirements and the system operating conditions. The composition changes during stack service life. Therefor an optimal initial composition was selected with the lowest mean melt temperature over stack life. A manufacturing recipe was developed for sub-scale (0.1 m^2) tapes. Tests showed that the electrolyte and matrix are compatible and good wetting was obtained.
Anodes with different chromium content have been fabricated, and have been tested under system conditions in combination with different types of cathodes.
Standard anodes and Li/Na-carbonate tapes have been produced for stack-test T4

8.20 Adaption of NiO cathode for Li/Na-electrolyte
$LiCoO_2$ was the selected cathode material at the start of this activity. In out-of-cell tests the wetting and filling behaviour of $LiCoO_2$ cathodes with Li/K and Li/Na carbonate has been investigated. The wetting of the cathodes with Li/K is excellent, for Li/Na wetting and filling were poor. Adjustment of the start-up procedure resulted in performances > 900 mV in lab-cell tests and of about 800 mV in bench-cells. The start-up procedure is not directly applicable in stacks. During the longer stack start-up time the cathodes become reduced, leading to unacceptable grain growth and loss of performance, as was observed in stack-tests, carried out under a previous program. Adaptations in the cathode recipe did not lead to significant improvements, nor did pre-assembly conditioning. An adjusted start-up procedure and other measures were tested in two stack-test, which were part of a parallel project at ECN and resulted in high Ohmic losses and cracks in the matrix. Two major problems are identified with $LiCoO_2$ cathodes, namely the stability of the cathode and the interaction of the cathode and the separator plate. The development risk to solve both problems was considered too high and therefore it was decided to focus to NiO cathodes in combination with Li/Na-carbonate. The choice was also made when model predictions of NiO cathodes under system conditions showed that a service life exceeding 25,000 hrs can be achieved.

The distribution of electrolyte over the anode and cathode is one of the performance determining factors. The distribution is governed by the microstructure of the electrodes. The combinations of electrodes were tested in lab- and bench scale tests and the best combination was selected. Also the filling degree of the electrodes strongly influences the performance. A model, validated with experimental data, was developed to support the developments. Based on the model and on experimental data the optimal cathode thickness was determined. Tests on bench scale cell did not show satisfactory results. Therefore, cathodes with standard thickness with a low filling degree were applied in stack test T4 and the performance was sufficient. However, a strong temperature dependence of the performance was observed which turned out to be an intrinsic property of Li/Na-carbonate. Also in single cell tests with Li/Na-carbonate showed a low reproducibility. The addition of potassium to the Li/Na mixture increased the performance and reproducibility and decreased the temperature dependence as was validated in a series of tests.

The electrode filling degree was optimised to meet the performance and the endurance requirements. The recycle cells of stack 3 face the most severe conditions in terms of electrolyte evaporation in the SMARTER™ system. Based on these conditions an optimal ternary Li/Na/K (60/20/20) mixture was selected.

During testing of the selected electrolyte composition and filling degree combination it was found that the binder burnout of electrolyte tape plays a crucial role. Electrolyte recipes with other binders brought no improvement. Adaptations in the start-up procedure showed good results in small-scale tests and the procedure was validated in stack test T5.

8.30 Corossion protection

At the start of this activity the ‘state of the art’ corrosion protection of the anode side of the bipolar plate in the active area is a nickel layer. The estimated lifetime is in the order of 10,000 hours due to the deterioration of the nickel coating by internal oxidation of bipolar plate constituents diffusing into the nickel layer. Other areas of the bipolar plate are protected by an aluminium layer that transforms in a non-conductive $LiALO_2$ layer, having an estimated endurance over 25,000 hours. The challenge of this task is to specify a process for the active area that combines the excellent protection of aluminium with good electrical conductivity.

Two procedures were explored. The first is a high temperature one step process, in which the metal parts are aluminised and heat-treated. This method, however, is not easily applicable to full-size separator plates. The second procedure is a two step process and requires lower temperatures. To demonstrate the feasibility of this process several corner sections of a DIRSEP™ bipolar plate have been manufactured. Also a bench cell test running for more than 3000 hours was carried out to test a flange that was manufactured with the developed procedure. The Ohmic resistance of his cell remained unchanged at a low level and post test analysis showed no sign of corrosion.

8.40 Component fabrication

Under this activity all porous components have been produced for the stack-tests, the lab- and bench-cell tests and out-of-cell tests.

8.50 Evaluation
The development of the porous components has been evaluated regularly during the project. At several occasions decisions were taken to changes the course.

Work Package 9: Reforming catalyst research

BG carried out this work package. The work links closely with the developments in WP6, 7, 8 and 10.

9.10 Comparative out-of-cell tests with Li/K and Li/Na
A series of tests have been carried out to investigate deactivation of BG catalyst in a Li/Na environment. In the first tests, the catalyst was run under the same reforming conditions for periods of 0, 50, 100 and 1000 hours. After each period the full catalyst bed was discharged and analysed in three sections.
The final test involved removing a small amount of catalyst from the top of the bed and replacing the depleted reservoir of carbonate. This test was run for up to 2000 hours and has provided the most severe Li/Na environment for the catalyst and for the first time, a significant fall in the intrinsic activity (as measured using a differential micro-reactor) was observed. In each test there has been a notable increase in nickel crystallite size, and reduction in total surface area with time, but the measured temperature profiles did not show significant loss of effective activity in the reactor. Apart from the last test, the amount of Na taken up by the catalyst was less than the amount of K taken up in tests with an equivalent time on-stream. On the basis of catalyst characterised by such physical measurements so far, it is therefore concluded that deactivation in Li/Na appears to be less than in the equivalent Li/K tests. This will be confirmed by carrying out further severe lifetime tests in Li/K.

The excellent performance of the BG catalyst in the presence of Li/K electrolyte was demonstrated in many tests in previous projects. Under this activity a series of tests have been carried out to investigate deactivation in a Li/Na environment. Each test showed a notable increase in nickel crystallite size, and reduction in total surface area with time, but the measured temperature profiles in the test reactor did not show significant loss of effective activity. The amount of Na taken up by the catalyst was less than the amount of K taken up under comparable conditions and it was concluded that deactivation in Li/Na appears to be less than in the equivalent Li/K tests. The surface areas and activities indicate a lifetime of the catalyst of at least 20,000 hours.

9.30 Literature survey of methods of catalyst fabrication
Loading pellets into the separator plate hardware is labour intensive. Coating the hardware with a thin film of catalyst material may be better in the long term. There are many methods of depositing thin films of catalyst onto metal hardware. Of the liquid media methods, sol-gel processing affords good prospects. The gaseous media processes such as PVD, CVD and flame-assisted deposition and electrostatic-assisted vapour deposition techniques also appear attractive. The various methods of catalyst coating have been compared.

9.40 Theoretical and experimental analysis for predicting catalyst loading
Calculations of the effectiveness factors of fresh and aged BG catalyst and test with thin film

coated stainless steel indicate that a thin loading of catalyst is required in the stack. Further tests have been conducted using the BG micro-reactor to establish the intrinsic reaction rate constant for the BG and for an ICI catalyst. Also a reforming rate equation that applies under DIR-MCFC conditions has been determined on a theoretical base and on experimental data in order to improve the ECN stack model.

9.50 Specification of optimised catalyst

Within this project in most occasions catalyst small pellets have been used that can be manufactured commercially. Spheres or extrudates of the material cannot be made commercially although other shapes are possible, including smaller granules. Having considered all the options for optimised catalyst particle size, it was felt that the present size pellets offer the best prospects in the near term, but in the longer term, better optimisation may be possible for coated material.

9.60 Investigation of pre-reforming

Higher hydrocarbons are removed by pre-reforming of natural gas. This reduces the risk of carbon deposition, which may occur when the gas is heated to temperatures above about 650°C. For external reforming MCFC and also for internal reforming SOFC systems operating above 800°C, some degree of pre-reforming is essential. For DIR MCFC systems this is not the case since pre-heating to such high temperatures is not necessary. This has been confirmed throughout this project since all of the out-of-cell tests carried out by BG until recently used a feed of desulphurised natural gas. In no test any problem of carbon deposition occurred through decomposition of higher hydrocarbons.

9.70 Specification of pre-reformer
Work in the previous task has demonstrated that there is no need for a pre-reformer. BG has earlier designed a suitable pre-reformer for an SOFC system, in the EC supported project JOE-CT95-015.

9.80 Fabrication of catalyst for the test programmes
Several kilograms of catalyst have been prepared for the ECN stack tests as required by the project.

9.90 Characterisation of catalysts discharged from tests
Catalyst discharged from out-of-cell reactor tests and stack tests at ECN have been routinely analysed during the project using well established techniques: elemental chemical analysis, X-ray diffraction, electron microscopy, surface area analysis by nitrogen and hydrogen chemi-sorption and temperature programmed reactions. In addition, a micro-reactor system has been set up to measure the intrinsic activity of fresh and used catalysts. Results obtained with catalyst from the various stack tests were passed to ECN for consideration in Work Package 6.

Work Package 10: Stack test programme 0.1 m2

This work package has been carried out by ECN.

10.1 Set up, control and execute a stack test programme for the development of 2nd generation DIR components and operating conditions
Under this activity the preparation of the detailed work plan, the reporting, the frequent discussions with other work package managers,
and the planning of the 0.1 m² stack test programme were carried out. An overview of the stack tests with the main parameters and the results obtained is presented in the next table.
Table 1: Overview of 0.1 m² stack tests, showing the chronology, the parameters changed, and the results from the stack tests. Stack tests T8 and T9 were specified and prepared, but not executed within the contracting period.

Test Name	Chron	Cells	Time hrs	ER/ DIR	Separator DIRSEP ™	Electrolyte	Filling	Start-up	Perfor-mance	Gastight Internal	Gastight External
T1	1	3	170	DIR	v 1.0	Li/K	30%	1	84%	80%	2%
T4	2	3	1062	DIR	v 1.0	Li/Na	20%	1	100%[1]	50%	5%
T2a	3	2	104	ER	v 2.0	Li/K	36%	1	60%	100%	100%
T2b	4	3	205	ER	v 2.0	Li/K	30%	2	85%	50%	100%
T6	5	3	1324	DIR	v 2.1	Li/Na/K	34%	2	83%	25%[4]	75%[2]
T3	6	3	166	DIR	v 3.0	Li/Na/K	20%	2	94%[3]	1%	100%
T3b	7	3	291	DIR	v 3.1	Li/Na/K	20%	2	96%	30%	100%
T7	8	20	993	DIR	v 2.2	Li/Na/K	20%	2	94%	25%[4]	100%
T5	9	3	168	DIR	v 3.1	Li/Na/K	29%	3	72%	100%	100%
T5b	10	3	177	DIR	v 2.1/3.2	Li/Na/K	29%	3	95%	100%	100%
T8	11	3	3000	DIR	v 3.2	Li/Na/K	29%	3			
T9	12	20	1000	DIR	v 3.3	Li/Na/K	29%	3			
Target		for full-size design							660 mV Sm2/exh	< 1% @ 0.2 bar	< 1% @ 0.3 bar

[1]) performance is not 100% in all possible stack conditions.
[2]) probably 100%, major leakage outside stack.
[3]) performance of 2 cells, the third cell had an assembly error.
[4]) assembly errors in 2 cells; all other cells 100%

Stack tests T8 and T9 were specified and prepared, but not executed within the contract period. The stack tests are reported below in chronological order.

10.20 3-cells stack test T1: Basic design separator plate construction

The aim of this test is to determine the gas tightness of the DIRSEP™-v1.0 separator plate as developed in WP7, in combination with the standard porous components and catalyst.
The internal gas tightness is close to the target value. The leakage of this test was far higher than specified due to matrix cracks that were formed because of deformations of the separator plate. The performance was lower than calculated by the wet-seal leakage, although still increasing at the time the test was shut-off. It was concluded to increase the stack holding force for the next stack test (T4) with the same separator plate.

10.50 3-cells stack test T4: Development of porous components, adaption to $(Li/Na)_2CO_3$

T4 is the first stack test with Li/Na carbonate and adapted components. An initial electrode filling degree of 20% has been applied. The separator plate is the DIRSEP™-v1.0, which was also used in T1. The performance is excellent under SMARTER™ stack 2 and stack 3 conditions, but significant (15%) lower under stack 1 conditions, where diffusion limitations start to play a role.

10.30 stack tests T2a and T2b: Separator plate DIRSEP™-v2, gas tightness

Two tests were carried out to determine the gas tightness of the second version of the DIRSEP™ separator plate. T2a was a 2-cells test with an increased stack holding force to ensure the contact between separator plate and matrix in the seal area. T2b was a 3-cells stack test with a lower holding force. No catalyst was built in and the tests were carried out under ER conditions. The external gas tightness in both tests was excellent and the targeted value was met. The crossover was within specification for the first test and near to specification for the second.
The performance achieved in test T2a was low. A reason is the overfilling of the cathode with electrolyte, due to insufficient removal of the binder removal during start-up. T2b was start-up with a modified procedure, to improve binder removal. Its performance was higher than T2, but below specification, due to high Ohmic losses.

10.70 3-cell stack test T6: Development of porous components

The aim of this stack test is to determine the performance of the porous components at conditions according to the interface specifications. The electrolyte composition is a ternary mixture of $(Li/Na/K)_2CO_3$. The electrode filling degree is chosen such that a stack service life of 25.000 hrs under SMARTER™ conditions can be achieved.
The high filling degree caused a slow oxidation of the cathode and a low

initial but in time increasing performance. At the end of the test 83% of the specified performance was reached. The external gas tightness was close to the specifications, the internal gas tightness was below specified due to an assembly error of one of the cells.

10.40 3-cells stack test T3: DIRSEP™-v3, gas tightness

Separator plate DIRSEP™-v3 was tested in combination with the standard porous components Li/Na/K electrolyte and catalyst. A low electrode filling degree was applied and the modified current collectors have an improved flexibility. Two cells performed well, the third was not performing. Post test visual inspection showed that an anode and a current collector of the non-performing cell had been interchanged during assembly. The internal gas tightness was 25%, the external gas tightness 75% of the targeted value.
It was decided to repeat this test, since the results did not form a sufficient base to start the 20 cells stack test T7.

The test was repeated under coded T3b. All features were equal to test T3, except for the use of standard current collectors. The test showed two excellent performing cells and one performing 80%. Post test inspection showed that the low performance was caused by insufficiently removal of the electrolyte binder. The internal gas tightness was a factor 3 below specs, the external gas tightness was good.

10.80 20 -cells stack test T7: Basic stack design

The test is aimed at the determination of the performance and temperature distribution in a 20 cell SMARTER™ stack and to compare the results with the 3-D stack model. The stack test facility is adapted to handle the two fuel outlet flows (exhaust and recycle), and feed SMARTER™ fuel compositions including CO to the stack. The stack is instrumented with over 70 thermocouples to allow a detailed comparison between the measured and the calculated temperature profiles.
The performance of 14 cells is according to the specs, 5 cells have a 10 % lower performance, and 1 cell has a bad performance. The lower performance was most likely due to overfilling of the cathodes during start-up. The internal gas tightness was according to the specs for 18 out of 20 cells. The external gas tightness is good, some leakage outside the stack was observed.
The temperature profile of the stack showed, as expected, a much smaller temperature dip under SMARTER™ conditions than under 3BG5 conditions. Also the performance is higher under SMARTER™ conditions. The measured performance and temperature profile agree very well with those calculated from the 3-D stack model. This also holds for the stack behaviour under dynamic load changes. The 3-D MCFC SMARTER™ stack model has been experimentally verified and serves as a valuable tool for the stack development.

10.60 3 -cells stack test T5: Verification of gas tightness and catalyst loading

The major objective of this test is verification of DIRSEP™-v3.1 with respect to

internal gas tightness. Additional measures in the transition zone are taken to prevent crossover. The external gas tightness was earlier demonstrated in test T3b. An additional goal is to determine the start-up and performance characteristics for the active components with a estimated service life of 25,000 hrs.
During the start-up, an external short-circuit over the stack occurred. The stack was subsequently cooled down, the short circuit was removed and the stack was restarted.
The gas tightness of the stack, both before and after the thermal cycle, was according to the specifications and milestone M2 has been met.
The performance of the stack is low due to the short-circuiting. Post test analysis showed that the electrolyte was unevenly distributed over the electrodes, the anodes had a very high filling degree, the cathodes were almost unfilled. A clear explanation could not be given. No residual binder was found in the cells, indicating that the start-up procedure functioned as required.

The stack has been repeated under code T5b to determine the performance under optimal start-up conditions. The top and bottom separator plates are of type DIRSEP™-v2.1 because insufficient DIRSEP™-v3.2's were available.
Internal and external gas tightness both complied with the specifications. The cell performances was close (95%) to the target. Post test inspection showed no residual binder. The applied start-up procedure and the electrolyte filling degree have been validated and have been applied in the stack tests T8 and T9.

10.90 3–cells stack test T8: Verification of porous components development
The preparations for this stack test to achieve milestones M4:'2nd generation porous component verification stack test' were within, but the start-up was after the contractual end date. Additional funding for the stack tests comes from Novem. The test has been start-up and performs well. Milestone M4 will be achieved after 3,000 hours of continuous operation.

10.100 20 -cells stack test T9: Demonstration 2nd generation DIR technology
The test stand is slightly modified for continuous application of SMARTER™ conditions. The stack has been assembled within the contractual period, but the start-up took place after the contractual end date. The stack performed well and milestone M5 has been achieved.

Work package 11: Evaluation

This work package was led by BCN, whereas all partners contributed.

11.10 Mid-term assessment
Mid 1997 a mid-term assessment has been carried out. The project partners discussed the results of the project at that stage. Next, a formal MTA discussion was held with the representative of the European Commission.

11.20 End-term assessment

At the end of 1998 an end-term assessment was held involving all project partners. In March of 1999 the formal end-term meeting was held.

5 RESULTS AND CONCLUSIONS

The overall objectives of the project have been met:

1. A specific combined heat and power (CHP) application of a DIR-MCFC in the range of 200 - 2000 kWe and fuelled with natural gas or other methane containing fuels has been selected
2. A preliminary system design (SMARTER™) has been made for this application with optimal reduction of the system complexity and investment costs
3. A second generation DIR-MCFC stack has been developed, based on the system requirements, with an innovative approach on life-time extension and with highly efficient thermal, hydraulic and electrolyte management.

The milestones M4: '2nd generation porous component verification stack test' and M5:_'Demonstration 2nd generation DIR technology' are achieved after the contractual end date.

A more detailed summary of the results and conclusions is presented below.

1/ The commercial sector, especially hospitals, hotels and leisure centres are an attractive initial markets for DIR-MCFC systems were. Also there exist opportunities in the light process industry. The optimal system size was determined to be around 400 kWe with delivery of steam, hot water and/or cold.
2/. Competing technologies are the grid, combined with boiler systems, gas engines and gas turbines.
3/ Liberalisation of the energy market gives large uncertainties for the near future. A consequence of the increased competition in power generation is lower market prices for electricity and less willingness of the energy sector to invest in new technologies.
4/ A highly innovative system and stack concept (coded the SMARTER™ system) combines maximum simplicity, a high reliability and availability, and a wide operational window in order to obtain the lowest cost of energy (electric and thermal).
5/ Dynamic analysis of the SMARTER™ system showed a remarkable stability and good controllability.
6/ A full scale, system dedicated, internal manifolded, co-flow Direct Internal Reforming (DIR) stack with a predicted service life of 25,000 hours has been designed, making use of advanced design tools and models.
7/ The solutions for the critical stack issues have been verified in a large number of lab and bench cell tests, in 12 stack tests on 0.1 m² scale and in various dedicated out of cell tests. In the final 20 cells stack test, the integration of all solutions was demonstrated successfully under representative (system) conditions.
8/ The DIRSEP™ separator plate, the active components, including the catalyst and metal part corrosion protection form a solid technological base. Endurance models and post test analysis show that a service life of 25,000 hours can be achieved.
9/ A detailed cost analysis, based on a production volume of 50 systems per year and a stack production volume of 200 MW/year, resulted in a total system cost level of 3000 EURO/kWe. This is considered more than twice the level for competing technologies.
10/ Technical breakthroughs, significant changes in emission regulations, or strongly increased fuel prices are prerequisites for further commercialisation of the technology.

6 EXPLOITATION PLANS AND ANTICIPATED BENEFITS

6.1 Functional analysis of the project

A 400-kWe DIR-MCFC system module has been developed. Such a system can be applied for on-site co-generation, supplying power (AC/DC), heat (warm water, steam) and cold at high efficiencies and low environmental impact. Operation can be either grid connected or in island mode. The primary focus is on natural gas of European quality as feedstock but also other methane containing feedstock could be applied, such as bio-gas. Larger system sizes are obtained by combining more modules. The market sector may be either commercial (hospitals, hotels, etc.), or light process industry (food, manufacturing etc.), or agricultural (green houses).

6.2 Summary of exploitable results

No	Title of exploitable result	Partners (result owners) involved	Exploitation intention
1	DIRSEP™ stack design	BCN/ECN	Building a Proof of Concept installation as intermediate step before Demonstrations/Prototype Systems
2	Reforming Catalyst coated on Stainless Steel substrate	BG-Technology	Continue to develop and optimise coated catalyst in collaboration with potential user(s).
3	SMARTER™ system design	BG-Technology	Building a Proof of Concept installation as intermediate step before Demonstrations/Prototype Systems. Alternatively: applications in other fuel cell technologies to be developed by BG-Technology
4	Header Box compact system design	BCN/Stork	Building a Proof of Concept installation as intermediate step before Demonstrations/Prototype Systems

6.3 Short description of the results

6.3.1. DIRSEP™ stack design

The DIRSEP™ stack design is a set of specifications for a pre commercial, internal manifolding, full size MCFC stack with internal reforming. The stack is designed to function under internal and external pressure differences up to 0.3 bar, enabling its operation within a simple (without expensive pressure control equipment), low cost system. Although optimised to the SMARTER™ system design, for improved temperature profile and electrical performance of the stack, the stack design can be adapted to other internal manifolding MCFC systems (IR or ER). The developed stack has a lifetime over 25,000 hours. The costs for the full size stack were calculated at 1030 EUR/kW_{AC} at a production rate of 200 MW_{AC}/year.

Further cost reduction to 530 EUR/kW_{AC} is feasible by a limited number of rather straightforward developments.

6.3.2. Reforming Catalyst coated on Stainless Steel substrate

Conventional steam reforming catalyst is manufactured industrially in the form of particles or pellets. They cannot be used in the DIR-MCFC because of insufficient activity at the operating cell temperature and high susceptibility to poisoning by alkali, which originates from the molten carbonate electrolyte. BG has developed a particular catalyst material, which overcomes the problems associated with alkali poisoning, and has demonstrated long lifetimes in DIR-MCFC cells and stacks. Most recently, BG has investigated methods of coating such catalyst, in the form of a thin film, onto MCFC stack hardware. This will have the advantage of ease of manufacture for the MCFC developer, when compared with the use of conventional pelleted catalyst material. Several methods of catalyst deposition are currently being evaluated. A further advantage of using coated catalyst is that of cost reduction, both in the method of catalyst deposition and in the reduced amount of catalytic material that is needed for an MCFC stack.

6.3.3. SMARTER™ system design

The SMARTER™ stack concept is a generic design of a high temperature fuel cell stack that incorporates internal reforming. Although devised during the Advanced DIR-MCFC project, the concept could be applied equally to solid oxide as well as internal reforming molten carbonate fuel cell stacks. The stack concept contains two different types of cells. The anode off gas of one type of cells is recycled to the stack inlet, while the anode off gas of the other type is fed to the anode combustion. The SMARTER™ stack design can be applied to both co-flow and cross-flow geometry’s and gives high electrical efficiencies and improved stack temperature distributions.

The SMARTER™ stack has been tested on sub-scale (0.1 m^2) within the Advanced DIR-MCFC development project. Tests have shown that the operating regime for the stack is less demanding than conventional stacks and offers good prospects for longer lifetimes, since less thermal strain is placed on the stack materials. Calculations suggest that electrical efficiency may be improved by 5 percentage points compared with more conventional designs of fuel cell stack.

6.3.4. Header Box compact system design

The Header Box compact system design is a result of the combination of the following functions:

- Conduction of gases to the stacks and system components
- Mechanical support of the stacks
- Provisions for measuring equipment and valves
- Mixing of fuel in the anode section and air in the cathode section

The combination of these functions has led to a design in the shape of a flat box in the bottom of a container that encapsulates the system. Walls are implemented to separate the gases, and conduct them to / from the stacks, the hot blower, the burner and the air compressor

6.4 Protection of the results

Patent Title	By	Status	File nr.	File date
Method for the realisation of tolerance for external pressure differences	BCN/ECN	Appl.	1009061 (NL/PCT)	
Method for the realisation of tolerance for internal pressure differences	BCN/ECN	Appl.	1009062 (NL/PCT)	
An electric power generation system	BG	Filed	GB9 9623327.5	08.11.96
An electric power generation system	BG	Filed	GB9 9621540.5	16.11.96
Bepaling van stromingsverdeling (Determination of flow distribution)	ECN	Filed	40354	06.05.96
Werkwijze voor plaatvor-mige componenten (Procedure for plate like components)	ECN	Appl.	OA 1003238	30.05.96
SERIES CONNECTED FUEL CELL SYSTEMS	BG plc / ECN	Appl.	OA 1004513	13-12-96
The design of the header box for gas distribution and gas conduction for fuel cell systems	BCN/Stork	Appl.	1010264 (NL)	98-10-07
AN ELECTRIC POWER GENERATION SYSTEM USING FUEL CELLS	RG Fellows BG	Filed	9721682.4 (GB)	14-10-97
AN ELECTRIC POWER GENERATION SYSTEM	RG Fellows BG	Filed	9723012.3 (EUR)	Nov 97
FUEL CELL AND CATALYST FOR USE THEREIN	BG	Filed	9424886 (GB) 5622790 (US)	Dec 1994 22-04-97
METHOD OF COATING CATALYST	BG	Appl.	(GB)	July 1999

6.5 Exploitation strategy

6.5.1 General

The project results will be exploited by the project partners and fall into category A according to the Joule Technology Implementation Plan Guidelines (dated 05-06-97).
Results are to be kept confidential. BCN and its stakeholders will take decisions on when and how the technology in this project will be exploited. After decisions from BCN, its partners and Dutch stakeholders on the when and how of the exploitation of this project result, complementary partners may be sought. Options are further R&D, License agreements, joint ventures and/or financial support.

6.5.2. Reforming Catalyst coated on Stainless Steel substrate

BG Technology will seek to offer the catalyst technology to developers of MCFC, and other appropriate fuel cells and process. Exploitation will be sought on a non-exclusive basis, which may take the form of licence agreement, financial support of further R&D, or development in the form of a joint venture.

6.5.3. SMARTER™ system design

BG will take decisions on when and how the technology of the SMARTER™ stack concept will be exploited. The rights of the other partners in the Advanced DIR-MFC project will be acknowledged when the concept will be exploited. BG is prepared to discuss licence agreement with third parties. BG is also seeking financial support to further develop the concept and will consider joint venture arrangements.
Activity 1. Critical evaluation of the project results by the Dutch stakeholders, in view of an assessment of the market for CHP in the near and mid term future. (July to October 1999)
Activity 2. Preparation of a follow-on programme (October 1999 to January 2000)

6.5.4. Exploitation activities and timetable:

Activity 1: Critical evaluation of the project results by consortium members and Dutch stakeholders, in view of an assessment of the market for CHP in the near and mid term future (July to October 1999)
Activity 2: Preparation of a follow-on programme (October 1999 to January 2000
Activity 3: Further development of catalyst (July 1999 to January 2000)
Activity 4: Preparation of an industrial programme to exploit the activities of BG Technology within this programme (January 2000 to January 2001).

6.6 Project benefits / Exploitation Potential

6.6.1. DIRSEP™ stack design

Potential application of the result is in the CHP-market in which case electric utilities are the end-users. Application sectors resulted from the market study performed as part of the project. Identified were hospitals, leisure centres, hotels, etc. The typical size of systems for these sectors is between 300 and 400 kWe.
Main innovative features are:
- The SMARTER™ configuration by which an optimisation of electrical performance and temperature profile is acquired. The latter aspect reduces electrolyte evaporation rate and hence improves the lifetime of the stack.
- The tolerance for pressure differences, by which pressure regulation control of the system can be greatly simplified and thereby the system costs reduced.

Major barrier for the application of the result is the costs of the MCFC-system, which are not competitive with conventional CHP-systems, like gas engines and turbines.

6.6.2. Reforming Catalyst coated on Stainless Steel substrate

The work on catalyst development has identified materials that have good long-term prospects in the aggressive environment of the MCFC. The catalyst may be used in other applications (e.g. medium temperature SOFC) which do not have such a demanding operating regime. Development of coated catalyst material is at an early stage but the long-term prospects are good. It is envisaged that over the period to March 2000, further optimisation can be carried out to firm up the critical success factors. BG is also applying catalyst-coating technology in the development of a Compact Reformer concept, which will be applicable to many markets that require hydrogen or syngas, including fuel cell plants. Fuel Cell developers have shown keen interest in the internal reforming catalyst devised by BG Technology. It is sought after for a wide range of market applications, where the internal reforming stack technology is preferred. This is not restricted to DIR-MCFC, but may be applied, for example to the SOFC stack. BG Technology is also seeking to exploit its knowledge of catalysts in other related areas, such as pre-reforming for SOFC systems and in fuel processing for solid polymer or PEM fuel cell systems.

6.6.3. SMARTER™ system design

The SMARTER™ stack concept is applicable to developers of SOFC and MCFC, where some internal reforming is carried out within the stack. The concept is applicable to all scales of stack although >100kW scale is envisaged as the most practicable. The SMARTER™ stack concept offers better efficiency (+5% of MCFC and +10% for SOFC) than conventional stack designs. It also has advantages in terms of temperature gradients, and therefore can be expected to be beneficial in terms of stack lifetime. The potential barrier to commercialisation is the need for anode recycle, however, recycle blowers have been identified, that are available for this duty.