EVALUATION AND SCALE-UP OF INTERMEDIATE TEMPERATURE (700°C) SOLID OXIDE FUEL CELL TECHNOLOGY

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2. TITLE: VALUATION AND SCALE-UP OF INTERMEDIATE TEMPERATURE (700°C) SOLID OXIDE FUEL CELL TECHNOLOGY

3. ABSTRACT

At the outset of the project there were no suitable commercial supplies of gadolinia-doped ceria (CGO) electrolyte and strontium doped lanthanum cobalt ferrate (LSCF) cathode powders. Such powders which were available were of low performance and only available in sub kilogramme quantities. Rhodia have been successful in developing a CGO powder which when sintered gave a ceramic with high ionic and low electronic conductivity. The processing of the powder was subsequently scaled-up to multi-kilogramme batch size, and supplies of powder transferred to ECN and CERAM as required. In a similar way, LSCF powder has been developed and its manufacture scaled-up to multi-kilogramme batch size.

The development of PEN (Positive-Electrolyte-Negative) structures has seen the scale-up of both tape-casting and viscous plastic processing (VPP) from 5 x 5 cm and 3.5 cm diameter components to 10 x 10 and 12.5 cm diameter. Close control over processing and firing schedules has enabled the enhancement of performance and long-term stability of PEN's with ECN cells in particular showing low degradation over long periods of operation.

GASTEC have assessed a range of catalyst materials for their efficiency and effectiveness in operation at intermediate temperature with both methane and natural gas. The best of these was manufactured by Rhodia and used in a redesigned stack reformer during the demonstrator trial.

One of the prime reasons for investigating intermediate temperature systems was the potential for use of cheaper ferritic stainless steels as system components particularly as interconnects. Although ferritic alloys had the potential for this application, at the start of the project no data was available on their performance in fuel and oxidant streams or on the effects of alloy composition. Similarly contacts and sealing systems had been developed for high temperature systems and no data was available on their suitability for ITSOFC. Two closely related alloys based on 17% Cr ferritic stainless steel with stabilising additives were shown to be stable during long-term corrosion tests at both Siemens and Sulzer. The corrosion of selected alloys with ceramic coatings was shown to be palliative and result in high conductivity interfaces. Characterisation of tested alloys showed chemical changes consistent with the behaviour observed.

The demonstrator test was carried-out in a Sulzer HEXIS system using the specially designed reformer containing the developed catalyst, interconnect plates incorporating design modifications for intermediate temperature operation, and ECN optimised PEN's.

This 3-year development and evaluation of materials and fabrication processes for ITSOFC has resulted in a successful demonstration of the components developed. A 120 mm 5-cell stack was operated over 2000 hours at high fuel utilisation using steam reformed CH_4 at temperatures between 630° to 675°C. Cost effective materials were largely used resulting in a 45% reduction of costs compared to state of the art SOFC stacks. The demonstration of a large stack was, however, only partially successful due to the inherent thermomechanical weakness of the key component, the CGO electrolyte.

4. PARTNERSHIP

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5. OBJECTIVES

The principal objective of this project was the fabrication and evaluation of intermediate temperature (700 C) planar Solid Oxide Fuel Cell (SOFC) stacks developing at least 1 kW electrical power using natural gas. Self-supported ceramic electrochemical cells incorporating $Ce_{0.9}Gd_{0.1}O_{1.95}$ based electrolytes (130 m thick) were scaled-up from approximately 25-100 cm ^{2} area. After appropriate testing these PEN structures were assembled in two different SOFC stack designs already developed by two of the partners, Siemens and Sulzer. Ferritic stainless steel with appropriate functional layers was used as the bi-polar plate material. Following preliminary evaluation of the performance of short stacks a larger stack was developed with the aim of producing at least 1 kW of electrical power at 700°C with power densities in excess of 0.2 Wcm $^{-2}$.

These objectives remained valid throughout the course of the project.

Two of the participants, Siemens and Sulzer intended to use intermediate temperature SOFC stacks in their current, or in modifications of their current SOFC systems. Reduction in the temperature of operation of these systems to 750°C would realise the benefits of being able to use lower cost readily worked alloys as interconnect plates. Lower operating temperatures would also allow cheaper materials of construction to be used in balance of plant.

(In the final stages of the project Siemens made a policy decision to concentrate their future efforts on the Westinghouse tubular SOFC design for large-scale power generation, and to discontinue work on their planar system. Sulzer's exploitation plans, however, remained valid throughout the project.)

The participants of the project envisaged the initial industrial application of the technology to be in the areas of small to medium size power generation, either in isolation or as part of a CHP system. Outside the project, however, ITSOFC systems were being considered for other applications, eg transport. UK government through its Advanced Fuel Cell programme carried-out a review of the applications of ITSOFC during the course of this project. The results of the review were published during 1998.

Estimates for the market for ITSOFC identified replacement of older generating plant, and expansion of power generation as the major segments, with a potential demand for SOFC in these segments, of 500 million ECUs per year.

The main benefits of SOFC from the environmental point of view were identified as reduced fuel consumption and reduced levels of harmful exhaust gases. This may be enhanced in distributed power systems by an increased efficiency when compared to conventional plants. The emissions levels for SOFC plants are less per kWh than conventional plant (for NOx, the emissions may be an order of magnitude lower).

Noise pollution from SOFC systems was also expected to be significantly lower than competing plant. Due to the minimum number of moving parts (restricted to the balance of plant) noise levels may be as low as 55 dB. The absence of the need for smokestacks and cooling towers make SOFC suitable for incorporation into urban sites as low profile sub systems enclosed within buildings.

A beneficial environmental impact provided by the ITSOFC system in comparison with the high temperature system was the reduced dependence on materials containing significant amounts of Chromium. Experimental evidence showed:

a) High temperature systems operating with Ca or Sr doped $LaCrO_3$ ceramic bi-polar plates suffered degradation problems associated with the loss of volatile Cr vapour.

b) Other high temperature systems which operate with metallic interconnects used high Chromium alloys to minimise oxidation problems, these also suffer from progressive Cr volatilisation.

In both cases the volatilised Cr condensed in the cooler parts of the device.

The carcinogenic nature of the hexavalent form of Chromium is well established, and the risk of conversion of the volatilised Cr to that form is significant. This has as yet unknown implications for the repair, reclaim, and disposal of material from used high temperature SOFC systems.

It was felt that ITSOFC operating at 750°C with low Cr ferritic stainless steel interconnects should not suffer from these problems.

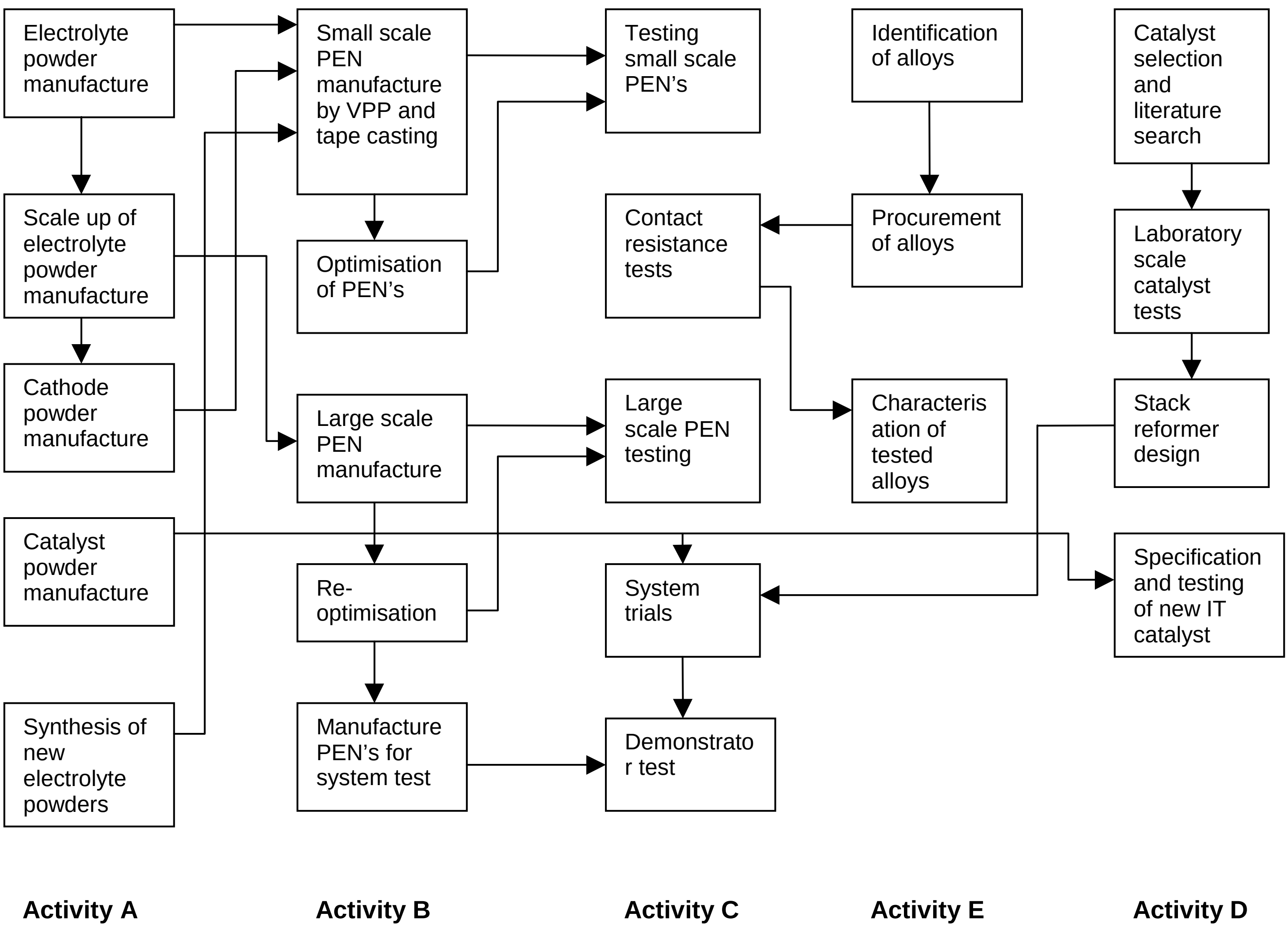
6. TECHNICAL DESCRIPTION

The project comprised five activity areas, each of which was led by a nominated partner. The activity areas were:

	Activity	**Lead partner**
A	Supply of ceramic powders	Rhodia
B	Fabrication of PEN structures	ECN
C	Testing of PEN's	Sulzer
D	Development and evaluation of catalysts	GASTEC
E	Optimisation and quality control procedures for stack components	CERAM Research

Each of the activities contained sub-tasks which were interdependent on each other. The extent of this interdependence is illustrated in Figure 1.

Figure 1



Progress in Activity Areas

Activity A

Activity A comprised:

- Task A.1: Electrolyte Powders
- Task A.2: Cathode Powders
- Task A.3: Reforming Catalysts

- Task A.4: Alternative Electrolyte Compositions

Activity A was carried out by Rhodia.

Electrolyte Powders

At the outset of the project Rhodia had established the ability to manufacture 1 kg batches of $Ce_{0.9}Gd_{0.1}O_{1.95}$ (CGO) powder.

Initial 1 kg batches of CGO powders were shown to have good characteristics with respect to stoichiometry, sintering behaviour and electrical conductivity. This preliminary powder was found to sinter to > 97% at 1400°C and the sintered body exhibited good conductivity at 500°C. This powder was assessed by ECN and CERAM Research for suitability to their processing routes. This initial assessment showed that whilst the powders supplied were suitable for tape-casting at ECN (with some process modification), the surface area was too high for the powder to be used with the VPP process at CERAM. Firing trials were carried-out to reduce the powder surface area and, after confirmation of the powders suitability, a modified powder specification was agreed between Rhodia and CERAM.

Samples were manufactured by uniaxial pressing at a pressure of 1 tonne/cm^2. These were then fired to 1400°C for 2 hours in air. The electrical conductivity (bulk and grain boundary) as measured by AC impedance was found to be lower than that measured using DC techniques.

At temperatures below 350°C the activation energies for AC and DC conduction were found to be similar at about 0.84 eV. Above 350°C and up to 900°C the activation energy was reduced to 0.69 eV. At 700°C the total electrical conductivity was measured as 0.036 S/cm. These values agreed closely with tests carried-out on similarly prepared samples at CERAM.

Scaling-up the processing of CGO powder required the use of larger reactors, filters, and furnaces etc, and this inevitably resulted in a change in powder characteristics. The sintering behaviour of initial large volume batches was noted to be different to earlier small-scale batches, with de-densification (as indicated by an inflexion point in the dilatometry curve) occurring above 1300°C resulting in a reduction in fired density at 1400°C to about 94%.

Cathode Powders

An initial batch of $La_{0.6}Sr_{0.4}Fe_{0.8}Co_{0.2}O_3$ (LSCF) powder was produced and evaluated by ECN and CERAM. The surface area of this batch was measured as 10 m^2/g. The initial evaluation of this powder indicated that its activity was too high to be accommodated by the firing schedules in operation at the time.

Subsequently two further batches of 2 kg each were manufactured with reduced surface area and increased particle size. These powders were evaluated and found to be suitable for both ECN and CERAM processes. Further batches of powder to this specification were manufactured and used throughout the project.

Alternative Reforming Catalysts

Following the Gastec's studies (see task D), Rhodia had prepared different powders based on the composition of 1% Platinum, loaded on Lanthanum-stabilised Alumina for evaluation.

After trials, 2.400 kg of reformer catalyst have been prepared with ball shape and shipped to Sulzer for reforming of 1 kW system. Samples of the final catalyst had been proof tested previously by Gastec prior this milestone stack.

Doped Lanthanum Gallate as an Alternative Electrolyte

At the request of the other partners in the project, the synthesis of a doped lanthanum gallate as an alternative electrolyte to CGO was developed. The composition chosen was $La_{0.9}\ Sr_{0.1}\ Ga_{0.8}\ Mg_{0.2}\ O_{2.85}$ (LSGM). Several conventional chemical processing routes were evaluated and a preliminary 50g batch with acceptable purity was produced.

Characterisation of this preliminary batch indicated average particle size of between 4 and 5 microns. The density of pressed pellets sintered at 1500°C for 6 hours was found to be approximately 96% of theoretical.

Ball milling of the powder reduced the particle size to 1-2 microns but did not improve the sintered density. A minor phase impurity (less than 5% molar) identified as $SrLaGaO_4$ was found in both the powder and the sintered discs.

The structure of the LSGM was found to be tetragonal with the following lattice constants:

- a = 5.524 nm

- c = 7.804 nm

The conductivity obtained was 0.02 S/cm at 700°C (DC measurement) with no grain boundary contribution detected below 300°C.

An observed shift between AC and DC conductivity was attributed to grain boundary contribution.

Activation energy values were calculated from $Ln(\sigma.T) = \int (1/T)$.   Below 350°C AC measurements indicate an activation energy of 1.02 eV, and between 300-700°C the activation energy from DC measurements was 1.11eV.

Activity B

Activity B comprised:

- Task B.1: Manufacture of Small-scale Cells for Initial Evaluation

- Task B.2: Optimisation of Electrode Microstructures

- Task B.3: Scale-up of PEN Structures

Activity B was carried out by ECN and CERAM.

Two routes were used to manufacture small and scaled up PEN structures:

1. Electrolyte manufactured using tape casting and electrodes applied by screen printing – process carried out by ECN.

2. Electrolyte manufactured using viscous plastic processing (see below) and electrodes applied by screen printing – process carried-out by CERAM.

Using powders supplied by Task A, PEN structures or cells were fabricated by tape-casting and screen printing techniques. The initial size of the PEN structures is 5 x 5 cm square for Siemens and 3.5 cm diameter for Sulzer. The size the PEN structures are to be scaled up to is 10 x 10 cm square for Siemens and 12 cm diameter for Sulzer. PEN structures of these dimensions were delivered to Siemens and Sulzer for evaluation within Task C.

Throughout the course of the project ECN cells evolved in four phases from a base cell configuration through three optimised designs to a final design, which was used in the demonstrator trial. CERAM cells evolved through two designs. These evolutionary steps were made in response to changes in powder performance, largely arising as a result of scale-up, and cell performance requirements arising from the test programme in Task C.

The initial aim of Task B was to manufacture small-scale cells using powders provided by Rhodia in Task A and deliver them to Siemens and Sulzer for further evaluation within Task C. The sizes of the electrolytes were 5 x 5 cm square for Siemens and 3.5 cm circular for Sulzer. The start point for ECN was previous knowledge in which PEN structures had been developed using Praxair powders. With these powders ECN had previously developed a PEN production route based on tape-casting. The start point for CERAM was prior experience in the use of VPP with electrolyte powders for HTSOFC.

The performance of the cells incorporating CGO electrolytes had to be increased by optimisation of the microstructure of the electrolyte / electrode / gas three phase boundary and the size of the two phase electrode / gas boundary. The performance target for 700°C operation was defined as 0.25 W cm^{-2} at 0.7 V. The processing steps for PEN production had to be economic and suitable for up-scaling with commercial, large scale production in mind.

Double and single layer screen printed cathodes were evaluated in conjunction with optimised anode composition resulting in the OPT2 cell as follows:

- Electrolyte: Tape cast Rhodia 10GCO powder.

- Cathode: Screen print Rhodia LSFC powder batch.

- Anode: Screen print commercially available anode powder. Electrodes are co-fired.

Further developments aimed at reducing firing temperature resulted in the OPT3 cell, which was used in the demonstrator trial.

Manufacturing routes in order to be able to produce up-scaled PENs were developed at both ECN and CERAM. Comparative testing showed that although the electrolytes from both systems were exhibited similar performance the ECN anode system was superior in its stability. In month 31 the decision was taken to build-up a 1 kW system with ECN cells and the Sulzer stack technology. 70 up-scaled cells were delivered to Sulzer for assembly into the stack.

Activity C

Activity C comprised:

- Task C.1: Evaluation of Small Square PEN and Short Stack Assemblies (SSA)
- Task C.2: Evaluation of Large Square PEN and SSA Units
- Task C.3: Optimisation of Small Circular PEN s and SSA Units
- Task C.4: Optimisation/Evaluation of Large PEN s and Stacks
- Task C.5: Evaluation of 1kW HEXIS System

Tasks C1-C2 were carried out by Siemens.

Tasks C3-C5 were carried out by Sulzer.

For the Siemens system the PEN structures were required to be hermetically sealed into the interconnect separator plate. This had been achieved in the high temperature design by use of a glass seal.

An initial series of sealing studies were carried out at Siemens, to determine the materials and processing required to affect a suitable gas seal to the PEN. Eleven CGO plates were delivered by ECN to Siemens for sealing experiments. The electrolyte was sealed onto stainless steel under load at 850°C in a 90% Ar 10% H2 atmosphere using Schott D263 glass onto 1.4742 stainless steel.

The electrolytes cracked on cooling from the sealing temperature. The crack patterns indicated, however, that the stresses in the electrolytes cooled in air were compressive whereas the cracks, which occurred in those cooled in Ar/H2, were tensile.

From these results it was felt that in the stack the compressive and tensile stresses would cancel each other's effects and the electrolyte should survive.

Initial tests on two 5 x 5 cm PEN’s supplied by ECN in combination with 1.4727 steel interconnect showed a difference in performance for different sealing geometries. In one case (PEN 62-1) the PEN was sealed to the interconnect on the anode side only. In the other (PEN 58-2), both sides were sealed to the steel. The IV characteristics after a reduction procedure in dry hydrogen and oxygen showed a current density of about 0.54 A/cm^2 at 0.4 V.

Operating under galvanostatic conditions with dry hydrogen and air, a degradation rate of 10% in 1000 hours was observed with PEN 58-2 whereas under the same conditions PEN 62-1 showed a degradation rate of 4% in the same time.

Testing of 35-mm button cells at Sulzer commenced with testing in an open cell housing using alloy 446 current collectors with 1 x 1 pin structures.

With cells from batches 1 and 2, high power densities up to 0.4 W/cm2 were obtained within the first 5 hours of the electrochemical test at 715°C. All cells showed degradation induced either by cell cracking or caused by electrode degradation predominantly on the anode side. Even so, power densities of > 0.15 W/cm2 were obtained after 500 hours of operation.

Ohmic losses due to the current collectors remained low during the 500 hours tests and compared well with resistivities determined in contact coating tests (Activity E).

These baseline tests on 35-mm button cells were instrumental in identifying the critical features in the development of PEN's towards scale-up. Items identified by the small-scale tests at Sulzer included degradation of performance and ohmic losses in the electrodes and at the contact.

Testing of Scaled-up Cells

Eight large area cells (100 x 100 mm) made by ECN and CERAM Research were tested in short stacks applying metallic housings, glass seals, ceramic spacers and $LaCoO_3$ cathode contact layers. Both cell types showed good initial performance at 675°C operating temperature (CR-cells: OCV = 0.88V – 0.90V, mean cell voltage = 0.59V at 350mA/cm², ECN-cells: OCV = 0.90V – 0.93V, mean cell voltage = 0.65V at 350mA/cm²).

Compared to single cell measurements in ceramic housings at ECN the short stacks with ECN cells showed equal performance levels. These results confirmed that the specified project target (200 mA/cm² at 0.7 V) could be achieved.

After measuring V/I characteristics, long term tests with constant load were performed up to 1300 hours. The CERAM cells degraded very rapidly. It was assumed that adherence problems of the anode caused this degradation. ECN cells showed a better long-term stability. Nevertheless, observed degrading rates of 4%/1000h were still considered too high for a technical application.

Post Test Analysis

After the long-term tests the short stacks were cooled down to room temperature applying shield gas (Ar) on the anode side. During removing the weight on top of the stacks the cell areas opened slightly. The glass sealing was cracked inside the glass matrix. The stack parts were disassembled and documented by photos.

Tests with ECN cells:

- In two cases (LZ98/07 layer1,2) the anode adhered partly on the Ni meshes.

- In three cases (LZ98/07, layer 1 and LZ98/08 both layers) the colour of the anodes and Ni meshes was totally grey. This indicated that no severe cell cracking occurred during stack operation.

- Layer 2 of LZ98/07 had leakage problems.

- The adherence of the cathode contact layer was poor.

Tests with CERAM cells:

- Adherence of anodes was a severe problem.

- Anodes and Ni-meshes show large green areas, which indicates leakage problems (mainly cell cracking) during stack operation.

- In the stack LZ98/09 spacers of MgO were used for sealing purpose, which should fit well to the ferritic steel interconnectors in respect of thermal expansion. Nevertheless, all of these spacers were damaged. It was felt that this was a problem of shaping the spacers by laser cutting, which probably induced micro-cracks in the ceramic.

The interconnector plates withstood the tests very well. They could be refurbished easily for further use.

Work at Sulzer included a trial to determine the influence of current collector design on stack performance. Stacks were operated at 650°, fuel and air-massflows were varied and the corresponding OCV measured. The results indicated that a new design of interconnect was required for ITSOFC stacks, and this was manufactured for the demonstrator trial.

The influence of operating temperature and fuel utilisation on the performance and stability of the stack was also investigated. The results of the parametric study were influenced by the cracking of the PEN elements in some of the stacks, complicating the true comparison of data. However, the following trends were observed:

- Influence of temperature on performance was small (as observed in ø35 mm cells).

- Degradation was faster at higher temperatures and higher current densities.

It was, however, unclear how far the behaviour of the performance depended on the start up procedure of the stack.

The performance of the stack may be summarised as follows:

- ASR (2.0g/h H_2): 1.0 to 0.75 Ωcm^2.
- OCV: 0.91 V to 0.85 V.
- Operation: 630°C to 680°C without cracking.
- Performance: 22 W with 34% el Eff at 75% FU.
- Degradation: High at high current.
- Cracking probability: Low.

A 5-cell stack using ECN-CGO-PEN technology for system qualification was operated with CH4. The stack was prepared with standard stack assembly procedures using OPT3 CGO-cells. The thermally integrated steam reformer was prepared according to an optimised configuration using 1% Pt-catalyst manufactured in the 2 kg-batch quantity by RHODIA. Samples of the catalyst had been proof tested previously by GASTEC prior to the milestone stack.

The 5-cell stack was operated for more than 2000 hours despite some degradation. The stack showed the expected performance at a given current of 20 Amps

On the basis of these and other results the decision was taken to use ECN cells in the demonstrator trial.

Demonstration Trial

A 62 cell stack was prepared using redesigned interconnect plates and nickel mesh current collection on the anode side. After installation of the reformer with a volume filling of 1% Pt catalyst, the fixation of the system stack and the wiring of the diagnostics connections the stack was started up. Control of the start-up procedure used algorithms derived for HTSOFC which were unable to accommodate the temperature excursions, which occurred in the stack when the fuel supply was switched on. The resulting thermal stress caused some of the cells to crack resulting in a reduced performance. Given these conditions, however, the performance of the demonstrator agreed with that of the five-cell stack.

Activity D

Activity D comprised:

- Task D.1: Evaluation of Novel Reforming Catalysts
- Task D.2: Scale-up and In-situ Testing of Novel Reforming Catalyst

Activity D was carried out by GASTEC and Sulzer.

A literature review was carried-out which indicated that the bulk of industrial reforming processing took place at high temperature (900°C) and at pressure up to 30 bar. Most processes employed a nickel catalyst carried-on a MgO, Al_2O_3, or magnesium aluminate spinel substrate. Sometimes an alkali additive was used to limit the degree of carbon formation. Steam/methane ratios used varied from 3-3.5.

The requirements of intermediate temperature reforming, however, were:

- Operating temperature: 650 – 700°C
- Pressure: 1 – 3 bars
- Steam/methane: 2.5

From the results of the literature review a test programme focussing on commercially available Ni catalyst, Pt, and Rh was defined. The programme included tests of, activity at fixed temperature, sulphur poisoning, carbon deposition, and characterisation.

The catalyst materials included in the examination were:

- Nickel catalyst (Ni).
- Low concentration platinum on alumina (PtL).
- High concentration platinum on alumina (PtH).
- Low concentration platinum on lanthanum stabilised alumina (PtLS)
- Ruthenium on lanthanum stabilised alumina (RuS).

The results of the assessment indicated:

- Only the nickel catalyst was sensitive to deactivation by coke formation.
- The nickel catalyst was also very sensitive to sulphur poisoning.
- The ruthenium catalyst showed severe deactivation by sulphur poisoning as well as time dependent deactivation under normal reforming conditions.
- The platinum catalysts show only marginal deactivation under operating conditions and are most resistant to sulphur deactivation.
- High platinum concentrations have no clear benefit over low concentrations.

Using these results and others a specification was drawn-up for a catalyst which was manufactured by Rhodia.

Reformer performance trials at both Gastec and Sulzer indicated that improvements to the reformer design should yield improved performance. A new design of reformer was developed which gave better mixing of the fuel and water vapour, and due to the use of smaller grain size catalyst, less "slippage" of methane.

Trials were carried-out at Sulzer using the improved reformer design feeding 35 mm diameter cell housing. The reformer was charged with platinum catalyst (1% concentration) on stabilised alumina and a cell operated for 780 hours. The conversion achieved using the 1% Pt catalyst was up to 83% at a S/C ratio of 2.8 and an outlet temperature of 655°C. No cell degradation or deactivation of the catalyst was observed in 780 hours of operation.

Ultimately, in a five cell stack test, a conversion of 92% was achieved at a catalyst temperature of < 650°C and a steam/carbon ratio of 2.3. Decreasing the steam/carbon ratio to 2 in order to improve cell efficiency resulted in a reduction in conversion to 80%. Reducing the catalyst temperature at this point to below 600°C reduced the conversion further to 70%.

Activity E

Activity E comprised:

- Task E.1: Selection and Optimisation of Ferritic Stainless Steel Components

- Task E.2: Development of Quality Control Procedures for Ceramic Components

Activity E was carried out by Avesta, Sulzer, Siemens, and CERAM.

Selection and Optimisation of Ferritic Stainless Steel Components

The requirements of ferritic stainless steel interconnect and the criteria for their selection for examination were established as follows:

Requirements:

- Close match of thermal expansion to that of the PEN structure.

- The oxide film interface resistance to be low and stable.

- Ageing and embrittlement characteristics to give interconnects adequate strength and ductility for the life of the cell/stack.

- Material in the form suitable for machining or forming.

Considering these requirements and the general requirements for high temperature application a number of criteria for selection were established. These included:

- Chromium content high enough for good oxidation resistance.

- Chromium not so high as to promote embrittlement.

- Avoid compositions containing silicon or aluminium additions as these were known to give a high resistance oxide film.

- The presence of carbide stabilising agents to improve oxidation resistance and limit carburising effects in service.

The compositions of a range of commercial stainless steels were collated together with information on their properties, and candidate steels were selected by Avesta, Siemens and Sulzer.

Screening tests were carried-out at Siemens, in which the total conductivity of metal/contact coating/cathode sandwich structures of 2.9 cm^2 area were measured at 700°C for 500 hours, and at Sulzer using a four terminal system and different coatings. Both laboratories identified the alloys 1.4510 and 1.4509 as yielding sufficiently low resistivity in contact with selected coatings to merit longer term testing. Siemens also selected 1.4742.

Long-term contact conductivity tests with the alloys FeCr16Ti, FeCr17NbTi and FeCr18 MoTi were started to collect data for more than 5000 hours at the upper limit of the future stack operating temperature of 725°C. After 4100 hours the electrical resistivity values of all three alloy/ceramic contact samples were still below 6 mΩ, corresponding to a contact area resistivity of

0.018 Ωcm^2. The degradation rates per 1000 hours range from 2.32 $m\Omega cm^2$ for FeCr16Ti to 1.97 $m\Omega cm^2$ for FeCr17NbTi and 0.9 $m\Omega cm^2$ for FeCr18 MoTi. The contact resistivity was expected to stay below 0.1 Ωcm^2 for 40,000 hours assuming a constant degradation rate. This was deemed an acceptable value.

Characterisation of the corrosion scale created during these tests was carried-out using SEM, SNMS, and GDOES techniques. The results obtained showed:

- Titanium was found to diffuse in significant amounts within the oxide scale formed on the 1.4509 steel exposed to the fuel cell atmospheres (inlet and exit gases), and in contact with the contact coating at 700°C for 5,000 hours.

- This observation indicated a behaviour similar to that observed following high temperature oxidation in air and is known to be beneficial to oxidation resistance.

- It was suggested that titanium diffused from fine particles precipitated at the metal/oxide interface into the scale.

- Assuming the oxidation behaviour of the 1.4509 steel to be consistent with high temperature oxidation characteristics, other compositions can be formulated for further improvements.

7. RESULTS AND CONCLUSIONS

The exploitable results arising from the project include the following.

No	Title of Exploitable Result	Partners (Result Owners) Involved	Exploitation Intention
1	Electrochemical machining of interconnector plates.	Siemens	-
2	Low resistance contact layers for intermediate temperature fuel cells.	Siemens, Sulzer	-
3	Identification of standard ferritic stainless steels with useful interface electrical characteristics.	Siemens, Sulzer, Gastec, Avesta, Sheffield	Joint Product Development
4	Definition of a manufacturing route for optimised PEN structures.	ECN	Manufacture
5	Definition of a manufacturing route for electrolyte plates.	CERAM	R & D activities in oxygen generation and electrochemical reactors.
6	An improved reform reactor with a suitable catalyst for the Sulzer HEXIS IT-SOFC.	Gastec: Advising catalyst manufacturer on catalyst recipe. Rhodia: Production of reform catalyst. Sulzer: Production of reformer filled with catalyst Marketing and sales.	Licensing

- The 3-years development and evaluation of materials and fabrication processes has culminated in a successful demonstration of the components developed.

- A › 120 mm 5-cell stack was operated over 2000 hours at high fuel utilisation's using steam reformed CH4 at temperatures between 630° to 675°C.

- The use of cost effective materials resulted in a 45% reduction of costs compared to state of the art SOFC stacks.

- The demonstration of a large stack was only partially successful due to the inherent thermomechanical weakness of the key component, the CGO-electrolyte. Under realistic system environment and operating conditions extended cracking of the cells occurred which led to large temperature gradients in the stack.

Exploitation Plans and Anticipated Benefits

The exploitation intentions of the partners in respect of the result are shown in the above Table.

During the final year of the project Siemens announced its intention to concentrate its future activities on the tubular SOFC design developed by Westinghouse. As a result they would not pursue activities in planar systems beyond their existing commitments. Consequently Siemens will not exploit their results.

The benefits, which have arisen from the project, may be summarised as follows.

Benefit	To Whom
A commercially viable European source of high quality electrolyte, and cathode powders has been established.	European SOFC manufacturers.
Manufacturing processes for SOFC components have been developed and refined.	Component manufacturers, Systems manufacturers.
Low cost steels for interconnect and other components have been identified.	Systems manufacturers.
Rationale for development of improved alloys has been established.	Technology providers.
Catalysts and reformer designs for low temperature reforming have been established.	Systems manufacturers, Fuel processors.
An understanding of the design limitations and performance requirements of ITSOFC has been established.	European Systems manufacturers.

8. ILLUSTRATION

1 kW HEXIS System used in the Demonstrator Trial