SCALE UP OF A MULTI-FUNCTIONAL SOLID OXIDE FUEL CELL TO MULTI-TENS OF KILOWATT LEVEL (MF-SOFC)

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ABSTRACT

The project MF-SOFC, partly funded by the European Commission, is aiming to scale up the Rolls-Royce SOFC technology from kW to tens of kW scale. The consortium brings together the wide range of skills and experience required to achieve this goal from fundamental materials science to end user knowledge. Due to a focus on the requirements of a commercial product, the first generation stacking concept was redesigned to achieve a higher power density. The manufacturing processes were refined and scaled to meet the increase in requirements for components. The cell development activity combines the experience of Rolls-Royce and Risø National Laboratory with the aim of improving the durability of the multi-cell module. Mechanical modelling to evaluate and predict the reliability of the ceramic structures is led by Imperial College London and has been carried out in parallel with the design process. Testing has been carried out in purpose-built test facilities, in France and the UK. Results from both locations are in good agreement and the high power density support module demonstrated performance that would meet the requirements of initial commercial products.

INTRODUCTION

Scale Up of a Multi-Functional Solid Oxide Fuel Cell to Multi-Tens of Kilowatt Level (MF-SOFC) is an European Commission Framework V funded project which aims to develop an SOFC stack with a nominal rating of ~20 kWt. The project is led by Rolls-Royce along with the partners of Risø National Research Laboratory, Imperial College London, Gaz de France and Advanced Ceramics Ltd.

Between 1987 and 1992, Rolls-Royce undertook an evaluation of fuel cell technologies, considering the various types, and concluded that the solid oxide fuel cell was likely to be
the most relevant option for the company's energy businesses. In June 1992, a cautious but active SOFC programme commenced with the aim of learning about the technology, building an understanding of its strengths and weaknesses, enabling the company to make sound decisions on its involvement with the technology. From these initial studies, it was not evident that the state-of-the-art SOFCs provide a suitable technology to meet customer requirements of high power-to-weight ratio, rapid load following, and affordability. An SOFC concept was conceived by Rolls-Royce which aimed to combine the benefits of planar and tubular approaches, with the fundamental fuel cell building blocks or 'modules' combining the best features of both philosophies. To distinguish it from other contemporary types, it was called the integrated planar solid oxide fuel cell (IP-SOFC). In the simplest form, the concept is based on series connected cells fabricated on a fuel carrying porous support tube. A schematic of the cell arrangement is shown in Figure 1.

![Figure 1. Schematic cross section of IP-SOFC concept.](image)

The MF-SOFC project aims to develop the Rolls-Royce IP-SOFC technology from kW level to tens of kW level. The consortium brings a wide range of skills and extensive SOFC experience required to complete this task. To achieve this goal, the multi-cell MF-SOFC module, which is the building block of the system, required scale up in rating and performance in line with commercial targets of durability, reliability and cost. Previous details of the IP-SOFC concept can be found in references (1, 2).

**STACK DESIGN AND BUILD**

In parallel with stack development, Rolls-Royce has completed extensive market and system studies. This work concluded that capital cost targets for the stack and system would be best met by a pressurised hybrid system at 1 MW scale. This work also showed that the most important parameters in reducing system cost ($/kW) is stack volume (kW/litre) and corresponding weight. An evaluation of the first generation multi-channel support module in this context revealed that an improvement in stack volumetric power density was required to meet the cost targets. The support structure was, therefore, re-worked resulting in a revised concept based on tubes with significantly reduced thickness.

A remaining challenge was to develop a stack concept around the cell tubes (modules). The requirement was for a stack structure that provides simple low-cost fuel manifolding,
simple reliable sealing and mechanical compliance to accommodate the temperature variations seen during start-up and shut down.

The selected solution is a 'serpentine bundle'.

Figure 2 shows an early test bundle. With a little care it can be resolved that ten modules are connected in flow-series with respect to their internal flow of fuel. This configuration directly benefits achieving required fuel utilisation by guiding the flow for all ten modules past the anodes of each individual module. By using the widely established technique of anode flow recirculation, additional flow can be achieved and overall utilisation requirements can then be met without being forced to operate the cells at excessive current density. For the purposes of the MF-SOFC project, this recirculation is simulated by feeding appropriate gas mixtures. A separate program of work has confirmed in principle that this recirculation can be achieved by use of a purpose developed pump based on an ejector with advanced flow characteristics. The large flow achievable on the fuel side coupled with the temperature of operation enables the use of a porous support tube without drop in performance.

Another benefit of the serpentine design is its ability to provide overall geometrical compliance, even if built from stiff ceramic materials. This broadens the range of concepts that can be used when manifolding the bundles into larger stack units or “strips.” The strips each produce several kW and the MF-SOFC stack will consist of an array of these strips. Strip level manifolding requires an order of magnitude less components than would otherwise be required to manifold individual tubes if the serpentine bundle design were not used.
This overall MF-SOFC stack enables issues at bundle, strip, and multi-strip level to be investigated and de-risked under realistic conditions without at the same time attempting the harder challenges of operating under pressure with tightly coupled reforming. Lessons learned from MF-SOFC enable a subsequent EC framework V project, PIP-SOFC, to address these additional challenges with basic stack technologies in terms of materials and geometry at least in part established.

MANUFACTURE OF MODULES

The IP-SOFC technology has been specifically designed to be a technology with a low-cost manufacture. Complex gas-phase deposition processes used in other SOFC programs were precluded from the onset for economic reasons. The solution was found in the use of fabrication routes including extrusion, screen-printing, and slurry spraying. These are all currently used in low-cost, high-volume ceramic and electronic industries. This approach to realising a commercial SOFC technology was seen as the key to providing a cost effective technology.

A program of manufacturing development has provided an established, well-controlled manufacturing process definition. The processes have been fully documented and have allowed the fabrication of consistent components with little variation.

The manufacturing technology was initially proven at Rolls-Royce but during this program, the manufacture of the porous support module was transferred to Advanced Ceramics Limited (ACL), a 'small-to-medium enterprise' based in Stafford (UK). This, in combination with existing equipment, has provided manufacturing capacity of support structures and modules in line with the program requirements. ACL has extensive experience of technical ceramic manufacture and, within this project, is responsible for the implementation of technology to prove concept and capability of production scale-up.

Additional equipment (Fig. 3) has been absorbed in line with production requirements for the program. This, in combination with existing equipment, has provided the capability to support the required support structure and module manufacture.

The change to the high power density support tube has significantly increased module output per batch of material. In addition development of the manufacturing process and improvement in quality control has also led to an increase in manufacturing yield.

Continued improvement and iteration of the extrusion process has been carried out in parallel with the need to meet program requirements. Consequently, the increasing volumes manufactured at ACL have further defined the limits of the process by the use of statistical process control (SPC).

Additional screen-printing capability that incorporates a visual alignment system has increased accuracy and consistency during printing. Significant effort has also been applied in the development of procedures and process control leading to an improvement in yield. Refinements in the cell design have incorporated control of alignment and
registration of the multiple layers. This has enabled inline inspection at each stage of the printing process thus improving product quality.

Figure 3. Support structures (foreground), extrusion equipment (background).

Further automation of the process is continuing and important relationships with equipment suppliers have been established. This information and expertise is being used to reduce the operator input and cycle times, ensuring that the process can be scaled-up rapidly and economically to satisfy future requirements.

MODULE PERFORMANCE AND DURABILITY STUDIES

The cell development effort has been carried out in close collaboration with Risø National Laboratories. Risø has many years of experience in SOFC research and development and is widely respected in the field. There is also a long history of collaboration with Rolls-Royce spanning several EC programs, giving Risø an intimate and detailed knowledge of the IP-SOFC concept and the challenges to bring the technology through to reality. Much of the effort on the program has been the optimisation of the various screen-printed layers within the cell array to optimise the interaction between durability and performance.

An important aspect of this work is to minimise the environmental impact of the fabrication processes. It is essential that any vehicle systems used are as environmentally benign as possible while still retaining compatibility with adjacent materials and manufacture processes. A large effort has been spent at Risø researching and developing ceramic vehicles for use in the wet ceramic processes. A major challenge has been integrating these alternative systems into the manufacturing process while maintaining the quality and standard currently achieved.

An investigation was carried out to evaluate the applicability of ceria-NiO anodes in the IP-SOFC concept (3). Modules containing both YSZ and ceria based cermets were studied, and a detailed comparison between the two revealed that the Ni-CGO anode gave
only a slightly improved performance than that based on Ni-YSZ. The Ni-CGO was more active than Ni-YSZ for direct CO oxidation, whereas there was little difference between them when operating on H₂-CO₂ mixtures, where a mixture of H₂ and CO would be present. This suggests that Ni-YSZ anodes should be suitable for operating on reformed hydrocarbon mixtures, provided that they display sufficient durability.

Although a slight improvement in performance was observed for the Ni-CGO anode, an undesirable reaction between ceria and zirconia was observed leading to anode degradation. Therefore the YSZ-NiO was selected for use in the cells in the program as it provided a better durability for minimal drop in performance.

Work has also been carried out to further optimise the cathode microstructure. This work has revolved around manipulation of the composite and current collecting layers. This has resulted in stable structures that have exhibited excellent durability as shown in Figure 4. Similar levels of durability have been demonstrated in module tests carried out within other EC programs currently underway and demonstrate good reproducibility in module behaviour (4). The improvements in the manufacturing protocols have also had a benefit in this regard allowing a stable baseline for future development and scale up.

The standard test vehicles for cells used throughout MF-SOFC has consisted of 7 and 20 cell arrays. In general, the area specific resistance of these modules is around 1 Ωcm². This is higher than exhibited by previous reported short 3-cells stacks (1). This difference is attributed to geometric compromises that are required to balance performance against the ability achieve high yield manufacture. It has been calculated that the performance is sufficient for use in an entry-level 1 MW pressurised system. Figure 5 shows the results of 7-cell and 20-cell modules. It can be seen that these are comparable and illustrate the scalability of the concept.
Extensive studies were carried out at Imperial College London with the objective of assessing and improving the mechanical reliability of the cells and stacks in both manufacture and operation. The work carried out encompasses stress analysis of components using finite element modelling (FEM), experimental measurement of the thermo-mechanical properties of the materials involved, and experimental verification of the modeling predictions (such as thermal shock resistance). In addition, new materials solutions are being developed to transfer current between modules.

The mechanical properties that are being studied include elastic modulus, flexural strength and fracture toughness for the principal materials (such as the support tube material and the functional materials) as a function of temperature. In addition, residual stress measurements are made on cell structures and related to the thermal expansion and mechanical properties of the constituent materials. Fracture mechanics approaches are then applied to assess their mechanical stability under normal operation or fault conditions.

The FEM method is used to compute the stresses induced in the complex modules and bundles resulting from the thermal fields or misalignments in manufacture. The analysis is fully elastic and requires as input the measured values of the elastic moduli of the constituent materials. This analysis has proved invaluable in the verification of the module and stack design under a range of operating conditions. For example, Figure 6 shows the predicted distribution of tensile stress in the support tube near a joint between two tubes that is induced by a total misalignment displacement of 1 mm in a bundle of 10 tubes. The maximum tensile stress is localised and approximately 15 MPa. This is comfortably below the fracture stress of the support tube material measured in the mechanical tests (30-50 MPa, depending on the pore volume fraction).
This work is carried out in parallel to the stack design activity. Utilisation of these tools has provided a direct link to further steer the design process and provide critical input into the final design for modules and bundles.

![Figure 6. Results on stress analysis on the bundle.](image)

**TESTING PROGRAMME**

During the project, work on module and stack testing has been led by Gaz de France (GdF). As one of the world's largest public service utilities, GdF has extensive knowledge of end users' requirements, ensuring commercial imperatives are taken into account.

During the initial stages of the program, parallel testing facilities were set up in Rolls-Royce in Derby (UK) and GdF in Paris (France). The testing facilities in Paris allow detailed experimentation to be carried out (Figure 7). The multi-cell modules can be tested in a computer-controlled system under an extensive range of gas compositions and flow rates. This can emulate operating conditions representative of a final commercial stack. Pressure sensors and comprehensive gas analysis capability allow the performance to be characterised, providing essential data required for stack modelling activities.

The test results obtained in the two facilities showed good correlation for open circuit voltages and when tested under load. Each facility has used modules fabricated in a similar manner and the similar test results provide a high level of confidence in the validity of the testing procedure. The testing programme also showed an excellent reproducibility. This has provided a reproducible baseline upon which the effect of changes in design or manufacturing process can be evaluated.

Typical test results are shown in Figures 8 and 9. Figure 8 shows the effect of fuel flow on performance for the higher power density module design. The dependence on fuel flow is markedly less than experienced with the multi-channel design. The results showed very little performance improvement above a fuel flow rate of 2 Nl/min. Figure 9 shows the effects of H$_2$ dilution with CO. In combination with other dilution studies...
extrapolation of single module results to bundle size indicate that 75% fuel utilisation is achievable under commercial operating conditions.

Figure 7. Test facilities at GdF in Paris.

Figure 8. Effect of fuel flow rate on performance.
Figure 9. Effect of H$_2$ fuel dilution with CO.

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REFERENCES

1. F. J. Gardner, M. J. Day, N. P. Brandon, M. N. Pashley, M. Cassidy, Journal of Power Sources, 86, 122 (2000).

2. M. J. Day, Proceedings of the 4$^{th}$ European Solid Oxide Fuel Cell Forum, Lucerne, I, p.113, (2000).

3. N. Lapena-Rey, N. T. Hart, R. D. Collins, N. P. Brandon, N. Bonanos, Proceedings of the 5$^{th}$ European Solid Oxide Fuel Cell Forum, Lucerne, 2, p. 883 (2002).

4. Component Reliability (CORE) - EU Funded Project – NNES-2000-00148.