

SOFC technology development at Rolls-Royce

F.J. Gardner, M.J. Day, N.P. Brandon^{*}, M.N. Pashley, M. Cassidy

Energy Conversion Group, Rolls-Royce Strategic Research Centre Sinfin A-28, Rolls-Royce, PO Box 31, Derby DE24 8BJ, UK

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Abstract

Fuel cells have the prospect for exploiting fossil fuels more benignly and more efficiently than alternatives. The various types represent quite different technologies, with no clear winner, yet. Nevertheless, the high temperature MCFC and solid oxide fuel cell (SOFC) types seem better suited to power generation in a hydrocarbon fuel economy. Presently, the costs of MCFCs and SOFCs are too high to compete directly with contemporary power generation plant. Seeking to overcome the drawbacks of first generation fuel cells, over the past 7 years an innovative second generation SOFC concept has been evolved in the Rolls-Royce Strategic Research Centre, with encouraging results. It is distinguished from other types by the name: Integrated Planar Solid Oxide Fuel Cell (IP-SOFC). It is a family of integrated system concepts supporting product flexibility with evolutionary stretch potential from a common SOFC module. Fabrication of the key component of the IP-SOFC, the “multi-cell membrane electrode assembly (multi-cell MEA) module” carrying many series connected cells with supported electrolyte membranes only 10 to 20 μm thick, has been proved. Development of the internal reforming subsystem, the next big hurdle, is now in hand. Following an outline of its salient features and test results, the methodology and results of recent IP-SOFC stack costing studies are presented, and the continuing research and development programme indicated. © 2000 Elsevier Science S.A. All rights reserved.

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1. Introduction

The Rolls-Royce of today is a global company. It employs some 40,000 people worldwide, providing power and propulsion systems and support services for civil aerospace, defence and energy markets. As a power and propulsion systems integrator, Rolls-Royce buys in some 80% of the value of its products, creating employment in supplier companies around the world. Rolls-Royce's energy businesses include power generation, gas pumping and ship propulsion, based on aero-derivative gas turbine and diesel engine plant. Its product portfolio spans the range 1 to ~ 100 MW.

Rolls-Royce has a strategic interest in environmental issues, and is in the vanguard of developments to reduce noise, carbon dioxide and harmful emissions from its products and from its industrial processes. The Company is

committed to certifying all its businesses worldwide to ISO 14001, the international standard for environmental management systems, by the end of 2000.

Fuel cells promise to generate electricity from fossil fuels more efficiently and more benignly than alternative systems. For Rolls-Royce, they present both threats and opportunities for its energy businesses.

Between 1987 and 1992, occasional studies of second generation (mainly solid polymer, but also solid oxide) fuel cell technologies were undertaken in Rolls-Royce, leading to the view that the solid oxide fuel cell (SOFC) was likely to be the most relevant fuel cell for the Company's energy businesses. In June 1992, a cautious but active SOFC programme was commenced in the Corporate Applied Science Laboratory, now subsumed into an extended Corporate Strategic Research Centre. This department is responsible for evaluating emerging technologies that may impact the Rolls-Royce businesses, but which are beyond the focus of the main business units.

The aims of the programme are: to learn the technology of SOFCs, building an understanding of their strengths and weaknesses, enabling the Company to make sound deci-

^{*} Corresponding author. T.H. Huxley School of Environment, Earth Sciences and Engineering, Imperial College of Science, Technology and Medicine, London SW7 2BP, UK.

sions on the technology; and to give Rolls-Royce affordable technology for an SOFC power system.

2. Genesis of the Integrated Planar Solid Oxide Fuel Cell (IP-SOFC)

Initially, a more detailed review of the literature on SOFCs was undertaken, a process updated from time to time. Being a solid state technology, the SOFC has the potential for a wide range of design variants, and this is reflected in the literature [1]. Three broad classes were identified, the tubular, the bipolar monolithic and the bipolar planar. Members of the latter two classes have the common feature of a bipolar member (bipolar plate) which prevents mixing between the reactants of adjacent cells, and effects interconnection of the cells in electrical series. With the tubular types (of which there are at least two), cell interconnections are effectively integrated into the cell construction.

From an electrical engineering viewpoint, the bipolar stacking arrangements of contemporary monolithic and planar SOFCs have short current paths, giving low cell resistances and high power density. But from a mechanical engineering perspective, bipolar stacking gives a monolithic structure in a high temperature device constructed from brittle ceramic components with rather high thermal expansion coefficients. The components cannot expand and contract freely, whether or not they are sintered as a monolith, or built from pre-sintered planar components. With this intrinsic lack of thermal expansion compliance, in service cracking problems are to be expected, resulting in cross-over of reactants and loss of performance.

Tubular SOFC stack designs, on the other hand, have the freedom to expand and contract without constraint. These are much more elegant from a mechanical engineering perspective, although from an electrical viewpoint they have rather long current paths, giving high resistance cells and a low power density, compared with bipolar variants. High manufacturing cost [2] coupled with a low power density appeared to be major drawbacks of tubular types, drawbacks which may limit their long-term competitive position.

From these initial studies of the literature, it was not a priori evident that SOFCs would be a worthwhile option for Rolls-Royce's energy businesses, many of which require high power to weight ratio, rapid load following, and "affordability".

Thus, Rolls-Royce examined whether such drawbacks could be overcome, or whether they are intrinsic to SOFCs. To provide a vehicle for our studies, a SOFC stack design was conceived, integrating as much functionality as seemed desirable in a unit designed to aid learning and understanding of SOFC technology issues. This concept, substantially a cross between tubular and planar geometries, seeks to borrow thermal expansion compliance from the former,

and low cost component fabrication from the latter. To distinguish it from contemporary types it is called the IP-SOFC.

3. The aims of the IP-SOFC

The new concept embodies three desirable themes [4]:

- SOFCs with supported electrolytes around 10–20 μm thick, thought likely to enable efficient operation at lower temperatures (below 800°C) [3];
- A recuperative semi-indirect internal steam reforming subsystem, providing flexibility in managing the steam reforming endotherm to prevent local temperature excursions;
- An exothermic partial oxidation reforming subsystem for low power operation, providing the potential to operate the stack at zero and low power in a self-sustaining mode.

The question "Can a SOFC stack with these features be evolved which is affordable to develop, to buy, and to operate?" is needed to be answered. One which:

- May be manufactured by adapting intrinsically cheap fabrication approaches, like the bipolar planar SOFC seems to be;
- Could use its materials efficiently, especially the expensive interconnect materials;
- Can expand and contract freely, like the tubular SOFC;
- Will scale up in power to the multi-megawatt range, of most interest to Rolls-Royce's energy businesses, with some scope for exploiting economies of scale, an attribute seemingly lacking in contemporary fuel cells;
- Puts minimal demands on the balance of plant required to support the stack, making the stack the high value component of the power system;
- Will be operable from zero to full power, producing electricity efficiently and benignly.

4. Key features of the IP-SOFC

To avoid the bipolar plates of contemporary planar SOFCs, a multi-cell stacking configuration, illustrated in Fig. 1, was adopted. This was borrowed from earlier work in Rolls-Royce on a novel solid polymer fuel cell concept evolved in a study, carried out in conjunction with Johnson Matthey, of a Residential Total Energy Module for the South Coast Air Quality Management District and the Southern California Gas of Los Angeles. As with the earlier work, the motivation here was not only to avoid the weight and cost of bipolar plates, but also to avoid the intrinsic monolithic structure of the bipolar stacking configuration.

Fabrication of the supported electrolyte film was perceived to be both a key requirement and a major technological challenge. We decided to avoid gas phase fabrica-

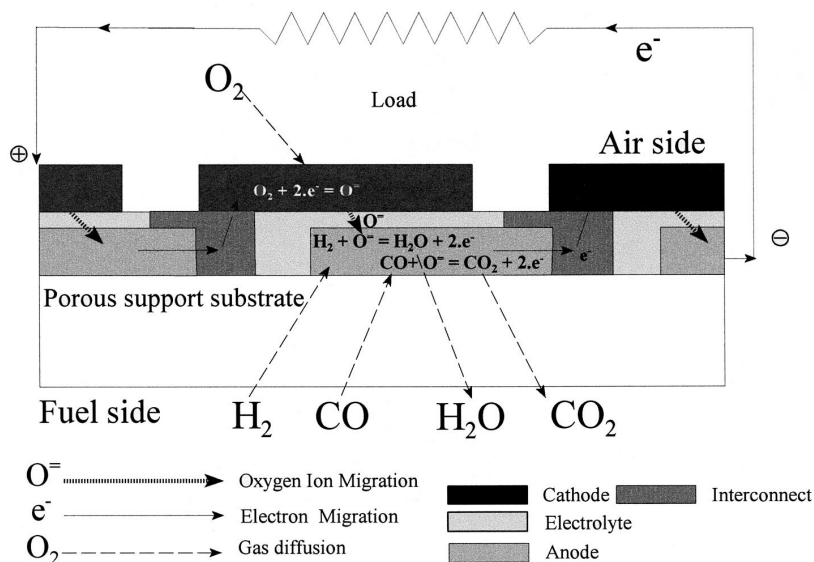


Figure 1 Illustrating the multi-cell MEA concept

Fig. 1. Illustrating the multi-cell MEA concept.

tion routes, such as the various chemical vapour deposition approaches and plasma spraying: the former, too demanding in unaffordable equipment, the latter, unlikely to give high quality dense supported electrolyte films of the thickness required, 20 μm or less. After a brief study of alternatives, wet slurry printing approaches were adopted with progressively encouraging results.

From the outset, it was decided to develop a multi-cell array carrying three series connected cells, rather than single cells, on a 50 mm by 50 mm porous ceramic substrate. Starting with the electrolyte, printing approaches were developed for the anode/electrolyte subsystem, the interconnect subsystem and then the cathode. Development of the “multi-cell membrane electrode assembly” (multi-cell MEA) is reported in Ref. [5]. The three-cell array

(Fig. 1) continues to be the primary unit for studying improvements in cell performance.

A lot of money is needed to develop fuel cells, but what is most needed is much patience and perseverance. This painstaking process took around 4 years before interconnected cells with adequate performance was achieved. Compatibility between the substrate and the cell functional components proved to be the major difficulty, but one which was successfully overcome. The SEM micrograph of the electrolyte sub-cell of a three-cell array in Fig. 2 shows the excellent bonding now obtained, particularly between the anode and the electrolyte.

The interconnect is a possible weak point in the multi-cell stacking arrangement of Fig. 1. Firstly, sealing integrity must be achieved. Secondly, a low resistance is

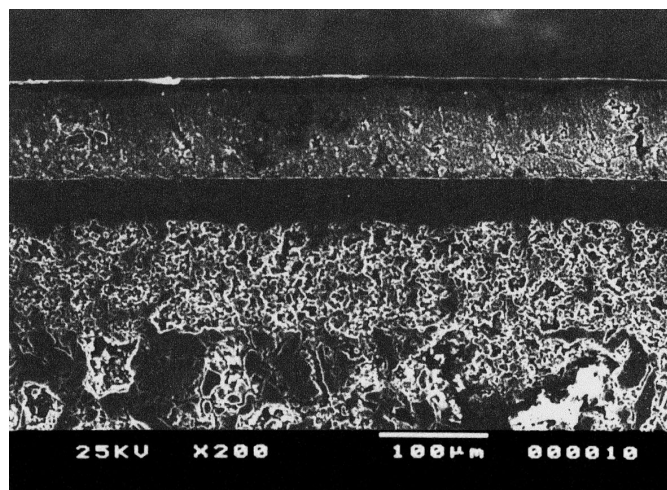


Fig. 2. Micrograph of a supported electrolyte cell deposited by screen printing.

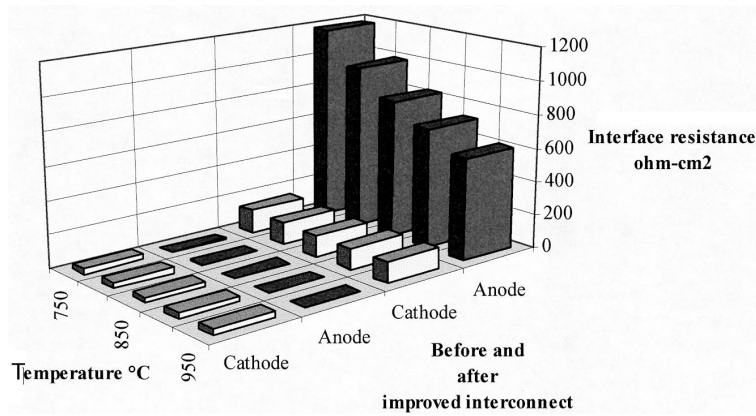


Fig. 3. Illustrating electrode/interconnect interfacial resistances before and after improvements to the interconnect.

vital, as the current density through the interconnect may be an order of magnitude greater than that through the anode/electrolyte/cathode subsystem. Problems with sealing and high interconnect resistance were overcome by attention to the detailed design, materials selection and microstructures of the interconnect subsystem and its components. Fig. 3 illustrates the reductions in electrode/interconnect interfacial resistances realised in this process.

Having achieved an adequate performance at the three-cell level ($\sim 1 \Omega \text{ cm}^2$ mean cell area specific resistance [5]), scale-up of the multi-cell MEA was commenced. The design for scaling up the multi-cell MEA, the “multi-cell MEA module”, is illustrated in Fig. 4. This double-sided

module is the basic building block of the IP-SOFC stack. Note that air flows over the exterior surface, across the outward facing cathodes. Development of the multi-cell MEA module is described in Ref. [6].

Scale-up of the multi-cell MEA has proceeded through three steps: seven cells over a 100-mm span; 20 cells over a 300-mm span; and 30 cells over a 420-mm span. 420 mm is the longest span we can print with our current equipment. Fig. 4 is a photograph of a 30-W multi-cell MEA module with test results. It is double-sided with two multi-cell MEAs, one on each side. Each multi-cell MEA has 20 cells deposited over an active area of 300 mm by 60 mm. The test results refer to 950°C operating tempera-

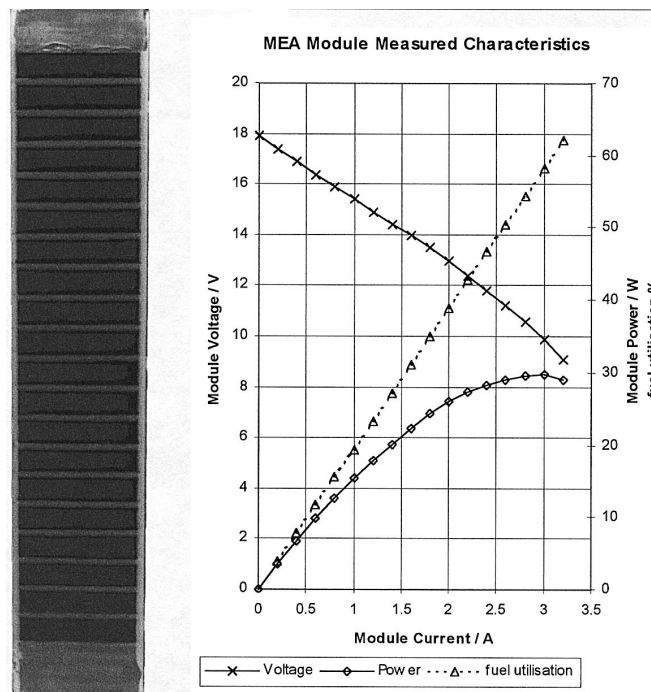


Fig. 4. Thirty watt multi-cell MEA module with measured performance characteristics.

ture, and a fuel inlet molar composition of 66% H₂, 17% H₂O and 17% N₂, the latter emulating the CO₂ content of fully reformed methane.

At this juncture, the cell pitch of the multi-cell MEA module is a non-optimal 15 mm to ease print registration difficulties with the larger print area. This gives a mean cell area specific resistance of 1 to 1.2 Ω cm² at 950°C. As experience with sintering protocols and shrinkage tolerances is gained, expected to reduce the cell pitch to the preferred 10 mm, used in the three-cell MEAs. This will reduce the mean cell area specific resistance to ~0.6 Ω cm² at 950°C, assuming the same functional materials technology. With further functional materials improvements, already demonstrated at the three-cell MEA level, the mean cell area specific resistance should fall to ~0.4 Ω cm² at 950°C. Such improvements are expected to increase the peak power of the 30-W multi-cell MEA module of Fig. 4 by a factor of two to three.

5. Current research projects

Funding for most of this work has come from Rolls-Royce, and from the UK Department of Trade and Industry's (DTI) Advanced Fuel Cells Programme managed by the Energy Technology Support Unit at Harwell Laboratory. The DTI's Advanced Fuel Cells Programme has supported the work over the past 7 years through various projects. These are reported in Refs. [4–6]. Key subcontract support has been provided over the years by CERAM Research, Unitec Ceramics, Imperial College, Keele, Birmingham, Napier and Brunel Universities.

We also benefit from European Union Framework 4 Programme funding, and from the DTI/EPSC LINK "Applied Catalysis and Catalytic Processes" Programme funding. The latter is one of a number of DTI/EPSC LINK programmes that sponsors collaboration between UK universities and industry, providing up to 50% of eligible project costs. EPSC (the Engineering and Physical Sciences Research Council) is the UK Government body that provides funds to the universities in the DTI/EPSC LINK, and other EPSC sponsored, programmes. DTI provides support funding to the industrial partners in the DTI/EPSC LINK programmes.

Our current projects are listed below:

(1) In the DTI's Advanced Fuel Cells Programme, we are:

- (a) Developing a kilowatt IP-SOFC stack containing 36 30-W multi-cell MEA modules. This project is well in hand;
- (b) Increasing MEA module production capacity and quality, in a project recently started. This project also aims to improve cathode durability and to develop co-sintering protocols for the functional components of the multi-cell MEA;

(c) To develop a thermally self-sustaining IP-SOFC stack of ~5 kW, with subsystems for cold start-up and low power operation, following on from the kilowatt stack, above.

(2) In an EU Framework 4 Brite-Euram Programme project (LOCO-SOFC, Low-Cost Fabrication and Improved Performance Of SOFC Stack Components) together with our partners, we are improving the performance of SOFC cells over 800–1000°C, focusing on cells manufactured by inexpensive approaches. This project is led by Risø National Laboratory (DK). The other partners are: Institut National Polytechnique de Grenoble (F), Ecole Polytechnique Federale de Lausanne (CH), Gaz de France, Napier University Ventures (UK) and Innovision R&D (DK).

(3) Finally, in the DTI/EPSC LINK "Applied Catalysis and Catalytic Processes" Programme we are working with Imperial College, Keele University, ICI Syntex and Advanced Ceramics to develop the internal reforming subsystem of the multi-cell MEA module.

6. Recent progress in multi-cell MEA performance

Progress in our LOCO-SOFC Project (2 above), led by Risø National Laboratory, has been particularly encouraging. Recent results are given in Fig. 5. These are mean cell characteristics of multi-cell MEAs with three series connected cells tested with humidified hydrogen and air. Thus, they include cell interconnection losses as well as electrolyte resistance, electrode polarisation and electrode current distribution losses.

In Fig. 6, the mean cell area specific resistance (ASR) vs. temperature characteristic, derived from these results, is compared with a previous characteristic measured shortly before the LOCO-SOFC project started. The reduction in area specific resistance at lower temperatures is particularly beneficial, enabling the stack to be designed for operation with lower air inlet temperatures, the greater air inlet to exit temperature rise easing air pre-heating requirements, among other considerations. To assess the indicative costs of IP-SOFC stacks using such cells, such ASR characteristics are used to extrapolate test results to stack operating conditions in design simulation studies.

7. Indicative cost projections for the IP-SOFC

The IP-SOFC Programme in Rolls-Royce encompasses IP-SOFC stack concept development, technology development, and manufacturing feasibility evaluation. Concept development seeks to provide direction while manufacturing feasibility evaluation seeks to provide realism, to the programme. The concept development stage usually fixes ~80% of the eventual cost of a product. The concept development stage is the best time to reduce costs. From

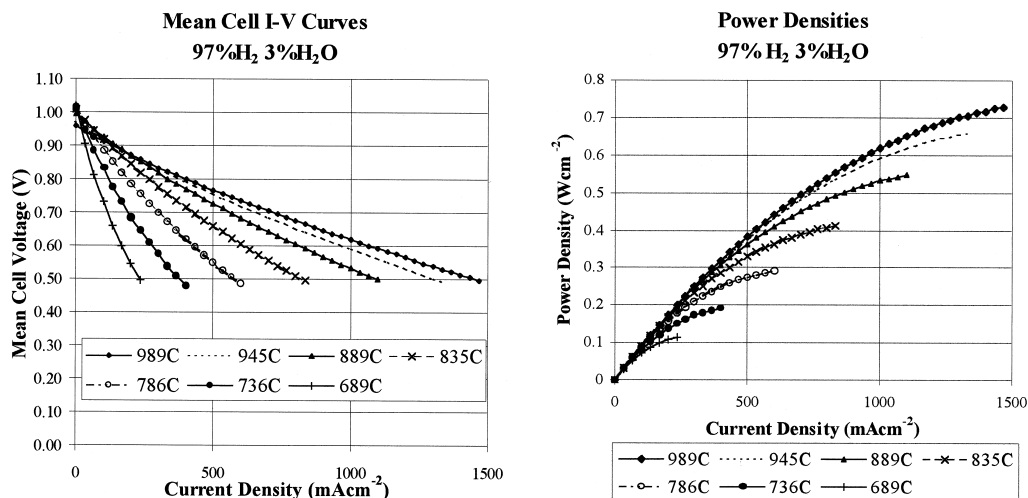


Fig. 5. Recent multi-cell MEA performance characteristics from the EU Brite-Euram LOCO-SOFC project.

time to time Concept Reference Design and cost studies are carried out to guide concept development. The First Concept Reference Design cost study is reported in Ref. [4]. This suggested indicative costs in the range £400–600/kW for stack gross efficiencies in the range 52% to 67%, for early volume production (75 MW pa) stacks of ≈ 1250 kW rating. These costs are far too high.

The high specific cost of the First Concept Reference Design motivated changes to simplify the IP-SOFC stack design. The aim was set to reduce predicted stack costs to less than the £200/kW (\$300/kW), generally believed to be the commercial target for introduction of SOFCs. As a result, the IP-SOFC is now a flexible family of integrated system concepts with:

- Sub-megawatt power generation options with potential stack efficiencies of 55% to 68% LHV;

- Multi-megawatt combined gas turbine cycle options with potential gross cycle efficiencies of 70–80% LHV.

Further Concept Reference Design and cost studies have been carried out recently, on both these cases. The procedure adopted for estimating costs for the IP-SOFC stack is as follows: (1) Computer-aided Concept Reference Designs were developed for: (a) Sub-megawatt IP-SOFC stacks for stand-alone applications, (b) Multi-megawatt IP-SOFC stacks for combination with a gas turbine bottoming cycle; (2) Bills of materials were derived for these; (3) A model of the processes for fabricating components and for constructing the alternative IP-SOFC stacks was drawn up, which together with the bills of materials, facilitated the costing of materials used, equipment required, labour needed, energy used and infrastructure required; (4) Cost breakdowns were derived for producing: (a) 500 sub-megawatt IP-SOFC stack units per year

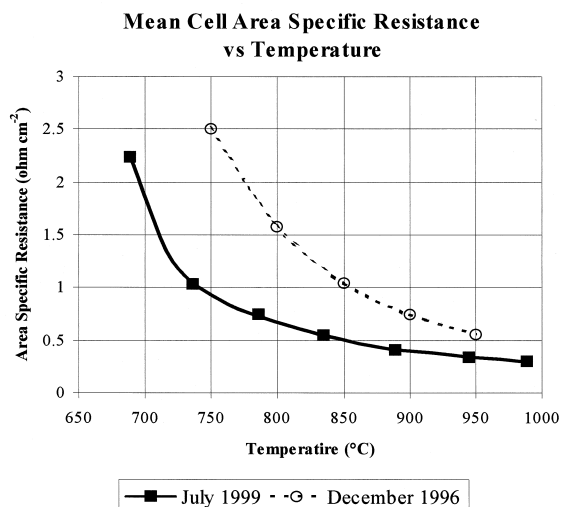


Fig. 6. Illustrating the improvement in multi-cell MEA resistance characteristics won in the EU Brite-Euram LOCO-SOFC project.

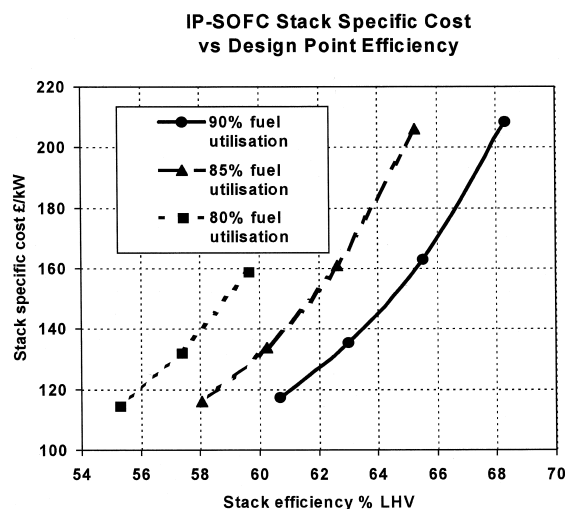


Fig. 7. Indicative specific stack costs of IP-SOFC stacks for sub-megawatt stand-alone applications.

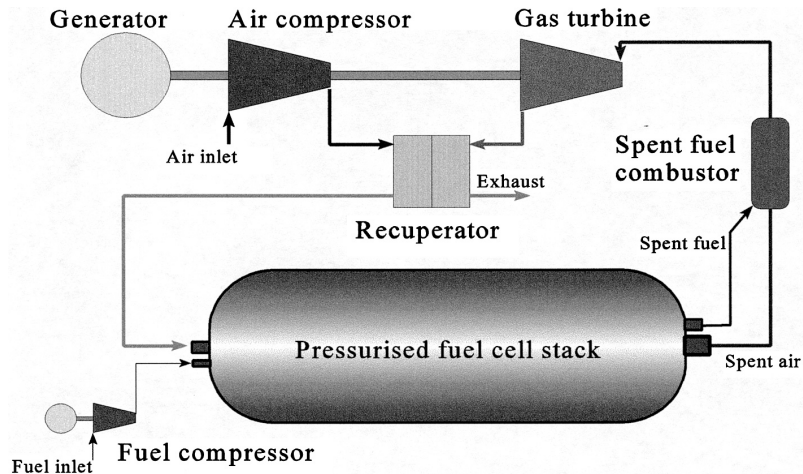


Fig. 8. Schematic of a recuperated SOFC/gas turbine cycle.

(~ 100 – 200 MW/year), (b) 100 multi-megawatt IP-SOFC stacks units per year (~ 1000 – 2000 MW/year); (5) Steady state design simulations were carried out for the cases 1.a and 1.b, above, giving process parameters throughout the internal reforming stacks at alternative design full load current levels. The improved mean cell area specific resistance characteristics given in Fig. 6 were used in these simulations; (6) Results from 4 and 5 were used to gauge the sensitivity of stack specific cost to stack efficiency and design full load rating.

Indicative cost projections on this basis for sub-megawatt stacks (of say 200–500 kW rating) for stand-alone applications are given in Fig. 7. Stack specific cost is plotted against stack design point efficiency, with fuel utilisation as a parameter. Cost projections are below the £200/kW (\$300/kW) target, significantly below if we

choose a relatively low stack design point efficiency by driving the stack harder to get more power out of the same unit. With the cell data in question, £120/kW (\$180/kW) appears to be the lower limit. At this cost level, stack gross efficiencies are 56–62%, depending on fuel utilisation efficiency. If we are prepared to pay £200/kW (\$300/kW) stack gross efficiencies of 65–68% are indicated.

Turning to the IP-SOFC/gas turbine combination, the schematic system analysed is given in Fig. 8. A recuperated cycle is assumed, with a high temperature recuperator. In this cycle, the IP-SOFC stack and spent fuel combustor replace the usual combustion chamber. Spent fuel is combusted down stream of the stack, maximising the turbine inlet temperature (TIT). Stack specific cost vs. stack design point efficiency is plotted in Fig. 9. Combined cycle gross efficiency is also plotted (on the RH axis). At the

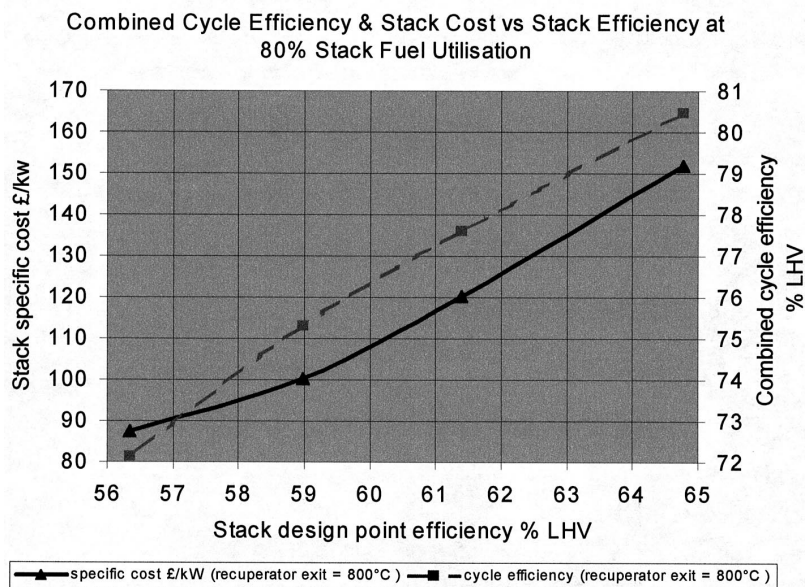


Fig. 9. Indicative costs of multi-megawatt IP-SOFC stacks for combination with gas turbine bottoming cycles.

lower stack design point efficiencies specific cost falls below £100/kW (\$150/kW). Even so, gross combined cycle efficiencies of 73% to 75% LHV are indicated. At the higher stack design point efficiencies, efficiencies of 80% LHV are indicated, with stack costs of ~ 150/kW (\$225/kW).

8. Conclusion

In summary: (1) Following on from previously reported work [4], improvements simplifying the IP-SOFC stack concept have been introduced. As a result, the IP-SOFC is now a flexible family of integrated system concepts, based on a universal multi-cell MEA module, giving: (a) Sub-megawatt power generation options with potential stack efficiencies of 55% to 68% LHV, (b) Multi-megawatt combined gas turbine cycle options with potential gross cycle efficiencies of 70–80% LHV. (2) Supported electrolyte multi-cell MEAs have been demonstrated to have a high performance capability (Figs. 5 and 6). (3) If this performance can be achieved at multi-cell MEA module level (the basic building block of the IP-SOFC stack), projected specific stack costs should be in the range £100–200/kW (\$150–300/kW, 150–300euro/kW), depending on the stack design point efficiency preferred.

Much further work is needed before the IP-SOFC is ready for commercial demonstration:

- (A) Stack development and scale-up to commercial ratings is required, simultaneously achieving durable high performance and reliability;
- (B) Development of a stable internal reforming subsystem is required, first at the multi-cell MEA module level, and then at the stack level;

- (C) Development of a pilot component fabrication and supply infrastructure is required, before;
- (D) Significant technology demonstration, followed by commercial demonstration, can be mounted.

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