

# Critical issues and future prospects for molten carbonate fuel cells

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## Abstract

The molten carbonate fuel cell (MCFC) has several potential advantages over low-temperature fuel cells by virtue of its operating temperature of 650 °C. This temperature allows the reforming of, for example, methane from natural gas in the fuel cell stack itself, resulting in reduced systems cost and increased efficiency. In addition, high temperature waste heat is available for industrial processes or bottoming cycles. Furthermore, CO, which is produced in almost all fossil fuel conversion processes, can be used as fuel instead of acting as a poison as in other types of fuel cell. Drawbacks of MCFCs are the high corrosivity of the electrolyte at the operating temperature and the need for a continuous supply of CO<sub>2</sub> to the cathode. Research into and development of MCFCs actually started in 1950 by Ketelaar and Broers when they investigated an earlier idea of Davtyan. Since then, a lot of progress has been made with respect to understanding the cell mechanisms, improving the materials, the performance, the manufacturing techniques and up-scaling. This resulted a few years ago in proof-of-principle tests at the 100 kW level. At present, the MCFC is in the first demonstration phase with full-scale systems at the 250 kW to 2 MWe level, marking the transition from fundamental and applied R&D to product development or from a technology push to a market pull situation. This paper reviews the most important remaining as well as expected new issues to be resolved.

*Keywords:* Molten carbonate fuel cells; Development

## 1. Introduction

The molten carbonate fuel cell (MCFC) demonstration projects under construction are the first of their kind. The stacks are the product of a long-term laboratory development [1] and represent already a somewhat out-dated technology. The systems have been designed without previous experience other than that from test-scale installations. In some cases, dedicated system components have been developed and will be operated for the first time, some user's requirements being incorporated. These systems are extremely complicated and thus expensive. They are not representative for future competitive systems and they do not demonstrate the potential benefits to their full extent.

Extensive market analysis with respect to individual applications should guide product development, user's requirements and competing technologies. These parameters will clearly define new targets in terms of cost, performance, reliability, operation, maintenance, etc., the cost being the most important target. New system concepts should therefore be designed, engineered and demonstrated providing in an interactive process, new boundary conditions, requirements and specifications for sub-systems and individual components. Because of the many features involved and their inter-rela-

tions, optimization of the total system to reach commercial competitiveness will prove to be a tedious process.

## 2. System costs

Cost can be defined in several ways, but the most important figure is the cost per unit of useful energy produced or, alternatively, the cycle-life cost. These figures are needed by the consumer or user to determine the profitability, and by the manufacturer to determine the capital cost or prospects for investment. Hardware, engineering and other costs can be quantified. However, there is also a number of non-quantifiable cost parameters that will add additional constraints. Also, external factors, site specific and locally determined cost factors will have to be taken into account.

An example of the latter can be found in a recent IEA study [2]. In this study the investment feasibility was determined for a 500 kWe combined heat and power (CHP) installation for a specific industrial application. Fig. 1 gives the relative results for seven participating countries (A–G). The tariff structure is the main reason for the large differences shown with respect to fuel, power and heat in the individual countries. This indicates clearly that the often quoted cost goal of US \$ 1500/kWe is not more than a first approximation which certainly is not generally applicable.

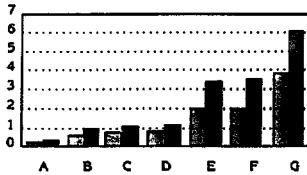


Fig. 1. Comparative room for investment for a 500 kW MCFC combined heat and power plant in seven IEA countries for a typical industrial application and pay-back times of 5 and 15 years. A: Norway, B: Sweden, C: The Netherlands, D: UK, E: Italy, F: Germany, and G: Japan.

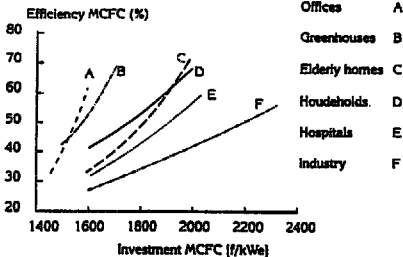


Fig. 2. Electric efficiency and capital cost at which a 500 kW MCFC CHP plant is competitive with a heat engine in different applications.

In an additional study for Dutch conditions [3], the profitability of this 500 kW installation was determined for six different applications and compared with a competitive heat engine. Fig. 2 shows the electrical efficiency (the total efficiency of the system was kept constant at 85%) and the capital cost at which the profitability of the MCFC is similar to that of the competing heat engine. Again, there is an inter-relationship between the application and its requirements. This study also showed that, especially for higher annual load factors, the investment cost is more important with respect to the profitability than the electric efficiency of the MCFC system.

The potential for the cost reduction is different for stacks and the balance-of-plant (BOP) components. The stack consists of a large number of components for which high volume production will lead to a very substantial cost reduction which is essentially limited by the cost of the raw materials. The BOP, however, consists mainly of off-the-shelf components which are being produced in high volume for other applications. In this case, series production will have only a minor effect and cost reduction will have to be obtained through the design of highly simplified and integrated systems. The latter may be supported by newly developed auxiliaries, controllers and regulations [4].

3. System design

For the general theory of fuel cell system design, and of MCFC systems in particular, reference is made to Refs. [5,6].

In designing advanced systems, heat and mass transport, pressure and power control, system control and regulation are the most important factors. For further optimization, exergy analysis or pinch technology may be applied.

The MCFC operates at a sufficiently high temperature in order to reform methane over a catalyst. Thus, by incorporating the reforming catalyst in the stack and preferably in the anode chamber of the individual cells (direct internal reforming), the heat of the exothermal cell reaction for the endothermal reforming reaction is provided on site. This reduces heat and mass transport for reforming to an absolute minimum. In addition, the hydrogen produced by the reforming reaction is consumed on site by the cell reaction, thereby bringing the reforming reaction to completion instead of equilibrium which, in turn, causes an increase in efficiency. Fig. 3 illustrates the principle. Compared with a system with external reforming of natural gas, internal reforming has the obvious following advantages: elimination of the reformer with its associated equipment and of the water knock-out system, a high degree of heat integration, a lower cooling demand with its associated parasitic losses and an increased stack and system efficiency. A drawback is a somewhat higher cost for the stack due to the incorporation of the reforming catalyst. Another drawback may be poisoning of the catalyst by the electrolyte. Therefore, some developers have chosen for the indirect internal reforming option, in which the reforming catalyst is incorporated in the stack in separate cells. This will result in a more complicated stack design and a somewhat lower efficiency. Based on the principle of internal reforming further optimization is possible as illustrated in Ref. [7]. For advanced system concepts in the power range of 250 kW to several MWe internal reforming may result in a cost reduction of 50% or more as compared with external reforming systems.

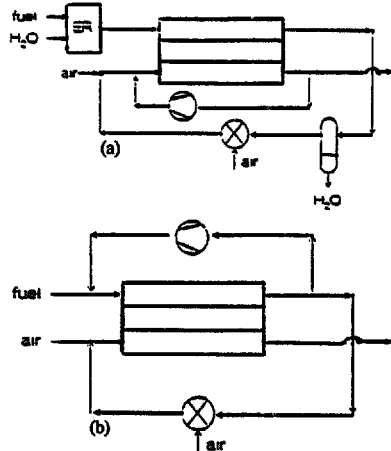


Fig. 3. Concepts of (a) external and (b) internal reforming systems.

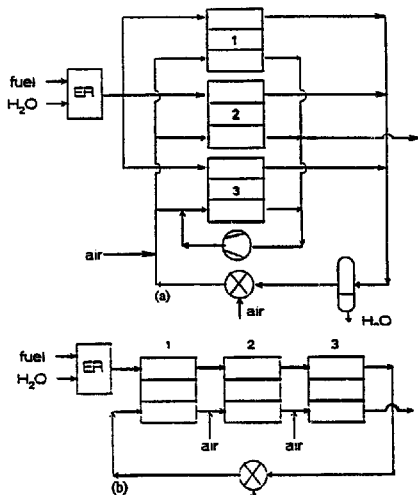


Fig. 4. Standard (a) operation and (b) networking of stacks.

A second major option is the networking of stacks [8]. Traditionally, stacks in a system are operated in parallel with respect to the process gas flows. By connecting the stacks in series and injecting relatively cold process gases in between stacks (see Fig. 4), the cooling demand will be strongly reduced. In addition, at a constant total fuel utilization, the fuel utilization for each stack can be reduced and the stack efficiency increased. Overall, the cooling demand with its associated parasitic power consumption will be reduced, the high temperature cathode recycle blower and the water knock-out systems can be eliminated, hot valves for stack control can be replaced by low temperature valves and the stack and system efficiencies will be increased. This will lead to a substantial cost reduction, while in a first approximation Ref. [8] quotes system efficiency increases of 10% for external reforming and 5% for internal reforming. Also this concept offers many opportunities for further optimization.

#### 4. Stack issues

Extensive reviews on stack parameters have been presented by Selman [9] and Penner et al. [10]. These can be grouped into four main categories: cost; endurance; reliability, and performance. Further stack development will raise new issues and change the boundary conditions for a number of them. These boundary conditions concern temperature level, sustained temperature differences, thermal shock and controlled thermal cycles, pressure level, sustained pressure differences, pressure shock, rates of power and gas compo-

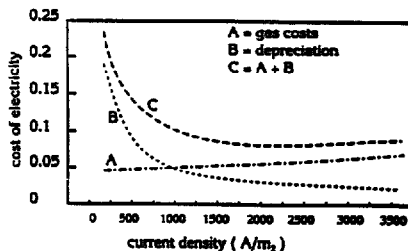


Fig. 5. Stack contribution to the cost of electricity of a dispersed power plant vs. current density.

sition change, etc. As many factors are involved and various options exist for resolving each of the issues, further development and optimization will certainly lead to an acceptable solution. The main effort should be directed to the cost and life-time parameters, while the optimization performance may be used as a trade-off parameter.

Two parameters contribute to the cost of electricity ( $C_{el}$ ) of a dispersed power generator: the first is directly proportional to the gas price ( $G$ ) divided by the electrical efficiency ( $\epsilon_{el}$ ), and the second is the depreciation of the stack which is proportional to its capital cost ( $I$ ) divided by its service life ( $L$ ) and its electrical power density ( $P_{el}$ ):

$$C_{el} = \frac{G}{\epsilon_{el}} + \frac{I}{LP_{el}} \quad (1)$$

For a constant gas utilization, the power density is directly proportional to the current density, and an increase in power density results in a proportional increase in current density.

This will lead to a decrease in electrical efficiency. Fig. 5 shows the results [11], indicating a broad minimum in the cost of electricity with respect to current density. In addition, however, other factors should be taken into account: increase in cooling demand, and probably a reduction of service life. The general statement, therefore, that cost reduction should be obtained by increasing the stack performance should at least be regarded with caution.

#### 5. Stack cost

Stack costs should of course be compatible with the capital cost of the system. As a rule of thumb, the stack would contribute to about 1/3 of the system cost at full scale production levels. It is generally accepted that the cost of raw materials would constitute about 80% of total stack costs.

Stack costs are to a large extent determined by the design. However, the today's large area flat plate designs offer little room for further cost reductions, except by increasing production volumes.

Materials costs, as indicated above, will be the determinant factor. Opportunities for further cost reduction should be

found in reducing the amount of materials used and in the use of cheaper materials. Less material will lead to thinner components, which may have an adverse effect on endurance and reliability. But there is ample room for reduction regarding the current collectors, the electrodes, if their specific performance can be increased, and in the electrolyte matrix if its strength can be improved and the cathode dissolution problem solved. Great cost reduction can also be expected when lithium salts are produced in big quantities. On the other hand, more expensive materials may be necessary to solve the cathode dissolution problem.

The separator plate, at present, is made of a 0.5 mm thick sheet metal. Cheaper base metals can only be used if sufficiently cheap corrosion protection techniques can be developed. On the other hand, fundamental research in the corrosion process and the development of model alloys may result in the final application of more expensive materials.

Sintering of the electrodes is the most important cost factor, giving possibilities for further development and cost reduction, e.g. by speeding up the sintering procedure. Although tape-casting is probably the cheapest manufacturing technique for the porous components, further cost reduction seems to be possible by fine tuning of the slurry formulations and speeding up of the drying step. Finally, also minor adjustments in the separator plate design are possible to reduce manufacturing costs.

Performance is also an important factor in determining the investment cost per installed kW. As discussed above, performance improvements will be traded-off, in a first step, for direct cost reduction of materials and manufacturing and probably also life time where possible. The remainder can then be used for further system optimization.

In a recent ECN study for the Dutch Fuel Cell Corporation BCN, an inventory was made of a number of cost reduction options for each stack component. Their direct effect on other stack components and their properties was assessed and their chances for successful development were estimated. This shows that for an annual production rate of 200 MWe, stack costs of US \$ 500 per kWe are feasible, including assembly, pressure vessel and acceptance testing at the factory. Material costs for internal reforming may be up to 25% higher.

## 6. Stack life

Stack life is an essential factor in determining the cost of electricity, as shown above. In addition, analysis of well-developed systems shows that an increase in stack life from 3 to 5 years will increase the prospects for investment for the whole system by 25 to 30%. The goals for stack service life range from 25 000 to 40 000 h. The main issues that determine the endurance are cathode dissolution, separator plate corrosion, electrolyte inventory and, for internal reforming, catalyst poisoning.

The cathode consists of NiO, which slowly dissolves in the electrolyte during operation. It is then transported towards

the anode and precipitates in the electrolyte matrix as Ni. These processes lead to a gradual degradation of cathode performance and the cell being shorted. Both of these processes limit stack life under atmospheric reference gas conditions to about 25 000 h [12]. Under real operating conditions, however, life times are much shorter. Factors that influence these processes are the acidity or basicity of the electrolyte, operating pressure, concentration of CO<sub>2</sub> and water in the process gases. By making use of these factors, a number of partial solutions to extend stack service lives have been developed. It is, however, doubtful whether combinations of these measures will be sufficient to reach the target 40 000 h, or even more in the future. In a search for alternative cathode materials, LiCoO<sub>2</sub> showed a dissolution rate which is one order of magnitude lower than that of the standard material, while it is much less dependent on the operating conditions, no metal being precipitated in the electrolyte matrix [12]. In spite of the higher cost of cobalt, compared with that of nickel, this former material is attractive in that its life time may reach 100 000 h or even more.

The second important issue is the corrosion of the separator plate being dependent on the substrate, possible protective layers, composition of the electrolyte, local potential and gas composition, and the oxidizing and reducing atmospheres at the cathode and anode, respectively. Stainless steel 310 or 316 is used as the base material. In the wet seal area it is protected by aluminization and in the anode active area by a nickel layer.

During operation a protective oxide layer is formed in the cathode active area. When growing, this layer tends to spall, certainly if subjected to thermal cycles, and thus will impair the hydraulic behaviour of the stack. How far this is detrimental is not yet clear.

The situation on the anode is much more important because alloying elements from the substrate as well as oxygen from the anode diffuse into the nickel layer [12]. These form oxides that will destroy the nickel layer at a point in time that depends on its thickness. For most of the coatings life time seems to be limited to the order of 10 000 h. Research to avoid this problem are in progress.

Full aluminization of the wet seal area, finally, will be effective up to 20 000 h. Improvement of the application process will further extend this period.

The electrolyte is consumed by various processes. Lithiation of the cathode, the formation of lithium chromite or lithium aluminate in the anode, transformation of aluminium oxide fibres in the matrix and the initial corrosion of the cell hardware are almost time independent. Later corrosion rates are rather slow and also creep seems well under control. Potassium carbonate, however, reacts with the available water to form potassium hydroxide which has a relatively high partial pressure. Its evaporation will seriously limit the stack life time [12]. The process is strongly dependent upon the outlet temperature, the pressure, the flow rates and the gas composition. These are determined by the system, and calculations based on experimental evidence show that under

realistic operating conditions target life times cannot be met. Some relief may be expected from substitution of  $K_2CO_3$  by  $Na_2CO_3$ . It may well prove necessary to develop a system for supplying additional electrolyte to the stack during its service life.

Poisoning of the reforming catalyst is another factor in direct internal reforming. Responsible processes are the evaporation of electrolyte from the cell components and condensation on the catalyst (which is the coldest spot of the cell) and by liquid creep within the cell. Progress has been made in avoiding this effect [13]. As an alternative, more expensive indirect internal reforming options are being developed.

In summary, great development efforts supported by fundamental research will be needed to reach target life times. But sufficient options are available and recent long-term experiments are sufficiently encouraging to predict that the short-term milestone of 25 000 h and the later milestone of 40 000 h can be met.

## 7. Stack reliability

Like other fuel cells, the MCFC does not contain any moving parts, so it is in principle highly reliable. The reliability, therefore, is to a large extent dependent on endurance issues. Other mechanisms, their rate of occurrence, propagation and statistics are hardly known or unavailable. Theoretical models are lacking. In some cases, however, stacks have been operated for prolonged times with failed, but self contained, cells. In these cases the current was probably maintained by the inverse cathode reaction. Future system lay-outs will probably require the stacks to be very sturdy in order to withstand pressure, thermal and power shocks. These may occur by unintentional plant outages. Special attention in this respect will be needed for the electrolyte matrix which is the weakest part of the cell. Alternatively, the system should be designed to protect the stack from such occurrences but this will certainly increase total plant capital cost.

## 8. Stack performance

Cell and stack performance is determined by the Nernst, ohmic and polarization losses. The first are influenced by system lay-out and operating requirements and will probably not be taken into consideration in order to optimize the stack performance because of consequences for system capital cost and profitability.

Ohmic losses, both at the anode and cathode, can probably be reduced by another 50% as a result of improved design and reduction of electrode thickness [10].

There may be an opportunity for a reduction of polarization at the anode while the cathode offers appreciably more room

for reduction. This gain will be, at least in part, traded for a reduction of material inventory and cost. Overall, it is expected that future stacks will operate at power densities of 1–2  $kWe/m^2$  and per cell which is comparable with today's 1.25  $kWe/m^2$ .

## 9. Conclusions

Molten carbonate fuel cells are in their first demonstration phase, marking the transition from fundamental and applied research and development towards product development. Further system development and engineering will be guided by a detailed market analysis and will be characterized by simplicity and a high degree of integration of functions. This will raise new issues which, together with the remaining issues of cost, endurance, reliability and performance, will have to be resolved within shifting boundary conditions. Although substantial development efforts supported by fundamental research are still needed, the available knowledge and number of alternatives will probably make it possible to produce pre-commercial units by 2000 at a capital cost of US \$ 2000–4000/ $kWe$ . Pre-competitive commercial units may be expected some years later by which time further cost reduction to full competitiveness will be guided by extensive operating experience and increased volume production, continued financial support and dedication of all parties concerned being a prerequisite.

## References

- [1] J.A.A. Ketelaar, in L.J.M.J. Blomen and M.N. Mugerwa (eds.), *Fuel Cell Systems*, Plenum, New York, 1993, pp. 19–35.
- [2] L. Sjunnesson, personal communication.
- [3] D. Jansen, personal communication.
- [4] C.M. Seymour, *Advances in system design and development*, personal communication.
- [5] M.N. Mugerwa and L.J.M.J. Blomen, *Fuel Cell Systems*, Plenum, New York, 1993, pp. 201–244.
- [6] M. Krumpelt, R. Komar and K.M. Myles, *J. Power Sources*, 49 (1994) 37–51.
- [7] P. Kraus, *J. Power Sources*, 49 (1994) 53–59.
- [8] J.G. Winer, M.C. Williams, D.K. Archer and J.F. Osterle, *Molten carbonate fuel cell networks: principles, analysis and performance*, 28th IECEC, Atlanta, GA, USA, 8–13 Aug. 1993.
- [9] J.R. Selman, in L.J.M.J. Blomen and M.N. Mugerwa (eds.), *Fuel Cell Systems*, Plenum, New York, 1993, pp. 395–463.
- [10] S.S. Fenner, *Energy*, 20 (1995) 331–470.
- [11] G.J. Kraay, personal communication.
- [12] L. Plomp, R.C. Makus, E.F. Sitters and G. Rietveld, *Ext. Abstr., Fuel Cell Seminar, San Diego, CA, USA, 1994*, Courtesy Associates, Washington, DC, USA, pp. 164–167.
- [13] T. Murahashi, M. Matsumura and H. Urushibata, *Ext. Abstr., Fuel Cell Seminar, San Diego, CA, USA, 1994*, Courtesy Associates, Washington DC, USA, pp. 230–233.