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Development of second generation direct internal reforming molten carbonate fuel cell stack technology for cogeneration application

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Abstract

A system-dedicated stack is the basis for second generation direct internal reforming molten carbonate fuel cell (DIR–MCFC) stack development. Therefore, an internally manifolded, co-flow stack with robust sealing properties and lifetime >25 000 h is being developed. Stack design, validation of design tools and obtaining the specifications for full size stack components are the prime objectives. © 1998 Elsevier Science S.A.

Keywords: Molten carbonate fuel cell stack; Direct internal reforming; Internal manifolding; Modelling

1. Introduction

ECN together with partners Stork, Schelde and BG plc carries out a 3-year programme on second generation stack technology for direct internal reforming molten carbonate fuel cell (DIR–MCFC) stack development within the framework of the collaborative European Advanced DIR–MCFC Development project [1].

The consortium consists of the Dutch Fuel Cell Corporation (BCN) (Stork, Schelde Systems and ECN), BG plc, Gaz de France and Sydkraft AB. ECN has started MCFC development in 1986, and has carried out 44 stack experiments since 1989. An overview on the characteristics of these stack experiments is given in Table 1.

2. Advanced DIR–MCFC development programme

The Advanced DIR–MCFC programme is strongly 'top-down' driven (i.e. market led), ensuring that the resulting cogeneration plant meets all technical, economic, legislative and environmental requirements. Three fields are covered, i.e. (a) market analysis [2], (b) system development [3] and (c) stack development. This paper is presenting the status of the stack development.

From the market analysis, cogeneration for hospitals was identified as a preferred market entry application at a size of 400 kWe. Essential is the development of simple and low 'cost-of-electricity' systems, which can be operated within the stack interface conditions. In the selected system, series connection of stacks occurs and the stack operates at ambient pressure [1]. In the stack development programme a DIR–MCFC, internal manifolded, co-flow stack for operation in the envisaged system is being developed.

Specific objectives for the stack include sustained pressure differences across the stack and extension of the lifetime to 25 000 h. Activities include: (l) stack modelling and

Table 1

Overview of stack experiments at ECN

Type	External reforming	Direct internal reforming
No. of experiments	32	12
Maximum pressure	0.6 MPa	0.1 MPa
Maximum active area	0.33 m^2	0.1 m^2
Maximum operating time	6343 h	5137 h
Maximum power	14 kWe (33 cells; 0.33 m^2)	2.5 kWe (20 cells; 0.1 m^2)

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validation in order to define the stack operating window for acceptable performance and degradation, (2) development of a new separator plate design and (3) optimization, characterization and manufacturing of porous components for use in realistic system operating conditions. BG plc researches and supplies the catalyst to the development programme.

3. Stack development issues

The system demands for the stack have been translated into a stack design and a stack component development programme. The design for full scale (1 m^2) stacks incorporates:

- thermal management with DIR, i.e. the steam reforming reaction of methane absorbs approximately 60% of the heat produced in the cells;
- flow and pressure management for cathode series connected stacks, i.e. the flow distribution within stacks should be homogeneous for obtaining high fuel utilisation;
- stack operating strategy with degradation of stack components.

For designing a stack, the following design tools were developed:

- 3-D stack model, consisting of integrated
  - – electrochemical description of the cell;
  - – hydraulic description of internal manifolding, co-flow stack;
  - – thermal balance with internal reforming, heat production, heat loss and conduction;
- cost of investment model for an insulated and confined stack module. This guides the design solutions on the basis of cost-effectiveness, and will be used to assess the stack module cost at the end of the programme.

The 3-D stack model has been validated with various

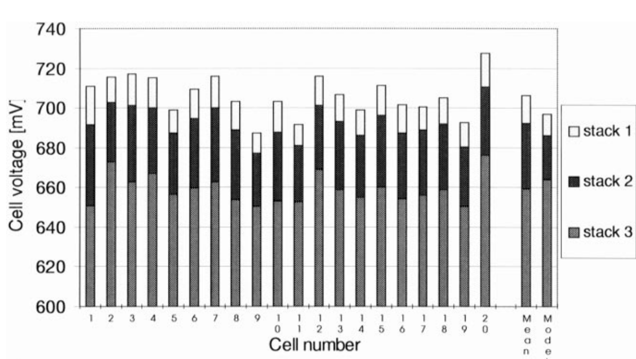

Fig. 1. Performance of a 20-cell stack designed for 3-D stack model verification tested at 3 different system conditions with $j = 1775$ A/m^2, denoted stack 1, stack 2 and stack 3.

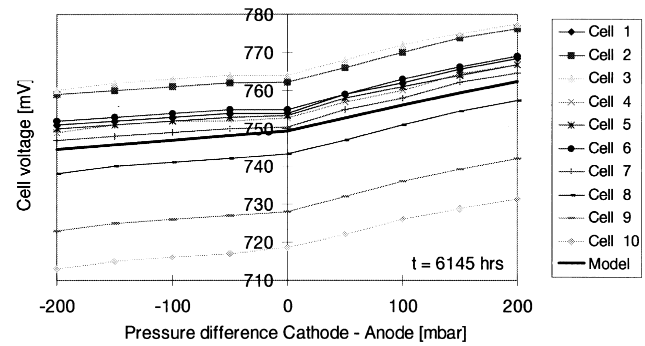

Fig. 2. Performance of stack and model at pressure differences between fuel and oxidant up to 200 mbar. At the left side the oxidant is at atmospheric pressure, at the right side the fuel.

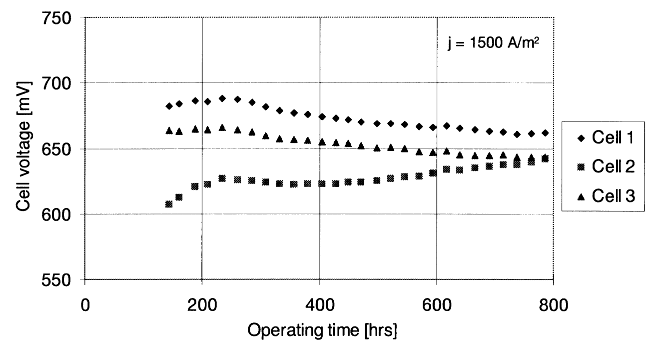

Fig. 3. Performance of $LiCoO_2$ cathode in a 3-cell stack experiment of 0.1 m^2.

dedicated cell and stack experiments. An example of the verification of the electrochemical model of the cell with the measured cell voltages at different system conditions is shown in Fig. 1. This 20 cell DIR stack experiment has run for over 5000 h with state-of-the-art components in order to validate the model with many different simulated gas and temperature conditions of various series connected stacks in one of the preferred systems [3]. Differences between the mean measured cell voltage and the model predictions are smaller than 10 mV (c.f. Fig. 1, the bars ‘mean’ and ‘model’).

4. Separator plate development issues

For a simple system design it is required that the stack should have robust sealing properties. The separator plate under development is based on the external reforming ECN FLEXSEP design. Adaptations are required in order to accommodate the reforming catalyst. In addition special attention is paid to the stack seals and to improvements in corrosion protection. Material development of separator plate coating is necessary for obtaining a lifetime >25 000 h, since the state-of-art nickel coating at the fuel side has a projected lifetime of only 10 000 h.

Table 2

Lifetime for state-of-the-art and alternative materials

Component	State-of-the-art	Endurance (1000 h)	Alternatives (>40 000 h)
Anode	Ni-Cr, Ni-Al	40.80	–
Cathode	NiO	6–25	$LiCoO_2$
Matrix	Reinforced $LiAlO_2$	>25	
Electrolyte	Li/K 62/38	7–18	Li/Na
Separator base material	AISI 310 S	>25	New base materials
Separator			
Coating anode	Ni-layer	10	New coating
Wet-seal	Aluminium	>25	
Catalyst	BG plc	?	

The design of the so-called DIRSEP separator plate for the programme comprises:

- internal manifolding for the sealing properties;
- an interior at the fuel side to integrate the catalyst; supplied by BG plc;
- a soft rail, for compliance during assembly and to accommodate for the thickness decrease of the active cell components during life.

The cell voltages for varying pressure differences between anode and cathode for a leak tight 10-cell stack experiment up to 0.2 bar are shown in Fig. 2. The measured voltage dependence on pressure is in accordance with the model calculations for leak tight stacks. This means that the goals for leak tightness of the stack are attainable.

5. Material development issues

5.1. NiO cathode

The issue for material development is lifetime improvement. The lifetime estimates for state-of-art materials and ambient pressure conditions are presented in Table 2, together with alternatives. Model predictions on life time in $(Li/Na)_2CO_3$ as the electrolyte have shown that the system derived stack operating conditions (ambient pressure, low P_{CO_2} and P_{H_2}) favour life times well over the projected 25 000 h.

5.2. $LiCoO_2$ cathode

For $LiCoO_2$ cathodes, a life time over 40 000 h is predicted from the model calculations. The performance of $LiCoO_2$ cathodes on bench-scale experiments [4] are comparable to NiO cathodes. However, stack testing with 0.1 m^2 $LiCoO_2$ cathodes shows lower performances (Fig. 3) and it is concluded that for $LiCoO_2$ additional adaptations of the stack technology are required. Stack experiments in the current programme will be carried out with NiO cathodes, making $LiCoO_2$ an option for the longer term.

6. Conclusions

It is concluded that:

- the ECN 3-D stack model describes the stack performance within 10 mV and therefore is a valuable tool for stack design and development;
- the DIRSEP separator plate under development offers possibilities for robust sealing requirements;
- model calculations on NiO cathode and $(Li/Na)_2CO_3$ electrolyte predict life times over 25 000 h under the prevailing system conditions.

Acknowledgements

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References

[1] P.J. Kortbeek and R.G. Ottervanger, *J. Power Sources, 71 (1–2)* (1998) 223.
[2] P.J. Kortbeek, R.G. Ottervanger and A.L. Dicks, *Proc. Electrochem. Soc. 97–4*, pp. 203–211.
[3] R. Fellows, E.W. Sloetjes and R.G. Ottervanger, *J. Power Sources, 71 (1–2)* (1998) 138.
[4] L. Plomp, J.B.J. Veldhuis, E.F. Sitters and S.B. v.d. Molen, *J. Power Sources, 39* (1992) 369.