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Stack networking for system optimisation: an engineering approach

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Abstract

Direct internal reforming molten carbonate fuel cell (MCFC) systems are generally more efficient and more simple than equivalent external reforming systems. It is demonstrated that even better performance and system simplicity can be achieved by connecting the stacks in networks with the cathode flows and/or anode flows in series. The advantages and disadvantages of such networks are presented. Within the EC's Joule III programme, stack networking has been explored for direct internal reforming-molten carbonate fuel cell (DIR-MCFC) systems. This study resulted in the selection of a system which employed anode recycle with the anodes connected in parallel and the cathodes connected in series. It achieved high performance, yet required few system components. In the project 'Advanced DIR-MCFC development', led by BCN (P. Kortbeek and R. Ottervanger, *J. Power Sources, 71 (1–2)* (1998) 223), this system is being assessed with regard to cost, operating window, and controllability. The approach is to design the system to conform to the requirements of ECN's stacks. At the same time the MCFC stack development at ECN (G. Kraaij et al., *J. Power Sources, 71 (1–2)* (1998) 215) is geared towards fulfilment of the requirements in such a system with respect to performance, pressure management and lifetime, so the development of the system and the stack have an iterative character. A novel, patented, stack design with two anode outlet streams has been proposed by BG. It can be used in the chosen system and gives increased power output. © 1998 BG plc. Published by Elsevier Science S.A.

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1. Introduction to the 'Advanced DIR-MCFC' project

The project 'Advanced DIR-MCFC Development' [3] is led by BCN [1] in the Netherlands and is part-funded by the European Commission within the Joule III programme. It is a collaboration between the companies listed above as well as ECN, Stork Alpha Engineering, Royal Schelde, Gaz de France, and Sydkraft. The aim of the project is to develop direct internal reforming (DIR) molten carbonate technology and assess its commercial potential.

The first task in this 3-year project, which began in 1996, was to determine the most promising application for natural gas-powered direct internal reforming-molten carbonate fuel cell (DIR-MCFC) technology by examining various market sectors. It was concluded that combined heat and power (CHP) at a scale of about 400 kW_e represents the most competitive market niche, and that suitable applications include hospitals, hotels, and leisure centres. The main competition in these applications comes from gas engines, though MCFC systems will have the advantages of lower emissions, higher efficiency and the ability to generate high-pressure steam.

This project has an integrated approach with close collaboration between the stack developers and system designers. This is important as stack and system should not be developed in isolation. It will be seen later in this paper how the selection of the system configuration is subject to the requirements of the stacks, and, conversely, how the design targets of the stacks are set by the system requirements.

2. Introduction to series connection

Commercial fuel cell systems will often consist of more than one stack since there is a practical upper limit to the

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size of a single stack. When internal manifolding is used, the need for an even flow distribution limits the height of the stacks. With external manifolding there is still a limit to the stack height, since the stacks need to be easily transported. Cell area is limited by manufacturing considerations and by the fact that gas pressure losses increase with cell length for a given volumetric power density. For these reasons molten carbonate stacks may be limited in practice to 125–300 kW_e. Therefore, since multi-stack systems are anticipated, it is important to consider how they are to be connected, but there is very little in the literature on this subject.

The possibility of connecting the stacks in series with respect to the gas flows has been discussed by Wimer and Williams [4] who show that higher voltages can be achieved by series connection since, in principle, the voltages of the upstream stacks can exceed the Nernst voltage at the outlet of the downstream stack. An example of a system with series connection of external reforming stacks is covered by a patent filed by a consortium of Japanese companies [5]. Series connection of external reforming stacks has also been proposed by ECN [6].

The concept of series connection is shown schematically in Fig. 1, which indicates that the streams between the stacks can be cooled by heat exchangers or they can be mixed with fresh fuel or air. Fig. 1 shows a single-pass flow scheme, but it will be appreciated that when recycle loops are allowed, a very large number of configurations become possible. It will be shown that connecting the stacks in series with respect to the gas flows generally gives higher efficiencies than equivalent parallel-connected systems.

It also gives low per pass fuel and oxidant utilisations in each stack, while not requiring high-temperature recycle blowers. On the other hand, the cumulative pressure losses are likely to be greater in series-connected systems. These advantages and disadvantages are discussed in more detail for MCFC systems in Section 3 and Section 4, which deal with the cathode and anode flows respectively.

3. Series connection of the cathode streams

Series connection of the cathode gas flows does not require anode series connection, so is discussed here independently of the fuel flow configuration.

3.1. Advantages of series connection of cathode flows

3.1.1. Increased stack cooling

Consider a system with series connection of cathodes and no cathode recycle, where the cathode gas is cooled between the stacks. Nearly all the cathode flow passes through each stack and performs stack cooling on each pass. In contrast, in a parallel connected system with no cathode recycle the flow divides upstream of the stacks and only a proportion of the gas flows through each stack. Consequently the cathode flow per stack is less in this parallel connected case than in the series connected case. Thus series connection of the cathodes gives more stack cooling than parallel connection for the same stack inlet and outlet temperatures.

In parallel-connected systems the stack cooling can be enhanced by introducing cathode recycle, but this would reduce the O_2 and CO_2 partial pressures at the stack inlets and have a negative effect on the voltage.

Now, in an internal reforming stack, heat is generated by the electrochemical reaction and removed by the gas flows and by the reforming reaction. Therefore, the enhanced cooling achieved by series connection allows an increase in the current, leading to improved electrical efficiency. That is to say, series connection (without recycle) allows a higher overall fuel utilisation than parallel connection (without recycle) for the same stack inlet and outlet temperatures.

3.1.2. Lower per pass O_2 and CO_2 utilisations

The per pass O_2 utilisation is defined as the total consumption of O_2 in a stack divided by the O_2 flow into the stack. It has been established by ECN that low per pass utilisations are needed to prevent the oxidant concentration from becoming too low at any point in the stacks. This is because it is not possible in practice to achieve a uniform flow distribution in the stack, and the cells that receive the lowest cathode flows can become depleted in the oxidant gases at their outlets if the utilisations are high. Series connection achieves low per pass utilisations without the need for cathode recycle. A parallel connection system, on the other hand, requires much of the cathode outlet gas to be recycled to the inlet to reduce the per pass utilisations, and this probably demands either a high-temperature recycle blower with its additional cost and power consumption, or a low-temperature blower with additional heat exchanger(s). Injectors have also been proposed as a means of driving the recycle, but they are practical only with low recycle rates. A parallel connected system would require a recycle rate of 67% to give the same per pass oxidant utilisations as the downstream stack in a series-connected three-stack system.

3.1.3. Better stack temperature distribution

The higher cathode flow (compared with a parallel connected system without cathode recycle) gives a more even

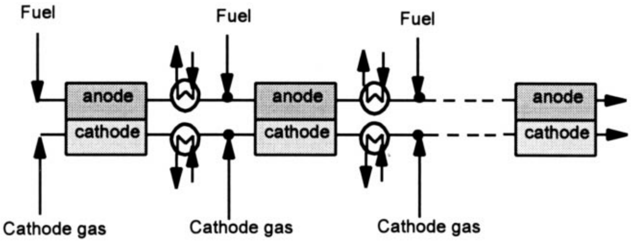

Fig. 1. The concept of series connection.

temperature distribution in the stack and this improves stack performance.

3.1.4. High CO_2 and O_2 concentrations

Series connection gives high CO_2 and O_2 concentrations in the upstream stacks, giving low cell polarisation resistances and high Nernst voltages. Parallel connection with cathode recycle gives lower inlet oxidant concentrations, since the recycled gas is relatively depleted.

3.1.5. Lower carbonate evaporation rates

The rate of carbonate evaporation is inversely proportional to the CO_2 partial pressure, so we can expect the loss of carbonate to be greatly reduced in the upstream stacks where there are high CO_2 concentrations.

3.1.6. Fewer control or trim valves needed

For many arrangements of series connection it is not necessary to put control valves on the cathode inlet streams of every stack.

3.2. Disadvantages of series connection of cathode flow

3.2.1. More difficult stack pressure management

There is a maximum permitted pressure difference between the anode and cathode channels in the stacks. There is also a limit to the pressure difference between the stack and the surrounding atmosphere, and connecting the stacks in series makes it more difficult to satisfy these constraints.

3.2.2. Greater susceptibility to the failure of a stack

Wimer et al. [4] have pointed out that with series connection it may not be possible to shut down one stack without affecting the other stacks in the line.

3.2.3. Increased cathode degradation

The higher CO_2 concentrations in the upstream stack(s) will increase the rate of cathode degradation by NiO dissolution if NiO cathodes are used.

3.2.4. Higher pressure drop requiring a higher duty air blower

Series connection requires the air blower to give a higher pressure lift than does parallel connection.

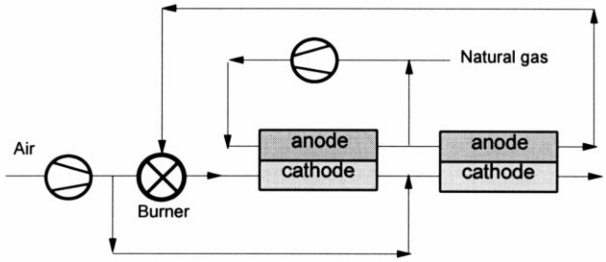

Fig. 2. A two-stack system.

3.2.5. Possible high fuel consumption at hot standby

Fuel consumption is needed at hot standby to compensate for heat losses. In a parallel connected system with cathode recycle, the recycle blower can be used at hot standby to maintain a high cathode gas through-put. This minimises the temperature drop across the stack. Many series-connected systems do not have this advantage.

4. Fuel flow configurations

The issues surrounding the connection of the anode streams are more complicated than those of the cathode flows, and a complete assessment is beyond the scope of this paper.

4.1. In external reforming systems

Series connection of non-reforming stacks gives low per pass fuel utilisations. Another advantage of series fuel flow is that higher voltages are achieved when the stacks are connected electrically in series [4].

4.2. In internal reforming systems

4.2.1. Carbon deposition

Several techniques can be used to prevent carbon deposition from the fuel in internal reforming stacks, but two of the most attractive are anode gas recycle and steam addition. Fig. 2 shows a two-stack system in which carbon deposition is suppressed by recycling anode gas from the outlet of the first stack to the inlet of the same stack. It is, of course, possible to recycle anode gas from the exit of the second stack to the inlet of the first stack, but such schemes are generally less efficient and require the recycle blower to give a higher pressure head.

4.2.2. Thermal management

In internal reforming systems the distribution of the natural gas is critical to the heat balances in the stacks, since the reforming reaction is so endothermic. In Fig. 2 all the natural gas is fed to the first stack so there is very little reforming in the second. Thus the second stack relies largely on the cooling effect of the cathode gas to remove the heat generated by the electrochemical reaction. Consequently the second stack in Fig. 2 has to be much smaller than the first. It can be seen that systems such as this, in which there are both internal reforming stacks and non-reforming stacks, require stacks of different sizes and different designs. Conversely, if half of the natural gas is fed to the inlet of the second stack then both stacks become internal reforming and can be identical in design. However, such schemes have little advantage over equivalent systems with parallel connection of the anode flows.

5. System simulations

Many systems have been simulated within the current project by Stork Alpha Engineering using Aspen, and by BG using a spreadsheet-based simulation tool. These have included systems with 1, 2, 3, and 4 stacks with combinations of series and parallel connection. Other features that have been simulated include cathode recycle, pressurisation, single-pass anode gas flow, anode recycle, the use of an injector to drive the anode recycle, and combinations of internal reforming and non-reforming stacks.

The stack models have been one-dimensional and have employed cell resistance formulae derived by ECN, and have used finite reforming kinetics. The one-dimensional models have been verified against ECN's 3-dimensional model, which in turn has been compared with experimental results.

When comparing the performance of different system configurations it is important to give careful consideration to the parameters that are kept constant. For our system selection exercise we chose to use the same fuel consumption and the same total cell area for each system. We also fixed the stack inlet and outlet temperatures. Thus direct comparisons could be made between the rival configurations. Note that other parameters such as overall fuel utilisation, per pass fuel utilisation, air flow rate, current, and voltage, were not kept constant and varied considerably.

6. The stack requirements and the system selection process

The choice of system configuration is very dependent on the requirements of the stack. The approach within the 'Advanced DIR-MCFC' project has been to develop the stack and the system to meet the requirements of each other. They should not be designed in isolation. In particular, 3-dimensional modelling of internally manifolded stacks by ECN has shown that the per pass utilisations of both the fuel and the oxidant should be kept below certain maximum values, as explained below.

The flow distribution of the cathode gas will inevitably be non-uniform because of engineering tolerances and pressure losses in the manifold channels, with the cells at the top of the stack receiving less flow than those at the bottom. This can lead to depletion of oxygen or carbon dioxide at the outlet of those cells where the flow is least, and cause cell degradation as a consequence. The problem can be overcome by ensuring that the per pass oxidant utilisations are relatively low, and this can be achieved by employing series connection or cathode gas recycle.

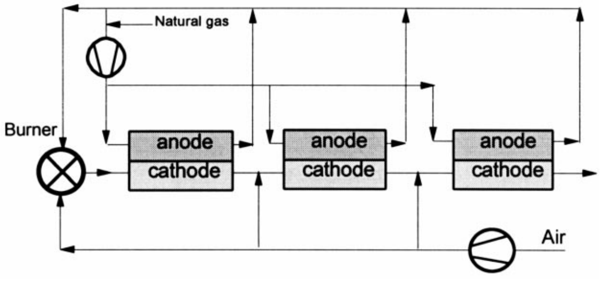

Fig. 3. System 3BG5.

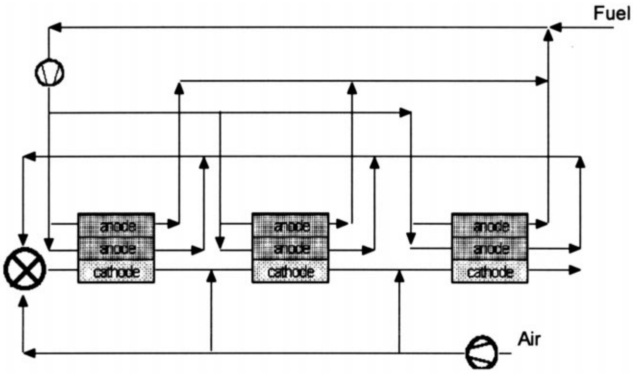

Fig. 4. The 'Smarter' system.

Similarly, for the anode flow, a low per pass fuel utilisation is required in order to prevent the local hydrogen concentration becoming too low at the outlets of some of the cells. For this reason we prefer systems with anode recycle over those with single-pass fuel flow.

Another important stack issue that influences the choice of system is that of pressure differences. Many of the most promising systems have two or three stacks in series with respect to the fuel and cathode gas flows, resulting in stack pressures of typically 200 mbar above ambient. Moreover, large pressure differences between the anode and cathode flows are expected in the stacks. ECN are developing designs to allow the stacks to be tolerant to such pressure differences.

A short-list was made of the leading systems, and these were assessed by a multi-disciplinary team which looked at the issues of performance, simplicity, safety from carbon deposition, 3-D stack simulations, ease of start-up, stack lifetime, pressure requirements, and controllability.

7. The chosen configuration

A system known as 3BG5 was selected for further development and is shown in Fig. 3. It has three stacks in series with respect to the cathode flows and in parallel as far as the fuel flows are concerned. The cathode flows between the stacks are cooled by injecting a relatively small quantity of ambient air which is controlled by inexpensive room temperature valves. There is a hot blower to recycle some of the anode off-gas from the stack outlets to the inlets, while the remainder to the anode exit gas is burnt in the feed air upstream of the first stack.

System 3BG5 combines the advantages of series cathode connection and parallel anode connection. The series cathode connection gives an electrical efficiency about 4 percentage points higher than an equivalent system with the stacks connected in parallel, while the anode flow scheme allows the stacks to be of the same size and design. Moreover, the pressures in the system are less than for similar systems with series connection of the anode flows. Note that there are very few balance of plant components in this system: there are no heat exchangers except any required for fuel pre-treatment, and the series connection has eliminated the need for a cathode recycle blower.

A novel stack concept, known as the ‘Smarter’ stack will now be presented, and it will be shown how it can be used in the configuration given above.

8. ‘Smarter’ stacks

The ‘Smarter’ stack concept is presented in more detail in a separate paper in this journal [7].

In brief, the concept is to manifold the cells so that two anode exit streams are created. One of the exit streams is recycled to the inlet, while the other is passed to the burner. Therefore, we have two groups of cells: one group, known as recycle cells, feeds the anode recycle stream, and the other group, known as exhaust cells, supplies the burner. The two types of cells are mixed so that there is good heat transfer between them. Now, when a high recycle rate is used this specialisation of the cells causes the recycle stream to be substantially less depleted than the exhaust stream, and this leads to high Nernst potentials within the stack. Furthermore, the high recycle rate means that the fresh methane is greatly diluted by the (relatively undepleted) recycle stream and the reforming reaction is spread more evenly across the stack. Thus, the temperature dip near the stack inlet can be reduced or eliminated completely.

Although the Smarter concept requires the recycle blower to give a high flow rate, it is believed that this will not greatly increase the cost of this blower.

Fig. 4 shows schematically how Smarter stacks can be used in the chosen system configuration. As in Fig. 3, the cathodes are connected in series and the anodes in parallel. The only difference is that each stack now has the two types of anode channel.

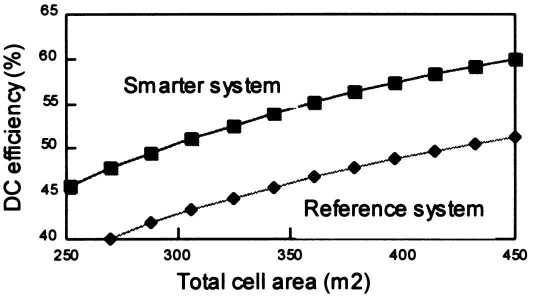

Fig. 5. Stack DC efficiencies for a fixed fuel flow rate of 1 mol/s of CH_4.

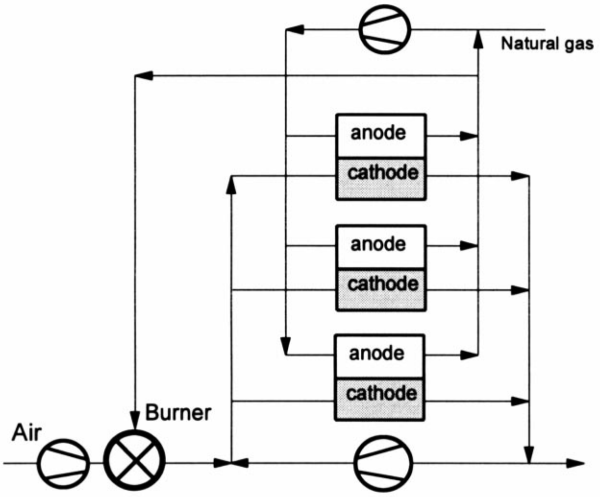

Fig. 6. Reference parallel-connected system.

This system has been simulated with a fixed fuel flow rate and a recycle rate of 80%. Fig. 5 shows how the DC efficiency of the system depends on the total active cell area. A simple parallel-connected system (shown in Fig. 6) has been simulated for comparison. It can be seen from Fig. 5 that the chosen Smarter system gives an efficiency improvement of about 8% compared to the conventional parallel-connected system. About half of this improvement is attributable to the series connection of the stacks, and about half to the inclusion of ‘Smarter’ stacks.

9. Developments within the ‘Advanced DIR-MCFC’ project

9.1. Stack development

The stack development at ECN has included stack tests and experiments with a Li/Na electrolyte (see Ref. [2]).

Reforming catalyst research has continued at BG and has concentrated on a patented nickel based catalyst (see Ref. [8]).

9.2. The hot blower

Stork has assessed the feasibility of the hot recycle blower and has selected a leading manufacturer. Much of the technology of the anode recycle blower is standard, although the seal arrangement will need verification. The sealing issue is important since combustible and toxic gases are recycled.

9.3. The burner

A novel atmospheric pressure homogeneous combustor has been built and tested at BG. In these tests the fuel and air flows were selected to be appropriate to the systems presented in this paper. That is to say, air was supplied to the burner at room temperature while the fuel was pre-heated to the expected stack outlet temperature. The composition of the fuel was varied to simulate a range of overall fuel utilisations and the combustor exit gas was sampled for CO. It was found that good combustion was sustainable for overall fuel utilisations as high as 88%. Tests have also shown that natural gas can be burned directly in the burner, as will be required by the system during hot-standby and low load operation. Further burner development within the project will concentrate on scale-up and is planned at both Shelde and BG.

9.4. Desulfurisation

Gaz de France and BG are investigating desulfurisation techniques for natural gas that do not require hydrogen for hydrogenation of the sulfur-containing compounds. The use of activated carbon at room temperature looks promising. Also under consideration is ICI's 'Puraspec' technique, which involves a single pass through a modified zinc oxide bed.

9.5. Mechanical integration

It has been realised for some time that in a cost-effective system, pipe lengths should be kept as short as possible and the hot component should be located together to minimise heat losses. Therefore, a compact design of the system layout has been produced within the 'Advanced DIR-MCFC' project by Stork Alpha Engineering. The three stacks, the hot blower, and the burner are situated in a mechanically and thermally integrated module which is easily transportable.

9.6. Dynamic simulations

The dynamic behaviour of system 3BGS has been simulated and control strategies have been explored. The consequences of stack degradation and part-load performance have also been assessed. Further dynamic modelling is planned (see Ref. [9]).

10. Conclusions

1. Connecting the stacks in series with respect to the cathode flows improves efficiency and removes the need for a cathode recycle blower.
2. Series connection of the anode streams can also give increased system efficiencies, but for internal reforming systems does not generally allow the stacks to have the same design.
3. The chosen system, to be developed within the 'Advanced DIR-MCFC' project, has three identical stacks which are connected in series with respect to the cathode flows and in parallel with respect to the anode flows.
4. The chosen system with 'Smarter' stacks is 8% more efficient than an equivalent parallel connected system with the same total active cell area.

Acknowledgements

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