

## A novel configuration for direct internal reforming stacks

Richard Fellows

*Gas Research and Technology Centre, Ashby Road, Loughborough, Leicestershire LE11 3GR, UK*

---

### Abstract

This paper presents a stack concept that can be applied to both molten carbonate fuel cell (MCFC) and solid oxide fuel cell (SOFC) internal reforming stacks. It employs anode recycle and allows the design of very simple system configurations, while giving enhanced efficiencies and high specific power densities. The recycle of anode exit gas to the anode inlet has previously been proposed as a means of preventing carbon deposition in direct internal reforming (DIR) stacks. When applied to a normal stack this reduces the Nernst voltages because the recycle stream is relatively depleted in hydrogen. In the concept proposed here, known as the ‘Smarter’ stack, there are two anode exit streams, one of which is depleted, while the other is relatively undepleted. The depleted stream passes directly to the burner, and the undepleted stream is recycled to the stack inlet. By this means high Nernst voltages are achieved in the stack. The concept has been simulated and assessed for parallel-flow and cross-flow MCFC and SOFC stacks and graphs are presented showing temperature distributions. The ‘Smarter’ stacks employ a high recycle rate resulting in a reduced natural gas concentration at the stack inlet, and this reduces or eliminates the unfavourable temperature dip. Catalyst grading can further improve the temperature distribution. The concept allows simple system configurations in which the need for fuel pre-heat is eliminated. Efficiencies are up to 10 percentage points higher than for conventional stacks with the same cell area and maximum stack temperature. The concept presented here was devised in a project part-funded by the EU, and has been adopted by the European Advanced DIR–MCFC development programme led by BCN. © 1998 BG plc. Published by Elsevier Science S.A.

*Keywords:* Internal reforming; Fuel cell systems; Anode recycle

---

### 1. Introduction

Natural gas is widely available and is the preferred fuel for stationary power generation applications. Before it can be converted electrochemically in fuel cell stacks, it must be reformed to produce hydrogen and carbon monoxide. In MCFCs and SOFCs this can take place within the stacks themselves: a process known as internal reforming. A system layout for an internal reforming SOFC system is shown in Fig. 1, in which carbon deposition in the stack is prevented by the recycle of anode exit gas to the anode inlet stream. There is no external reformer or need for water treatment or steam raising, and this system is potentially very simple and cost effective, but suffers from two major problems. Firstly, the anode exit gas dilutes the inlet fuel, thus reducing the open circuit, or Nernst, voltage. Secondly, and more importantly, the reforming reaction is highly endothermic and creates a strong temperature dip near the inlet of the stack. This may result in unacceptable thermal stresses and will reduce the stack performance because the

electrical resistance of the cells increases rapidly when temperature falls.

A number of techniques to reduce the endothermic effect have been proposed, examples being:

1. to perform a significant proportion of the reforming outside the stack [1];
2. to reform in the stack in separate compartments (a process known as indirect internal reforming), or to employ a combination of direct and indirect internal reforming [2];
3. to dilute the incoming fuel by employing a high recycle rate.

The use of high anode recycle rates allows us to keep the system simplicity of Fig. 1, but means that the fuel will be depleted at all points in the stack.

In this paper a further option, known as the ‘Smarter’ stack, is presented. It uses a high anode recycle rate, but overcomes the dilution problem by recycling relatively undepleted anode gas.

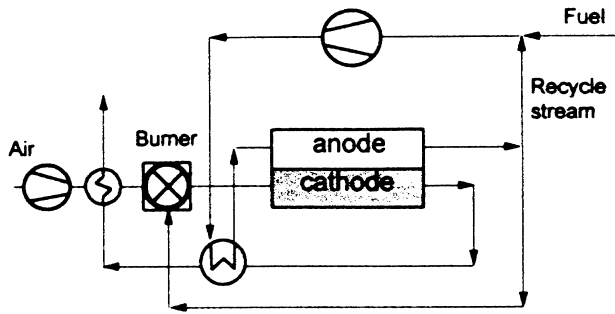


Fig. 1. An SOFC system with a conventional stack.

## 2. The ‘Smarter’ concept

‘Smarter’ stands for separately manifolded anodes rendered to exhaust and recycle. The idea is to have two anode exit streams, one of which is depleted while the other is substantially less depleted. The more depleted stream, known here as the anode exhaust stream, passes directly to the burner, and the other stream, the anode recycle stream, is recycled to the inlet.

One way of producing these two anode outlet streams is indicated in Fig. 2, which shows only the anode gas flows. There are two outlet manifolds which are arranged so that anode gas from alternate cells passes to the exhaust stream, while the other cells feed the recycle stream. Those cells which supply the exhaust stream and recycle stream will be called exhaust cells and recycle cells, respectively. The two types of cell alternate so that there will be good heat transfer from the exhaust cells, which are net generators of heat, to the recycle cells where most of the reforming occurs. Now, it will be seen that if the recycle cells were to receive the same inlet flow rate of anode gas as the exhaust cells, they would be essentially identical; the two exit streams would have the same composition, and the ‘Smarter’ concept would not be achieved. However, consider the case where the recycle cells receive 90%, say, of the total inlet stream. The per pass utilisation of the recycle cells will then be one ninth of that of the exhaust cells. Thus, the recycle stream will be considerably less depleted than the exhaust stream.

The anode gas flow configuration in Fig. 1 consists, topologically, of a feed stream, an exhaust stream, and a recycle loop. The recycle loop is a circuit comprising the recycle stream, the stack inlet stream, the anode channels in the stack, and the stack exit stream. Note that the recycle blower can be placed anywhere in the recycle loop and that the recycle flow rate depends on the speed of the blower, whereas the exhaust flow rate is fixed by the feed flow rate because of the need for an overall mass balance.

In Fig. 2, the flow configuration is topologically identical to that in Fig. 1. The only difference is that the exhaust cells are now outside the recycle loop. This means that the flow to the recycle cells depends on the blower, but that to the exhaust cells does not. To produce the ‘Smarter’ effect it is necessary only that the blower be sized to produce a large

flow rate. To minimise the electrical power consumption of the blower it is necessary to keep the pressure losses in the recycle loop as small as possible. Thus, the anode channels of the recycle cells should be wide because they are in the recycle loop. If the recycle cells and the exhaust cells have the same design then the exhaust cells will have a lower pressure drop and the exhaust stream will consequently have a higher pressure than the recycle stream.

## 3. The advantages of ‘Smarter’ stacks

In this section the merits of ‘Smarter’ stacks are discussed in general terms, while in following sections the concept will be applied separately to MCFC and SOFC stacks of parallel-flow and cross-flow types.

### 3.1. Stack performance advantages

For a stack of a fixed total cell area, in a system with a fixed fuel flow rate, the ‘Smarter’ stack concept has been shown to give an efficiency improvement of about 5% points for MCFCs and about 10% for SOFCs compared to equivalent systems with conventional stacks. The reasons for this improvement are as follows:

- Improved temperature distribution: as state above, the high recycle rate dilutes the inlet fuel giving relatively low methane concentrations at the stack inlet. This greatly reduces the reforming reaction rate there and the reaction is spread more evenly across the stack, producing a more favourable temperature distribution. A significant amount of reforming can occur in the recycle channels near the stack outlet. Any unreformed methane that leaves the recycle cells is, of course, returned to the stack inlet.
- Higher Nernst voltages: the ‘Smarter’ stack gives higher Nernst voltages at both the inlet and the outlet. The inlet of the ‘Smarter’ stack receives anode gas which is relatively undepleted and already substantially reformed giving high Nernst voltages

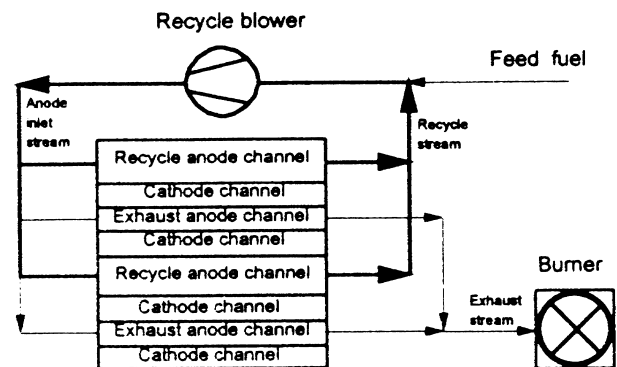


Fig. 2. The ‘Smarter’ concept applied to a parallel-flow stack.

there. In a conventional stack with anode recycle, on the other hand, the inlet hydrogen concentration is actually less than that of the spent exhaust stream. Depleted fuel is experienced by the outlet of only the exhaust cells, whereas in a conventional stack all the cells see spent fuel.

- Lower anode overpotentials: the higher hydrogen partial pressures give reduced cell resistances.
- Increased stack cooling: the higher recycle rate enhances the stack cooling by the anode gas.

### 3.2. System advantages

- Fuel pre-heating not required: the high flow rate of the recycle stream generally makes an additional fuel pre-heater unnecessary:
- Reduced concentrations of higher hydrocarbons: the higher hydrocarbons present in the natural gas feed are greatly diluted by the recycle stream so they are less likely to crack and result in carbon deposition. The pre-reformer, often recommended to convert the higher hydrocarbons [3] in the feed fuel, may no longer be necessary, though this needs to be confirmed by experiment. If the pre-reformer can be omitted then, in the solid oxide case, we may also be able to omit the desulphuriser since the stack itself is relatively tolerant to sulphur [4].
- Option of sensible heat reforming: a sensible heat reformer could be conveniently installed in the recycle stream. The high recycle rate would allow this stream to provide the energy required to perform some or all of the reforming in this external reformer. This idea will not be considered further in

this paper, but it may have advantages, particularly in the MCFC case, since it allows the reforming catalyst to be periodically replaced.

## 4. Molten carbonate

### 4.1. Co-flow

The ‘Smarter’ concept is being developed for co-flow MCFC stacks within the European BCN programme [5,6].

Fig. 3 shows schematically a simple MCFC system with a co-flow ‘Smarter’ stack.

It has been simulated with a one-dimensional stack model which accounts for finite reforming kinetics and includes formulae for cell resistance. The fuel flow is 1 mol/s of CH<sub>4</sub> and the total cell area is 440 m<sup>2</sup>. The recycle rate is 79%. That is to say, 79% of the total anode inlet stream is delivered to the recycle cells. Fifty percent of the cells are recycle cells, and 50% are exhaust cells. The minimum Boudouard safety factor (BSF) occurs at the stack inlet and is 1.02. The BSF is defined according to

$$BSF = \frac{P_{CO_2}}{(P_{CO})^2 K}$$

where  $K$  is the equilibrium constant for the reaction



and  $P_{CO_2}$  and  $P_{CO}$  are the partial pressures of CO<sub>2</sub> and CO for the gas if it were to go to equilibrium with respect to the reforming and shift reactions at the local temperature. Thus, according to the principle of equilibrated gas [7] carbon should not form when the BSF is >1. This method of predicting carbon deposition does not account for reaction kinetics in non-equilibrium mixtures, but will be used here

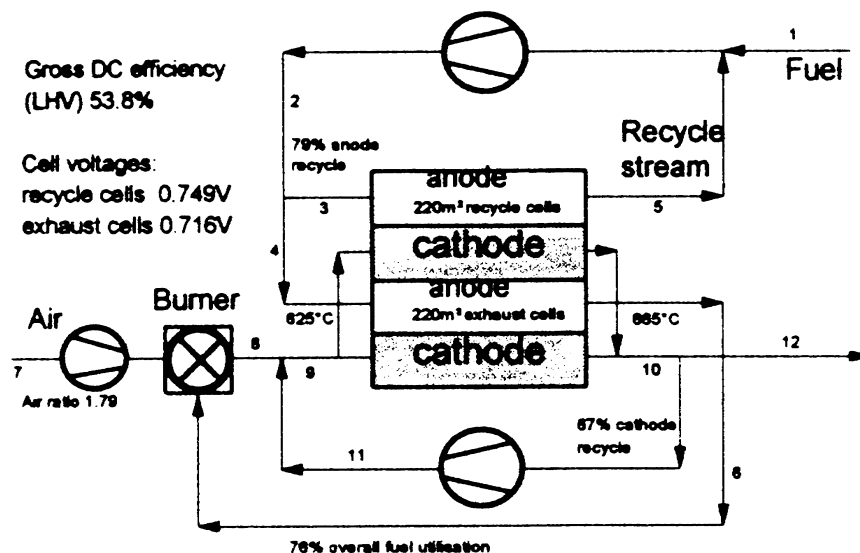


Fig. 3. A co-flow MCFC system with ‘Smarter’ stack.

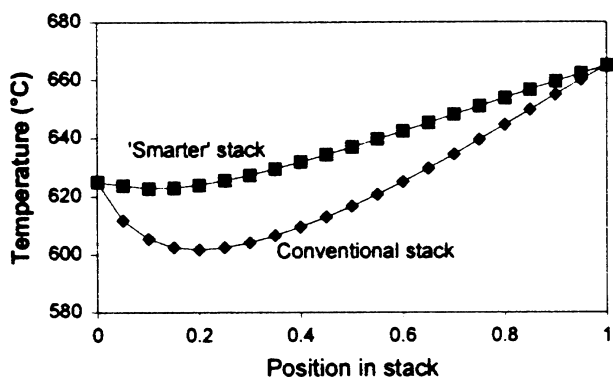


Fig. 4. Temperature profiles for co-flow MCFC stacks.

since relevant experimental data are not yet available.

The DC efficiency of the system is 53.8% on a lower heating value basis. For comparison the system shown in Fig. 3 has been simulated with a conventional stack of the same total cell area and with the recycle rate chosen to give the same minimum BSF at the inlet, and with the same stack outlet temperature. The predicted DC efficiency was 49.1% in the latter case, which is 4.7% less than with the ‘Smarter’ stack. When the additional power consumption of the recycle blower is taken into account the difference may drop to about 4%. An additional improvement in efficiency of about 1% can be achieved with the ‘Smarter’ stack by omitting the catalyst from near the inlet of the recycle cells. Thus, the total advantage of the ‘Smarter’ concept may amount to about 5%. For the system with the conventional stack to achieve this efficiency improvement the total cell area would have to be increased by about 30%.

Fig. 4 shows the stack temperature distributions that are achieved with the ‘Smarter’ stack, and with the conven-

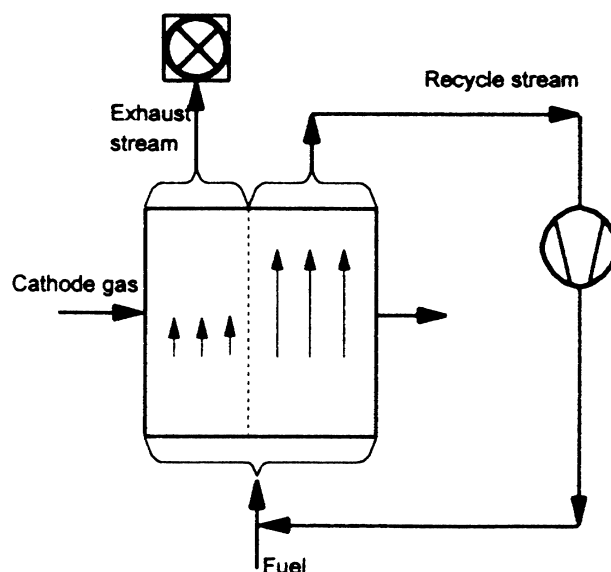


Fig. 5. A method of applying the concept to a cross-flow stack.

tional stack. It can be seen how the ‘Smarter’ concept has reduced the temperature dip by distributing the reforming reaction more evenly across the stack.

Table 1 gives the stream data for the simulation shown in Fig. 3.

#### 4.2. Cross-flow MCFC

The ‘Smarter’ principle can be applied to cross-flow stacks in the same way as described above for parallel flow stacks, with alternate cells manifolded to the recycle and exhaust streams. Alternatively, the stack can be divided as shown in Fig. 5, where the part of the stack furthest from the cathode inlet feeds the recycle stream, while that closest

Table 1

Stream data for co-flow MCFC system with ‘Smarter’ stack

Stream number	1	2	3	4	5	6	7	8	9	10	11	12
	Fuel feed	Anode inlet	Recycle inlet	Exhaust inlet	Recycle outlet	Exhaust outlet	Air feed	Burner outlet	Cathode inlet	Cathode outlet	Cathode recycle	Cathode exhaust
Temperature (°C)	400.00	625.00	625.00	625.00	665.00	665.00	25.00	565.00	625.00	665.00	665.00	665.00
Molar flow rates:												
H <sub>2</sub>			3.52	2.78	0.74	3.52	0.43					
CH <sub>4</sub>	1.00		1.14	0.90	0.24	0.14	0.00					
CO			3.69	2.92	0.78	3.69	0.52					
CO <sub>2</sub>			7.20	5.69	1.51	7.20	3.54					
H <sub>2</sub> O			3.72	2.94	0.78	3.72	1.57	2.00	6.00	6.00	4.00	2.00
O <sub>2</sub>							3.58	3.1	6.26	4.73	3.15	1.58
N <sub>2</sub>							13.45	13.45	40.36	40.63	26.91	13.45
Total	1.00	19.28	15.23	4.05	18.28	6.05	17.03	22.61	58.67	54.09	36.06	18.03
Enthalpies (kW)	-58.00	-3782.00	-2988.00	-794.00	-3707.00	-1669.00	0.00	-1669.00	-2682.00	-1520.00	-1013.00	-507.00

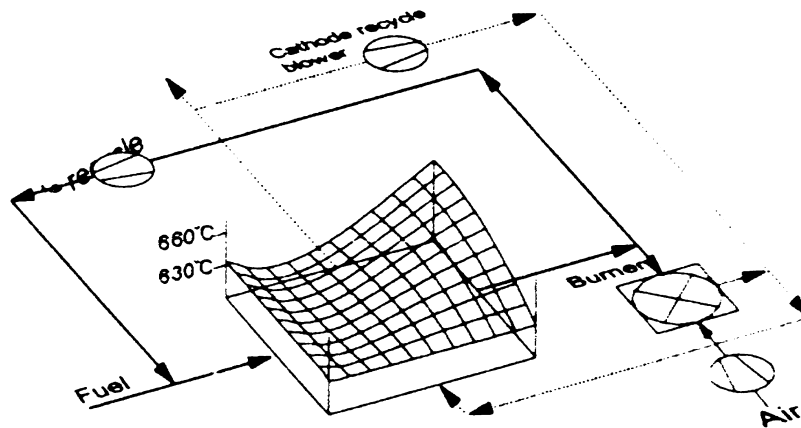


Fig. 6. A conventional cross-flow MCFC system showing stack temperature distribution.

to the inlet delivers anode gas to the exhaust stream. This arrangement does not require complicated manifolding and produces a favourable temperature distribution.

With conventional cross-flow internal reforming stacks a hot-spot occurs at the corner furthest from the anode and cathode inlets as shown in Fig. 6 (see also [8–10]).

With a ‘Smarter’ stack the hot-spot is greatly reduced because some reforming occurs in that corner, and this can be enhanced by omitting the reforming catalyst from the inlet portion of the recycle channels as shown in Fig. 7. Simulations have shown that the system with the ‘Smarter’ stack, shown in Fig. 7, gives an efficiency of 49.6%, whereas the conventional system in Fig. 6 has an efficiency of 43.2% when compared on the same basis with the same maximum stack temperature. The difference between these efficiencies is 6.4%, of which about 5% is attributable to the ‘Smarter’ concept and the rest is due to the catalyst grading.

## 5. Solid oxide

For internal reforming high temperature solid oxide fuel

cells the problem of the endothermic temperature dip is greater than for molten carbonate cells. This is because the reforming rate can be very fast and the cell internal resistance is very sensitive to temperature. For some stack architectures the temperature gradients at the stack inlet can cause unacceptable thermal stresses. The improved temperature distribution produced by the ‘Smarter’ concept can, therefore, be a great advantage for solid oxide stacks.

A disadvantage is that the high recycle rate requires a hot recycle blower which must run at high temperature. Blowers that run at solid oxide temperatures have been produced, but they require that the pressure losses in the recycle loop be kept to a minimum.

The simulations presented here were performed using the formulae for cell resistance and reforming rate suggested by Achenbach [11]. However, current work at BG indicates that the reforming kinetics are more complicated than one would suppose from the relatively simple equations given in the literature. For small stacks, in-plane heat transfer can have a significant effect on the temperature distribution [12]. Here we assume large stacks and do not account for in-plane heat transfer.

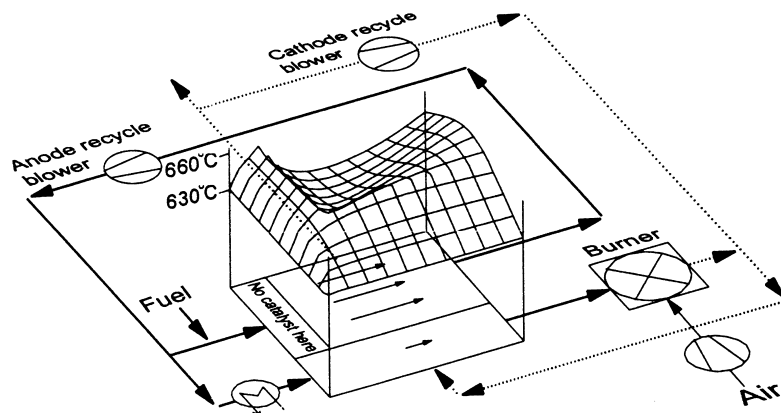


Fig. 7. A cross-flow MCFC ‘Smarter’ system.

Table 2

Stream data for Fig. 8

	Stream number										
	1 Fuel	2 Anode inlet	3 Recycle inlet	4 Exhaust inlet	5 Recycle outlet	6 Exhaust outlet	7 Air feed	8 Heated air	9 Burner outlet	10 Cathode outlet	11 System exhaust
Temperature (°C)	50.00	910.00	910.00	910.00	950.00	950.00	25.00	750.00	910.00	950.00	288.00
Molar flow rates:											
H <sub>2</sub>		24.66	22.93	1.73	24.66	0.47					
CH <sub>4</sub>	1.00	1.34	1.25	0.09	0.34	0.00					
CO		12.59	11.71	0.88	12.59	0.36					
CO <sub>2</sub>		0.35	0.33	0.02	0.35	0.64			1.00	1.00	1.00
H <sub>2</sub> O		1.23	1.14	0.09	1.23	1.53			2.00	2.00	2.00
O <sub>2</sub>							8.64	8.64	8.23	6.64	6.64
N <sub>2</sub>							32.49	32.49	32.49	32.49	32.49
Total	1.00	40.17	37.36	2.81	39.17	3.00	41.13	41.13	43.72	42.13	42.13
Enthalpies (kW)	-74.00	-803.00	-747.00	-56.00	-729.00	-557.00	0.00	925.00	368.00	381.00	-544.00

5.1. Co-flow SOFC

Fig. 8 shows an SOFC system containing a co-flow ‘Smarter’ stack. The air is pre-heated by a heat exchanger and further heated by the burner which is located upstream of the stack. The burner may alternatively be located downstream of the stack but the heat exchanger would then be required to heat to a higher temperature and simulations have shown that there would not be a significant improvement in performance. Table 2 gives the stream data from the simulation of the system in Fig. 8. A recycle rate of 93% is assumed and the air is pre-heated to 750°C. One in every three cells is a recycle cell and the minimum BSF is 1.17. The predicted DC efficiency is 58.4%.

An equivalent system with a conventional stack would still require a high recycle rate to prevent an unacceptably large temperature dip in the stack. An efficiency of 46.9% is predicted for such a system with the same stack outlet temperature.

5.2. Cross-flow SOFC

Fig. 9 shows a cross-flow equivalent of Fig. 8. The same anode manifolding arrangement is assumed as for the co-flow case. This gives a temperature distribution that is much smoother than is given by normal cross-flow SOFC stacks. The predicted efficiency is 54.2%, which is a little less than for co-flow, and an equivalent system with a conventional stack would give about 41.7%.

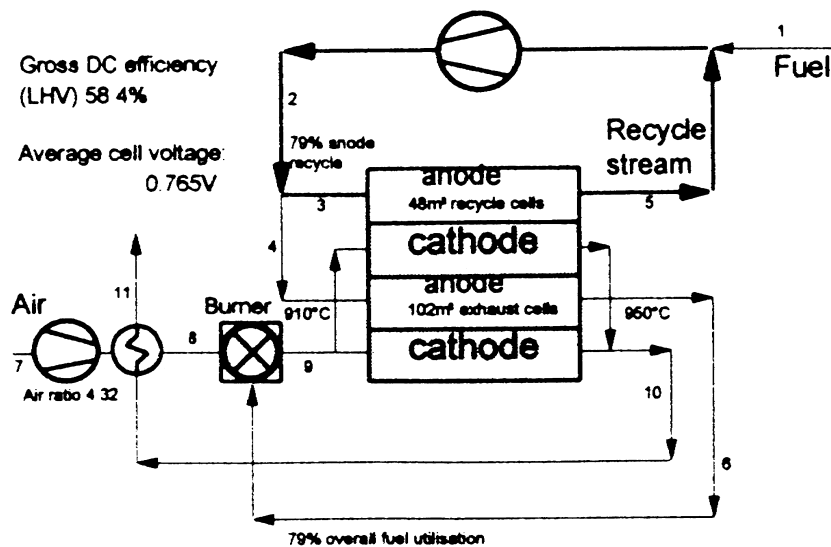


Fig. 8. A co-flow SOFC system with a ‘Smarter’ stack.

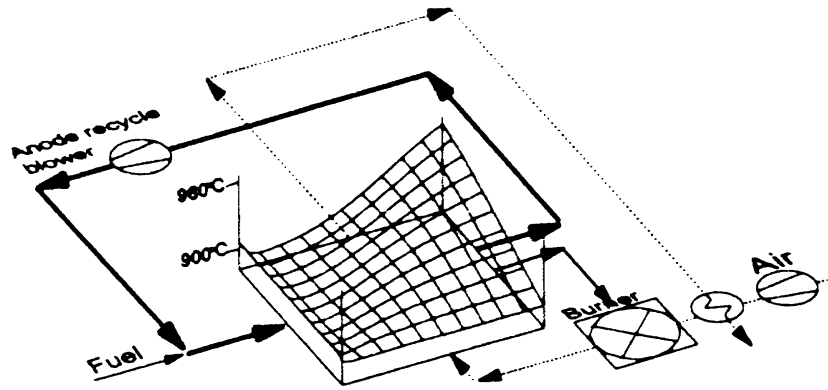


Fig. 9. A cross-flow 'Smarter' SOFC system.

## 6. Conclusions

1. The concept of recycling relatively undepleted anode gas can be applied to direct internal reforming co-flow and cross-flow stacks of both molten carbonate and solid oxide fuel cell types.
2. The temperature dip in the stack is greatly reduced or eliminated, and an efficiency increase of up to 10% is expected.
3. A high temperature recycle blower is needed, but the requirements for fuel processing and pre-heating are reduced.

## Acknowledgements

The MCFC stack calculations have used cell resistance equations supplied by ECN [13].

## References

- [1] A.L. Dicks, *J. Power Sources*, 61 (1996) 113.
- [2] A. Sasaki, T. Shinoki and M. Matsumura, *1996 Fuel Cell Seminar*, Orlando, FL, 1996.
- [3] E. Achenbach, E. Riensche and G. Unverzagt, *1st Eur. Solid Oxide Fuel Cell Forum*, Lucerne, Switzerland, 1994, pp. 153–162.
- [4] A.J. Appleby and F.R. Foulkes, *Fuel Cells Handbook*, Van Nostrand Reinhold, New York, p. 585.
- [5] R. Fellows, E.W. Sloetjes and R. Ottervanger, *J. Power Sources* 71 (1–2) (1998) 138.
- [6] P.J. Kortbeek and R.G. Ottervanger, *J. Power Sources* 71 (1–2) (1998) 223.
- [7] G.H.J. Broers and B.W. Treijtel, *Adv. Energy Conversion* 5, (1965) 365.
- [8] K. Aasberg-Petersen, P. Simonsen, P.S. Christensen, S.K. Winther, E. Jørn, C. Olsen and E.J. Jensen, *1996 Fuel Cell Seminar*, Orlando, FL, 1996.
- [9] Chindemi, G. Belmonte and A. Maladrino, *1st Eur. Solid Oxide Fuel Cell Forum*, Lucerne, Switzerland, 1994, pp. 363–372.
- [10] T. Kahara, M. Kunikata, M. Takeuchi, S. Takashima, T. Komo and H. Fujimura, *1994 Fuel Cell Seminar*, San Diego, CA, 1994, pp. 222–229.
- [11] E. Achenbach., *J. Power Sources*, 49 (1994) 333.
- [12] C. White, P. Jackson and N. White, *1st Eur. Solid Oxide Fuel Cell Forum*, 1994, Lucerne, Switzerland. pp. 267–276.
- [13] N.J.J. Dekker, *1994 Fuel Cell Seminar*. San Diego, CA, 1994, pp. 156–159.