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Optimisation of the cell shape for industrial MCFC stacks

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Abstract

The aim of this work is to compare Molten Carbonate Fuel Cell (MCFC) stack behaviour when square and rectangular cells are used, and to discuss the optimisation of their operating conditions. In particular, experimental data have been collected for a stack with square cells of 0.1-m^2 area to validate a stack simulation model; then, the effects of area increase of up to 0.75 m^2 have been predicted, proving the advantage of working with rectangular cells in order to obtain a better temperature and pressure drop management. Moreover, the operating conditions of a rectangular stack have been analysed and optimised when reformed natural gas, as well as coal gas, are used as fuel. The results presented and discussed have been used to guide design and operation of a rectangular stack constructed by Ansaldo Ricerche (ARI).

Keywords: Molten carbonate fuel cells; Rectangular shape; Operating condition optimisation

1. Introduction

Fuel cell systems are a promising technology for electrical energy production with low environmental impact. In particular, Molten Carbonate Fuel Cells (MCFC) represent an interesting fuel cell family under study at the pre-commercialisation level.

The basic technology considered here results from industrial experience of Ansaldo Ricerche (ARI) with a MCFC development process [1,2] carried out in cooperation with the group working at the Department of Environmental Engineering of Genoa University on fuel cell system simulation.

The scale-up has been extended to test stacks with cross-flow feeding system based on external manifolds increasing the cell area up to about 0.75 m^2. Experimentation at this size was successfully carried out with a square stack [3]. Recently, tests on a new stack configuration, based on a rectangular shape to optimise temperature and pressure drop management, have been carried out. The temperature has to remain within specific constraints during stack operation to avoid possible cell material component damage, and pressure drops have to be controlled to avert gas cross-over between anode and cathode, as well as leakage across manifold seals.

The aim of this work is to compare stack behaviour when square and rectangular cells are used and discuss the optimisation of their operating conditions. Experience has shown that a detailed simulation, coupled with analysis and investigation of experimental phenomena, are the best guides to optimising this new technology solution; thus, the comparison is based on simulation results obtained by means of a theoretical model, which takes account of mass, energy and momentum balances. The electrochemical kinetics in the model are based on a semi-empirical relationship where phenomenological parameters have been calibrated from experimental data collected from a square stack tested in 1998.

2. Theoretical model

The three-dimensional theoretical model, developed by the authors and presented elsewhere [4], takes account of mass, energy and momentum balances at cell and stack level; the corresponding computational code has been developed in Fortran language.

Moreover, in the model, like the electrochemical reactions, the water gas shift reaction is also evaluated; the latter occurs on the anodic side and has been assumed to

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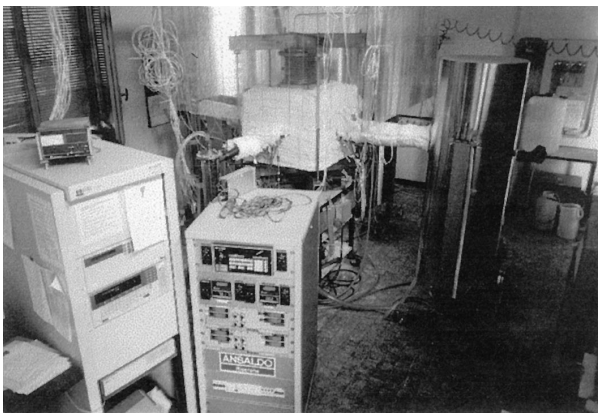

Fig. 1. Photo of the square stack with 14 cells of 0.1 m^2 of area during the operation.

be at thermodynamic equilibrium on the basis of experimental experience.

The electrochemical cell performances are evaluated on the basis of an electrical resistance R, which locally accounts for voltage losses ΔV with respect to the thermodynamic voltage E_0, assuming a linear dependence of voltage as a function of current density J:

$$\Delta V = E_0 - RJ. \quad (1)$$

The resistance R has been written as a semi-empirical relationship [4] obtained starting from a theoretical approach, and takes account of the partial pressure p_i of the chemical species i and of the solid local temperature T:

$$R_{\text{tot}} = \frac{A e^{E/T}}{\prod_i p_i^{\beta_i}} + c_{iR} + D e^{F/T}. \quad (2)$$

Where c_{iR}, is a constant related to the ohmic resistance of the contacts; β, E and F are function of the chemico-physical properties of the cell; and A and D are phenomenological coefficients evaluated by experimental data as shown in the following.

The first factor of Eq. (2) gives the polarisation resistance, whilst the sum of the remaining two gives the internal resistance.

This simplified model well describes cell behaviour if the reactant utilisation factors are not too high and diffusion is not limiting in the porous electrode. In order to take account also of diffusion phenomena, a correction coefficient related to the limiting current has been introduced in the model in terms of a constant value identified on the basis of experimental data (see Section 3).

The simulation results show the distribution of gas and solid temperatures, electrical current density, Nernst voltage, polarisation, internal resistance, pressure drops, and compositions and flow rates of the gaseous streams on the plane of each cell of the stack.

3. Model validation

An initial model calibration was made on the basis of specific test series on single square cells; good agreement was obtained between simulation and experimental results [4] working at a low utilisation factor.

Furthermore, the eventual stacks, using the same basic technology and materials, but in which the matrix thickness has been increased in order to improve mechanical resilience, were expected to operate at high utilisation factors. Thus, in the light of these considerations, a new detailed validation of the electrode model seemed necessary.

With reference to Eq. (2), the parameters β, E and F, related to chemico-physical proprieties of the cell, have not been changed, and also c_{iR} has been assumed equal to the value identified to simulate a previous similar stack with good results [4]; the other coefficients and limiting diffusive effects have been evaluated by considering the experimental data collected from a new stack.

The latter, shown in Fig. 1, consisted of 14 square cells of 0.1 m^2 of area. This stack technology was based on an externally manifolded cross-flow feed system and standard components [1]; the anodes were a Ni/Cr alloyed powder, the cathodes a lithiated nickel oxide structure and the electrolyte a lithium carbonate and potassium carbonate mixture. The stack has been tested during 1998 and operated for about 2200 h with more than 2000 h on load.

D has been calculated on the basis of the experimental internal resistance measured by means of the current interruption method [5], whilst A and the correction parameter due to diffusion have been identified by fitting experimental voltage/current density curves.

Finally, the parameters used to simulate stack behaviour related to Eq. (2) are the following: $\beta = 0.667$, $A = 7.4 \times 10^{-7}$ Ω cm^2 atm, $c_{iR} = 0{,}3$ Ω cm^2, $D = 8.4 \times 10^{-5}$ Ω cm^2, $E = 11\,400$ K, $F = 6596$ K.

Assuming the above values, good agreement between measured and calculated electrochemical performance has been obtained, as shown, for example, in Fig. 2 for the

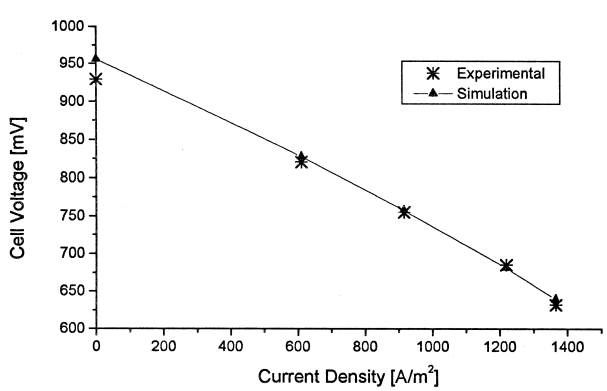

Fig. 2. Comparison between measured and calculated average stack voltage with reference to the tested square stack.

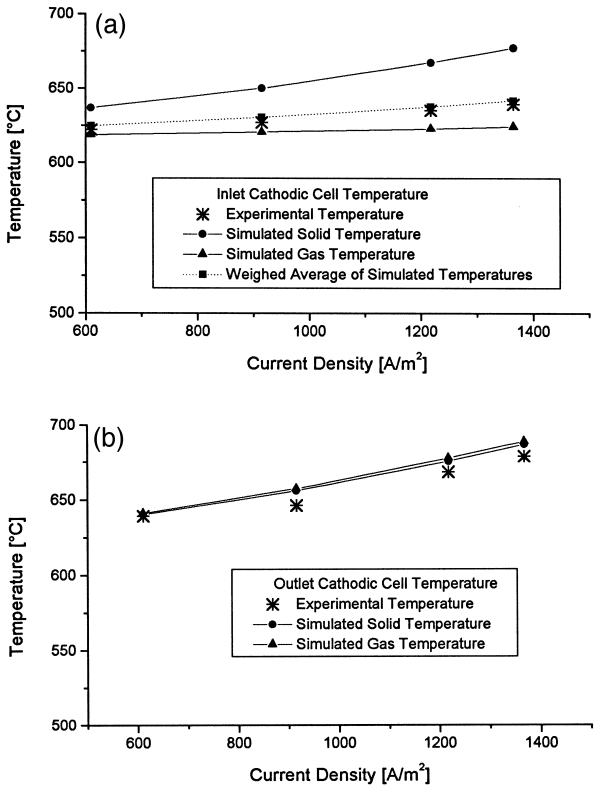

Fig. 3. (A and B) Comparison between measured and calculated temperatures respectively at cathodic gas inlet and cathodic gas outlet for the tested square stack.

square stack with the following operating conditions — fuel composition: H_2 21.2%, CO_2 18.7%, N_2 36.7%, H_2O 22.4% (total flow rate 3.63 N m^3/h); oxidant composition: CO_2 9.8%, N_2 77.4%, O_2 12.8% (total flow rate 21.9 N m^3/h); atmospheric pressure.

Agreement between measured and calculated temperatures has been verified. Fig. 3A and B shows, respectively, the temperature at the inlet and outlet of the cell along the oxidant flow direction under the same operating conditions as in Fig. 2. The cell temperature has been measured by thermocouples placed in the cathodic gas distributor, so partially in contact with the solid and partially with the gas. The simulation results show that, at the cell outlet, where the thermal regime approaches equilibrium, the calculated gas and solid temperatures are very similar and fit the experimental data well, whilst at the cell inlet, experimental temperature fits very well with an average temperature between solid and gas calculated according to the thermocouple position (see Fig. 3A and B).

On the basis of the good agreement obtained, the validated model has been used to predict and analyse the scale-up of stacks with bigger cell area and different shape.

4. From square to rectangular shape

The scale-up process of MCFC technology has been conducted to test stacks with increasing cell area of up to about 0.75 m^2, following the successful experiments carried out with a square shape stack [3]. However, to ensure proper stack behaviour, it is necessary to take account of several operating constraints [6] to avoid cell material component damages or cross-over and leakage problems (see Table 1). The simulation results showed that the temperature and the pressure drops can be too high locally when a square stack of the above size is fed with a reformed natural gas and the working point is set up at a current density higher than 1350 A/m^2.

Fig. 4A and B shows the calculated maps of cell temperature and pressure drops of cathodic gas for a hypothetical stack with 0.75 m^2 square cells working at a reference current density of 1500 A/m^2 and 3.5 atm (fuel composition: H_2 57.1%, CO_2 27%, N_2 14.3%, H_2O 1.6%; anodic total flow rate: 16.18 N m^3/h; oxidant composition: CO_2 7.2%, N_2 59.2%, O_2 10%, H_2O 23.6; cathodic total flow rate 243.14 N m^3/h).

The maximum cell temperature derived is 1018 K, whilst the maximum permissible is about 973 K. Moreover, both the maximum cell differential pressure (34.9 mbar) and pressure drops of cathodic gas (35 mbar) are higher than the advisable values.

As the gas inlet temperature is already at the minimum value of 853 K, to reduce cell temperature further, a higher cathodic total flow rate could be used, but this would lead to further adverse pressure drops if the operating pressure is not increased; one solution is to use a rectangular stack, where cell direction traversed by the cathodic gas is shorter than on the anodic side.

A parametric analysis has been carried out to identify the optimum edge length to be traversed by cathodic gas, whilst maintaining constant effective cell area (about 0.74 m^2 on 0.75 m^2 of total area). Fig. 5 shows that, with a length below 67 cm, the limit of the cathodic pressure drop is 20 mbar. In order to ensure a sufficient safety margin and to reduce the temperature with increasing cathodic flow rate, a length of 61 cm has been chosen. In this case,

Table 1
Operating condition constraints for stack safe running

Maximum cell local temperature	$\leq$ 973 K	To avoid cell material component damage
Minimum feeding gas temperature	$\geq$ 853 K	To avert electrolyte solidification
Maximum cell cross pressure	$\leq$ 20 mbar	To avert gas cross-over between anode and cathode
Maximum cathodic gas pressure drop	$\leq$ 20 mbar	To avert leakage across manifold seals

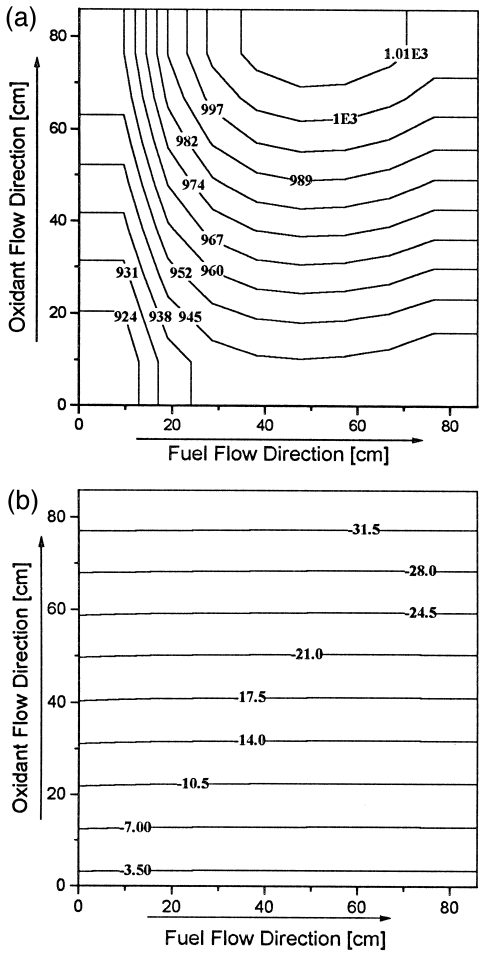

Fig. 4. (A and B) Calculated distribution of solid temperature [K] and pressure drops of cathodic gas [mbar] on the cell plane of an hypothetical square stack with 0.75 m^2 working cells.

the side traversed by anodic gas is 121 cm, with a gas pressure drop of about 3 mbar.

A stack of 15 cells with the above geometry has been constructed and tested in a test facility at the

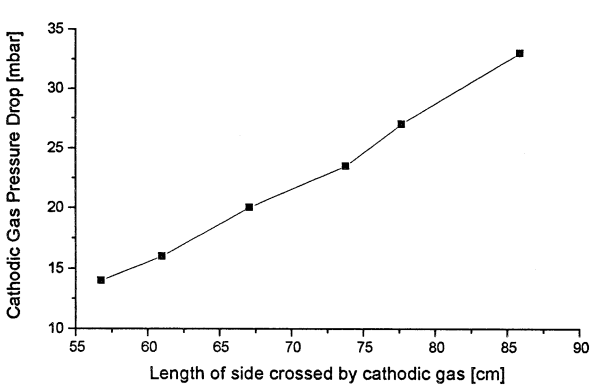

Fig. 5. Calculated total pressure drop of cathodic gas for different length of the cathodic side.

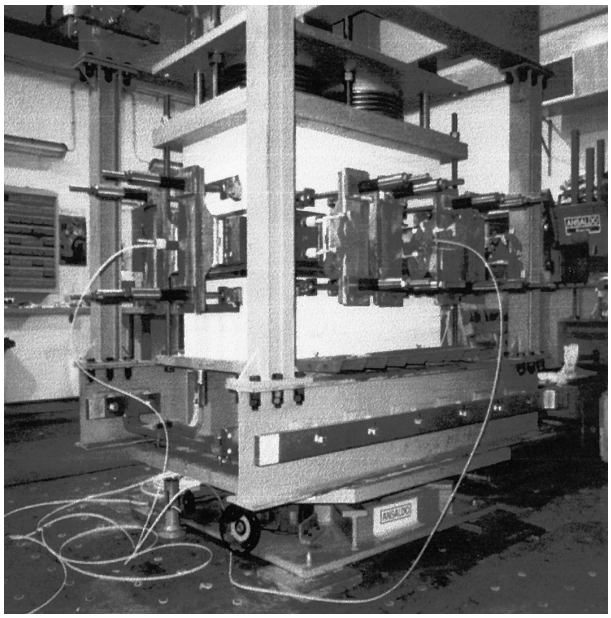

Fig. 6. Photo of the rectangular stack with 15 cells of 0.75 m^2 of area.

PEP/IBERDROLA site of Guadalix (Spain). A photograph of this stack is shown in Fig. 6.

In the following paragraph, the optimised operating conditions for a similar stack are discussed using a feed of reformed natural gas, as well as coal gas.

5. Operating condition optimisation for a rectangular stack

5.1. Reformed natural gas feed

On the basis of the results shown in Section 4, the maximum temperature over the cell plane has been reduced by feeding a cathodic total flow rate that was increased by 20%.

In this way, the maps reported in Fig. 7A and B have been calculated for stack cell temperatures and cell differential pressures.

Fig. 7A always shows the same temperature distribution, but the maximum value is now much lower (about 974 K). Moreover, both the maximum cell differential pressure (about 18 mbar) and the cathodic pressure drop (about 20 mbar) are acceptable, as shown in Fig. 7B.

Fig. 8 shows the effect of cathodic total flow rate on maximum cell temperature: a flow rate increase greater than 20%, apart from implying too high pressure drops, does not yield significant temperature reduction.

Another possible expedient to reduce cell temperature could be anodic gas diluted with an inert component. Even if for a working stack integrated in a power plant, where the anodic gas has a fixed composition related to the

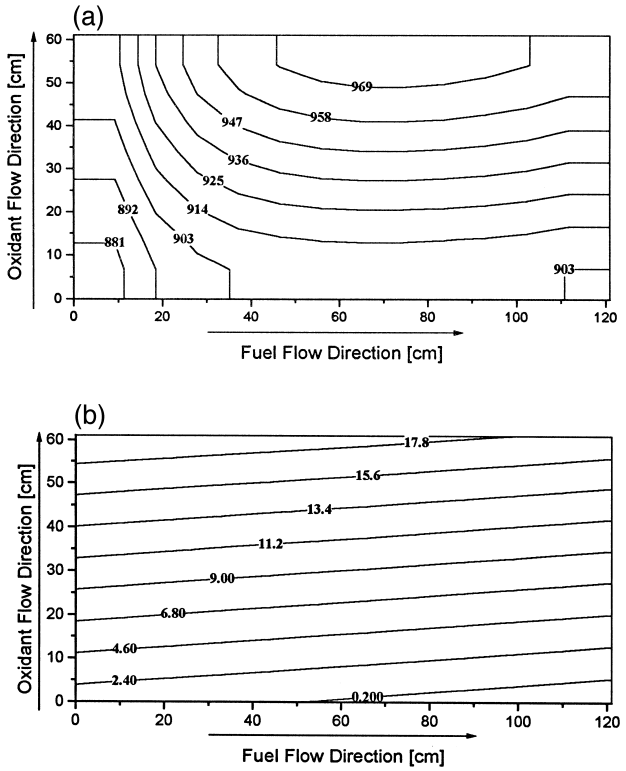

Fig. 7. (A and B) Calculated distribution of solid temperature [K] and cell cross pressure [mbar] on the cell plane of the rectangular stack when reformed natural gas is fed and operating conditions are optimised by means of cathodic flow rate increasing.

chosen fuel processor system, this solution can not be considered; in this case it has been analysed because the test facility at Guadalix, where the rectangular stack has been tested, allowed a gas blood of N_2.

Calculations show that it is sufficient to increase the N_2 flow rate to about 40 N m^3/h to obtain an acceptable stack maximum temperature of 973 K without increasing the cathodic total flow rate (see Fig. 9).

In this case, the temperature distribution in the stack cell plane is slightly different because of the higher thermal inertia of the anodic gas; the maximum temperature is

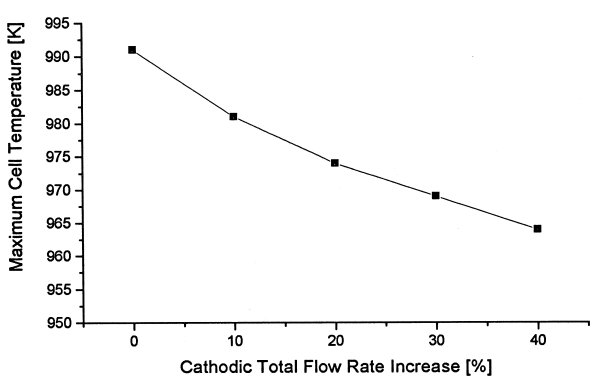

Fig. 8. Calculated maximum temperature in the rectangular stack for different cathodic flow rates when natural gas is fed.

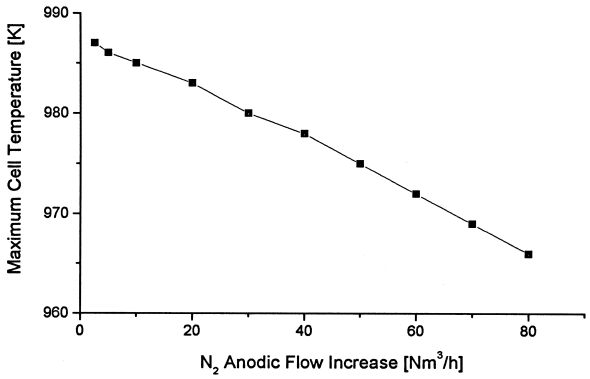

Fig. 9. Calculated maximum temperature in the rectangular stack for different anodic N_2 flow rate when reformed natural gas is fed.

then located nearer the gas outlet. Moreover, this choice does not involve either a high anodic pressure drop, maximum is 8 mbar, or high cell differential pressure, maximum is 16 mbar (see Fig. 10B).

Finally, Fig. 11 compares the voltage/current density curves calculated in the three cases studied.

In terms of performance, the model considered associates the dilution of anodic gas composition by nitrogen with a higher equilibrium thermodynamic voltage, whilst the increase in the cathodic gas flow rate, at constant

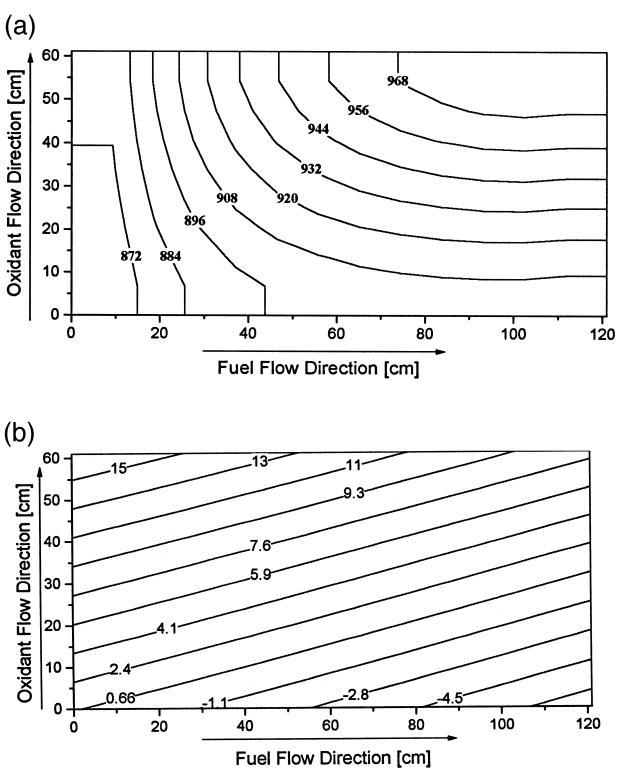

Fig. 10. (A and B) Calculated distribution of solid temperature [K] and cell cross pressure [mbar] on the cell plane of the rectangular stack when reformed natural gas is fed and operating conditions are optimised by means of dilution of anodic composition adding N_2.

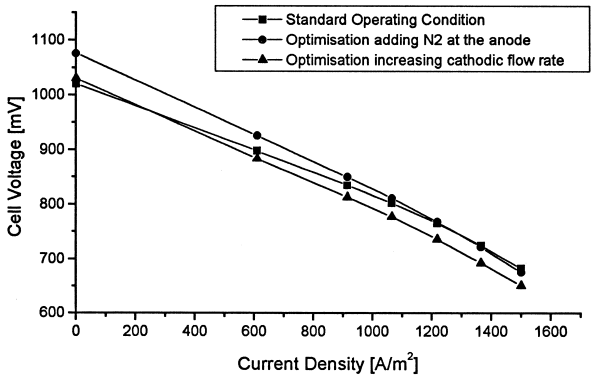

Fig. 11. Comparison of the rectangular stack performance calculated under standard operating conditions and in optimised condition when natural gas is fed.

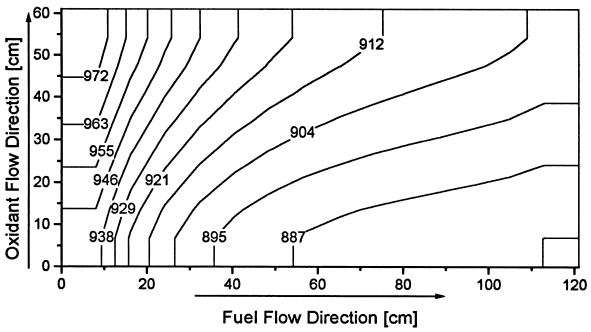

Fig. 13. Distribution of solid temperature [K] in the cell plane of the rectangular stack when coal gas is fed and operating conditions are optimised by means of lowered current density.

oxidant composition, does not have significant effect on the electrochemical performance. However, in both cases, although the electrical resistance is higher than for the unoptimised case due to the lower operating temperature, the strategy is preferable because the present satisfactory performance is compatible with safe stack operation.

5.2. Coal gas feeding

The stack behaviour when coal gas is used has been analysed due to the present interest in environmental applications, which allows energy production starting from different fuel types [6].

Coal gas is typically lean in hydrogen and characterised by a high CO content. MCFC stacks can operate with this type of gas compositions; in fact, their high operating temperature allows the use of non-noble catalysts, which are insensitive to certain fuel contaminants, like CO, which poison other cells. In particular, stack operation using a fuel with CO concentration up to 62% have been successfully carried out by ARI in a 100-kW MCFC plant; those experimental results are at the moment under scrutiny [7].

Furthermore, CO is directly converted in these cells by virtue of the water gas shift reaction $CO + H_2O \leftrightarrow H_2 + CO_2$, thus, supplying additional H_2 for the electrochemical reaction.

As the water shift reaction is exothermic, with this fuel thermal conditions are more critical. In particular, the maximum temperature is always too high. Reducing the gas inlet temperature to 853 K, and increasing the cathodic total flow rate as much as allowed by pressure drop constraints, still creates too high temperature that is up by 30%. It is not relevant to dilute the anodic composition in this case, because the aim of this analysis is to study the behaviour with this specific anodic gas.

In the light of these results, a new operating point has been proposed at lower current density, maintaining a reasonably high cathodic total flow rate.

Fig. 12 shows that at the selected operating pressure, the maximum current density achievable in respect of present temperature constraints is about 1000 A/m^2.

Fig. 13 shows the optimised temperature distribution in the plane of stack cells (fuel composition: H_2 10%, CO 22%, CO_2 0.4%, N_2 3%, H_2O 64.6%; anodic total flow rate: 26.3 N m^3/h; cathodic total flow rate 364.4 N m^3/h; operating pressure 3.5 atm).

The temperature distribution is different with respect to the cases previously presented; this is due to the exother-

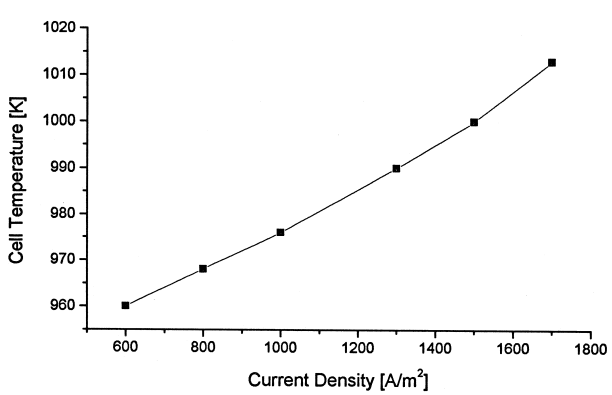

Fig. 12. Calculated maximum temperature in the rectangular stack at different current densities when coal gas is fed.

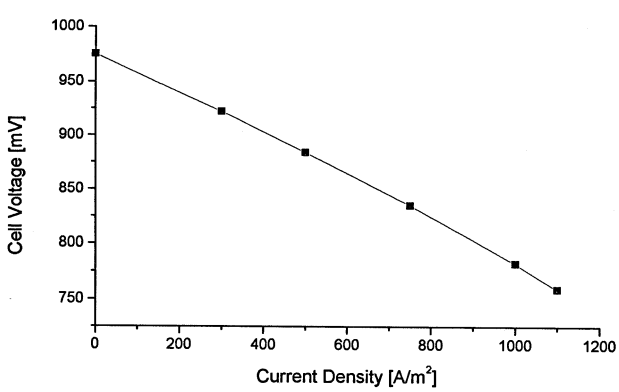

Fig. 14. Calculated performance curve for the rectangular stack in the optimised operating condition when coal gas is fed.

mic shift reaction that transfers the maximum heat near the anodic gas inlet, where most CO reacts.

Fig. 14 shows a voltage/current density curve evaluated at the above operating conditions.

These results show that operation of the MCFC stack using coal gas as anodic fuel is possible and gives good performance, provided that an appropriate operating point is chosen.

6. Conclusions

MCFC stack behaviour has been theoretically analysed on the basis of experimental data. In particular, cell shape has been discussed, which indicates the advantages of working with rectangular cells where the cell direction traversed by the cathodic gas is shorter than the anodic side. With such geometry, it is possible to feed the stack at greater flow rate of oxidant without achieving too high a pressure drop; additional cell cooling occurs, thanks to fresh cathodic gas.

By means of a stack model validated with experimental data, operating conditions have been optimised for a rectangular stack with reference to different fuels, namely, reformed natural gas and coal gas. In the first case, safe operating constraints can be satisfied by increasing the cathodic total flow rate or diluting the anodic composition with inert gas. In the second case, the exothermic water shift reaction changes the thermal operating regime, requiring modification of the working point.

All the considerations presented have been of help designing the MCFC stack with 15 rectangular cells of 0.75 m^2, just tested in Spain following guidance from these simulation results; consequent experimental data are now under study.

Acknowledgements

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