Heat and Mass Transfer in a Molten Carbonate Fuel Cell*

(Performance and Temperature Distribution in a Cell Stack)

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A large-scale stack model of a molten carbonate fuel cell was developed for evaluating the variation of cell performance and temperature in the cell-stacking direction. A numerical method for the performance and three-dimensional temperature distribution in the stack was proposed and examined. For the temperature distribution, calculated results, taking the heat transfer at the end plate surface into account, were compared with the measured temperature distribution of the cell stack with good agreement. For several stacks composed of 12 cells with different gas flow configurations, the total stack performance and the temperature distribution were investigated analytically. The temperature distribution of the cell becomes more uniform due to the effect of heat exchange in the stacking direction.

Key Words: Molten Carbonate Fuel Cell, Temperature Distribution, Numerical Analysis, Electrochemical Reaction, Heat Transfer, Stack Performance

1. Introduction

The molten carbonate fuel cell (MCFC), known as a second-generation fuel cell, has been anticipated to be an efficient and practical energy generation system. Efforts have been made to enlarge the cell capacity through a larger scale and higher stacking of the cell.

The MCFC works at a high temperature of 650°C and produces heat in the power generation mode due to resistance, polarization, and so forth incidental to transfer of ions in the electrolyte plates and electrons in electrodes during the electrochemical reaction. This heat causes an increase of the cell temperature which, in turn, promotes the electrochemical reaction; then the reaction and temperature influence each other. Additionally, the cell temperature has a close relation to thermal stress, vaporization of electrolytes, and corrosion of the cell component in addition to the performance. Accordingly, the cell temperature must be kept within a suitable range for good performance,

lifetime, and reliability. This requires technologies for the cooling and temperature analyses in connection with the cell reaction.

There have been a few investigations to date on reaction and temperature distribution in the fuel cell relating to heat and mass transfer. Gidaspow and Baker⁽¹⁾ performed a heat conduction analysis under such assumptions as a constant heat generation rate accompanying the electrochemical reaction within the cell. They obtained a temperature distribution for the coflow-type fuel cell. Wolf and Wilemski⁽²⁾ obtained a temperature distribution by taking the cell reaction into consideration using a numerical analysis with a porous electrode model. Spradlin⁽³⁾ also studied current density and temperature distribution by dividing a cell into parts for various flow types. Ito et al.⁽⁴⁾ analyzed the heat and mass transfer within the cell by treating it as an electrochemical reactor from the viewpoint of irreversible thermodynamics. Watanabe and Nikai⁽⁵⁾ investigated the effects of heat transfer, gas flow rate and so forth upon the temperature distribution by treating the fuel cell as a heat exchanger and comparing experimental results of the model cell with calculated results. Recently, the authors examined effectiveness and problems in the proposed analytical model and method by comparing results of measurements and analyses for temperature distribu-

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tion in the cell during power generation⁽⁶⁾. But, in all the fuel cell models mentioned above, no treatment was applied for the cell stack. As the cell stack height becomes taller in the future, importance should be attached to the effects of temperature distributions within the plane or in the cell- stacking direction, as well as flow distribution upon the total stack performance. Therefore, a 3-dimensional temperature distribution analysis for the stack cell is needed to realize longer lifetime and higher reliability through development of a stack structure having a uniform temperature distribution.

In this report, the authors propose a model and a method to analyze stack performance and temperature distribution as a cell stack, based on the temperature distribution analysis model⁽⁶⁾ coupled to the electrochemical reaction for a single cell. Comparisons are made between analytical and measured results for the temperature distribution. Also, the effects of a combined cell structure with different directions of gas streams upon the stack performance and the temperature distribution are studied by using the analytical model and method.

2. Structure of Cell Stack

An example stack structure of the MCFC is shown in Fig. 1. The cell stack successively piles up single cells consisting of an electrolyte plate, anode,

Fig. 1 Stack structure of molten carbonate fuel cell and cross-sectional view of a cell

cathode, and separator plate which forms part of the gas flow channel and separates anode and cathode gases. In addition to these main elements, the stack is equipped with an internal manifold to supply and exhaust gases of each cell, gas headers placed at both the top and bottom ends of the stack, and some other components. Surrounding the cell are sealing surfaces, to which the electrolyte and separator plates are tightly adhered to prevent gas leakage.

As for the gas flow configuration, there are coflow and counterflow in addition to the crossflow as shown in Fig. 1; these depend on the flow direction of the anode and cathode gases. The reactant gas, supplied from the gas header, passes through the manifold and is distributed to each cell. In the gas channels of the separator, the gas reaches the exit while changing its volume due to production and consumption in the electrochemical reaction, and it is carried outside of the stack.

In the stack, heat is produced along with the power generated. The produced heat is transferred to the gas streams and removed from the stack. The heat is also released by convective and radiative heat transfer between the outside surface of the stack and the surroundings. As for cooling the cell down, there is a process gas cooling system in which cathode gas, as a mixture of air and carbon dioxide, flows through in an amount in excess of that required for the reaction. Another method is placement of cooling plates between cells. In this study, the process gas cooling system is examined; here, the flow rate and direction of the cathode gas are considered to have a large effect on performance and temperature distribution, and thus become important factors in achieving a uniform stack temperature.

3. Analytical Model and Method

3.1 Analytical model

For calculation of the current density distribution in the cell required for analyses of performance and temperature distribution of the stack, the thin-film cylindrical model⁽⁷⁾ was used because it is simple and easy to deal with and many cells were to be considered.

The analytical model for the temperature field uses the form for the single cell previously proposed⁽⁶⁾ and piles it up to the number of cells in the stack. Other model features are that the direction of gas introduction can be chosen for every cell, and the flow channel direction in the separator can be changed depending on this flow direction. Accordingly, it allows analysis of performance and temperature distribution of the stack with different combinations of flow configuration types.

3.2 Analysis method

Performance and temperature analyses of the stack were performed by dividing it into elements and then numerically calculating. A plan of the divided stack temperature field and its cross section in the stack direction are illustrated in Figs. 2 and 3, respectively. The division can be unequal; for instance, the size of a divided element for the seal or reaction area may be changed at will. Temperature boundary points were set at the top and bottom end plates, and the boundary conditions dealt with were: (1) heat transfer with ambient atmospheric gas, (2) adiabatic boundary, (3) fixed temperature, (4) fixed heat flux and (5) periodic boundary.

In the performance calculation for the stack, local current density i was calculated for every divided element with respect to the load current I as an operating condition, based on the flow rate of reaction gas, temperature of electrolyte plate and direction of gas introduction to each cell. This i has to satisfy Eq. (1). The relationship of i to the cell voltage V which represents the performance is given by Eq. (2):

$$I = \sum_{n=1}^N i_n \cdot \Delta x \cdot \Delta y \quad (1)$$

$$V = E_n - i_n \cdot Z_n - \eta_{An} - \eta_{cn}, \quad (2)$$

where N : total number of division for reaction region; E : open circuit voltage (V); Z : ohmic impedance ($\Omega \cdot \text{cm}^2$); η_A , η_c : anode and cathode overpotential (V); and subscript n : number of divided elements.

When the current density distribution and the cell voltage which satisfy Eqs. (1) and (2) were obtained for each cell, temperature distribution was calculated

Fig. 2 Plan of the divided stack temperature field in the numerical computation

Fig. 3 Cross section of the divided stack temperature field in the stacking direction

from mass and heat balance equations of Eqs. (3) through (6), and heat generation flux q per unit reaction area is given by Eq. (7):

(1) Mass balance

$$\frac{\partial \dot{G}_i}{\partial X} = - \frac{i}{n_i \cdot F} \quad (3)$$

where G_i : gas flow rate of i -th component (mol/h·m); F : Faraday constant (26.8 A·h/mol); and n_i charge number of i -th component.

(2) Heat balance

$$\frac{\partial}{\partial x} (\dot{G} C_{pg} T_g) = A_{eg} \alpha_g (T_e - T_g) + A_{sg} \alpha_g (T_s - T_g) + \sum \dot{g}_i C_p T_i \quad (4)$$

$$A_e \lambda_e t_e \frac{\partial^2 T_e}{\partial x^2} + A_e \lambda_e t_e \frac{\partial^2 T_e}{\partial y^2} + A_{es} \lambda_{es} t_e \frac{\partial^2 T_e}{\partial z^2} = A_{eg} \alpha_g (T_g - T_e) + \dot{q} \quad (5)$$

$$A_s \lambda_s t_s \frac{\partial^2 T_s}{\partial x^2} + A_s \lambda_s t_s \frac{\partial^2 T_s}{\partial y^2} + A_{se} \lambda_{se} t_s \frac{\partial^2 T_s}{\partial z^2} = A_{sg} \alpha_g (T_g - T_s) \quad (6)$$

where T : temperature ($^{\circ}\text{C}$), α : heat transfer coefficient ($\text{W}/\text{m}^2 \cdot \text{K}$); C_p : specific heat capacity ($\text{Wh}/\text{mol} \cdot \text{K}$); λ : thermal conductivity ($\text{W}/\text{m} \cdot \text{K}$); t : thickness of cell component (m); $\dot{g}_i$: molar flux of species i entering the stream (mol/h·m); A : ratio of gas/hardware interface area to cell area; subscripts g : gas, e : electrolyte plate, s : separator plate, eg : between electrolyte plate and gas, sg : between separator plate and gas, and se : between separator and electrolyte plates.

$$\dot{q} = i \left[- \frac{T \Delta S}{nF} + (E - V) \right] \quad (7)$$

where ΔS : entropy change for the formation of water [$\text{Wh}/(\text{mol} \cdot \text{K})$], and T : reaction temperature (K).

Equations (3) through (6) were made into finite difference representations in accordance with the divided element mentioned above. Temperature node $T(ijk)$ of the electrolyte plate is illustrated as an example in Fig. 4, which shows positions and numbers of other nodes having a thermal relation to it. Separator temperature is similar to this, and nine points, including the objective node, were used on the solid side. On the fluid side, the total number of nodes is four. The difference in node numbers is caused by the one-dimensional flow model and the upwind finite difference representation adopted here. However, the finite difference equation regarding the temperature of all elements is expressed in the form of Eq. (8), allowing temperatures on the solid and fluid sides to be calculated at the same time.

The numerical computation of Eq. (8) was performed using a biconjugate gradient method (BCG method) involving incomplete LDU decomposition to the matrix $A^{(6)}$, because this method is considered suitable for the solution of large and scattered asym-

metric matrices such as this.

$$\sum_{\nu}^9 A^{(\nu)}(ijk) \cdot T(i'j'k') = S(ijk) \quad (8)$$

If the cell stack is higher, and consequently analyses for a great number of cells are needed, the stack model can be divided into several blocks by the above calculation method. The number of cells in one block is automatically set up in accordance with the array volume determined by the memory size of the computer used. Then, temperature in the block was calculated by applying a fixed temperature condition at the element (anode gas) neighboring the lower block and the element (separator) adjacent to the upper block as the boundary condition.

Thermal conductivity λ_{se} in Eqs. (5) and (6) is the apparent one which takes such items as contact resistance between electrolyte and separator plates into account, and it was set to the value of 1.0 W/m·K from measured results⁽⁶⁾. Convergence was accelerated by setting λ_{se} to zero at the beginning of the calculation and by giving the above value only when the average temperature variation of the stack was below 5°C. In addition, stabilization of the solution was obtained by transferring the term of the contact heat conduction into $S(ijk)$ on the right-hand side of Eq. (8).

Returning to the previous performance calculation

Fig. 4 Position and numbers of temperature nodes having a thermal relation to electrolyte plate

Fig. 5 An example of analytical results of 3-dimensional temperature distribution in electrolyte plate and performance for 12-cell stack with crossflow configuration

tion after the temperature distribution was obtained, both calculations were repeated until the temperature variation was below 1°C on the average.

4. Analytical Results of Performance and Temperature Distribution

4.1 Analytical results

Figure 5 illustrates example analytical results of the 3-dimensional temperature distribution in the electrolyte plate and the performance for a stack of 12 cells with an electrode area of 3 600 cm². Flow type in the stack was assumed to be crossflow; temperature boundary conditions at the upper and lower end plates and side faces were natural convection; and heat transfer coefficient α was 12 W/m²·K. Other assumptions and thermal properties of the solid parts used in the analysis were the same as in the previous report⁽⁶⁾. The width of the seal area was 5 cm, and the stack was divided into 5 100 elements (10×10×51) in total. Furthermore, the inlet gas flow rate of each cell was assumed to be the same.

As for the temperature distribution in the stacking direction, Fig. 6 shows temperatures on the average and at the cell center of each cell in the above state. Except for a few cells near the upper and lower end plates, neither temperature differs greatly in the stacking direction, so they can be said to be nearly uniform.

Figure 7 shows the voltage of each cell in this case. As in the temperature distribution profile in Fig. 6, cell voltages at the upper and lower end plates are around 2% lower than those of the central cells in the stacking direction. This drop in performance is an effect of cell temperature. Central and bottom cells have differences in their average temperatures of 17°C and in their cell voltages of 20 mV, and this analytical result for the cell voltage distribution may be regarded as reasonable from the fact that the temperature dependency of the cell performance has been deter-

Fig. 6 Temperature distribution in the stacking direction

mined to be about 1.0 to 1.5 mV per 1°C in the power generation test. Only the voltage of the No. 10 cell shows a bit larger rate of decrease when compared with the tendency of temperature distribution; this is an influence from a calculation error of the voltage.

As for the temperature distribution within the cell plane, as in the results of the single cell already reported⁽⁶⁾, a steep temperature gradient is produced in the cathode gas streamwise direction, and the highest temperature appears at the cathode exit. This tendency is common to each cell. Shown in Fig. 8 is the maximum temperature difference ΔT which is the difference of the maximum and minimum temperatures of each cell in this case. The same figure has results for values of the heat transfer coefficient multiplied by factors of 5 and 10, respectively, in order to study effects of boundary conditions at the stack end plates. This point was examined in consideration of the fact that the heat transfer would be greater than under the assumed condition of natural convection, because of heat conduction, and so forth through fixtures such as stack fasteners which are additionally mounted to the top and bottom portions of the stack in practical use.

Fig. 7 Voltage of each cell for the 12-cell stack shown in Fig. 5

Fig. 8 Maximum temperature difference within each cell

ΔT in the stacking direction shows a trend to reach its maximum near the stack center, and it gets smaller as the cells become closer to the top or the bottom, with the difference being about 40°C. Additionally, the greater α is, the smaller ΔT becomes as a whole, and its influence is found to be larger for cells closer to the top or bottom.

4.2 Comparison between analytical results and measured values

Figure 9 compares measured values of the gas temperature distribution within the cell reported previously⁽⁶⁾, and the present analytical results. Structural parameters of the electrode model for every cell were adjusted so that the calculated cell voltage matched the measured one. Figures 9(a) and (b) illustrate a $990\text{ cm}^2 \times 4$ cell stack (object: cell No. 3) and a $3600\text{ cm}^2 \times 10$ cell stack (object: cell No. 5), respectively. Their comparison from the standpoint of temperature difference shows that the difference is small in Fig. 9(a), which has a small number of cells, whereas it is large in Fig. 9(b), with a larger number of cells, but there is good agreement between analytical results and measured values in both cases. This tendency is caused by the influence of temperature boundary conditions at the end plates as studied previously⁽⁶⁾. The temperature difference is considered to be smaller for the short cell stack because the end effect becomes notable, as described in section 4.1, and this was proved by analytical results here in which heat transfer at the top and bottom of the stack was considered.

The temperature distribution tendency in the analytical results exhibits hardly any difference with that of the single cell already reported⁽⁶⁾. Also, as in

Fig. 9 Comparison of analytical results and measured values⁽⁶⁾

the results of the single cell, a difference can be seen between measured and analyzed results at the inlet area of the anode gas.

4.3 Change in stack performance and temperature distribution with combination of types of the gas flow configuration

Estimating the effect of the gas flow configuration type on the cell performance and temperature distribution is an important problem for uniform stack temperature⁽⁹⁾. Thus, the performance and temperature distribution were analytically compared for six kinds of 12-cell stacks with different combinations of flow configuration types, in which the flow direction of the cathode gas was chosen for each cell. The result are tabulated in Table 1. The objective cell was the 6th one, and the analytical conditions were the same as those in Fig. 5.

For the cell performance, a correlation with mean temperature of the electrolyte plate is recognized, and both the cell voltage and the total voltage are higher for the cell with higher average temperature, showing the same temperature dependency as described in section 4.1.

As for the temperature distribution, both maximum temperature and temperature difference ΔT are greater in stacks of No. 2, with crossflow configuration, and No. 5, when compared with others. The flow type of the No. 5 is that in which the cathode gas crosses at right angles for every cell, and the maximum temperature is raised because cells with the counterflow configuration share half of the whole stack. By contraries, the maximum temperature is lower in the No. 1 type with the coflow configuration, in which the minimum temperature is also low and ΔT is as high as 103°C. The No. 3 is a stack in which the direction of the cathode gas stream is opposite that in the adjacent cells, and both maximum and

minimum temperatures are high, but the temperature difference is 89°C, and lower than that in No. 1. The No. 4 is one of the types in which the cathode gas flows cross each other, as in No. 5, and it is a combination of Nos.1 and 3. The temperature distribution therefore shows values between them. The No. 6 looks like No. 4, but it differs in its composition, having three kinds of cells with different types of flow configuration equally shared along the stack; namely, this stack uses both crossflow and counterflow for the cathode gas flow. As a result, No. 6 has the smallest ΔT of 76°C, and it can be regarded as the best combination of flow configuration types through overall considerations of performance and temperature distribution.

Next, the combination of different flow configuration types was studied regarding changes in the cell temperature distribution. Shown in Fig. 10 are analytical results of temperature distribution, as an example, in the electrolyte plate of stack No. 3, which uses counterflow for the cathode gas flow. A comparison with the crossflow configuration of No. 2, shown in Fig. 5, shows that No. 3 exhibits a large difference and the region of highest temperature has moved near the center, with the temperature distribution becoming symmetrical about the direction of cathode flow.

Figure 11 illustrates the temperature profile of the electrolyte plate in the direction of cathode gas flow at the central part of the 5th cell in stack No. 3, along with the temperature distribution of the cathode gas in the 5th and 6th cells. The case of crossflow is similarly shown in Fig. 12 for the purpose of comparison. When all the cathode gas flows from one direction as shown in Fig. 12, the electrolyte plate and two cathode gases have the same trend in their temperature profile. On the other hand, it is found from Fig. 11 that the electrolyte plate temperature is affected by not only the cathode gas of the 5th cell, but also the gas temperature of the adjacent 6th cell. In other

Table 1 Comparison of performance and temperature distribution for six kinds of 12-cell stacks with different combinations of flow configuration

No.	1	2	3	4	5	6
Gas flow in cell stack						
Total voltage V	8.86	8.87	9.05	8.95	9.04	8.99
Cell voltage V	0.740	0.744	0.759	0.746	0.759	0.752
Average temperature °C	654	656	674	662	669	667
Maximum temperature °C	710	753	735	717	752	715
Minimum temperature °C	607	594	646	621	611	639
Temperature difference °C	103	159	89	96	141	76

Fig. 10 Analytical result of temperature distribution in electrolyte plate of stack No.3 (counterflow for the cathode gas flow)

Fig. 11 Temperature profiles in the direction of cathode gas flow at the central part of the 5th cell in stack No. 3

words, the temperature at the inlet area becomes higher than that in Fig. 12 due to an effect of the elevated gas temperature of the 6th cell, and conversely, it becomes lower at the outlet area due to the 6th cell's cathode inlet cooled gas.

As mentioned above, the temperature distribution of each cell in a stack is affected not only by the gas flowing in the said cell, but also considerably by the gas temperature of the neighboring cells. The results in Table 1 imply that the temperature distribution in a cell becomes more uniform and stack cooling characteristics can be improved with the promotion of heat transfer in the stacking direction, taking the combination of gas flow configuration types into consideration.

5. Conclusions

The 3-dimensional performance and temperature distribution of molten carbonate fuel cell stacks were analyzed. The following results were obtained:

(1) A comparison of the analytical results of temperature distribution and measured results at power generation showed good agreement on the point of temperature difference within the cell plane. The analytical model and method with which heat transfer at the ends of the stack is taken into consideration proved to be effective for analyzing the temperature distribution of the cell stack.

(2) Heat transfer at the upper and lower ends of the stack cells had a large influence on the cell temperature distribution, and the greater the heat transfer was, the more uniform was the temperature distribution obtained in the cell.

(3) Because the temperature distribution in a cell is also influenced by the gas temperature of the neighbor cells, heat transfer between cells is promoted and uniformity of the stack temperature distribution can

Fig. 12 Temperature profiles for stack No. 2 with crossflow configuration

be obtained by combining cells with different types of flow configuration.

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