Current density analysis in a molten carbonate fuel cell unit with non-uniform inlet flow and high anode gas utilization

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Keywords: Current density, Molten Carbonate Fuel Cell, Non-uniform, Anode gas Utilization.

ABSTRACT

This study investigates the electrical performance of a planar molten carbonate fuel cell unit with cross-flow configuration when both the anode gas utilization gets higher and anode gas inlet flows are non-uniform. A software package in this study solves two-dimensional, simultaneous, partial differential equations of mass, energy, and electro-chemistry, without considering stack direction variation. The results show that the anode gas utilization increases with a decrease in the molar flow rate, and the average current density decreases when the molar flow rate drops. Additionally, non-uniform inlet profile of the anode gas will induce a happening of non-reaction area. This non-reaction area deteriorates the average current density and then deteriorates the electrical performance to −4%.

INTRODUCTION

A molten carbonate fuel cell (MCFC) includes two anode and cathode electrodes, as well as one electrolyte, between the anode and cathode, delivering carbonate ions from the cathode to the anode. Molten carbonate fuel cells operate at high temperatures of about 650°C, and use methane or ethanol as anode gas due to internal self-reformation. The planar-type molten carbonate fuel cell stacks easily and is desirable for many applications. Several studies investigated performance simulation under different conditions, since anode gas rate, inlet temperature, operation pressure, cell size, etc. affect temperature and current density distribution in a MCFC unit.

Sampath and Sammells (1980) analyzed a current density distribution on a MCFC unit with an isothermal assumption, and discussed the effect of gas concentrations, flow rates and operating pressure on the performance of the MCFC unit. Wolf and Wilemski (1983) used a two-dimensional numerical model to simulate the non-isothermal performance of a MCFC unit with considering co-flow, counter-current, and cross-flow configuration. Kim et al. (2002) investigated the effect of water-gas shift reaction on the performance of a MCFC unit by applying a numerical method, and their results indicated that this effect of the shift reaction on the voltage distribution is small. Standaert et al. (1998) employed a quasi-isothermal approach to describe the behavior of non-isothermal MCFC unit of co-flow type. This analytical expression for the local current density and the cell voltage is very accurate up to high fuel utilizations by comparison with numerical calculations. Mangold and Sheng (2004) reduced nonlinear model for a planar MCFC unit with cross-flow configuration to describe temperature field dynamics with sufficient accuracy and less computation time.

In practical application, a MCFC is constructed by stacking a lot of units for providing a higher power. Therefore, there are many literatures focused on the thermal and electrical analysis of a MCFC stack. Kobayashi et al (1989) and Fujimura et al (1992) investigated the temperature and current density distribution in a MCFC stack by a numerical method. Their results showed that the upper and lower ends of the stack cells had a large influence on the cell temperature distribution. Yoshiba et al (1998) analyzed the temperature and current density distribution in a MCFC stack with five gas flow geometries, and found that the co-flow type induced the highest net output power. Bosio et al. (1999) used a
total cell resistance formula proven by experimental data to analyze temperature distribution and current density distribution for a single cell and a 20-layer stack of MCFC by numerical code. Their numerical results showed that all the cells behave identically, but the end cells are slightly different due to their special geometry. Yoshiba et al (2000) used a numerical method to calculate the temperature and current density distribution of a co-flow MCFC stack, and found that greater increase in partial internal resistance and greater insufficiency of fuel gas in a particular cell will result in a larger cell voltage difference.

Because the anode gas and cathode gas need manifolds to lead them into each stack, and distributors to assign the gases to channels in each stack, manifold position and distributor geometry influence flow rate distribution in the inlet section. Flow rate pattern in both the frontal entrance area and stacking direction are therefore non-uniform in practice. Few literatures have investigated non-uniform inlet flow effect on anode gas cell temperature and current density. Hirata and Hori (1996) used the numerical method to examine relationships between planar and stacking direction gas flow uniformities, and cell performance in a co-flow type molten carbonate fuel cell. Although their results indicated that stacking direction gas flow uniformity is about 2 to 10 times greater than that in the planar direction, the inlet flow mal-distribution in the planar direction will produce more complex temperature distribution and current density distribution due to the cross-flow pattern. Okada et al (2006) presented an investigation of the gas distribution in a large-scale MCFC stack with an internal reforming and co-flow configuration, and found that the improvement of the flow distribution in the reforming unit affected the uniformity of the temperature distribution in the cell area. Recently, Liu et al (2006) investigates the temperature and current density distributions in a molten carbonate anode fuel cell unit with cross-flow configuration when the inlet flows of the anode gas and the cathode gas are mal-distributed in eight patterns. Their results showed that the non-uniform inlet flow slightly affects the mean temperature and mean current density, but worsens the distribution of temperature and current density for most mal-distribution patterns.

Anode gas utilization is an important factor, because it represents an economical operation of a fuel cell when the anode gas utilization is higher. Examining the previous literature indicates that the anode gas utilization is over 70%, and authors’ previous research considered the anode gas utilization only up to 25% in Lie et al (2006). Therefore, this paper plans to extend previous research to the current density distribution analysis of a molten carbonate fuel cell when both the anode gas inlet flow is non-uniform and the anode gas utilization gets higher. Because the anode gas molar flow rate dominantly affects the current density of a fuel cell, the non-uniform inlet flow effect must induce an appearance of non-reaction on the cell plane when the anode gas utilization gets higher. This non-reaction area also decreases the overall current density of a cell and deteriorates the electrical performance of a molten carbonate fuel cell.

![Fig. 1 Schematic diagram of a unit of solid oxide anode gas cell in cross-flow.](image)

**ANALYSIS**

This study investigates a 0.6m×0.6m molten carbonate anode gas cell unit with anode gas and cathode gas cross-flow, as shown in Figure 1. The anode gas flows in the x direction, and the cathode gas flows in the y direction. Additionally, this study combines the anode, electrolyte, and cathode to form a unit cell for simple analysis. Due to separator ribs, both flows are considered to unmixed flow, i.e., each flow is composed of many flow tubes parallel to each other. Manifold position will induce different mal-distribution profiles in the inlet section, as mentioned above. According to the results in Liu et al (2006), the anode gas and cathode gas molar flow rate dominantly affects the current density and the cell temperature, respectively. For simplifying the current density analysis, this paper considers the cathode gas inlet is uniform and the anode gas inlet has three profiles, as shown in Figure 2. Before formulating the governing equations, this study assumes: 1.) Inlet temperature and mole fractions of species in anode gas and cathode gas are constant and uniform. 2.) Thermal properties of anode gas, cathode gas, cell, and separator are constant except for anode gas and cathode gas specific heat capacities. 3.) Cell and separator boundary are adiabatic. 4.) Neglect z direction change. 5.) Separator cross-section geometry is unchanged throughout the x-y plane. 6.) Cell voltage is uniform over the x-y plane. 7.) Neglect the anode gas water-shift reaction.
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\[ H_2 + CO_3^{2-} \rightarrow H_2O + CO_2 + 2e^- \]  

The reaction in cathode is

\[ O_2 + 2CO_2 + 4e^- \rightarrow 2CO_3^{2-} \]  

Mass balances are formulated for each species molar flow rate change in anode gas and cathode gas with species consumption linking the local current density.

\[ \frac{1}{L_y} \frac{dn_{m,j}}{dx} = \pm \frac{i}{n_F} \]  

\[ \frac{1}{L_z} \frac{dn_{m,j}}{dy} = \pm \frac{i}{n_F} \]  

Meanwhile, \( n \) is the molar flow rate of the k-component, \( n_e \) is the number of electrons transferred in the reactions of the anode and the cathode, and the plus/minus symbol represents molar flow rate increase or decrease dependent on reactant or product species. In Eq. (3), anode gas reactants and products include hydrogen, carbon dioxide, and water. The reactant and product in cathode gas include oxygen and carbon dioxide in Eq. (4).

This research conserves energy for anode gas, cathode gas, cell, and separator, respectively.

For anode gas,

\[ \frac{\partial}{\partial x} \left( \sum n_{m,j} c_{p,j} T_i \right) = (h_a)_{m,j} \left( T_i - T_{m,j} \right) + (h_a)_{m,j} \left( T_i - T_{m,j} \right) + \frac{i}{2F} c_{p,CO_2} T_i \]  

For cathode gas,

\[ \frac{\partial}{\partial y} \left( \sum n_{m,j} c_{p,j} T_i \right) = (h_a)_{m,j} \left( T_i - T_{m,j} \right) + \frac{i}{2F} c_{p,CO_2} T_i \]

For cell,

\[ (k) \frac{\partial^2 T}{\partial x^2} + (k) \frac{\partial^2 T}{\partial y^2} + (ka)_{m,j} \left( T_i - T_{m,j} \right) + (ha)_{m,j} \left( T_i - T_{m,j} \right) + \frac{i}{2F} c_{p,CO_2} \left( T_i - T_{m,j} \right) + q_e = 0 \]

For separator,

\[ (k) \frac{\partial^2 T}{\partial x^2} + (k) \frac{\partial^2 T}{\partial y^2} + (ka)_{m,j} \left( T_i - T_{m,j} \right) + (ha)_{m,j} \left( T_i - T_{m,j} \right) = 0 \]

Each gas species specific heat capacities in anode gas and cathode gas are a function of temperature based on the results of Koh et al. (2002), and this study adds one carbon and one half of oxygen specific heat capacity as carbonate ion specific heat capacity. In Eq. (7), \( q_e \) is heat generation occurring in the cell unit due to electrochemical reactions and cell internal losses, and this heat generation occurs over the x-y plane as follows.

\[ q_e = -\Delta H \times \frac{i}{2F} V \cdot i \]

\[ \Delta H = -240506 - 7.3835 T_i \]

Meanwhile, \( \Delta H \) is enthalpy change per mole of chemical reaction, calculated as a temperature function. In Eq. (7) and (8), \( k \) is thermal conductivity due to contact resistance between cell and separator in the z direction, and its value is set to 1.0 W/m K. In Eqs. (3) - (6), this paper considers the non-uniform profile of the molar flow rate of the anode gas as follows.

\[ n_{m,j}(0,y) = \frac{N_m}{L_y} \left( \frac{2d}{L_y} y + 1 - d \right) \]

Based on the Nernst equation, this study calculates the Nernst voltage as follows.

\[ E = E_0 + \frac{RT}{2F} \ln \left( \frac{P_{P_{o2}} P_{P_{CO_2}}}{P_{P_{CO_2}} P_{P_{o2}}} \right) \]

\[ E_n = 1.2723 - 2.7654 \times 10^{-4} T \]
Meanwhile, $E_0$ is the reversible voltage under standard conditions, according to Koh et al. (2002). According to the results in Bosio et al. (1999), this study uses the total cell resistance, including that due to cathode polarization, the electrolyte tile contribution, and the Ohmic resistance of the contacts. Note that this total cell resistance did not include the concentration polarization, because Bosio et al. (1999) assumed the diffusion is non-limiting in the electrode. The cell voltage is the Nernst voltage minus the over-potentials, as follows.

$$V = E - iR_{\text{tot}}$$  \hspace{1cm} (14)

$$R_{\text{tot}} = \frac{A e^{\beta_o T} + c_v + D \cdot e^{T/\beta_o}}{\prod_{i} \beta_i}$$  \hspace{1cm} (15)

where the parameters are $\beta_o = 0.67$, $A = 1.38 \times 10^7 \ \Omega \ m^2 \ Pa^{0.67}$, $B = 11400 \ K$, $c_v = 0.348 \times 10^{-4} \ \Omega \ m^2$, $D = 4.8 \times 10^{-8} \Omega \ m^2$ and $F = 6596 \ K$ (Bosio et al., 1999).

The above simultaneous equations of the MCFC contain seven unknown variables, which are mole flow rate of each species ($n_{H_2}$ and $n_{O_2}$), anode gas temperature ($T_{ag}$), cathode gas temperature ($T_{cg}$), cell temperature ($T_h$), separator temperature ($T_s$), current density ($i$), and cell voltage ($V$). The mass equations are used to determine mole flow rate of each species, and energy equations are used to determine the temperatures. Nevertheless, in Eq. (14), both current density and cell voltage variables must be evaluated. Therefore, this study assumes that the cell voltage is uniform over the reaction area of the cell and then calculates the current density using Eq. (14). This paper employs a software package, FlexPDE, to solve the governing equations, because it flexibly solves partial differential equations by a finite element method. The validation of this software package had done in the literature by Liu et al. (2006).

**RESULTS AND DISCUSSION**

This study considers the anode gas flow rate is 0.00621-0.0621 mol/s, the cathode gas flow rate is 0.0263-0.1841 mol/s, the inlet temperature of anode gas and cathode gas are 858 and 867 K, the operation pressure is 3.5 atm, the operation voltage is 0.8 V, and the deviation of the non-uniform profile is 0.5. Figure 3 shows that the current density distribution in pattern A and pattern B when the anode gas and cathode gas flow rate is 0.0621 and 0.1841 mol/s, respectively. In this figure, the profile with bold line is the current density distribution in uniform pattern, and the profile with color represents current density distribution in pattern A or pattern B. Examining the current density distribution in uniform pattern indicates that the current density mainly decreases along the anode gas flow direction from 1842 to 1315 A/m². In the cathode gas flow direction, the current density slightly rises because of the decrease of total resistance in Eq. (15). In Fig. 3(a), the current density has more severe reduction in the corner of the anode gas exit and the cathode gas inlet. In this corner, the hydrogen concentration becomes lower because the progressively increasing anode gas profile induces less anode gas flowing through this area. Oppositely, the area with apparent current drop happens in the corner of the anode gas exit and the cathode gas exit, because the anode gas non-uniform profile is progressively decreasing. Comparing the Fig. 3(a) and 3(b) indicates that the current density drop in the corner of Fig. 3(b) is slightly larger than that of Fig. 3(a).

Figure 4 shows that the current density distribution in pattern A and pattern B when the anode gas and cathode gas flow rate is 0.01242 and 0.0526 mol/s, respectively. In this figure, the profile with
bold line is the current density distribution in uniform pattern, and the profile with color represents current density distribution in pattern A or pattern B. Examining the current density distribution in uniform pattern indicates that the current density mainly decreases along the anode gas flow direction from 1846 to 347 A/m². This current density range is clearly lower than that in Fig. 3. Because both the anode gas and cathode gas molar flow rate reduces in Fig. 4, the current density becomes lower due to the less hydrogen and oxygen concentration. In Fig. 4(a), the current density has a reduction in the corner of the anode gas exit and the cathode gas inlet. In this corner, the current density rapidly decreases from 1848 to 138 A/m². This means the region is close to a non-reaction area. The less anode gas flow rate and non-uniform inlet flow induces the anode gas molar flow rate is fewer in this area, so the hydrogen is almost used up and the current density becomes near zero.

Similarly, the current density severely drops in the corner of the anode gas exit and the cathode gas exit in Fig. 4(b), and this corner occur the non-reaction situation.

Figure 5 shows that the current density distribution in pattern A and pattern B when the anode gas and cathode gas flow rate is 0.00621 and 0.0263 mol/s, respectively. The non-reaction area of Fig. 5(a) is apparently larger than the same area of Fig. 4(a), because the anode gas molar flow rate is the least in this study. Even in uniform pattern, the current density distribution happen a severe drop in the anode gas exit, as shown in the bold line. This means the anode gas is almost used up when the anode gas flows out the cell reaction in uniform pattern. Examining the average current density over the reaction area shows that it is the lowest one in Figs. 3 to 5. When the inlet flow is non-uniform, the current density in the corner of the anode gas exit of Fig. 5(a) and 5(b) drops to zero.
because of the progressively increasing and decreasing profile of the anode gas inlet molar flow rate.

Promoting the anode gas utilization in an anode gas cell is more economical, but the global current density will drop with an increase in anode gas consumption and decrease the power of a anode gas cell. Moreover, the non-reaction area will happen and more deteriorate the global current density when the anode gas molar flow rate decreases, the anode gas utilization will increase accompanying the flow rate decrease. The effect of non-uniform inlet flow on the average current density becomes more apparent when the anode gas utilization is close to 73%, and then becomes slighter along the increase of gas utilization. In this figure, the relative change of average current density raise to ~4% in Pattern B.

![Fig. 6 Relative change of average current density in non-uniform pattern.](image)

**Table 1. Average current density and anode gas utilization at different inlet molar flow rate and patterns**

<table>
<thead>
<tr>
<th>Molar Flow Rate</th>
<th>Uniform</th>
<th>PatternA</th>
<th>PatternB</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( \frac{i}{(A/cm^2)} )</td>
<td>( U_f ) (%)</td>
<td>( \frac{i}{(A/cm^2)} )</td>
</tr>
<tr>
<td>( n_f=0.0621 )</td>
<td>1655</td>
<td>25</td>
<td>1636</td>
</tr>
<tr>
<td>( n_f=0.01242 )</td>
<td>998</td>
<td>73</td>
<td>981</td>
</tr>
<tr>
<td>( n_f=0.00621 )</td>
<td>606</td>
<td>90</td>
<td>601</td>
</tr>
</tbody>
</table>

**CONCLUSIONS**

This study investigates the effect of non-uniform inlet flow on the electrical performance of a MCFC unit. This work employed a software package to solve the simultaneous mass, energy, and electrochemistry equations. With considering three flow patterns and three molar flow rates, this research analyzed the current density distributions at different conditions. The results show that the anode gas utilization increases with a decrease in the molar flow rate, and the average current density decreases with the decrease in the molar flow rate. In addition, non-uniform Pattern A and B will induce a happening of non-reaction area in the corner of the anode gas exit. This non-reaction area deteriorates the average current density and deteriorates the electrical performance to ~4% when the anode gas molar flow rate is 0.01242 mol/s and anode gas utilization is 73%.

**REFERENCES**


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S.F. Liu and H.S. Chu: Current density analysis in a molten carbonate fuel cell.


NOMENCLATURE

- area of heat transfer area per base area (m² m⁻²)
- specific heat capacity (J mol⁻¹ K⁻¹)
- deviation of inlet flow profile in the stacking direction
- Nernst voltage (V)
- reversible open circuit voltage (V)
- Faraday’s constant (96485 As mol⁻¹)
- heat transfer coefficient (W m⁻² K⁻¹)
- current density (A m⁻²)
- conductivity (W m⁻¹ K⁻¹)
- length in the x or y direction, indicated by subscript x or y (m)
- molar flow rate of anode gas or cathode gas per unit width, indicated by subscript ag or cg (mol m⁻¹ s⁻¹)
- molar flow rate of anode gas or cathode gas, indicated by a subscript ag or cg (mol s⁻¹)
- number of electrons transferred in reactions of anode and cathode
- pressure (Pa)
- universal gas constant (8.314 J mol⁻¹ K⁻¹)
- total cell resistance (Ω m²)
- temperature (K)
- cell voltage (V)
- x direction
- molar fraction indicated by subscript k
- y direction

Greek symbols

δ thickness (m)

Subscripts

ag anode gas
c cell
cg cathode gas
k components in anode gas or cathode gas
s separator
x x direction
y y direction
高燃料使用率下非均勻入口流率對單體熔融碳酸塩燃料電池之電流密度場分析

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摘要

本篇論文在探討平板式融熔碳酸鹽單體燃料電池在交叉流模式及高燃料使用率下，非均勻入口流率對燃料電池電性能之影響分析。在忽略各特性在堆疊方向上之變化下，本篇利用一套裝軟體分別求解二維之質量守恆、能量守恆及電化學反應微分方程聯立式。結果顯示燃料使用率會隨著莫爾流率之增加而減少，而平均電流密度則會隨者莫爾流率之減少而減少。此外，非均勻的入口流率會在高燃料使用率之狀況下產生非反應區之出現。此非反應區會減低平均電流密度並使電性能下降 4%。