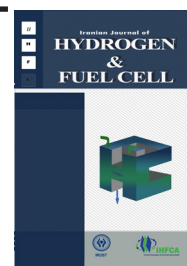


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## New mathematical modelling and dynamic simulation of a molten carbonate fuel cell

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### Abstract

In this study, a more accurate model of a fuel cell of molten carbonate was also used to determine input and output control variables and to investigate the behavior of the system with respect to those variables. A more complete kinetic was also implemented to increase the effectiveness of the presented paper. The input variables, methane and cell voltage, are included in the fuel flow rate of cell. The output of the model is the flow resulting from the cell which is a function of the electrochemical reaction rate, and accordingly also a function of the state variables quantities. In the following, the model was used to simulate step response of each input and then the dynamic behavior of cell was analyzed. The results indicate that as the fuel flow rate into the cell lessens, the productivity of the fuel gets higher. Also, in the analysis of the fuel cell it was seen that the temperature of molten carbonate depends strongly on the amount of combustion of compositions in the combustion chamber. As the inlet concentration of methane, hydrogen and carbon monoxide increased, the heat liberated from combustion was more and increasing the system temperature which results in increased thermal stress in the molten carbonate fuel cell.

## 1. Introduction

It is possible to transform the chemical energy of fuels into electrical energy in gaseous power plants and to transmit it as electricity; hence, this should allow people access energy everywhere and provide for their needs with various electrical apparatus. A fuel cell is a kind of galvanized cell that can produce electrical

energy with a series of electrochemical reactions, when the free energy of the reactants is greater than that of the products. The produced electrical energy can be used in applications such as domestic uses, in portable equipment, military industries, and transportation [1-3].

Side products of fuel cells are mainly heat which can be used to increase the efficiency of the production.

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In addition to less air pollution with respect to gaseous power plants, it also has a higher theoretical efficiency. This is due to their mechanism which in a series of reactions directly transforms energy existing in the fuel into electron current, it also benefits from mechanical waste avoidance in sectors like the boiler and turbine of a power plant [1, 4, 5].

In 2001, Michael Lukas et al. [6] introduced a simulated model of a zero dimensional mathematical model of a molten carbonate fuel cell adapted by several ordinary differential equations. In this model, the water-gas shift reaction was presented and solved numerically as an algebraic equation showing the equilibrium of this reaction. The empirical model utilized in the Ding et al. [7] was employed to calculate the voltage drops. Finally, a storage with a capacity of two megawatts was simulated.

During the same time, Koh et al. [8] also introduced a model to predict the pressure and temperature profile in MCFC storage. This model was based on fluid dynamics and heat transfer in a fuel cell of molten carbonate, and it predicted dynamic pressure and gas temperature extensively in the fuel cell. The results of pressure drop and temperature in the simulation were compared with empirical data of a molten carbonate fuel cell in a pilot scale and the results showed that the profile was predicted accurately. Then, the effect of boundary temperature on the profile was investigated in the cell. The results showed that the model accuracy was satisfactory for the conductivity boundary condition. Also, it was shown that there was no difference between 2D and 3D model accuracy. Moreover, the impact of scale increase on the temperature gradient was studied. Results showed that the increase of scale does result in variations in the temperature gradient.

Sundmacherz [9-12] introduced another paper in this area. In this complimentary work an extensive dynamic model for MCFC was introduced with a transverse arrangement that included mass, heat, and electrical charge equations, and all of these models were conducted dimensionless. The equations were solved to find distribution of concentration, temperature, molar rate of gas and current density in

the cell. By employing this model, these profiles can be obtained in a steady state condition. Sundmacherz claimed that this model can be used in order to simulate, optimize, and MCFC control in every aspect. He also claimed that this model can be extended to other high temperature fuel cells simulation like Solid Oxide Fuel Cells (SOFCs). At the end, the results acquired in the simulation were compared with the results of other models.

Chudej et al. [13] expressed the behavior of a MCFC with a bunch of Partial Differential Equations (PDEs) obtained based on physical and chemical rules. This model was used as a valuable tool for identification and controlling a system in order to increase its lifetime. In this model, system control was emphasized especially with the aim of high temperature gradient avoidance. The model introduced in this work is 1D and includes 3 PDEs which were solved with the lines approach. Software was employed to solve the equations. Additionally, Kim et al. [14] used a 3D numerical solution of conservation of mass, momentum, energy and electrochemical equations in MCFC for comparison of to compare electrochemical performance. In this work, three arrangements of co-current, counter-current and cross-current for gases in two channels were investigated, and in every case the concentration, temperature, and current density were obtained so that the pressure drop was calculated. The results of this work showed that the co-current arrangement has the lowest pressure drop along the electrolyte. Temperature and current density distribution are smoother in this arrangement than the others. However, a counter-current stream was found to be the best stream arrangement in the performance steady state, because it had the lowest voltage drop. More currently, Lee et al. [15-17] investigated the effect of temperature on the performance of MCFC in steady state conditions. They founded reaction kinetic was accelerated by increasing the cell temperature.

The majority of the previous studies concentrated on steady state simulation of molten carbonate fuel cells. However, the main priority of this study is on a dynamic simulation of molten carbonate fuel cells. In this study, a more accurate model of the fuel cell

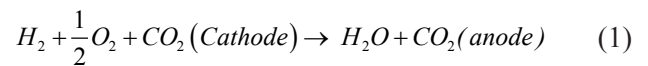
of molten carbonate was also used to determine input and output control variables and then the behavior of the system was investigated with respect to those variables. A more complete kinetic was also implemented to increase the effectiveness of the presented paper. The input variables include the fuel flow rate of the cell which is methane and cell voltage. The output of the model is the flow resulting from the cell which is a function of the electrochemical reaction rate, and accordingly of state variable quantities. In the following, the model at every input under the change of step was simulated and then the dynamic behavior of cell was analyzed.

## 2. Mathematical Modelling

The electrolyte of the molten carbonate cell is the melted salt of alkali metal carbonates. Lithium and potassium or lithium and sodium carbonate salt are often used. This salt was confined in the porous matrix of  $\text{LiAlO}_2$  which was in a molten state at the operation temperature of this type of fuel cell (600-800 °C) and electrical current was passed by the  $\text{CO}_3^{2-}$  ions movement. The overall view of a molten carbonate fuel cell with an electrochemical reaction of the electrodes is shown in Figure 1.

As can be seen in Figure 1, unlike other types of fuel cell, in the cathode channel it is necessary to have Carbon dioxide ( $\text{CO}_2$ ) in the molten carbonate cell in order for it to be consumed in electrochemical

reactions. When this species was combined with oxygen on the cathode electrode it yielded carbonate ions ( $\text{CO}_3^{2-}$ ). On the other hand, these ions combined with hydrogen on the anode electrode and yielded  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . This produced ion movement in the electrolyte from cathode to anode. The electron movement in the outer circle from anode electrode to cathode electrode is the consequence of two electrochemical reactions transmitting  $\text{CO}_2$  from cathode to anode. The outcome of these two reactions is expressed by equation 1 as follows:



Nernst voltage can then be calculated from equation 2-3 as follows:

$$E = E^0 + \frac{R.T}{2.F} \cdot \ln\left(\frac{P_{\text{H}_2\text{a}} \cdot P_{\text{O}_2\text{c}}^{0.5} \cdot P_{\text{CO}_2\text{c}}}{P_{\text{H}_2\text{Oa}} \cdot P_{\text{CO}_2\text{a}}}\right) \quad (2)$$

$$E^0 = 1.2723 - 2.7645 \times 10^{-4} \cdot T \quad (3)$$

Also, by considering their lost, cell voltage can be obtained from equations 4-7.

$$V_{\text{cell}} = E_{\text{eq}} - i \cdot (\eta_{\text{an}} + \eta_{\text{cat}} + \eta_{\text{ohm}}) \quad (4)$$

$$\eta_{\text{an}} = 2.27 \times 10^{-9} \exp\left(\frac{E_{\text{act an}}}{R.T}\right) \times P_{\text{H}_2}^{-0.42} \times P_{\text{CO}_2}^{-0.17} \times P_{\text{H}_2\text{O}}^{-1} \quad (5)$$

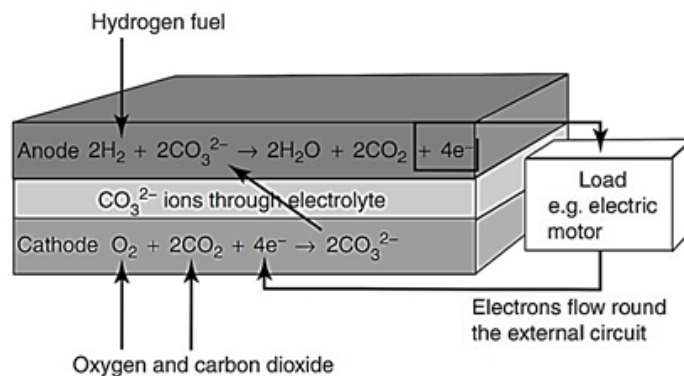


Fig. 1. Schematic of a molten carbonate fuel cell with electrochemical reaction.

$$\eta_{cat} = 7.505 \times 10^{-10} \exp\left(\frac{E_{act,cat}}{R.T}\right) \times P_{O_2}^{-0.43} \times P_{CO_2}^{-0.09} \quad (6)$$

$$\eta_{ohm} = 0.45 \times 10^{-5} \exp\left[8600\left(\frac{1}{T} - \frac{1}{923}\right)\right] \quad (7)$$

So, in addition to the partial pressure (concentration) of hydrogen, oxygen, and water vapor the partial pressure of carbon dioxide affected the Nernst voltage in both channels. The kinetic equation of chemical reactions in the molten carbonate are shown in Table 1.

In the molten carbonate fuel cell, fuel gases can combine and then burn the output gases of the anode channel with air in a combustion chamber instead of oxygen, and carbon dioxide injected into the cathode channel produced gases which were a combination of carbon dioxide, oxygen and some nitrogen and water vapor which can be conducted into the cathode channel. The second benefit of molten carbonate fuel cell is to increase the temperature of input air to the cathode

channel in addition to the necessary carbon dioxide provision. Otherwise, the input air has to be warmed up prior to entering to fuel cell like some other types of fuel cells. Accurate details of mathematical model are introduced in Table 2 and the physical properties and operational condition are shown in Tables 3 and 4 [9-12, 18].

The dynamic simulation of these systems uses the methods employed in the steady simulation or line approach which are a combination of intended methods and the analytical approach. It can discretize partial differential equations in the location dimension, but the mentioned method can conserve equations continuously in time dimension. Then, allocating these equations to each of the blocks, each partial differential equation transforms to an ordinary differential equations system that can be solved. In the dynamic simulation, considering the fact that the calculation cost is high, the fewer number of points in the dynamic simulation could have special importance in discretization. On the other hand, more points could give more accurate responses. So, researcher

**Table 1. Chemical reaction with kinetic equation in the molten carbonate fuel cell**

Chemical Reactions	Kinetic equation
$CH_4 + H_2O \rightarrow CO + 3H_2$	$R_1 = \frac{K_1}{P_{H_2}^{2.5}} \left( P_{CH_4} \cdot P_{H_2O} - \frac{P_{H_2}^3 \cdot P_{CO}}{K_{eq,1}} \right) / DEN^2$
$CO + H_2O \rightarrow H_2 + CO_2$	$R_2 = \frac{K_2}{P_{H_2}} \left( P_{CO} \cdot P_{H_2O} - \frac{P_{H_2} \cdot P_{CO_2}}{K_{eq,2}} \right) / DEN^2$
$CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$	$R_3 = \frac{K_3}{P_{H_2}^{3.5}} \left( P_{CH_4} \cdot P_{H_2O}^2 - \frac{P_{H_2}^4 \cdot P_{CO_2}}{K_{eq,3}} \right) / DEN^2$
	$DEN = 1 + k_{CO} \cdot P_{CO} + k_{H_2} \cdot P_{H_2} + k_{CH_4} \cdot P_{CH_4} + k_{H_2O} \cdot \frac{P_{H_2O}}{P_{H_2}}$
	$K_i = K_{i,T_r} \cdot \exp\left(\frac{-E_i}{R} \cdot \left(\frac{1}{T} - \frac{1}{T_r}\right)\right) \quad , \quad k_j = k_{j,T_r} \cdot \exp\left(\frac{-\Delta h_j}{R} \cdot \left(\frac{1}{T} - \frac{1}{T_r}\right)\right)$

Table 2. Mathematical modelling of molten carbonate fuel cell

Domain	Boundary
<b>Anode and cathode channels</b>	
Mole balance	
$C \left[ \frac{\partial x_A}{\partial t} + V_x \frac{\partial x_A}{\partial x} \right] = \frac{1}{V_{ch}} \cdot \sum_j (R_{Aj} - x_A \cdot \sum_{\beta=1}^N R_{\beta j})$	$x_A(x=0, t) = x_{A0}$ $x_A(x, t=0) = x_{Ai}$
Energy balance	
$C C_p \left[ \frac{\partial T_{a/c}}{\partial t} + V_x \frac{\partial T_{a/c}}{\partial x} \right] = \frac{q_{a/c}}{V_{ch}} - \frac{1}{V_{ch}} \cdot \sum_j R_j \cdot \Delta h_j$	$T_{a/c}(x=0, t) = T_{a/c,0}$ $T_{a/c}(x, t=0) = T_{a/c,i}$
$q_T = q_{a/c} + \sum_j R_j \cdot \Delta h_j \quad q_c = A \cdot h_c \cdot (T_s - T_c) \quad q_a = A \cdot h_{as} \cdot (T_s - T_a)$	
<b>Solid Phase (electrodes, electrolyte, Separator Plates)</b>	
Energy balance	
$\tilde{n}_s \cdot C_{ps} \cdot \frac{\partial T_s}{\partial t} = K_s \cdot \left( \frac{\partial^2 T_s}{\partial x^2} \right) -$	$\frac{\partial T_s}{\partial x}(x=0, t) = 0$
$\frac{R_{elec} \cdot \Delta h_{elec} - A \cdot [h_{as} \cdot (T_s - T_a) + h_{cs} \cdot (T_s - T_c)] - i V_{cell}}{V_s}$	$\frac{\partial T_s}{\partial x}(x=L, t) = 0$
	$T_s(x, t=0) = T_{s,i}$

should use an optimized number of points which are the minimum number of points having acceptable accuracy.

### 3. Results and Discussions

A dynamic simulation of the system is necessary for the system control goals, steady conditions alteration, startup, shutdown and opposition to turbulences. As discussed in detail above, an ordinary differential

equation system needs to be solved to screen the dynamic behavior of the system. In the present project, the method of lines has been employed to solve this system. In this approach, which is a combination of, analytical and numerical methods, each PDE equation was transformed to an ODE equation with equation discretization. This approach decreased the complexity of the equations but increased the number of equations and consequently the volume of calculations. In this project, the system of equations are discretized in place by the finite difference method

**Table 3. Operational condition of molten carbonate fuel cell**

Simulated Parameters	Value
Operational pressure	1 bar
Cell thickness	1 cm
Thickness of electrodes	0.8 mm
Electrolyte thickness	1 mm
Cell Area	0.5 m×1 m
Convection Coefficient of Anode Channel	83.86 W.m <sup>-2</sup> .K <sup>-1</sup>
Convection coefficient of cathode channel	92.96 W.m <sup>-2</sup> .K <sup>-1</sup>
Activation energy for anode reaction	55000 J.mol <sup>-1</sup> .K <sup>-1</sup>
Activation energy for cathode reaction	77000 J.mol <sup>-1</sup> .K <sup>-1</sup>
Gas heat capacity for anode channel	3.5R
Gas heat capacity for cathode channel	3.5R

**Table 4. Physical property of fuel cell particles**

Physical Properties	Electrolyte	Cathode Electrode	Anode Electrode	Separator Plates
Density (Kg.m <sup>-3</sup> )	1914	6794	8220	8000
Heat capacity (J.Kg <sup>-1</sup> .K <sup>-1</sup> )	4000	43524	444	500
Heat Conduction (W.m <sup>-1</sup> .K <sup>-1</sup> )	25	2.5	25	25

and were continuous in the time direction, then the obtained system of ODE was simulated in MATLAB software. For solving the obtained dynamic equations, ode23s was selected by trial and errors of solvers in MATLAB, but they were not found to be acceptable for our needs due to their stiffness. Then, ode15s was selected but was also rejected because of their low speed in solving the equations. Among other options, ode23t and ode23tb were not convergent. So, the only accountable solver was ode23s and the necessary runs were conducted with it. In this section, responses of state variables of systems to step input in the entry voltage of the system and flow rate of input fuel to the cell were discussed to study the dynamic behavior of the system.

### 3.1. Validation

One of the most significant parts of the simulation was to validate the proposed model with experimental data to prove the validity of its ability to predict real events. In this section, we compare the results of the simulation with experimental data existing in the

literature so that the accuracy and rationality of the simplifying assumptions of the model are verified. The document of Brouwer et al. [18] was chosen for our validation. They prepared an MCFC in an experimental scale and part of the polarization curve was presented. Their cell was maintained at a constant temperature by virtue of an experimental heater. To replicate this experimental work in our model, we first removed the solid phase temperature from variables of states so that we could keep the entire cell at a constant temperature. Second, we removed the combustion chamber from our model and used the composition of constituent of cathode from their work. Finally, we used all of their parameters. The result is shown in Figure 2.

The model mean error at 1 atm was almost 5.3%. While, much has been done to make our model resemble the above-mentioned experimental work, some of our model's parameters, such as the electrode's thickness, and were not perfectly similar to the work of Brouwer et al. Other sources of error can be other simplifying assumptions such as: constant properties (heat capacity, activation energy of electrochemical

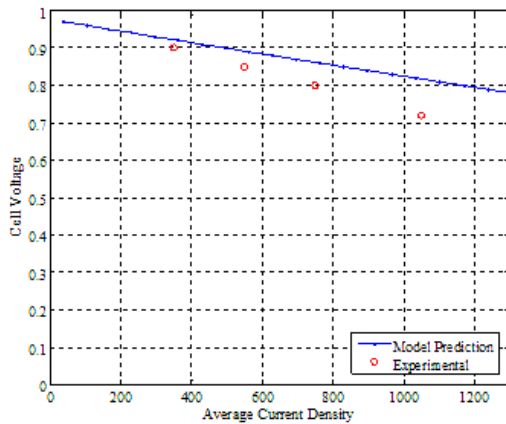


Fig. 2. Polarization curves for experimental data and model prediction.

reactions), ideal gas, and constant pressure for gas throughout the cell.

### 3.2. Step response of cell voltage

In this section, a step input of +5, +10, -5, and -10 percent of the cell voltage were allocated to the dynamic model of the system and the results of the simulation were introduced. Amongst the state variable of the system the behavior of the important variables of current density, solid temperature, hydrogen and methane concentration in the anode channel and oxygen concentration in the cathode were introduced in Figure 3 to 6.

As can be seen from Figure 3, voltage increase in the cell results in a reduction in current density in the cell and vice versa. Comparing the four diagrams introduced in the figure indicated that the amount of system productivity in these four step changes were not the same which shows the system non-linearity. However, the most significant point is the existence of two phases in the behavior of current density. The first phase is related to couple change of current density, gas concentration and gas temperature, and the second phase includes slow changes resulting from solid temperature.

As can be seen in Figure 4, as the voltage cell increases the solid temperature also increase and vice versa. This behavior indicates that the temperature of the MCFC

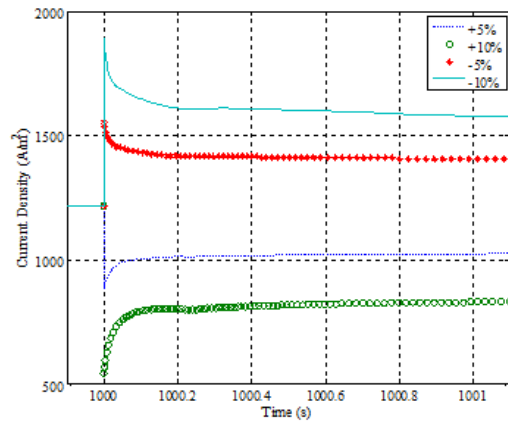


Fig. 3. Dynamics behavior of current density of fuel cell to step change of cell voltage.

solid was affected by the combustion reaction of spies more than other factors in the combustion chamber. When the system voltage increased, current density and thus hydrogen consumption reduces, so the amount of remaining hydrogen and also the amount of remaining methane increases based on the equations of equilibrium chemical reactions of water and gas in vapor in equilibrium with hydrogen concentration. Based on the fact that in the simulation all the fuels in the combustion chamber were assumed to be consumed liberating a lot of heat and the resulting gas ws injected into the anode channel giving the cell access to more heat.

Voltage increase led to current density decrease which caused the process to consume less oxygen. This

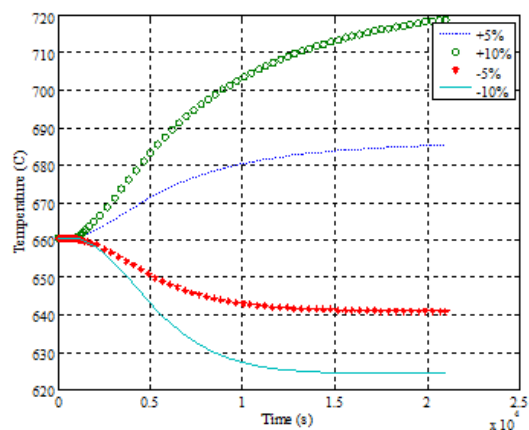
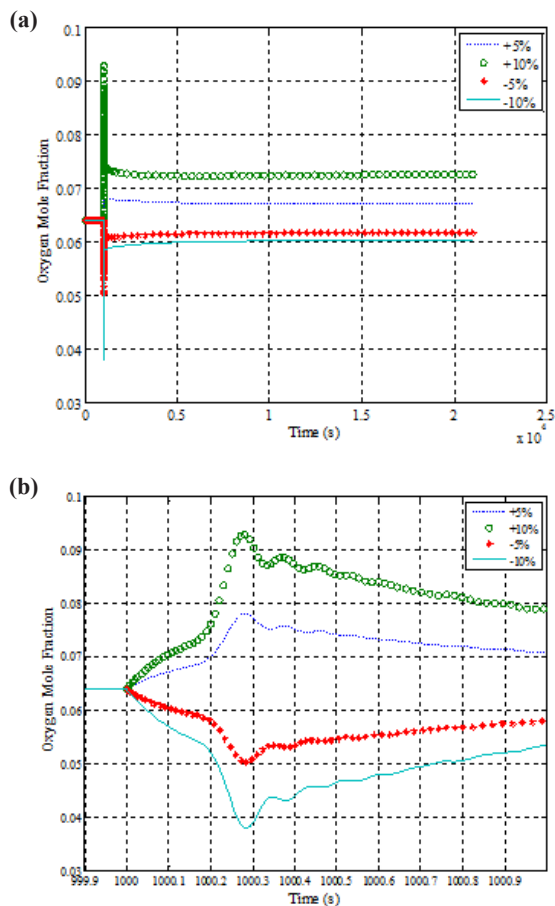


Fig. 4. Dynamics behavior of solid phase temperature of fuel cell to step change of cell voltage.

behavior was also seen in the cell voltage reduction. In addition to current density reduction, the oxygen consumption was reduced by the voltage increase, and the concentration of oxygen increased in the output stream of the cathode channel. But, taking into account that voltage increase results in an increase in flammable gases concentration in the combustion chamber which in turn requires more oxygen, so that the oxygen concentration in the input stream to the cathode channel decreases decreasing the oxygen concentration in the output cathode channel. These changes are shown in Figure 5. Oxygen concentration changes include various dynamics like other state variables.



**Fig. 5. Dynamic behavior of oxygen mole fraction at the cathode channel outlet (a) Total Response (b) Initial Response.**

By increasing the cell voltage, which is equivalent to current density reduction, hydrogen consumption decreases and increasing the hydrogen concentration

at the end of the cell. Three different steps were observed in the dynamic behavior of the hydrogen concentration (Figure 6(a)). In the first step increasing the voltage decreased the current density as well as hydrogen consumption which resulted in an increase in the hydrogen increase. In the second step a hydrogen concentration reduction resulted from a reduction in the methane concentration which is in equilibrium with the hydrogen concentration. In the third step an increase in the hydrogen and methane concentration in the output of the anode channel burned more constituents and increased the temperature of the cell which led to an increase in hydrogen concentration. Therefore, reforming reactions advanced more as can be clearly observed in the diagram of methane concentration (Figure 6(b)). An increase in cell voltage and subsequent current density reduction means hydrogen consumption was reduced in the anode channel increasing the hydrogen concentration in this channel. Therefore, the concentration of methane, which is in equilibrium with the hydrogen concentration, increases. These changes are given in Figure 6(b). As the voltage increases result in a current density reduction, the hydrogen concentration decreases so that the hydrogen concentration increases and prevents the transformation of methane, which results in an increase in the methane concentration. However, reforming reactions gradually advanced by increment of cell temperature to a point and methane concentration reduced to certain level.

### 3.3. Step response of fuel flow rate

In this section the input to the anode channel of the system is simulated by entering a step input of +5, +10, -5, -10 percent to fuel flow rate. Results of simulations are given in Figures 7 to 10.

As it is seen in Figure 7, an increase of fuel flow rate results in more current density and vice versa. Importantly, in the analysis of this diagram it can be seen that the system productivity is larger in a negative step change than a positive step change. The reason for this phenomena is the high fuel productivity at low fuel flow rates. This is because the remaining time



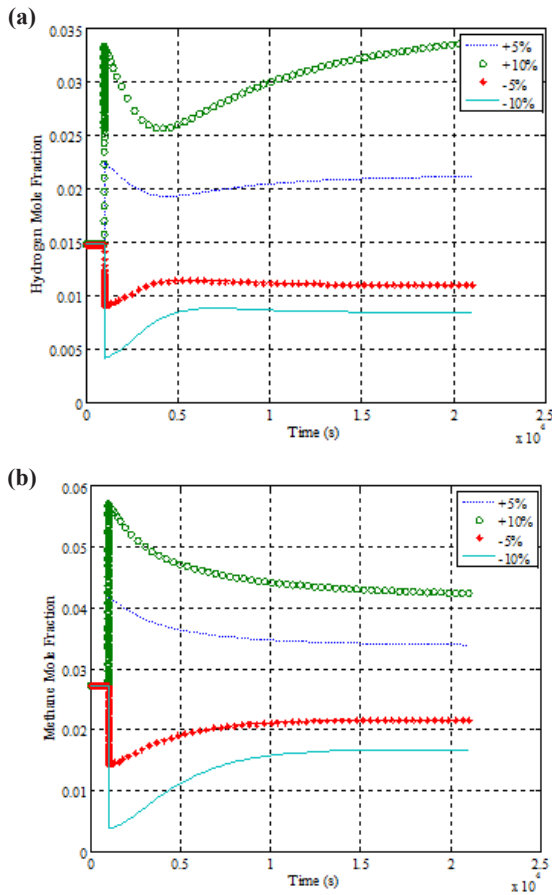


Fig. 6. Dynamic behavior of (a) hydrogen, and (b) methane molar fraction at the anode channel outlet.

for fuel at low flow rates is sufficient to give the opportunity of reaction. Owing to the fact that increasing the fuel flow rate results in higher amounts

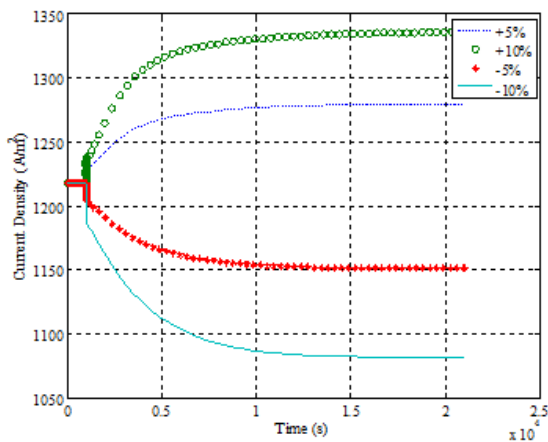


Fig. 7. Dynamics behavior of current density of fuel cell to step change of fuel flow rate.

of reactions which liberates more heat, increasing cell temperature and vice versa. This behavior is clearly seen in Figure 8.

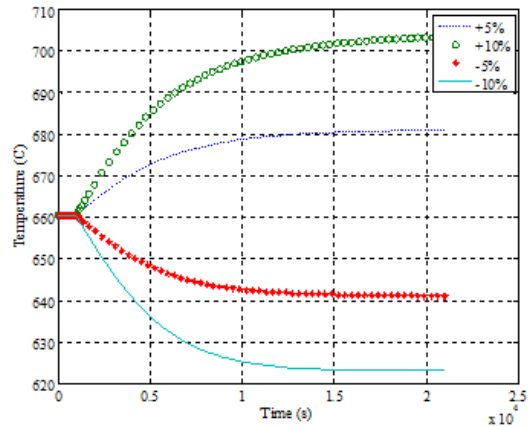


Fig. 8. Dynamics behavior of solid phase temperature to step change of fuel flow rate.

In Figure 9, it can be seen that increasing fuel flow rate reduces oxygen concentration and vice versa. The reason is that as the fuel flow rate increases, the flow rate of usable gases increases in the combustion chamber and more oxygen is consumed in the chamber; consequently, less oxygen reaches the cathode in the output of the chamber. Due to rapid dynamic behavior of oxygen concentration, the changes is plotted in Figure 9 (b).

Figure 10 (a) shows that increasing the fuel flow rate increases the hydrogen concentration in the anode channel. This is due to the fact the anode electrochemical reaction is not able to consume all added hydrogen. Three steps are shown in the diagram of hydrogen molar component change (Figure 10(a)): In the first step the methane concentration increases with an increase of fuel cell flow rate, which leads to an increment of hydrogen concentration. In the second step hydrogen decreases when using methane, because their concentrations are in equilibrium. In the third step: cell temperature increases more by burning  $H_2$  and  $CH_4$  and reforming reaction increases which leads to more hydrogen production. Considering Figure 10(b), it can be concluded that increasing the methane flow rate decreases the methane concentration due to the fact that it leads to higher cell temperature which

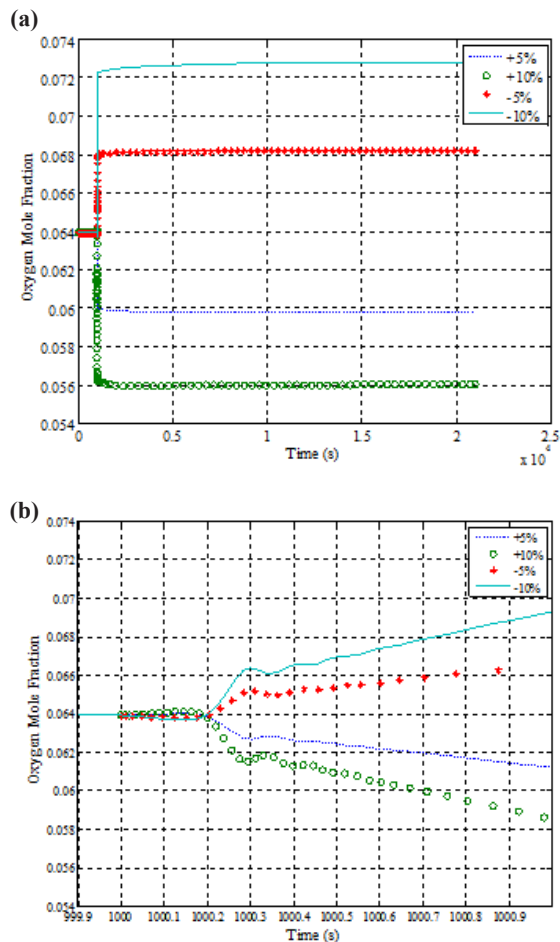


Fig. 9. Dynamic behavior of oxygen mole fraction at the cathode channel outlet (a) Total response (b) Initial response.

rapidly advances reforming reactions. Increasing the fuel flow rate increases the methane flow rate and accordingly the methane concentration. However, as time goes forward, reforming reactions advance more owing to higher cell temperature which leads to methane concentration reduction.

#### 4. Conclusions

Investigation of the performance of fuel cells based on theoretical modeling is superior to experimental measurements due to the fact that empirical approaches are time consuming and costly. In this paper, an extensive model for a molten carbonate fuel cell was introduced by employing partial differential

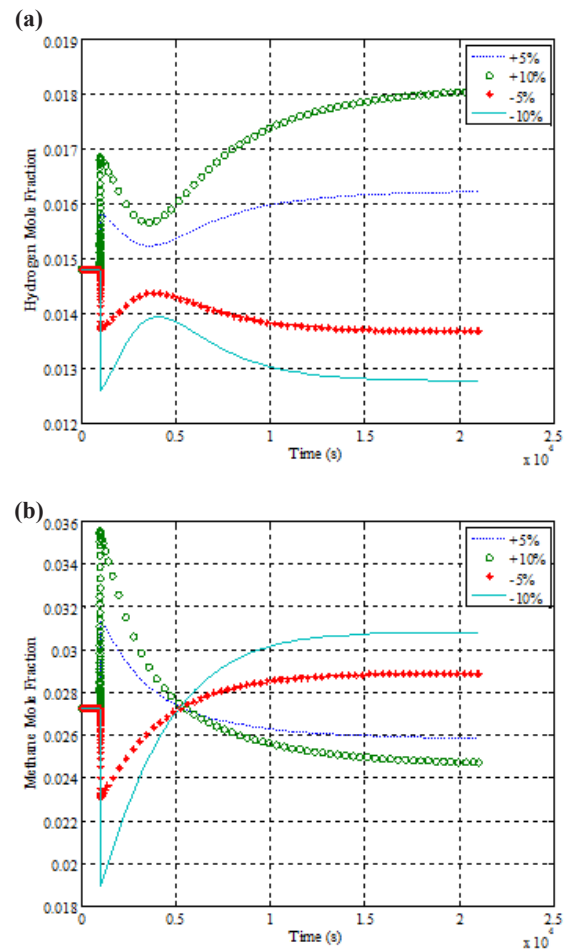


Fig. 10. Dynamic behavior of (a) hydrogen , and (b) methane mole fraction at the anode channel outlet.

equations. Then, the MCFC model was introduced by method of lines that was simulated in a dynamic state and the results of the simulation with step response in cell voltage and input fuel flow rate were studied. Results show that in the dynamic simulation with an input step of fuel flow rate, the system productivity in the negative echelon is smaller than the input positive echelon. This indicates that as fuel flow rate into the cell is less, the productivity of fuel gets higher. Also, in the analysis of the fuel cell, it has been seen that the temperature of molten carbonate depends strongly on the intensity of combustion in the combustion chamber. As methane, hydrogen and carbon monoxide concentration entering the combustion chamber increase, the heat released from combustion elevates and system temperature increases which results in

increasing thermal stress in the cell. Hence, methane and hydrogen flow rates are fundamental factors in controlling temperature and electrical current generation in molten carbonate fuel cell.

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