

INFLUENCE OF RADIATIVE HEAT TRANSFER ON VARIATION OF CELL VOLTAGE WITHIN A STACK

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ABSTRACT

In this study, a numerical investigation of cell-to-cell voltage variation by considering the impact of flow distribution and heat transfer on a stack of cells has been performed. A SOFC stack model has been previously developed to study the influence of flow distribution on stack performance (Burt, et al., 2003). In the present study the heat transfer model has been expanded to include the influence of radiative heat transfer between the PEN (positive electrode, electrolyte, negative electrode) and the neighboring separator plates. Variations in cell voltage are attributed to asymmetries in stack geometry and nonuniformity in flow rates. Simulations were done in a parallel computing environment with each cell computed in a separate (CPU) process. This natural decomposition of the fuel cell stack reduced the number of communicated variables thereby improving computational performance. The parallelization scheme implemented utilized a message passing interface (MPI) protocol where cell-to-cell communication is achieved via exchange of temperature and thermal fluxes between neighboring cells. Inclusion of radiative heat transfer resulted in more uniform temperature and voltage distribution for cases of uniform flow distribution. Non-uniform flow distribution still resulted in significant cell-to-cell voltage variations.

INTRODUCTION

In recent years emphasis has been placed on the development of affordable clean power sources. This has caused much speculation about the use of fuel cell technology in various endeavors; e.g., automobiles, stationary power generation, portable power supplies, etc. There are many fuel cell types, with the most common ones being: phosphoric-acid fuel cells (PAFC), solid-oxide fuel cells (SOFC), molten-carbonate fuel cells (MCFC), alkaline

fuel cells (AFC), proton exchange membrane (PEM), and Direct methanol fuel cells (DMFC) (Wójtowicz, et al., 2002). Regardless of the type of cell, stacks of cells in series can be used to generate desired voltage output and power. The SOFC shows a high potential for being an efficient and clean solution for stationary based power generation.

At the heart of a solid oxide fuel cell is the solid electrolyte (usually made of stabilized zirconia) which at temperatures greater than 600°C conducts oxygen ions from the porous cathode to the porous anode (Billingham, et al., 2000). At the triple interface (where the anode, electrolyte, and anode gas channel meet) oxygen ions react with hydrogen and carbon monoxide to form water and carbon dioxide, respectively. The anode electrode, electrolyte, and cathode electrode together are called PEN. Pressures are atmospheric or greater. In general, the gas flow through the anode and cathode gas channels results in forced convective heat transfer. SOFC usually operate at high temperatures in the range (700-1100°C) utilizing a variety of fuels (i.e. hydrogen gas, hydrocarbons, and carbon monoxide) (Billingham, et al., 2000, Yuan, et al., 2003, and Krotz, 2003). At these elevated temperatures thermal radiation emitted from the solid elements of the fuel cell should become a noticeable portion of the heat transfer within the stack. This paper considers the role of radiative heat transfer on uniformity in cell performance within a stack.

In order to reduce cost and increase fuel cell performance suitable designs must be developed. Effective computational models provide insight into design performance and allow developers the tools needed to start with a good design. Detail and speed are important considerations when developing useful tools. Generally one is traded off for the other in an attempt to find a cost effective solution. Detailed analysis of a SOFC stack using computational fluid dynamics (CFD) modeling requires immense resources and computational time and quickly becomes unreasonable for the modeling of large stacks

(stacks of 100+ cells). A less detailed 1-D model provides a means for the study of large stacks by reducing the problem to a performance study based on changes in the streamwise direction. An efficient and robust one-dimensional model of a fuel cell stack allows for quick parametric studies and hence is cost effective in design analysis. This is the approach used in the present analysis.

Careful distribution of fuel and oxidizer flow must be made to provide uniform and maximum power generation. Recently a one-dimensional single cell model was extended to the case of a stack of cells (Burt, et al., 2003) using domain decomposition for parallel execution of the code. In this paper the impacts of radiative and convective heat transfer as well as non-uniform flow distribution on cell-to-cell voltage variations are considered for a 5 cell planar SOFC stack. A relatively simple radiation model is included to provide an initial assessment of the impact of radiative transfer; in future work this model will be improved. The primary objective is to study and understand possible reasons for cell-to-cell performance, and the physical factors that may mitigate these variations, so that new strategies can be planned to achieve uniformity among individual cells.

COMPUTATIONAL MODEL

The fuel cell stack has been divided into computational domains using domain decomposition with each cell solved as a separate process under a multitasking environment. Communication between domains or processes was accomplished using the Message Passing Interface (MPI) library. Each cell was further divided into smaller control volumes. A one dimensional model was implemented where the variations in the streamwise (x-) direction are explicitly calculated but those in the other directions are accounted for via integral approximations. The fuel cell anode gas channel, electrolyte plate, cathode gas channel, and separator plate were divided in the streamwise direction into control volumes. Each control volume of the fuel and air gas channels was required to satisfy the following governing equations for mass, momentum, and energy, respectively:

$$\nabla \frac{\partial \rho}{\partial t} + (\rho u A_{xs})_w - (\rho u A_{xs})_e = \dot{m}_{\text{surf}} \Delta x l_w \quad (1)$$

$$\nabla \frac{\partial (u\rho)}{\partial t} + (u\rho u A_{xs})_w - (u\rho u A_{xs})_e = \sum F_x \quad (2)$$

$$\nabla \frac{d(e\rho)}{dt} + (e\rho u A_{xs})_w - (e\rho u A_{xs})_e = \dot{Q}_{\text{net}} \quad (3)$$

All symbols used in this paper are defined in the nomenclature section. Specie mass conservation was satisfied using

$$\frac{\nabla \partial (\rho Y_k)}{\partial t} + (\rho Y_k u A_{xs})_w - (\rho Y_k u A_{xs})_e = \dot{\omega}_k \Delta x l_w \quad (4)$$

In Eqs. (1-4) it is assumed that changes in the x-direction are small therefore diffusion terms are neglected i.e. boundary layer assumption. The energy equation (3) is used to determine the temperature, and the current density is determined from a simplified Butler-Volmer equation (Gemmen et al., 2000). Then, the current density and temperature are used to calculate appropriate fluxes which are introduced as source (or sink) terms for each of the conservation equations. The molar flux of a given species k is obtained from the current density using:

$$\omega_k = \frac{-i_{\text{den}}}{2\alpha_k F} \quad (5)$$

The PEN and separator plate are considered to be made of solid material therefore only the energy equation was solved; in these regions the energy equation, Eq. (3) can be simplified to:

$$\nabla \frac{d(e\rho)}{dt} = \dot{Q}_{\text{net}} + \dot{Q}_{\text{gen}} \quad (6)$$

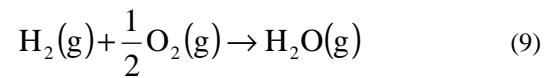
The radiative and convective heat flux through the surface are included in \dot{Q}_{net} (see Eq. 16), and the heat source, \dot{Q}_{gen} , is obtained from ohmic heating and heat released by the formation of H₂O resulting in the following expression:

$$\dot{Q}_{\text{gen}} = i_{\text{den}}^2 R + \frac{i_{\text{den}} \Delta H_{\text{H}_2\text{O}}}{nF} \quad (7)$$

Pressure, P, is calculated from:

$$P = \rho R_u T \quad (8)$$

The electrochemistry model is based on the assumption that the overall chemical reaction occurring in the fuel cell is:



Calculation of the cell potential starts with the Nernst Equation:

$$E = E^0 + \frac{R_u T}{2F} \ln \left[\frac{[X_{\text{H}_2}][X_{\text{O}_2}]^{1/2}}{[X_{\text{H}_2\text{O}}]} \right] + \frac{R_u T}{4F} \ln \frac{P}{P^0} \quad (10)$$

The pressure is assumed to be the same for both the anode and cathode gas channels. The reversible potential at

standard state conditions is obtained from the change in the standard Gibbs free energy.

$$E^0 = -\frac{\Delta G^0}{nF} \quad (11)$$

The corrected cell potential, E_{cor} , is obtained by subtracting the ohmic (L_R), concentration (L_C), and activation (L_A) losses (i.e. overpotentials) from the ideal Nernst potential, E :

$$E_{cor} = E - L_R - L_C - L_A \quad (12)$$

The overpotentials are related to the current density through empirical relations that represent Volmer-Butler equation in the limit of uniform species concentration

$$L_R = i_{den} R_{net} \quad (13)$$

$$L_C = -\frac{R_u T}{nF} \ln \left(1 - \frac{i_{den}}{i_L} \right) \quad (14)$$

$$L_A = \frac{R_u T}{n\alpha F} \ln \left(\frac{i_{den}}{i_0} \right) \quad (15)$$

A quasi-steady gas option was used whereby the gas flow was determined from empirical steady state relations, e.g. a steady state friction coefficient equation. This allowed large time steps to be used with the time marching scheme to reach a steady state solution. More details about the mathematical model can be found in previous work (Burt, et al. 2003, Gemmen, et al., 2000).

Convective and Radiative Heat Transfer

When considering the heat flux from the PEN and separator plate there are two main modes of heat transfer. Convective heat transfer between the solid and gas phase, and radiative heat transfer between the solid and the neighboring solid surfaces. These are both included in \dot{Q}_{net} , the net boundary heat flux through the top and bottom surfaces of the computational volume, in Eq. (6). Thus \dot{Q}_{net} is obtained from

$$\dot{Q}_{net} = \dot{Q}_{conv}|_n - \dot{Q}_{conv}|_s + \dot{Q}_{rad}|_n - \dot{Q}_{rad}|_s \quad (16)$$

The convective heat transfer rate is given by

$$\dot{Q}_{conv} = h_c A_{xs} (T_{surf} - T_{env}) \quad (17)$$

An empirical Nusselt number correlation is used of the form

$$Nu = f(Re, Pr) \quad (18)$$

which, in turn, is used to calculate h_c from

$$Nu = \frac{h_c L}{k} \quad (19)$$

In general the radiative heat transfer between two surfaces 1 and 2 can be calculated from

$$\dot{Q}_{rad} = A_1 F_{12} (\epsilon_1 \sigma T_1^4 - \epsilon_2 \sigma T_2^4) \quad (20)$$

Initially, when considering the radiative heat transfer several simplifying assumptions were made (Mills, 1995). First, the gas medium between the surfaces is considered to be nonparticipating. The PEN and separator plate are considered to have black surfaces (having an emissivity, $\epsilon = 1$). The view factor, F , is assumed to be 1. Therefore all emitted radiation is considered to be absorbed by the surface of the opposite plate. With these simplifications the radiative heat transfer rate, \dot{Q}_{rad} , can be expressed as

$$\dot{Q}_{rad} = A_{xs} \sigma (T_1^4 - T_2^4) \quad (21)$$

It is not known what the exact radiative properties are for a generic fuel cell. Therefore in this study the worst case is compared to the best case. Equation (21) gives a conservative estimate for the heat transfer rate with maximum emissivity. Considering heat absorbed by a participating medium (i.e. gases in the anode and cathode channels) and modeling the surfaces of the electrodes and separator plates as grey surfaces would reduce the overall influence of the radiative heat transfer.

Creation of a Stack Model

In our simulation approach we exploit the modular structure of a fuel-cell stack, which enables a straightforward application of the domain decomposition technique for parallel implementation of the code. This is done by simulating each cell with a separate process in a multi-processor computing environment. Since the processes inside each fuel cell are relatively independent from other cells and are coupled only through the well defined fluxes (i.e. heat, mass, current), and the given boundary conditions (voltage, temperature, pressure, etc.), it is possible to arrange a stable and time accurate parallel iteration procedure for a coupled solution of cell properties in the stack without excessive communication overhead. In this implementation temperature, thermal (convective and radiative) fluxes, time step, and termination bit are communicated between processes using MPI (Message

Passing Interface) to form the stack geometry. This parallel solver can be run on a distributed memory computer platform, such as a Beowulf cluster.

A stack of cells can be modeled using several instances of individual cells. MPI library calls are used to communicate variable data between neighboring cells. Each cell in a stack is considered to be in series therefore the same total current is maintained by all cells. The smallest time step is used by all cells, because the stack of cells must be solved using the same time stepping.

Because the separator plate is not porous only thermal transfer must be considered between neighboring cells. The temperature of the separator plate of each cell is communicated with the neighboring cell below it. The convective and radiative heat fluxes are then calculated using the received temperature from the neighbor. These fluxes are then shared between cells and are used when calculating the temperature of the separator plate, anode gas channel, and electrolyte plate at the new time level.

Each cell process is executed independently therefore it is important for a break signal to be communicated to all cells. In this way if one cell must stop execution all cells will also stop. Through careful communication it is therefore possible to solve a stack of cells using individual processes. The limited number of variables that must be communicated makes decomposition of a fuel cell stack a prime candidate for parallel programming.

APPLICATION

The stack model was applied to the case of a 5 cell planar SOFC stack. Figure 1 depicts the physical geometry of a single cell (or unit cell) when visualized in a one-dimensional sense. The fuel and oxidizer are introduced to the cell in a coflow configuration. For illustration, in Figure 1 the stack has been divided in the x-direction into five computational nodes (denoted by i , and used with finite volume analysis to discretize the governing equations) using dashed lines. The active area of the cell is modeled by the computational nodes 1, 2, and 3. In this study the cell was divided in the streamwise direction into 20 computational nodes. T_{Top} and T_{Bottom} are extra storage arrays used for communication of temperature between neighboring cells in the stack. In the case of the top and bottom cells these arrays are also used to specify ambient temperatures useful for modeling heat transfer to the stack surroundings. In the current study the heat flux from the fuel cell stack to the surroundings is zero, representative of a perfectly insulated wall boundary condition on the top and bottom of the stack.

The physical geometry of the stack is defined by the length of the cell in the streamwise direction, and the thickness of each component. These dimensions are listed in Table 1. The PEN has a thickness defined by the sum of the thickness of anode electrode, electrolyte, and cathode electrode. In addition to the thickness of electrolyte plate the anode and cathode gas channel gap thicknesses must be

specified. For the current study an electrolyte supported cell geometry was considered.

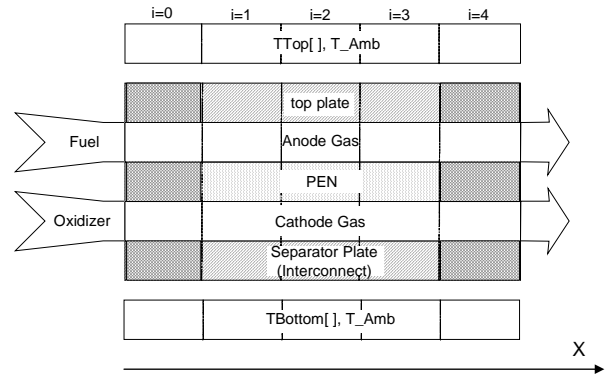


Figure 1 Physical geometry of a single cell

Table 1 Physical dimensions of single fuel cell with Electrolyte support

SOFC Component	Thickness,[m]
Cell Length	1.5E-01
Anode Gas Channel Gap	1.0E-03
Cathode Gas Channel Gap	3.0E-03
Electrolyte Thickness	1.8E-04
Anode Electrode Thickness	5.0E-05
Cathode Electrode Thickness	5.0E-05
Separator Thickness	2.0E-03

Material properties and model parameters listed in Table 2 were taken from an earlier single cell model (Gemmen, et al., 2000). The stack is considered to be homogeneous with all the cells being constructed with the same physical dimensions and material properties. The inlet and outlet boundary conditions applied to the governing equations (continuity, energy conservation, species-mass conservation, and momentum equation) and electrochemistry model are specified as model parameters in Table 2.

Table 2 Material properties and Model Parameters

Cell Heat Capacity [J/kg-K]	8.00E+02
Cell Density [kg/m ³]	1.50E+03
Separator Heat Capacity [J/kg-K]	4.00E+02
Separator Density [kg/m ³]	8.00E+03
No. Axial Nodes	20
Anode Inlet Temperature [K]	1.17E+03
Anode Inlet Pressure [Pa]	1.01E+05
Anode Exit Pressure [Pa]	1.01E+05
H ₂ Anode Inlet Mole Fraction	9.70E-01
H ₂ O Anode Inlet Mole Fraction	3.00E-02
Cathode Inlet Temperature [K]	1.17E+03
Cathode Inlet Pressure [Pa]	1.01E+05
Cathode Exit Pressure [Pa]	1.01E+05
O ₂ Cathode Inlet Mole Fraction	2.10E-01
N ₂ Cathode Inlet Mole Fraction	7.90E-01

A numbering convention was used to identify the cells in a manner similar to levels in a building (with cell number increasing in the vertical direction). The inlet velocities prescribed are given in Table 3. For the non-uniform case the flow was redistributed from cell 0 to cell 1. These inlet conditions were imposed for both cases with and without radiative heat transfer.

Table 3 Prescribed inlet velocity [m/s]

Cell number	Uniform flow distribution		Non-uniform flow dist.	
	anode	cathode	anode	Cathode
4	4.07E-01	1.21E+00	4.07E-01	1.21E+00
3	4.07E-01	1.21E+00	4.07E-01	1.21E+00
2	4.07E-01	1.21E+00	4.07E-01	1.21E+00
1	4.07E-01	1.21E+00	4.88E-01	1.21E+00
0	4.07E-01	1.21E+00	3.26E-01	1.21E+00

Fuel utilization is of major concern and therefore fuel mass flow rates are generally low to insure high H₂ utilization.

RESULTS AND DISCUSSION

Variations in cell voltage are present as a result of asymmetries in stack geometry and flow rate. In this study steady state solutions are compared for a 5 cell planar SOFC stack providing a total current of 600A. Air was supplied to the cathode gas channel such that there was c.a. 20% oxygen utilization (Table 3). The higher mass flow rate resulted in the cathode side of the cell being cooler than the anode side. This resulted in non-uniform temperature distributions throughout the stack (Figure 2a).

Results with and without radiative heat transfer can be compared by considering the pseudo 2-D solution obtained. The individual components of the fuel cell are considered using one-dimensional layers e.g. fuel channel (layer 1), PEN (layer 2), air channel (layer 3) etc. Communication of fluxes and current density, between layers (as explained above) allows for what may be called a pseudo 2-D solution to be obtained. This approach used in the single cell model is expanded to include multiple cells in a stack with temperature, heat flux, and total current communicated between neighboring cells. Thus, the model has effectively become two-dimensional via layering and stacking. This allowed extracting two-dimensional temperature contours (see Figures 2a&b), albeit the grid in the transverse direction (y-direction normal to the flow direction) is coarse due to the nature of the model.

Figures 2a&b show that the addition of radiative heat transfer improved the uniformity of the temperature distribution within the stack (Figure 2b).

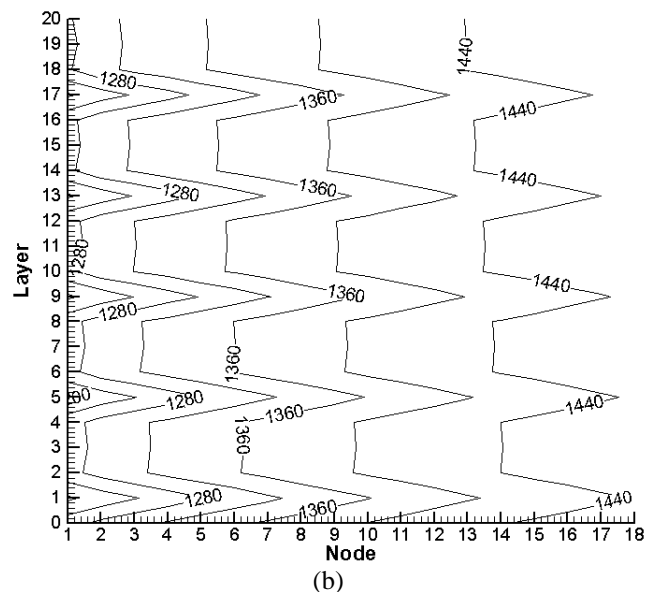
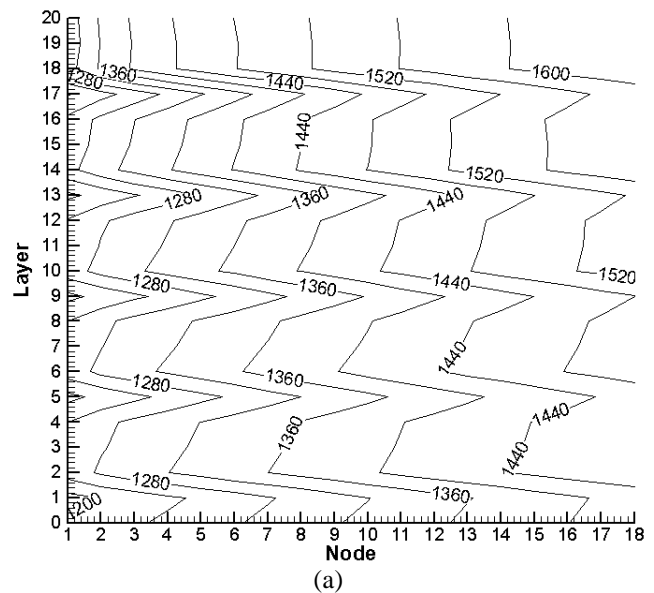


Figure 2 Temperature contours for uniform flow case (a) without radiative heat transfer (b) with radiative heat transfer

The open circuit (or Nernst) voltage and the Ohmic losses are functions of the temperature. Therefore variations in cell voltage are observed when there are temperature variations. When considering heat transfer only in a purely convective form even under uniform flow conditions noticeable variations in cell performance are observed (Burt, et al., 2003). Figure 3 shows that for the non-radiative case (purely convective heat transfer) a variation of 3.6% was obtained. When radiative heat transfer was included the same trend in voltage variation occurred but with a variation of only 0.4%.

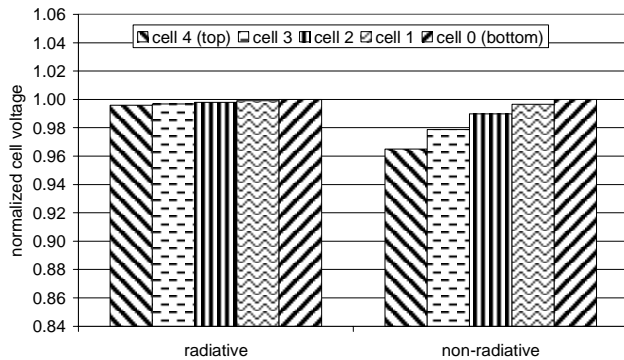


Figure 3 Variation in cell voltage for uniform flow distribution with and without radiative heat transfer; In each case the cell voltage is normalized with the highest cell voltage.

Even larger variations in cell voltage were observed while performing a parametric study on the impact of flow distribution. It was shown that the greatest voltage variation occurred when a 20% of the fuel flow in the bottom cell (cell 0) was directed to the neighboring cell (cell 1) (Burt, et al., 2003). Radiative heat transfer did not decrease the voltage variation caused by this non-uniform distribution. In both this study and the previous study it was found that a redistribution of the fuel mass flow rate resulted in up to 12.3% variation in cell voltage (Figure 4). This variation mostly results from the mole fraction of H_2 in Nernst Equation (Eq. 10). However the voltage variations resulting from temperature distribution were much smaller when radiative heat transfer was included.

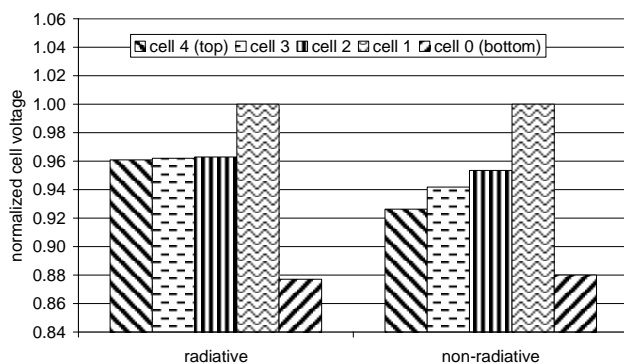


Figure 4 Variation in cell voltage for non-uniform flow distribution with and without radiative heat transfer; In each case the voltage is normalized with the highest voltage.

Although the inclusion of the radiative heat transfer reduces cell-to-cell voltage variations, significant variations are still present due to mal-distribution of fuel and oxidizer flow within the stack.

CONCLUSIONS

The results of this study indicate that the variations in voltage among cells in a stack can be influenced by mal-distribution of fuel and oxidizer flows. Cell-to-cell voltage variations occur partially due to the temperature non-uniformities within a stack of cells which results from asymmetry in planar stack design (Burt, et al., 2003). The temperature gradient results in convective and radiative heat transfer among solid-gas and solid-solid components within the stack which helps to mitigate the temperature non-uniformities. When only convection/conduction heat transfer is considered large variations in cell performance was observed. The inclusion of radiative heat transfer in the mathematical model improved the uniformity of the temperature distribution within the stack thus leading to more uniform cell voltages. For the case of uniform flow distribution with radiation heat transfer the cell-to-cell voltage variation was found to be 0.4%. This is much smaller than the 3.6% variation obtained previously when considering only conduction/convection heat transfer. However, redistribution of 20% of the fuel mass flow rate resulted in a 12.3% variation in cell voltage which is comparable to the case computed previously without radiation. The present radiation model is quite simple in that it accounts only for first order effects. These trends need to be verified in a future work with more refined radiation model.

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NOMENCLATURE

A_1	Area of surface 1 [m ²]
A_{XS}	Cross-sectional area [m ²]
e	Energy per unit mass [J/kg]
E	Open circuit potential [V]
E^0	Potential at standard state conditions [V]
E_{cor}	Corrected potential [V]
F	Faraday constant [C/mol]
F_{12}	Shape factor from surface 1 to 2 [1]
F_x	Forces in x-(streamwise-)direction [N]
G	Gibbs free energy
ΔH_{H_2O}	Heat of formation for H ₂ O [kJ/kgmol]
h_c	Convective heat transfer coefficient [W/m ² K]
i_{den}	Current density [A/m ²]

i_0	Exchange current [A/m^2]
k	Thermal conductivity [$W/m\ K$]
l_w	Width of control volume [1 m]
L	Cell length [m]
L_A	Activation loss [V]
L_C	Concentration loss [V]
L_R	Ohmic loss [V]
\dot{m}_{surf}	Mass flux through the surface [$kg/m^2\ s$]
n	Number of participating electrons
Nu	Nusselt number [1]
P	Pressure [Pa]
P^0	Reference pressure [Pa]
Pr	Prandtl number [1]
\dot{Q}_{conv}	Convective heat transfer rate [W]
\dot{Q}_{gen}	Rate of heat generation [W]
\dot{Q}_{net}	Net heat transfer rate [W]
\dot{Q}_{rad}	Radiative heat transfer rate [W]
R	Resistance [Ω]
Re	Reynolds number [1]
R_{net}	Net resistance [Ω]
R_u	Universal gas constant
t	time [s]
T	Temperature [K]
T_{surf}	Surface temperature [K]
T_{env}	Temperature of the environment [K]
u	velocity in x-direction [m/s]
Δx	Length of control volume in x-direction [m]
X_k	Mole fraction [1]
Y_k	Mass fraction [1]

Greek

α	Transfer coefficient [1]
ε	Total emittance [1]
σ	Stefan-Boltzmann constant [$W/m^2\ K$]
ρ	Mixture density [kg/m^3]
$\dot{\omega}_k$	Rate of formation and destruction of specie k
∇	Volume [m^3]

Subscripts and Superscripts

1	Surface 1
2	Surface 2
k	k^{th} specie
e	East face of control volume
env	Environment
H_2	Hydrogen gas
H_2O	Water vapor
n	North face of control volume
O_2	Oxygen gas
s	South face of control volume
surf	Surface of control volume
w	West face of control volume
xs	Cross-sectional area

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