ANALYSIS OF NONUNIFORM CELL VOLTAGE DISTRIBUTION IN A PEMFC STACK

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ABSTRACT
The nonuniform cell voltage distribution greatly influences design complexity, heat management and system performance, such as life time, reliability and output power, etc., in a real PEMFC system. To get insight into the corresponding influence factors, a fully coupled non-isothermal, electrochemical and transport 3D model for a 10-cell PEMFC stack with coolant channels is set up in this paper and the nonuniform characteristics are reconstructed under the defined operating conditions by simulations. It is found that the temperature profile in the stack has an important impact on the cell voltage distribution through influencing the distribution of activation potential and ohmic overpotential.

1 INTRODUCTION
For practical application, by having multiple cells electrically connected in series and/or parallel, the voltage and/or current of a PEMFC stack will be multiplied to output a required high power. However, connecting a number of cells to form a stack may result in some undesired phenomena, such as the uneven distributions of reactant gas flows, by-product water accumulation and nonuniform heat transfer and removal [1], and furthermore, lead to the nonuniform distributions of individual cell voltages and finally, cause the overall performance drop and the life time decrease. Hence, it is critical for these issues to be dealt with optimal design and safe operations of a PEMFC stack.

To the related topics, considerable efforts have been reported [1–18]. J. P. Hensel et al [2] showed the importance of cell-to-cell fuel flow balancing on fuel cell through experimental data and presented a fuel cell energy management technique that demonstrated the ability to improve stack performance. A. Friedl et al [3] investigated the interdependence between the internal stack gas distribution configuration and single cell/overall stack performance by operating a laboratory-scale fuel cell stack with different gas distribution configurations and different operating parameters. C. Y. Wen et al [4] experimentally investigated the effects of various combinations of bolt configuration and clamping torque on the corresponding contact pressure distributions and performances of a 10-cell stack. Measurement of ohmic voltage losses in individual cells of a PEMFC stack was presented by T. Mennola et al [5]. Some experiments on the hydrogen pressure drop in various operating pressures, temperatures, flow rates and stack current conditions were carried out by P. Pei et al [6].

J. Park and X. Li [7] developed a non-isothermal stack model to analyze the effects of flow variance and temperature distribution on the performance of a PEMFC stack. Their results indicated that the temperature effect was dominant on the cell voltage variance when the flow variance was small for sufficiently uniform distribution of reactant flows among the cells in the stack. S. Y. Choe et al [8–11] proposed highly dynamic PEMFC stack models, taking into account the most influential property of temperature. In general, these models were lumped parameter model, and could not deal with the spatial distributions...
of some important variables, such as reactant concentration, temperature, current density, and so on.

In [12], a complete 3D CFD simulation was developed to study how the baseline operation and original design of the stack impact the local temperature, water content, water transport, and kinetic variables inside the individual cells. Though the uneven distributions of cell voltages and temperature were described, it was a lack of deep analysis of the relationship between them. C. H. Cheng and H. H. Lin [13] performed numerical simulation of transport phenomena in a 6-cell PEMFC stack by adjusting the gas channel size of individual cells to obtain evenly distributed cell voltages. One of their conclusion was that an even voltage distribution can be obtained if the distribution of reactant gas is uniform among the cells and not drying out in the cells. However, the energy conservation was not considered in their model and the temperature effects on the degree of the nonuniform distribution of single cell voltage was not taken into account.

The above brief review shows that there are few papers pursuing the original reason of nonuniform cell voltage distribution inside the stack. This paper is devoted to analyzing the key factors related to this problem and proposing operation strategies to eliminate the undesired nonuniform cell voltage distribution.

2 NUMERICAL MODEL

2.1 Assumptions and domain

Assumptions made are as follows [8, 19]:

1. Ideal gas mixture.
2. Isotropic and homogenous electrode, catalyst layers and membrane.
4. No contact resistance.
5. Negligible ohmic overpotential due to electronic resistance.
6. Neglecting the inherent manufacturing tolerances of the cells.

Based on these assumptions, a layer-based model for a 10-cell stack with coolant channels is developed. The schematic diagram of the simulation domain for the stack is shown in Fig. 1, where 1, 2, and 3 denote the cathode inlet, coolant inlet, and anode inlet; 4, 5, and 6 the anode outlet, coolant outlet, and cathode outlet, respectively. For the sake of simplicity, ten cells are numbered from right to left in increasing order, starting at #1.

The input variables for the model are stack output voltage, reactant mass flow rate, the gas components fraction, temperature, pressure, and relative humidity of reactants as well as the temperature and flow rate of coolant at the inlets.

The model is described as follows based on the above assumptions and domain. Static behaviors of single cell in the stack under different coolant mass flow rates will be described in section 3.

2.2 Governing equations

A fully coupled non-isothermal, electrochemical and transport 3D model of PEMFC stack, consisting of five conservation laws of, i.e., mass, momentum, species, energy, and charge, can be presented in a concise form as follows:

\[
\nabla \cdot (\rho \vec{u}) = 0 \tag{1}
\]

\[
\nabla \cdot (\rho \vec{u} \vec{a})/\varepsilon^2 = \nabla \cdot (\vec{\tau} + \vec{p} + S_a) \tag{2}
\]

\[
\nabla \cdot (\rho \vec{u} \vec{Y}_i) = -\nabla \cdot (\vec{J}_i) + S_i \tag{3}
\]

\[
\nabla \cdot (\rho \vec{u} E) = \nabla \cdot (k_{eff} \nabla T) + S_T \tag{4}
\]

\[
\nabla \cdot (\sigma_m \nabla \phi_m) + S_m = 0 \tag{5}
\]

\[
\nabla \cdot (\sigma_c \nabla \phi_c) + S_c = 0 \tag{6}
\]

The species conservation (Eq. (3)) includes the coolant besides hydrogen, water and oxygen. This is an improvement in contrast with Ref. [13]. The physical, electrochemical, heat and mass transport properties and the source terms \(S_a\), \(S_i\), \(S_T\), \(S_m\) and \(S_c\) are referred to [20–22].

To solve the above PDEs, the corresponding boundary conditions shall be defined as follows:

Inlet boundaries: The anode/cathode inlet velocities can be expressed by the respective stoichiometric flow ratios, i.e., \(\zeta_a\) or \(\zeta_c\), defined at a reference current, \(I_{ref}\), as:

\[
u_{in,a} = \frac{I_{ref} M_{H_2} \zeta_a}{2F \rho_a A_{in,a} Y_{H_2}} \quad \text{and} \quad \nu_{in,c} = \frac{I_{ref} M_{O_2} \zeta_c}{4F \rho_c A_{in,c} Y_{O_2}} \tag{7}
\]

The inlet molar fraction, \(X_{H_2O}\), is determined by the inlet pressure and humidity condition:

\[
RH = \frac{X_{H_2O} P}{P_{sat}(T)} \tag{8}
\]

where \(RH\) is the relative humidity, \(P\) the absolute pressure, and \(P_{sat}(T)\) the water vapor saturation pressure which is a function of temperature only [20]. Based on Eq. (8) and the relationship between mass fraction and molar fraction shown in [21], the mass fraction of each species can be computed. Therefore, once we specify the inlet \(P, T, RH, \zeta_a, \zeta_c\), and \(I_{ref}\), the mass fraction, \(Y_i\), and inlet velocity, \(u_{in}\), shall be derived.
Outlet boundaries: Fully developed or no-flux conditions are applied:
\[ \frac{\partial \vec{u}}{\partial n} = 0, \quad \frac{\partial P}{\partial n} = 0, \quad \frac{\partial T}{\partial n} = 0, \quad \frac{\partial Y_i}{\partial n} = 0, \quad \frac{\partial \phi_m}{\partial n} = 0, \quad \frac{\partial \phi_s}{\partial n} = 0 \] (9)

Walls: No-slip and impermeable velocity condition and no-flux condition are applied:
\[ \vec{u} = 0, \quad \frac{\partial P}{\partial n} = 0, \quad \frac{\partial Y_i}{\partial n} = 0, \quad \frac{\partial \phi_m}{\partial n} = 0 \] (10)

In addition, the boundary conditions for the electronic phase potential, \( \phi_s \), and temperature, \( T \), at the bipolar plate outer surfaces can be expressed as [19, 20]:
\[
\begin{align*}
\phi_s &= 0, \quad T = T_0 \quad \text{Anode bipolar plate of cell #1;} \\
\phi_s &= V_{\text{stack}}, \quad T = T_0 \quad \text{Cathode bipolar plate of cell #10;} \\
\frac{\partial \phi_s}{\partial n} &= 0, \quad \frac{\partial T}{\partial n} = 0 \quad \text{Otherwise.}
\end{align*}
\] (11)

### Table I. Geometrical and operating parameters

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas channel depth</td>
<td>2.0 mm</td>
</tr>
<tr>
<td>Gas channel and shoulder width</td>
<td>2.0 mm</td>
</tr>
<tr>
<td>GDL thickness</td>
<td>0.3 mm</td>
</tr>
<tr>
<td>Catalyst layer thickness</td>
<td>0.01 mm</td>
</tr>
<tr>
<td>Number of fuel cell in stack</td>
<td>10</td>
</tr>
<tr>
<td>Porosity of GDL/catalyst layer</td>
<td>0.8/0.6</td>
</tr>
<tr>
<td>Thermal conductivity of membrane</td>
<td>2 W/(m·K)</td>
</tr>
<tr>
<td>Thermal conductivity of catalyst layers</td>
<td>8 W/(m·K)</td>
</tr>
<tr>
<td>Thermal conductivity of GDL</td>
<td>8 W/(m·K)</td>
</tr>
<tr>
<td>Thermal conductivity of bipolar plate</td>
<td>220 W/(m·K)</td>
</tr>
<tr>
<td>Permeability of GDL</td>
<td>10^{-12} m^2</td>
</tr>
<tr>
<td>Electronic conductivity in the GDL/land</td>
<td>5000/10^6 /(Ω·m)</td>
</tr>
<tr>
<td>Anode/cathode inlet pressure</td>
<td>1.0/1.0 atm</td>
</tr>
<tr>
<td>Stoichiometry, ( \zeta_a/\zeta_c )</td>
<td>3.0/3.0</td>
</tr>
<tr>
<td>Inlet temperature of gas flows, ( T_0 )</td>
<td>353 K</td>
</tr>
<tr>
<td>Inlet temperature of coolant</td>
<td>333 K</td>
</tr>
<tr>
<td>Inlet humidity at both sides</td>
<td>40%</td>
</tr>
<tr>
<td>Open circuit voltage, ( V_{\text{oc}} )</td>
<td>9.5 V</td>
</tr>
<tr>
<td>Stack output voltage, ( V_{\text{stack}} )</td>
<td>6 V</td>
</tr>
<tr>
<td>Reference current, ( I_{\text{ref}} )</td>
<td>1.8 A</td>
</tr>
</tbody>
</table>

### Numerical procedures:

The governing equations, Eq. (1)-(6) along with their appropriate boundary conditions, are discretized by the finite volume method, and solved in the commercial CFD software package, Fluent (version 6.3.26), by the SIMPLE (semi-implicit pressure linked equation) algorithm. The source terms and physical properties are implemented in a UDF (user-defined function) and the species and charge transport equations are solved through the software’s user-defined scalars. For the sake of accuracy and convergence, a hexahedral mesh with 487,326 grid cells in the computation domain is used. Geometrical and operating parameters used are listed in Table I. In addition, the overall mass balances of species are checked besides the equation residuals as an important convergence criterion. The mass balance check of these species ensures the correctness of the obtained numerical results, mathematically. For all the simulations to be presented in next section, the errors of mass balance of species (i.e., \( \text{H}_2, \text{H}_2\text{O}, \text{O}_2 \)) are all less than 1% and the equation residuals smaller than 5% [25–27].

### 3 RESULTS AND DISCUSSION

Fig. 2 shows the output voltages of each cell inside the stack. It can be seen that the output voltages of the ten cells are not identical. Cell #2 gives the highest voltage, 0.6218V, and it is followed by the cells #1, #3, and #10, whose output voltages are all above 0.6V. Nevertheless, the lowest output voltage is revealed to cell #6, as 0.5865V, followed by the cells #7, #5, #8, #4, and #9, whose output voltages are all below 0.6V. Note that the cells which give the lower output voltage are positioned in the middle of the stack. In contrast, the cells having the higher voltage are located at two ends. In order to investigate the main influencing factors for the nonuniform voltage distribution inside the stack, we also demonstrate cathode activation potential and membrane ohmic overpotential, which are the major voltage losses, in Fig. 2 [23, 24].

Because of the same current output of each cell, we can deduce that the nonuniform distributions of the cathode activation potentials shall be influenced mainly by temperature profile.
according to the simplified Butler-Volmer equation [23]:

\[ \eta_c = \frac{RT}{\alpha_c F} \ln \left( \frac{i}{i_0} \right) \]  

(12)

As seen in Fig. 2, the highest cathode activation potentials belong to the cells #6 and #5, given as 0.1712V and 0.1708V, respectively. However, the lowest cathode activation potentials are positioned at the cells #10 and #9, given as 0.1630 V and 0.1646V, respectively. The difference between the highest and lowest cathode activation potentials is 0.0082V, which is mainly due to the temperature difference under the same current. Compared with the largest overall performance difference value of the cells, 0.0353V, the activation potentials difference is a small proportion.

On the other hand, it is shown in Fig. 2 that the highest and lowest membrane ohmic overpotential are located at the cells #7 and #2, whose value are 0.1486V and 0.1192V, respectively. Note that the largest difference among the membrane ohmic overpotential of the cells is 0.0295V, which occupies 83.5% of the output voltage difference, hence, it is the major influencing factor for the nonuniform voltage distribution inside the stack under low relative humidity. As discussed in [23,24], membrane ohmic overpotential can be expressed by:

\[ \Delta V = I \left( \frac{\delta_m}{\sigma_m A_m} \right) \]  

(13)

where \( I \) is the current, \( \delta_m \) the thickness of the membrane, \( A_m \) the active area of the membrane, and \( \sigma_m \) the ionic conductivity of the membrane, and defined as:

\[ \sigma_m = (0.005139 \lambda - 0.00326) \exp \left[ 1268 \left( \frac{1}{303} - \frac{1}{T} \right) \right] \]  

(14)

As seen in Eq. (13), membrane ohmic overpotential depends on the ionic conductivity under the constant current, membrane thickness, and the active area of the membrane. Furthermore, it is found that ionic conductivity also relies on temperature when the expression of water content \( \lambda \) is given as:

\[ \lambda = \begin{cases} 
0.043 + 17.18a - 39.85a^2 + 36a^3 & 0 < a \leq 1 \\ 
14 + 1.4(a - 1) & 1 < a \leq 3 
\end{cases} \]  

(15)

where \( a \) is the water activity at the interface of membrane and gas phases in thermodynamic equilibrium. Water activity in the gas phase is calculated by:

\[ a = \frac{C_w RT}{P_{sat}(T)} \]  

(16)

Hence, temperature is decisive for the ohmic overpotential according to the discussion above. We will study the distribution of temperature in the stack, and try to find the distributions of important variables affected by it.

Fig. 3 gives the temperature distributions in membranes and coolant channels. The same spatial distributions of temperature variation at different layers can be observed, where the highest temperature of membranes and coolant channels are located around cells #5 and #6. The temperature decreases from the inner cells (cells #5 and #6) to outer ones (cells #1 and #10). The heat due to electrochemical reaction and ohmic heat is generated at the cathode MEA surface, which leads to the temperature increase in the cell. In addition, the poorest heat ejection in the middle of the stack results in the highest temperature as shown in Fig 3. Since the nonuniform distributions of the temperature inside the stack is found, the corresponding distributions of important physical quantities (i.e., relative humidity at both sides and proton conductivity) influenced by temperature will be described below.

![Fig. 3. Temperature distributions in membranes and coolant channels.](image)

Fig. 4 presents the relative humidity distributions in catalyst layers at both sides and proton conductivity of the membranes in each cell, respectively. It is interesting to find that the distributions of the relative humidity at both sides are almost identical but opposite to the distributions of temperature shown in Fig. 3. Because of the same output current of each cell, water generated at each one should be roughly the same. Hence, RH depends on temperature under the same inlet pressure according to Eq.(16). By Eq. (14), we can deduce that the distribution of the ionic conductivity should be converse of that of temperature, the trends of which can also be observed in Fig.4.

Since the distribution of the temperature has a strong impacts on the distribution of the above important physical quantities and then influences the degree of the nonuniformity of the individual cell performances, we will investigate the influencing
factors for the distribution of temperature and improve it.

Fig. 5 shows the distribution of coolant mass flow rate at each cell. It is found that the distribution presents a parabolic shape, which is converse of that of the temperature given in Fig. 3. Besides, the different distribution trends of humidified hydrogen and air can be observed as well, which will be discussed later in our next work. Therefore, we obtain that the different mass flow rates of each cell could lead to an uneven distribution of temperature. Furthermore, different coolant mass flow rates will be presented and the corresponding distributions of individual cells performance will also be analyzed in the following.

To further explore the influence of coolant mass flow rate impacts on the nonuniform distribution of single cell voltage, we choose five cases to investigate. The operation conditions and the corresponding current outputs, and voltage variances are listed in Table II.

It is shown in Table II that the output current increases with the coolant mass flow rate. This can be explained by the better hydrated membrane when the heat generated in the stack is carried away timely by the higher coolant flow rate. As said in the above section, under the good heat transfer condition, we can keep the membrane hydrated due to the water content that depends on the temperature. Furthermore, hydrated membrane can maintain high ionic conductivity of the membrane, which means low ohmic overpotential. Hence, the activation potential will increase with the diminish of the low ohmic overpotential under the constant open circuit voltage and output voltage. In addition, the output current will increase with the activation potential according to Butler-Volmer equation [23, 24] or Eq. (12).

Table II. Results compare obtained from different cases

<table>
<thead>
<tr>
<th>case</th>
<th>mass flow rate(^1)</th>
<th>current(^2)</th>
<th>voltage variance(^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1</td>
<td>(1.0 \times 10^{-3})</td>
<td>1.048</td>
<td>1.553 \times 10^{-3}</td>
</tr>
<tr>
<td>#2</td>
<td>(5.0 \times 10^{-4})</td>
<td>1.003</td>
<td>9.113 \times 10^{-4}</td>
</tr>
<tr>
<td>benchmark(^4)</td>
<td>(1.0 \times 10^{-4})</td>
<td>0.6219</td>
<td>1.444 \times 10^{-4}</td>
</tr>
<tr>
<td>#3</td>
<td>(5.0 \times 10^{-5})</td>
<td>0.5391</td>
<td>3.044 \times 10^{-4}</td>
</tr>
<tr>
<td>#4</td>
<td>(1.0 \times 10^{-5})</td>
<td>0.3767</td>
<td>3.237 \times 10^{-3}</td>
</tr>
</tbody>
</table>

\(^1\) Total coolant mass flow rate (kg/s) at (2) in Fig. 1.
\(^2\) Current output (A/cm\(^2\)) under the constant stack output voltage, 6V.
\(^3\) Voltage variance under the same stack output voltage, 6V.
\(^4\) The results obtained by using the operating parameters listed in Table I.

However, the single cell voltage fluctuation in the stack is becoming more obvious as the coolant flow rate increases/decreases from the benchmark case, according to the voltage variance listed in Table II. This is due to the significant nonuniform distributions of the temperature when the coolant flow rate gets higher or lower (see Fig. 6, 9 and 10).

Fig. 6 shows the distributions of single cell output voltage under different cases, from which the uneven individual cell performances are observed intuitively. Under cases #1 and #2, the single cell voltage increase from cell #2 to cell #9, which is opposite to activation potential (see Fig. 7), and accordant with that of ohmic overpotential (see Fig. 8). The reason for this phenomena is that the various activation potential induces the fluctuating cell voltage under the good heat ejection conditions (cases #1 and #2). However, the distribution of the output voltage is converse of that of activation potential and ohmic under poor heat transfer conditions (cases #3 and #4). Due to the poor cooling effects at case #3 and #4, ohmic overpotential increment is contributed to the consistent distribution of the above three variables.

Fig. 9 and 10 present the temperature distributions at membranes and coolant channels, respectively. The trends of the two ones are identical. As seen in Fig. 9, the highest temperature reaches above 400K under case #4, which will cause not only
Fig. 6. Distributions of single cell output voltages under different coolant mass flow rates.

Fig. 7. Distributions of single cell activation potentials under different coolant mass flow rates.

Fig. 8. Distributions of single cell ohmic overpotentials under different coolant mass flow rates.

Fig. 9. Distributions of single cell membrane temperature under different coolant mass flow rates.

The nonuniform of the cell performance, even the damage of the stack. Of course, cases #3 and #4 are two extreme conditions, which will not happen in practical operation. But the strong effects of temperature and/or cooling system on the voltage distribution inside the stack must be taken into account.

Fig. 11 shows the coolant flows distributions of single cell under the above cases. It is found the coolant mass flow rates distributions of single cell change with the magnitude of the total inlet mass flow rate. Under the middle or small total inlet mass flow rates (the benchmark problem and cases #3 and #4), the distribution of the flow shows a parabolic shape; however, the flows increase along the main flow direction under the large total mass flows (cases #1 and #2). By comparing the distribution of temperature with that of voltage and coolant flow under cases #3 and #4, we can find that the former one is converse of the two latter ones. However, a high flow rate also leads to the major fluctuation as shown in Fig. 9 and Table II. Therefore, we should keep the coolant flow rate in a reasonable range according to the energy balance and the layout of the cells inside the stack.
Fig. 10. Distributions of single cell membrane temperature under different coolant mass flow rates.

Fig. 11. Distributions of single cell coolant flows under different coolant mass flow rates.

4 CONCLUSION

A fully coupled 3D, non-isothermal, electrochemical and transport PEMFC stack model in which the coolant effect is considered, is presented in the paper. Based on this model, the nonuniform distribution of the single cell voltage in the stack is studied. By analyzing the main voltage loss distribution, e.g., cathode activation potential and membrane ohmic overpotential, we find that the fluctuation of the ohmic overpotential has important impacts on the uneven distribution of the single cell voltage. A further exploration is carried out to find the original reason of fluctuation of the ohmic overpotential. It is found that the nonuniformity is due to the different heat transfer in each cell. In addition, five cases with different coolant mass flow rates are presented. By comparing the effects of these cases, we conclude that both higher or lower flow rates can lead to the major fluctuation of the voltage. The uneven distribution of activation potential is contributed to the reason of nonuniform distribution of single cell output voltage under a large coolant flow rate. However, when a poor coolant flow rate is carried out, the influence of ohmic overpotential is a major factor. Based on this analysis, we find that the distribution of the temperature inside the stack determines the distribution of the activation potential and ohmic overpotential, then the degree of the nonuniformity of the single cell output voltage.

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**NOMENCLATURE**

- \( A \): area (m²)
- \( C_W \): molar concentration of water (mol/m³)
- \( E \): energy (J)
- \( F \): Faraday’s constant (96487C/mol)
- \( I \): current (A)
- \( \bar{J} \): mass flux (kg/m²·s)
- \( k \): thermal conductivity (W/m·K)
- \( m \): membrane
- \( M \): molecular weight (kg/mol)
- \( P \): pressure (atm)
- \( R \): gas constant (8.314J/mol·K)
- \( RH \): relative humidity
- \( S \): source term
- \( T \): temperature (K)
- \( \bar{u} \): superficial velocity (m/s)
- \( V_{stack} \): stack output voltage (V)
- \( X \): molar fraction of species
- \( Y \): mass fraction of species

**Greek letters**

- \( \varepsilon \): porosity
- \( \rho \): density (kg/m³)
- \( \zeta \): stoichiometry flow rate
- \( \sigma \): ionic/electrical conductivity (S/m)
- \( \lambda \): water content
- \( \mu \): viscosity (kg/m·s)
- \( \tau \): viscous stress (N/m²)

**Subscripts**

- \( a \): anode
- \( c \): cathode
- \( eff \): effective value
- \( f \): fluid
- \( i \): species
- \( m \): membrane
- \( ref \): reference value
- \( s \): solid phase
- \( sat \): saturation