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Understanding heat and charge transfer in a low temperature PEM Single Cell: a simulation tool development

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Abstract - *This work is a study of coupled charges and heat transfers phenomena on a low temperature PEMFC single cell. A one-dimensional steady state model was developed to compute current and temperature distributions in a PEMFC single cell whatever the operating conditions. The work comprises of three parts: Firstly, a literature survey was conducted to describe the principle of a PEMFC and the fundamental operations. A state of the art of the previous modeling works on the PEMFC was also presented in this part. Secondly, the developed model using the basic relations to describe heat and charges transfer phenomena occurring in the single PEM cell was presented. Finally, a simple simulation tool called FCvb was performed and optimized using Visual Basic Excel and referring to the previous relations in the developed model. To finalize this study, a conclusion and perspectives were presented in the last part of this work.*

Résumé - *Ce travail étudie les phénomènes des transferts couplés de charge et de chaleur au coeur d'une cellule de pile à combustible à membrane échangeuse de proton. Un modèle monodimensionnelle et en régime permanent été utilisé pour évaluer le courant et la tension déliverés par la pile ainsi que de visualiser la distribution de la température pour une cellule quelques soit les conditions opératoires. Ce travail comprend trois parties dont la première concerne une description générale de ce type de pile avec les opérations fondamentales ayant lieu, un état de l'art concernat ce genre de modélisation à fait aussi l'objet de cette partie. La deuxième partie décrit le modèle utilisé pour chaque type de transfert envisagé dans ce travail. La description de l'outil qui a été développé sous le nom de "FCvb tool "est intégrée dans la dernière partie, cet outil est sous Visual Basic Excel et il fait appel à toutes les formulations mathématiques du modèle choisi. Pour finaliser cette étude, une conclusion suivie des perspectives pour cet outil développé ont été mis evidence dans la partie conclusion.*

Keywords: Polymer Electrolyte Membrane Fuel Cells - Heat transfer - Charge transfer - Modelling tool.

1. INTRODUCTION

A Polymer Electrolyte Membrane Fuel Cells (PEMFC) is an electrochemical energy conversion device that combines a fuel with an oxidant, and converts chemical energy directly into electrical energy. It is composed of an anode and a cathode electrode, and a polymer electrolyte membrane between the electrodes [1].

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As well as presented in figure 1, hydrogen and oxygen are separately fed to anode and cathode channels, they diffuse through the porous electrodes and reach the catalytic layer respectively. At the catalytic layer of the anode, hydrogen splits into protons and electrons, and protons permeate through the membrane to the cathode side. The electrons are transported to the cathode by an external electric circuit. At the catalytic layer of the cathode, oxygen combines with protons and electrons to produce water.

The electrochemical reactions for the PEM fuel cell are shown in equations (1, 2 and 3), [2].

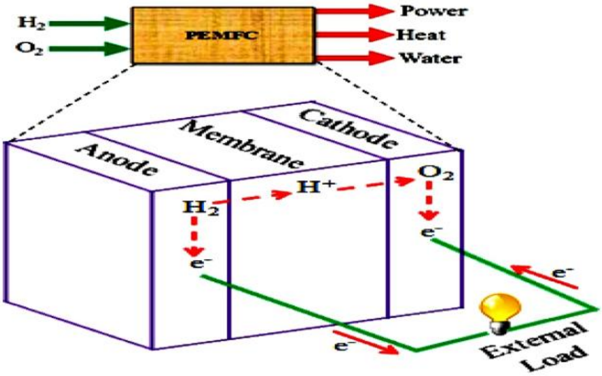

Fig. 1: PEMFC operations

Anode reaction-

$$H_2 \rightarrow 2H^+ + 2e^- \quad (1)$$

Cathode reaction-

$$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O \quad (2)$$

Overall reaction-

$$\frac{1}{2}O_2 + H_2 \rightarrow H_2O \quad (3)$$

They must be constantly fed with a fuel and oxidant. The fuel can be pure hydrogen or any hydrocarbon with high hydrogen content, and the oxidant is commonly air or pure oxygen. In this work, we are interested by understanding the charge and heat transfer phenomena in the heart of the PEMFC. A variety of models have been proposed in literatures, studying charge, heat and mass transfer phenomena using static or dynamic models, or even one, two or three dimensional approaches [1].

In the early1990's, Bernardi *et al.* [3] and Springer *et al.* [4] developed the first one-dimensional, steady state and isothermal models. These models were not complete but they provided the basis of fuel cells transport modeling by considering phenomena such as water transport in the polymer membrane and gas transport in the gas diffusion layers (GDLs). They also emphasized the importance of water management, the hydration of the membrane being one of the key issues for electrical efficiency [5].

Thirumalai *et al.* [6] completed a good sensitivity analysis on an earlier model [7] to find the most important operating conditions of the PEM fuel cell stack. They found that the following factors were particularly important: gas mass flows (pressure and stoichiometry of the inlet gases), operating temperature of the fuel cell stack, and relative humidity of the reactant gases, primarily of the anode gas.

Other models include that of Larminie *et al.* [8] which include the internal current and a simplified dynamic performance model of the fuel cell stack. Complex models use more detailed techniques and require detailed information on the materials and makeup of the fuel cell stack. Because of the complexity and interdependence of variables on the performance of the fuel cell stack an empirical equation can be used to predict the polarization curve. The advantage of this approach is that it is relatively simple to accurately predict a particular polarization curve. The disadvantage of this approach is that the polarization curve must be re-calculated for any change in operating conditions, such as humidity or temperature.

An empirical equation was presented in 1995 by Kim *et al.* [9] that gave an accurate prediction of the polarization curve without complete consideration for the electrochemistry. Another empirical model was used by Amphlett *et al.* [10].

When the analytical constants were not fully understood they went about finding an empirical equation for each parameter and for the entire irreversible losses. This technique issued as a supplement to analytical models and will be used in the proposed model. In our modeling tool we have envisaged to use the semi empirical model of Amphlett because it’s the most comprehensive one and which include more details on the voltage losses.

Numerous thermal model of the PEMFC have been published in the literature. Early efforts were made by Nguyen *et al.* [7] and Fuller *et al.* [11]. Nguyen *et al.* [7], developed a two-dimensional PEMFC model with one-dimensional heat transfer in the flow direction. This model considers phase change of the water in the flow channel as the only heat source, allowing convective heat transfer between the gas and solid phases.

On the other hand, Fuller *et al.* [11] a pseudo two-dimensional thermal PEMFC model with one-dimensional mass transfer in the through-membrane direction and one-dimensional heat transfer in the flow direction. Yi *et al.* [12] extended Nguyen *et al.* [11] model to include the entropic and irreversible reaction heats along with the phase change heat. However, this model allowed the temperature variation of the solid phase in the flow direction only, assuming uniform temperature in the through-membrane direction.

Wöhr *et al.* [13] developed a one-dimensional thermal model for heat and mass transfer in the through-plane direction, particularly for PEMFC stacks. Accounting for the entropic and irreversible reaction heats, they computed the temperature profile in the through-membrane direction and predicted the maximum temperature as a function of the number of cells contained in a stack. Rowe *et al.* [14] also developed a one-dimensional model in the through-membrane direction. Including entropic, irreversible and phase change heats, they further took account of Joule heating in the membrane and anode/cathode catalyst layers.

In our model, we have proposed to use a one dimensional steady state model, the main topic was to build and test a software tool with Visual Basic Excel to predict the PEMFC performances starting from operating conditions and with different technical data.

2. CHARGE TRANSFER PHENOMENA

In the PEM fuel cell, the chemical reaction is responsible for the transformation of chemical energy into electrical; nevertheless, this ideal conversion is accompanied by irreversible phenomena like those of activation, diffusion, electric and ionic conduction. To study the charge transfer in a PEM single cell, we propose to develop a simplified model that describes the real cell voltage starting from some empirical correlations found in literature.

2.1 Reversible cell voltage

The reversible cell voltage, or theoretical voltage, is a term used to define the cell voltage in open circuit (no current load), it is the maximum theoretical voltage of a stack produced at a given temperature with the partial pressure of the reactants and products known. Each element of the PEM fuel cell, (single cells), realizes the direct conversion of the hydrogen into electricity current, (electrical energy), in the theoretical case of a reversible reaction, [2].

The pressures of the products and reactants play a part in effecting the reversible voltage of the fuel cell. Basically increasing the pressure of the reactants increases the amount of reactants on the surface of the electro plates and so increases the frequency of chemical activity.

In this condition (open circuit), the cell voltage (E) is equal to E_{Nernst}. The effect of pressure on E_{Nernst} as shown in literature is presented in equation (4) and (5), [15].

$$E = E_T^0 + \frac{R \times T}{2F} \times \ln\left(\frac{P_{H_2} \times \sqrt{P_{O_2}}}{P_{H_2O}} \right) \times \sqrt{\frac{1}{P_0}} \tag{4}$$

While,

$$E_T^0 = 1.229 - 0.85 \times 10^{-3} \times (T - 298.15) \tag{5}$$

As a simplification, the partial pressures of H_2, O_2 and H_2O can be stated in atmospheres which reduce the term $(1/P_0)^{1/2}$ to 1, [16].

2.2 Real cell voltage

Anytime the PEM fuel cell is delivering energy to an external load, the voltage losses appear and the cell voltage become less than the reversible one. Three voltage losses can be found during this operation; Activation, Ohmic and Concentration losses [17].

2.2.1 Activation losses

The activations losses are related to driving the chemical reaction that occurs on the electrodes. A certain proportion of energy is needed to start the chemical reaction. This phenomenon produces a nonlinear voltage drop called activation polarization. These losses occur on both anode and cathode catalysts.

If only a single reaction is considered then the voltage loss due to activation polarization can be described in equation (6) by the Tafel empirical equation [16]:

$$\eta_{act} = A_{ac} \times \ln\left(\frac{i}{b}\right) \tag{6}$$

2.2.2 Ohmic losses

They are due to resistance to proton flow across the membrane, also included in this losses is the resistance originating from the non ideal electrode interconnections. An ideal membrane would freely conduct H^+ ions. Equation (7) shows the expression of the Ohmic losses expressed by Ohm's law [18]:

$$\eta_{ohm} = r_m \times i \tag{7}$$

2.2.3 Concentration losses

Also called mass transport losses, they relates to the reduction of the fuel's concentration in the gas channels. The fuel and oxidant are used at the surface of the electrodes. The incoming gas must then take the place of the used reactant.

The concentration of the fuel and oxidant is reduced at the various points in the fuel cell gas channels and is less than the concentration at the inlet value of the stack. To approximate this phenomenon, an empirical equation better describes the concentration losses given in equation (8) shows the expression of the concentration loss [2].

$$\eta_{conc} = m \times \exp^{(n \times i)} \tag{8}$$

where (m) and (n) are empirical coefficients respectively expressed in V and cm^2/mA, then the combined effects of all irreversibility's (losses) could be summarized in equation (9) which presents the real cell voltage according to each current density, pressure and temperature $V(P,T,i)$, [20].

$$V(P,T,i) = E - \eta_{act} - \eta_{ohm} - \eta_{conc} \tag{9}$$

A polarization curve is a characteristic graph of voltage versus current for a set of operating conditions. This curve is the most common output of the models and it is seen as the most important criteria. Modeling this graph is among the primary goal of the model proposed.

In FCvb tool, that we have developed, all the previous equations were put in the database of this tool. However, we have considered studying the performances of a PEM fuel cell with this tool [21]. The data input are shown in **Table 1**.

Table 1: Data input of the PEM fuel cell

Parameters	Units	Values
T	°C	70
A_{ac}	V	2.89×10^{-2}
b	mA/cm^2	0.035
r_m	$k\Omega/cm^2$	2.5×10^{-4}
m	V	1.6×10^{-5}
n	cm^2/mA	6×10^{-3}
P_{H2O} , P_{H2}, P_{O2}	bar	1

Many simplifications were taken in consideration as well as the water management and the temperature variations. However, ‘ i ’ should take the values where i [0, 1000] and a values of $E = 179\ \text{V}$ and $E_{80}^{0} = 1.182\ \text{V}$ were calculated by the tool, so we can observe that the value of E_{80}^{0} is smaller than E and this is due to the oxygen pressure (0, 21 bars) which has an impact on the logarithm part in the Nernst equation.

We try to vary the hydrogen and oxygen pressure, figure 2 presents the polarization curve under different pressures given by FCvb. We can observe that there is a relationship between the increase of the pressure and the cell potential, so each time we increase pressure it will be better for the cell performances.

Latha *et al.* [15] found experimentally that the performance of the fuel cell improves with the increase of pressure of the inlet gases. Figure 2 shows polarization curve shifts positively as the pressure increases from 1 to 4 atm.

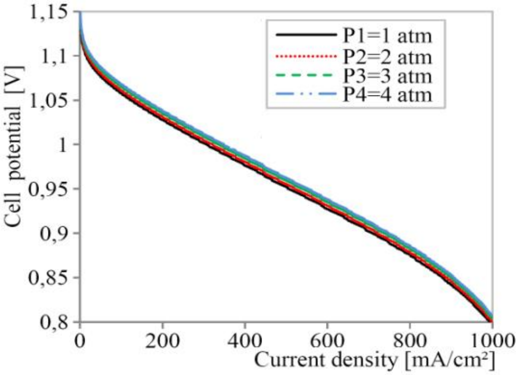

Fig. 2: Polarization curve under different pressure, T=80°C

We have also tried to vary the operating temperature of the PEM single cell, figure 3 gives the polarization curve under different temperature given by the graphical tool. We can observe that there is a remarkable difference between these different curves.

Indeed, the cell voltage increase each time the temperature decrease, so with high operating temperature we need to maintain the cell in better conditions by using a chiller device.

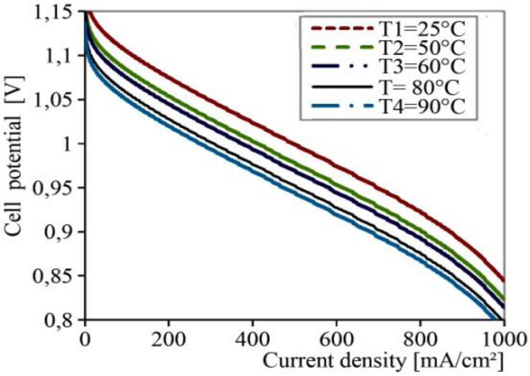

Fig. 3: Polarization curve under different temperature, $P_{H_2} = 1\text{atm}$; $P_{O_2} = 0.21\ \text{atm}$

Generally, In the case of Nafion membrane, water is easily condensed at lower temperature, water flooding may deteriorate gas diffusivity in the gas diffusion layer. The gas diffusivity of the fuel cell is improved with increased fuel cell temperature.

Therefore, fuel cell performance is improved at higher temperature. However, membrane conductivity decreases at higher temperature because of the reduction in the relative humidity of reactant gases and water content in the membrane. As the temperature increases, there will be a greater rate of water evaporation.

When the temperature reaches a critical temperature where the amount of evaporated water exceeds the amount of produced water, the membrane will start to dry out. The resistance will increase as the membrane dried out and thereby, decreases both the current and water production [16].

3. HEAT TRANSFER

The PEM fuel cell produces heat in an amount comparable to its electric power output so that the energy conversion efficiency is around 50 %. This heat generation stems from entropic heat of reactions, the irreversibility of the electrochemical reactions and Ohmic resistance or Joule heating. Roughly, the irreversible reaction heat, entropic heat and Joule heating account respectively for 55 %, 35 % and 10 % of total heat release [21].

3.1 Heat sources in the PEMFC single cell

Electrochemical reactions are exothermic, and water sorption or desorption, imply also heat production (or even consumption in some particular cases) at the electrode which is supposed to be homogenous. For the anode, and due to low relevant value, most of the authors in literature don't consider the heat production in this electrode.

The ionic resistivity of the membrane through which protons flow is at the origin of a volumetric heat source, in (W/m^3) distributed in its whole thickness [22], in this model we consider only this Joule heating as heat source given by equation (10), [23].

$$q_J = \frac{r_m}{\tau_m} \times I^2 \tag{10}$$

3.2 Heat transfer in the GDL/Membrane/GDL assembly

We have decided to study the GDL/Membrane/GDL zone in steady state. Both heat conduction and heat convection phenomena can be found in the PEMFC single cell, Chupin *et al.* [5], has demonstrated by using the Peclet number that the heat convection phenomena in the GDL/Membrane/GDL area is negligible comparing to the heat conduction's one. The heat conduction in equation (11) known as the Fourier's law is given as [24]:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} + \frac{\dot{q}}{k} = \frac{1}{\alpha} \times \frac{\partial T}{\partial t} \tag{11}$$

We consider only the heat transfer in the 'Ox' direction (perpendicular to the electrodes), the heat produced by the reaction, over potentials and the water desorption in the electrodes are not considered in this model.

No water phase change in the GDL, only the Joule heating is considered as the heat source in the middle of the membrane thickness ($x = 0$).

The electrodes are not considered to be an element of heat transfer due to their small thickness. The membrane is considered on a constant relative humidity. Figure 4 presents a schematic of the PEMFC single cell heat transfer model description.

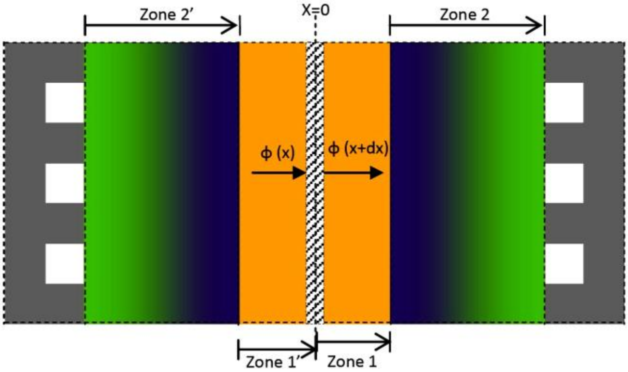

Fig. 4: The PEMFC single cell heat transfer model description

In the membrane (Zone 1) and (Zone 1'), the Joule heating is a uniform internal source, the membrane is considered isotropic and its thermal conductivity is constant and uniform, equation (11) become:

$$\frac{\partial^2 T}{\partial x^2} + \frac{q_j}{k} = 0 \tag{12}$$

In the GDL'S (Zone 2) and (Zone 2'), there is no heat source, equation (11) become:

$$\frac{\partial^2 T}{\partial x^2} = 0 \tag{13}$$

The FCvb tool, referring to the technical data in **Table 2** taken from [5, 24], gives the graphical results presented in figures 5, and 6. The parabolic temperature distribution in the membrane is representative of conductive heat transfer, but with a volumetric heat source.

The linear temperature distributions in the gas diffusion layer area are characteristic of purely conductive heat transfer, so the temperature increases linearly throughout the GDL till the membrane.

Table 2: The PEMFC single cell parameters values

Variables	Units	Values
eBP	m	10^{-3}
kBP	W/m.K	100
eGDL	µm	200
kGDL	W/m.K	1
em	µm	20
km	W/m.K	0.36
TBP =Twork	°C	70

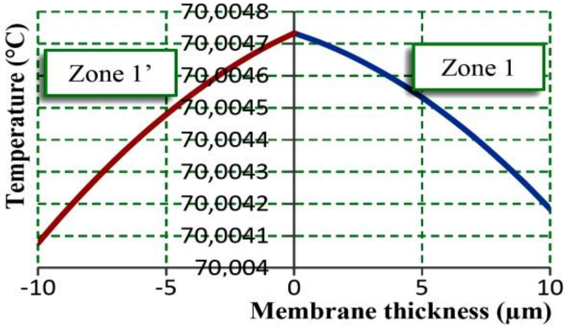

Fig. 5: Temperature distribution in the membrane

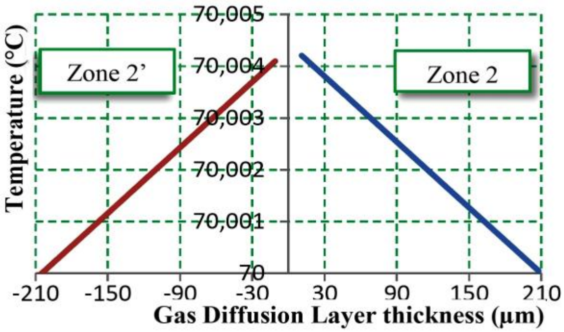

Fig. 6: Temperature distribution in the Gas Diffusion Layers

In order to test the effect of the current density on the temperature profile in the single cell, we propose to lunch another simulation using different values of the current densities, the results is presented in figure 7. The graph give temperature distributions in the anodic gas diffusion layer (-210 ≤ x ≤ -10 µm), in the membrane (-10 ≤ x ≤ +10 µm) and in the cathodic gas diffusion layer (+10 ≤ x ≤ +210µm).

As shown in figure 7, the temperature distribution in the given field depends strongly on current density, this is linked to the physical mechanisms at the origin of the main heat source and it's the Joule heating.

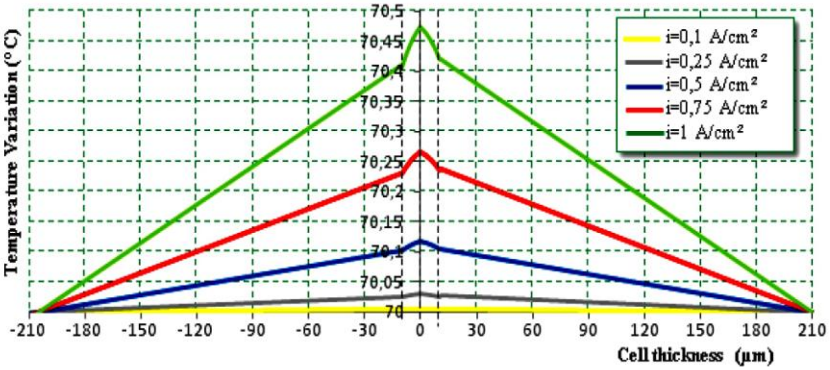

Fig. 7: Temperature distribution in the
Single cell under different current densities

4. CONCLUSION

Modeling studies of PEMFC have significantly contributed to understand, test and improve the design and performance of this energy generation technology system. The proposed modeling tool was developed to be used for PEM Fuel Cells charge and heat transfer modeling, it can provide several calculations and graphical results according to data input such as the Nernst Potential Cell and the various voltage losses and can also provide graphical of temperature distribution in the single cell area.

We can also make many tests on this tool including the pressure and temperature variation to predict the fuel cell characteristics.

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