Experimental validation of a PEM Fuel Cell Model

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Keywords: Proton Exchange Membrane Fuel Cell, model, Discretized GDL

ABSTRACT

This article proposes a pseudo 1-D, isothermal, lumped model of a Proton Exchange Membrane Fuel Cell (PEMFC) which is characterized by the presence of a discretized gas diffusion layer (GDL). Most of PEMFC models consider the pressure of reactant in the catalytic sites as the same as in the channels by neglecting the GDL. By the help of a discretized GDL, it is possible to consider this concentration more accurately at the catalytic sites by avoiding the partial derivative equations occurring by the application of the Fick's law. After the model's analytic description, this article deals with the validation of the model by comparing simulation and experimental. The results show good correlation both on polarization curve and on time domain data results, allowing to determine the state of the membrane more accurately.

1. INTRODUCTION

With the petroleum crisis of 1973 and 1979, governments became aware about the fact that our society was mainly dependent of a non-renewable resource. Adding to this the occurrence of major sanitary issues (Asian brown cloud, rise of asthmatic allergies etc.), an environmentally-friendly consciousness began to emerge as demonstrated by the creation of the WWF in 1969 or Greenpeace in 1979. In 1987, the Brundtland report introduce the concept of sustainable development defined such as the "development that meets the needs of the present without compromising the ability of future generations to meet their own needs" [1]. In this context, and due to a high power density combined with a solid electrolyte, low corrosion and relatively low operating temperature, the proton exchange membrane fuel cell (PEMFC) is a very promising technology for distributed generation and ground vehicle applications. PEMFC is an energy converter device which produces electricity, by oxydo-reduction reaction, by supplying hydrogen at the anode and oxygen (mostly ambient air) at the cathode. However, this technology is still restrained by several bottlenecks such as the price, the social acceptance of hydrogen and the durability. This article tackles the issue of the durability by proposing an experimental validation of a model able to predict the flooding at low current density, which is one of the most reversible faults that might occur in a PEMFC. An important quantity of models exist, developed according to the author's goals. For example, the development of a real time emulator in order to avoid issues related to the gaz supply and the ancillaries (Gao et al. [2], Marsala et al. [3]), it can be for control purposes (Pukrushpan et al. [4], Suh et al.[5], Talj et al. [6], Hernandez et al. [7], Bao et al. [8]), for the parameter identification or quantification of physical phenomenon (Springer et al [9], Nam et al. [10]) or to quantify the impact of ancillaries on the PEMFC system (Blunier et al [11], Mckay et al. [12]). For simple tasks, static models can be used and a good comparison of differents static models can be found in Saadi et al [13].

2. THE ANALYTICAL MODEL

In this paper, a model of a PEMFC has been developed in order to predict a voltage decrease caused by water flooding. As said before, many different models can be found in the literature, however only a few of them consider the flooding phenomenon and, among these models, only the model used by Mckay et al. [12] has been found which could be used for control problem. This model takes into consideration the diffusion of the species through the gas diffusion layer (GDL), where the model developed by Pukrushpan et al. considers the pressures in the channels as the same as the ones at the catalytic sites. Notice that the model developed by Mckay et al. [12] considers the species diffusion in the GDL. The partial differential equations are approximated by finite difference method, allowing the PDE to be expressed by simple differences. Some assumptions have been made to simplify the computations:

- All the gases are ideals;
- The stack temperature is isothermal;
- All the volumes are lumped;
- The load losses in the channels are neglected so the pressure are uniform inside;
- Hydrogen, nitrogen and oxygen do not pass through the membrane;
- The volume of liquid water will not restrict the gases volume;

2.1 Gas model in the channels

All gazes are considered as ideals and the mass of these gases can be described by the law of the mass conservation. As it is shown in the figure 1, the channels are fed with a mix of input gases (air or hydrogen) and water vapor, subsequently no liquid water enter the channels through the supply manifolds. However, at the output of the channels, liquid water can exist.

2.1.1 Gas model in the cathode channels

Considering the above assumptions, it can be possible to express:

$$\begin{aligned} \frac{dm_{O_2,ca,ch}}{dt} &= W_{O_2,ca,in} - W_{O_2,ca,out} + W_{O_2,ca}(K) & \frac{dm_{N_2,ca,ch}}{dt} &= W_{N_2,ca,in} - W_{N_2,ca,out} \\ \frac{dm_{O_2,ca,ch}}{dt} &= W_{O_2,ca,in} - W_{O_2,ca,out} + W_{O_2,ca}(K) & \frac{dm_{w,ca,ch}}{dt} &= W_{v,ca,in} - W_{w,ca,out} + W_{w,ca}(K) \end{aligned} \tag{1}$$

with $W_{x,n,p}$ in $kg.s^{-1}$ the mass flow of the species x in the channel n described at the location p (input, output or coming from the last discretized volume of the GDL), where x can be O_2 for oxygen, N_2 for nitrogen, w for water and v for vapor.

$$\begin{aligned} W_{O_2,ca,in} &= x_{O_2,ca,in}\, W_{da,ca,in} \\ W_{N_2,ca,in} &= \left(1 - x_{O_2,ca,in}\right) W_{da,ca,in} \\ W_{v,ca,in} &= \omega_{ca,in}\, W_{da,ca,in} \end{aligned} \tag{2}$$

With $\omega_{ca,in}$ is the ratio of vapor in the mixed gas and $x_{O_2,ca,in}$ is the ratio of oxygen in the input dry gas and can be expressed as follows:

$$\begin{aligned} \omega_{ca,in} &= \frac{M_v}{M_{dg}} \frac{\phi_{ca,in} p_{sat}}{p_{ca,in} - \phi_{ca,in} p_{sat}} \\ x_{O_2,ca,in} &= 0.21 \end{aligned} \tag{3}$$

With $\varphi_{ca,in}$ is between 0 and 1 and represents the relative humidity of the input gas at the cathode. p_{sat} is the saturation pressure, a monotonic function of the temperature which describes the maximum value of the vapor pressure before condensation and therefore liquid water occurrence as explained by Larminie et al. [14]. The function of p_{sat} is a polynomial interpolation of a thermodynamic table made by Sonntag et al [15].

$$p_{sat} = 6.859193 \times 10^{-4} T^4 - 0.74324595.T^3 + 304.1375.T^2 - 55613.63.T + 3831801 \tag{4}$$

with T is the stack temperature. From practical point of view, this temperature is refered as the one at the exit of the cooling system.

$$W_{ca,out} = k_{ca,out}\left(p_{ca,ch} - p_{ca,out}\right) \qquad W_{v,ca,out} = W_{ca,out} - W_{dg,ca,out}$$
$$W_{dg,ca,out} = \frac{1}{1+\omega_{ca,out}} W_{ca,out} \qquad W_{N_2,ca,out} = (1 - x_{O_2,ca,ch})\, W_{dg,ca,out} \tag{5}$$
$$W_{O_2,ca,out} = x_{O_2,ca,ch}\, W_{dg,ca,out} \qquad x_{O_2,ca,ch} = y_{O_2,ca,ch} \frac{M_{O_2}}{M_{dg,ca,ch}}$$

with $k_{ca,out}$ is the constant orifice at the output of the cathode and $p_{ca,ch}$ is the pressure in the cathode channels which can be expressed by:

$$p_{ca,ch} = p_{dg,ca,ch} + p_{v,ca,ch} \qquad p_{v,ca,ch} = min\left[p_{sat}, \frac{RTm_{w,ca,ch}}{M_v\, V_{ca}}\right]$$
$$p_{dg,ca,ch} = \frac{RT}{V_{ca}}\left(\frac{m_{O_2,ca,ch}}{M_{O_2}} + \frac{m_{N_2,ca,ch}}{M_{N_2}}\right) \qquad \omega_{ca,out} = \frac{M_{v,ca,ch}}{M_{dg,ca,ch}} \frac{\phi_{ca,ch}\, p_{sat}}{p_{ca,ch} - \phi_{ca,ch}\, p_{sat}} \tag{6}$$

with $M_{v,ca,ch}, M_{dg,ca,ch}, M_v, M_{O_2}, M_{N_2}$ are the molar mass of the vapor in the channel (which does not change with the standard one), the molar mass of the dry gas in the cathode channels (which is going to change because of the oxydo-reduction reaction that consume oxygen), the molar mass of the vapor, the molar mass of dioxygen and the molar mass of nitrogen, respectively. V_{ca} is the cathode volume and R is the universal gas constant.

$$M_{dg,ca,ch} = y_{O_2,ca,ch} M_{O_2} + (1 - y_{O_2,ca,ch}) M_{N_2}$$
$$y_{O_2,ca,ch} = \frac{p_{O_2,ca,ch}}{p_{ca,ch}} \tag{7}$$

Finally, it is possible to express the oxygen and water mass flows as:

$$W_{O_2,ca}(K) = N_{O_2}(K) M_{O_2} \varepsilon\, A_{fc}\, N_{cells}$$
$$W_{w,ca}(K) = (W_{l,ca}(K) + N_{v,ca}(K)\, M_v\, \varepsilon\, A_{fc})\, N_{cells} \tag{8}$$

With N_{cells} is the number of cell, ε is the porosity of the GDL material and A_{fc} is the nominal active area. Concerning $W_{l,ca}(K)$, it can be calculated using equation (20), another assumption is to consider the volume of liquid water negligible compared to the total channel volume. $N_{O_2}(K)$ is computed with the equation (19).

2.1.2 Gas model in the anode channels

Using the exact same pattern, it is possible to compute the hydrogen and water mass in the anode channels.

2.2 The diffusion of the species in the GDL

The diffusion of the species depends on the concentration gradient between two regions, the species will tend to move to a place where the concentration is lower. The molar flux can be expressed using the Fick's law:

$$N_j = -\langle D_j \rangle \frac{\partial c_j}{\partial y} \tag{9}$$

with $-\langle D_j \rangle$ is the effective diffusivity and c_j is the molar concentration of the specie j. Nam et al. [10] expressed the effective diffusivity as a function of the liquid water saturation s:

$$\langle D_j \rangle = D_j\, \varepsilon \left(\frac{\varepsilon - 0.11}{1 - 0.11}\right)^{0.785} (1 - s)^{\,2} \tag{10}$$

Where ε is the porosity of the GDL and D_j is the gas diffusion coefficient. The time derivative of the concentration is expressed as follows:

$$\frac{dc_j}{dt} = \frac{\partial N_j}{\partial y} + R_j \tag{11}$$

with R_j is the local reactant rate. This term is influent only on water which could change its phase. The liquid water transport is ruled by capillary effect and by applying the mass conservation law to a pore of a GDL. Then, one can express the liquid water saturation s, which describes the liquid water volume within a pore, as a function of the liquid water mass flow, W_l, and the molar evaporation rate, R_{evap}:

$$\frac{ds}{dt} = \left(\frac{1}{\rho_l \varepsilon A_{fc}}\right)\frac{\partial W_l}{\partial y} - \frac{R_{evap} M_v}{\rho_l} \tag{12}$$

with ρ_l is the liquid water density, ε is the GDL material porosity. The liquid water mass flow can be estimated using the capillary pressure gradients:

$$W_l = -\frac{\varepsilon A_{fc} \rho_l K K_{rl}}{\mu_l}\left(\frac{\partial p_c}{\partial S}\right)\left(\frac{\partial S}{\partial y}\right) \tag{13}$$

with K is the absolute permeability, K_{rl} is the liquid water relative permeability ($K_{rl} = S^3$), μ_l is the viscosity of liquid water, p_c is the capillary pressure and S is the reduced liquid water saturation which can be expressed as follow :

$$S = \begin{cases} \frac{s - s_{im}}{1 - s_{im}} \; pour & s_{im} < s \leq 1 \\ 0 \qquad pour & 0 < s \leq s_{im} \end{cases} \tag{14}$$

with s_{im} is the immobile saturation representing the point where the liquid flux becomes irregular occurring when $s < s_{im}$. The capillary pressure describes the pressure of a droplet of water over its surface, the Leverett J-function is used to quantify it as a function of the reduced liquid saturation:

$$p_c = \frac{\sigma \cos\theta_c}{\sqrt{\frac{K}{\varepsilon}}}\left[1.417S - 2.120S^2 + 1.263S^3\right] \tag{15}$$

with σ is the surface tension between air and water and θ_c is the contact angle of the droplet as described by Nam et al. [10]. The molar evaporation rate is described using the following expression:

$$R_{evap} = \gamma \frac{p_{sat} - p_v}{RT} \tag{16}$$

with γ is the volumetric condensation coefficient. As it can be seen, the problem confronted here to describe the diffusion of the gases along the GDL is full of partial derivative equations (PDE). These types of equations are clearly not convenient and Mckay et al. [12] had thought on considering the GDL as a three-discretized volume allowing the spatial derivative to be a simple difference between two volumes closed to each other. It is, then, possible to write:

$$\begin{aligned} \frac{\partial N_{j,e}}{\partial y}(k) &= \frac{N_{j,ca}(k) - N_{j,ca}(k-1)}{\delta y} \\ \frac{\partial c_{j,e}}{\partial y}(k) &= \frac{c_{j,ca}(k+1) - c_{j,ca}(k)}{\delta y} \\ \frac{\partial S_{j,e}}{\partial y}(k) &= \frac{S_{j,ca}(k+1) - S_{j,ca}(k)}{\delta y} \end{aligned} \tag{17}$$

with δy is the discretization width. This parameter is very important because it determines the importance of the discretization. Its value can be determined by:

$$\delta y = \frac{GDL\ thickness}{Discretization\ level} \tag{18}$$

Where the discretization level is an integer. From here, it is possible to express:

$$\begin{aligned} N_{j,e}(k) &= -\langle D_j(k)\rangle \frac{\partial c_{j,e}}{\partial y}(k) \\ \frac{dc_{j,e}}{dt} &= \frac{\partial N_{j,e}}{\partial y}(k) + R_{evap}(k) \end{aligned} \tag{19}$$

Equations (11) and (12) become:

$$\frac{ds}{dt}(k) = \left(\frac{1}{\rho_l \varepsilon A_{fc}}\right)\frac{W_{l,e}(k)-W_{l,e}(k-1)}{\delta y} - \frac{R_{evap}M_v}{\rho_l}$$
$$W_{l,e}(k) = -\frac{\varepsilon A_{fc}\rho_l K K_{rl}(k)}{\mu_l}\left(\frac{\partial p_c}{\partial S}(k)\right)\left(\frac{\partial S_e}{\partial y}(k)\right) \tag{20}$$
$$\frac{\partial p_c}{\partial S}(k) = \frac{\sigma cos\theta_c}{\sqrt{K/_\varepsilon}}\left[1.417 - 4.24\, S_e(k) + 3.789\, S_e(k)^2\right]$$

2.3 Membrane hydration

The molar fluxes at the membrane level are noted as the 0 section ($k = 0$) and can be described by the Faraday's law:

$$N_{H_2}(0) = \frac{I}{\varepsilon A_{fc}2F} \qquad N_{v,an}(0) = \frac{1}{\varepsilon}N_{v,mb}$$
$$N_{O_2}(0) = \frac{I}{\varepsilon A_{fc}4F} \qquad N_{v,ca}(0) = \frac{1}{\varepsilon}\left(N_{v,mb} + \frac{I}{A_{fc}2F}\right) \tag{21}$$

with F is the Faraday constant, I is the current and $N_{v,mb}$ is the vapor water flux considering the electro-osmotic drag and back diffusion phenomenon as describe by Springer et al. [9].

$$N_{v,mb} = n_d\frac{i}{F} - \alpha_w D_w\frac{c_{v,ca,mb}-c_{v,an,mb}}{t_{mb}} \tag{22}$$

with n_d is the electro-osmotic drag coefficient, i is the current density, α_w is a tunable parameter, D_w is the membrane water vapor diffusion coefficient and t_{mb} is the membrane thickness. The electro-osmotic drag coefficient, the membrane water vapor diffusion coefficient D_w and the water vapor concentrations are calculated by:

$$n_d = \frac{2.5\,\lambda_{mb}}{22}$$
$$D_w = 3.5\times 10^{-6}\left(\frac{\lambda_{mb}}{14}\right)\exp\left[\frac{-2436}{T}\right] \tag{23}$$
$$c_{v,e,mb} = \frac{\rho_{mb,dry}}{M_{mb,dry}}\lambda_e$$

with λ_{mb} is the membrane water content, $\rho_{mb,dry}$ is the dry membrane density, $M_{mb,dry}$ is the dry membrane molar mass and λ_e is the membrane water content next to the concerning electrode. The membrane water content is a linear interpolation of two polynomial functions representing the membrane water content at 30°C presented by Springer et al. [9] and at 80°C presented by Hinatsu et al. [16]. However, depending of the value of the water activity, the function can be assumed to be linear or constant.

$$\lambda_j = \begin{cases} \left(\lambda_j^{80^\circ C} - \lambda_j^{30^\circ C}\right)\left(\dfrac{T-303}{353-303}\right) + \lambda_j^{30^\circ C}\ for\ 0 \le a \le 1 \\ \left(\dfrac{16.8-\lambda_j^{a=1}}{2}\right)\left(a_j - 1\right) + \lambda_j^{a=1}\ for\ 1 < a \le 3 \\ 16.8\ for\ a > 3 \end{cases} \tag{24}$$

where the membrane water content is function of the water activity and given by:

$$\lambda_j^{30°C} = 0.043 + 17.81\,a_j - 39.85\,a_j^2 + 36\,a_j^3$$
$$\lambda_j^{80°C} = 0.3 + 10.8\,a_j - 16\,a_j^2 + 14.14\,a_j^3 \tag{25}$$
$$a_{mb} = \frac{a_{an}(1)+a_{ca}(1)}{2}\ \text{et}\ a_j(1) = \frac{p_{v,j}(1)}{p_{sat}}$$

with a is the water activity and j is the subscript defining the electrode next to the membrane surface or the membrane itself.

2.3 Voltage static model

As said before, this model takes the flooding phenomenon into account. This is achieved by using the apparent FC active area A_{app} which is the nominal FC active area reduced by the accumulation of the formation of a thin layer of liquid water:

$$A_{app} = A_{fc} - \frac{2m_{l,an,ch}}{N_{cells}\rho_l t_{wl}} \tag{26}$$

with $m_{l,an,ch}$ is the mass of liquid water accumulated in the anode channel and t_{wl} is the thickness of the liquid water accumulated in the anode channel and is considered as a tunable parameter. The scaling factor 2 is introduced to express that half of the active area is already blocked by the channel ribs. The equation to determine $m_{l,an,ch}$ is:

$$m_{l,an,ch} = max\left[0, m_{w,an,ch} - \frac{p_{sat}M_v V_{an}}{RT}\right] \tag{27}$$

From here, it is possible to define the apparent current density:

$$i_{app} = \frac{I}{A_{app}} \tag{28}$$

Then, the voltage can be expressed using:

$$v = E - U_{act} - U_{ohm} - U_{conc} \tag{29}$$

with E is the theoretical open circuit voltage, U_{act} represents the activation losses, U_{ohm} represents the ohmic losses and U_{conc} represents the concentration losses. The theoretical open circuit voltage is given by the Nernst equation (Barbir et al. [17]).

$$E = -\left(\frac{\Delta H}{2F} - \frac{T\Delta S}{2F}\right) + \frac{RT}{2F} ln\left(\frac{p_{H_2,an}(1)\sqrt{p_{O_2,ca}(1)}}{p_0^{1,5}}\right) \tag{30}$$

with ΔH is the difference in enthalpy from the standard conditions; ΔS is the difference in enthropy from the standard conditions, p_0 is the standard pressure, $p_{H_2,an}(1)$ is the hydrogen pressure at the anode side in the GDL discretized volume next to the membrane and $p_{O_2,ca}(1)$ is the oxygen pressure at the cathode side in the GDL discretized volume next to the membrane. The activation overvoltage represents the energy required to perform the electrochemical reaction and is described using the Butler-Volmer equation:

$$U_{act} = \frac{RT}{F} ln\left(\frac{i_{app}}{i_0}\right) \tag{31}$$

with i_0 is the exchange current density expressed as:

$$i_0 = K1\left(\frac{p_{O_2,ca}(1)}{p_0}\right)^{K2} exp\left[-\frac{E_c}{RT}\left(1 - \frac{T}{T_0}\right)\right] \tag{32}$$

Where $K1$ represents a tunable parameter related to the catalytic surface and to the exchange current density in standard conditions. $K2$ is a tunable parameter representing the pressure coefficient as described by Barbir et al. [17], E_c is the activation energy for the reduction of oxygen on platinum and T_0 is the standard temperature. Springer et al. [9] have shown that the ohmic overvoltage could be expressed as:

$$U_{ohmic} = \left[\frac{t_{mb}}{b_{11}\lambda_{mb} - b_{12}} e^{-1268\left(\frac{1}{303} - \frac{1}{T}\right)} + K3\right] i_{app} \tag{33}$$

Where $K3$ is a tunable parameter representing the resistance through the electrically conductive FC components, the values of b_{11} and b_{12} are constant and given by Springer et al. [9].

3. Model Validation

In order to validate the developed model, some polarization curves have been made in the FCLab laboratory (France) with different operating conditions. They show a good correlation between the experimental polarization curve and the simulated one. However, it has to be noted that inconsistencies exist concerning the experimental polarization curves and the experimental temporal data, constraining another adjustment of the coefficients $K1$, $K2$ and $K3$. The bench test used is fully equipped with barometers, flowmeters, thermometers, and hygrometers at the input and output of the FC. The FC used for the experimental part is a 20 cells with an active area of 100 cm^2 and a GDL thickness of 17 μm with a porosity of 0.84. Notice that the tests have been made with an open anode with recirculation and the humidity at both sides is regulated by boilers. The figures 1 and 2 show good correlation between experimentally obtained polarization curve and the simulated ones at different temperatures, relative humidity and stoichiometry computed as the ratio of the reactant oxygen/hydrogen mass flow over the oxygen/hydrogen inlet mass flow. Concerning the temperature, the test bench includes a cooling circuit to regulate the temperature, the stack temperature is considered to be the same as the one at the FC cooling system exit.

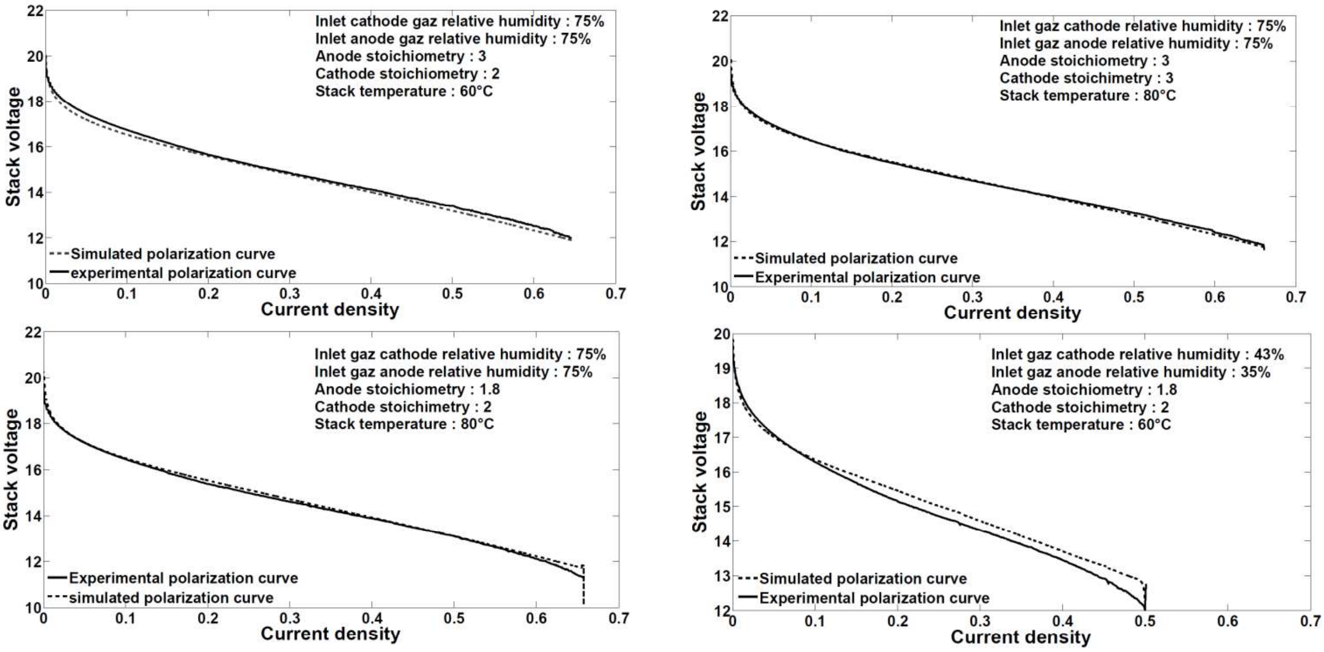

Figure 1: Experimental and simulated polarization curves with the stack voltage in volt and the current density in A.cm^2

It is possible to note an inconsistency at low relative humidity, which could be assumed as the occurrence of a drying process which is going to rise the membrane ionic resistance and will lower the catalyst surface as shown by Yousfi-Steiner et al. [18].

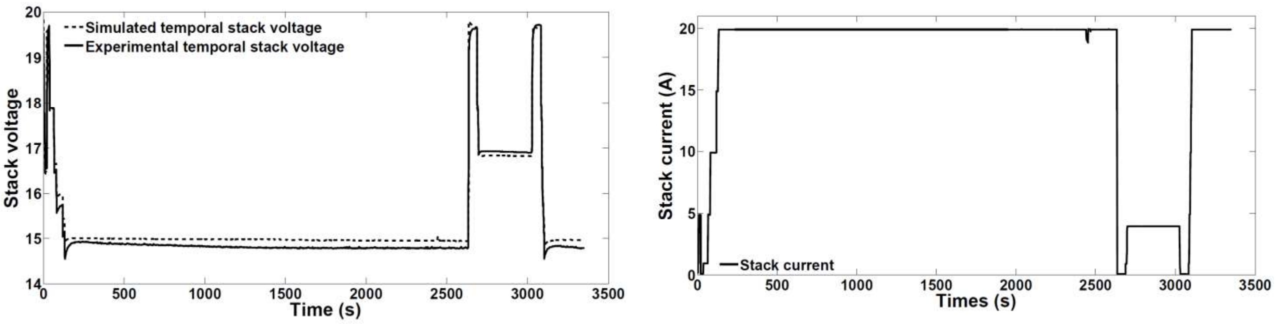

Figure 2: Experimental vs simulated voltage over the time with the stack current

The model gives satisfying results with temporal data. However, a transient response error appears, the model tends to have a time constant much shorter than the real one. It is supposed to be due to the approximation done on the GDL parameters. For example, considering the droplet contact angle constant or the porosity to be the one given by the manufacturer which is likely to degrade due to the ageing process. Nevertheless, this transient error does not look much critical as the time constant of flooding and drying is supposed to be of the order of one hundred seconds.

4. Conclusion

This article proposes a relevant model able to estimate the species concentration at the catalyst surface. Avoiding the spatial derivatives of the Fick's law by discretizing the GDL gives good results. However, it is important to consider a discretization level thin enough in order to keep the behavior as close as possible from reality, especially when speaking about its influence on water flux and having a computation time fast enough for control task. Another issue of such model is to have a good estimation of the different parameters, which has been clearly the biggest issue, despite disposing of a considerable amount of experimental data. To achieve this task, an optimization algorithm has been performed on polarization curve giving satisfying results. However, another constraint were to find coherent values for some settings in the literature close to physical ones in order to keep physical meaning.

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