Using electrical analogy to describe mass and charge transport in PEM fuel cell

P. Noiyinga, M. Hinaje, P. Thounthong, S. Raël, B. Davat

Abstract

This article deals with a PEMFC (proton exchange membrane fuel cell) electrical model based on a 1D analog representation of mass transport phenomena, and taking into account the influence of gas supply conditions on fuel cell voltage. By using an electrical analogy to describe transport of gaseous species in GDLs (gas diffusion layers), and water distribution in the membrane, the model can be directly implemented in standard simulation softwares used in electrical engineering (such as Saber, in our case), so that it can be easily employed for the simulation of fuel cell electrical systems. The paper explains how the analog model is obtained from mass transport equations, then it presents validation tests carried out in both steady-state and transient regimes on a PEM single cell bench.

1. Introduction

The use of fuel cells in electrical circuits needs an understanding of internal effects for an efficient management of the system, as well as an understanding of dynamic behavior in order to design and control the entire energy production unit with fuel cells, power electronic converters, and storage units. It is especially crucial to describe accurately the strong influence of operating conditions, such as temperature, gas flows, pressure and humidity, on fuel cell performances. For example, it is widely accepted that one of the key weak point of PEM fuel cell systems is their dynamic limitation. Indeed, the fuel cell system time constant is dominated by the compressor and the membrane hydration level, and may be several hundred of milliseconds. Therefore, fast load demands will cause significant voltage drop, particularly due to air starvation. This phenomenon is illustrated in Fig. 1, which presents the voltage experimental response of a 500 W PEM fuel cell system to a current step, obtained by means of a current controlled boost converter [1]. In this test, fuel flows are adapted to the fuel cell current, and the effect of the ancillaries time constant (namely the fact that fuel supply and delivered electrical current do not coincide in transient), can be clearly seen on the voltage curve.

Within the heart of a fuel cell three key processes that are strongly coupled take place [2]: mass transport in electrodes and electrolyte, electron conduction in electrodes and proton migration in electrolyte, and electrochemical kinetics at reactive sites. Many mathematical models have already been developed in order to describe locally these phenomena, through partial differential equations involving space and time. One of the earlier studies, by T. E. Springer et al. [3,4], led to an isothermal, one-dimensional, and steady-state model, relative to a PEMFC with a 117 Nafion membrane. This model focused on water transport in the membrane through diffusion and electro-osmotic drag, and introduced variable hydration as well as hydration dependent protonic conductivity. In another significant early work [5], D. M. Bernardi and M. W. Verbrugge developed a one-dimensional isothermal cell model to investigate factors that limit performance. Some other studies improved previous works, by including heat transfer description [6] or conservation equations [7] (i.e. transient modeling) in the general one-dimensional formulation. Many articles also focus on fuel cell two-dimensional mathematical modeling, the aim being often to investigate different aspects of water and heat management [8,9], of flow modes (coflow or counterflow) [10], or of gas distribution along channels [11].

According to results presented in the literature, mathematical fuel cell models are obviously accurate, and enable to study specific local effects. However, they are hardly used in a system approach, in which fuel cell models that are easy to handle are required. Moreover, they are most often only steady-state, which is clearly not sufficient to design a fuel cell based generation system comprising power electronic converters and storage units.

For the aforementioned reasons, many electrical circuit models have been developed. Among them, we first find models that have been obtained by extending a static current–voltage description to
transient regimes, often through adding a capacitor associated with the electric double layer phenomenon. This is seen for example in the case of works published by C. Wang et al. [12], and by S. Pasricha et al. [13]. In Ref. [14], such a circuit is used to establish a state model of the fuel cell, and then to achieve a control algorithm of the boost converter connected with the fuel cell. Some authors propose original solutions to describe fuel cell static or transient behaviors. In Ref. [15], the static characteristic is obtained by means of electronic components (bipolar transistors, diodes, resistances), so that the model has been implemented in a simulation software dedicated to electrical and electronic circuits (PSPICE). S. Lazarou et al. [16] use an ideal transformer and two saturable inductors to describe fuel cell dynamic responses to load changes. These kinds of models can give accurate results, but they are often empirical, and with the exception of fuel cell current, they can hardly describe the influence of operating conditions (temperature, gas flows, pressure, and humidity).

Secondly, many dynamic models of fuel cells (and of electrochemical devices in general) are based on Randles equivalent scheme associated with harmonic responses, obtained by the usual EIS technique under various loading conditions. The aim is to fit, usually by means of a numerical identification method (such as least square algorithm), a non-linear equivalent circuit with experimental impedance spectra. For example, A. M. Dhirde et al. have recently proposed such an equivalent circuit [17]. In this work, distributed elements, such as Warburg impedance and constant-phase element, are used to describe diffusion phenomena which take place within the fuel cell. In Ref. [18], authors employ fractional differentiation to approximate the Nernst diffusion transfer function in Laplace domain. I. Sadli et al. have shown that this diffusion impedance could be correctly represented by two RC cells [19]. Modeling fuel cells through AC impedance studies is most often physics-based, but fundamentally, theses studies generates only small signal models, which are theoretically not available in large signal conditions, because of non-linearities of electrochemical devices. In addition, include all operating conditions leads to a quite long and difficult parameter identification procedure.

Lastly, some works, such as [20–22], present fuel cell models that associate a standard description through electrical circuits, and macroscopic conservation equations of mass and energy. This enables prediction of transient evolution of gas flows and pressures, and to evaluate their influence on electrical behavior.

In this article, we focus on a PEM fuel cell electrical model implemented in simulation software commonly used in electrical engineering to design systems. This model is isothermal and includes transient description, double layer phenomenon, and 1D mathematical representation of mass transport phenomena in GDLs (gas diffusion layers) and membrane. For this purpose, we use an electrical analogy of gas and water diffusion equations, so that the model can be directly implemented in simulation software dedicated to electrical circuit. It can be therefore easily employed for the simulation of fuel cell electrical systems. It will be shown moreover how gas supply conditions and associated phenomena (influence of over-stoichiometric conditions on fuel cell voltage, transient air starvation) can be described.

In the first part, mass and charge transport equations, electrochemical kinetic equations, and boundary conditions of our fuel cell model are recalled. Following this, electrical analogy of mass transport equations in electrodes and membrane, and of associated boundary conditions, is detailed, as well as the model assembly.

Finally, an experimental validation carried out on a single cell system is presented.

2. One-dimensional PEM fuel cell mathematical model

Fig. 2 shows a schematic representation of the studied single PEM fuel cell. It is composed of a membrane sandwiched between two catalyst layers and two gas diffusion layers. Hydrogen (humidified or dry) is fed to the anodic compartment, and humidified air to the cathodic one, by means of gas distribution channels. In this section, we detail equations that describe the distribution of gaseous species in GDLs, the water content in the membrane, electrochemical overvoltages at membrane–electrode interfaces, and ohmic voltage drop across the membrane. The aim
is to calculate the fuel cell voltage in transient conditions. Assumptions and simplifications adopted in the present model are as follows:

1) the cell temperature remains constant and homogeneous throughout the cell
2) the ideal gas mixture is in a single phase
3) mass and charge transports are one-dimensional
4) the membrane is gas-tight, and homogeneous
5) all electrical contact resistances are neglected
6) catalyst layers are assumed like interfaces

As a result, only three computational regions will be considered: the anodic GDL, the membrane, and the cathodic GDL. Phenomena and associated laws that are taken into account in our one-dimensional transient fuel cell model are listed hereafter:

1) diffusion of species in GDLs (Knudsen diffusion law), and binary diffusion of gas mixtures in GDLs (Stefan–Maxwell diffusion law)
2) diffusion (due to concentration gradient) and electro-osmotic dragging (due to the membrane ionic current) of liquid water in the membrane
3) conductive transport (Ohm law) of electrons in electrodes, and of ions in the membrane

As mentioned before, electrochemical reactions that take place in catalyst layers are computed as boundary conditions at membrane/electrode interfaces.

### 2.1. Gas diffusion layers

The aim of mass transport calculation in GDLs is to determine partial pressures of hydrogen and oxygen at membrane–electrode interfaces, in order to evaluate electrode overvoltages. Secondly, partial pressures of water at membrane–electrode interfaces are needed to calculate the membrane water content at these boundaries. Thus, anodic and cathodic gas diffusion layers content the following unknowns: partial pressures of gases ($P_{H_2}$ and $P_{H_2O}$ at the anode, $P_O$, $P_{H_2}$, and $P_{H_2O}$ at the cathode), molar fluxes of gases ($J_{H_2}$ and $J_{H_2O}$ at the anode, $J_O$, $J_{H_2}$, and $J_{H_2O}$ at the cathode), and electrode potentials (denoted $\phi_s$).

#### 2.1.1. Gas transport

If both Knudsen and Stefan–Maxwell diffusions are taken into account, 1D gas mixture transport in GDLs is governed for each gas $i$ of the mixture by the following law [23,24]:

$$\frac{\partial P_i}{\partial x} = \frac{RT}{A_{cell}} \left( \frac{J_i}{D_{i,eff}} + \sum_{j \neq i} N_j J_j - P \cdot D_{j,eff} \right)$$

where $N$ is the number of species ($N_a = 2$ and $N_c = 3$), $J_i$, $P_i$, and $D_{i,eff}$ are the molar flow, the partial pressure and the effective Knudsen diffusion coefficient of the gas $i$, $D_{j,eff}$ is the effective Stefan–Maxwell diffusion coefficient of gases $i$ and $j$, and $P$ is the total pressure. The subscript $i$ is $H_2$ or $H_2O$ at the anode, $O_2$ or $N_2$ or $H_2O$ at the cathode. If $\varepsilon$ is the GDL porosity, the balance equation of gas $i$ can be written as:

$$\frac{\partial P_i}{\partial t} = \frac{RT}{\varepsilon \cdot A_{cell}} \frac{\partial J_i}{\partial x}$$

#### 2.1.2. Charge transport

Charge transport in GDLs is governed by Ohm’s law. Thus, the GDL electronic current $I_s$ and the electrical potential $\phi_s$ are linked by:

$$\frac{I_s}{A_{cell}} = -\sigma_{s,eff} \frac{\partial \phi_s}{\partial x}$$

where $\sigma_{s,eff}$ is the effective electronic conductivity of electrodes. In a 1D approach, the electronic current $I_s$ only depends on time, and is necessarily equal to the fuel cell current $I_{cell}$. As a result, voltage drop across GDL is simply described by a constant resistance.

#### 2.1.3. Boundary conditions for gas transport

Boundary conditions at membrane–electrode interfaces ($x = \delta_a$ and $x = \delta_a + \delta_m$) are linked to hydrogen and oxygen consumption, to water production, and to water molar flow continuity. This leads at the membrane–anode interface to:

$$\begin{cases} J_{H_2} = \frac{I_a}{2F} \\ J_{H_2O} = J_{H_2O_a} \end{cases}$$

where $J_{H_2O_a}$ is the membrane water molar flow, and $I_a$ the anodic Faraday current. It should be noticed here that this current is different from $I_{cell}$ in transient, because of the capacitive component due to the electric double layer that exists at membrane–anode interface. At the membrane–cathode interface, if $I_c$ is the cathodic Faraday current, boundary conditions are:

$$\begin{cases} J_{H_2} = \frac{I_c}{4F} \\ J_{H_2O} = J_{H_2O_m} + \frac{I_c}{2F} \end{cases}$$

In an actual experimental setup, both partial gas pressures (gas mixture composition) and gas molar flows are imposed at GDL inlets. For example, these boundary conditions can be expressed versus gas relative humidities (HR,), humidifier water temperatures ($T_{hum,in}$–$T_{hum,out}$), gas mixture pressures ($P_{in}$–$P_{out}$), and gas supply conditions (reference current $I_{ref}$, reactant stoichiometries $z_a$ and $z_c$). At the anode side, the saturation pressure being denoted $P_{sat}$ this leads to:


\[
\begin{align*}
P_{H_2O,in} &= HR_b - P_{sat}(T_{hum.a}) \\
P_{H_2,in} &= P_{a,in} - P_{H_2O,in}
\end{align*}
\]

and:

\[
\begin{align*}
J_{H_2,in} &= -c_a \frac{I_{ref}}{2F} \\
J_{H_2O,in} &= -\frac{P_{H_2O,in}}{P_{H_2,in}} J_{H_2,in}
\end{align*}
\]

At the cathode side \((x = \delta_a + \delta_m + \delta_c)\), boundary conditions are:

\[
\begin{align*}
P_{H_2O,in} &= HR_b - P_{sat}(T_{hum.c}) \\
P_{O_2,in} &= 0.21 \cdot (P_{c,in} - P_{H_2O,in}) \\
P_{N_2,in} &= 0.79 \cdot (P_{c,in} - P_{H_2O,in})
\end{align*}
\]

and:

\[
\begin{align*}
J_{O_2,in} &= -\frac{c_a}{4F} \\
J_{H_2O,in} &= \frac{P_{H_2O,in}}{P_{O_2,in}} J_{O_2,in} \\
J_{N_2,in} &= \frac{P_{N_2,in}}{P_{O_2,in}} J_{O_2,in}
\end{align*}
\]

Nevertheless, as there is no GDL outlet in 1D modeling, gas molar flows and gas partial pressures cannot be imposed simultaneously at GDL inlets. Most often, inlet partial pressures are imposed as boundary conditions. As a result, gases are necessarily supplied under strict stoichiometric conditions [7,25], what does not correspond to actual operating conditions. Indeed, reactants are usually supplied under over-stoichiometric conditions, what has a significant influence on fuel cell voltage. On the contrary, as shown in Fig. 1, air starvation phenomena may occur, especially during fast transient power demands [1]. In our 1D model, whether gas partial pressures, or gas molar flows, are used as boundary conditions at GDL inlets, depending on gas supply conditions. Thus, in the case of an over-stoichiometry gas supply, partial pressures are imposed at GDL inlets. Then, using Eqs. (6) and (8), boundary conditions at the anodic inlet \((x = 0)\) are as follows:

\[
\begin{align*}
P_{H_2O,(0)} &= P_{H_2O,in} \\
P_{H_2,(0)} &= P_{H_2,in}
\end{align*}
\]

and at the cathodic inlet \((x = \delta_a + \delta_m + \delta_c)\):

\[
\begin{align*}
P_{H_2O,(\delta_a + \delta_m + \delta_c)} &= P_{H_2O,in} \\
P_{O_2,(\delta_a + \delta_m + \delta_c)} &= P_{O_2,in} \\
P_{N_2,(\delta_a + \delta_m + \delta_c)} &= P_{N_2,in}
\end{align*}
\]

In the case of an under-stoichiometry gas supply, gas molar flows are imposed at GDL inlets, using Eqs. (7) and (9) as follows:

\[
\begin{align*}
J_{H_2,(0)} &= J_{H_2,in} \\
J_{H_2O,(0)} &= J_{H_2O,in}
\end{align*}
\]

and:

\[
\begin{align*}
J_{O_2,(\delta_a + \delta_m + \delta_c)} &= J_{O_2,in} \\
J_{H_2O,(\delta_a + \delta_m + \delta_c)} &= J_{H_2O,in} \\
J_{N_2,(\delta_a + \delta_m + \delta_c)} &= J_{N_2,in}
\end{align*}
\]

It will be explained in Section 3 how to compute such boundary conditions, when using electrical analogy.

### 2.1.4. Parametric laws

The saturation pressure of water vapor, which is needed to calculate water molar fractions at GDL inlets, depends on humidifier temperature. In our model, it is computed using the empirical expression of M. Wöhr [26], with \(T_{hum}\) in Kelvin, \(P_{sat}\) in Pa unit, \(A = 8.073\), \(B = 1656.39\) K, and \(C = 226.86\) K:

\[
P_{sat}(T_{hum}) = 100 \cdot 10^A e^{-A/T_{hum}}.
\]

For binary diffusion coefficients, we will use the empirical correlation given by M. F. Serican and S. Yesilyurt [27]:

\[
D_{ij}(T, P) = \frac{D_{ij}^{0}(T, P_0)}{P_0} \left(\frac{T}{T_0}\right)^{1.5},
\]

where \(T\) and \(P\) are the local fuel cell temperature and pressure.

Table 1 hereafter details values of \(D_{ij}^{0}(T, P_0)\) [27]. To account for GDL porosity \(\epsilon_g\), the effective diffusion coefficient is calculated with Bruggeman’s relation [28]:

\[
D_{ij,eff}(T, P) = D_{ij}(T, P) \cdot \epsilon_g^{1.5}.
\]

Last, it should be noticed that binary diffusion coefficients of gas \(i\) and gas \(j\) are linked by a reciprocal relation, called in this case Onsager’s reciprocal relation [29]:

\[
D_{ij,eff}(T, P) = D_{ji,eff}(T, P).
\]

### 2.2. Membrane

Fuel cell membrane behaves like an acid solution. It contains fixed sulfonic sites \(SO_3^-\) that dissociate water molecules in protons \(H^+\). As a result, both water and proton transports in the membrane strongly depends on the membrane hydration state. The aim of water transport calculation in the fuel cell membrane is to determine the membrane water concentration, in order to evaluate the voltage drop across the membrane. Secondly, water concentration and water molar flow at membrane–electrode interfaces are needed to calculate partial pressures of water at these boundaries.

Thus, the fuel cell membrane, in which water and ions are transported, has the following unknowns: water concentration, water molar flux and membrane potential, denoted \(c_{H_2O}, J_{H_2Oa}\) and \(\phi_m\), respectively.

However, the membrane hydration state is often represented, in particular in parametric laws, by the membrane water content \(\lambda\). This quantity is defined as the ratio between the number of water molecules and the number of sulfonic sites \(SO_3^-\) available in the membrane polymer. In practice, it approximately varies between 2 and 22. Water content can also be calculated versus membrane proton exchange capacity \(X_m\) (number of available sulfonic sites in the membrane per mass unit), membrane density \(\rho_m\) and water concentration as follows:

\[
\lambda = \frac{c_{H_2O}}{X_m \cdot \rho_m}.
\]

### 2.2.1. Water transport

The water transport in the membrane is a combination of two competing mechanisms [9,30]. One is due to the proton displacement from the anode to the cathode. As protons are solvated, they drag some water molecules with them. This phenomenon is called

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Binary diffusivities at (P_0 = 1) atm [27].</th>
</tr>
</thead>
<tbody>
<tr>
<td>(D_{ij}^{0})</td>
<td>Ref. temperature (T_{ref}) [K]</td>
</tr>
<tr>
<td>(D_{ij}^{0}_{H_2O,H_2O})</td>
<td>307.1</td>
</tr>
<tr>
<td>(D_{ij}^{0}_{H_2O,O_2})</td>
<td>308.1</td>
</tr>
<tr>
<td>(D_{ij}^{0}_{N_2,H_2O})</td>
<td>293.2</td>
</tr>
<tr>
<td>(D_{ij}^{0}_{N_2,N_2})</td>
<td>307.5</td>
</tr>
</tbody>
</table>
electro-osmotic drag. The other mechanism is water diffusion that
generally occurs from the cathode to the anode. This water flux
results from the water concentration gradient created in the
membrane by the electro-osmotic drag and the water produced by
the redox reaction at the cathode. Therefore, the total molar flow of
water transported inside the membrane is given by:

$$J_{H_2O_m} = \frac{n_d}{F} l_m - A_{cell} D_{H_2O_m} \frac{\partial C_{H_2O}}{\partial x}$$

(19)

where $l_m$ is the ionic current flowing through the membrane, $n_d$ is
the electro-osmotic drag coefficient (number of water molecules
dragged per proton), and $D_{H_2O_m}$ is the water diffusion coefficient.
The conservation of water quantity in the membrane can be written as:

$$\frac{\partial C_{H_2O}}{\partial t} = - \frac{1}{A_{cell}} \frac{\partial J_{H_2O_m}}{\partial x}.$$  

(20)

### 2.2.2. Charge transport

The membrane is assumed to be homogeneous. Therefore, as
sulfonic sites are fixed, there is no proton concentration gradient in
the membrane body, where local electroneutrality is verified. As
a consequence, proton transport in the membrane is only governed
by Ohm’s law. Thus, the ionic current $l_m$ and the electrical potential
$\phi_m$ are linked by:

$$l_m = - A_{cell} \frac{\partial \phi_m}{\partial x},$$

(21)

where $\sigma_m$ is the ionic conductivity of the membrane that greatly
depends on membrane hydration. In a 1D approach, the ionic current $l_m$
only depends on time, and is necessarily equal to the fuel cell current $I_{cell}$. As a result, voltage drop across the membrane is
described by a non-linear resistance:

$$\eta_m = \frac{l_{cell}}{A_{cell}} \int_{\delta_a}^{\delta_a + \delta_m} \frac{dx}{\sigma_m}.$$  

(22)

### 2.2.3. Boundary conditions for water transport

At membrane–electrode interfaces ($x = \delta_a$ and $x = \delta_a + \delta_m$), the continuity of water molar flow is considered. Thus, according to
Eqs. (4) and (5), we obtain:

$$\begin{cases}
J_{H_2O_m}(\delta_a) = J_{H_2O_1}(\delta_a) \\
J_{H_2O_m}(\delta_a + \delta_m) = J_{H_2O_1}(\delta_a + \delta_m) - \frac{I_c}{2F}
\end{cases},$$

(23)

$I_c$ being the cathodic Faraday current (different from the fuel cell
current $I_{cell}$ in transient, because of the capacitive component
due to the electric double layer at membrane–cathode interface).

Moreover, a phase change takes place for water (vapor phase in
GDLs, liquid phase in the membrane) at these interfaces. This
phenomenon is called sorption. Assuming thermodynamic equi-
librium between water vapor in the backing layers and liquid water
in polymer, sorption curves enable the calculation of water contents
versus water activity $a = \gamma_{H_2O}/\gamma_{H_2O_s}$ at membrane–electrode interfaces, as follows:

$$\begin{cases}
\lambda(\delta_a) = \lambda_{sorp}\left(\frac{P_{H_2O_1}(\delta_a)}{P_{sat}(I_{cell})}\right) \\
\lambda(\delta_a + \delta_m) = \lambda_{sorp}\left(\frac{P_{H_2O_1}(\delta_a + \delta_m)}{P_{sat}(I_{cell})}\right)
\end{cases}.$$  

(24)

### 2.2.4. Parametric laws

We will make use of parametric laws established by some
authors for Gore-Select membranes. Indeed, our fuel cell test bench
is equipped with this kind of membrane. Early studies of T. E.
Springer et al. [3] have proposed the following linear empirical law
for the electro-osmotic drag coefficient versus water content:

$$n_d = \frac{2.5}{22} \lambda.$$  

(25)

More recently, this coefficient has been measured for a Gore
membrane to the following constant value, over a wide range of
water activity (from 40% to 95%) [31]:

$$n_d = 1.07.$$  

(26)

In the same article, diffusion properties of Gore membrane are
studied, and authors propose for water diffusion coefficient versus
membrane water content and temperature the following empirical
relation [31]:

$$\begin{cases}
D_{H_2O_m} = f_D(\lambda) \cdot g_D(T) \cdot 10^{-10} & \quad (27) \\
f_D(\lambda) = \frac{2.563 - 0.33 \cdot \lambda + 0.0264 \cdot \lambda^2 - 0.000671 \cdot \lambda^3}{2} \\
g_D(T) = \exp\left(2416 \cdot \left(\frac{1}{303} - \frac{1}{T}\right)\right)
\end{cases}.$$  

To evaluate the membrane ionic conductivity, we will use the
following empirical law, established by Neubrand [32]:

$$\begin{cases}
\sigma_m = f_\sigma(\lambda) \cdot g_\sigma(T) \cdot 10^{-10} & \quad (28) \\
f_\sigma(\lambda) = 0.2658 \cdot \lambda + 0.0298 \cdot \lambda^2 + 0.0013 \cdot \lambda^3 \\
g_\sigma(T) = \exp\left((2640\exp(-0.6\lambda) + 1183) \cdot \left(\frac{1}{303} - \frac{1}{T}\right)\right)
\end{cases}.$$  

Last, the sorption phenomenon at membrane–electrode interfaces
will be described by empirical sorption curves of Hinatsu et al.
[33]. At 30 °C and 80 °C, these curves are defined as follows:

$$\begin{cases}
\lambda_{sorp}(30 °C) = 0.043 + 17.81 \cdot a - 39.85 \cdot a^2 + 36 \cdot a^3 \\
\lambda_{sorp}(80 °C) = 0.3 + 10.8a - 16a^2 + 14.1a^3
\end{cases}.$$  

(29)

To estimate interface water contents at an intermediate
temperature, a linear interpolation between these two experimental
formulas is made.

### 2.3. Voltage, overvoltages and currents

Fuel cell voltage is given versus electrode equilibrium potentials
$E_2$ and $E_c$, electrode overvoltages $\eta_a$ and $\eta_c$, membrane voltage drop $\eta_m$, and GDL electrical resistances $r_a$ and $r_c$ by:

$$V_{cell} = (E_c - \eta_c) - (E_a + \eta_a) - \eta_m - (r_a + r_c) \cdot I_{cell}.$$  

(30)

Electrode overvoltages are evaluated using simplified expressions
based on Butler–Volmer equation, in order to facilitate algo-
rithm convergence. The anodic overvoltage expression supposed that
both oxidation and reduction transfer coefficients are equal to
0.5, so that it can be written:

$$\eta_a = \frac{RT}{F} \ln \sinh\left(\frac{P_{H_2O,m}}{P_{H_2O}(\delta_a)} \frac{I_a}{A_{cell} \cdot \gamma_{H_2O}(\delta_a)}\right).$$  

(31)

where $I_{3a}$ is the anodic exchange current. The cathodic overvoltage
expression supposed that the oxidation component of $I_c$ is
negligible, compared to reduction component of $I_c$. However, the logarithm
argument theoretically obtained is slightly modified, so that $\eta_c$ is defined if $I_c = 0$. Finally, it gives:

$$\eta_c = \frac{RT}{F 2 \Delta c} \ln \left(1 + \frac{P_{O_2,\text{in}}}{P_{D_2, \delta_a + \delta_m / A_{cell}/3}} \right). \quad (32)$$

where $J_{Far}$ is the cathodic exchange current, and $\alpha_c$ is the reduction transfer coefficient.

At membrane—electrode interfaces, where redox reactions take place, fuel cell current $I_{cell}$ is split into a Faraday component verifying Butler—Volmer equation, and a capacitive component due to the electric double layers that exist at these interfaces. This results in:

$$\begin{cases}
I_a = I_{cell} - C_{\text{eff}} \frac{d(E_a + \eta_a)}{dt} \\
I_c = I_{cell} + C_{\text{eff}} \frac{d(E_c - \eta_c)}{dt}.
\end{cases} \quad (33)$$

3. PEMFC modeling using electrical analogy

3.1. Gas diffusion layers

Space discretization of Eqs. (1) and (2) leads to the following system, for the description of gas mixture diffusion in GDLs:

$$\begin{align*}
&\left(P_i(x + \Delta x) = P_i(x) - \left(R_i + \sum_{j=1, j \neq i}^{N} R_{ij}(x) \right) J_i(x) + \sum_{j=1, j \neq i}^{N} V_{ij}(x) \right), \\
&\left(J_i(x + \Delta x) = J_i(x) - C \frac{dp_i}{dt} \right).
\end{align*} \quad (34)$$

with:

$$\left\{ \begin{array}{l}
R_i = \frac{RT \cdot \Delta x}{A_{\text{cell}} \cdot D_{ij, \text{eff}}} \\
R_{ij}(x) = \frac{RT \cdot \Delta x}{A_{\text{cell}} \cdot D_{ij, \text{eff}}} \frac{P_i(x)}{P_j(x)} \\
V_{ij}(x) = \frac{RT \cdot \Delta x}{A_{\text{cell}} \cdot D_{ij, \text{eff}}} \frac{P_i(x)}{P_j(x)} J_j(x) \\
C = \frac{\varepsilon \cdot A_{\text{cell}} \cdot \Delta x \cdot RT}{\varepsilon}
\end{array} \right. \quad (35)$$

$\Delta x$ being the space discretization step. This non-linear system can be computed using an electrical analogy, for which pressures and molar flux are assimilated to voltages and currents, respectively. For illustration, Fig. 3 presents the equivalent electrical scheme of gas mixture diffusion in an element of fuel cell anode GDL. This equivalent scheme is composed of two electrical circuits (three in the case of the cathodic GDL), coupled to one another by partial pressures (resistances $R_{ij}$ in Eq. (35)) and molar flows (voltage sources $V_{ij}$ in Eq. (35)). To compute these couplings, as depicted in Fig. 3, each circuit of the equivalent electrical scheme (for example, circuit describing hydrogen transport in an element of anode GDL) receives from the other circuits their input partial pressures and molar flows (in our example: $P_{H_2,0}(x)$ and $J_{H_2,0}(x)$, respectively), what enables to calculate coupled components of the considered circuit (in our example: $R_{H_2,0,H_2}$ and $V_{H_2,0,H_2}$), according to Eq. (35). In return, it feeds the other circuits with its own input partial pressure and molar flow (in our example: $P_{H_2}(x)$ and $J_{H_2}(x)$), what enables to calculate coupled components of other circuits (in our example: $R_{H_2,0,H_2}$ and $V_{H_2,0,H_2}$), according to Eq. (35). Gas mixture diffusion in fuel cell GDLs is then described by associating in series such equivalent coupled circuits. In the analogical model presented here, a space discretization in 10 elements of both GDLs is used ($\Delta x_a = 0.1$ and $\Delta x_c = 0.1$).

3.2. Membrane

Space discretization of Eqs. (19) and (20) leads to the following system, for the description of water transport in the membrane:

$$\begin{align*}
&\lambda(x + \Delta x) = \lambda(x) - RW \cdot J_{H_2,0}(x) + V_W \\
&J_{H_2,0}(x + \Delta x) = J_{H_2,0}(x) - C_m \frac{d\lambda}{dt} \quad (36)
\end{align*}$$

with:

$$\left\{ \begin{array}{l}
RW = \frac{\Delta x}{A_{\text{cell}} \cdot D_{H_2,0} \cdot X_m \cdot \rho_m } \\
V_W = \frac{\eta L}{F} \cdot RW \cdot J_m \\
C_m = A_{\text{cell}} \cdot X_m \cdot \rho_m \cdot \Delta x
\end{array} \right. \quad (37)$$

FIGS. 4 AND 5 DEPICT INPUTS, OUTPUTS AND BOUNDARY CONDITIONS OF THE RESULTING ANODOIC AND CATHODIC MODELS. MODEL INPUTS, I.E. CURRENT REFERENCE $I_{eff}$, FARADAY CURRENT $I_f$ OR $I_c$, AND MEMBRANE WATER MOLAR FLOW $J_{H_2,0}(\delta_a)$ OR $J_{H_2,0}(\delta_a + \delta_m)$ AT MEMBRANE—ELECTRODE INTERFACES, ARE REQUIRED FOR SETTING BOUNDARY MOLAR FLOW CONDITIONS. MODEL OUTPUTS ENABLE FUEL CELL VOLTAGE CALCULATION (OVERVOLTAGE $\eta_a$ OR $\eta_c$) OR WATER SORPTION COMPUTATION (WATER PARTIAL PRESSURE AT MEMBRANE—ELECTRODE INTERFACES). BOUNDARY CONDITIONS (Eqs. (4) AND (5)) FOR MOLAR FLOWS AT MEMBRANE—ELECTRODE INTERFACES, Eqs. (6)—(9) FOR MOLAR FLOWS OR PARTIAL Pressures AT GDL INLETS) ARE DESCRIBED BY MEANS OF CURRENT SOURCES AND VOLTAGE SOURCES. IT SHOULD ALSO BE NOTICED HERE THAT FOR MORE CLARITY, COUPLINGS BETWEEN CIRCUITS WITHIN GAS TRANSPORT MODELS ARE NOT REPRESENTED.

AT GDL INLETS, THE USE OF DIODE FUNCTIONS ENABLE TO DISTINGUISH THE OVER-STOICHIOMETRY GAS SUPPLY CONDITIONS (FOR WHICH PARTIAL PressURES ARE IMPOSED), FROM THE UNDER-STOICHIOMETRY GAS SUPPLY CONDITIONS (FOR WHICH MOLAR FLOWS ARE IMPOSED). BY THIS WAY, AIR OR HYDROGEN STARVATION PHENOMENA, AS WELL AS THE INFLUENCE OF GAS FLOWS ON FUEL CELL PERFORMANCES, ARE INCLUDED IN THE MODEL, DESPITE THIS ONE IS 1D.
where $\Delta x$ is the space discretization step. Once again, this system can be computed using an electrical analogy, for which water content and molar flux are assimilated to voltages and currents, respectively. Fig. 6 presents the two equivalent electrical schemes of water transport in membrane that are implemented in our model. The first (Fig. 6a) is used for the left part of the membrane (anode side), and the second (Fig. 6b) for the right part of the membrane (cathode side). Water transport in the membrane is then described by associating in series these equivalent circuits. In the analogical model presented here, a space discretization in 10 elements of the membrane (5 elements for the left part of the membrane, and as many for the right part) is used ($D_x = d_m/10$).

Fig. 7 depicts inputs, outputs and boundary conditions of the resulting membrane model. Anodic and cathodic water partial pressures at membrane–electrode interfaces, i.e. $P_{H_2Oa}(\delta_a)$ and $P_{H_2Oc}(\delta_a + d_m)$ are required for setting boundary water contents. The ionic current $I_m$ flowing through the membrane is needed to calculate the electro-osmotic term in water transport (voltage term $V_w$ in Eq. (37)), and to evaluate the voltage drop $\eta_m$ across the membrane (Eq. (22)). The model outputs $J_{H_2Om}(\delta_a)$ and $J_{H_2Om}(\delta_a + d_m)$, which are water molar flows at membrane–electrode interfaces, are used as inputs of anodic and cathodic models (cf. Figs. 4 and 5) to set water flow conditions at these boundaries. The two other model outputs, i.e. the membrane resistance $R_m$ and the voltage drop $\eta_m$, simply enable the calculation of membrane electrical performances and fuel cell voltage. There are computed as follows:

$$
\begin{align*}
R_m &= \frac{\Delta x}{2 \cdot A_{cell} \cdot \sum_{k=1}^{N_m} \left( \frac{1}{\sigma_m(x_k)} + \frac{1}{\sigma_m(x_k + \Delta x)} \right)}, \\
\eta_m &= R_m \cdot I_m
\end{align*}
$$

$N_m$ being the number of membrane elements. Last, membrane boundary conditions on water contents (Eq. (24)) are described by means of voltage sources.

### 3.3. Fuel cell model

Fig. 8 presents the complete fuel cell model that has been implemented in Saber software. It associates:

1) analogical transport models previously described, which calculate gas flows and partial pressures in GDLs, water content and water flow in the membrane, electrochemical overvoltages at membrane–electrode interfaces (Eqs. (31) and (32) and voltage drop across the membrane (Eq. (38)),

2) a standard electrical scheme that compute the fuel cell output voltage as a function of electrode equilibrium potentials, electrode overvoltages, membrane voltage drop and GDL electrical resistances (Eq. (30)), and the fuel cell output current as a function of Faraday currents and double layer capacitors (Eq. (33)).

As depicted in Fig. 8, these two parts of the model are coupled, from transport models toward electrical model through electrochemical overvoltages and membrane voltage drop, and from electrical model toward transport models through electrochemical currents.

### 4. Simulations and experimental validation

#### 4.1. Fuel cell experimental setup

Experiments were carried out on a single proton exchange membrane fuel cell, based on a Gore Primea MEA. Membrane thickness is about 15 $\mu$m, and GDL thickness is equal to 400 $\mu$m. The cell active area is 100 cm$^2$. It is supplied with pure dry hydrogen from cylinders. Two pressure regulators decrease hydrogen pressure from 200 bars to 3 bars, and a third pressure regulator enables
to adjust hydrogen pressure at anode outlet. At anode inlet, hydrogen flow is set by a flux regulator.

The fuel cell cathode is supplied with air through a compressor and a pressure regulator that decreases air pressure down to 3 bars. As for anode, another pressure regulator enables to adjust air pressure at cathode outlet, and air flow is set at cathode inlet by means of a flux regulator. Moreover, air humidity is controlled by bubbling the gas through a water bath, which is maintained at a set-point temperature. Inlet air temperature is measured, in order to calculate its water molar fraction (according to Eq. (14), inlet air being supposed water saturated), and then its relative humidity (at stack temperature).

Fig. 5. Electrical analogy of gas mixture diffusion in fuel cell cathode.

Fig. 6. Equivalent circuits of water transport in a membrane element.
Fig. 7. Electrical analogy of water transport in fuel cell membrane.

Fig. 8. Electrical model of a PEM fuel cell.
The fuel cell rated current is 50 A. A cooling system keeps the stack temperature constant. An electronic load enables to impose a constant current to the fuel cell, up to 100 A. A second one, made in our laboratory by means of power bipolar transistors working in linear regime (rated current: 40 A, cut-off frequency: 10 kHz), enables to impose fast varying currents (such as current steps), and to perform impedance spectroscopy. Last, a “real time” electronic card (dSPACE DS1104 card) is used in combination with Matlab-Simulink mathematical environment for all necessary control functions such as reference setting (inlet gas flows, outlet pressures, and load current). Inlet gas flows are calculated versus stoichiometric coefficients \( \zeta_a \) and \( \zeta_c \) and reference current \( I_{\text{ref}} \) that can be set by an external signal (in particular when using an adapted flow supply), as follows:

\[
\begin{align*}
J_{\text{H}2} &= \frac{I_{\text{ref}} R T_0 \zeta_a}{2 F} \cdot 1000 \cdot 60 \\
J_{\text{air}} &= \frac{I_{\text{ref}} R T_0}{4 F} \cdot \frac{1}{0.21} \cdot \zeta_c \cdot 1000 \cdot 60 \\
&= \left[ \text{L}. \text{min}^{-1} \right], \quad (39)
\end{align*}
\]

with \( T_0 = 273 \, \text{K} \) and \( P_0 = 1.013 \cdot 10^5 \cdot \text{Pa} \). Other operating conditions are detailed in Table 2.

### 4.2. Model parameters

Geometric and physical parameters required for model simulation are recalled or given in Table 3. It should be noticed here that Knudsen diffusion phenomenon will not be taken into account. GDL ohmic resistances \( r_a \) and \( r_c \) (that can easily be calculated to 40 \( \mu \Omega \) have also been neglected. Anodic and cathodic double layer capacitors are assumed to be equal to twice the value obtained, by means of usual EIS technique, for the global double layer capacitor of the fuel cell.

### 4.3. Simulations and experimental validation in steady-state

Steady-state characteristics plotted hereafter have been obtained with constant gas flows, set by \( I_{\text{ref}} = 40 \, \text{A} \). The first test (cf. Fig. 9a) is associated with a stoichiometry \( 1-2 \) (\( \zeta_a = 1 \) at the anode side, and \( \zeta_c = 2 \) at the cathode side). It can be observed that there is quite good agreement between simulation results and experimental results. Thus, the model correctly describes the actual static behavior of the fuel cell when air is supplied with a double stoichiometry. However, there is an increasing difference between experiments and simulation, when the fuel cell current is near the limiting current imposed by hydrogen supply (i.e. 40 A in this case).

At \( \zeta_c = 1 \), model and experiment give quite different results, as shown in Fig. 9b. Indeed, if simulation obviously fits experiments for currents less than 20 A, it can be observed for higher currents that the measured fuel cell voltage drops more sharply than the simulated one. It seems that the fuel cell suffers from air starvation. This phenomenon should normally occur for a fuel cell current approximately equal to \( I_{\text{ref}} \). This is clearly not the case, and further investigation on the experimental setup has demonstrated that gas flow imposed at cathode inlet is significantly different than gas flow actually entering cathodic gas diffusion layer. Indeed, for the cell technology used here, gas flows in channels, parallel to the GDL, from the inlet to the outlet. As a result, a part of inlet gas (approximately 37% at the cathode) flows in channels to the outlet, without entering the GDL. This part of inlet gas cannot react, it is simply wasted. A small but actual discrepancy (approximately 5%) is also observed at the anode, which results in the increasing difference between experiment and simulation near the anodic limiting current (cf. Fig. 9a).

Fig. 10 presents membrane resistance obtained in steady-state with constant gas flows, set by \( I_{\text{ref}} = 40 \, \text{A} \), \( \zeta_a = 2 \) and \( \zeta_c = 4 \). The

### Table 2

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value or Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell temperature</td>
<td>( T )</td>
<td>333 , K</td>
</tr>
<tr>
<td>Humidifier temperature</td>
<td>( T_{\text{hum.c}} )</td>
<td>333 , K</td>
</tr>
<tr>
<td>Air inlet temperature</td>
<td>( T_{\text{air,in}} )</td>
<td>320 , K</td>
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<td>Anode relative humidity</td>
<td>( \frac{H_R}{X_{\text{ref}}} )</td>
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</tr>
<tr>
<td>Cathode relative humidity</td>
<td>( \frac{H_R}{X_{\text{ref}}} )</td>
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</tr>
<tr>
<td>Anode outlet pressure</td>
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</tr>
<tr>
<td>Cathode outlet pressure</td>
<td>( P_{\text{c,in}} )</td>
<td>1 , atm</td>
</tr>
</tbody>
</table>

### Table 3

<table>
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<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value or Reference</th>
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<tr>
<td>GDL thicknesses</td>
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<tr>
<td>Membrane thickness</td>
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<td>Cell active area</td>
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<td>GDL porosity</td>
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<td>S.-M. diffusion coefficients</td>
<td>( D_{\text{eff}} )</td>
<td>(16)–(18) , (27–29)</td>
</tr>
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<td>Electrode conductivity</td>
<td>( \sigma_s )</td>
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<tr>
<td>GDL resistances</td>
<td>( r_{\mu, f_c} )</td>
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<td>Dry membrane density</td>
<td>( \rho_m )</td>
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</tr>
<tr>
<td>Membrane proton exchange capacity</td>
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<td>Electro-osmotic drag coefficient</td>
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<td>(27) , (31)</td>
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<tr>
<td>Water diffusion coefficient</td>
<td>( D_{\text{H}<em>{2}O</em>{m}} )</td>
<td>(28) , (31)</td>
</tr>
<tr>
<td>Membrane ionic conductivity</td>
<td>( \sigma_m )</td>
<td>(29) , (32)</td>
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<tr>
<td>Anode exchange current density</td>
<td>( J_{\text{a}} )</td>
<td>5000 , A \cdot m^{-2}</td>
</tr>
<tr>
<td>Cathode exchange current density</td>
<td>( J_{\text{c}} )</td>
<td>0.5 , A \cdot m^{-2}</td>
</tr>
<tr>
<td>Cathode reduction transfer coefficient</td>
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<td>Double layer capacitors</td>
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<tr>
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<td>(15) , (26)</td>
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<tr>
<td>Water sorption curves</td>
<td>( \lambda_{\text{hump}} )</td>
<td>(30) , (33)</td>
</tr>
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</table>

Fig. 9. Fuel cell static characteristics. (a): \( I_{\text{ref}} = 40 \, \text{A}, \zeta_a = 1 \) and \( \zeta_c = 2 \). (b): \( I_{\text{ref}} = 40 \, \text{A}, \zeta_a = 1 \) and \( \zeta_c = 1 \).
measurement has been achieved by means of current step technique (measure of the immediate voltage drop caused by a current step added to a given steady-state DC regime). The experimental curve is compared with two simulated curves: one is calculated with gas flow conditions imposed at electrode inlets ($I_{\text{ref}} = 40\, \text{A}$, $z_a = 2$, $z_c = 4$), the other with corrected gas flow conditions ($I_{\text{ref}} = 40\, \text{A}$, $z_a = 2 \times 0.95$, $z_c = 4 \times 0.63$). Except for low current levels, there is quite good agreement between simulation and experiment, as far as air waste (due to direct air flow from cathode inlet to cathode outlet) is taken into account.

Last, we point out that, as for an actual experimental setup, the model describes fuel cell voltage dependence versus air flow conditions. Fig. 11 presents static characteristics simulated with $z_c = 1$ and $z_c = 4$. It can be observed that the voltage obtained with $z_c = 4$ is greater than the one corresponding to $z_c = 1$. This is first due to a higher oxygen partial pressure at membrane–cathode interface, implied by a greater air stoichiometry. The effect on output voltage increases with fuel cell current. In the example treated in Fig. 11, it represents 60% of the voltage gain at $I_{\text{cell}} = 20\, \text{A}$, and 92% at $I_{\text{cell}} = 35\, \text{A}$. Second, increasing air stoichiometry also implies a higher water partial pressure at membrane–cathode interface, and therefore a better membrane hydration in open circuit condition. However, this increase of membrane water content is partially or totally balanced (depending on the relative humidity of inlet air) by electro-osmotic drag phenomenon. As a result, its effect on output voltage decreases with fuel cell current. In the example treated in Fig. 11, it represents 40% of the voltage gain at $I_{\text{cell}} = 20\, \text{A}$, and only 8% at $I_{\text{cell}} = 35\, \text{A}$. These simulation results have still to be validated by experiment.

4.4. Simulations and experimental validation in transient

Fig. 12a presents experimental and simulated voltage responses to a current step at constant gas flows, set by $I_{\text{ref}} = 40\, \text{A}$, $z_a = 1$ and $z_c = 2$. The initial current is 0 A, and the step amplitude is 30 A. As stated previously, it can be observed that steady-state is correctly described by the model. However, one can also notice a significant difference between experiment and simulation in short times (the first 500 ms following the current step). Indeed, simulated voltage quickly drops to 0.607 V (obtained at $t = 52\, \text{ms}$) before increasing to the new steady-state (i.e. 0.662 V), whereas measured voltage slowly decreases to 0.657 V (obtained at $t = 510\, \text{ms}$) before increasing to nearly the same steady-state. As can be seen in Fig. 12b, which presents the simulated response of the membrane resistance to the considered current step, this difference is mainly due to the description of water diffusion in the membrane, the dynamic of which is clearly too high. The so-called fuel or air starvation phenomenon occurs during fast power demands (such as current steps), when gas flows are set by current level. It is mainly due to the slow time constant of the air flux regulator. This time constant is about 1 s in the case of our test bench. Fig. 13 shows experimental and simulated voltage responses to a current step, from 10 A to 30 A. Gas flows are set by the actual fuel cell current $I_{\text{cell}}$ ($t$), stoichiometries being $z_a = 2$ and $z_c = 4$ (as recommended by the cell manufacturer). When comparing step voltage responses given in Fig. 12 (obtained with constant gas flows) to Fig. 13, it can be concluded that our model includes the description of air starvation phenomenon. Nevertheless, as shown previously by the step response at constant gas flows, it is obvious that the accuracy has to be improved, notably by an actual parameter identification procedure that is still under study.
In this article, we detail a PEM fuel cell electrical model that includes transient description, double layer phenomenon, and 1D mathematical representation of mass transport phenomena in GDLs (gas diffusion layers) and membrane. For this purpose, electrical analogies of gas and water diffusion equations are used, so that the model is directly implemented in simulation software dedicated to electrical circuits and electrical systems design. Simulations have shown that gas supply conditions are taken into account although the model is one-dimensional. As a result, electrical behaviors associated with air stoichiometry, or with transient air starvation, are included in the model.

However, validation tests indicate that the model parameters have to be correctly defined, in order to increase accuracy in transient. We are presently working on defining specific tests for parameter identification. And even in steady-state, it seems that we have to investigate fuel cell behavior near limiting current conditions, in particular to predict more accurately cell voltage under transient air starvation condition.

References


Nomenclature

Latin letters

- a: water activity [-]
- A: active surface [m²]
- c: concentration [mol m⁻³]
- C: capacitor [F]
- D: diffusion coefficient of species [m² s⁻¹]
- Eeq: equilibrium potential [V]
- F: Faraday’s constant, 96472 [C mol⁻¹]
- J: exchange current density [A m⁻²]
- Jf: molar flow [mol s⁻¹]
- I: current [A]
n: charge number [-]
N: number of species in a gas mixture [-]
$n_e$: electro-osmotic drag coefficient [-]
P: pressure [Pa]
R: gas constant, 8.314 [J mol$^{-1}$ K$^{-1}$]
RH: relative humidity [-]
t: time [s]
T: temperature [K]
V: voltage [V]
x: 1D coordinate [m]
X: proton exchange capacity [mol kg$^{-1}$]

Greek letters
α: transfer coefficient [-]
δ: layer thickness [m]
ε: porosity [-]
η: surface overvoltage [V]
ζ: membrane water content [-]
ζ: stoichiometry [-]

ρ: density [kg m$^{-3}$]
σ: electrical conductivity [S m$^{-1}$]
ψ: electrical potential [V]

Superscripts and subscripts
a: anode
c: cathode
cell: fuel cell
dl: double layer
eff: effective value
GDL: gas diffusion layer
hum: humidifier
in: inlet
i, j: species (H$_2$, O$_2$, H$_2$O, N$_2$)
m: membrane
ref: reference condition
s: electronic phase
sat: saturated value
sorp: sorption