Effect of Water Content in the Gas Diffusion Layer of H$_2$/O$_2$ PEM Fuel Cell

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Abstract: The heart of a fuel cell is the membrane-electrode assembly consisting of two porous electrodes, where the electrochemical reactions take place, and the ionomer conductive membrane, which allows the proton exchange from the anode to the cathode. The porosity of the electrodes plays an important role in the fuel cell performance. One of the drawbacks presented by the porous electrodes is the accumulation of water in their structure, which implies a hindrance for the reactive gas transport to reach the catalytic reactive sites. In this paper, a mathematical model of a porous electrode, assuming single pores with uniform distribution, is introduced to determine the influence of water accumulation in the electrode on the fuel cell performance under different operating conditions. It is demonstrated that at low current densities, water accumulation has no effect in the fuel cell behavior, whereas at high current densities its performance is severely affected.

Key words: Fuel cells, porous-electrodes, electrode flooding.

Nomenclature

A	Electrode area (cm^2)
C	Concentration (mol·cm^{-3})
F	Faraday Constant (96487 C)
i	Current density (A·cm^{-2})
i_0	Exchange current density (A·cm^{-2})
i_L	Limit current density (A·cm^{-2})
M_-	Anode water balance (mol·s^{-1})
M_+	Cathode water balance (mol·s^{-1})
M_{max}	Water amount to complete flooding (mol·s^{-1})
p	Total pressure (atm)
p_{H_2}	H$_2$ pressure (atm)
p_{gas}	Air pressure (atm)
R	Ideal gas constant (8.3142 J·mol^{-1}·K^{-1})
T	Temperature (K)
E	Operation voltage (V)
E_{Nernst}	Nernst operation voltage (V)
W_{tr-EOD}	Water transport through the membrane (mol·cm^{-2}·s^{-1})
W^{hum}	Water inlet to the cell due to reactant gas humidity (mol·cm^{-2}·s^{-1})
W_{gen}	Water production at the cathode (mol·cm^{-2}·s^{-1})
W^{out}	Water exit from the cell (mol·cm^{-2}·s^{-1})
x	Molar fraction
a_{H_2O}	Water activity
x^{hum}	Molar fraction corresponding to humidification condition
V_{Ohm}	Ohmic loss (V)

Greek letters

σ	Membrane ionic conductivity (S·cm^{-2})
δ_{thick}	Membrane thickness (cm)
η_{act}	Activation overpotential (V)
η_{act}^{HOR}	Activation overpotential corresponding to hydrogen oxidation reaction (V)
η_{act}^{ORR}	Activation overpotential corresponding to oxygen reduction reaction (V)
η_{conc}	Concentration overpotential (V)
ρ	Density
ζ	Stoichiometry coefficient
α	Charge transfer coefficient
$\in^0$	Initial electrode porosity
$\in^{hum}$	Porosity corresponding to humidification condition
λ	Water content or local ratio H$_2$O/SO$_3^-$ in the membrane
η_{drag}	Electro-osmotic drag coefficient

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1. Introduction

Fuel cells are electrical power sources on which the chemical energy contained in a compound as hydrogen is spontaneously converted into electricity. The process is cleaner and more efficient than that based on burning fossil fuels, which is seriously harmful to the environment. As long as the reactant and oxidant are supplied, fuel cells can provide electricity and reach high efficiency, because they involve a single transformation step and unlike conventional heat engines, they are not under Carnot limitation.

Polymer electrolyte membrane fuel cells (PEMFCs) are very attractive as power sources in portable, transport and stationary applications, because they can deliver high power density at low temperatures, are noiseless and lightweight, have low maintenance and have no mechanical components. They are also friendly to the environment.

Nafion® membrane is usually employed in PEMFCs as ionomeric electrolyte, which is a peruorsulfonic acid membrane consisting of a hydrophobic polytetrauoroethylene backbone functionalized with hydrophilic sulfonic-acid at the end of lateral chains. The content of water in the membrane affects its ionic conductivity, because proton migration occurs in the ionomeric hydrated phase with participation of dissociated sulfonic groups [1-3]. In the search of highly efficient and reliable PEMFCs, considerable work has been done. Many internal and external factors affect their performance, such as catalytic performance, complex reaction pathways, materials degradation, mass transport, fuel cell design and assembly, operational conditions. [4-8].

A model of the processes taking place inside the fuel cells could help to understand what is the most important factors to improve and optimize its performance. Due to the complexity of the processes in real cells, one dimension and constant temperature are assumed, in most of the previously studied models [9-13]. In general, the models are able to give reasonable predictions of the cell performance in the low and intermediate current density ranges, but failed to reproduce the experimental curves at high current densities, where an abrupt potential drop is observed.

Some authors consider that the electrode porosity does not change during cell operation; however, it is well known that it can be affected by many factors when current is drained. One of the most significant factors is water accumulation in the electrode porous layer, which can provoke flooding, a non-uniform pore distribution and an increase in the reactant transport resistance [14].

The aim of this work is to evaluate the role of the relative humidity of the reactive gases needed to avoid the accumulation of water in the porous electrode. A simplified model which considers a H₂/O₂ PEMFC operating under steady state conditions at constant temperature with a single pore distribution is introduced and discussed.

2. Mathematical Model

The model to be considered takes the following premises into account:

The reactant concentration of both gases (H₂/O₂) is constant.

The fuel cell work under isothermal conditions.

The potential at the membrane electrode interface is constant.

The pressure of gases and fluids is constant.

There are no changes in the material properties in 20-90 ºC temperature range.

An ideal behavior of gases and fluids is considered.

A uniform pore distribution in the electrode is assumed.

The proton exchange membrane is always completely hydrated.

2.1 Description of the Fuel Cell

Typically, each individual PEMFC is made of a

layered structure consisting of an electrode-membrane-electrode assembly (MEA), which is constrained among two flow field plates, which plays two roles i) by collecting the current and ii) by supplying the reactants through the gas diffusion layer (GDL) to the catalytic active sites. The MEA comprises two electrodes separated by a thin Nafion® membrane (Fig. 1).

The electrodes are composed of two layers, the gas diffusion layer (GDL) and the catalytic layer (CL). The CL is the site where the electrochemical reaction takes place, whereas the GDL supplies the reactants (which run through the flow channels of the flow field plates) and it also drains the products from the CL (heat, water and electrons). The GDL needs to be efficient for the gas transporting from the gas flow channels to the electrolyte/electrode interface [15]. To achieve the goal it is necessary to maintain free void pores to ease the diffusive flow of the gases. It is well known that preventing water away from condensing in the pores and consequently from obstructing the gas pathway; the GDL material must be hydrophobic. Teflon treatments of the GDL reduce the amount of water in the pores facilitating the gas transport toward the catalytic sites.

It is important to stress that the GDL plays an essential role in regulating the flow of water away from the catalyst layer in PEM fuel cells. However, at higher current densities, the produced liquid water has to be removed with the effluent through the gas flow channels.

Some models assumed that water and gas move through separate pores. Some authors claimed that a distribution of pore sizes must be determined to follow the behavior of the GDL, closely. Thus Benziger et al. [16] suggest that water move through large pores and the small pores are kept free of liquid water.

During fuel cell operation (Fig. 2), H₂ reaches, by diffusion through the porous electrode, the anode reaction sites of the catalysts and the hydrogen
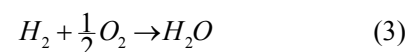
oxidation reaction (HOR) (as shown in Eq. (1)) takes place.

$$H_2 \rightarrow 2H^+ + 2e^- \tag{1}$$
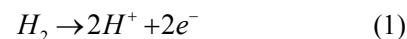
At the cathode, the oxygen reduction reaction (ORR) with protons and electrons takes place.

$$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O \tag{2}$$
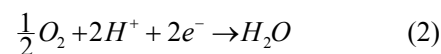
Consequently, the global fuel cell reaction is:

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \tag{3}$$

2.2 Water Accumulation in the Porous Electrodes

The water balance in the porous electrode (Fig. 3) results from the correlation between: i) the water inlet from the humidified reactant gases, ii) the water produced according to Eq. (2), iii) the internal water transport through the membrane between anode and cathode and iv) the water outlet from the humidified gases which have not reacted [17].
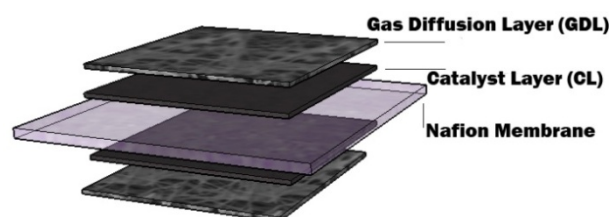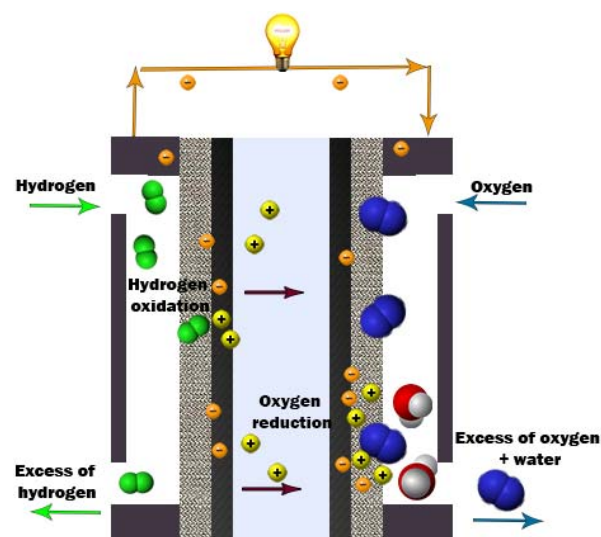

Fig. 1   Membrane-electrode assembly (MEA).

Fig. 2   Schematic drawing of a H₂/O₂ fuel cell and the reactions involved in a PEMFC.
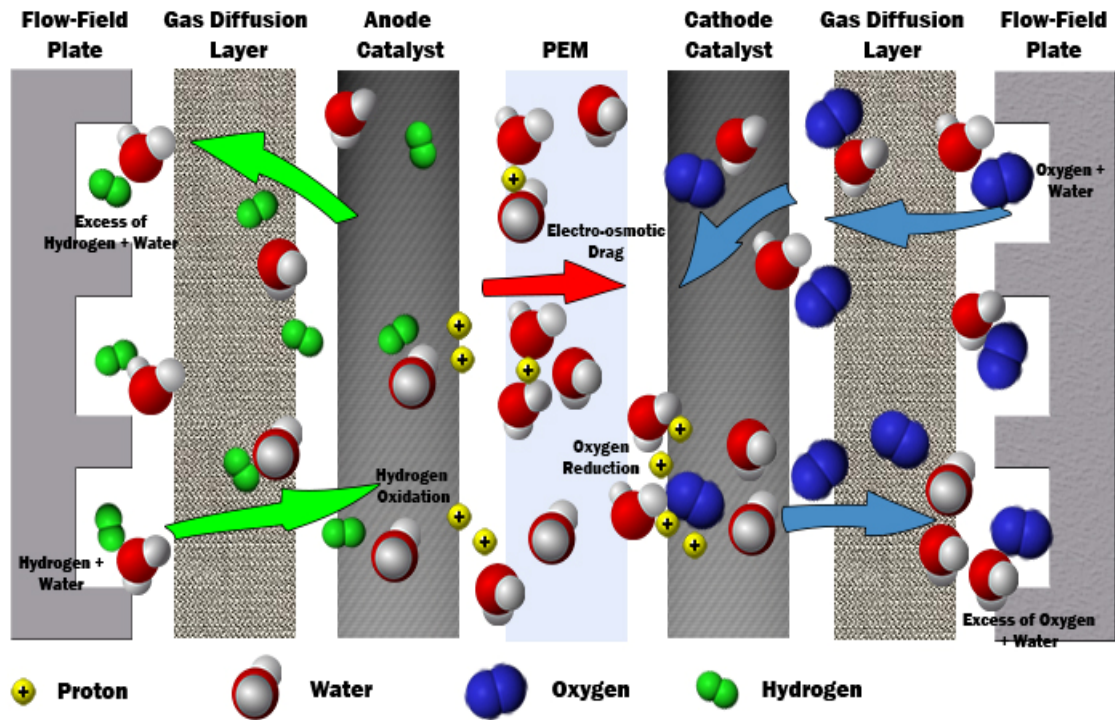
Fig. 3   Water transport in a typical PEM fuel cell.

The inner water transport through the membrane results from many contributions; among the most important, it can be mentioned:

• The electro-osmotic drag (EOD) associated to proton migration, which can be characterized by the electro-osmotic drag coefficient described by the number of water molecules moving with each proton [11].

• The back-diffusion due to the water concentration gradient between cathode and anode.

• The thermal osmotic drag (TOD) caused by differences in temperature along the membrane.

• The hydraulic permeation due to different pressures on each side of the membrane.

The model is developed considering that i) the fuel cell is working in an isothermal condition, ii) the water amount in the membrane is constant, iii) no pressure gradient is applied, consequently, the thermal osmotic drag, the hydraulic permeation and the back diffusion caused by concentration gradient can be neglected.

2.3 Water Inlet

Relative humidity is defined as the ratio of the actual water vapor pressure to the saturation water pressure, usually expressed in percentage. A given amount of water coming with the reactant gases (H$_2$, air/O$_2$) is taken into account in line with Eqs. (4) and (5) [10].

$$W_-^{hum} = \frac{i}{2F} \zeta \frac{x_-^{hum}}{1 - x_-^{hum}} \qquad (4)$$

$$W_-^{hum} = \frac{i}{4F} \zeta \frac{x_+^{hum}}{1 - x_+^{hum}} (1 + \frac{x_{N_2}^0}{x_{O_2}^0}) \qquad (5)$$

2.4 Water Produced

A given amount of water is obtained from the ORR and can be taken into account according to:

$$W_{gen} = \frac{i}{2F} \qquad (6)$$

2.5 Water Outlet

A certain amount of the reactive gases are not able to react in the PEMFC and they exit taking a certain

amount of extra water produced by the electrochemical reaction.

$$W_-^{out} = \frac{i}{2F}(\zeta - 1)\frac{x_w^{out^-}}{1 - x_w^{out^-}} \qquad (7)$$

$$W_+^{out} = \frac{i}{4F}\left[\zeta + (1 + \frac{x_{N_2}^0}{x_{O_2}^0}) - 1\right]\frac{x_w^{out^+}}{1 - x_w^{out^+}} \qquad (8)$$

2.6 Water Transport through the Polymeric Membrane

The electro-osmotic drag is associated to the protons transport from the anode to the cathode in the PEMFC. Springer et al. [11] define the number of water molecules for each transported proton as:

$$\eta_{drag} = \frac{2.5\lambda}{22} \qquad (9)$$

Being the dragged water amount:

$$W_{tr-EOD} = \frac{i}{F}\eta_{drag} \qquad (10)$$

2.7 Cathodic and Anodic Water Balance

According to the mass balance expressed in moles in each compartment the total amount of water results:

$$M_- = W_-^{hum} - W_{tr-EOD} + W_-^{out} \qquad (11)$$

$$M_+ = W_+^{hum} + W_{gen} + W_{tr-EOD} + W_+^{out} \qquad (12)$$

In the following equation, the variation of the porosity in the cathode is expressed as:

$$\varepsilon^{hum} = \varepsilon^0(1 - \frac{|M_+|}{M_{max}}), \, if \, M_{max} > 1 \qquad (13)$$

3. Effect of Water Accumulation in the PEMFC Performance

The fuel cell fed with H₂ and O₂ has a cell potential value of 1.229 V at standard conditions (25 ℃, activity or concentration of reactants and product equal 1 and generation of liquid water). When the activity of the reactive and products are different from 1, the cell potential should be corrected according to Nernst, equation and the cell potential value becomes lower than 1.229 V.

$$E_{Nernst} = 1.229 + \frac{RT}{nF}\ln\frac{\alpha_{H_2O}}{\alpha_{H_2}\alpha_{O_2}^{\frac{1}{2}}} \qquad (14)$$

The fuel cell performance can be determined by measuring the relationship between the fuel cell voltage and the drained current. The polarization curve (fuel cell voltage vs. current density drained) is depicted in Fig. 4, the curve shows important cell potential shifts from the E_{Nernst} value. Depending on the current density values, the fuel cell voltage losses can be associated to different resistances identified in three main current density regions, the first one at low current densities caused by the electron transfer reaction (activation overpotential) denoted as η_{act}; at mid-current densities, prevalent ohmic loss is attributed to the membrane resistance to transport hydrogen ions, we identified the ohmic loss as V_{Ohm}, Finally at high current densities, there is a limitation in the transport of the reactive to catalytic sites and we named it as concentration overpotential η_{conc}.

Thus, after taken into account all the potential losses, the cell potential can be written as:

$$E = E_{Nernst} + V_{Ohm} + \eta_{act} + \eta_{conc} \qquad (15)$$

3.1 Activation Overpotential

The activation overpotential is defined as the sum of the activation overpotential for each electrochemical reaction that takes place on each fuel cell electrode:
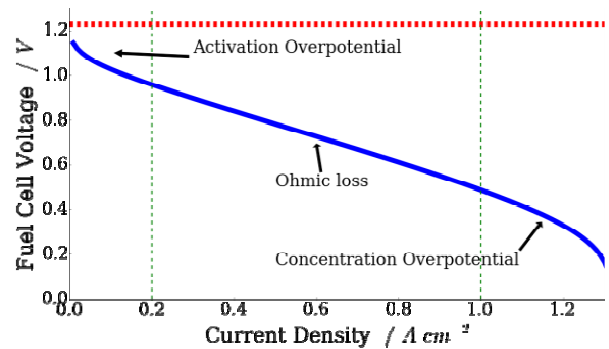

Fig. 4 Different overpotential contribution to the polarization curve. Ideal voltage 1.229 V (----), polarization curve (——).

$$\eta_{act} = \eta_{act}^{HOR} + \eta_{act}^{ORR} \qquad (16)$$

The HOR on Pt is a fast reaction, whereas the opposite occurs with the ORR, which is a slow catalytic reaction [18]. Consequently the η_{act}^{HOR} can be neglected. Then:

$$\eta_{act} = \eta_{act}^{HOR} \qquad (17)$$

The activation overpotential is written with regards to only ORR, its relation with the current density is expressed according to the classic Butler-Volmer equation as [19]:

$$\eta_{act} = -\ln\left(\frac{i}{i_0}\right)\left(\frac{RT}{\alpha F}\right) \qquad (18)$$

3.2 Ohmic Losses

Ohmic loss related to membrane resistance to hydrogen ions transport can be described by employing the Ohm law.

$$V_{ohm} = -iR_{Ohm} \qquad (19)$$

The ohmic resistance depends on the membrane conductivity and thickness [20, 21] expressed as:

$$R_{Ohm} = \frac{\delta_{thick}}{\sigma} \qquad (20)$$

3.3. Concentration Overpotential

The concentration overpotential is caused because the reactant cannot reach the catalytic sites. In the polarization curve, the mass transport influence is noticed at high current densities values [22-24].

$$\eta_{conc} = -\frac{RT}{nF}\ln\left(\frac{i_L}{i_L - i}\right) \qquad (21)$$

where,

$$i_L = -nF\frac{C_0}{\left(\dfrac{1}{h_m} + \dfrac{\delta}{D_g + \epsilon_{hum}^{\frac{1}{2}}}\right)} \qquad (22)$$

When liquid water accumulates in the GDL, the reactant transport throughout the flow-field channel to the catalyst layer is hindered and, consequently, it reduces the power output from the fuel cell. The liquid water accumulation is especially severe at the cathode where water is formed in the catalytic layer at the electrolyte/electrode interface. At high current densities water accumulation avoids oxygen from getting the electrode/electrolyte interface causing mass transport restrictions which limits the maximum current density drained. The ideal GDL should permit water to be removed from the electrode/electrolyte interface without hindering gas transport.

4. Results and Discussion

The values of current density and cell potential obtained after replacement of Eqs. (18), (19) and (21) in Eq. (15) are depicted and superimposed to the experimental current density vs. cell potential values obtained by Maggio et al [25] by using a PEMFC with H₂ and O₂ flux, cathode feed gas = Air, cathode gas pressure = 3 atm, T = 343.15 K. The simulated curve current density-cell potential shows an agreement with the experimental data according to Fig. 5.

Simulated polarization curves and power curves shown in Figs. 6 and 7, demonstrate the importance of the relative humidity content in the inlet air/O₂ gas. The values of the most significant parameters needed for calculations are shown in Table 1. From the analysis of the curves it can be concluded that a better fuel cell performance is possible with relatively low water content in the inlet air. The cell performance loss at high humidity values could be attributed to water accumulation in the pores of the GDL which
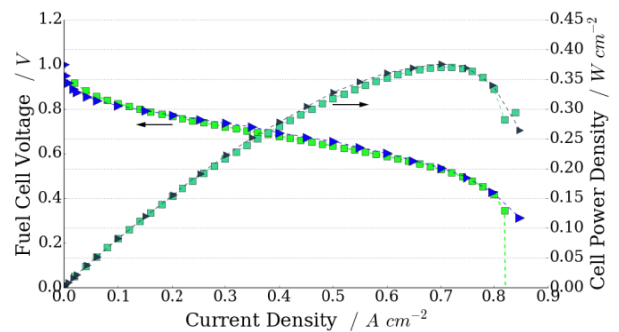

Fig. 5 Polarization curve of a H₂/O₂ fuel cell working at 70 ºC: experimental data of Maggio et al. [25] (-►-). Simulated data from Eq. (15) (-■-).
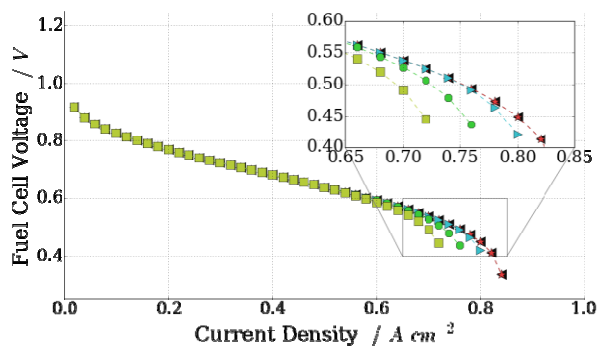
Fig. 6   Polarization curves of H₂/O₂ fuel cell at different relative humidity in air: RH = 20% (-◄-), RH = 30% (- -), RH = 40% (-►-), RH = 50% (-●-), RH = 60% (-■-). Polarization curves magnification in the current density region between 0.65 and 0.85 A·cm⁻².
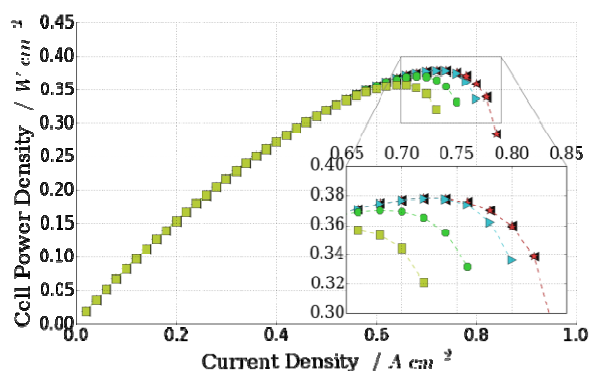

Fig. 7   Power curve of a H₂/O₂ Fuel Cell at different relative humidity: RH = 20% (-◄-), RH = 30% (- -), RH = 40% (-►-), RH = 50% (-●-), RH = 60% (-■-).

Table 1   Input data required for calculations.

Datum	Value
Cell temperature (K)	343.15
Cathode feed gas	Air
Thickness of the Nafion® membrane (μm)	183
Pressure of the gas inlet (H₂ and Air) (atm)	3
Ideal gas constant (J·mol⁻¹·K⁻¹)	8.31434
Faraday constant (C·mol⁻¹)	96,485.33
Initial electrode porosity	0.74
Cathode stoichiometry coefficient	3
Molar fraction of O₂ in air	0.21

prevents the gas from reaching the catalytic sites.

In Fig. 8, changes in the porosity of the GDL with the current density drained and different relative humidifities, is shown. Water can coalesce in the inside of the pore when the relative humidity of the gas is increased (as happens at high current densities), resulting in fewer pores available for gas ux. This brings
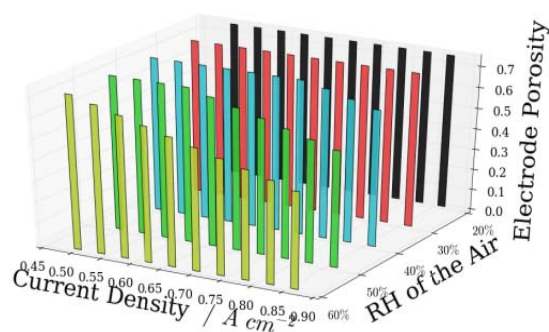

Fig. 8   Changes in the electrode porosity due to water accumulation caused by different relative humidity values in the inlet gas (air/O₂) and the current densities drained.
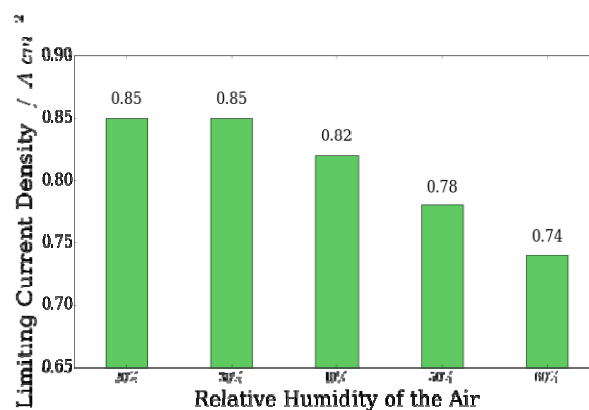

Fig. 9   Changes in the limiting current density with the relative humidity of the inlet gas (air/O₂).

about mass transport limitations and a decrease in the performance of the fuel cell. It is noticed that for relative humidity values higher than 30% there is a decrease in the porosity available when the current density drained is higher.

Fig. 9 depicts the limiting current density and its dependence on the air relative humidity: it is possible to predict the maximum allowed water content in the gas inlet to avoid affecting the maximum current density drained.

Both Figs. 8 and 9 draw attention to the influence of the water content in the porous electrode on the cell performance, thus, the higher the relative humidity percentage, the lower the pore accessibility.

5. Conclusions

To achieve an optimal PEMFC performance it is necessary to determine the maximum water content in

the cathode gas inlet in order to avoid flooding during fuel cell operation. A theoretical approach of how PEMFC works is proposed. Of the many parameters that play a part in improving the PEMFC behavior, the water content in the gas that fed the cathode is systematically studied in reference to PMFC performance by using a simple pore approach of the GDL.

A model is presented and discussed in which PEMFC operating under steady state and isothermal conditions together with a simple and uniform porous electrode structure, is assumed. From the current potential relationship obtained, it is concluded that for relative humidity values in the air higher than 30%, the fuel cell performance starts to decay. The losses in efficiency are ascribed to the partial flooding of the porous structure of GDL, which is more severe at higher current densities.

Acknowledgments

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