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Short communication

The dynamic and steady state behavior of a PEM fuel cell as an electric energy source

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Abstract

The main objective of this work is to extract information on the internal behavior of three small polymer electrolyte membrane fuel cells under static and dynamic load conditions. A computational model was developed using Scilab [SCILAB 4, Scilab—a free scientific software package, http://www.scilab.org/, INRIA, France, December, 2005] to simulate the static and dynamic performance [J.M. Correa, A.F. Farret, L.N. Canha, An analysis of the dynamic performance of proton exchange membrane fuel cells using an electrochemical model, in: 27th Annual Conference of IEEE Industrial Electronics Society, 2001, pp. 141–146] of this particular type of fuel cell. This dynamic model is based on electrochemical equations and takes into consideration most of the chemical and physical characteristics of the device in order to generate electric power. The model takes into consideration the operating, design parameters and physical material properties. The results show the internal losses and concentration effects behavior, which are of interest properties and researchers.

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1. Introduction

In the last decade, a group of researchers [5,6] published a series of papers on electrochemical problems relating to PEM fuel cells, from modeling to the problems of poisoning of the cell, their work aimed at the need for a model based on electrochemical equations to correctly model this new electrical power source. Almost 10 years later another group of researchers has focused on electrochemical reaction modeling [4], but like previous publications, their model is based on the assembling of a prototype fuel cell and laboratory experiments.

More recent work by Souza and Gonzales [15] has presented a mathematical model for the PEM Fuel Cell, and their modeling has allowed detailed studies. They discussed the electrocatalysis of the reactions and the design of water-management schemes to

0378-7753/\$ - see front matter © 2006 Elsevier B.V. All rights reserved. doi:10.1016/j.jpowsour.2006.04.149 cope with membrane dehydration. Mishra, Yang and Pitchumani [18], did a detailed numerical investigation of the transport and electrochemical phenomena involved in the operation of a single proton exchange membrane (PEM) fuel cell with reformate feeding with a view to developing optimal design and operating conditions.

Along with a large number of methodologies, Cheddie and Munroe [16] did a review of recent literature on proton exchange membrane fuel cell modeling, and they have categorized them as analytical, semi-empirical or mechanistic. Mechanistic modeling has received the most attention in the literature. In mechanistic modeling, differential and algebraic equations are derived based on the physics and electrochemistry governing the phenomena internal to the cell.

Mehta and Cooper [17] did an extensive review and analysis of PEM fuel cell designs and manufacturing. Their work had an important influence on the work we present here, Govelioglu and Stenger [19] developed research on computational fluid dynamics modeling of PEM fuel cells. They used a finite element method to solve the transport model coupled with flowing

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Nomenclature

Α	anode (or cathode) area/cell useful area (cm ²)
$c^*_{ m H_2}$	hydrogen concentration in the interface electrode
	membrane (mol cm^{-3})
$C_{\text{ion,m}}$	membrane ionic concentration (mol cm^{-3})
E^0	open circuit voltage (V)
F	Faraday constant (96487 $C \mod^{-1}$)
ΔFE_a	anode reaction activation energy $(J \text{ mol}^{-1})$
ΔFE_{c}	cathode reaction activation energy $(J \text{ mol}^{-1})$
ΔG	Gibbs free energy variation (-237.165 J)
ΔH	variation of enthalpy (-285.823 J)
<i>i</i> op	operational cell current
$i_{\rm L}$	electrode current limit
$i_{\rm L,a}$	electrode current limit in the anode
$i_{\rm L,c}$	electrode current limit in the cathode
I _{op}	operational electrical current (A)
$j_{ m op}$	cell operation current density $(A \text{ cm}^{-2})$
j_0	exchange current density in the electrode
- 0	$(A \text{ cm}^{-2})$
k^0	reaction degree intrinsic constant (cm s^{-1})
l	membrane thickness (cm)
$m_{\rm air}$	mass flow of air (dimensionless)
$m_{ m H_2}$	mass flow of hydrogen (dimensionless)
m_{O_2}	mass flow of oxygen (dimensionless)
n	number of moles involved in a reaction for each
	mol of H_2 (dimensionless)
na	number of moles of reagent in the anode
$n_{\rm c}$	number of moles of reagent in the cathode
$P_{\rm H_2}$	partial pressure feeding for hydrogen (atm)
P_{O_2}	partial pressure feeding for oxygen (atm)
K D	universal constant of gases (8314 J K mol ⁻¹)
ĸ _{cj}	resistance of gas spreaders and separating plates (Ω)
R _m	resistance of the electrodes (Ω)
ΔS	variation of entropy $(-163.2 \text{ J K}^{-1})$
$T_{\rm op}$	operational temperature (K)
$T_{\rm ref}$	reference temperature (298.15 K^{-1})
$V_{\rm atv}$	activation voltage drop (V)
$V_{\rm con}$	concentration voltage drop (V)
$V_{\rm ohm}$	Ohmic voltage drop (V)
$V_{\rm op}$	operational voltage (V)
Greek le	etters
α	electronic transference current (around 0.4 for the
	cathode and 1.0 for the anode)
$\alpha_{\rm c}$	cathode load transference coefficient (dimension-
	less)
α_{a}	anode load transference coefficient (dimension-
	less)
γ	membrane ionic conductance (S cm ² mol ^{-1})
$\delta_{\rm c}$	cathode Nernst diffusion layer thickness (cm)
δ_{a}	anode Nernst diffusion layer thickness (cm)
λ_{air}	stoichiometry of air (dimensionless)
$\lambda_{\rm H_2}$	stoichiometry of hydrogen (dimensionless)
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λ_{O_2}	stoichiometry of oxygen (dimensionless)
$ ho_{ m m}$	electrical resistivity of the membrane $(\Omega \text{ cm})$
$\phi_{ m H_2}$	relative humidity of hydrogen (%)
ϕ_{O_2}	relative humidity of oxygen (%)

in a porous medium, charge balance, electrochemical kinetics, and a rigorous water balance in the membrane.

These papers have contributed greatly to comprehension of the global electrochemical processes in a fuel cell, but did not improve understanding of the fuel cell as an electrical energy source, and that is our proposal in this work, i.e. to make a model [1] suitable for use with power electronics hardware.

The basic physical structure of a fuel cell used in the computer model consists of an electrolyte and a polymeric membrane, with a typical thickness of between 12 and 210 µm, which separates the porous anode and cathode from each other; they have a thickness of between 5 and 15 μ m. The porous and slender electrodes, make it possible to assemble a membrane electrode assembly (MEA). In a typical fuel cell, a flux of fuel which is rich in hydrogen (liquid or gaseous) is fed continuously through channels that keep it in contact with the anode and, simultaneously, a flux of an oxidant (in general the oxygen in the air) are also fed through different and separate channels, which make the contact with the cathode [2]. The layers in between are responsible for the gas diffusion process in the electrodes, with a thickness between 300 and 400 µm, they are made of an electrical conducting material, like porous carbon paper, a carbon based fabric with a Teflon[®] layer that prevents water adherence and make possible the fast diffusion of gases [3].

The process that takes place is a natural reaction, however due to its slow speed it is necessary to use a catalyst for the reaction to be efficient, the element used as catalyst at the present is platinum. The other two important elements in the fuel cell are, firstly the separating plates, to give mechanical stiffness and to direct the flow of gas through the cell, and secondly the cooling elements that have the function of extracting heat from the exothermal reactions, the ratio used for interspersing the cooling elements are used in a proportion that varies from 1:1 to 1:5 [5].

2. Mathematical modeling of the PEM fuel cell

A mathematical model that presents the correct answers for the fuel cell is of paramount importance, since it allows the development of electronic systems for feeding and control of fuel cells without the need for a great number of experimental prototypes. Electrically speaking, a fuel cell works basically as a current source. The voltage response for the cell will be studied from the current variation required by the load. The main operational parameters are: the operational electrical current, temperature, and the characteristics of gas transportation.

2.1. Activation losses

2.1.1. Cathode activation losses

The consideration on the effect of operating variables in the value of i_0 in the cathode can be taken as [6]:

$$i_{0c} = n_c FAk^0 (C_{\text{proton}}^*)^{1-\alpha_c} (C_{\text{H}_2\text{O}}^*)^{\alpha_c} (C_{\text{O}_2}^*)^{1-\alpha_c} e^{(\Delta FE_c/RT_{\text{op}})},$$
(1)

Working mathematically in the previous equation we can have for the cathode the following value for the activation voltage [4]:

$$V_{\text{atvc}} = \xi_1 + \xi_2 T_{\text{op}} + \xi_3 T_{\text{op}} \ln(C^*_{\text{O}_2}) + \xi_4 T_{\text{op}} \ln(i)$$
(2)

where

$$\xi_1 = \frac{\Delta FE_c}{n_c \alpha_c} \tag{3}$$

$$\xi_2 = \frac{R}{n_c F \alpha_c} \ln(4Fk^0) + \frac{R}{n_c F \alpha_c} \ln(A) + \frac{R}{n_c F} \ln(C_{H_2}^*)$$
(4)

$$\xi_3 = \frac{R(1 - \alpha_c)}{n_c F \alpha_c} \tag{5}$$

$$\xi_4 = \frac{R}{n_c F \alpha_c} \tag{6}$$

2.1.2. Anode activation losses

With a more detailed Eq. (1), taking in consideration the effect of operational variables in the value of i_0 at the anode as [6]:

$$i_{0,a}(A) = n_a FAk^0 C_{H_2}^* e^{(-\Delta FE_a/RT_{op})}$$
(7)

The activation voltage [4] in the cathode can be given by:

$$V_{\text{atv,c}}(V) = \xi_5 + \xi_6 T_{\text{op}} \ln(C_{\text{H}_2}^*) + \xi_7 T_{\text{op}} \ln(i)$$
(8)

where

Table 1

$$\xi_5 = \frac{\Delta F E_a}{n_a F \alpha_a} \tag{9}$$

$$\xi_6 = \frac{-R\ln(n_a FAk^0)}{n_a F\alpha_a} \tag{10}$$

$$\xi_7 = \frac{R}{n_{\rm a} F \alpha_{\rm a}} \tag{11}$$

As previously stated, the activation voltage at the anode will have lower values compared with the activation voltage at the cathode, and can be neglected in the operating behavior of the PEM fuel cell fed with pure hydrogen.

2.2. Resistive losses

To illustrate the empirics to obtain fuel cell operating parameters, the first part of the resistive losses (R_{ohm}) is given by the polymeric membrane and the second part by the electrodes, spreaders and other series components.

2.2.1. Polymeric membrane resistive losses

The resistance in Ohm for this membrane (R_m) which represents the resistance for transference of protons through the solid membrane [14], and can be computed by the two following equations:

$$R_{\rm m}\left(\Omega\right) = \frac{\rho_{\rm m}l}{A} \tag{12}$$

The resistivity values for the Nafion[®] [9] membranes (produced by DuPont) to complement the ohmic resistance can be computed through the following equation:

$$\rho_{\rm m} \left(\Omega \,{\rm cm}\right) = \frac{181.6 \left[1 + 0.03(i_{\rm op}/A) + 0.0062(T_{\rm op}/303)^2(i_{\rm op}/A)^{2.5}\right]}{\left[\lambda - 0.634 - 3(i_{\rm op}/A)e^{4.18((T_{\rm op}-303)/(T_{\rm op}))}\right]}$$
(13)

For the Nafion[®] membrane, the typical thickness in each case is indicated in Table 1 [8,12].

Therefore, λ is the ratio of the number of water moles for each sulfonic group in the membrane. The value of λ indicates the membrane relative humidity, being regulated by the feeding gases relative humidity, their stoichiometries and assembling characteristics involved in the efficiency of the gas transportation and membrane humidity maintenance. The literature indicates a value of approximately 14 for λ corresponding to a membrane in humidified air at 100%, and approximately to 22 the value corresponding to a water immersed membrane [4].

2.3. Concentration losses

2.3.1. Concentration losses at the cathode

The value for the current limit at the cathode $(I_{L,c})$ will be adopted [13] through the following equation:

$$i_{\rm L,c}(A) = \frac{RT_{\rm op}A\gamma C_{\rm ion,m}}{n_{\rm c}F\delta_{\rm c}}$$
(14)

2.3.2. Concentration losses at the anode

They will be simulated as in Eq. (14), and the value for the current limit at the anode $(I_{L,a})$ will be adopted through the fol-

rable 1			
Typical	thickness	of Nafion [®]	membranes

Membrane	Nominal thickness (mm)	Thickness when dry (mm)	Thickness when wet (mm)	Thickness when MEA (mm)
Nafion 117	0.178	0.183 ± 0.003	0.208 ± 0.005	0.148 ± 0.002
Nafion 115	0.127	0.141 ± 0.003	0.161 ± 0.003	0.100 ± 0.002
Nafion 113	0.089	0.091 ± 0.002	0.111 ± 0.002	0.075 ± 0.003
Nafion 112	0.051	0.050 ± 0.002	0.058 ± 0.003	0.040 ± 0.002



Fig. 1. Typical elemental operating scheme for a PEM fuel cell stack.

lowing equation:

$$i_{\rm L,a}(A) = \frac{RT_{\rm op}A\gamma C_{\rm ion,m}}{n_a F \delta_a}$$
(15)

2.4. PEMFC cell under dynamic load change

When a variation occurs in the load fed by a fuel cell, this is the cause for a variation in the cell operating voltage. However, a transient state exists due to the capacitance generated in the polymeric membrane due to an internal physical process called "double layer effect". [7]

The equation that represents the voltage behavior during load transients can be expressed as:

$$v(t)(\mathbf{V}) = v(\infty) + [v(0+) - v(\infty)]\exp\left(-\frac{t}{\tau}\right)$$
(16)

where $v(\infty)$ is the final value to which the voltage will go after the load variation, and v(0+) is the voltage initial value, before the variation of the load fed by the PEM fuel cell. The time constant (τ) is a measure of the necessary time for the transient analysis in RC circuits, computed by the following equation:

$$\tau(s) = R_{\rm th} C_{\rm dc},\tag{17}$$

and R_{th} is the Thevenin equivalent resistance of the circuit seeing from the capacitance.

3. Methods

In order to develop this study, computational modeling tools were built up to model the fuel cell behavior through a detailed theoretical base taking into consideration assembling and operating parameters.

With this purpose; to build the fuel cell simulator besides the operating parameters already mentioned (T_{op} , I_{op} , P_{H_2} , P_{O_2} , ϕ_{H_2} , ϕ_{O_2} , λ_{H_2} , λ_{O_2} , or λ_{air}), the constructive parameters (electronic transference coefficients, electrode reaction level constants, and the physicochemical properties of electrodes and membrane), and the dimensioning parameters for the fuel cell stack (number of cells, cell area and membrane thickness) were also considered. The simulation results are compared with practical experiments.

The experimental tests were made through controlled processes using different models and prototypes of PEMFC fuel cell stack as the system shown in the basic scheme of Fig. 1.

Under normal operation a frequency inverter is used to feed ac energy due to the voltage regulation showed by the fuel cell. An electronically controlled resistive load was used in order to make possible a better control of the necessary electrical load during laboratory experiments.

4. Results

4.1. PEM fuel cell with a load in steady-state

Fig. 3 shows the behavior of a cell with net area of 5 cm^2 , fed by hydrogen and pure oxygen with feeding pressures of 1 atm, stoichiometries of 1.1, humidification 100% under operating temperatures of 40, 60 and 80 °C for a defined loading, being the curves assembled from the cell voltage response according to the current requested by the load. Similar curves can be obtained fixing the temperature and changing other operating parameters.

Fig. 2 shows the behavior of the cell with the variation of temperature for the first prototype with constants described in Table 2 [20]:

Table 2	
Design characteristics for the first prototype	

Description	Symbol	Amount
Area of the cell	A _{cel}	5 cm ²
Number of cells	N _{cel}	1
Membrane	_	Nafion [®] 112
Electronic transfer coefficient (cathode)	$\alpha_{\rm c}$	0.43
Electronic transfer coefficient (anode)	α_{a}	1.0
Reaction grade constant (cathode)	$k_{\rm c}^0$	$2.7cms^{-1}$



Fig. 2. Behavior of a PEM fuel cell under static load with the temperature.



Fig. 3. Behavior of a PEM fuel cell under static load with the gas pressure.

Fig. 3 shows the behavior of the cell with the variation of pressure for the second prototype with constants described in Table 3 [20]:

The third prototype was used for experimental and dynamic simulation tests, it is a fuel cell stack built from two cells, assembled in a test bench with main design characteristics as shown Table 4 [20].

Table 3	
Design characteristics for the second prototyp	e

Description	Symbol	Amount
Area of the cell	A _{cel}	20 cm ²
Number of cells	N _{cel}	1
Membrane	_	Nafion [®] 112
Electronic transfer coefficient (cathode)	$\alpha_{\rm c}$	0.43
Electronic transfer coefficient (anode)	$\alpha_{\rm a}$	1.0
Reaction grade constant (cathode)	$k_{ m c}^0$	$3.5\mathrm{cms^{-1}}$

Table 4	
Design characteristics for the third prototype	

Description	Symbol	Amount
Area of the cell	A _{cel}	80 cm ²
Number of cells Membrane	N _{cel}	2 Nation [®] 112
Electronic transfer coefficient (cathode)	$-\alpha_c$	0.43
Electronic transfer coefficient (anode)	α_a	1.0
Reaction grade constant (cathode)	$k_{\rm c}^0$	$1.3 {\rm cm s^{-1}}$

4.2. Utilization of air instead of pure oxygen

For this analysis the second prototype was used with air or oxygen as a cathode reagent with an operating temperature of $50 \,^{\circ}$ C. The other cell parameters are the ones specified in Table 2. The values obtained experimentally and through computational simulation are indicated in Fig. 4 [20].

The result above shows the developed model efficiency and the success in the collection of constructive parameters, and also demonstrates the influence of the use of atmospheric air and pure oxygen.

Practically, this substitution, although showing loss of efficiency, is advantageous for commercial projects, avoiding the need for oxygen transport and storage, since one of the major challenges for the PEM fuel cells is to find an efficient way to store bulky amounts of gaseous fuels like hydrogen and oxygen.

The decreasing efficiency is due to the lower oxygen concentration in the composition of atmospheric air, and to the consequent increase in the cathode concentration effect (Fig. 5) [20]. The ohmic voltage drop and activation effect are not dependent on this parameter.

A second reason for this variation is the concentration effect on the cathode. With a decrease in concentration of the fuel gas, the maximum possible current in this electrode decreases [11]. Additionally the experiment was at a relatively low temperature (50 $^{\circ}$ C), which makes the cell response curve for high current



Fig. 4. Behavior of a PEM Fuel Cell for pure oxygen and air.



Fig. 5. Ohmic and activation losses when comparing feeding of pure oxygen and air.



Fig. 6. Concentration effects when comparing feeding of pure oxygen and air.

densities close to the saturation limit (the oxygen concentration in the membrane–electrode interface is close to zero). This phenomenon occurs in a region where the power is decreasing.

Fig. 6 shows also that the anode concentration effect is minimal, and due to this very small value, is irrelevant to the process [20].

5. Conclusion

As can be seen from this investigation, the coupling of a fuel cell with an inverter to produce ac current is not as simple as it seems. The fuel cell as an energy source changes its internal parameters dynamically for each load or operating point. The inverter design is not simple since its characteristics should be in agreement with the fuel cell variable parameters.

The study of the dynamic behavior shows that the electrical *RC* constant can change considerably with the control setup for

the stack with all internal parameters and inputs, and also shows that for a certain operating point, the *RC* constant during the load increase is larger than during the load decrease; opposite to the fact that the resistance in general increases with the value of current and temperature.

The implementation of this model allows the assembling of different sizes of PEMFCs, and shows the effects of variations on the many components of the performance of the cells.

Anode concentration and activation effects proved to be much less dependent of the type of feeding (air or pure O_2) than the effects at the cathode, as can be seen in Fig. 6. Ohmic voltage drops have a non-linear dependency with current and can be smaller than the anodic activation losses for a given operating current density. The developed computational simulator was demonstrated to be an efficient and valuable tool in the development of polymeric membrane fuel cell stacks; making possible an estimation of the cell performance [10].

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