Evaluation of the Dynamic Characteristics of Proton Exchange Membrane Fuel Cell Generating Systems

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***Abstract* – This paper makes an evaluation of the main dynamic characteristics present in electrical generating systems using Proton Exchange Membrane (PEM) fuel cells (FC). For this study it is used an electrochemical model for representation, simulation and analysis of the performance of low size generating systems using PEM fuel cells. The results of the model are used to predict the output voltage, efficiency and power of FC´s, the rates of hydrogen and oxidant (air) utilization, generated heat and water, as functions of the actual load current and of the constructive and operational parameters of the cells. Beyond conventional tests, total load insertion and rejection tests were accomplished to evaluate the dynamic response of the studied models in these extreme situations. The results guarantee a better analytical performance of these models with the consequent reduction of time and costs of projects using fuel cells as a primary source of energy.**

I. INTRODUCTION

Relative environmental reasons for the use of fossil fuels have been creating, along the years, a steadily increasing demand for new conversion technologies and systems of non-pollutants energy generation. Within this context, fuel cells (FC´s) have been showing up as a highly promising alternative for their high efficiency, low aggression to the environment, excellent dynamic response and superior reliability and durability in space, automotive and stationary applications. Especially, the Proton Exchange Membrane Fuel Cells (PEMFC) look as a great alternative as an element of distributed sources of energy. PEMFC produces water as residue; operate at low temperatures, what allows fast start-up, and use a solid polymer as the electrolyte, what reduces construction, transportation and safety concerns.

In its operation, a FC produces electrochemical power due to the passage of a rich gas in hydrogen through an anode and of oxygen (or air) through a cathode, with an electrolyte among the anode and cathode to enable the exchange of electrical charges (ions). The ion flow through the electrolyte produces an electrical current in an external circuit or load. Any hydrocarbon material, in principle, can be used as fuel, irrespective to be gas, liquid or solid. These materials have, however, to pass through a reformer to liberate the hydrogen from the carbon. The natural gas, for example, is reformed through vapor and high temperatures. In normal operation, a simple FC typically produces 0.5 to 0.9 V. For use in energy generation systems, where a relatively high power is needed, several cells must be connected in series, forming a stack.

To allow the evaluation of the PEMFC dynamic performance in low size electrical energy generation systems, to reduce cost and time during the projects and tests, the existence of a reliable mathematical model present itself as an important tool to the growing use of FC´s. In this paper, an PEMFC electrochemical model is presented to determine the resulting cells output voltage for each operating point. The model, also, allows the determination of stack power and efficiency, the rates of fuel and oxidant and the generated heat and water. In power generation systems, the dynamic response is of extreme importance for the planner of control and management systems; especially when there is injection of energy into the mains grid, when the control system should decide which amount of power the FC will supply to the mains grid, as a function of the load demand. The dynamic response of the FC should be compatible with the variations of a random load curve. So, a special attention is given to the FC dynamic response, differently of most of the models presented in the literature [1-5].

This way, the paper presents FC performance evaluation using commonly situations found in electrical power generation, like insertion and rejection of load and the best generating point. It is possible, also, the development of several control techniques for operation of the PEMFC, as the ones using Fuzzy Logic and Hill Climbing Control. For practical evaluation, the parameters of a Mark V cell, manufactured by the Canadian company Ballard, are used, whose operation and data are well known in the literature, allowing the comparison of the simulation results with practical tests.

II. BASIC FUEL CELL OPERATION

A PEMFC converts the chemical energy of a fuel, just as the hydrogen (H_2), and an oxidizer, just as the oxygen (O_2), in electrical energy. The outline of a typical PEMFC is illustrated in Fig. 1. On the anode side of the cell, the fuel is supplied under certain pressure. The fuel for this model is the pure gas H_2, although other hydrocarbon gases can be used. The fuel spreads through the electrode until it reaches the anode catalytic layer where it reacts to form protons and electrons:

$$H_{2(g)} \rightarrow 2H^{+} + 2e^{-} \qquad (1)$$

The protons are transferred through the solid electrolyte to the cathode catalytic layer. On the cathode side of the cell, the oxidizer (air or O_2) flows in the channels of the plate and it spreads through the electrode until it reaches the cathode catalytic layer. The oxygen is consumed with the protons and electrons and the product, liquid water, is produced with residual heat in the surface of the catalytic particles. The cathode electrochemical reaction is represented by:

$$2H^{+} + 2e^{-} + 1/2O_2 \rightarrow H_2O + \text{heat} \qquad (2)$$

Then, the global chemical reaction of FC is represented by the reaction:

$$H_2 + 1/2O_2 \rightarrow H_2O + \text{heat} + \text{electrical energy} \qquad (3)$$

III. MODEL FORMULATION

The output voltage of a single cell can be defined as the result of the following expression:

$$V_{FC} = E_{Nernst} - V_{act} - V_{ohmic} - V_{con} \qquad (4)$$

E_{Nernst} is the thermodynamic potential of the cell and it represents its reversible voltage; V_{act} is the voltage drop due to anode and cathode activation, a measure of the voltage drop associated with the electrodes; V_{ohmic} is the ohmic voltage drop, a measure of the ohmic losses associated with the conduction of the protons in the electrolyte and internal electronic resistances; and V_{con} represents the voltage drop resulting of the mass concentration of oxygen and hydrogen [8]. E_{Nernst} represents the FC open circuit voltage and the other terms represent reductions in this voltage to supply the cell useful voltage, V_{FC}, for a given operation current. The terms of (4) are discussed and modeled bellow.

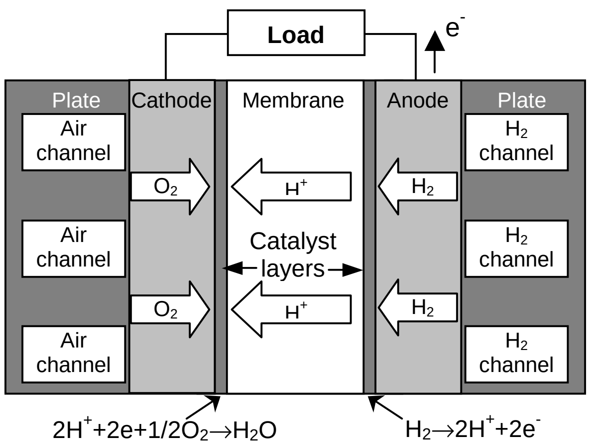

Fig. 1. Basic PEMFC operation

3.1. Cell reversible voltage

The cell reversible voltage (E_{Nernst}) is the cell potential obtained in the open circuit thermodynamic balance (without load). In this model, E_{Nernst} is calculated starting from a modified version of the Nernst equation, with an extra term to take into account changes in the temperature with respect to the standard reference temperature, 25°C [1]. This is given by:

$$E_{Nernst} = 1.229 - 0.85.10^{-3}(T - 298.15) + 4.31.10^{-5}T\left[\ln\left(p^{*}_{H_2}\right) + \frac{1}{2}\ln\left(p^{*}_{O_2}\right)\right], \qquad (5)$$

where $p^{*}_{H_2}$ and $p^{*}_{O_2}$ are the partial pressures (atm) of the hydrogen and of the oxygen, respectively. Variable T denotes the cell operation temperature (K) and T_{ref} is the reference temperature.

3.2. Activation overpotential

As shown in [1], the activation overpotential, including anode and cathode, can be calculated by

$$V_{act} = -[\xi_1 + \xi_2.T + \xi_3.T.\ln\left(c^{*}_{O_2}\right) + \xi_4.T.\ln(i)] \qquad (6)$$

where i is the cell operating current (A), $c^{*}_{O_2}$ is the concentration of oxygen in the cathode catalytic interface (mol/cm^3) and the ξ's represent parametric coefficients, based in kinetic, thermodynamical and electrochemical laws.

3.3. Ohmic overpotential

The ohmic overpotential results from the electrons transfer resistance in the collecting plates and carbon electrodes and the proton transfer resistance in the solid membrane. The membrane equivalent resistance is calculated by:

$$R_M = \frac{\rho_M l}{A} \qquad (7)$$

where ρ_M is the membrane specific resistivity (Ω.cm), A is the cell active area (cm^2) and l is membrane thickness (cm).

The following numeric expression for the membrane resistivity is used:

$$\rho_M = \frac{181.6\left[1 + 0.03\left(\frac{i}{A}\right) + 0.062\left(\frac{T}{303}\right)^2\left(\frac{i}{A}\right)^{2.5}\right]}{\left[\lambda - 0.634 - 3\left(\frac{i}{A}\right)\right].\exp\left[4.18\left(\frac{T-303}{T}\right)\right]} \qquad (8)$$

where: 181,6/(λ-0,634) is the specific resistivity (Ω.cm). The parameter λ is an adjustable parameter influenced by the membrane preparation procedure and it is a function of the relative humidity and stoichiometry of the anode gas. It can have a value in the order of 14 under 100% of relative humidity and 22 or 23 under oversaturated conditions.

Using the value of (7) for the membrane resistance, the following expression determine the ohmic overpotential:

$$V_{ohmic} = i.R_M \qquad (9)$$

3.4 Concentration overpotential or mass transport

The mass transport affects the hydrogen and oxygen concentrations. This, for its turn, causes a decrease of the partial pressures of these gases. Reduction in these pressures depends on the electrical current and physical system characteristics.

To determine an equation for the calculation of this voltage drop, it is defined a maximum current density, J_{max}, under which the fuel is being used at the same rate of the maximum supply speed. The current density cannot surpass this limit because the fuel cannot be supplied to at a larger rate. Typical values for J_{max} are in the range from 1000 to 1500 mA/cm^2. Thus, the voltage drop due to the mass transport can be calculated by:

$$V_{con} = -B.\ln\left(1 - \frac{J}{J_{\max}}\right), \tag{10}$$

where B (V) is a constant depending on the cell and its operation state and J is the actual cell current density (A/cm^2).

3.5 Dynamics of the cell

In fuel cells there exists a phenomenon known as "charge double layer". This phenomenon is of extreme importance for the cell dynamics understanding: whenever two differently charged materials are in contact there is a charge accumulation on the surfaces or a load transfer from one to other. The charge layer on the interface electrode/electrolyte (or close to the interface) is a storage of electrical charges and energy, and, in this way, it behaves as an electrical capacitor. If the current changes, it will take some time for the load (and the associated voltage) to vanish (if the current decreases) or to increase (if the current increases). Such a delay affects the activation and concentration potentials. It is important to point out that the ohmic overpotential is not affected, since it is related in a linear form to the cell current through the Ohm's Law. Thus, any current change causes an immediate change in the ohmic voltage drop. In this way, it can be considered that a delay of first order exists in the activation and concentration voltages. The time constant, τ (s), associated with this delay is the product:

$$\tau = C.R_a, \tag{11}$$

where C represents the equivalent capacitance (F) of the system and R_a the equivalent resistance (Ω). The value of the capacitance is of some few Farads. The resistance R_a is determined from the steady-state values of the cell current and of the activation and concentration voltages and current, through the equation:

$$R_a = \frac{V_{act} + V_{con}}{i} \tag{12}$$

3.6 Power generation

A PEMFC electrical energy generation system may be represented according to Fig. 2 which shows the stack with the feeding of hydrogen, oxygen (air) and water for refrigeration, as well as its output products, hot water and electricity. The overall stack output voltage is represented by V_s. The reformer is also represented, to obtain hydrogen for the cells starting from a fuel with hydrocarbon. Number of system components will depend, mainly, of the total stack power.

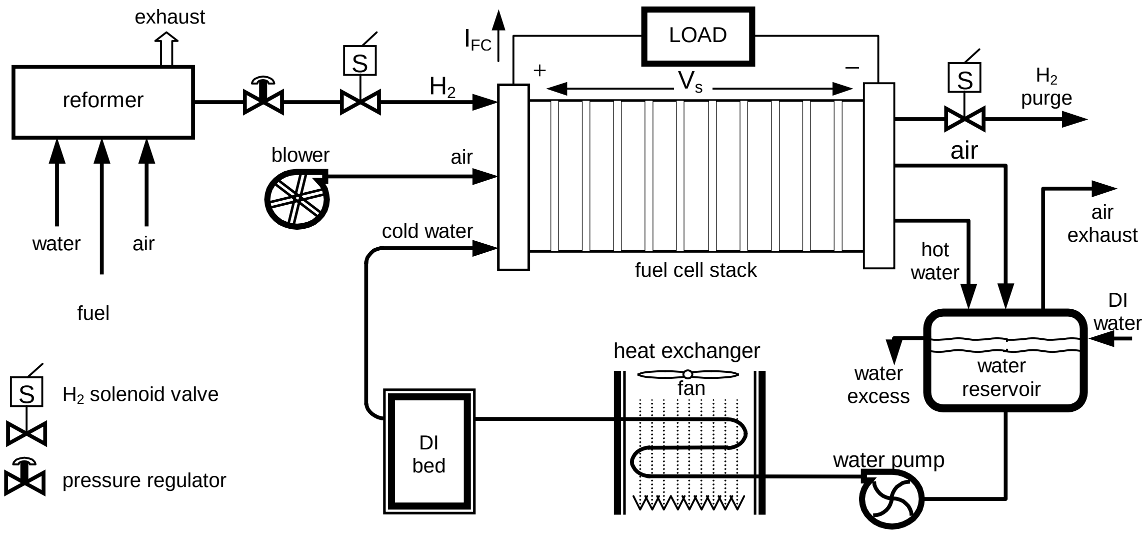

Fig. 2. Generation system with PEMFC

The stack electrical output energy is linked to a load, represented by a block in the diagram of Fig. 2. There is no restriction with respect to the load type, since the power supplied by the stack is enough to feed it. For example, in systems of energy injection into the mains grid, the load can be a combination of a boost DC/DC converter, an AC/AC converter and an isolation transformer. In isolated systems it can be a pure resistive load (heating) or a resistive-inductive load (motor). In any case, the cell current density J (A/cm^2) is defined by the following expression:

$$J = \frac{i}{A} \tag{13}$$

The instantaneous electrical power P_S (W) supplied by the stack to the load can be determined by the equation:

$$P_S = V_S.i, \tag{14}$$

The FC efficiency can be determined from the equation:

$$\eta = \mu_f \frac{V_{FC}}{1.48}, \tag{15}$$

where μ_f is the fuel utilization coefficient, generally in the range of 95%, and 1.48 V represents the maximum voltage that can be obtained using the largest value of the cell enthalpy.

3.7 Fuel cells fluids

In the FC stack operation, some additional factors must be taken into account, as: hydrogen and air utilization rates, generated heat and water. The oxygen utilization rate, kg/s, can be expressed by:

$$\dot{O}_2 = 8{,}29.10^{-8}\,\frac{P_S}{V_{FC}} \tag{16}$$

Equation (16) allows the calculation of the oxygen utilization for any FC system. In normal situation, however, the oxygen will be obtained from air. So, (16) must be adequated to calculate the air utilization rate. The oxygen molar proportion in the air is 0.21 and the air molecular mass is $28.97*10^{-3}$ kg/mol. Besides that, the air supplied to the stack will not be the amount just needed to the chemical reactions. That is, the air will be supplied in a stoichiometry ratio bigger than the unit. Calling the stoichiometry ratio as κ, the equation to calculate the air utilization rate, in kg/s, is:

$$\dot{air} = 3{,}57.10^{-7}.\kappa.\frac{P_S}{V_{FC}} \tag{17}$$

For FC´s feeding by pure hydrogen, the hydrogen utilization rate, in kg/s, can be calculated by:

$$\dot{H}_2 = 1{,}05.10^{-8}\,\frac{P_S}{V_{FC}} \tag{18}$$

When another hydrocarbon fuel is to be used, the calculus will depend on the amount of carbon monoxide present in the fuel. Also, in hydrogen FC, the water production rate, in kg/s, can be determined by the equation:

$$H_2\dot{O} = 9{,}34.10^{-8}\,\frac{P_S}{V_{FC}} \tag{19}$$

Another important aspect of FC operation is the heat generated. The difference between the actual cell voltage and the theoretical value (1.48 V) represents the energy that will be converted in heat. This value, in Watts, is calculated by:

$$Heat = P_S\left(\frac{1{,}48}{V_{FC}} - 1\right) \tag{20}$$

IV. SIMULATION RESULTS

4.1. Model validation

For model validation, one single cell Ballard Mark V was simulated which was fed with gases H_2 and O_2, using the membrane Dupont Nafion 117. The parameters used for this simulation are presented in the Table I.

It should be made a proviso with relationship to the cell and model parameters. The main proposal of the model is to supply the system operating characteristics in a possible more precise way, allowing the development and improvement of electrical energy generating systems using this new and highly promising technology. However, there is a series of parameters involved in this model, empiric some of them and others of difficult determination. For the simulations that proceed, the parameters were obtained in the literature and the results agree fully with the ones presented by several authors in the literature. With the technology development, more precise parameters and of easier determination can be obtained, due to the great and growing worldwide interest in the subject. For the current situation, a model that can be used as a block in the construction of simulators of generation systems using PEMFC, with reasonable results and good dynamic response, comes as a quite interesting and up-to-date option to benefit planners and researchers of the subject.

Table I – Simulation parameters (FC Ballard Mark V)

Par.	**Value**	**Par.**	**Value**
T	343,15 K	ξ_1	-0,948
A	50,6 cm^2	ξ_2	$0{,}00286+0{,}0002.\ln A+(4{,}3.10^{-5}).\ln c^*_{H_2}$
l	178 μm	ξ_3	$7{,}6.10^{-5}$
$c^*_{O_2}$	1.10^{-4} mol/cm^3	ξ_4	$-1{,}93.10^{-4}$
$c^*_{H_2}$	1.10^{-4} mol/cm^3	λ	23,0
$p^*_{O_2}$	1 atm	B	0,016 V
$p^*_{H_2}$	1 atm	J_{max}	1500 mA/cm^2

The cell polarization curve represents the FC output voltage as a function of the current density in steady state. The result obtained with the simulation is presented in Fig. 3, which also shows the practical results presented in [1] and [10]. The simulation results show great likeness with the experimental ones. For normal operation current density, with values above 0.1 A/cm^2 and below 1.3 A/cm^2, the error was less than 2%. This figure is completely acceptable taking into account the difficulty to find the right values for the different model parameters.

The resulting PEMFC efficiency and power density are represented in Fig. 4. Of the presented data, it can be observed that the cell voltage and efficiency present higher values for low current densities and power. For higher values of power, the efficiency and the voltage present smaller values. Therefore, it should match the best cell operation point, taking into account an efficiency and a voltage adequate to feed the load. That is to say, one cannot work with a very high voltage (and, consequently, high efficiency) because the possible output power would be very reduced, meaning that the cell should be over estimated for the application. It cannot also operate with a very high output current, because, in this case, the cell output voltage and efficiency would be very reduced, besides decreasing the useful life of the cell. A compromise should be established among the demand of the load and the power supplied by the cell. The control algorithm should decide when the FC assumes the demanded load, even when this is too high, or when it should just supply part of the load power, for not damaging temporary or permanently the cell. Using this model, the action of an FC can be analyzed for certain practical conditions of load and, therefore, to develop the generation control algorithm.

From data presented in Fig. 4, it can be noticed that, for a certain value of current density, there will be a maximum value of power density. This characteristics, also common to other sources of energy (for example, asynchronous generation and photo-voltaic cells) allows the use of control algorithms such as the Hill Climbing Control - HCC, in that it searches, always, the point of maximum generated power of a certain source of energy [9].

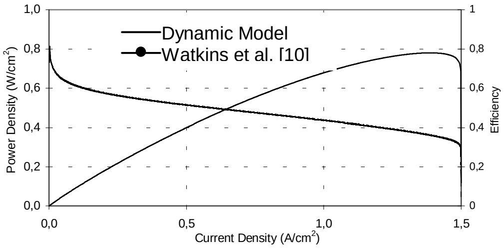

Fig. 3. PEMFC polarization curve

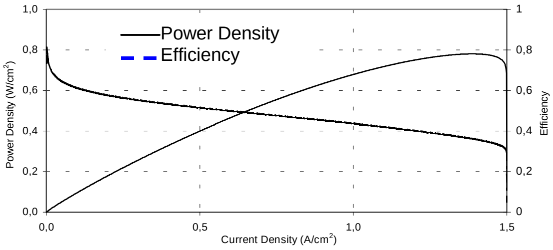

Fig. 4. PEMFC efficiency and power density

4.2. Total load insertion and rejection test

Results from the model for one single cell can be extrapolated for an association in series of FC´s, resulting in an output voltage, V_s, that is the sum of the individual cell voltages. In the same way, it is possible to obtain the total stack output characteristics against the load current.

To evaluate the system response to load changes, it was simulated some extreme conditions commonly found in generating systems: total load insertion and rejection. For the simulations that proceed, the tests correspond to the use of a PEMFC stack Ballard Mark V, consisting of an association of 35 cells, with an active area of each cell of 232 cm^2, with a power of 5 kW @ 960 mA/cm^2. The capacitor used to evaluate the dynamic response is of 3 Farads. The parameter B, used for this stack model, is of 0.05V. The other parameters are the same ones as described in the Table I. In agreement with the model, described in section 3, the partial pressures of hydrogen and oxygen influence the resulting stack voltage. In the simulations that proceed, air was used as the oxidizer. This way, the partial pressure of oxygen becomes 0.2095 atm [8].

For the total load insertion and rejection tests, the stack is considered initially without load, which means, no current. In the instant of time of 3 seconds starting from the beginning of the simulation, the load current is increased to 200 A. In the time instant of 6 seconds, the load is rejected, and the current goes to zero, as showed in Fig. 5. This load current characteristic represents an extreme situation in PEMFC systems operation, and it is used to evaluate the dynamic response of the generation system as showed in Fig. 5

Fig. 6 presents the response of the resulting voltage as a function of the simulation time. The initial voltage is 39,64 V, and corresponds to the no load value. It can be noticed that, at the instant of load insertion (time = 3 s), there is an instantaneous voltage variation and, after an attenuated response, it reaches its final value. The instantaneous variation is due to the ohmic potential, that initially is zero (because there is no current) and, after the load insertion, is 3.27 V. The attenuated voltage response is due to the activation and concentration potentials, as detailed in section 3. The voltage steady state value for a load current of 200 A is 23.68 V. When the load is rejected, at instant 6 seconds, it is noticeable the instantaneous voltage change followed by an attenuated response, until the no load value of 39.64 V is again reached.

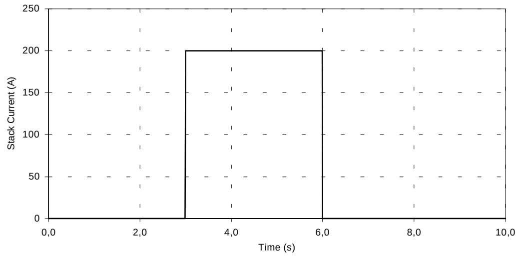

Fig. 5. Stack current

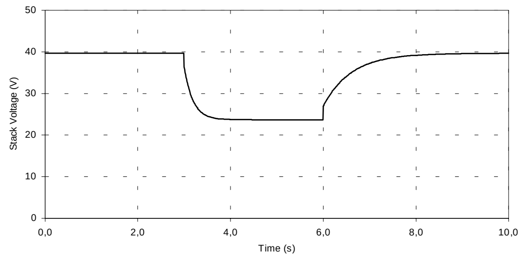

Fig. 6. Stack voltage

Fig. 7 presents the stack power response starting at zero. After the load insertion, there is a transient period in that the power presents values above 5kW, with an instantaneous maximum value of 7.27kW, for a brief time interval. After the transient period, the power supplied by the cell falls to a close value to its rated power, of 4.74kW. In an energy generation system with FC, attention should be given for these power picks, because it will demand a considerable effort of the stack to assist the load demand. If the power pick goes very high, it may be necessary to use another sources of energy in parallel with the FC such as batteries, solar or thermal. This way, the power picks can be absorbed by this other source of energy, not damaging the cells. When the load is rejected, the output power is suddenly reduced to zero. The electrical stack energy delivery in this test is in the order of 4.08Wh.

Fig. 8 shows the resulting efficiency for the total load insertion and rejection tests. The graph shows that, initially, the efficiency is null, since there is no output power. After the load insertion the efficiency presents a maximum value of the order of 66.6%. After, the efficiency begins to drop to its final value, of the order of 43.4%. The values obtained for the efficiency just consider the electrical output power, not considering the use of the water and heat generated. If these are considered, the value for the efficiency will be increased in a proportion that depends on the amount of the generated heat. This evaluation is out of the scope of this work and it will be evaluated in another paper. When the load is rejected, the efficiency falls to zero, since there is no output power.

Fig. 9 shows the water production rate in this test. The total amount of water mass produced is about 1.96×10^{-3}kg. As the water density is 1.0g/cm^3, this amount corresponds to 1.96cm^3. The electrical energy supplied in the test is about

4.08*10^{-3} kWh. So, extrapolating these results, for every 1.0kWh of electrical energy, the stack produces 480.4cm^3 of water. This value is consistent with the values presented in the literature [8], taken into account a variable stack voltage.

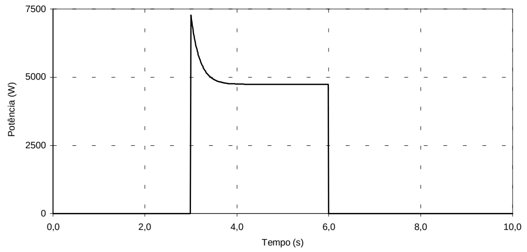

Fig. 7. Stack output power

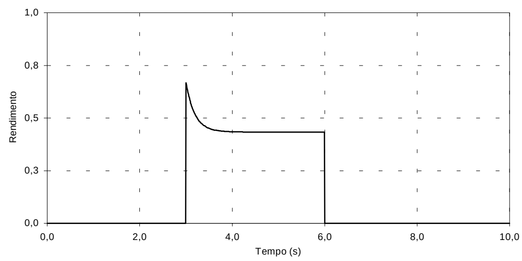

Fig. 8. Stack efficiency

Fig. 10 presents the generated heat in the stack operation. It can be observed that there are a large amount of heat been generated: about 5.63kW after reaching the steady state regime. Comparing this value with the electrical output power (about 4.74kW), it is noticeable that more than 50% of the total power produced in the cells are converted in heat. Actually, this heat must be removed from the stack, using either circulating water or air. If the heat is used in a combined heat and power system, it is better to use water, as this water can be used in the system.

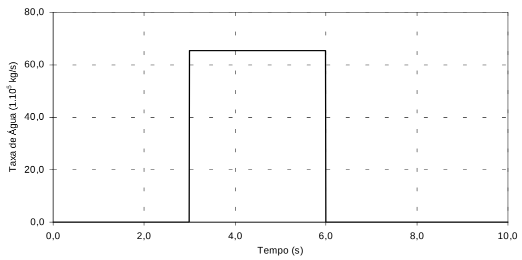

Fig. 9. Water production rate

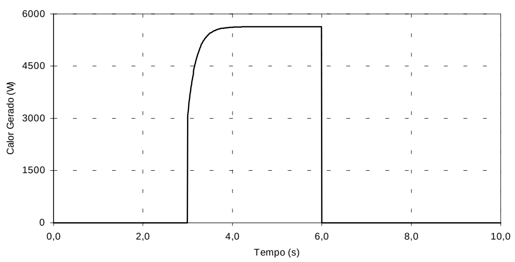

Fig. 10. Heat generated

V. CONCLUSION

In this study, most of the parameters used in PEM fuel cells were taken into account to build a realistic dynamic model for the PEMFC performance analysis in the situations commonly met in low size energy generating system. However, it must be said that a model producing the fuel cells characteristics with high precision degree as a function of the stack load current, is still a challenge. However, the results agree with the results presented in the current literature on fuel cells, validating the model as a block that can be used in the modeling of complete power generation systems. The results obtained with tests of load variations suggest the use of algorithms such as the Hill Climbing Control (HCC) for the control of the power supplied to the load. With the use of this model, these and other algorithms can be debugged and tested in laboratory, without putting the cell integrity and the load in danger besides readily making available all the models and sizes of cells of the which the parameters are available.

The total load insertion and rejection test demonstrated that the resulting voltages present a component that varies instantly with the variation of the current, known as the ohmic overpotential.. There are, still, two components, the activation and the concentration overpotential, that are responsible for the attenuation of the voltage variation as a function of the current variation of the cell. This voltage dynamic variation has significant reflexes on the supplied power, as it could cause power picks, as demonstrated by several authors (Fig. 6). This characteristic should be taken into account during the project time and specification of a FC for energy generation systems. Other sources of energy may have to be used to attenuate the abrupt power oscillations, since these may cause temporary or permanently damage to the cells.

The results have also shown the importance of using as a combined heat and power systems. As the stack efficiency in of the order of 50% or less, there are large amounts of heat been generated.. This is ideal for distribuited generating systems, where hot water and elerctricity are of interest.

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