DESIGN OF AN EXTRA-HUMIDIFICATION SYSTEM FOR PEM FUEL CELL

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Abstract. *Major efforts to reduce greenhouse gas emission have increased the demand for pollution-free energy sources. Fuel cells have attracted great attention in recent years for their high power density, high efficiency and zero emissions. Significant improvements in polymer electrolyte membrane (PEM) fuel cell technology have been achieved over the past decade. However, the performance, stability, reliability and cost for today's fuel cell technology are not enough to replace traditional stationary and mobile power sources. A number of fundamental problems must be overcome to improve those limitations. The conductivity of the membrane is proportional to its water content. The chemical reaction forms water, but when the temperature increases, the reaction air coming into the fuel cell has a drying effect; i.e., the amount of water removed from the fuel cell is higher than the amount of water produced by the chemical reaction. In this research, a humidifier, which transfers humidity and heat from the output air to the input air through a permeable membrane, is considered. The warm and damp air leaving the fuel cell passes over one side of the permeable membrane. Some of the water condenses on one side of the membrane and is evaporated by the drier air going into the cell on the other side. The amount of water and heat transferred from the output air to the input air is proportional to this difference. An analysis was performed to quantify the influence of the extra humidification.*

Keywords: *Polymer electrolyte membrane fuel cells, temperature, humidifier, extra humidification control.*

1. INTRODUCTION

The global warming is caused mainly by burning of fossil fuels (oil, diesel, gasoline, etc.) that emit millions of tons of pollutants. Besides, the certainty that those fossil fuels are non-renewable resources allows more researches in cleaner energy, and particularly for vehicles. In this way, fuel cell (FC) has a special attention because it can be applied in urban transport and improves the actual environmental situation (Silveira et al, 2009).

Fuel cells appear like a promising technology for energy generation. Among several technologies in the present, the PEMFC (proton exchange membrane fuel cell) is the most appropriate for vehicles application, because it combines durability, high power density, high efficiency, good response and it works at relatively low temperatures. Besides that it is easy to turn it on and off and it is able to support present vibration in vehicles (Silveira et al, 2009).

The development of fuel cell technology has grown over the past forty years due to several factors, including new materials in the area and the growing demand for clean sources of efficiency energy (Linardi, 2010).

The concept of a new energy conversion system called the fuel cell begins to interest in the growing population and is no longer an issue confined to the technical-scientific community and industry. This concept is applied of environmental preservation, development of non-polluting vehicles and distributed energy generation with greater efficiency (Linardi, 2010).

The Brazil is a first in using a hybrid system that combines hydrogen fuel cells and batteries. This strategy allows for increased fuel economy and rationalize the generated energy is harnessed for the possibility of supplying the batteries with energy produced by the load cell during periods when the vehicle is stopped (for loading / unloading or traffic lights), and the recovery of braking energy by regeneration (MME, 2009).

The project involves the acquisition, operation and maintenance of up to five fuel cell buses, which will operate in the Metropolitan Corridor São Mateus / Jabaquara (São Paulo) – as part of a four year program. A hydrogen fueling station (producing the hydrogen by electrolyzing water) will be built to fuel the buses.

2. AUTOMOTIVE APPLICATIONS IN PEMFC

Several studies have been undertaken for improving the system for automotive applications. The results indicate that it is necessary extra-humidification at temperatures above 60^{o}C (Dicks and Larminie 2003; Huizing et al., 2008). The internal wetting refers to the addition of water directly into the PEMFC, and the gases are heated outside and moist inside the PEMFC (Vasu et al., 2008). Other method can be applied to keep water reaction in PEMFC within the membrane (Huizing, et. al., 2008). External humidification in the gases are heated and humidified externally to the entrance of PEMFC (Vasu et al., 2008), involving the use of a humidification unit for PEMFC providing humidified gases which usually involves recycling heat and humidity from the gas output of the fuel cell (Huizing et al., 2008).

The parameters that influence the HR (relative humidity) reagent gas in a humidifier depend on the residence time of gas in the humidifier, the humidifier outlet temperature, and pore diameter membrane (Vasu et al., 2008).

To achieve the required humidification, various types of external humidifiers have been developed. The most common humidifiers are the external spray, bubbling gas membrane humidifier, and evaporative cooling system (Kang, Min and Yu, 2010).

In this paper, a project is proposed for a humidifier using continuous steam output of the PEMFC. The proposed humidifier exploits the humidity output of the PEMFC, rendering unnecessary the application of a water tank. The use of water reservoirs can be used for applications in laboratories, but are impractical for mobile applications.

2.1 Bus Operation

The Brazilian Fuel Cell Bus has a capacity for 45 kg of hydrogen in nine tanks, also has three high-performance batteries. The average hydrogen consumption is 15 kg/100km. Powered by hybrid electric traction (hydrogen fuel cell + battery), the vehicle has a range of 300 km, running on hydrogen; and if necessary, it can run over 40 km using only batteries (Oliveira, 2009).

2.2. Interest in the use of a FC in buses for urban transport

A ground transport vehicle that seems particularly interesting for adoption of a fuel cell is the bus for urban transport. It is possible also for another kind of transports and another application.

The main reasons for use in buses are (Santarelli 2002; Silveira et al., 2009):

- The dimensions and structure of the bus allow installation of fuel cells and their auxiliaries, including the hydrogen tanks (if it were stored in gaseous form on the bus). The weight percentage increase would represent a lower problem compared with small cars.
- The ratio between the engine power and the weight of the vehicle is low: a bus of 18 tons with a medium speed of 300 km/h has a power of 230 kW.
- The bus circulates in the center of the urban areas where the air pollution is serious. In these areas, a zero emission vehicle could gain high revenue in terms of social costs connected with air pollution.
- A FC technology is, at present, much more expensive than traditional engines. Buses are bought and managed in fleets of a large number of units, and this allows an investment and maintenance cost reduction.
- The urban transport service is usually managed by a public administration, which is more interested in the acquisition of social benefits and which is less bound by short term revenue of the investment.
- Some characteristics of a fuel cell engine are particularly interesting for a bus: the engine is quiet, vibrations are absent and the electric engine has a smooth operation, increasing the comfort for users.

3. PEMFC MODELING

To size the humidifier is necessary to calculate the inlet temperature of PEMFC (t_4) and the humidifier outlet temperature (T_6). T_5 is the operating temperature of the cell output and t_3 is the temperature of the ambient air that enters the humidifier (approximately 20^{o}C). It is also necessary to identify the relative humidity PEMFC inlet and air stoichiometry (λ).

In this paper, we proposed a humidifier countercurrent arrangement for promoting heat exchange between the gases, allowing heat exchange faster.

3.1 Operating parameters of system components humidification

The operating conditions of PEMFC are adapted to the design of the Brazilian bus, powered by hydrogen.

To simplify the gas flow conditions T_5, T_6 for RH_5, RH_6 is called the upper circuit. And the air flow conditions t_3, t_4 for RH_3, RH_4 is called the lower circuit, Figure 1.

In this paper we applied a control temperature technique for PEMFC based on the maximum operating temperature. The maximum operating temperature is the maximum temperature at which the PEMFC can operate in ideal conditions (RIASCOS and PEREIRA, 2009).

The temperature also influences the voltage of the fuel cell. Thus, the higher the temperature, the greater the voltage produced. Basically, a PEM fuel cell model consists of an electrochemical part and a thermodynamical part. These models are applied to analyze the behavior and the dependence between the variables of the PEMFC (RIASCOS and PEREIRA, 2009).

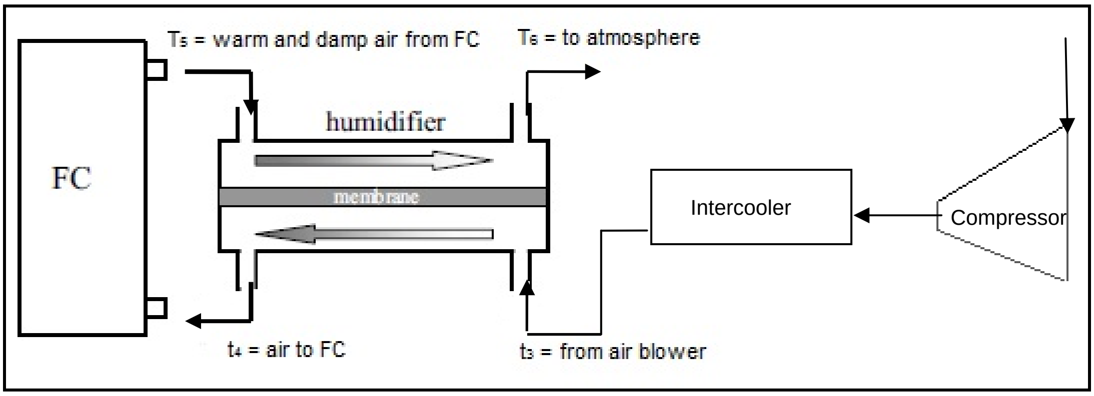

Figure 1. Humidification system using the exit air.

3.2 Electrochemical Model

Although fuel cell technology development requires a complex multidisciplinary effort the basic concept of fuel cell operation is very simple. A fuel cell is an electrochemical device that converts chemical energy, typically from hydrogen, directly into electrical energy. Similar to a battery, a fuel cell consists of two electrodes (anode and cathode) and an electrolyte.

The electrochemical model permits us to calculate the cell voltage. The output voltage V_{FC} of a single cell can be defined as the result of the following expression (10) (DICKS and LARMINIE, 2003; RIASCOS and PEREIRA, 2009):

$$V_{FC} = E_{Nernst} - V_{act} - V_{ohmic} - V_{con} \quad (1)$$

The E_{Nernst} represents the reversible open-circuit voltage and can be defined as the result of the following expression (eq. (2)) ; V_{act} is the voltage drop due to the activation of the anode and cathode (also known as activation over potential); V_{ohmic} is the ohmic voltage drop (also known as ohmic over potential), a measure of the ohmic voltage drop resulting from the resistances of the conduction of protons through the solid electrolyte and electrons through the electric circuit; and V_{con} represents the voltage drop resulting from the reduction in concentration of the reactants gases or, alternatively, from the transport of mass of oxygen and hydrogen (also known as concentration over potential) (Outeiro et al., 2008).

$$E_{Nerst} = 1{,}229 - 0{,}85.10^{-3}(T - 298{,}15) + 4{,}31.10^{-5}T\left[\ln(P_{H_2}) + \tfrac{1}{2}\ln(P_{O_2})\right] \quad (2)$$

Where P_{H_2} and P_{O_2} (atm) are the hydrogen and oxygen pressures, respectively, and T (K) is the operating temperature.

The activation potential is relative to the losses by slow chemical reactions. The energy required to initiate the reaction is called activation energy, and, in some reactions, can delay the process. The activation losses (V_{act}) occur when the cell current density is low (DOE, 2000) and can be defined as the result of the following expression (Riascos and Pereira, 2009):

$$V_{act} = -[\xi_1 + \xi_2 T + \xi_3 T\ln(C_{O_2}) + \xi_4 T\ln(I_{FC})] \quad (3)$$

Where $\xi_1 = i = 1, \ldots, 4$ are specific coefficients for each type of cell; I_{FC} (A) is the electric current, and C_{O_2}(atm) is the oxygen concentration.

$$V_{ohmic} = I_{FC}(R_M + R_C) \quad (4)$$

Where R_C (Ω) is the resistance to electron flow and R_M (Ω) is the resistance to proton transfer through the membrane. R_M e ρ_M can be defined as the result of the following expression respectively (Riascos and Pereira, 2009):

$$R_M = \frac{\rho_M l}{A} \tag{5}$$

$$\rho_M = \frac{181{,}6\left[1+0{,}03\left(\frac{I_{FC}}{A}\right)+0{,}062\left(\frac{T}{303}\right)^2\left(\frac{I_{FC}}{A}\right)^{2{,}5}\right]}{\left[\psi-0{,}634-3\left(\frac{I_{FC}}{A}\right)\right]\exp\left[4{,}18\left(\frac{T-303}{T}\right)\right]} \tag{6}$$

Where ρ_M (cm. Ω) is the specific resistivity of the membrane, l (cm) is the membrane thickness, A (cm^2) is the active area of the membrane, and ψ is a specific coefficient for each type of membrane. V_{con} , can be defined as the result of the following expression (Riascos and Pereira, 2009):

$$V_{con} = -B\ln\left(1-\frac{J}{J_{max}}\right) \tag{7}$$

Where B (V) depends on the type of cell, J_{max} is the maximum electric current density, and J is the electric current density produced by the cell. $J = J_{OUT} + J_n$, where J_{OUT} (A/cm^2) is the output electric current density and J_n (A/cm^2) represents the fuel crossover and internal loss current (Riascos and Pereira, 2009). Figure 2 illustrated the voltage for the simulated conditions.

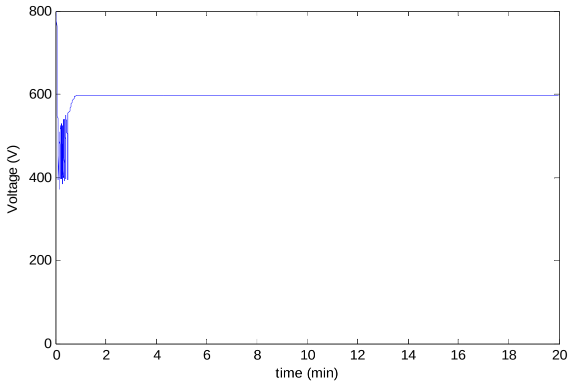

Figure 2. Voltage.

For a stack with n cells connected in series the voltage Vs can be calculated by eq. (8) (Outeiro et al., 2008):

$$V_S = nV_{FC} \tag{8}$$

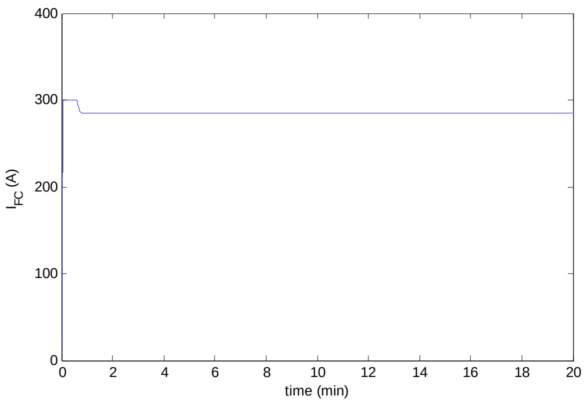

Figure 3. Electric current.

3.3 Thermodynamical Model

The calculations of the relative humidity (RH) and the temperature essentially compose the thermodynamical model (Riascos and Pereira, 2009).

Air humidity should be above 80% to avoid excessive drying and its below 100%, to avoid possible flooding in the electrodes. The controlled flow of air can be used to regulate the humidity of PEM fuel cells, together coupled to a control system to ensure the system temperature fuel cell (Riascos e Pereira, 2009). To calculate the RH of the output air can be calculated by eq. (9):

$$RH_{out} = \frac{P_{w_{in}} + P_{w_{gen}}}{P_{sat\_out}} \tag{9}$$

Where $P_{w_{in}}$ is the water partial pressure in the input air, $P_{w_{in}} = P_{sat\_in} RH_{in}$. $P_{w_{in}}$ is the water partial pressure generated by the chemical reaction; and P_{sat_out} is the saturated vapor pressure in the output air. $P_{w_{gen}}$ is calculated from equation (10):

$$P_{w_{out}} = P_{sat\_out} RH_{out} \tag{10}$$

$P_{w_{gen}}$ is calculated from equation (11):

$$P_{w_{gen}} = \frac{42,1 P_{air}}{\lambda - 0,188} \tag{11}$$

Where P_{air} is the air pressure (atm) and λ is the air stoichiometric relationship. Figure 4 illustrated the evolution of the absolute humidity for the simulated conditions.

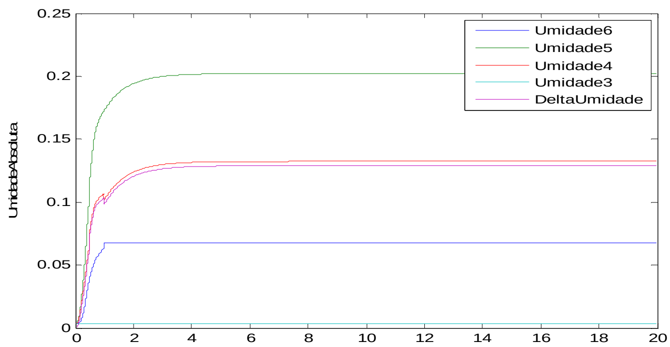

Figure 4. Variables without extra humidification in PEMFC.

The Figures 5 and 6 illustrate the amount of air and the mass flow of water generated in the humidifier for the PEMFC.

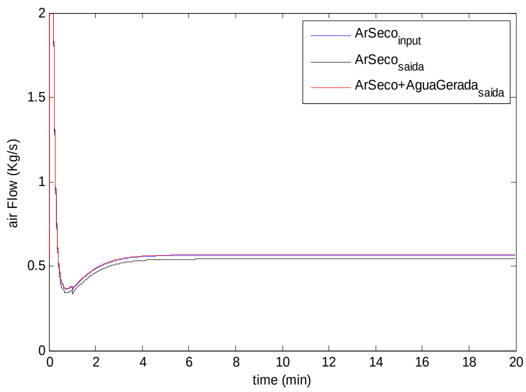

Figure 5. Air flow.

P_{sat} is calculated from equation (22) (Riascos and Pereira, 2009):

$$P_{sat} = -0,01751 + 0,016786 \exp\left(\frac{T}{22,33}\right) \tag{22}$$

The Figure 7 illustrates the variation of relative humidity in PEMFC.

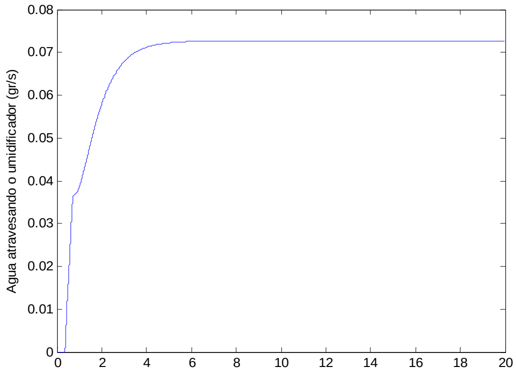

Figure 6. Flux mass through the humidifier.

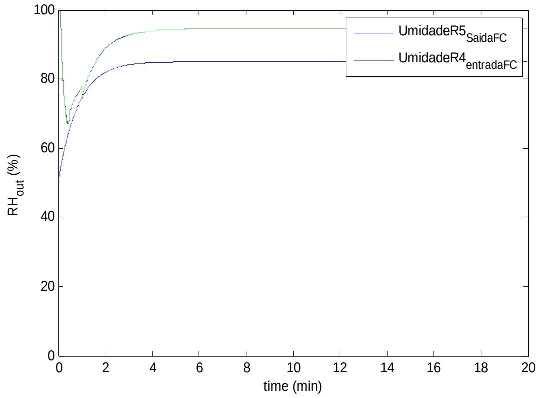

Figure **7.** RH_{in} and RH_{out} in PEMFC.

3.4 Temperature

According to Eqs. (1) (2) and (5), the operating temperature influences the fuel cell voltage; the higher the temperature, the better the voltage (V_{FC}). The variation in temperature in the PEM fuel cell is obtained with the following differential equation (13) (RIASCOS and PEREIRA, 2009):

$$\frac{dT}{dt} = \frac{\Delta Q^{\cdot}}{MC_s} \qquad (13)$$

Where M (kg) is the whole stack mass, *C*s (J/kg K) is the average specific heat capacity of the stack, and $Q^{\cdot}$ is the rate of heat variation (difference between heat generated by chemical reaction and heat removed). The rate of heat generated in a PEM fuel cell is calculated from Eq (14):

$$Q_{ger} = Pow_s\left(\frac{1.48}{V_{FC}} - 1\right) \qquad (14)$$

Where Pow_s is the power produced by the stack.

Three types of heat removed are considered:
Q_{rem_1}: heat removed by the reaction air flowing in the stack.
Q_{rem_2}: heat removed by the refrigeration system.
Q_{rem_3}: heat exchanged with the surroundings.

The heat removed by the reaction air is calculated by Eq. (15).

$$Q_{rem_1} = {}_{ar}C_{ar}\Delta T \tag{15}$$

Where $m_{air} = 3.57.\ 10^{-7}\ \lambda\ Pows\ /V_{FC}$ is the mass of used air (kg/s), C_{air} = 1004 (J/kg K) is the air heating capacity, and T is the difference between the operating and environment temperatures, (Riascos and Pereira, 2009):

To calculate the heat removed by the refrigeration system, Eq. 26 is applied (Riascos and Pereira, 2009):

$$Q_{rem_2} = \eta_{ref} Pow_{ref} \Delta T \tag{16}$$

Where Pow_{ref} is the power of the refrigeration system and η_{ref} is the efficiency of the refrigeration system. To calculate the heat removed by the surroundings, Eq. (17) is applied:

$$Q_{rem_3} = Pow_{amb} \Delta T \tag{17}$$

To calculate the heat removed (Q_{rem_t}) eq. (18) is applied:

$$Q_{rem_t} = Q_{rem_1} + Q_{rem_2} + Q_{rem_3} \tag{18}$$

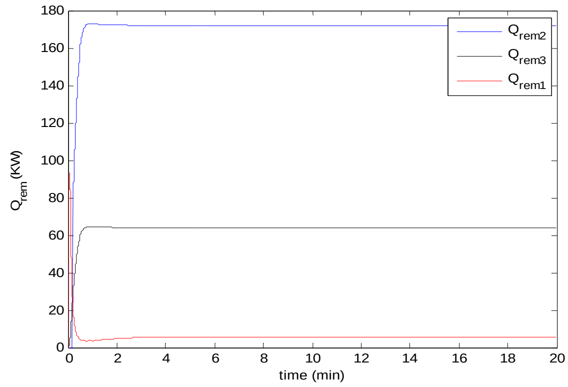

Figure 8. Removed heat.

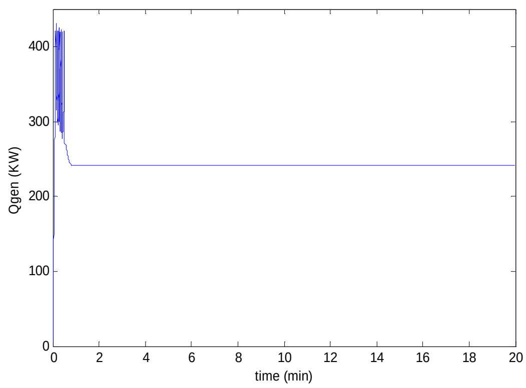

Figure 9. Generated heat.

The difference between heat generated by chemical reaction and heat removed is calculated according to eq. (19):

$$\Delta Q = Q_{ger} - Q_{rem_t} \tag{19}$$

Figure 11 illustrates the evolution of temperature.

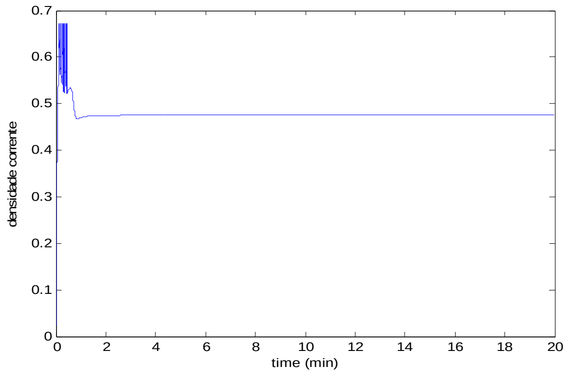

Figure 10. Current density (A/cm^2).

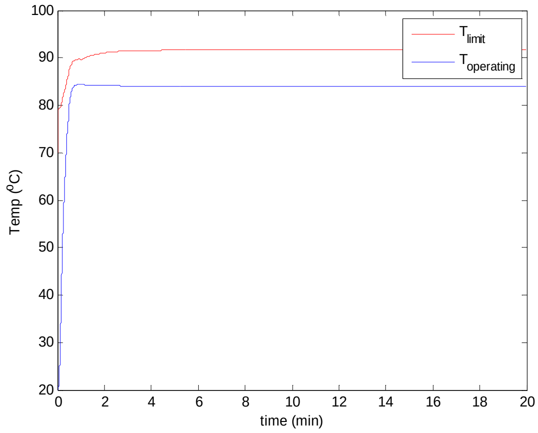

Figure 11. T_{limit} and operating temperature.

3.5 Compressor

When a gas is compressed, it must do work on the gas, generating an increase in temperature. In a reversible process and adiabatic (no heat loss), the increase in pressure and temperature can be calculated by (20) (Dicks and Larminie 2003):

$$\frac{T_2}{T_1} = \left(\frac{P_2}{P_1}\right)^{\frac{\gamma-1}{\gamma}} \tag{20}$$

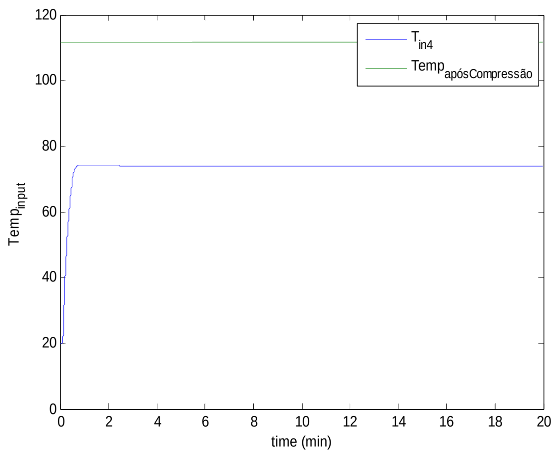

Figure 12. Inlet temperature (t_4) and compression temperature.

Where T_2 is a outlet temperature of compressor; $T_2^{'}$ is an isentropic temperature of compressor; γ is a ratio of the specific heat capacities of the gas, C_P / C_V (aprox. 1.4) and C_p = 1004 (J/kg.K), Figure 12.

3.6 Water Flow from the PEMFC and Humidifier

The amount of water condensing in the humidifier in the upper circuit is the difference between the gas absolute humidity at conditions T_5, RH_5 and the absolute humidity at conditions T_6 and RH_6:

$$P_{agua_5} = RH_{saida} P_{sat} \quad (21)$$

$$RH_5 = \frac{0,622\, P_{agua_5}}{P_{ar} - P_{agua_5}} \quad (22)$$

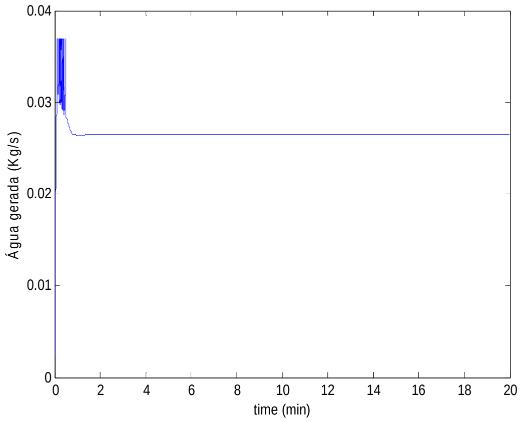

Figure 13. Water flux in PEMFC *versus* time.

To calculate the outlet temperature of the humidifier can be used for energy balance in the humidifier by eq. (23) (24):

$$Q_{vc} = -(m_5 H_5 + m_3 H_3) + (m_6 H_6 + m_4 H_4) \quad (23)$$

$$m_6 H_6 = (m_5 H_5 + m_3 H_3 - Q_{vc} - m_4 H_4) \quad (24)$$

The enthalpy can be calculated by eq. (25):

$$H_i = 1,006 T_i + RH_i (2501 + 1,775 T_i) \quad (25)$$

Where i = 3,4,5,6. The temperature outlet humidifier can be calculated by eq. (26):

$$T_6 = \frac{m_5 H_5 + m_3 H_3 - m_4 H_4 - Q_{vc}}{m_6 (1 + RH_6 1,778)} - RH_6 \frac{2501}{1 + RH_6 1,778} \quad (26)$$

Absolute humidity (AU) from the air entering the PEMFC is the sum of the humidity that goes through the membrane plus the moisture in the air after compression. The absolute humidity of the air after compression should be the same as the environment, since no moisture was removed or added in the compressor.

The temperature t_4 should allow the complete absorption of moisture UA_4, (i.e. $RH_4 < 100\%$). Thus, the humidifier should be designed to allow a heat transfer such that $RH_4 < 100\%$. Then the conditions of gas and air in all the stages are defined.

4. CONCLUSION

Fuel cells are a source of clean energy that helps to reduce emissions of greenhouse gases in the atmosphere, environmental conservation and health and welfare of the population.

According to the analysis presented, attach a humidifier to the fuel cell system increases the system performance, and reduces problems such as dry membrane, facilitates the temperature control.

Through the mathematical model of the PEMFC, it was possible to simulate the evolution of the variables of the PEMFC system for applications in the Brazilian buses powered by hydrogen using Matlab®.

For the simulation it was possible to determine the temperature and relative humidity of the fuel cell, these variables are essential for the design of the humidifier given the objective of this paper.

The results show that it is possible to construct a humidifier with compatible dimensions for automotive applications.

5. REFERENCES

Ballard. “Brazil Bus Project”. 12 December 2010. <http://www.ballard.com/files/pdf/Brazil_Bus_Project_Release.pdf>.

Department of Energy (DOE), 2000. “Fuel Cell Handbook”. Fifth Edition, Morgantown: National Technical Information Service, U.S. Department of Commerce.

Dicks, A. and Larminie, J., 2003. “Fuel Cell Systems Explained”. 2nd ed.: John Wiley & Sons.

EMTU - Ônibus Brasileiro a Hidrogênio. 20 November 2010. <http://www.emtu.sp.gov.br/h2/index.htm>

Huizing, R.; Fowler, M.; Mérida, W.; Dean, J., 2008. “Design methodology for membrane based plate-and frame fuel cell humidifiers”. Journal of Power Sources, Ontario: v. 180, pp. 265-275.

Kang, S.; Min, K. and Yu, S. “Two dimensional dynamic modeling of a shell-and-tube water-to-gas membrane humidifier for proton exchange membrane fuel cell”. International Journal of Hydrogen Energy. Seoul: n. 35, p. 1727–1741, 2010.

Linardi, M., 2010. “Introdução à ciência e tecnologia de células a combustível”, Artliber, São Paulo, Brazil, 350 p.

Matsumoto, E. Y., 2002. “Simulink 5: Fundamentos”, Érica, São Paulo, Brazil, 158 p.

Ministério de Minas e Energia. Projeto ônibus brasileiro a hidrogênio: tecnologias renováveis para o transporte urbano no Brasil. <www.mme.gov.br/programas/onibus_hidrogenio/noticias/noticia1.html>.

Ogden, J. M.; Steinbugler, M. M. and Kreutz, T. G., 1999. “A comparison of hydrogen, methanol and gasoline as fuels for fuel cell vehicles: implications for vehicle design and infrastructure development”. Journal of Power Sources, Princeton: n. 79, pp. 143–168.

Oliveira, M., 2009. “Coletivo a hidrogênio: Ônibus montado no Brasil faz parte de experimentos mundiais para redução da poluição do ar”. Revista Pesquisa Fapesp. São Paulo: ed. 160. <http://www.revistapesquisa.fapesp.br/extras/imprimir.php?id=3882&bid=1>.

Outeiro, M.T.; Chibante, R.; Carvalho, A. S.; Almeida, A.T., 2008. “Review: A parameter optimized model of a Proton Exchange Membrane fuel cell including temperature effects”. Journal of Power Sources, Coimbra: n.185, pp. 952–960.

Programa das Nações Unidas para o Desenvolvimento (PNUD). “Células de Hidrogênio como Combustível para o Transporte Urbano”. 21 January 2010. <http://www.pnud.org.br/projetos/meio_ambiente/visualiza.php?id07=105>.

Riascos, L.A.M., 2008. “Relative Humidity Control”. Journal of Power Sources, Santo André: n.184, pp. 204-211.

Riascos, L.A.M. and Pereira, D. D., 2009. “Limit operating temperature in polymer electrolyte membrane fuel cells”. Journal of The Electrochemical Society, Santo André: n.156, pp. B1051-B1058.

Santarelli MGL, Cali, M and Bertonasco A., 2002. “Different fuelling technologies for urban transport bus service in an Italian big town: economic, environmental and social considerations”. Energy Conversion and Management; v. 44, n.14, pp.353–70.

Silveira, J. L.; Braga, L. B.; Souza, A. C. C.; Antunes, J. S.; Zanzi, R., 2009. “The benefits of ethanol use for hydrogen production in urban transportation”. Renewable and Sustainable Energy Reviews. Guaratinguetá: n.13, pp. 2525–2534.

Vasu, V.; Tangirala, A. K.; Viswanathan, B.; Dhathathreyan, K. S., 2008. “Continuous bubble humidification and control of relative humidity of H_2 for a PEMFC system”. International Journal of Hydrogen Energy, Tamilnadu: n. 33, pp. 4640–4648.

Zhang, J.; Tang, Y.; Song, C.; Cheng, X.; Zhang, Jiujun; Wang, W. 2007. “PEM fuel cells operated at 0% relative humidity in the temperature range of 23–120C”. Electrochimica Acta. Vancouver: n. 52, pp. 5095–5101.

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