CONTROLLING OPERATING TEMPERATURE IN PEM FUEL CELLS

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Abstract. Proton exchange membrane (PEM) fuel cells have attracted great attention in recent years as a promising replacement for traditional power sources. However, a number of fundamental problems must be overcome to improve their performance and to reduce cost. A control system is needed to ensure that all variables remain in secure limits ovoiding breakdowns. In this article, a control technique for PEM fuel cells based on the optimum temperature is proposed. The optimum temperature is the maximum temperature in which the fuel cell can operate preserving a minimum recommended stoichiometry and good relative humidity. Also the influence of operating conditons of air pressure, temperature and humidity of environment are analized.

Keywords: PEM Fuel Cell, Temperature Control, Fuel Cell Model, Operating Temperature

1. INTRODUCTION

Major efforts to reduce greenhouse gas emission have increased the demand for pollution-free energy sources. Fuel cell has attracted great attention in recent years as a promising replacement for traditional stationary and mobile power sources, especially due to their high power density and low greenhouse gas emissions.

Fuel cell is an electrochemical device that generates electricity, similar to batteries, but which can be continuously fueled. Under certain pressure, hydrogen (H_2) is supplied into a porous conductive electrode (the anode). H_2 spreads through the electrode until it reaches the catalytic layer of the anode, where it reacts, separating protons and electrons. The H^+ protons flow through the electrolyte (a solid membrane), and the electrons pass through an external electrical circuit, producing electrical energy. On the other side of the fuel cell, oxygen (O_2) spreads through the cathode and reaches its catalytic layer. On this layer, O_2, H^+ protons, and electrons produce liquid water and residual heat as sub-products (Larminie and Dicks, 2003).

Significant improvements in proton exchange membrane (PEM) fuel cell technology have been achieved over the past decade. However, the performance, stability, reliability, and cost for the present fuel cell technology are not enough to replace internal combustion engines. A number of fundamental problems must be overcome to improve their performance and reduce their cost.

The control, design, and optimum operation of fuel cell require an understanding of the dynamics when there are changes in electrical current, voltage, or load. A control system is needed to ensure that the flow rate and temperature of fuel and air are within prescribed limits during normal operation at variable loads, as well as during system start-up and shut-down.

The fuel cell performance is influenced by the water content in the membrane; the conductivity of the membrane is proportional to its water content. The chemical reaction forms water, but when 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 water produced by the chemical reaction. As a rough approximation, several references suggest that PEM fuel cells working below 60 (^{o}C) do not need extra-humidification on the input reactants and PEM fuel cells working over 60 (^{o}C) need extra-humidification. Besides, most commercial equipment executes the control of temperature over a fixed set point.

The optimal temperature evolves according to the operating conditions. In this research, a control technique that calculates the exact optimal temperature on different operational condition is introduced.

This paper is organized as follows. In section 2, the basic concepts for the mathematical model of a PEM fuel cell are introduced. Section 3 introduces the proposed control technique and presents simulation tests. In section 4, main conclusions are reported.

2. THE FUEL CELL MODEL

Many mathematical models of PEM fuel cell can be found in the literature (Correa et al., 2004; Fouquet et al., 2006; Promislow and Wetton, 2005). Basically, a model of PEMFC consists of an electro-chemical and thermo-dynamical sub-models. Correa et al. (2004) introduce an electro-chemical model of a PEMFC; to validate the model, the polarization curve obtained with this model is compared to the polarization curve of the manufacturing data sheet. In (Riascos et al., 2006; Riascos et al., 2007), the thermo-dynamical part of the model is included to study the effects of different types of faults.

2.1. The electrochemical model

The output voltage V_{FC} of a single cell can be defined as the result of the following expression (Larminie and Dicks, 2003):

$$V_{FC} = E_{Nernst} - V_{act} - V_{ohmic} - V_{con} \tag{1}$$

E_{Nernst} is the open circuit voltage of the cell representing its reversible voltage:

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

where: P_{H2} and P_{O2} (atm) are the hydrogen and oxygen pressures, respectively, and T (K) is the operating temperature.

V_{act} is the voltage drop due to the activation of the anode and the cathode:

$$V_{act} = -\left[\xi_1 + \xi_2 \cdot T + \xi_3 \cdot T \cdot \ln(c_{O_2}) + \xi_4 \cdot T \cdot \ln(I_{FC}) \right] \tag{3}$$

where: ξ_i ($i = 1...4$) are specific coefficients for every type of fuel cell, I_{FC} (A) is the electrical current, and c_{O_2} is the oxygen concentration.

V_{ohmic} is the ohmic voltage drop associated with the conduction of protons through the solid electrolyte, and electrons through the internal electronic resistance:

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

where: R_C (Ω) is the contact resistance to electron flow, and R_M (Ω) is the resistance to proton transfer through the membrane:

$$R_M = \frac{\rho_M \cdot \ell}{A} \tag{5}$$

where: ρ_M ($\Omega \cdot cm$) is the membrane specific resistivity, ℓ (cm) is the membrane thickness, A (cm^2) is the membrane active area, and ψ is a specific coefficient for every type of membrane.

V_{con} represents the voltage drop resulting from the mass transportation effects, which affects the concentration of the reacting gases:

$$V_{con} = -B \cdot \ln\left(1 - \frac{J}{J_{máx}} \right) \tag{6}$$

where: B (V) is a constant depending on the type of FC, J_{max} is the maximum electrical current density, and J is the electrical current density produced by FC. In general, $J=J_{out}+J_n$ where J_{out} is the real electrical output current density, and J_n represents the fuel crossover and internal current loss.

The specific parameters for the fuel cell are presented in Tab. 1.

Considering a stack composed by several fuel cells, the output voltage can be assumed to be $V_S=nr \cdot V_{FC}$, where nr is the number of cells composing the stack. However, constructive characteristics of the stack, such as flow distribution and heat transfer, could influence the output voltage of each cell (Chang et al., 2006; Freunberger et al., 2006; Santis et al., 2006; Wokaun et al., 2006; Kim et al., 2005)

2.2. The thermo-dynamical model

The calculation of the relative humidity and the operating temperature of the fuel cell essentially compose the thermo-dynamical model.

2.2.1. Temperature

The variation of temperature in the fuel cell is obtained with the following differential equation:

$$\frac{dT}{dt} = \frac{\Delta \dot{Q}}{M \cdot C_s} \tag{7}$$

where: M (kg) is the whole stack mass; C_s (J/K·kg) is the average specific heat capacity of the stack; and $\Delta \dot{Q}$ is the rate of heat variation (i.e., the difference between the rate of heat generated by the cell operation and the rate of heat removed). Three types of heat removed are considered: heat by the reaction air flowing in the stack (Q_{rem1}), by the refrigeration system (Q_{rem2}), and by heat exchanged with the surroundings (Q_{rem3}).

TABLE 1.
Parameters of the fuel cell, (Riascos et al., 2008).

Parameter	Value
nr	4
A	62.5 (cm^2)
ℓ	0.0025 (cm)
P_{O_2}	0.2095 (atm)
P_{H_2}	1.47628 (atm)
R_C	0.003 (Ω)
B	0.015 (V)
ξ_1	-0.948
ξ_2	$0.00286 + 0.0002 \cdot \ln A + (4.3x10^{-5}) \cdot \ln c_{H_2}$
ξ_3	$7.22x10^{-5}$
ξ_4	$-1.06153x10^{-4}$
ψ	23
J_n	0.022 (A cm^2)
J_{max}	0.672 (A cm^2)

The rate of heat generated in a fuel cell is calculated from the following equation (Larminie and Dicks, 2003):

$$\dot{Q}_{ger} = Pow_s \cdot \left(\frac{1,48}{V_{FC}} - 1 \right) \tag{8}$$

where: Pow_s is the power produced by the stack
   The heat removed by the reaction air is calculated by Eq. (9).

$$\dot{Q}_{rem_1} = \dot{m}_{air} \cdot C_{air} \cdot \Delta T \tag{9}$$

where: $\dot{m}_{air} = 3.57x10^{-7} \cdot \lambda \cdot Pow_s / V_{FC}$ is the mass of used air (Kg/s); $C_{air} = 1004$ J/Kg·K, is the air heating capacity; ΔT is the difference between the operating and the environment temperature.
   To calculate the heat removed by the refrigeration system, Eq. (10) is applied.

$$\dot{Q}_{rem_2} = \eta_{blower} \cdot Pow_{blower} \cdot \Delta T \tag{10}$$

where: Pow_{blower} is the power of the refrigeration blower; $\eta_{blower} = 0.4$ is the efficiency of the refrigeration blower
   To calculate the heat removed by the surroundings, Eq. (11) is applied.

$$\dot{Q}_{rem_3} = Pow_{surrounding} \cdot \Delta T \tag{11}$$

The operating temperature affects the fuel cell performance. Figure 1 illustrates experimental text to quantify this effect on the polarization curve (Yan et al., 2006).
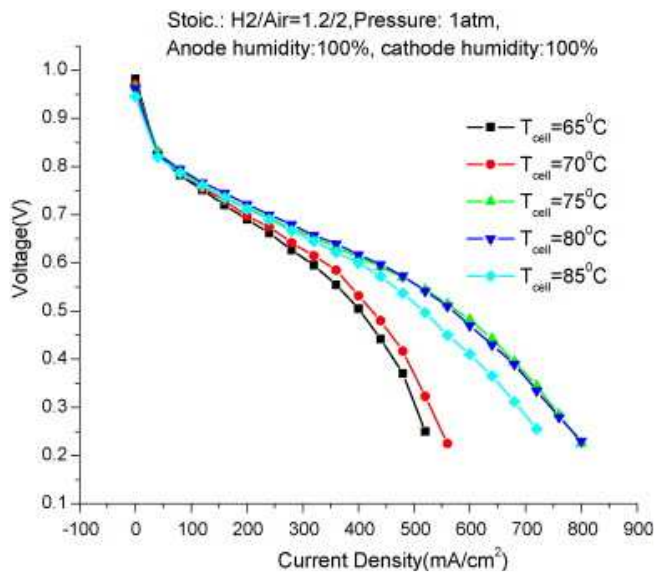
Figure 1. Polarization curve at selected temperature (25 cm^2 fuel cell with triple-serpentine flow pattern, hydrogen stoichiometry = 1.2, air stoichiometry = 2, (Yan et al., 2006).

2.2.2. Relative humidity

A correct humidity level should be maintained in the fuel cell. This level is measured through the relative humidity. If the relative humidity is much smaller than 100 %, then the membrane dries out and the conductivity decreases. On the other hand, a relative humidity greater than 100 % produces accumulation of liquid water on the electrodes, which can become flooded and block the pores, making gas diffusion difficult. The result of these two conditions is a fairly narrow range of normal operating conditions.

In (Kim et al., 2005), the water and thermal management in fuel cell systems were analyzed considering humidification at the cathode and anode. Forms of humidification can include liquid water injection, direct membrane humidification, recycling-humidification and many other methods; in (Chan et al., 2007), the parameters that affect the liquid water flux through the membrane and gas diffusion layer are analyzed.

Figure 2 associates the variation of temperature and relative humidity for different air stoichiometric ratios (λ=2, λ=4 e λ=8). The stoichiometry λ is the relationship between inlet air divided by the air necessary for the chemical reaction.

Figure 3 illustrates the effects on the performance of a fuel cell with variation in the relative humidity. In this figure, the polarization curve with different relative humidity on the cathode side (CRH) is illustrated. According to the figure, the best performance occurs at about 70% (Yan et al., 2006).
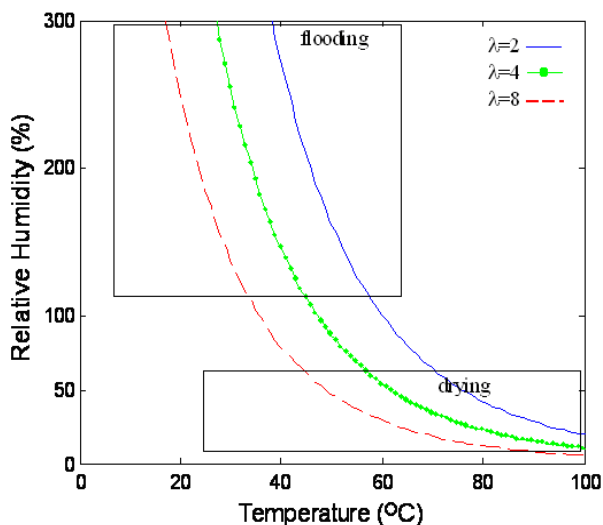
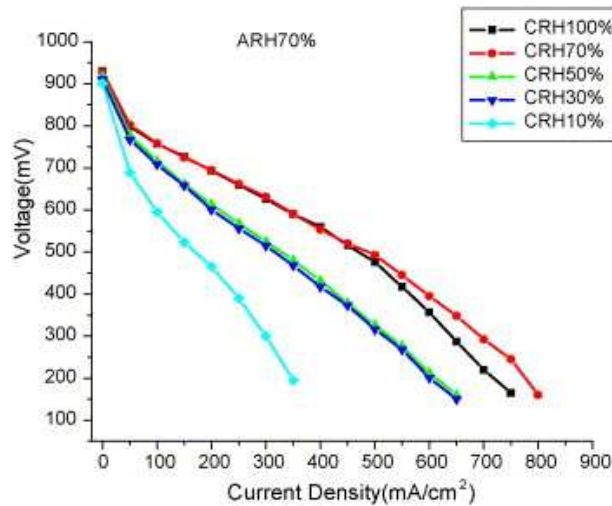Figure 2. Temperature and relative humidity for λ=2,4,8.

Figure 3. Polarization curves as function of feed gas humidity fuel cell (25 cm^2 fuel cell with triple-serpentine flow pattern, hydrogen stoichiometry = 1.2, air stoichiometry = 2, (Yan et al., 2006).

For a good concentration of O$_2$ in the air through the entire fuel cell, λ should be bigger than 4. The rate of air stoichiometric flow influences both the availability of O$_2$ as well as the humidity of the membrane. A low rate limits the availability of O$_2$ because the air is depleted of O$_2$ when it reaches the end of the airflow channels. Also, a very high rate can dry out the membrane.

When the temperature increases, the reaction air has a drying effect and reduces the relative humidity. Low relative humidity can produce a catastrophic effect on the polymer electrolyte membrane, which not only totally relies upon high water content, but also is very thin (and thus prone to rapid drying out).

To calculate the relative humidity of the output air, the balance of water is established:

output = input + internal generation,

or in terms of the water partial pressure: $P_{Wout} = P_{Win} + P_{Wgen}$.

And, also $RH_{out} \cdot P_{sat\_out} = P_{Wout}$, then RH_{out} is

$$RH_{out} = \frac{P_{w_{in}} + P_{w_{gen}}}{P_{sat\_out}} \tag{12}$$

where: P_{Win} is the water partial pressure in the input air; P_{Wgen} is the water partial pressure generated by the chemical reaction; P_{sat_out} is the saturated vapor pressure in the output air.

$P_{Win} = P_{sat\_in} \cdot RH_{in}$

where, RH_{in} is the relative humidity of the input air.

P_{Wgen} is calculated from the next equation, (Larminie and Dicks, 2003):

$$P_{w_{gen}} = \frac{42,1 \cdot P_{air}}{\lambda - 0.188} \tag{13}$$

where: P_{air} is the air pressure (atm) and λ is the air stoichiometric relationship.

The air stoichiometry (λ) to maintain a desired relative humidity is calculated according to Eq. (14), (Riascos, 2008).

$$\lambda = \frac{42.1 \cdot P_{air}}{RH_{des} \cdot P_{sat\_out} - P_{w_{in}}} - 0.188 \tag{14}$$

where: RH_{des} is the desired relative humidity to maintain saturated condition, normally between 80% and 100%, (Chan et al., 2007).

P_{sat} is the saturated vapor pressure:

$$P_{sat} = -0.01751 + 0.016786 \cdot \exp\left(\frac{T}{23.55}\right) \tag{15}$$

3. OPTIMAL TEMPERATURE

The optimal temperature ($T_{optimal}$) is the highest temperature in which the PEMFC can operate preserving a recommended output RH (in this case, RH_{out} on saturated conditions) and a minimum recommended stoichiometry. $T_{optimal}$ is obtained by combining Eq. (14) and Eq. (15).

$$T_{optimal} = 96.25 + 23.55 \cdot \ln\left[\frac{1}{RH_{des}} \cdot \left(\frac{0.421 \cdot P_{air}}{\lambda + 0.188} \right) + P_{w_{in}} + 0.01751 \right] \tag{16}$$

Figure 4 illustrates $T_{optimum}$ as a function of the temperature (T_{in}) and relative humidity of the input air (RH_{in}) simultaneously, (λ=2). From Fig. 4, it is observed that in the best condition ($RH_{in} = 100\%$, $T_{in} = 60\,°C$, $P_{air} = 1$ atm), the operating temperature should be smaller than 76 °C.

The air stoichiometry influences both the availability of oxygen as well as the humidity of the membrane. A low stoichiometry reduces the availability of oxygen because the air is depleted of oxygen when it reaches the end of the airflow channels. The effects on the PEM fuel cell performance with different level of fuel utilization and air stoichiometry utilization were tested in (Yan et al., 2006). In general, the maximum efficiency occurs at about 80% of fuel utilization (H_2) and 25% of air utilization.

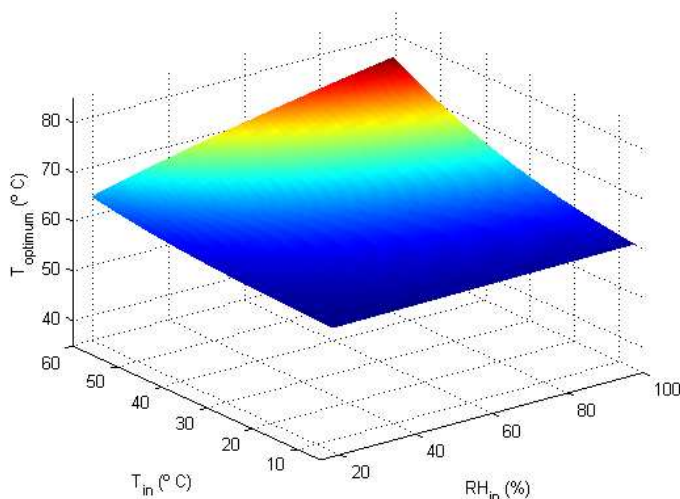Figure 5 illustrates the limit operating temperature as a function of stoichiometry and output relative humidity.

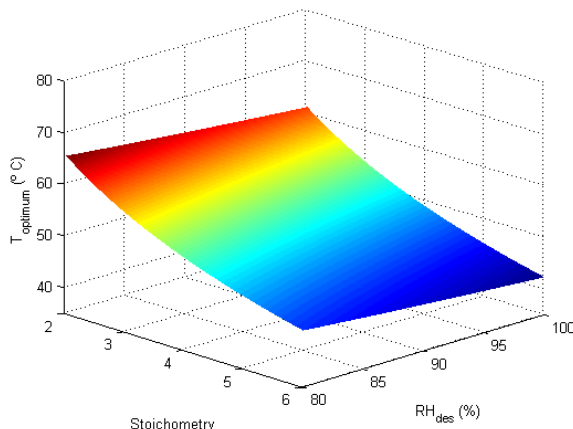Figure 4. $T_{optimum}$ vs. T_{in} and RH_{in} (RH_{out}=100%).

Figure 5. $T_{optimum}$ vs. RH_{out} and stoichometry, (T_{in}=25°C RH_{in}=50%).

Figure 6 illustrates $T_{optimum}$ for different pressures when T_{in} is modified and RH_{des}, RH_{in} and λ are constants (RH_{des} = 100%; $RH_{in} = 50\%$; $\lambda = 2$).
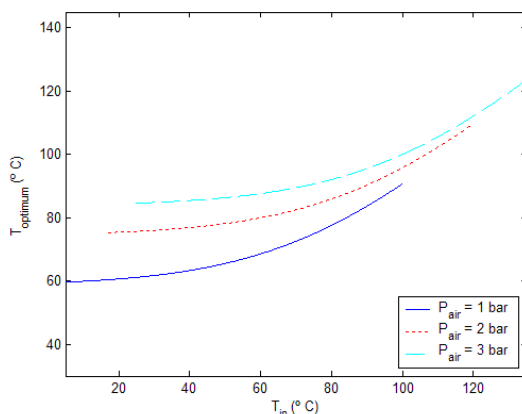
Figure 6. $T_{optimum}$ vs. T_{in}

Figure 7 shows the $T_{optimum}$ for different pressures when RH_{in} is modified and RH_{des}, T_{in} and λ are constants (RH_{des}=100%; T_{in} = 25 ºC ; λ = 2).

The operating conditions of a PEMFC are more sensitive to changes in air temperature than to input relative humidity. Notice that P_w at 60 ºC, RH_{in} = 50%, is more than three times than at 25 ºC and RH_{in} = 100%.
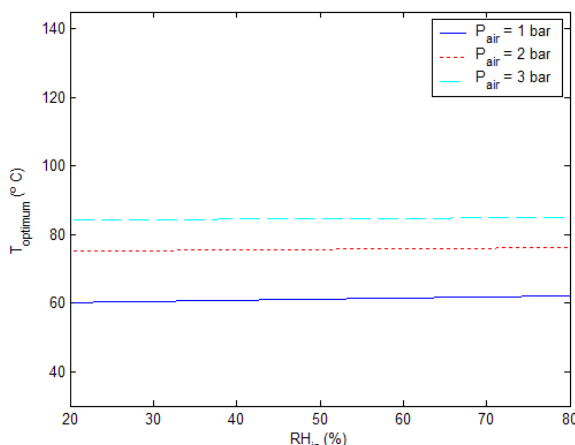

Figure 7. $T_{optimum}$ vs. RH_{in}

Considering Eq. (16), RH_{des} and λ (i.e. a minimum recommended λ) are given conditions; then the limit operating temperature should be modified based on the air input conditions.

In the case of PEM fuel cells without extra-humidification, the air input conditions change according to the environment conditions and, in general, variation of no more than 2 (ºC) per hour can be considered.

On the other hand, in PEM fuel cells with extra-humidification, the variation of temperature and relative humidity of input air can produce significant variation in the PEM fuel cell performance, and then $T_{optimum}$ should be continuously calculated.

In general, PEMFC with extra-humidification works more efficiently, between 20 and 40%, (Yu and Ziegler, 2006). But in some applications (such as portable electronics), the extra size and weight of the humidifier should be avoided.

Figure 8 illustrates the evolution of some PEM fuel cell variables. The variables are: electrical current (I_{FC}), temperature, stoichiometry (λ), and heat removed. Initially, the PEM fuel cell supports a constant-load demand; and the control system adjusts the air-reaction volume to maintain the humidity at the desired value (RH_{des}).

The simulation begins at environment temperature, approx. 25 (ºC). The temperature increases slowly as a consequence of a high inertia of the thermo-dynamical state. In this PEM fuel cell, the refrigeration system can be turned-on when the operating temperature is higher than 40 (ºC), Q_{rem2} represents the heat removed by the refrigeration system.

At t=30 minutes, the thermo-dynamical state is almost stable, then step-variations of load are preformed at t=30 and t=45 minutes to analyze the transient response (reduction in 50% and the return to 100%, respectively).
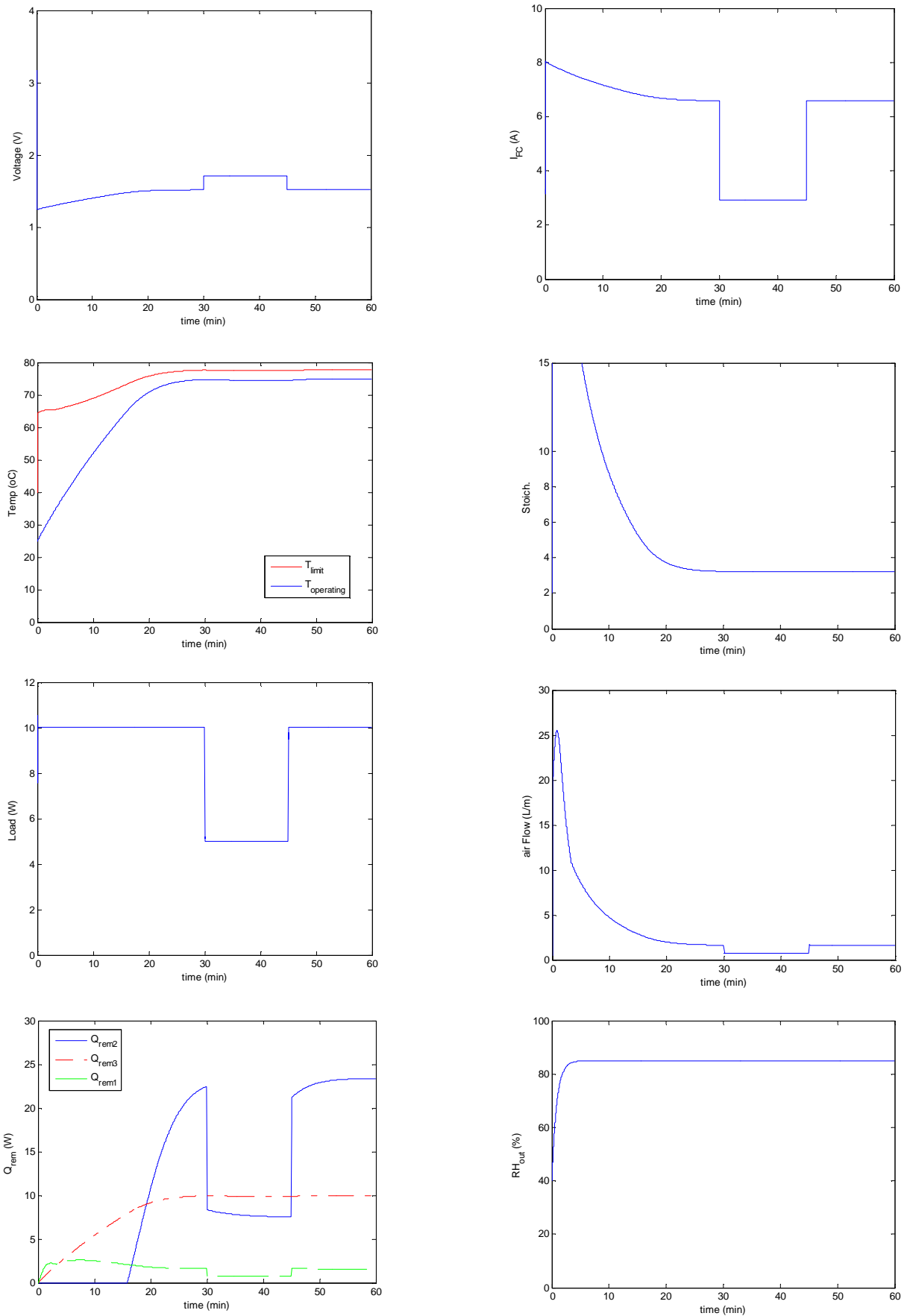
Figure 8. Evolution of the PEM fuel cell variables applying a optimal temperature strategy.

A control technique for the refrigeration system was tested. A PI (proportional-integral) controller was considered. The control signal is based on the difference (the error) between $T_{optimal}$ and the operating temperature. The airflow volume is adjusted by the control system; in this case RH_{out} is kept constant (85%) and stoichiometry is higher than 3.

A control technique for operating with optimal RH_{out} is implemented, (Riascos, 2008). Experimental tests show agreement between analytical results and validation tests.

Also in (Na and Gou, 2008) applied a thermal equivalent circuit for the design of a PEM fuel cell temperature controller. The controller is based on a PI-controller with small gains

4. CONCLUSIONS

The operation of PEM fuel cells require a control system to ensure that humidity and temperature are within the prescribed limits. In this research, a control technique considering the optimal operating temperature is introduced.

A PEM fuel cell model was applied to analyze the evolution and to establish the dependence among the variables. From the mathematical model, the evolutions of some variables that can be difficult to monitor in a real machine are observed (such as stoichiometry, heat removed by refrigeration, by reaction air, etc.). Moreover, tests that can imply permanent damage to the equipment can be avoided (such as tests in very dry or over-heat conditions). In addition, predictions about the evolution of those variables can be tested, optimizing time and resources. The results show that the optimal temperature control strategy is stable and consistent under different operational conditions.

The control strategy implemented in this work considers the desired relative humidity and the minimum air stoichiometry as given conditions, and then the adjustment on the optimal operating temperature basically depends on the input air conditions. In PEM fuel cells without extra-humidification, the limit operating temperature is relatively constant, since the input air conditions are relatively constant. In this case, the limit operating temperature can be considered a constant value and continuous calculation is not necessary.

Conversely, in PEM fuel cells with external humidifier, the limit operating temperature strategy requires a continuous calculation of $T_{optimal}$, since the input air conditions can cause significant variations.

5. ACKNOWLEDGMENT

6. NOMENCLATURE

A	area of membrane (cm^2)
E_{Nernst}	open circuit voltage
I_{FC}	electrical current of the fuel cell (A)
J_{max}	maximum density of current ($A \cdot cm^2$)
J_n	fuel crossover ($A \cdot cm^2$)
ℓ	thickness of membrane (cm)
nr	number of cells
PEM	polymer electrolyte membrane
P_{H_2}	hydrogen pressures (atm.)
P_{O_2}	oxygen pressures (atm.)
P_{sat}	saturated vapor pressure
P_W	partial pressure of water (atm).
Q_{gen}	generated heat (W)
Q_{rem}	removed heat (W)
RH	relative humidity
R_C	electrode resistance (Ω)
R_M	membrane resistance (Ω)
T	temperature
V_{act}	activation voltage drop
V_{con}	concentration voltage drop
V_{FC}	voltage of a single fuel cell
V_{ohmic}	ohmic voltage drop
V_s	stack voltage

Greek letter

λ	air stoichiometric relationship

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8. RESPONSIBILITY NOTICE

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