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Water Activity Control in PEMFC Electrical Energy System

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ABSTRACT:
This work is designed to study the energy management of a Proton Exchange Membrane Fuel Cell (PEMFC), which is based on the activity and management of water and its effect on the electrolyte membrane (FC) as a function of the influx of hydrogen and oxygen, by taking into account the influence of humidification on the implementation of this electrical system to avoid drying and flooding that can cause deterioration of the FC. The cell voltage and system efficiency are also influenced by current density and operating temperature, and simulation results on MATLAB/Simulink are discussed.

KEYWORDS: Water Activity, PEMFC, Drying and Flooding, Diffusion Layer (DL), Catalyst Layer (CL), Modeling and Control, RH.

1. INTRODUCTION

In order for the membrane to remain completely hydrated, the relative humidity of the inlet gases is generally maintained at a high value for good proton conductivity on the one hand and on the other hand, the excess of liquid water at the pores of the catalyst layer and the gas diffusion layer results in greater resistance to mass transport. The major problem of water management to prevent degradation or end-of-life of PEMFC is to maintain a sufficient water balance that is pleasant between the drying of the membrane and flooding.

This article presents a recent study on water management, including modeling and simulation for the characterization of this strategy, by detecting system performance parameters and flood mitigation pathways [1].

Water transport mechanism in electrolyte membrane FC is complex, based on the backscatter phenomenon, which is defined as a function of the moisture gradient between the gases of the cathode and the anode. Backscattering is hard to quantify because it depends on the inlet and outlet humidity and also on the hydration state of the membrane that leads to the electro-osmosis phenomenon which allows the movement of H+ ions in the electrolyte. An approximation of the electro-osmotic flow is possible because protons move in the membrane only in hydrated form of H3O+. The flow of water through the membrane by electro-osmosis is in terms of the produced current and the number of water molecules per H+ ion [2].

Thus phenomena are likely to occur locally, even after cell design simulations, which take into account the movement of electro-osmotic water and the re-diffusion of water from the cathode to the anode in cell configurations and actual operating conditions [2].

2. OPERATING PRINCIPLE OF PEMFC

PEMFC (Proton Exchange Membrane Fuel Cell) are good hydrogen and oxygen converters in electricity, whose reference material is Nafion, these are fuel that can be used at temperatures below 100 ° C with a wide range of power whose choice of operation is due to its simplicity of operation, and its quick start and its solid electrolyte which is sandwiched between two porous carbon electrodes comprising a thin layer of a platinum-based catalyst for the reduction of oxygen and the oxidation of hydrogen and on which the following reactions take place [2] [3].

-Anode: electron emitter whose reaction is Fig.1:

$$2H_2 \rightarrow 4H^+ + 4e^- \quad (1)$$

-Cathode: receiving electron with the reaction is

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad (2)$$

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-Carrier membrane:

$$H_2 + \frac{1}{2}O \rightarrow H_2O + heat + electrical\ energy \qquad (3)$$

To have higher voltages, the multiple cells of the PEMFC are stacked in series. The FC produces 0.7 to 0.8 V [4].

In this paper in order to optimize the performance of a PEMFC and to acquire a long lifespan, it is desired to maintain the moisture content of the membrane constant.

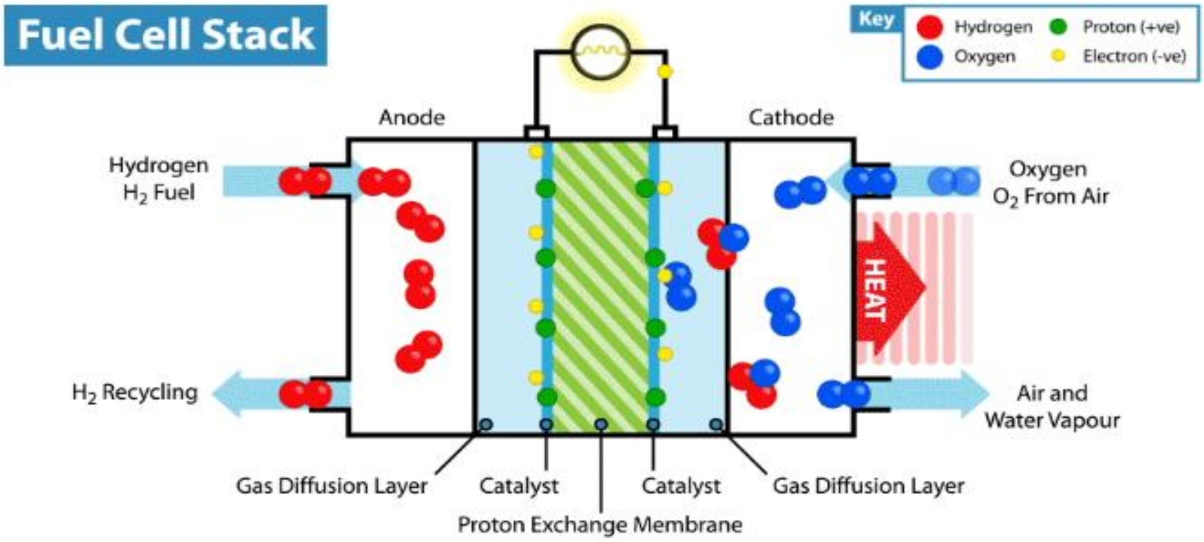

Fig. 1. Principle of Operation of a PEMFC.

A PEMFC consists of Fig. 2 [5]:

1-Fluidic domain:

- Oxygen input of pure air into the cathode.
- Hydrogen input into the anode.

2-Water management domain: water production.

3-Thermodynamic domain: electricity production (the core of the cell).

4-Electrical domain: production of a constant target voltage Vcell to a load.

5-Thermal domain: heat production.

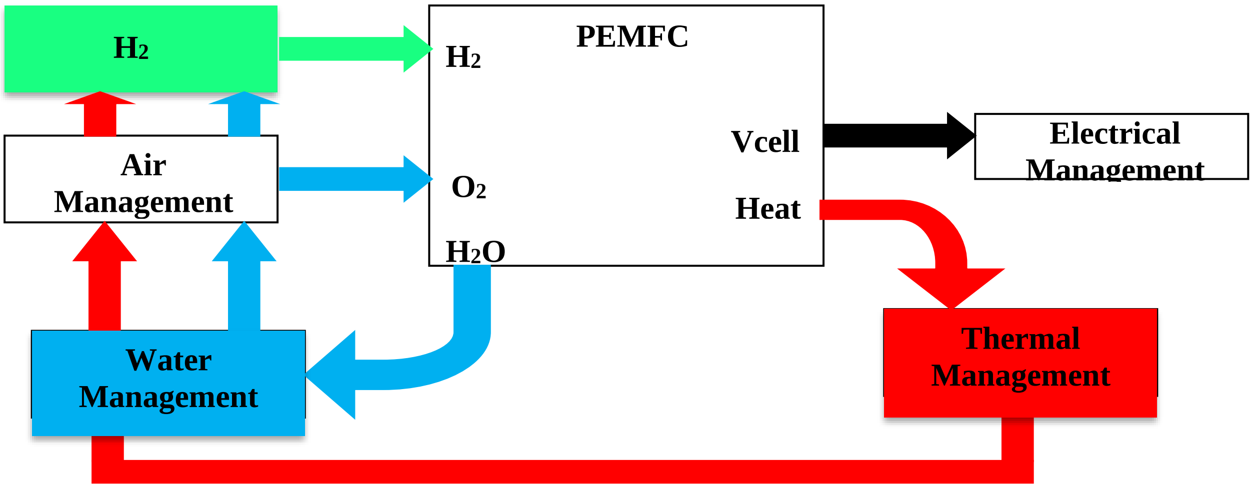

Fig. 2. Operating fuel cell model [6].

3. WATER MANAGEMENT PROBLEMS IN FUEL CELLS

One of the essential problems of each type of FC is its electrolyte. For Alcalin Fuel Cell (AFC), the electrolyte is liquid, which led either to the flooding of the electrodes in case of excess thereof, or their drying in case of leakage; for PEMFC, the electrolyte is an acidic ionic polymer membrane.

At temperatures above 90°C, the membrane is difficult to work, it no longer retains water and the migration of protons does not ensure. Researches are underway to replace Nafion with new polymer materials to increase the performance of PEMFCs at high temperatures of 160-180 °C to prevent anode poisoning by carbon monoxide, to increase the activity of the platinum catalysts of the electrodes and to improve the thermal efficiency of the electrical system [7].

The major problem of the membrane is that it must be sufficiently moistened (it can break in case of drought), which imposes that the air and the fuel are totally humidified to maintain the membrane properly hydrated on the one hand, on the other hand, water is an omnipresent element in the PEMFC, coming from both the humidification of the input gases and the electrochemical reactions of the cell. This is the main parameter of the operation and thus degradation of coupling between all the physical and electrochemical phenomena of the FC (electronic exchange, electrochemical reactions, transport of gas and liquid water in porous media and thermal transfer), the water must be evacuated and the influence of the Relative Humidity (RH) of the membrane is remarkable due to the induced voltage drop and high current densities as shown in Fig. 5.

The geometry of the gas distribution channels also influences the water management and performance of the PEMFC.

These different processes have a significant impact not only on the performance but also on the sustainability of the PEMFC [8] [9] [10].

3.1. Flooding and Drying Case of Membrane

The current research shows that water management in (PEMFC) is very important to expect the best performance and durability of the PEM fuel cell. In favor of this, a several solutions have been proposed to improve the management of the water in the fuel cell. Among them are solutions for modifying the structure of the stack as: Insertion of a porous wick into the electrolyte proposed by Watanabe et al [11]. Insertion of a porous seal in bipolar plates proposed by Gu et al [12] and change of the geometry of the bipolar plates proposed by Vanderborgh et al [13]. However, the most recent solution that is adopted in our work is the humidification of input gases on (PEMFC) systems. However, this solution causes the problems of flooding or drying in the membrane of (PEMFC). Previous investigations approved by Refs [14] and[15] have made it possible to evaluate the interval in which the relative humidity of the input gases (RHin) of (PEMFC) must be to avoid problems of flooding or drying the membrane (RHin), where this interval is between (40–70%). Nevertheless, one of the major chal- lenges to control hydration in the PEM fuel cell is to define exactly when the fuel cell is flooded or dry. To know that the relative humidity (RH) sensor measures only the outlet or inlet water content in the cathode or anode, moreover the voltage curve of the flooded and the dry are approximately superposed as shown in the experimental in Fig. 3. It is impossible to make a diagnosis of that cases (flooded or dry) caused the voltage drop. The challenge of this paper is to make a diagnosis of the healthy fuel cell (flooded or dry) using fuzzy logic inference. It will be applied to the impedance for case classification. For that, the first goal is to design and make a new improved (PEM FC) model for flood and drying diagnosis [16].

4. MODEL TEST AND VALIDATION

The goal of this section is to test and verify the performances of the proposed models. The equations of (PEMFC) models are implemented as shown in Fig. 4 with parameters given in Table 1. The result relating to the voltage/current and relative humidity characteristic is presented in 3D at Fig. 5. This result is according with experimental data of manufactured in Fig. 3.

4.1. Thermodynamic Domain

The voltage of a cell given according to the law of ohm:

$$V_{cell} = E_{nerst} - V_{act} - V_{ohm} - V_{con}$$

Where, ENerst is the Nernst equation, Vact is the activation loss, Vcon is the concentration loss and Vohm is the Ohmic loss.

The overall tension is as follows [17]:

$$V_{cell} = 1,29 - 0,85.10^{-3}.(T - 298,15) + 4,31.10^{-5}.T.\left[ln(P_{H_2}) + \frac{1}{2} ln(P_{O_2}) \right] -$$
$$\left[\xi_1 + \xi_2.T + \xi_3.T.ln(C_{O_2}) + \xi_4.ln(I_{stack}) \right] -$$
$$B.ln\left(1 - \frac{J}{J\max}\right) - I_{stack}.\left(\frac{t_m}{\sigma} + Rc\right)$$

(4)

P_{H_2} : is the partial pressures of hydrogen (atm), P_{O_2} : is the partial pressures of oxygen (atm) and T the fuel cell temperature (°K).

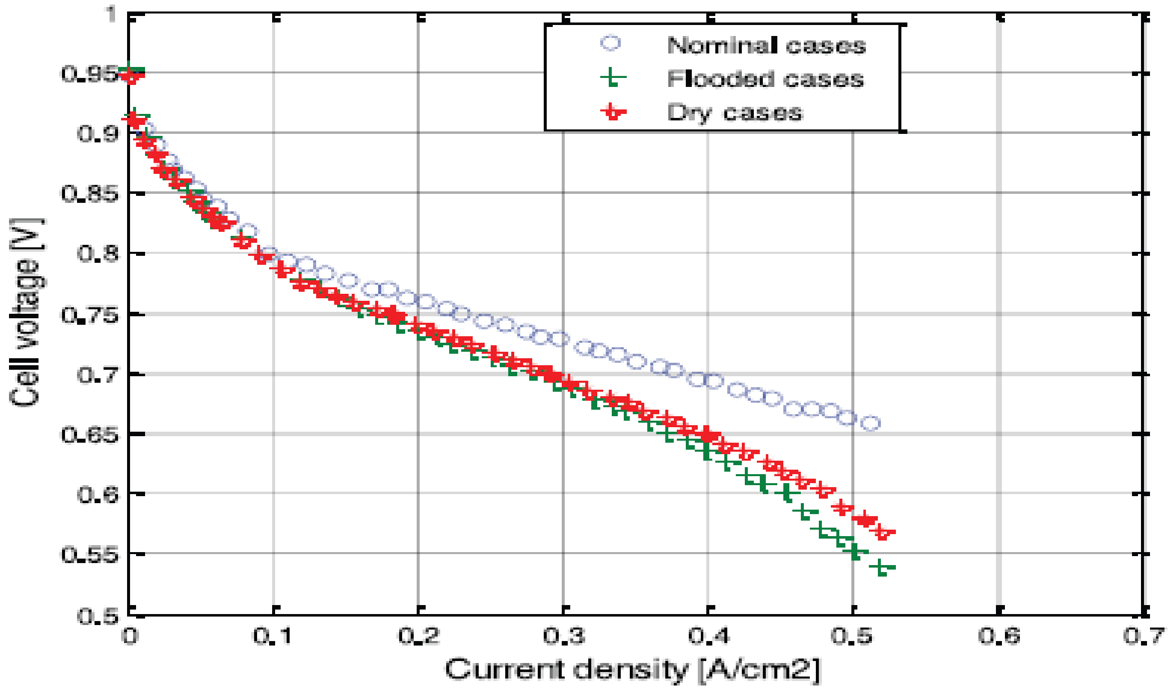

Fig. 3. Experimental polarization curves for a fuel cell working under flooded, dry. In addition, nominal conditions measured by Fouquet and all [14]

$\xi_1 = -0{,}948$,

$\xi_2 = 0{,}00286 + 0{,}0002.ln(\mathrm{A}) + \left(4{,}3.10^{-5}\right).ln(C_{H_2})$

$\xi_3 = 7{,}6.10^{-5}$ and $\xi_4 = -1{,}93.10^{-4}$ are experimentally defined parametric coefficients.

$B = \frac{RT}{nF} = 0{,}016V$: is a parametric coefficient, which depends on the fuel cell and its operation state and J: represents the actual current density of the cell (A/cm2).

J_{max} = 1.5A/cm2 is the current density limit.

$Rc = 0.00003\ \Omega$ is the stack internal resistance and tm is the membrane thickness.

[17]: give the concentration of oxygen Co_2 dissolved in a water film interface in the catalytic surface of the cathode in (mol/cm3):

$$Co_2 = \frac{Po_2}{5{,}08.10^6.e^{(-498/T)}} \tag{5}$$

The electrochemical equations of state developed by M. Y. El-Shark.

$$\frac{d}{dt}(P_{H2}) = \frac{RT}{V_{an}}(q_{H_2in} - q_{H_2out} - q^{r}_{H_2}) \tag{6}$$

$$\frac{d}{dt}(P_{O2}) = \frac{RT}{V_{an}}(q_{O_{2in}} - q_{O_2out} - q^{r}_{O_2}) \tag{7}$$

$$\frac{d}{dt}(P_{H_2O}) = \frac{RT}{V_{an}}(q_{H_2Oin} - q_{H_2Oout} - q^{r}_{H_2O}) \tag{8}$$

Table 1. The Model Parameters [15] [16].

PARAMETERS	VALUES
Faradays Constant, F	9684600 C/Kmol
Universal gas Constant, R	8314.47 j/Kmol.K
Number of cells, N_0	100
Hydrogen valve constant, K_{H2}	4.22*10^{-5} Kmol/S.A
Oxygen valve constant, K_{O2}	2.11*10^{-5} Kmol/ S.A
Stack internal resistance, Rc	0.00003 Ω
Constant, K_r	0.996*10^{-6} Kmol / S.A
Surface, A	50.6 cm^2
Membrane thickness, t_m	178 µm
Current stack, Istack	60 A
Temperature T	333 (°K)
Vanode=Vcathode	S.tm
τ_{O2}	6.74 (s)
τ_{H2}	3.37 (s)

There are three relevant contributions to the molar flow of hydrogen:

The flow injected at the entrance of the PEMFC.

The fuel flow that participates in the chemical reaction.

The fuel flow coming out of the PEMFC.

$$q_{H_2} = q_{H_2\,in} - q_{H_2out} - q^r_{H_2} \qquad (9)$$

It is therefore possible to express the molar flow rate of hydrogen leaving the anode $q_{H_2\,out}$ as a function of the pressure inside electrode P_{H_2} by the relation:

$$q_{H_2\,out} = K_{H_2} P_{H_2} \qquad (10)$$

The relationship (6) becomes:

$$\frac{d}{dt}(P_{H2}) = \frac{RT}{V_{an}}(q_{H2in} - K_{H2}.P_{H2} - 2.K_r.I_{stack}) \qquad (11)$$

This gives:

$$\frac{d}{dt}(P_{H2}) + \frac{R.T.K_{H2}}{V_{an}}P_{H2} = \frac{RT}{V_{an}}(q_{H2in} - 2.K_r.I_{stack}) \qquad (12)$$

$$P_{H2} = \frac{1/K_{H2}}{1+(V_{an}/RT.K_{H2}).S}.(q_{H2in} - 2.K_r.I_{stack}) \qquad (13)$$

Posing: $$\tau_{H2} = \frac{V_{an}}{R.T.K_{H2}} \qquad (14)$$

τ_{H2}: The reaction time constant of the hydrogen system

$$P_{H2} = \frac{1/K_{H2}}{1+\tau_{H2}.S}.(q_{H2in} - 2.K_r.I_{stack}) \qquad (15)$$

A similar operation is done for oxygen, we obtain:

$$P_{O2} = \frac{1/K_{O2}}{1+\tau_{O2}.S}.(q_{O2in} - 2.K_r.I_{stack}) \qquad (16)$$

Also: $$\tau_{O2} = \frac{V_{an}}{R.T.K_{O2}} \qquad (17)$$

P_{H_2inc} , P_{O_2inc} and $p_{H_2O_{inc}}$: The controlled partial pressures of each gas inside the fuel cell (atm) at Fig. 2. V_{an} : is the volume of the anode (l), q_{H_2} and q_{O_2} are the flow rate inlet of each gas and q_{H_2O} is the water inlet flow rate by air (kmol/s).

q_{H_2out} and q_{H_2out} are the outlet flow rates of each gas, and $q_{H_2O_{out}}$: water vapor on (kmol/s); q_{H_2in} and q_{O_2in} : the inlet flow rates of hydrogen and oxygen of the cathode and anode; q^r_{H2}, q^r_{O2} , and q^r_{H2O} : usage, production and reaction of the gases and water.

The input variables are (Ifc, P_{H2}, P_{O_2}, P_{H_2O} , I_{stack} and RH).

The output variables are (q^r_{H2}, q^r_{O2}, q^r_{H2O} , Vcell, Tfc, and Pcell).

The reaction rates of hydrogen, oxygen and water given on (kmol/s):

$$q^r_{H_2} = 2q^r_{O_2} = q^r_{H_2O} = \frac{N_0.I_{stack}}{2.F} = 2.K_r.I_{stack} \qquad (18)$$

N_0 : is the number of cells in the PEMFC stack, F is Faradays Constant = 9684600 C/Kmol.

$$q_{H_2out} = K_{H_2}P_{H_2}; q_{O_2out} = K_{O_2}P_{O_2}; q_{H_2Oout} = K_{H_2O}P_{H_2O} \qquad (19)$$

σ_m: Is the specific conductivity of the membrane proton (Ω.cm) which depends on water activity in the membrane obtained experimentally by the following equation:

$$\sigma_m = (0,00519\lambda\ - 0,00324).\exp\left(1268.(\frac{1}{303} - \frac{1}{T})\right) \qquad (20)$$

The water content of the membrane given by λ:

$$\lambda = \begin{cases} 0,043 + 17,81.RH - 39,85.(RH)^2 + 36.(\mathrm{RH})^3, & 0 \le RH \le 1 \\ 14 + 1,4.(\mathrm{RH} - 1) & 1 \le RH \le 3 \end{cases} \qquad (21)$$

4.2. Relative Humidity Formula

To study the influence of air and water management on humidification problems L. Boulon and others have developed this formula. When the relative humidity is given by the following equation (%) [18]:

$$RH = \frac{P_{H_2O}}{P_{sat}(T)} = \frac{P_{exit}\left(\dfrac{0.420 + \lambda_a\psi}{\lambda(1+\psi) + 0.210}\right)}{10^5 \exp\left(13.7 - \left(\dfrac{5120}{T_{air} + 273.15}\right)\right)} \qquad (22)$$

P_{H_2O} : The partial pressure of water vapor and P_{sat} : The saturated vapor pressure.

ψ : is an adjustable parameter with a possible maximum value of 23.

5. WATER MANAGEMENT AND THESE VARIATIONS

In order to have good energy behaviour of PEMFC, it is necessary to understand the variations of the parameters of output with those of entry for the controlled ones. Fig. 4 shows the variations of the output parameters of the PEMFC cell (Vcell, Pcell, and RH) as a function of the variations of the different entry parameters (Ifc, q_{H_2in} , q_{O_2in} , q_{H_2Oin}) and characterizing the uncontrolled PEMFC model.

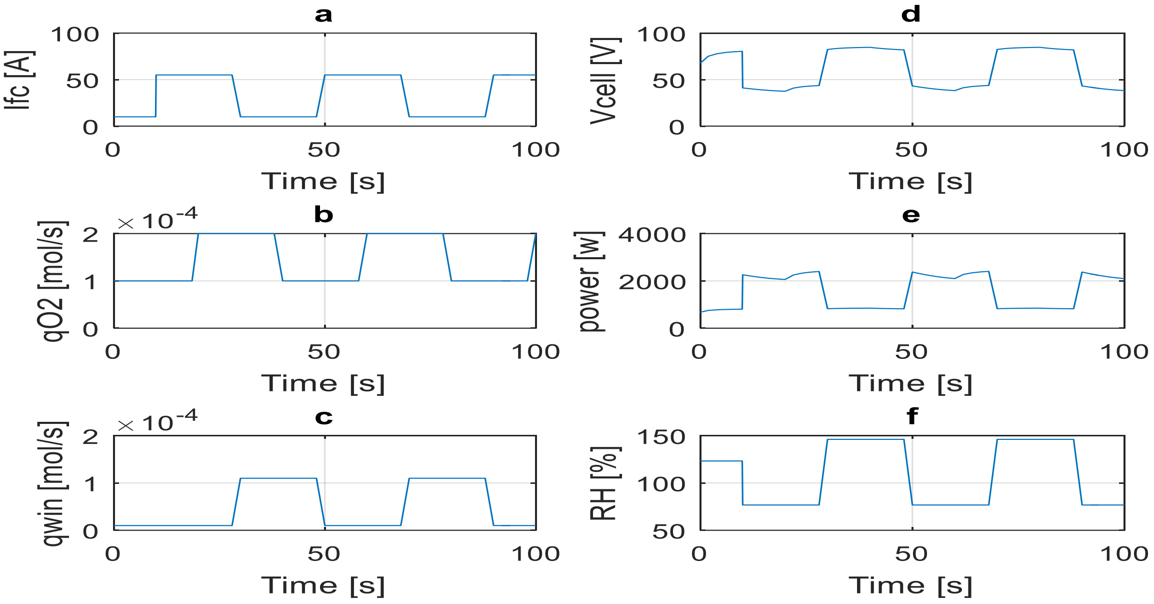

Fig. 4. The dynamic characteristics of a PEMFC (Ifc, q_{O_2in} , q_{H_2Oin} , Vcell, Pcell and RH).

In Fig. 4, The relative humidity exceeded 100% (50%< RH< 150%), for proper functioning a control is envisaged to make this relativity around 100%. [19], [20].

a) Fig.5 estimates the relative humidity at the output of the fuel cell with simultaneous changes in the humid airflow in the fuel cell q_{H_2Oin} and current density at a constant stack temperature of 90°C. The idea is to determine humid airflow at the inlet of fuelcell qwin for staying in the objective zone, in addition, characterizing the flooding or drying of the fuel cell heart (membrane and electrodes).

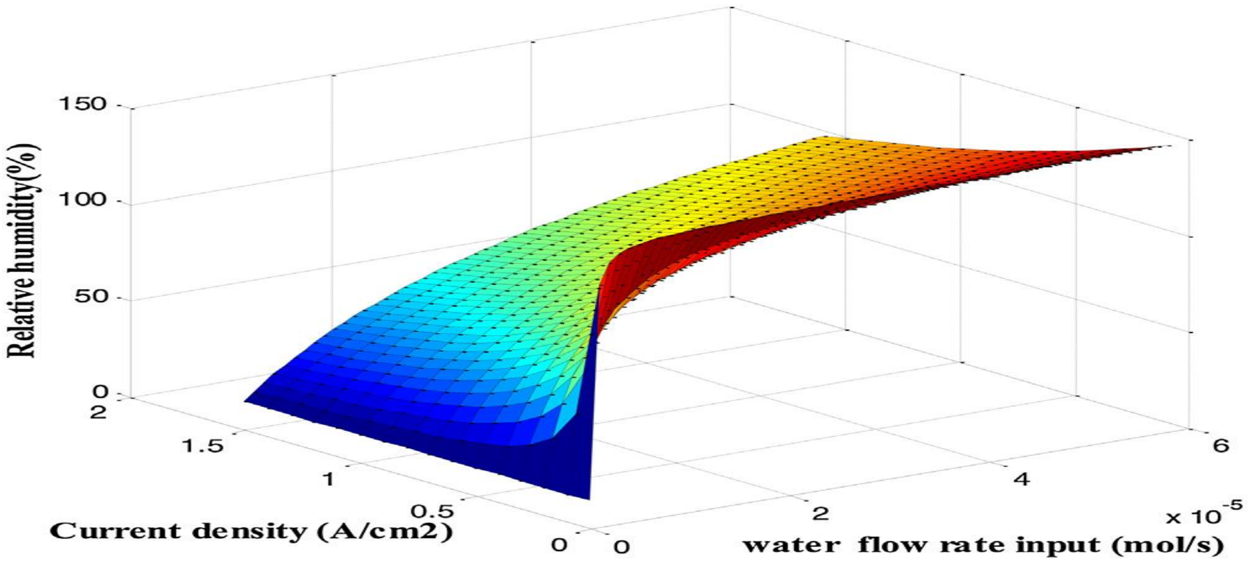

Fig. 5. Variation of the relative humidity as a function of flow of water entering by the air and the density of current.

b) Fig.. 6, shows that when the RH reaches 100% the cell power is at its maximum with increases relative humidity in this value, and power decreases rapidly for high current densities [21].

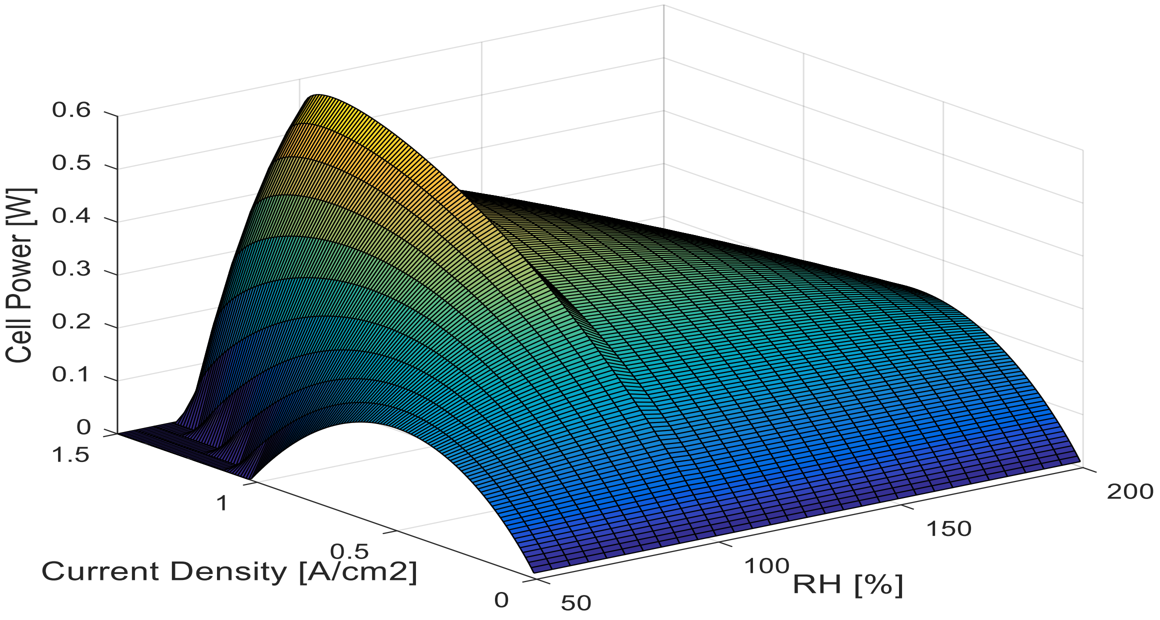

Fig. 6. Variation of Cell Power as a Function of Current Density and Relative Humidity with (PH2=PO2=1(atm) and T=90°C).

c) Three different parts can be distinguished in Fig. 7

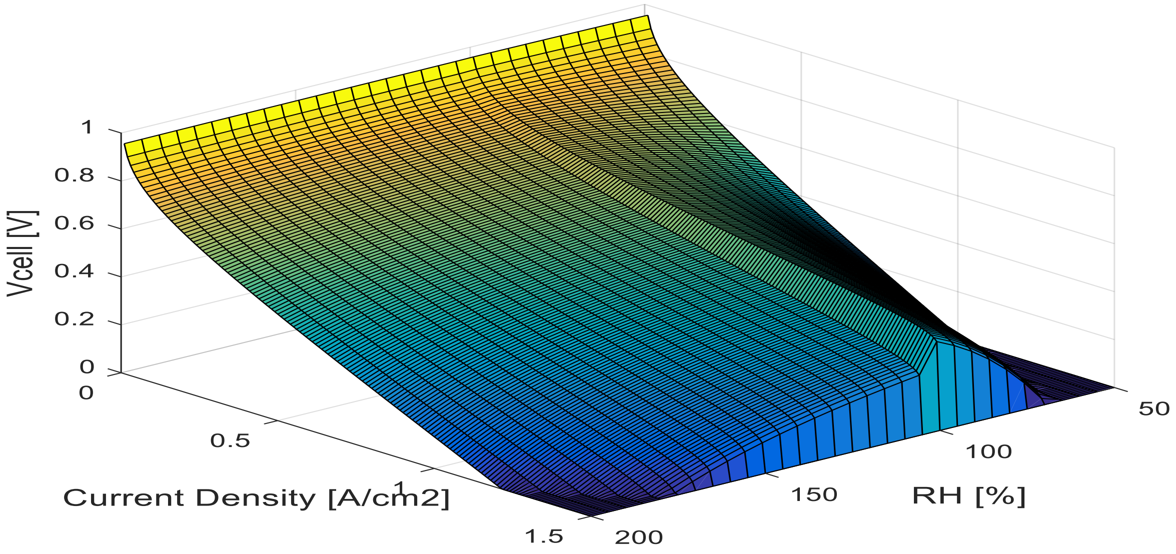

Fig. 7. Variation of Cell Voltage as a Function of Current Density and Relative Humidity with (PH2=PO2=1(atm) and T=90°C).

6. WATER ACTIVITY CONTROL IN PEMFC

Control difficulties may come due to low airflow rates or due to fast and fluctuating input dynamics. Nevertheless, the control scheme must provide useful insight to mark the boundaries of the PEMFC system applicability [24].

The control of the dynamic model of the core of the PEMFC cell has been developed in order to characterize the voltage of the battery according to the power demanded. This model is based on the ideal gas equation or the control of the partial pressures of hydrogen, oxygen and the rate of the wet relativity allowed us to specify the behavior of the molar flow rates necessary for the proper implementation of the system. The effect of hydrogen molar flow on PEMFC voltage and power response is detected.

$R_{H/O}$: it is the reaction oxidant fuel Hydrogen oxygen which is about 1.6

For the controlled hydrogen flow assumed in a hydrogen cylinder K = 1 and the time constant τ =5s, it is given experimentally in Fig. 8.

6.1. Hydrogen Supply

The hydrogen is extended to a pressure close to atmospheric pressure. It can come from a bottle of hydrogen. The control of the hydrogen is carried out by a flow meter controlled via a regulator PI by the equation (15) in Fig. 8, before passing through a water saturator. At the outlet of the water saturator, a filter is placed in order to retain the drops of water that could be carried away by a humidified fraction of the hydrogen. The two streams are then mixed and ready to be introduced into the stack. Just at the entrance, sensors are installed for relative humidity, temperature and the relative pressure of hydrogen.

6.2. Oxygen Supply

Oxygen can come from a bottle of pure oxygen or air. In this article, pure oxygen and compressed air are successively used. Its cotrole is dependent on the incoming hydrogen molecules. At the inlet, the dry air is expanded to a pressure close to atmospheric pressure by the equation (16) in Fig. 8.

Dry air and moist air are mixed and the relative humidity, the temperature and the relative pressure of oxygen are placed at the entrance of the chimney.

6.3. Water supply

The relative humidity of the FC is controlled by the relation (22) via a simple PI controller depending on the flow of water entering the core of the electrolyte membrane and the temperature of the air. This humidity will be controlled at relative humidity reference RH = 100%, so that the error is low for a good operation of the PEMFC system.

7. RESULTS AND DISCUSSIONS

Liquid water saturation is an indicator of the degree of flooding inside the porous electrodes and channels [25].

A PI controller is used with a DC/DC chopper to control the parameters of the system.

In the fluidic domain the flows q_{H_2} , q_{O_2} and q_{H_2O} are controlled for our situation. The purpose of determining optimum dry and wet air flows is to have the ability to work with constant humidity. To avoid the problems caused by the accumulation of impurities (nitrogen). As hypothesis, the cell is powered by pure and perfect hydrogen and oxygen.

At a constant temperature, the control of the parameters allows to see the variations of the (voltage, power and relativity) of the system with different operating parameters (current and incoming pressures of (hydrogen, oxygen and water)) in a simulation time of 100 seconds.

In order to estimate relative humidity at its 100% reference value using relation (22), the control strategy is to set the voltage Vdc at the output of the model to a constant reference value of 6OV. The relative humidity is controlled around 100%, as shown in Fig. 9.

For water management to work properly, this type of control must move the fuel cell from flooding and drought case [26].

Much of the produced water recirculates backscatter and is brought back to the anode without ever leaving the membrane; Excess water is evacuated by the airflow, which explains well the increase in the humidity rate at the outlet of high-current air [4].

Vcell: is the voltage produced by the PEMFC.

Vdc: is the voltage supplied by the DC/DC converter to the payload.

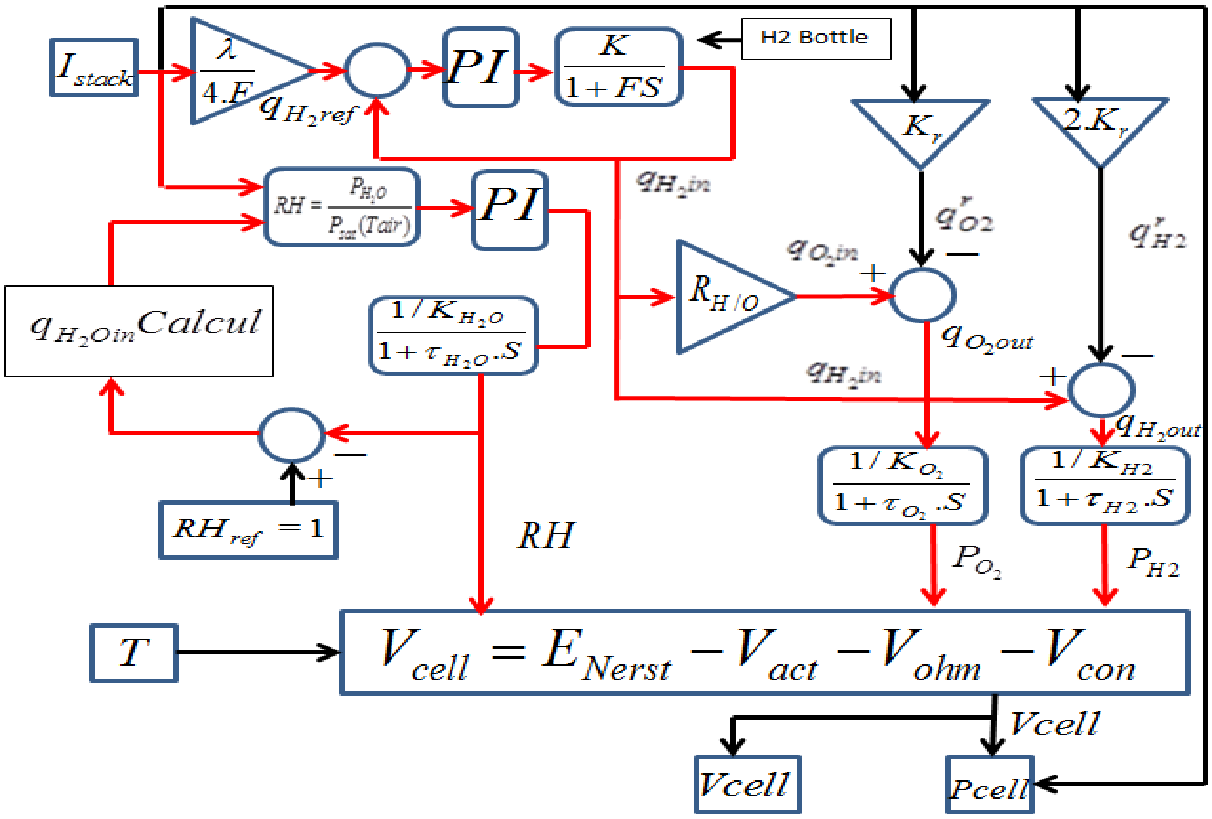

Fig. 8. Synoptic of PEMFC System Control .

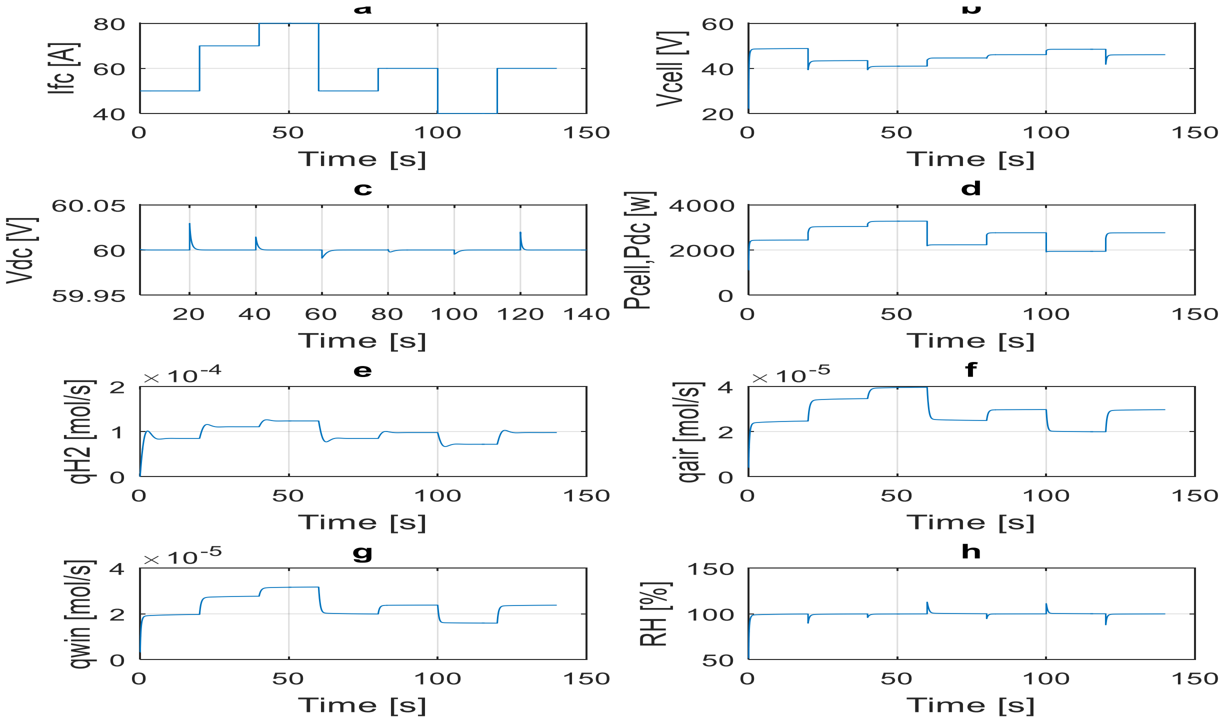

Fig. 9. Control of PEMFC Operating Variable.

8. CONCLUSION

Water management has been the focus of many researches in recent years, it is a critical issue for maximum performance and duration of PEMFC, and these performances are due to the transport and the distribution of internal water of the body of the fuel cell. The effect of water distribution can also be observed in the relative humidity at the inlet and outlet of the anode and the cathode. To ensure that the membrane remains fully hydrated, the relative humidity of the inlet gases and generally maintained at a high value and the pores of the catalyst layer and the gas diffusion layer are frequently flooded by an excess of liquid water, resulting in greater resistance to water transport.

Water management strategies should be addressed taking into account the design of the system as a whole especially in a porous electrode that would be a promising medium for the final solution of water flooding harassing PEMFC.

It is therefore necessary to maintain a balance of subtle liquid water between the flood and the drying of the membrane (The membrane must always remain saturated with water to allow the displacement of the H+ ions) to achieve an ideal performance of this energy system and avoid the degradation of the FC, which is the main problem of water management.

So the water content and the temperature are main parameters for a high yield and conductivity of the membrane.

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