Analyze of Impedance for Water Management in Proton Exchange Membrane Fuel Cells Using Neural Networks Methodology

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ABSTRACT

The objective of this work is to define and to implement a simple method to assess the impacts of relative humidity and operating time on the fuel cell impedance. The method is based on the physical model of Randles with CPE and a mathematical tool for identifying various parameters based on the least squares' method. The objective of the theoretical model development is the model implementation of the control system and water management of predictive diagnostics. Artificial neural networks are used to create the optimum impedance model. The model is applied for the identification of all resistors (internal resistors measured at high frequency, biasing resistors measured at high frequency) which are characterized by a high sensitivity for both cases, the flooding or drying of the cell heart (membrane and electrodes). This model is able to easily generate Nyquist diagram for any condition of relative humidity and operating time, it helped define the stack hydration status. Based on the obtained results, the model demonstrated a best flexible response, accurate and fast. The developed model can be integrated into a water management control system in PEM fuel cells.

I. Introduction

Fuel cell is generator that directly convert the internal energy of a fuel into electrical energy, using a controlled electrochemical process, without harmful gas emissions. Compared to the other types of renewable energy technology, fuel cell has a stable power and a high-efficiency compared to the internal combustion engines and an independence from fossil resource use. PEM is a fuel cell type that works in the low temperature (70-120°C), it is characterized by a high efficiency and one of promising technologies in future [1-6].

PEMFC converts fuel chemical energy into the electrical energy. Typical outline PEMFC is showed in Fig. 1 [2]. In first cell side that is named anode, the fuel is feed under certain pressure. The used fuel in this model is the pure hydrogen, although other gas compositions can be used in this process. [6].

For obtained an electric current of this reaction, the hydrogen dissociation into proton and electron at the anode which is given by Eq. (1), and the oxygen reduction at the cathode Eq. (2) these two electrodes are separated by a membrane, which is conducting the protons from the anode to the cathode. [6].

$$H_2 \rightarrow 2H^+ + 2e^- \qquad Anode \tag{1}$$

$$O_2 + 4e^- \rightarrow 2O^{2-} \qquad Cathode \tag{2}$$

The produced proton passes through the membrane and the electrons are forced to pass through the external circuit, producing electricity, this process is presented in Fig. 1.

The fuel cell successful operation is linked to a water management problem in the gas feed channels of the fuel cell.

The conductive membrane of the proton must be saturated with water, which is essential to ensure a good ionic conductivity [3]. For this reason, the membrane continuous drying led directly to a stack mandatory arrest, it better works after a while of use, because the water is produced in the cathode (hydration of the membrane), after this commissioning time, the excess water must be continuously removed to avoid the flooding and the waterlogging of the cell (gas transport blockage in the electrodes). [6]. Consequently, water management is one of the most critical parameter of PEM fuel cell design [4].

The objective of this presented work is to propose and implement a new and simple method to assess the impacts of the relative humidity and operating time on the impedance. The present method base on the model of Randles changes (CPE) and a mathematical tool for determining various parameters based on the least squares method. The theoretical model development objective is the implementation of control model and the predictive diagnostic water management tool. The artificial neural networks are used to obtains the optimum impedance.

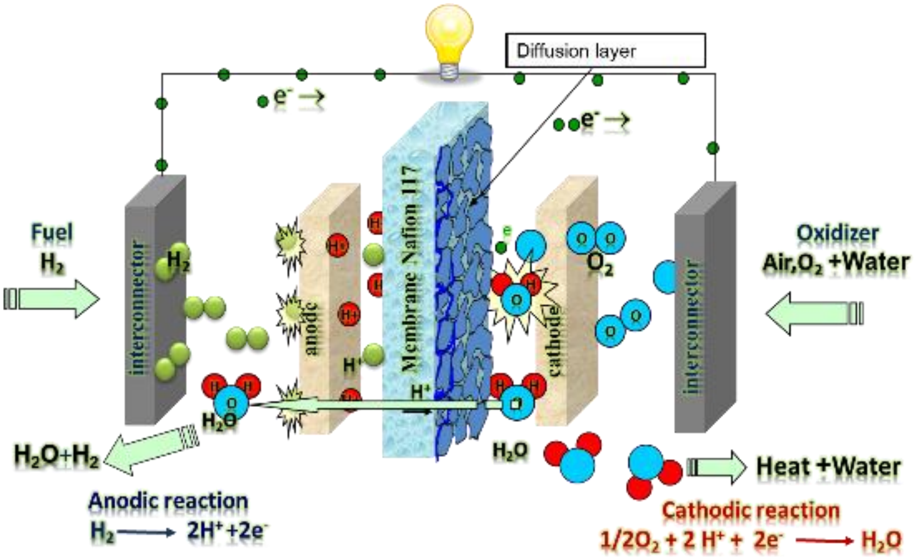

Figure 1.Basic Fell Cell Operation [6].

II. PEM Fuel Cell impedance model

When there is no mass transport limitation, the redox reaction is simply represented by an equivalent electrical circuit of parallel RC cells. [6]. However, when there are considerable variations of the interfacial concentrations on electrodes, the Randles cell is a common and practical way of electrochemical cell modeling by an equivalent circuit [7-9]. It consists of four elements: two resistors, R_m is the electrolyte ohmic resistance and R_p is the polarization resistance, a constant phase element (CPE), which represent some experiments phenomena [10], and the impedance of diffusion called Warburg impedance.

The diffusion impedance general expression for a finite length diffusion layer Z_w is given by. [4-6, 11]:

$$Z_w(j\omega) = \frac{RT}{n^2F^2S\sqrt{j\omega}} \frac{\tanh\sqrt{((j\omega)/D)\delta^2}}{C\sqrt{D}} \tag{3}$$

The relation (3) can be re-written as:

$$Z_w(j\omega)=\frac{RT}{n^2F^2S\sqrt{j\omega}}\frac{1}{C}\frac{1}{D}\delta\frac{\tanh\sqrt{\left(j\omega\left(\delta^2/D\right)\right)}}{\sqrt{\delta^2/D}} \tag{4}$$

Which leads to the definition of two parameters, a time constant:

$$\tau_d=\frac{\delta^2}{D} \tag{5}$$

In addition, the resistance:

$$R_d=\frac{TR\delta}{n^2FSCD} \tag{6}$$

This leads to the final expression of the concentration diffusion impedance

$$Z_w(j\omega)=R_d\frac{\tanh\sqrt{(\tau_d\, j\omega)}}{\sqrt{(\tau_d\, j\omega)}} \tag{7}$$

The standard plane capacitor exhibits the first-order behavior, CPE impedance is defined by [13, 14]:

$$Z_{CPE}(j\omega)=\frac{1}{Q(j\omega)^{\alpha}} \tag{8}$$

With a value of α usually ranging between 0.5 and 1

Fuel cell total impedance is composed by two impedances, one impedance for each electrode, which are in series with the internal resistance R_m. We assume that the rate limiting reaction is the oxygen reduction is directly related to the cathode, and we neglect the anode impedance contribution to the cell impedance. Thus, the retained fuel cell equivalent circuit model is represented by Fig. 2, and the overall impedance is. [6] :

$$Z_T(j\omega)=R_m+\frac{1}{Q(j\omega)^{\alpha}+\left(1/\left(R_P+Z_W\right)\right)} \tag{9}$$

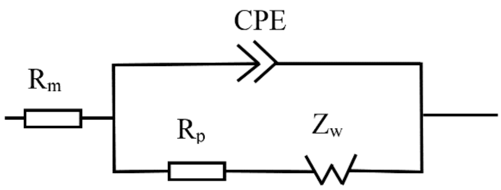

Figure 2. Equivalent circuit proposed by Fouquet et al. [4].

III. PEMFC dynamic characterization by an Artificial Neural Network model

Neural networks are now an actual tool of resolution of several problems that cannot be resolved by analytic or conventional methods [6]. The neural network method principle is based on the function approximations. In this study, we used a neural network of type feed-forward with a supervised training to build a network of four main construction steps. [6]:

- ✓ RNA block construction.
- ✓ Data acquisition (Learning base).
- ✓ Network test.

Neural network is composed of three principal layers, namely: input layer, hidden layer and output layer. Each neuron is connected to all next layer neurons. [6].

The used transfer function in the hidden layer is a sigmoid function; it is given by the following [14, 15]:

$$f(u)=\frac{1}{1+e^{-(d.u)}} \tag{10}$$

With d is the curve slope. The hidden layer input is given by the following equation [14, 15]:

$$u=\sum_{j=1}^{n}\left(w_{ij}x_i+b_i\right) \tag{11}$$

Where xi is the input signal, wij is the weight on the connection between neurons ith and jth in the hidden layer and bi is the bias.

When the output layer function is linear, the network model equation is given by the following equation [14, 15]:

$$y_k u=\sum_{j=1}^{N}\left(w^0{}_{ij}u_i+b_i\right)=\sum_{j=1}^{N}w^0{}_{ij}\,f\left(\sum_{j=1}^{N}\left(w_{ij}x_i+b_i\right)\right) \tag{12}$$

Where yk, is the output signal from kth output neuron, w_{ki}^{0} is the weight of ith output ui to the kth neuron in the output layer.

In this work, the ANN bias values and the weight are updated according to the dynamic gradient descent algorithm [16].

Fig. 3, represented the model architecture of neural network used in this work. The Neural network input layer part is composed of three neurons, the first is linked at the humidity rate, the second is linked at the operation time and the third is linked at the frequency. Hidden layer part is composed of two under hidden layers with twenty neurons of the first and ten neurons for the second; the output layer is composed of two output neurons. (Re (Z), Im (Z)).

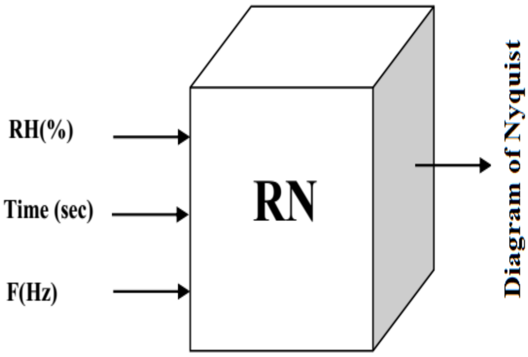

Figure 3. PEMFC Neural networks model.

For proceed to PEMFC parameter simulation, we completed the MATLAB code development that based on the model already presented by using Neural networks toolbox. Hyperbolic tangent sigmoid transfer function ("tansig ") is also used for the parameter estimations in the hidden layer and the linear transfer function ("purelin") is used to the output layer.

Mean square error is shown in Fig. 4. After 100000 iterations, the ANN mean square error reaches a very low value that equal 1.5977 × 10-18.

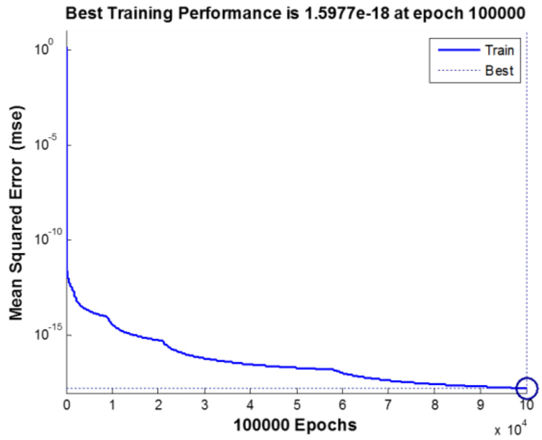

Figure 4. Training mean squared errors for NNT impedance model.

IV. Model test and validation

To establish our neural network learning, a database selection is mandatory. The model learning parameters values are given in Table 1 for four impedance spectra for both flooding and drying cases of the fuel cell membrane [4-6]. The identifying Randles model parameters with CPE is made by factorial design of (DoE) methodology.

Once the neural network-training phase is complete, we go later in the testing phase of impedance spectrum quality estimates for the both cases (drying and flooding of the fuel cell) membrane, for us the proceeds we presented another database, which is totally different to the training base for the verification and validation of the procedure used. [6]

In the cell heart flooding case, two validation tests are presented, the input necessary parameters for the first test are taken from Fig. 8 presented by Fouquet et all. [4], where the time and the relative humidity are 500 secs, RH = 100% and the frequency range is 0.1 Hz to 1 kHz. The input parameters of the second test are taken from the Fig. 8 presented by Fouquet et all [4] where the time and the relative humidity are 1600 sec and RH = 100% and the frequency range is 0.1 Hz to 1kHz.

In the cell heart drying case, the input parameters are taken from the Fig. 10 presented by Fouquet et al. [4], where the time is about 180 sec and RH is 15%, in a frequency range of 0.1 Hz to 1 kHz.

The experimental results and the model results are presented in the Fig. 5, the comparisons between experimental and numerical results of each test indicate that the used procedure is reliable.

Table 1. Model parameters and cell voltage during the flooding and drying [4-6].

Time (sec)	*RH (%)*	R_m	Q	R_p	R_d	τ_d	U
500	15	0.00512	0.952	0.0099	0.0051	0.1155	4.06
3700	15	0.0088	0.62	0.013	0.0101	0.1835	3.35
500	100	0.00398	1.109	0.008	0.0034	0.0872	4.18
3700	100	0.00416	0.936	0.0163	0.0312	0.0947	3.3

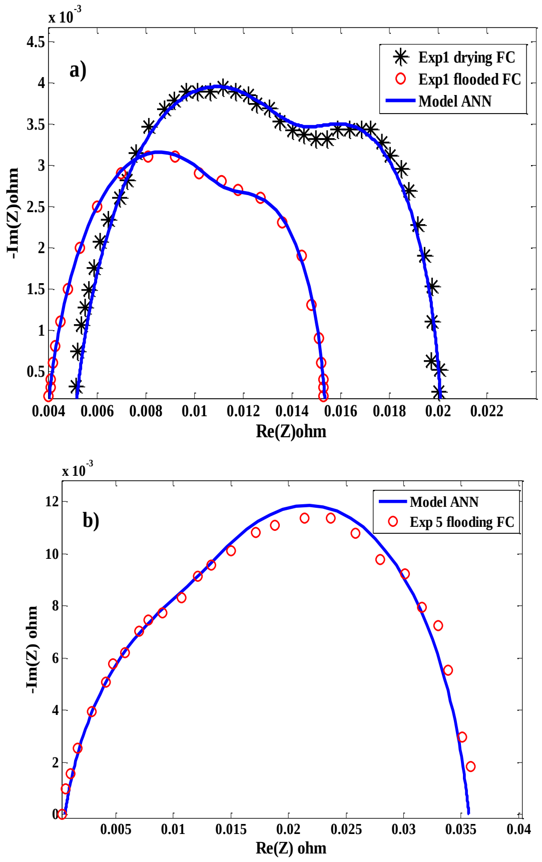

Figure 5. Model validation for a PEMFC flooding and drying cases.

V. Simulation results and discussion of the impedance response

Impedance progressive behavior simulation is realized in the time varies from (500 secs to 2500 sec) at dehydrating conditions RH=10% (dry gas). Spectra Nyquist plots from PEMFC are characterized by the effect of dehydration. Fig. 6, show a right lateral shift by the membrane resistance increase Rint. Under high load conditions, the current density increasing, which provokes a water production increasing at the cathode, this can clearly lead to the flooding case.

Impedance progressive behavior simulation is realized by a time varying from (500 secs to 2500 sec) under flooding conditions RH=100%. The spectra Nyquist plot from PEMFC characterized the effect hydration (Fig. 7), in this case, the large variations are absent in the high-frequency arcs. This behavior is induced by the fuel cell current density increasing, which provokes a water production increasing at the cathode; this can clearly lead to the flooding case. The flooding initial stage is characterized by a slow decrease in the fuel cell output voltage, also, the liquid water begins to accumulate in the channels and constrict the gas flow, thus increasing the diffusion which is related to equivalent circuit parameter Rd and time constant τd.

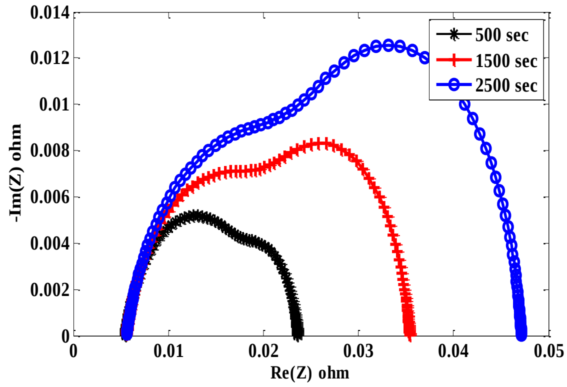

Figure 6. Nyquist diagrams according to the different operating time for a dehydrating conditions of RH=10%.

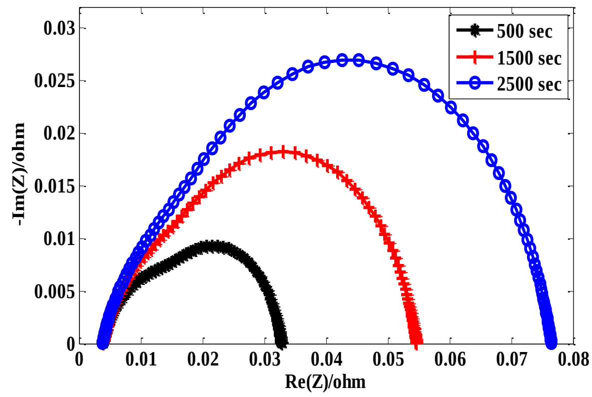

Figure 7. Nyquist diagrams according to the different operating time for a flooding conditions of RH=100%.

The behavior of impedance phase is simulated with a relative humidity variable (RH %) for a time of 500 secs and 2500 sec. The spectra Nyquist plot of PEMFC characterizes the hydration effect. (Fig. 8), (Fig. 9).

Fig. 11 and Fig. 12 show that the measured value of resistance at a high frequency Rint increases with the time, when the relative humidity of the cathode decreases from 100% at 10%. The resistance measured at low frequency Rpol gradually increases, according to the time when the relative humidity of the cathode increase.

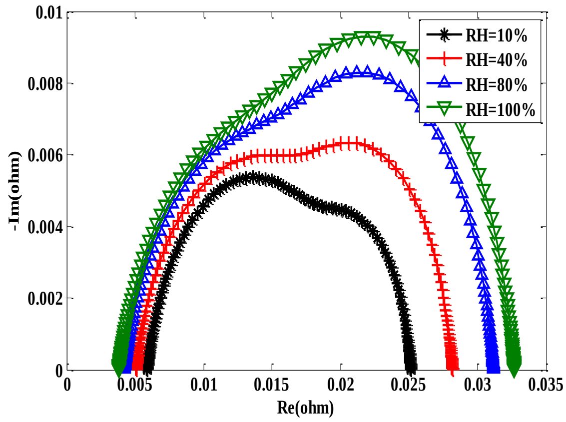

Figure 8. Relative humidity effect on PEMFC state at 500 sec.

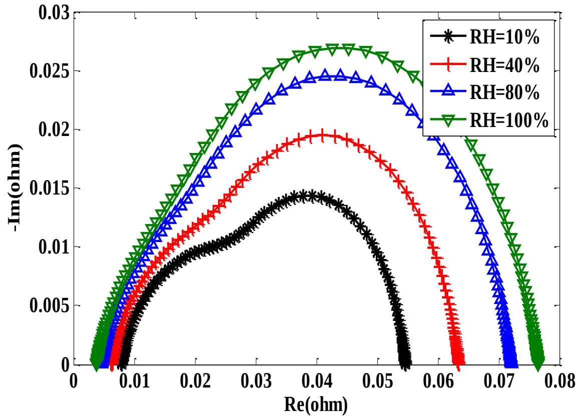

Figure 9. Relative humidity effect on PEMFC state at 2500 sec.

The impedance phase behavior is simulated with varying temperatures for a time of 500 secs to 2500 sec. The PEMFC Nyquist spectrum characterizes the drying effects and flooding (Fig. 10a), (Fig. 10b).

The increase in temperature causes a progressive sliding of the spectra on the right side of the real axis as a function of time, which also causes a reduction in the water content in the membrane and an increase in the membrane resistance.

In these figures, it can also be seen that the resistance measured at low frequency of the stack Rpol; gradually decreased according to the time when the temperature increases in both cases a and b.

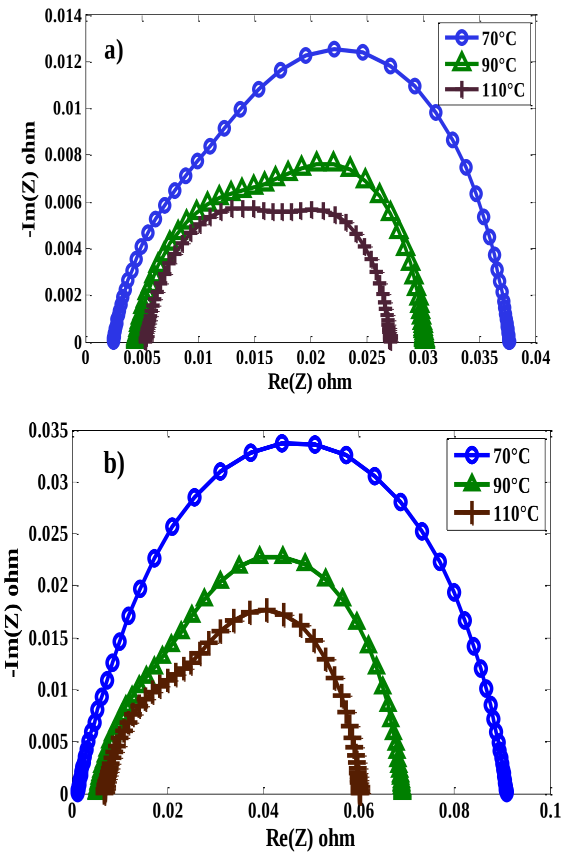

Figure 10. Nyquist diagrams according to the different temperature. a) Case t=500sec, b) Case t=2500sec.

VI. Conclusion

In this work, we have presented a new model for the reproduced of the Randles model with CPE, which represents the Proton Exchange Membrane Fuel Cell impedance. ANN was used to create a PEM impedance optimal model for impact evaluation of relative humidity, operation time and temperature of the fuel cell impedance. This reliable and robust approach, allowed us to identify a set of two parameters (the resistance measured at high frequency and the resistance measured at low frequency) which exhibiting high sensitivity to flooding or drying of the cell heart (membrane-electrodes).

The resistance measured value at a high stack frequency Rint decreases with time, when the cathode relative humidity increases from 10% at 100%. The resistance measured at low stack frequency Rpol gradually decreases, according to the time when the cathode relative humidity will decrease.

The resistance measured value at a high frequency of the stack Rint increases with time, when temperature increase from 70°C at 110°C.

The resistance measured at low frequency of the stack Rpol; gradually decreased according to the time when the temperature increases.

This proposed approach we have allowed to identify two parameters sets that have shown a high sensitivity to fuel cell flooding or drying (The resistance measured value at a high frequency of the stack Rint and the resistance measured at low frequency of the stack Rpol).

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