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Dynamic Model of a PEM Electrolyzer based on Artificial Neural Networks

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Abstract: *Hydrogen production by electrolysis is emerging as one of the most promising ways to meet future fuel demand; likewise, the development of models capable of simulating the operation of electrolysis devices is indispensable in the efficient design of power generation systems, reducing manufacturing costs and resources savings. The nonlinear nature of the Artificial Neural Network (ANN) plays a key role at the development of models for predicting the performance of complex systems. The behavior of a Polymer Electrolyte Membrane (PEM) Electrolyzer of three cell stack (100 cm^2 of active area) was modeled successfully using a Multilayer Perceptron Network (MLP). This dynamic model has been trained to learn the internal relationships that govern this electrolysis device and predict its behavior without any physical equations. The electric current supply and the operation temperature were used as input vector able to predict each cell voltage behavior. A reliable accuracy (< 2%) was reached in this work after comparing the single cell performance of the real electrolyzer versus the ANN based model. This predictive model can be used as a virtual device into a more complex energy system.*

Keywords: Artificial Neural Network (ANN), Model, Multilayer Perceptron Network (MLP), PEM Electrolyzer

1. INTRODUCTION

Energy production based on combustion of fossil fuels represents a severe negative impact on world economics and environment [1]. Electrochemical energy production is projected as an alternative power source; hydrogen has been identified as a promising fuel for sustainable energy supply provided that it is obtained from renewable energy [2-5]. The impending increase in the demand for hydrogen is a consequence of the technological developments which make use of it in a wide range of applications in the energy sector. However hydrogen generation is one of the critical aspects due to the high energy amount needed to obtain it. Different processes are analyzed: photo-dissociation of water, thermochemical cycles and microscopic organisms as algae, etc; despite the fact that these processes are still far from practical use. Water electrolysis is a well-established technology and one of the most widely used methods for producing high purity hydrogen [6]. Water electrolysis in a Proton Exchange Membrane (PEM) device is characterized by high efficiencies and suitable current density even at low temperatures. In comparison to the traditional alkaline electrolyzers, in which corrosive potassium hydroxide (KOH) solution is used as electrolyte, systems based on PEMs have a number of advantages, such as ecological cleanliness, considerably smaller mass-volume characteristics and essentially, a high degree of gas purity [7]. There is also the opportunity to obtain compressed gases directly from the electrolyzer at an increased level of safety. High manufacturing costs and production of PEM electrolyzers are the main disadvantages that limit their use. The production costs are dependent on the cost of electricity and a large energy supply is often needed due to high anode overpotentials [8-10].

Several models have been proposed to simulate PEM electrolyzer systems based on physic and electrochemical phenomena.

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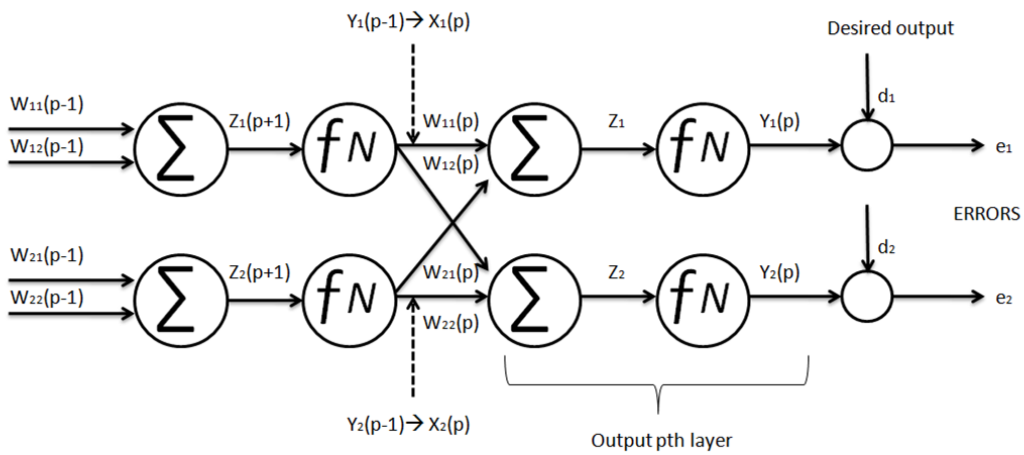

Figure 1. A Multi-layer Perceptron Network.

[11, 12]. Analytical models are an adequate tool to understand the effect of basic variables on the electrolyzer performance [13]. Semi-empirical models allow designers and engineers to predict the electrolyzer performance as a function of different operation conditions (temperature, pressure, water flow) using simple math equations [14]. However, this kind of models requires high level of knowledge of the process parameters as well as assumptions in order to simply them, for this reason they lose accuracy. Nevertheless it is possible to reach performance modeling using black-box models such as Artificial Neural Networks (ANN´s) [15-17]. These models are based on a set of measurable input parameters such as current supplied and temperature and they can predict the behavior of interesting output parameters such as cell voltage. A short stack PEM electrolyzer was characterized and used to build a dynamic model based in Artificial Neural Network (ANN).

1.1. Artificial Neural Network

An Artificial Neural Network (ANN) is inspired by the structure and functional aspects of biological neural networks; it can be regarded as a black box model able to give certain output data as a response of specific input values combination, depending on the effective identification of the main operation factors in the system performance. ANN's are widely accepted as a technology used in many engineering applications such as in control systems, pattern recognition and modeling, offering an alternative way to handle with complex problems. ANN´s are data driven methods, in the sense that is not necessary to postulate tentative formal models and then estimate their parameters, they can learn from examples, are fault tolerant and are able to handle noisy and incomplete data, as well they are able to deal with non-linear problems, and, once trained, can perform predictions at very high speed.

This paper specifies the data acquisition process and ANN´s design for the dynamic model of a PEM electrolyzer stack by using the Backppppnnuhvupropagation (BP) learning algorithm for a Multilayer Perceptron Network (MLP) [18].

The topology of the network is defined by the neurons organization. The MLP is organized by setting the number of neurons in the input and output layer according to the specific application, and optionally added hidden layers; fig. 1 shows an example of the topology of an ANN.

1.2. ANN´s learning process

The learning process make possible to adjust speed and accuracy. The idea with the BP algorithm is feedback to the neural-network, the errors generated when its outputs differs from the desired outputs. For this reason, a least mean square (LMS) error function, e, is introduced, according to:

$$e \overset{\Delta}{=} \frac{1}{2} \Sigma_{\kappa} (d_{\kappa} - Y_{\kappa})^{2} \tag{1}$$

$k=1,2,\ldots,N$, being N the number of neurons in the output layer, d the desired output, and Y the ANN´s output.

During this procedure, local gradients of e with respect to the weights W_k, are calculated, which later can be used for adjusting the old weights in (4). The goal of the learning is to find the optimal weights that minimize this error. The instantaneous error provides an indication of the model´s current performance. The weights are adjusted in order to improve the current performance of the model. The major assumption is that the weights correction ΔW_{kj} is proportional to this gradient, with a constant, η, also known as the learning rate:

$$\Delta W_{kj}(m) = -\eta \frac{\partial e}{\partial W_{kj}} \tag{2}$$

$$W_{kj}(m+1) = W_{kj}(m) + \Delta W_{kj}(m) \tag{3}$$

From the MLP definition, data X_j is presented at the input layer, each unit input is weighted and added to produce a net output Z_k:

$$Z_k = \Sigma_k W_{kj} X_j \tag{4}$$

The BP algorithm requires the use of differentiable transfer functions, due to the calculation of the local gradients. The choice of the best activation function f_N results from various test carried out on each layer pursuing a minimum error goal.

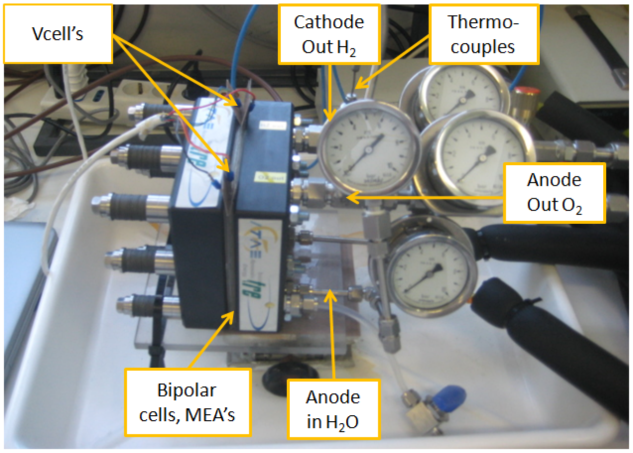

Figure 1. Three cells stack PEM electrolyzer.

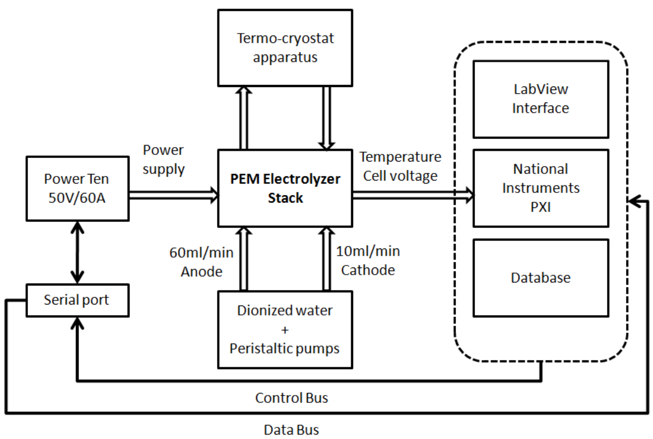

Figure 2. Schematic representation of the house-made test station.

$$Y_k=f_N(Z_k) \tag{5}$$

To accelerate the convergence in learning process, we use the Levenverg-Marquardt algorithm which optimizes the gradient descent method of the BP.

Once described the MLP learning algorithm, the relevant task consist about define the structure of the network based on the particular application of modeling a electrochemical device, the reliable performance of the ANN based model depends on the appropriate selection of the input-output pairs of variables that rules the behavior of the system and the availability of a sufficient number of patterns (experimental tests) from which the network can learn before it is tested. Later in this paper we describe the variable's selection process and training patterns of the neural network proposed.

2. EXPERIMENTAL

2.1. PEM Electrolyzer

In the water electrolysis process O-H bonds of water molecules are broken by both electromotive force and the catalytic action of the anode electrocatalyst when DC voltage is supplied.

$$H_2O \rightarrow \frac{1}{2}O_2 + H_2 \tag{6}$$

In PEM electrolyzers allows proton migration and separates H_2 from O_2 gases. The hydrogen protons, H, migrates through the membrane and recombines at the cathode forming H.

A short PEM electrolyzer stack was assembled and tested. The Membrane and Electrode Assembly (MEA) were made using Nafion® 115 (Ion Power). IrO_2 anode catalyst was prepared by a colloidal method followed by thermal treatment [10]. The anode electrocatalyst was deposited onto one side of the membrane, whereas a Pt/C catalyst-based was used as cathode. A Ti mesh was used as backing layer for the anode and carbon cloth for the cathode. The geometrical area of each MEA was 100 cm^2. MEAs were assembled in a short stack of three cells connected in series by tightening at 7 Nm using a dynamometric wrench. The stack hardware (fig. 2) included end-plates and bipolar plates of stainless steel due to their high resilience to electrochemical corrosion in acidic environment.

The electrochemical tests were carried out at CNR-ITAE with an in-house made test station (fig. 3) which included a hydraulic circuit, mainly consisting of pre-heaters and water condensers; a stack temperature control module which consisted of a thermocryostat device and thermocouples allocated inside the stack close to the inlet and outlet of the water flow and always keeping the temperature gradient lower than ±2 °C at the various current densities. In order to keep constant the operation temperature, deionized water was supplied to the anode and cathode compartments by a peristaltic pump at a flow rate of 60 ml min^{-1} and 10 ml min^{-1}, respectively. A power source (Power Ten model R62B) was used to supply the electrical energy to the electrolyzer. The overall stack voltage and the voltages of the various cells were measured by Advanced Measurements high common mode rejection ratio digital voltmeters. All the instruments of the test station were controlled by a Labview^{TM} software and PXI National Instruments interface boards. The test station also included separate instrumentation for the electrochemical diagnostics.

3. METHODOLOGY

The design of the neural-network is based on the understanding the process and identifying how each variable affects the PEM Electrolyzer performance. This knowledge allows to choose the appropriate data to train the neural-network. Nevertheless it is important that the training patterns must be well distributed throughout the operation range in order to avoid the overfitting problem. Once the fewest dominant variables are recognized, the neural network training process can start. From the test station and monitor module (see fig. 4), a data base was obtained for the PEM Electrolyzer.

Two variables were used as inputs to the neural network model: electrolyzer electric current and the operating temperature in each single electrolyzer cell (table 1), while cell voltage was observed and defined as output. Although hydrogen production rate is desired as the main output, this can be estimated according to Faraday's law denoted by:

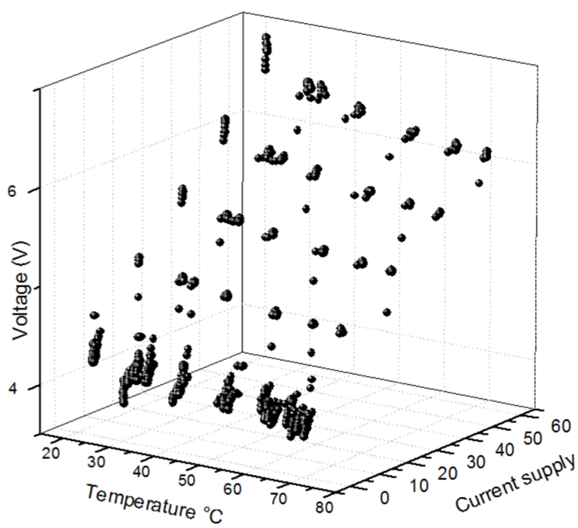

Figure 4. Main set of variables and their distribution in the electrolyzer operation space.

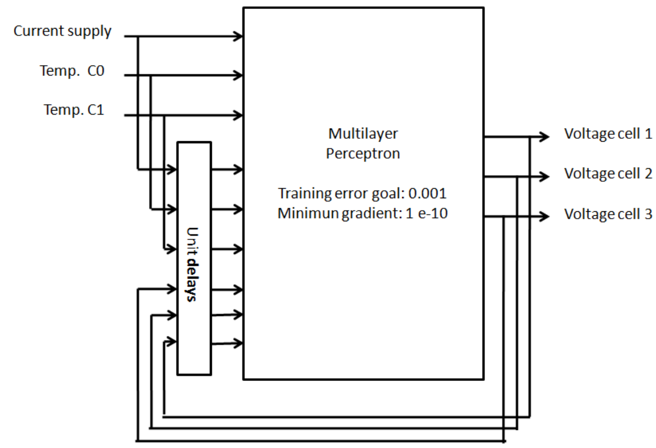

Figure 5. Recurrent Multilayer Network.

$$N_{H_2} = \frac{n_c I}{nF}\eta_F \tag{7}$$

Hydrogen flow rate N_{H2} (mol s^{-1}) is directly proportional to: the current I through each electrolyzer cell, the number of cells n_c and the Faraday efficiency n_F. The Faraday efficiency is defined as the ratio between the experimentally determined and theoretically estimated amount of hydrogen produced at a fixed current. Losses can occur from oxygen and hydrogen cross-over through the membrane with consequent chemical recombination [19]; n is the numbers of moles of electrons per moles of reacted water and F is the Faraday constant.

The test station is used to observe the operation of the device within an operating space and get the training database. Current steps of 15 amps were supplied to the stack into a range of 1 to 60 amps. The ambient temperature was ranged from 23°C to 75°C. A total of 30 pairs of current-temperature patterns were extracted from the stack, each measurement was performed with duration of one minute at a sample rate of two seconds. Once data-base is acquired, the following step is the separation in training and validation patterns. As from a total of 900 confident measures, 60% were used to train the network and 20% for testing and 20 % for validation in order to evaluate the robustness of the model [20]. An additional random sequence of current steps at different temperatures was also extracted from the stack in order to evaluate the predictive performance of the ANN based model. The input and output variables for the ANN´s learning and activation function were normalized to be within the range of (-1, 1), this process was necessary for the faster and better learning of the neural model, the minimum and maximum values of the training data were used after for the validation process.

Table 1. Input variables

Current supply (A)	Temperature setting (°C)
1	[23,25]
15	[35,40]
30	[45,50]
45	[55,60]
60	[65,70]
	Tmax.

4. RESULTS AND DISCUSSION

The basic electrochemical characterization of the PEM stack electrolyzer and active stack components was reported in a previous paper [18]. In this work, we have focused our efforts on stack behavior modeling by using the artificial neural network approach. In this regard, the experiments were addressed to this specific purpose.

The Levenberg-Marquardt [21-23] training process was carried out by testing several architectures and activation functions, in order to reach a minimum error goal settled in 0.001. Since no theory enables yet to determine the amount of hidden layers and neurons for the correct modeling process, the choice of the best architecture was made after testing many topologies. This selection was done evaluating the commitment between speed and accuracy in the prediction process for every model. The feedback connections originated from the output neurons have an important impact on the learning capability of the network, and on its performance. Moreover, the feedback loops involve the use of particular branches composed of unit-delay elements (denoted by z^{-1}), which results in a nonlinear dynamical behavior due to the nonlinear nature of the neurons. Non linear dynamics play a key role in the performance prediction of the system. Besides, since the BP algorithm is a gradient descent method, the activation functions must be differentiable, so that, *logistic* activation functions were used for hidden layers, and *hyperbolic tangent* function for the output layer. This selection was made after several tests carried out. Finally the 9 inputs 12

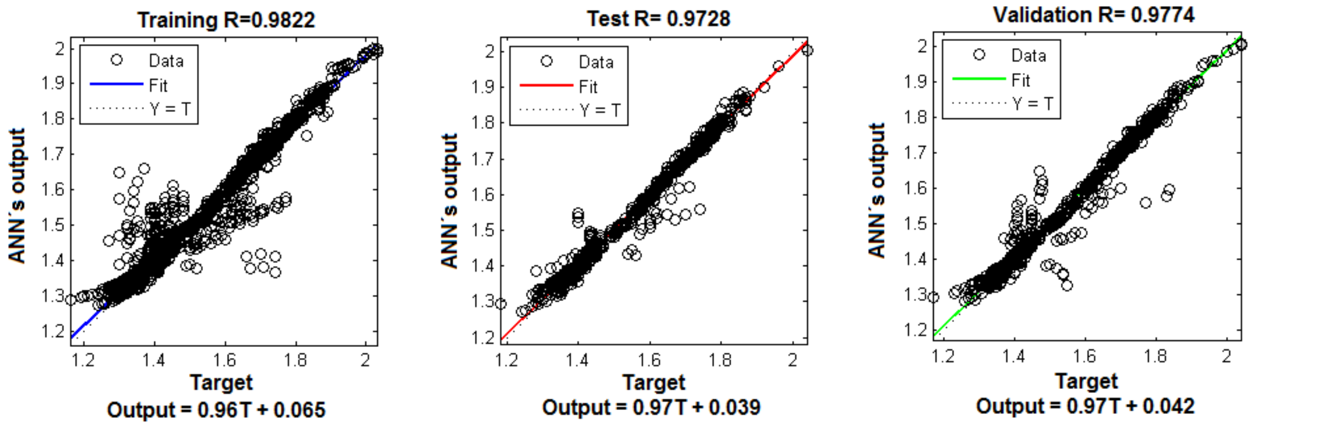

Figure 6. Rate of correlation of the output variable (stack voltage) by linear regression for training, testing and validation processes.

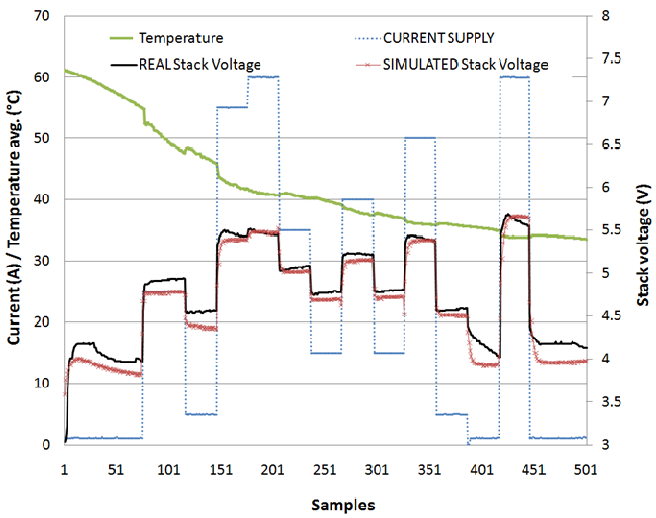

Figure 7. Random current steps supplied to the stack and comparison between the stack real voltage performance versus the ANN based model output (Sampling period is 2 seconds).

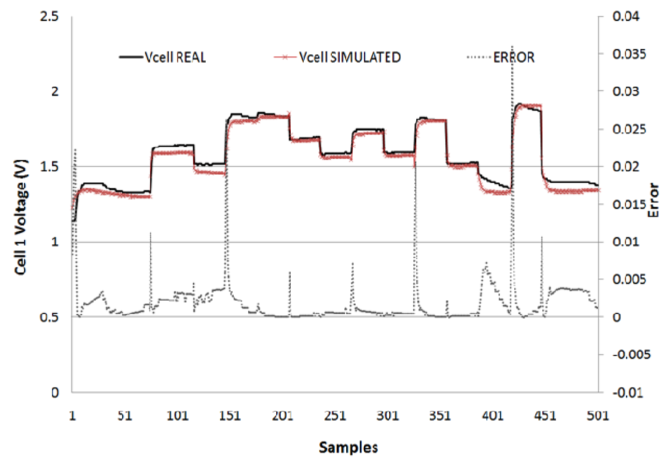

Figure 8. Comparison between the stack real voltage performance versus the ANN based model output in cell 1 (sampling period is 2 seconds).

hidden neurons and 3 outputs (fig. 5) architecture brought the best performance for this particular application.

Voltage linear regressions for training, testing and validation processes were calculated in order to evaluate the correlation between experimental data from the PEM electrolyzer and simulated data from the ANN model. A correlation rate of R= 0.9822 (fig. 6) was attained, indicating the success of the training process. In order to illustrate the performance of the network as a predictor, the mentioned sequence of random current steps from 1 A to 60 A was tested during a gradual decrement of temperature. This validation test is observed in figure 7, where the total stack voltage is compared against the simulated performance of the ANN based model. Figs. 8, 9 and 10 compare the voltage behavior of every single cell and the calculated error. The maximum error dropped in every cell is showed in table 2.

In each step current, a peak error occurs during ~2 s. Later it decreases and follows the system performance. The maximum error calculated was 4.89% in Vcell 2, first and third cells have a similar behavior, particularly at the beginning of the sequence and when a step from 1A to 60 A is supplied to the stack. In order to measure the total performance of the model, is necessary to calculate the error during the entire sequence (see fig.11). The error associated to each distribution was calculated by the Mean Square Error (MSE).

$$MSE=\frac{1}{N}\Sigma_{i=1}^{N}(A-T)^2 \qquad (8)$$

Where A represents the simulated voltage output for every cell and T the real experimental values taken from the stack. For the stack voltage output value in the validation sequence, the highest error obtained from comparing real values versus simulated data was = 0.039 V (1.96%) which is the lowest error value reported so far [15-17].

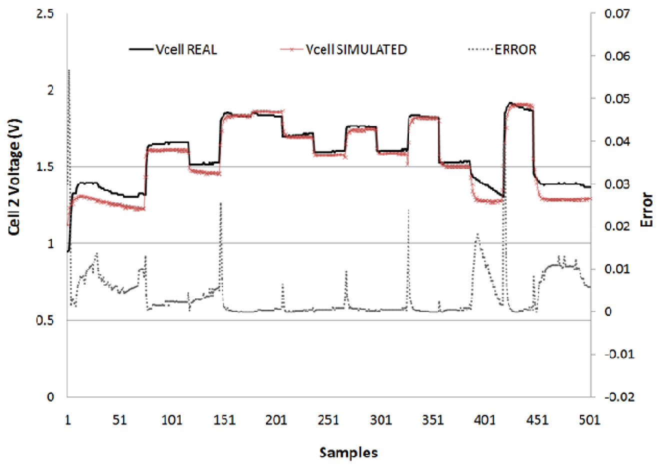

Figure 9. Comparison between the stack real voltage performance versus the ANN based model output in cell 2 (sampling period is 2 seconds).

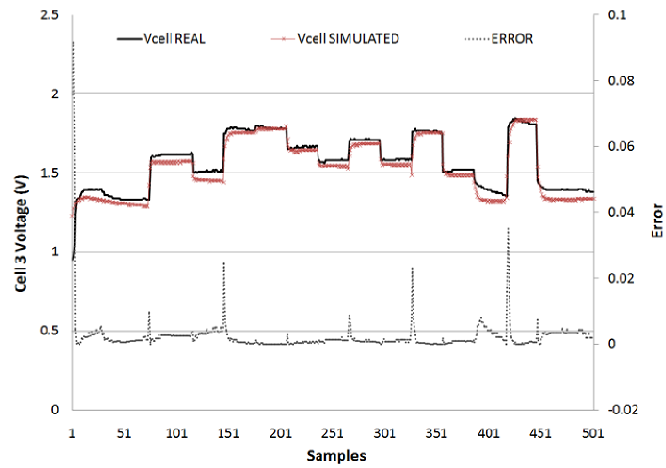

Figure 10. Comparison between the stack real voltage performance versus the ANN based model output in cell 3 (sampling period is 2 seconds).

5. CONCLUSIONS

Modeling systems allows the analysis and study of physical systems with the idea to optimize their performance saving time and resources. When modeling is done by using experimental data which cover M × N dimensional space, ANN based model shows good dynamic performance prediction since it takes into account the non-linearities, the non-modeled dynamics, and the non-measurable noise. Modeling of a PEM Electrolyzer stack was developed using an ANN as alternative approach when physical variable relationships are not well known. The Multilayer Perceptron Network with Levenberg-Marquardt learning algorithm designed in this work showed excellent accuracy in modeling and performance prediction of the output cell voltage for this particular application. The present ANN model considered a three inputs (current supply and two stack temperatures) vector and its respective delays units. The maximum error achieved for the stack voltage prediction was 1.96%. This work extends the use of ANN´s as modeling tool for a PEM Electrolyzer, achieving an excellent degree of accuracy in performance prediction.

6. ACKNOWLEDGMENTS

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Table 2. Maximum Error measured in every single cell

	Vcell 1 (V)	Vcell 2 (V)	Vcell 3 (V)
Max. Error	0.0360 4.76%	0.0567 4.89%	0.0918 4.78%

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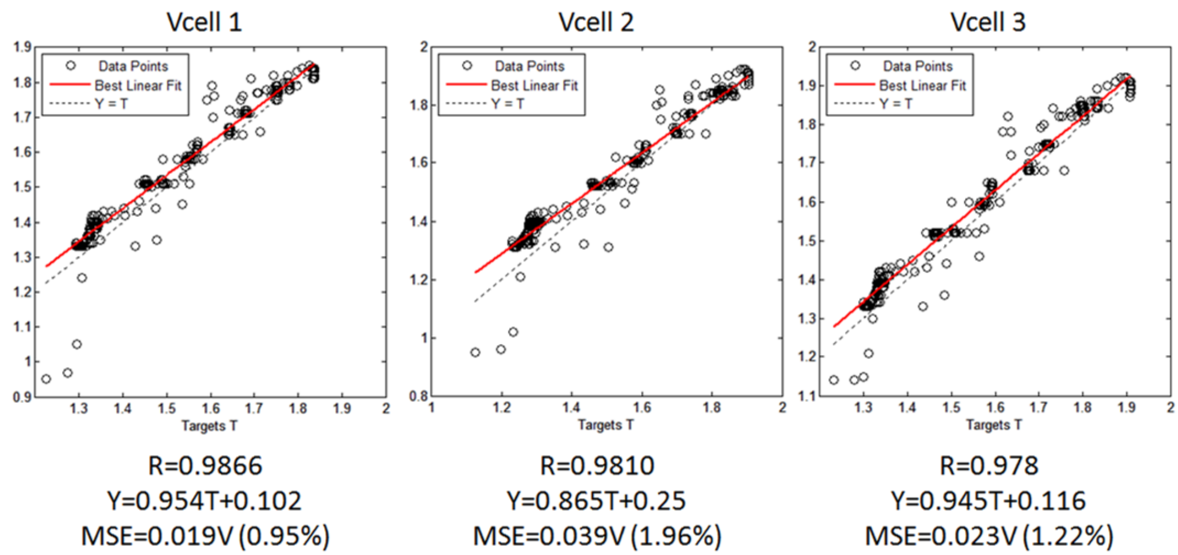

Figure 11. Correlation rate obtained from the validation sequence and Mean Squared Error for each cell

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