Artificial Intelligence Techniques for the Estimation of Direct Methanol Fuel Cell Performance

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Abstract. Artificial neural networks and neuro-fuzzy inference systems are well known artificial intelligence techniques used for black-box modelling of complex systems. In this study, Feed-forward artificial neural networks (ANN) and adaptive neuro-fuzzy inference system (ANFIS) are used for modelling the performance of direct methanol fuel cell (DMFC). Current density (I), fuel cell temperature (T), methanol concentration (C), liquid flow-rate (q) and air flow-rate (Q) are selected as input variables to predict the cell voltage. Polarization curves are obtained for 35 different operating conditions according to a statistically designed experimental plan. In modelling study, various subsets of input variables and various types of membership function are considered. A feed -forward architecture with one hidden layer is used in ANN modelling. The optimum performance is obtained with the input set (I, T, C, q) using twelve hidden neurons and sigmoidal activation function. On the other hand, first order Sugeno inference system is applied in ANFIS modelling and the optimum performance is obtained with the input set (I, T, C, q) using sixteen fuzzy rules and triangular membership function. The test results show that ANN model estimates the polarization curve of DMFC more accurately than ANFIS model.

Keywords: Direct Methanol Fuel Cell, Artificial Neural Networks, Neuro-Fuzzy, Modelling, Performance

1. Introduction

In recent years, the depletion of fossil fuel reserves, global warming and other environmental concerns have accelerated researches on fuel cell technology. Among various types of fuel cells, polymer electrolyte (PEM) hydrogen and direct methanol fuel cells (DMFC) are the most intensively searched as principal candidates for stationary and portable electrical energy sources. Liquid methanol has advantages of easy fuel storage and higher energy density compared to hydrogen. But, slow anode kinetics and methanol crossover are still major barriers to its commercialization.

Researches on fuel cells are mainly focused on the development of the various components of the membrane-electrode assembly (MEA), especially on the anode catalyst and PEM. On the other hand, the development of elaborated mathematical models is another active research area, as they are valuable tools to analyse ultimate details of electrochemical and physical phenomena occurring in various parts of the fuel cell [1- 7]. In this context, 3D models have been developed in order to detect the effects of operation conditions as well as the other structural-spatial and material properties on the complex mass transport, electrode kinetics, and two-phase flow in various parts of the fuel cell [8, 9]. These computational fluid dynamics (CFD) models which need commercial softwares and also a huge amount of thermo-physical parameters values to be estimated are usually used for single cell analysis but they are not suitable for analysis, design, optimization and control tasks at system level. Thus, semi-analytical or empirical models find still applications in these areas.

Empirical models can be divided as parametric and non-parametric ones. Parametric empirical models are based on pre-determined relations derived from scientific knowledge on activation, ohmic and mass transfer losses [10-13]. They correlate well fuel cell output as a function of current density, temperature, and fuel-oxidant concentrations, but they usually lack of other process variables affecting fuel cell performance. Furthermore, the model parameters have to be determined under different operating conditions for different systems.

In recent years, fuzzy logic and artificial neural networks (ANN) have been extensively used as effective non-parametric modelling tools capable of estimating non-linear nature of complex systems by means of sufficiently representative data sets. ANN offer exciting advantages, such as learning, adaptation, fault-tolerance, parallelism and generalization. Highly nonlinear complex relationships are implicitly put down in the weights of the network without the need of an explicit mathematical relation. On the other hand, fuzzy logic provides an inference morphology that enables to approximate human reasoning capabilities to be applied to knowledge-based systems. Fuzzy modelling is concerned with the construction of fuzzy inference systems that can predict and explain the behaviour of ill-defined or complex systems which requires otherwise an excessive number of parameter data and sophisticated numerical software for model solving. In recent years, various advantages of these two modelling approaches are beneficiated by hybrid approaches such as adaptive neuro-fuzzy inference system (ANFIS). Besides the modelling purposes, these techniques are successfully applied in process control for various engineering applications.

Despite the broad applications of these artificial intelligence techniques, a few studies have been conducted on the modelling of various types of single fuel cells [14-19] and cell stacks [20, 21]. In this study, feed-forward ANN and ANFIS models have been used to predict the polarization curve of DMFC, using current density, fuel cell temperature, methanol concentration, gas and liquid flow-rates as operating variables for a given MEA, channel geometry and dimensions. Various aspects of these modelling techniques have been investigated and comparative results on the test performance have been given.

2. Theory

2.1 Feed -Forward ANN

Inspired from the human brain, ANN is a layered organization of neurons. Among various types of organization (architecture), the most popular is multilayer feed-forward ANN consisting of an input layer, a series of hidden layers and an output layer, as exemplified in Fig.1 . The weighted sum of the input signals fed to a neuron is transformed to a normalized output signal by means of an activation function and transferred to the subsequent layer. The most popular activation functions are “tansig” and “logsig” function defined mathematically as

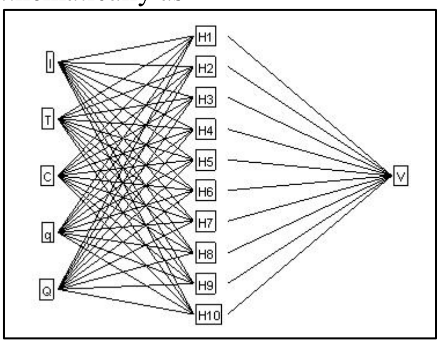

Figure 1. One hidden layered feed-forward ANN

$$\textit{Logsig activation function}: \; y = 1/(1+e^{-x}) \tag{1}$$

$$\textit{Tansig activation function}: \; y = 2/(1+e^{-2x}) - 1 \tag{2}$$

Mathematically, ANN is a black box containing a weight matrix optimized by a training process. For prediction or forecasting purposes, ANN model is trained usually by supervised learning using a data set organized usually in matrix form with rows corresponding to cases (experimental results) and columns corresponding to variables. The most widely used training algorithm for multilayer feed-forward ANN is the back-propagation. During the training process, model outputs are recalculated until some function of the network errors (the differences between model and experimental outputs) is minimized iteratively to yield finally the optimum weight matrix. "Overtraining" which falls the forecasting capability of the network due to the structure excessively adopted to the training data is avoided by using an independent check data; the training process is terminated when checking error tend to increase even if the training error continues to decrease. The ultimate success of the model is always assessed by an independent test data.

2.2 Fuzzy inference system

Fuzzy inference is a logical method that interprets the values in the input vector (x), and based on some set of fuzzy "*if-then*" rules, assigns values to the output vector (y):

$$IF\ x \text{ is } A \quad THEN \quad \text{y is } B \tag{3}$$

Where A and B are labels of fuzzy sets, e.g., "low", "high". Each fuzzy set is characterized by appropriate membership functions that map each element to a membership value between 0 and 1. Triangular, trapezoidal, Gaussian and disigmoidal types are examples of widely used membership functions.

A fuzzy inference system consists of three conceptual components namely, a *rule base* comprising fuzzy rules, a *database* defining the membership functions of the fuzzy sets used in the fuzzy rules and a *reasoning mechanism* which performs the inference procedure. A fuzzy inference process is realized in five steps:

1) Fuzzification of crisp inputs by means of membership functions
2) Application of fuzzy operations to antecedent parts
3) Application of implication to consequent parts
4) Aggregation of rules outputs into a single fuzzy set
5) Defuzzyfication of the final fuzzy set into a single crisp output.

Tagaki-Sugeno fuzzy inference system is widely used in data based modelling. The *i*.th rule for a first order linear Sugeno model with two input variables (x_1, x_2) is given as ;

$$\text{IF } (x_1 \text{ is } X_{1,i}) \text{ AND } (x_2 \text{ is } X_{2,i}) \text{ THEN } y_i = p_{i,0} + p_{i,1}x_1 + p_{i,2}x_2 \tag{4}$$

where p constitutes a tunable parameter set . The crisp output, y, is the weighted sum of the rule outputs , y_i

$$y = \sum W_i y_i \tag{5}$$

Where W_i is the weight of the *i*.th rule.

2.3 *Fuzzy and Neuro-Fuzzy Modelling*

Fuzzy modeling involves two phases namely, structure identification and parameter optimization. Structure identification encompasses the selection of input variables, the specification of the fuzzy inference system, the rule base set, the type and number of the membership functions. The parameter optimization is the fine-tuning of model parameters to best fit the input-output data set. Neural networks learning techniques are successfully applied in this phase.

Adaptive neuro-fuzzy inference system (ANFIS) is proposed to integrate fuzzy logic and ANN. ANFIS incorporates a Sugeno type fuzzy inference system into adaptive feed forward ANN structure, as illustrated in Figure 2. For training ANFIS, a hybrid algorithm has been developed by combining the gradient method and linear regression, which is faster than the classical back-propagation method [22].

The functionality of neurons in different layers of ANFIS is summarized as follows:

Layer 1: Neurons are adaptive; membership functions of input variables are used as activation functions, and parameters in this layer are referred to as antecedent (usually non-linear) parameters.

Layer 2: Neurons are fixed with outputs representing the firing strengths of the rules.

Layer 3: Neurons are fixed with outputs representing normalized firing strengths.

Layer 4: Neurons are adaptive with parameters referred to as consequent (constant or linear) parameters.

Layer 5: The single neuron is fixed with output equal to the sum of all the rules outputs (eq. (3)).

3.0 Experimental

3.1 Experimental Set-up

A commercial MEA provided by Fuel Cell Store was used in experimental study, with the following specifications:

Membrane type: Nafion 117
Anode catalyst: % 20 (1:1 Pt-Ru) /C with 2 mg/cm^2 loading
Cathode catalyst: Pt black with 2 mg/ cm^2 loading
Gas diffusion and backing layer: Carbon cloth (for both electrodes)
Effective surface area: 25 cm^2

The single cell unit has flow field of serpentine geometry: the flow channels are 1.5 mm wide, 1 mm deep and 38 cm total length. The rib between the channels is 1 mm. A computer controlled fuel cell test station (Fideris, USA) was used to obtain the polarization curves.

3.2 Experimental design

Fuel cell temperature, gas and liquid flow-rates and methanol concentration were selected as operating variables. The operating ranges of these variables are given in Table 1 and chosen as broad as possible to encompass practical ranges considered in the literature survey. The inlet air temperature and pressure were kept constant at 25 ^{o}C and 5 bar.

Polarization curves were obtained for 35 different operating conditions according to the second order experimental plan given in Table 2 where each operating variable is examined at six different levels. Furthermore, 3 central replicates were included in the plan to estimate the experimental error.

Table 1. Operating variables and ranges

Variable		range
Methanol concentration	C (M)	0.25- 3.25
Liquid flowrate	q (ml/min)	2- 14
Air flowrate	Q (ml/min)	450- 1200
Temperature	T (^{o}C)	25- 65

4.0 Results and discussion

MATLAB neural networks and fuzzy toolboxes were used in the modelling study. A polarization data comprising 460 data points collected according to the experimental plan was partitioned in three subsets in the following proportions; 60 % for training, 20 % for check and 20 % for test. The same datasets were used for both modelling studies. The data points from the activation, ohmic and mass-transfer limiting regions of polarization curves were distributed as evenly as possible to three data subsets. Five input variables were considered for the modelling; current density I, Temperature T, methanol concentration C, liquid flow-rate q, and gas flow-rate Q. The polarization data of three center replicate experiments were used to assess the experimental error on voltage values. The mean standard deviation was calculated as 9.8 mV, corresponding to percent relative error of 4.1 %.

Table 2. The experimental plan

Run no.	q (ml/min)	Q(ml/min)	C (M)	T (ºC)
1	5	450	1	35
2	5	450	1	55
3	5	450	2.5	35
4	5	450	2.5	55
5	11	450	1	35
6	11	450	1	55
7	11	450	2.5	35
8	11	450	2.5	55
9	5	950	1	35
10	5	950	1	55
11	5	950	2.5	35
12	5	950	2.5	55
13	11	950	1	35
14	11	950	1	55
15	11	950	2.5	35
16	11	950	2.5	55
17	8	700	1.75	25
18	8	700	1.75	65
19	8	700	0.25	45
20	8	700	3.25	45
21	2	700	1.75	45
22	14	700	1.75	45
23	8	200	1.75	45
24	8	1200	1.75	45
25	8	700	1.75	45
26	8	700	1.75	45
27	8	700	1.75	45
28	8	700	1.75	30
29	8	700	1.75	60
30	8	700	0.59	45
31	8	700	2.91	45
32	3	700	1.75	45
33	13	700	1.75	45
34	8	313	1.75	45
35	8	1087	1.75	45

The maximum number of training epochs was set to 300 for both ANN and ANFIS modelling. Meanwhile, all the training stopped early by validation control. The root of the mean sum of squares of the networks errors (RMSE) was used as the termination criteria.

4.1 ANN modelling

A feed-forward ANN architecture with one hidden layer, and one linear neuron in the output layer was used in the modelling study. The input data was scaled between {-1 1} for an efficient training. "Levenberg-Marquardt optimization method was used in the training process. The training process was started from several different initial conditions and the ANN with the minimum RMSE was accepted as the best.

Various subsets of input variables and number of hidden neurons were investigated to determine the optimum ANN structure. On the hidden layer, "tansig" and "logsig" activation functions were comparatively used from performance of view. Generally, tansig function performed better than logsig function. Table 3 gives the cumulative results of RMSE for different input subsets. As seen, by increasing the number of neurons, the generalization performance of ANN increased up to an optimum value and then it decreased due to the over-fitting caused by the excessive number of parameters. Furthermore, during the selection of most relevant input set, the initial examination showed that the base input set (I, T, C) was insufficient to estimate accurately the cell voltage. This is the situation of most empirical models where liquid and gas flow dynamics are usually neglected. By the inclusion of liquid flow rate into input set, the model performance was increased. But, further addition of the air flow-rate, the model performance is not increased as expected; the optimum number of neurons was less but the RMSE value was higher. In conclusion, minimum RMSE was obtained with the input set (I, T, C, q) and twelve hidden neurons; the total number of network parameters (weights and biases) is 73. The minimum value of RMSE is 10.8 mV, very similar to the mean standard deviation of the experimental error (9.8 mV) ; this supports the statistical validity of the model. The model fit is shown in Figure 2.

Table 3. Test RMSE values for various input sets

Number of neuron	Set (I,T,C)	Set (I,T,C,q)	Set (I,T,C,q,Q)
4	20.2	18.9	16.9
6	16.9	15.8	15.0
8	16.3	14.6	15.1
10	16.5	11.4	**11.6**
12	13.8	**10.8**	11.7
14	13.3	15.4	**13.1**
16	17.6	15.8	14.4

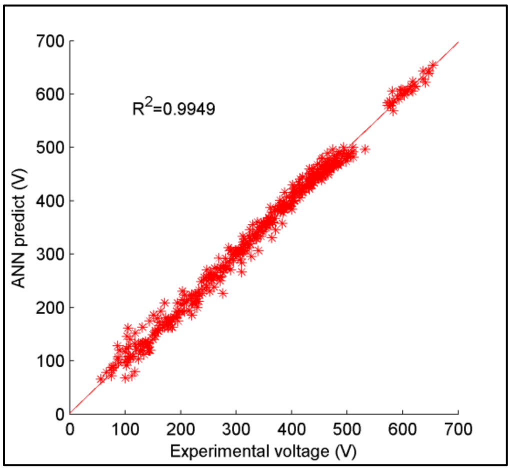

Figure 2. The fit of ANN model with experimental voltage values

4.2 ANFIS modelling

The rule base set was formed by means of “grid partition” method. Two membership functions were defined for each input, thus the number of fuzzy rule was equal to 2^N in which gived also the number of output membership functions. Zero and first order Sugeno inference systems were tried, first order system giving much higher performance.

Various subsets of input variables and various types of transfer functions namely triangular, trapezoidal, gaussian and disigmoidal transfer functions, were tried in the modelling study. RMSE values of various input sets and using different membership functions are given in Table 4. The highest test performance was obtain again with input set (I, T, C, q) and with triangular membership function. The optimum ANFIS architecture with 16 rules and 104 parameters is shown in Fig.3. The fit between model outputs and experimental values is plotted in Fig. 4.

Table 4. Test MSSE values for various input sets and membership function types

	Membership function type			
Input set	Trimf	Trapmf	Gaussmf	Dsigmf
I, T, C	25.2	81.9	65.5	44.2
I, T, C, q	**15.5**	92.4	112.8	43.8
I, T, C, q, Q	21.2	183.0	92.4	49.6

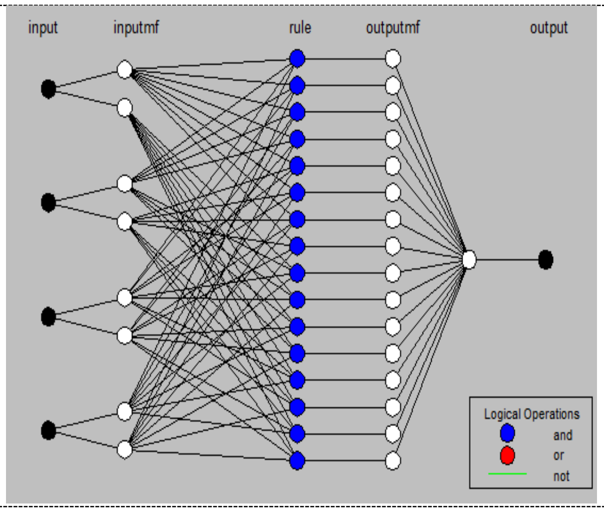

Figure 3. ANFIS structure with 4 inputs and 16 rules

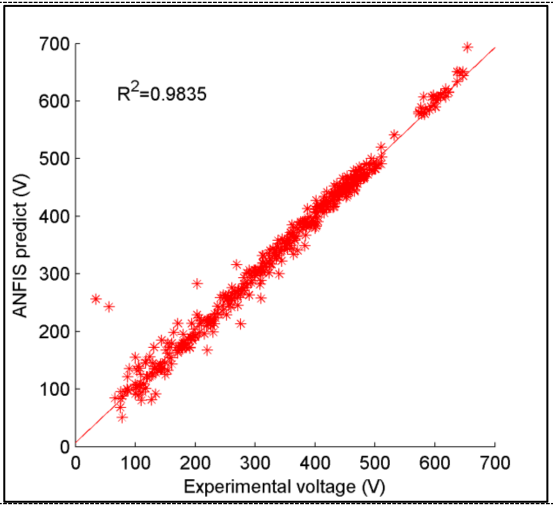

Figure 4. The fit of ANFIS model with experimental voltage values

Finally, when performance comparison between the two modelling techniques was made according to R^2 value of the model fit (0.9949 for ANN against 0.9835 for ANFIS) it was seen that feed-forward ANN model predicted more successfully the voltage values of DMFC. Simulated I-V curve shown in Fig. 5a and the corresponding power curve depicted in Fig. 5b approximate very well the experimental values.

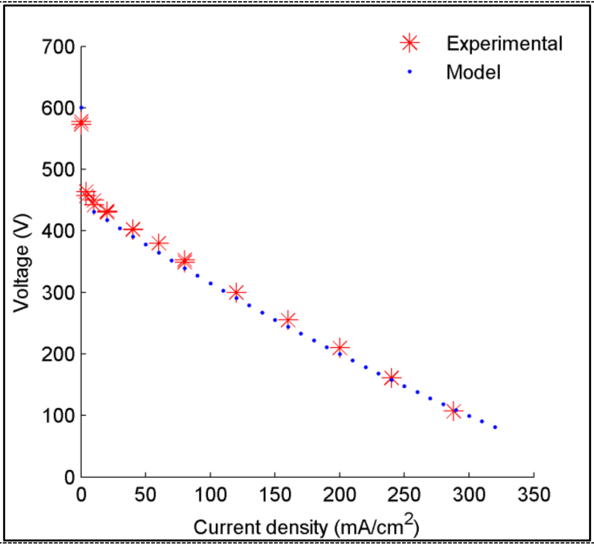

Figure 5-a. ANN model simulation I-V curves

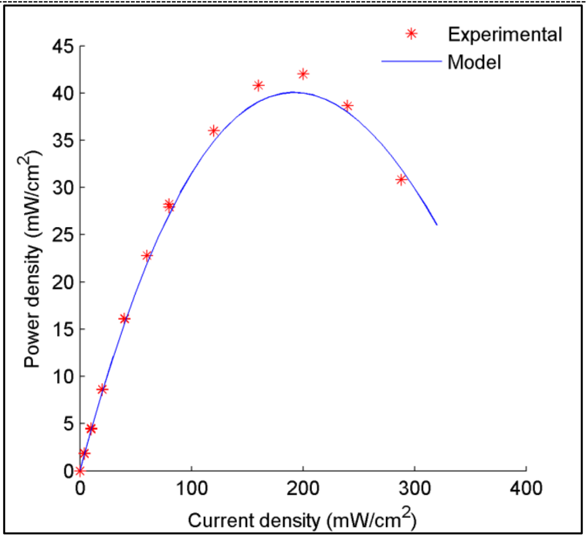

Figure 5-b. ANN model simulation I-W curves (Operating conditions: T = 55 ºC, C = 2.5 M, q = 11 ml/ min, Q = 950 ml/min)

5.0 Conclusion

In both modelling approaches, in addition to principal inputs (current density, temperature, methanol concentration) liquid flow rate was detected as an effective input influencing the cell voltage; liquid dynamics affects the mass transfer rates in flow channels through the backing and diffusional rates to catalyst particles surfaces. Meanwhile, fuel cell voltage is not so sensitive to air flow rate. For the optimum feed-forward ANN model with twelve hidden tansig neurons, minimum test RMSE was found as 10.8 mV which is the estimate of the mean standard deviation of the experimental error (9.8 mV); this result supports further the statistical validity of the model. In the case of ANFIS model, the minimum RMSE value (15.5 mV) was obtained with an architecture using triangular membership function and 16 fuzzy rules. Finally, further statistical tests demonstrated that feed-forward ANN model predicted more successfully the voltage values of DMFC.

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