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# Control Applications (ANFIS/Fuzzy/PID) over Mathematical Model of DMFC System: Experimental and Simulation Studies

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Environmental pollution, depletion of fossil fuel reserves and energy needs of a growing world population, are among the important issues concerning the future of energy. In this context, fuel cells are considered as alternative energy sources. In this study, one cell direct methanol fuel cell system, consisting of various sensors and control elements and controlling system are built. Experimental temperature control is conducted with fuzzy control technique over the systems. Then, useful and simple mathematical model that can adequately represent the system is established and the parameters of this model are determined by particle swarm optimization (PSO) method using experimental data. In addition, ANFIS model of the system with the 4-input and 1 output is established through the polarization curves. Also, membership function types and numbers of input variables having an impact on the ANFIS modeling are investigated. Beside to the experimental PID concentration control with sensor, ANFIS is tested for the sensor-less concentration control over the mathematical model in the simulation studies.

**Keywords:** Control; ANFIS; Fuzzy; PID; DMFC model

## 1. INTRODUCTION

Fuel cells are green power source that converts chemical energy direct into electricity. They are widely researched worldwide by many researchers due to a lot of advantages. They are clean, silent, efficient energy conversion systems. Besides, direct methanol fuel cells, (DMFCs) which are considered especially for portable electronic devices, have some superiority such as, low operating temperature, rapid start up, high energy density, no need for a reformer [1-3]. There are many operating characteristics including methanol concentration, reactants flow rates, cell temperature, pressure etc. that affects the performance of the DMFC system, but undoubtedly one of the most important of which is methanol concentration. The methanol concentration in the anode side must be

well adjusted due to concentration decayed (in a closed loop flow) and methanol crossover from the membrane which reduces cell power and fuel efficiency. The simply way to control the methanol concentration is to use concentration sensor. But cost, weight and volume of system is an important factor for commercialization. Also, these sensors based either chemical or physical phenomena should have many requirements. Sensor-less concentration control is another way to handle this task. In the last decade, a few sensor-less methanol concentration control algorithms have been published. Chen et al. [3] published an algorithm that utilized the cell current and voltage values, they fed a specific amount of methanol in a mixing tank, and they measured the characteristic values (V, $i_{cell}$) at the end of the determined specific monitoring period and before the end of the monitoring period, and then they took a new step by comparing these values. Chen et al. [4] modified their previous IR-DTFI algorithm and shortened monitoring period they publish another algorithm ‘’IR- CIDTFI ‘’and they evaluate algorithm performance over 40w portable DMFC system over the operating characteristics [5]. Ha et al. [6] build a control logic that can predict the rate of methanol consumption; firstly they carried out a set of experiment for specific concentrations at different operating conditions by varying temperature and current, and developed a relevant equation to correlate the total consumption rate of methanol. In the study of Arisetty et al. [7] they used bisection optimization algorithm, by starting from 2M concentration they searched optimal value by comparing voltage response. Lian and Yang [8], followed such a way to control the concentration, once they built empirical equation to determine ideal output voltage, they feed a specific amount of methanol in to the mixing tank and monitored loading and voltage, and eventually they compared them and took action according to load and voltage response. In the next study of Lian and Yang [9] they changed the previous algorithm a little and instead of determining the ideal output voltage by empirical equation they use reference voltage value experimentally detected. Recently Chang et al. [10] used Artificial Neuro Fuzzy Inference System (ANFIS) model to predict the needed amount of methanol to control the system. They handled dosing pump active time by means of voltage and current measurements. An et al. [11] recently proposed new algorithm, once algorithm predict the methanol consumption rate by early used equation [6] they used it for maintaining the stack temperature at a determined level which has close correlation with concentration. After that they modified their algorithm and they tested it at varying ambient temperatures [12]. In this study, firstly experimental setup was installed including several sensors and control elements and, a software has a user interface was launched based on Labview. Then a mathematical model was established that unknown parameters were determined by using experimental data with the particle swarm optimization method. Thus model was ensured to represent the system in a good manner in the simulation study. And studies were performed to establish the ANFIS model to control the methanol concentration in various operating conditions at varying temperatures, methanol flow rates, concentration and current density. Finally an ANFIS control algorithm was offered to control the concentration, over the mathematical model, causing drop in cell voltage. In our future study we are intended to accomplish this algorithm in real time system.

## 2. IMPORTANCE OF METHANOL CONCENTRATION

Although there are many impressive operating characteristic that affect the direct methanol fuel cell including temperature, reactant flow rates and pressures; the methanol concentration has a decisive impact on performance and efficiency of the DMFC. Currently, methanol crossover is one of the critical unsolved DMFC problems, which has undesired results on the performance and efficiency of the DMFC system. Firstly, permeated methanol that reacted at the cathode side reduces the fuel efficiency, and cause mixed potential by consuming the free electrons at the cathode side. Besides, permeation of methanol causes wasting cathode catalyst active sites and poisoning by the carbon atoms in the methanol. The methanol crossover intensely depends on methanol concentration at the anode compartment. And the methanol concentration at the anode compartment depends on current density can be drawn.

In addition to the crossover problem, the unreacted methanol which is returned back to the methanol reservoir also decreases the concentration in the tank. It is clear above statements that, for each current density an optimal concentration appears and the need to control the methanol concentration in adequate manner arises [3, 13]. Related schematic diagram is illustrated in Fig. 1.

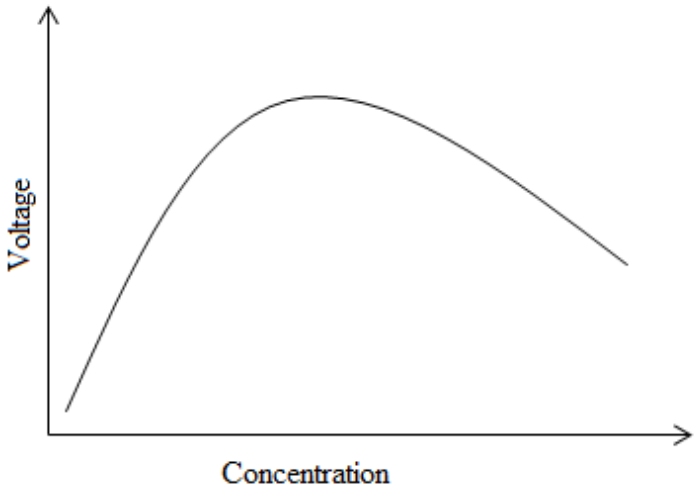


**Figure 1.** Schematic visualization of concentration dependence of voltage at fixed current density

The first way comes to mind to control the methanol concentration is to utilize a sensor. However, sensor to be selected should fulfill several requirements. Namely, sensors based on electrochemical principles may lead to degradation of MEA, also sensors based on sound speed, density or refractometry may create problem due to their sensitivity to the carbon dioxide bubbles, these sensors may also be affected to the pulse signal sending to the pumps. Further, several important factor such as; temperature dependence, weight, size, complexity, cost, calibration task should be considered in the use of these sensors. As a second alternative to control the methanol concentration, which is a prominent issue in recent years, is sensor-less methanol concentration control [14].

## 3. ANFIS STRUCTURE

ANFIS is a kind of artificial neural network based on the Takagi - Sugeno type fuzzy logic inference systems. Artificial neural network is used to determine the optimal values of model parameters of fuzzy inference system. Fuzzy inference system is gained an adaptive feature by using data sets for training the artificial neural network. In training the neural network, a hybrid algorithm containing widely used back-propagation learning method and the least squares method is used [15].

*Adaptive Neuro Fuzzy Inference Systems (ANFIS):*

As it is known there are certain disadvantages of the fuzzy logic system;

- The necessity of determining the rule base, this process is usually solved by referring an expert knowledge. This is a time consuming process fraught with problems.
- Clearly, it is need to determine the membership functions necessary for the formation of fuzzy sets. For example, If Gaussian membership functions were chosen, and then what would be the parameters of the Gaussian membership function?

For this purpose ANFIS allows to determine the rule base and the membership functions by using the data sets.

ANFIS has an adaptive network structure consisting of nodes and bonds between these nodes. It is adaptive because some or all of the nodes have influence on the end nodes. The relationship between input and output nodes are learned using an algorithm. The basic structure of a sample ANFIS, which has two inputs and one output, is shown in Fig. 2.

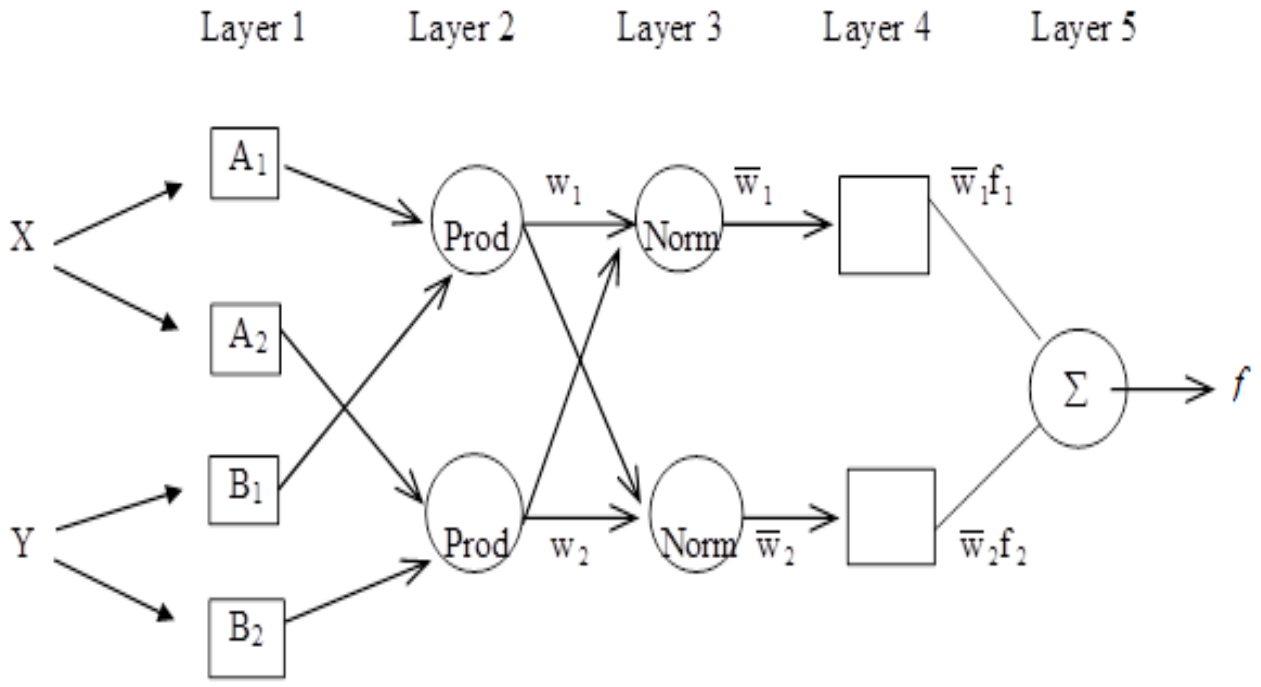


**Figure 2.** Structures of Sugeno type ANFIS system with two inputs and one output

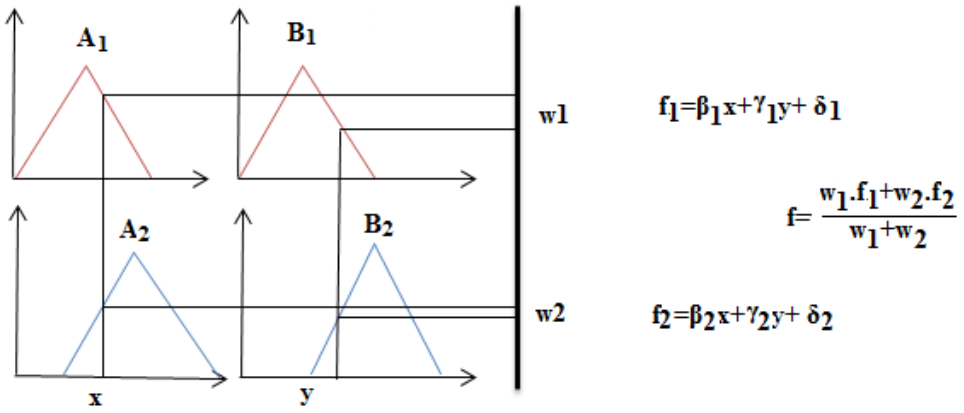


**Figure 3.** Sugeno type fuzzy sets with two inputs (membership functions)

The rules are as the form below in Sugeno type ANFIS system with two inputs is given in Fig. 3.

*If* $x$ *is* $A_1$ *and* $y$ *is* $B_1$ *THEN* $f_1 = \beta_1 x + \gamma_1 y + \delta_1$

*If* $x$ *is* $A_2$ *and* $y$ *is* $B_2$ *THEN* $f_2 = \beta_2 x + \gamma_2 y + \delta_2$

The main steps are listed below applied for training of the network.

*Layer 1.*

The output of the each node in the layer is calculated from the Eqs. (1)-(7).

$$O_{1,i} = \mu_{A_i}(x) \qquad for\ i = 1,2 \tag{1}$$

$$O_{1,i} = \mu_{B_{i-2}}(y) \qquad for\ i = 3,4 \tag{2}$$

Here, $O_{1,i}(x)$ indicates the membership degree of X and Y (linguistic variables)

Any of a variety of functions can be chosen as a membership functions (Gaussian, triangular, trapezoidal, etc.), but here Gaussian function is used for demonstration purposes. The general expression of Gaussian function is below.

$$\mu_A(x) = \frac{1}{1 + \left| \frac{x - \lambda_i}{\varepsilon_i} \right|^{2\sigma_i}} \tag{3}$$

Here, $\varepsilon_i, \sigma_i, \lambda_i$ are the parameters to be learned of the antecedent of the rule

*Layer 2*

In this layer determining the weight of the membership function is carried out, usually 'AND' norm is used.

$$O_{2,i} = w_i = \mu_{A_i}(x)\mu_{B_i}(y), \quad i = 1,2 \tag{4}$$

*Layer 3*

The average weight of each rule is determined in this layer.

$$O_{3,i} = \overline{w}_i = \frac{w_i}{w_1 + w_2} \tag{5}$$

*Layer 4*

In this layer consequent part of the rules is multiplied the weighted average to obtain weighted rule outputs.

$$O_{4,i} = \overline{w_i} f_i = \overline{w_i}(\beta_i x + \gamma_i y + \delta_i) \quad (6)$$

Here, the each output has parameters should be determined ( $\beta_i, \gamma_i, \delta_i$ )

*Layer 5*

In the last layer, weighted input variables are collected to be able to obtain the corresponding output.

$$O_{5,i} = \sum_i \overline{w_i} f_i = \frac{\sum_i w_i f_i}{\sum_i w_i} \quad (7)$$

After this stage, the parameters belong to the membership function that constitute antecedent and consequent of the rules is need to be determined.

There are many approaches to accomplish the determination process. But commonly used one is the hybrid learning algorithm (least squares and gradient descent), this stage include heavy computational effort.

All parameters can be gathered in a general group as follow.

$S$ = Total parameter set

$S_1$ = Antecedent parameter set

$S_2$ = Consequent parameter set

As mentioned before ANFIS uses two learning algorithm, both forward and backward propagation

- In the forward pass. $S_1$ is kept constant and $S_2$ is obtained according to least squares method.
- In the backward pass. $S_2$ is kept constant and $S_1$ is obtained via the gradient descent method.

## 4. MATHEMATICAL MODELING

Analytical models are effective approaches used to predict the system behavior. Considering DMFC, multiple input and output structure turns into system into a black box that modeling hard. In the literature 1, 2 or 3-dimensional analytical model, which using different kinetic and transport approaches, can be found. However, many of them typically contain complex non-linear relationships. Here, the aim is not to build/use a complex model to represent systems perfectly, is to choose a simple and useful model that can represent the system sufficiently. Therefor selected model to be used here is taken from the study of Sundmacher et al. [16].

Kinetics and transport phenomena that take into account in the mathematical model are given below;

Convective transport of methanol in the anode side

Methanol electrochemical oxidation

Oxygen electrochemical reduction

Mass transfer of methanol and carbon dioxide at the anode diffusion layer

Crossover of methanol driven by Electro-osmosis, diffusion and pressure gradient from the membrane

Electrochemical methanol oxidation at the cathode side

A schematic diagram is illustrated in Fig. 4.

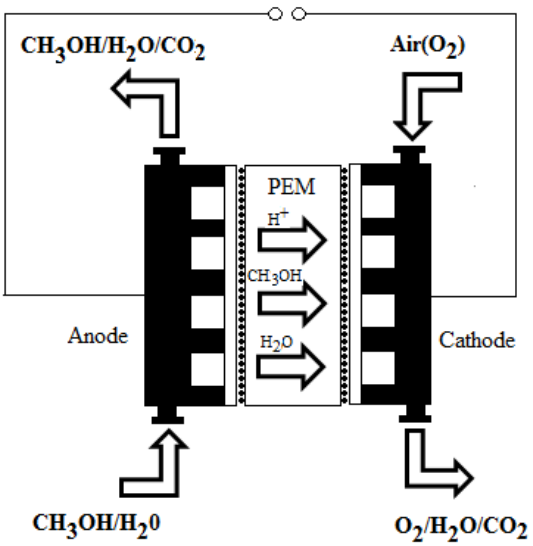


**Figure 4.** Schematic operation diagram of DMFC

The multi-step reaction mechanism for methanol electro-oxidation on Pt, suggested from McNichol [17] is summarized follows (R1)-(R5) reactions.

$$3Pt + CH_3OH \longleftrightarrow Pt_3 — COH + 3H^+ + 3e^- \quad (R1)$$

$$3Ru + 3H_2O \longleftrightarrow 3Ru - OH + 3H^+ + 3e^- \quad (R2)$$

$$Pt_3 — COH + 2Ru - OH \longleftrightarrow Pt — COOH + H_2O + 2Pt + 2Ru \quad (R3)$$

$$Pt — COOH + Ru — OH \longleftrightarrow CO_2 + H_2O + Pt + Ru \quad (R4)$$

$$3/2O_2 + 6H^+ + 6e^- \longleftrightarrow 3H_2O \quad (R5)$$

When installing the Mathematical model the following assumptions were made.

- Isothermal Fuel cell operation
- Single-Phase system, one dimensional heat and mass transfer
- Transfer coefficient of Methanol and carbon dioxide at the anode side are equivalent
- Carbon ad oxygen molecules do not diffuse into the membrane
- Ohmic losses in the current collector and the electrical connections are negligible
- Mass transfer resistance in catalyst layer can be negligible due to lower compared to the diffusion layer
- Excess oxygen supply (hence no need for oxygen balance)

The expression of reaction rates according to the Butler-Volmer rate expression are given with Eqs. (8)-(12). As a result, mole balances written according to the concentration at the catalyst surface, dissociative chemisorption of methanol is taken as rate-determining step.

$$r_1 = k_1 \exp(\frac{\alpha_1 F}{RT}\eta_a)\{\Theta_{Pt}^3 c_{CH_3OH}^{CL} - \frac{1}{K_1}\exp(-\frac{F}{RT}\eta_a)\Theta_{Pt_3-COH}\} \quad \text{rate-determining step} \quad (8)$$

$$r_2 = k_2 \exp(\frac{\alpha_2 F}{RT}\eta_a)\{\Theta_{Ru} - \frac{1}{K_2}\exp(-\frac{F}{RT}\eta_a)\Theta_{Ru-OH}\} \quad (9)$$

$$r_3 = k_3\{\Theta_{Pt_3—\mathrm{COH}}\Theta_{Ru\text{-}\mathrm{OH}}^2 - \frac{1}{K_3}\Theta_{Pt\text{-}\mathrm{COOH}}\Theta_{Pt}^2\Theta_{Ru}^2\} \quad (10)$$

$$r_4 = k_4\{\Theta_{Pt\text{-}\mathrm{COOH}}\Theta_{Ru\text{-}\mathrm{OH}} - \frac{1}{K_4}c_{CO_2}^{CL}\Theta_{Pt}\Theta_{Ru}\} \quad (11)$$

$$r_5 = k_5 \exp(\frac{\alpha_5 F}{RT}\eta_c)\{1 - \exp(-\frac{F}{RT}\eta_c)(\frac{p_{02}}{p^\theta})^{3/2}\} \quad (12)$$

Molar balances Eqs. (13)-(16) and charge balance Eqs. (17)-(18) are as follows.

*Anode compartment:*

$$\frac{dc_{CH_3OH}}{dt} = \frac{1}{\tau}(c_{CH_3OH}^F - c_{CH_3OH}) - \frac{k^{LS}A^s}{V_a}(c_{CH_3OH} - c_{CH_3OH}^{CL}) \quad (13)$$

*Anode catalyst compartment:*

$$\frac{dc_{CH_3OH}^{CL}}{dt} = \frac{k^{LS}A^s}{V_a^{CL}}(c_{CH_3OH} - c_{CH_3OH}^{CL}) - \frac{A^s}{V_a^{CL}}n_{CH_3OH}^M - \frac{A^s}{V_a^{CL}}r_1 \quad (14)$$

$$\frac{dc_{co_2}}{dt} = \frac{1}{\tau}(c_{co_2}^F - c_{co_2}) - \frac{k^{LS}A^s}{V_a}(c_{co_2} - c_{co_2}^{CL}) \quad (15)$$

$$\frac{dc_{co_2}^{CL}}{dt} = \frac{k^{LS}A^s}{V_a^{CL}}(c_{co_2} - c_{co_2}^{CL}) + \frac{A^s}{V_a^{CL}}r_1 \quad (16)$$

$$\frac{d\eta_a}{dt} = \frac{1}{C_a}(i_{cell} - 6Fr_1) \quad (17)$$

*Cathode catalyst compartment:*

$$\frac{d\eta_c}{dt} = \frac{1}{C_c}(-i_{cell} - 6F(r_5 + n_{CH_3OH}^M)) \quad (18)$$

Mass flux of the methanol through the membrane is expressed in Eq. (19),

$$n_{CH_3OH}^M = \frac{D_{CH_3OH}^M}{d^M}c_{CH_3OH}^{CL}\frac{Pe\ \exp(Pe)}{\exp(Pe)-1} \quad (19)$$

Equation (20) gives the Peclet number, Pe is;

$$Pe \equiv \frac{vd^M}{D_{CH_3OH}^M} \quad (20)$$

As a result, the open circuit voltage consisting ohmic losses and the sum of the over potentials is expressed in Eq. (21) as follows.

$$U_{cell} = U_{cell}^{\theta} - \eta_a + \eta_c - \frac{d^M}{\kappa^M} i_{cell} \quad (21)$$

For detailed information and data related the model can be accessible relevant studies [16-19].

## 5. EXPERIMENTAL

### *5.1 Experimental Setup*

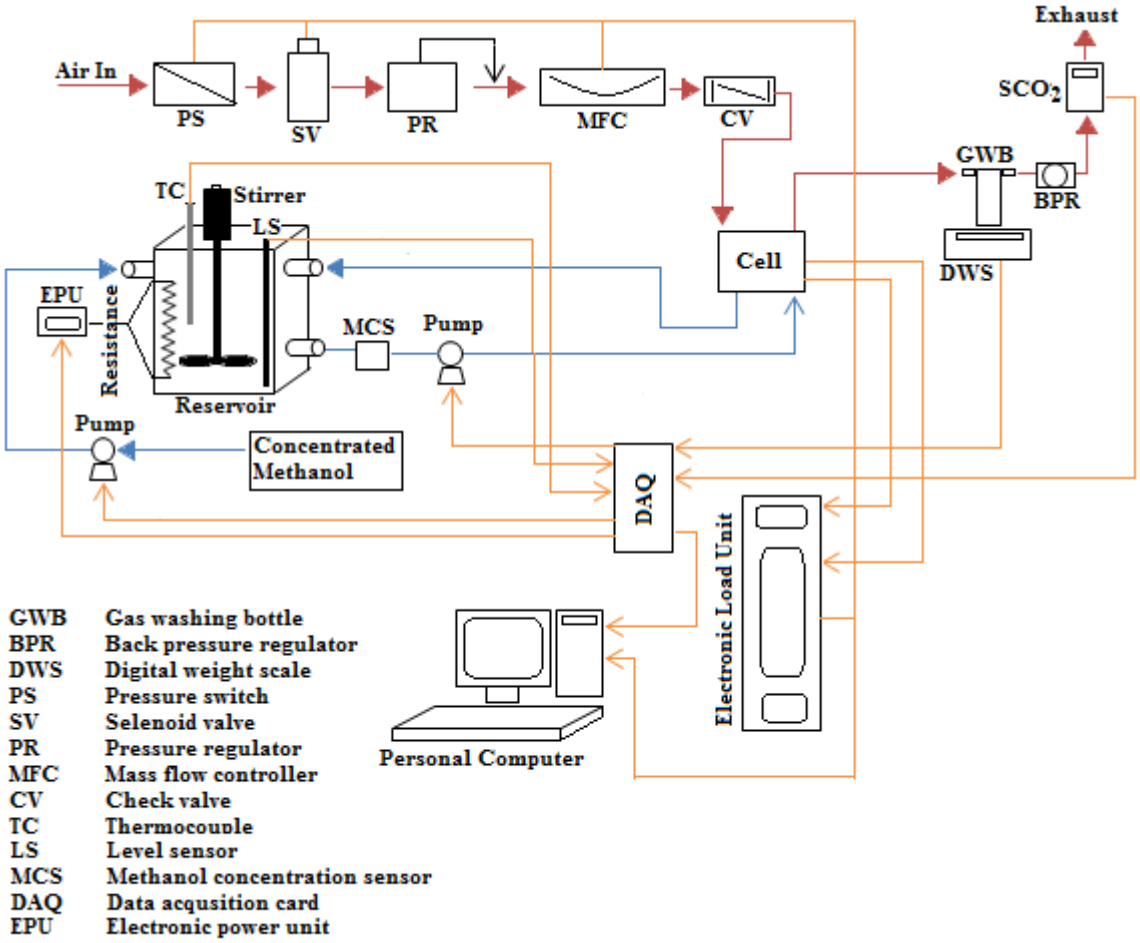


**Figure 5.** Flow chart of the DMFC system

Flowchart of the experimental system is as shown in Fig. 5. Sensors and final control elements were connected to the computer via the PCI 6221 DAQ board (data acquisition) of the National Instrument firm. And electronic load unit was connected via RS-232 (Serial port) to personal computer. Also as seen here, concentrated methanol is fed to the methanol reservoir to keep the methanol concentration at a certain level. In order to online monitor the volts, current, power, temperature and level information, and control the process elements such as pumps, heaters, mixers, etc. a Labview program was programmed. A screenshot of the program can be seen from Fig. 6.

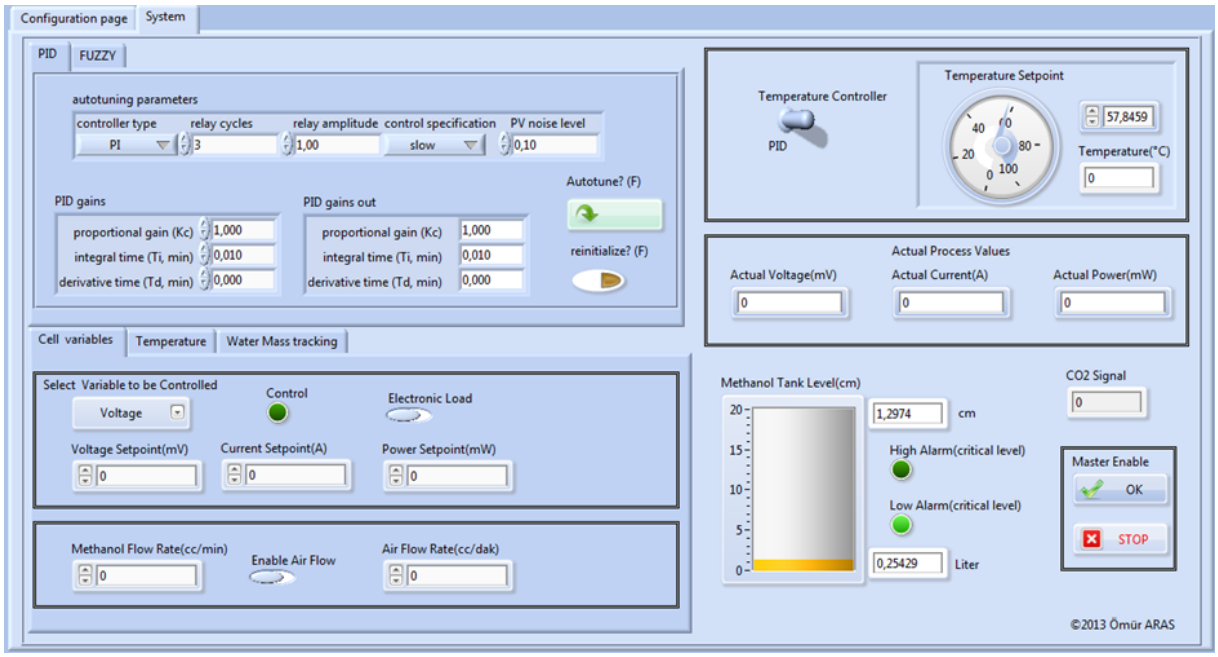


**Figure 6.** A screenshot from the interface of the developed DMFC software

*5.2 Temperature Control*

Also it is required to control the temperature adequately at any point, which unquestionably impacts on system performance. For this, Fuzzy Logic controller was applied to the system by varying the set point stepwise. While the system was initially at room temperature, set point was changed respectively, 35, 45, 55 and 65 °C and the system response was analyzed.

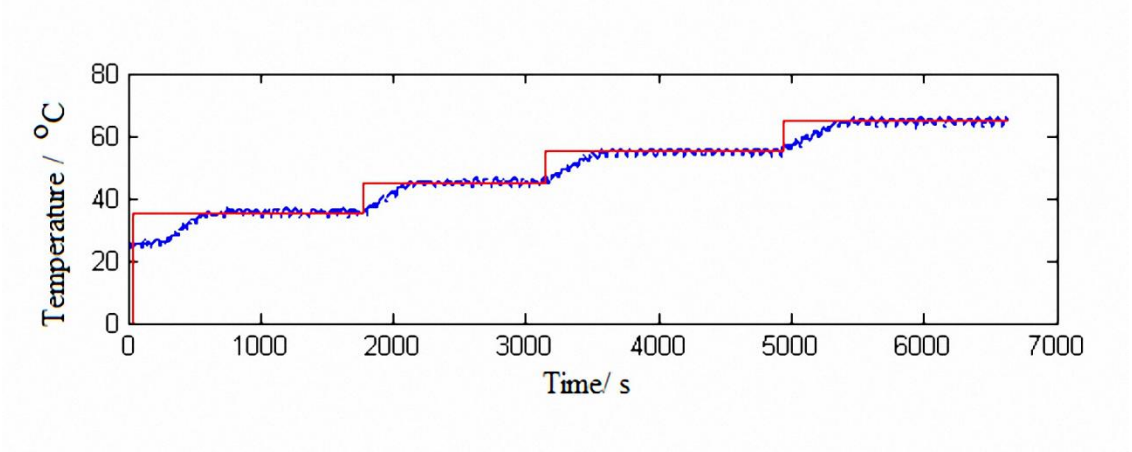


**Figure 7.** Reservoir temperature control with Fuzzy Logic Controller

As can be seen from the controller trial result in Fig. 7 the reservoir temperature value was kept in desired value with good performances by means of fuzzy controller.

*5.3 Methanol Concentration Control with Sensor*

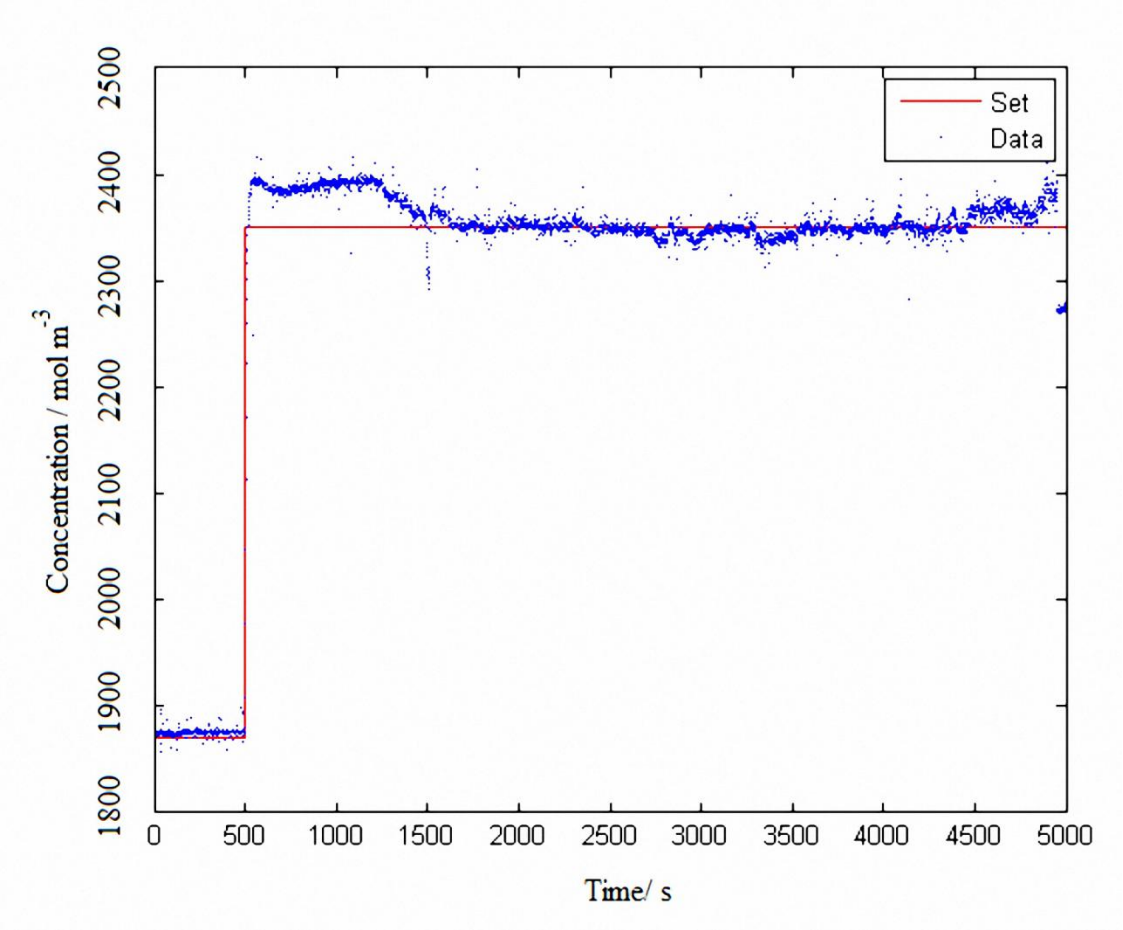


**Figure 8.** Control application with methanol concentration sensor

We need to control the methanol concentration to collect the data at a fixed concentration. Thus with the aid of density based Issys MicroCMS® sensor adapted to the system, PID control was applied to control the methanol concentration. In accordance with the servo control logic, while system at a certain concentration (1.87 Molar) set point was changed to 2.35 molar.

The graphics of the experiment are given in Fig. 8. Concentration reached the set point band after an overshoot and was tried to keep at the set point. Meanwhile high noise can be easily seen from the graph in the measurement.

## 6. RESULTS AND DISCUSSION

*6.1 Determination of Mathematical Model Parameters*

To accomplish the simulation studies the unknown model parameters; $k_1$, $k_2$, $k_3$, $k_4$, $k_5$, $K_1$, $K_2$, $K_3$, $K_4$ and $k^{LS}$ should be determined. Since $k_2$, $k_3$ and $k_4$ have no impact on the model (reaction rate

goes to zero) [16], they extracted from the list. It was benefited from the experimental data to find the other unknown parameters.

Dynamic models was run out in a specific (adequate) time to ensure the system became steady state for every current point in the polarization curve, and then the sum of difference between the experimental data and the steady state values was optimized by particle swarm optimization(PSO) method to find the unknown parameters.

Optimum model parameters including rate constants and equilibrium constants maintaining via PSO are below.

*Optimal parameters:*

$k_1$: 0.0221 $mol(m^2 s)$

$k_2$: 7.666e-06 $mol(m^2 s)$

$K_1$: 0.0152 $K_2$: 0.001

$K_3$: 0.688 $K_4$: 0.0010

Also optimum $k^{LS}$ values obtained for each concentration are shown in Fig. 9.

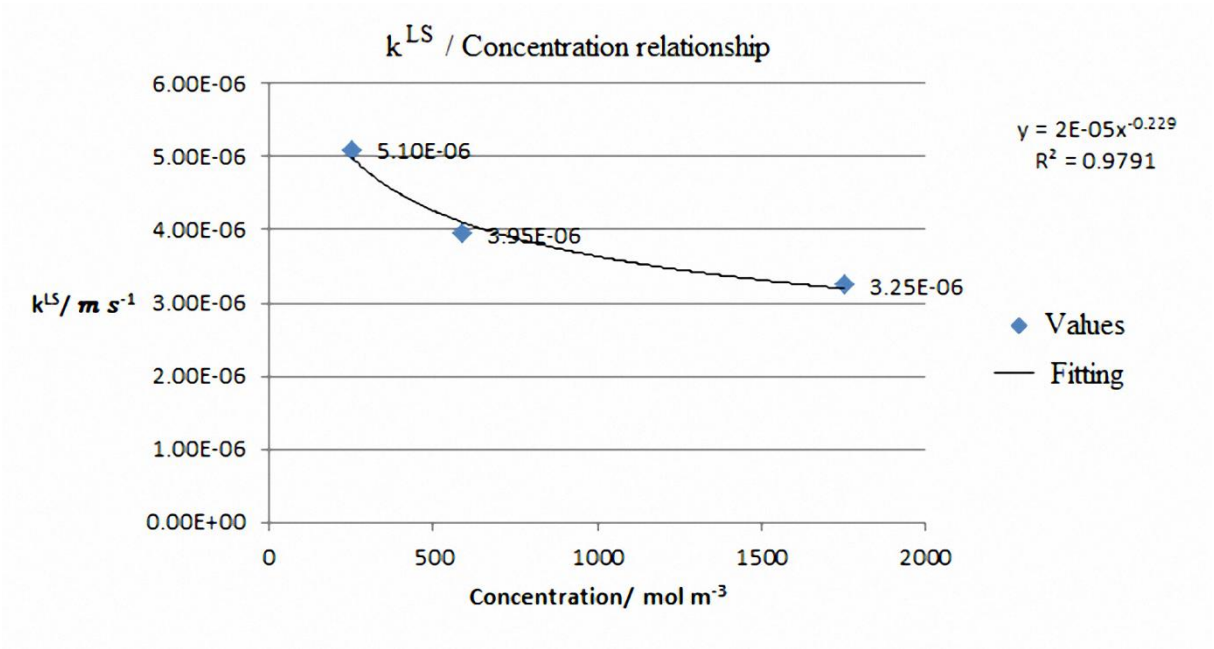


**Figure 9.** Relationship between the Concentration and mass convection coefficient

To test the mathematical model experimental studies conducted at 3 different concentrations value (0.25, 0.59, and 1.75 molar) were used here. Other experimental conditions of the experiments are shown below.

Methanol flow rate (q) = 8 $cc\ min^{-1}$

Temperature (T) = 45 °C

Air flow rate (Q) = 400 $cc\ min^{-1}$

Fitting graphs between the model result and experimental data are given in Fig. 10, Fig. 11 and Fig. 12.

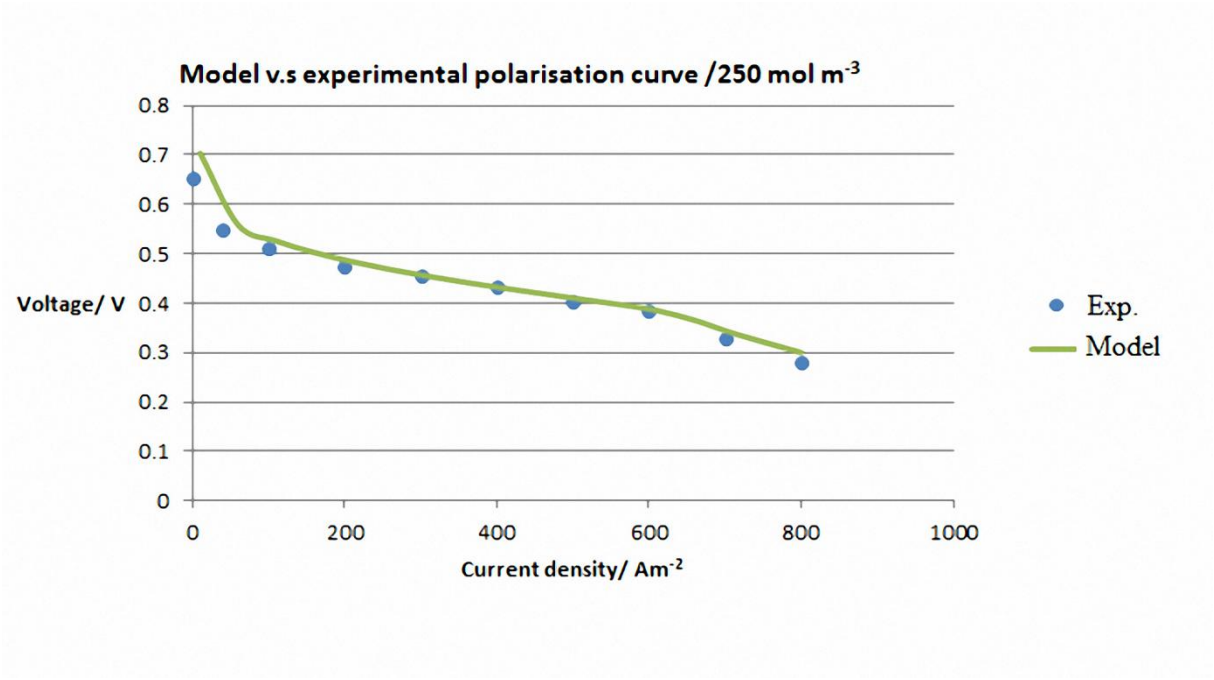


**Figure 10.** Model fitting with the experimental data at 250 mol $m^{-3}$

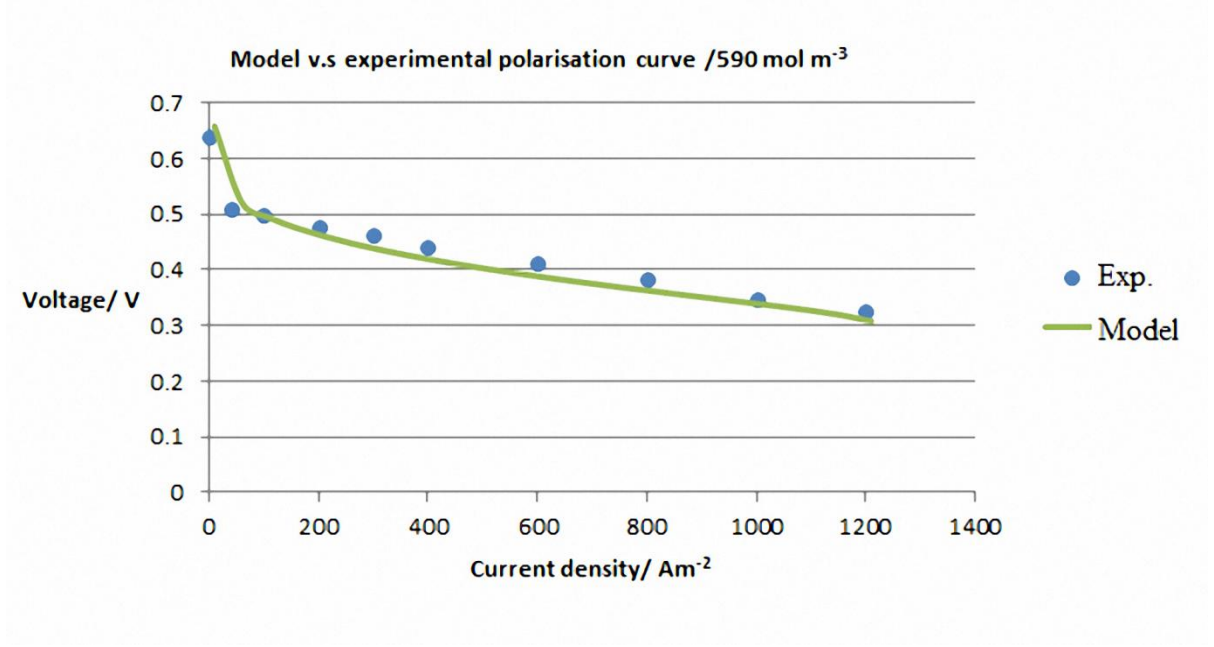


**Figure 11.** Model fitting with the experimental data at 590 mol $m^{-3}$

The model which parameters were optimized fits enough with the experimental data in almost every region of the polarization curve as can be seen from related figures. The experimental data are well compiled to the model for open-circuit voltage, the limiting current and ohmic losses values. Therefore it now could be used in simulation studies.

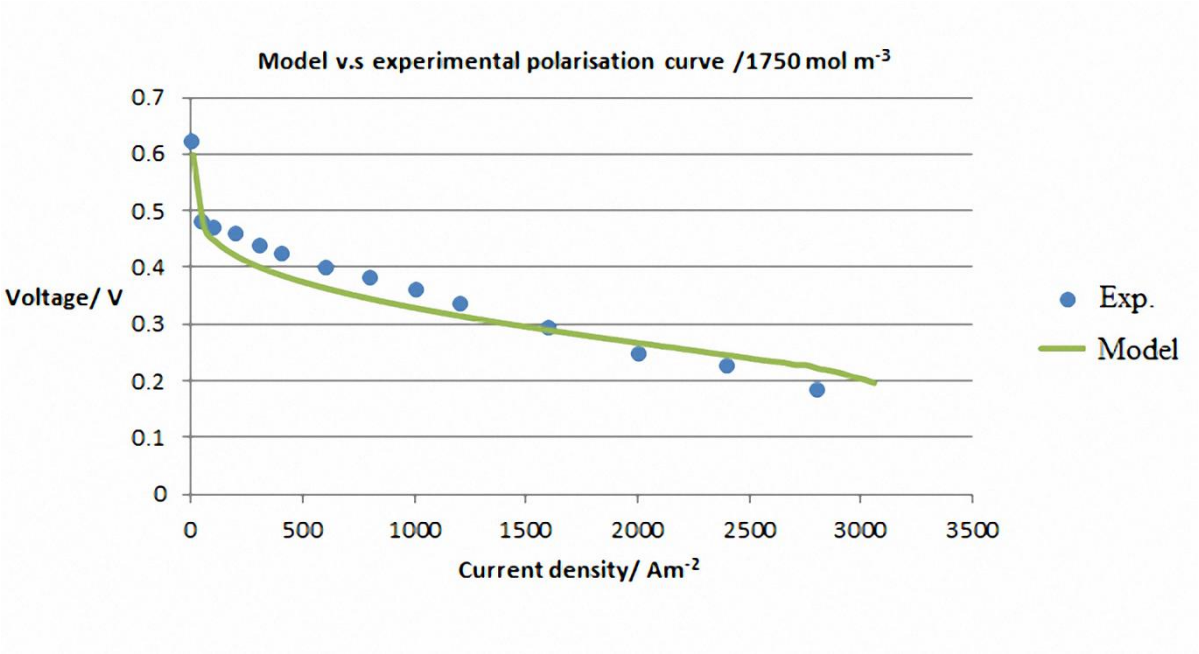


**Figure 12.** Model fitting with the experimental data at 1750 mol $m^{-3}$

*6.2 Training ANFIS*

In this section polarization curves obtained with the MEA purchased from Fuelcellstore © were used for ANFIS modeling, which having the following characteristics.

Membrane type: Nafion 117

Anode catalyst: 4 mg $cm^{-2}$ 1:1 Pt-Ru

Cathode catalyst: 2 mg $cm^{-2}$ Pt black

Gas diffusion layer: Carbon cloth

Respectively methanol flow rate (q), methanol concentration (c), the cell temperature (T) and current density (I) was taken as the input variables, corresponding cell voltage (V) is modeled as output variable in ANFIS. The experimental results handled with data in the following ranges were used for ANFIS modeling study in Matlab.

| | |
|---|---|
| Liquid Flow Rate (q) | 2-14 cc $min^{-1}$ |
| Methanol Concentration (c) | 0.25-3.25M |
| Temperature (T) | 25 to 65 ° C |
| Current density (I) | depends on the operating conditions |

As membership function widely used triangular (trimf) and Gaussian (gaussmf) functions tested also the number of membership functions (2 or 3) is changed to obtain the best result. Actually, data to be entered in ANFIS are also important factor.

In total 450 data, 60% of it for training, 20% of it to control and 20% of it were used to test the ANFIS model. However, how these data are entered to ANFIS? The first 270 of the data obtained in a specific methodology (systematic) (60%) for training, the next 90 (20%) for the tests and remaining for the control. Is this the right approach? The fact is that, in this case the results depend on the data set are

obtained. Consequently, in order to get stable results and to eliminate this dependence in any case, the data sets were randomized with the aid of random number generator; this procedure was performed 20 times for each attempt. Meanwhile, to get result from each attempt in the same condition, number generator was reset after 20 randomizing. This minimum, maximum and average results obtained from this 20 trials, are given in Tables 1 and 2.

**Table 1.** Test results handled with trimf function (different membership function number)

| | Trimf2222 | | | Trimf3333 | | |
|---|---|---|---|---|---|---|
| | Min | Max | Mean | Min | Max | Mean |
| Training MSSE | 13.60 | 127.98 | 28.02 | 9.,34 | 119.87 | 44.28 |
| Control MSSE | 16.12 | 800.64 | 196.42 | 16.66 | 807.47 | 307.33 |
| Test MSSE | 16.92 | 779.36 | 251.79 | 32.47 | 594.52 | 268.14 |
| | Trimf3222 | | | Trimf2322 | | |
| | Min | Max | Mean | Min | Max | Mean |
| Training MSSE | 12.33 | 145.99 | 28.86 | 11.59 | 124.59 | 26.62 |
| Control MSSE | 14.68 | 536.39 | 177.78 | 15.56 | 1013.01 | 176.75 |
| Test MSSE | 14.76 | 731.11 | 210.40 | 14.09 | 614.70 | 218.63 |
| | Trimf2232 | | | Trimf2223 | | |
| | Min | Max | Mean | Min | Max | Mean |
| Training MSSE | 11.49 | 144.54 | 28.11 | 12.10 | 15.73 | 14.31 |
| Control MSSE | 14.36 | 627.03 | 202.34 | 16.37 | 975.32 | 220.61 |
| Test MSSE | 16.32 | 602.17 | 199.01 | 17.70 | 847.76 | 236.69 |

**Table 2.** Test results handled with gaussmf function (different membership function number)

| | Gaussmf2222 | | | Gaussmf3333 | | |
|---|---|---|---|---|---|---|
| | Min | Max | Mean | Min | Max | Mean |
| Training MSSE | 29.04 | 33.71 | 30.56 | 24.50 | 257.99 | 51.63 |
| Control MSSE | 33.8 | 240.26 | 72.74 | 44.95 | 653.60 | 185.03 |
| Test MSSE | 37.11 | 155.01 | 57.76 | 56.43 | 4350.51 | 844.81 |
| | Gausmf3222 | | | Gausmf2322 | | |
| | Min | Max | Mean | Min | Max | Mean |
| Training MSSE | 27.68 | 33.01 | 29.71 | 28.77 | 33.44 | 30.50 |
| Control MSSE | 33.68 | 1248.32 | 190.67 | 32.99 | 283.69 | 89.53 |
| Test MSSE | 37.76 | 145.15 | 57.06 | 36.08 | 192.26 | 65.45 |
| | Gausmf2232 | | | Gausmf2223 | | |
| | Min | Max | Mean | Min | Max | Mean |
| Training MSSE | 28.43 | 32.90 | 29.92 | 25.87 | 29.73 | 27.32 |
| Control MSSE | 33.25 | 1289.01 | 210.97 | 34.69 | 167.15 | 71.75 |
| Test MSSE | 38.32 | 148.29 | 60.76 | 35.96 | 276.87 | 78.83 |

These results also were obtained with the criteria of root mean square errors (MSSA).

As can be seen from Table 1, trimf function was used for the input variables and membership functions numbers written beside it. For example trimf2232; trimf is membership function and the numbers of membership function were taken as q: 2, c: 2, T: 3 and I: 2. The same situation is valid for Table 2, gaussian was selected as the membership functions and the numbers of membership function were changed.

It is apparent from Table 1 and 2 that when trimf function is used, all minimal values are lower than the gaussian results. But when the data are changed for training, test and control i.e. different data were used in ANFIS, gaussmf function is better on average.

Best results, both trimf and gaussmf type membership functions when used, were obtained in the case where the number of membership functions was taken as '3-2-2-2', i.e. this results were obtained when the number of membership functions for the methanol flow 'q', was taken as 3.

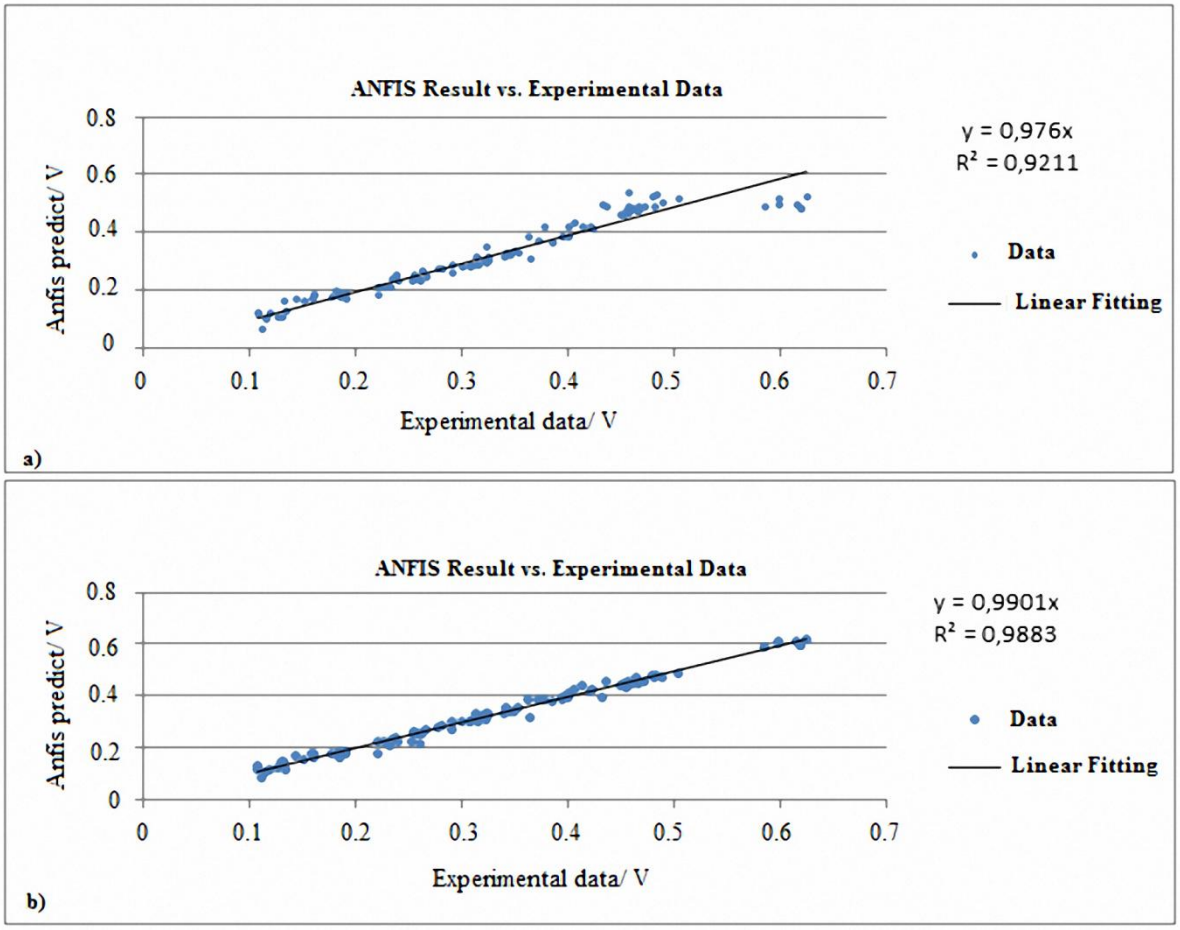


**Figure 13.** ANFIS-experimental data fitting graphs for a) gaussmf b) Trimf

Latest, a graph was plotted to see the fit results of experimental data with the ANFIS. In the sense that higher $r^2$ and slope close to 1, ANFIS results, as can be seen from Fig. 13, provides further compliance with the trimf function.

*6.3 Concentration Control with ANFIS (Sensorless concentration control)*

Matlab simulation studies were carried out to control the concentration of methanol with ANFIS. Fig.14 summarizes the control logic. Input variables are entered to ANFIS and cell operating voltage is taken as the output. As a result if the model output is less than the output of ANFIS, methanol in certain concentrations is fed into the tank at a certain flow rate with the pump and this begins to increase the concentration so that the voltage too.

Based on the power density curves, methanol concentration was tried to control near the points, which are described below, where performance is the highest.

$70\,mA\,cm^{-2}$ current density at 0.3Molar

$100\,mA\,cm^{-2}$ current density at 0.5Molar

$250\,mA\,cm^{-2}$ current density at 1.75Molar

As seen from Fig. 15 while methanol concentration is gradually decrease, a drop in the cell voltage is experiencing. During this period controller compares the ANFIS reference voltage results with the cell voltage. If there is exist negative difference between them, pump begins to feed concentrated methanol into the tank. Here, concentrated methanol flow rate was kept high to see the changes quickly. As a result, the cell voltage related the methanol concentration is trying to keep a particular value (as interconnected) depending on operating conditions.

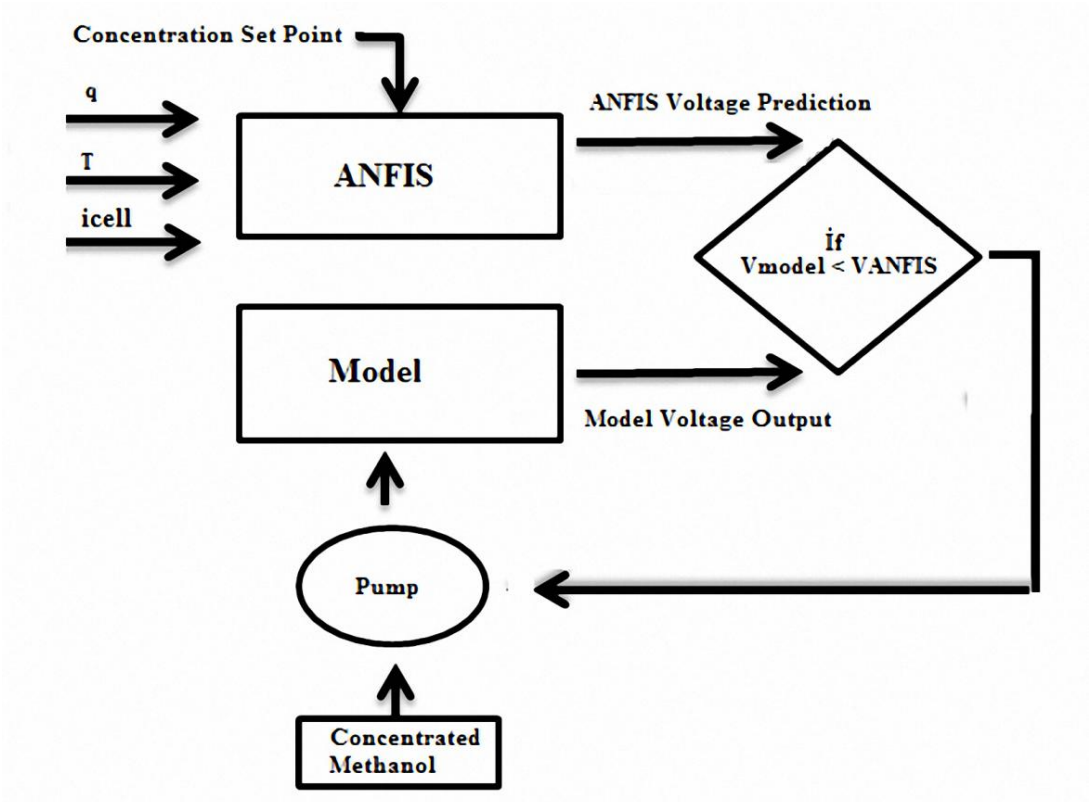


**Figure 14.** Methanol concentration control scheme with ANFIS

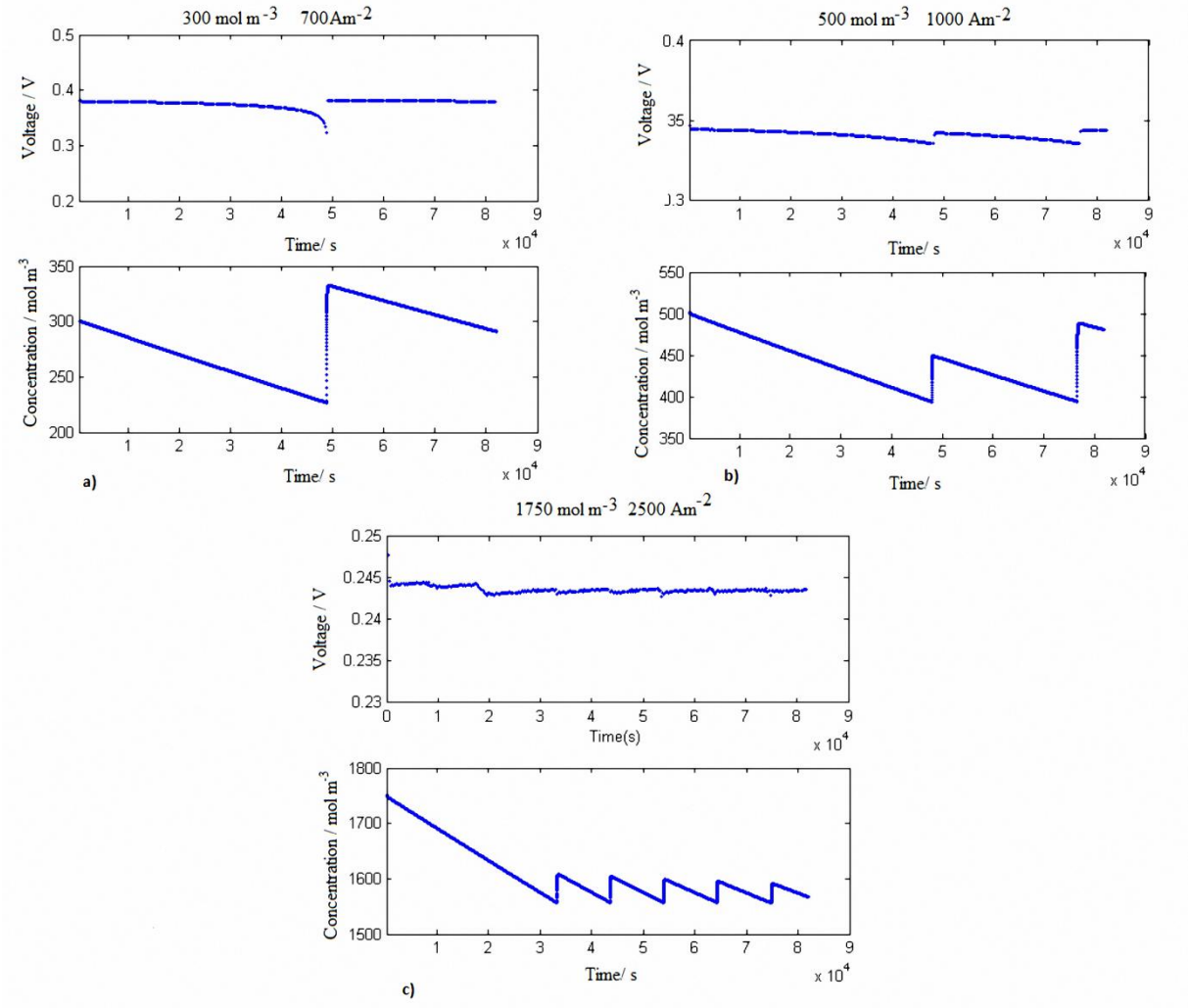


**Figure 15.** Control of methanol concentration with ANFIS at a) 300 mol $m^{-3}$ while current density is 700 A $m^{-2}$ b) 500 mol $m^{-3}$ while current density is 1000 A $m^{-2}$ a) 1750 mol $m^{-3}$ while current density is 2500 A $m^{-2}$

Consequently, as can be seen unlike the similar studies proposed control algorithm; does not need a particular monitoring period for intervention to concentration so longer working time at low or high concentrations is not disadvantage. Intensive mathematical operation, complex equations are not required (i.e. compute the pump active time, or methanol consumption rate etc.). Unlike other studies concentration is manipulated by estimating the cell reference voltage over a wide operating range (load, flow rate, temperature and concentration) whereby the reference voltage data, which obtained directly from the system itself to establish the ANFIS model, can be obtained at any operating point.

## 7. CONCLUSION

Establishing the experimental system is the first step of this study. Sensors such as level, concentration, carbon dioxide etc., and final control elements such as computer controlled power unit

and pump etc. were connected to the computer with the PCI 6221 DAQ card (data acquisition). The Electronic load and the air supply unit connected to the computer with the serial port (RS-232). After this stage, a program with an interface was developed for online tracking the cell and sensor values, also controlling the process elements. After all these preparations, firstly the methanol feed temperature was controlled by the Fuzzy Logic (fuzzy logic) control algorithm. Also PID control algorithm was applied to control the methanol concentration with sensor. As the system is controlled desirably; polarization curves obtained using a commercially MEA by changing the parameters such as methanol flow rate, methanol concentration, temperature, and current density was used in ANFIS modeling. Membership functions type was changed as trimf or gaussmf to investigate their effects also the number of membership functions of each input variables (2 or 3) was investigated. Ultimately, it was observed ANFIS model gave pretty good results when using the trimf as membership function in the case numbers of membership function is 3,2,2,2. Then a useful mathematical model that can represent the system sufficiently is established and unknown parameters of this model such as reaction rates, equilibrium constants, and mass transfer coefficient were determined through the experimental data with the particle swarm optimization method. And polarization curves obtained with the model were compared with the experimental data showed a good fit in all regions of the polarization curves. Then, this model was used to control the methanol concentration with ANFIS. It was seen in the works carried out different operating conditions that decreasing occurred in the cell voltage caused by the concentration drop is sensed by the controller and concentrated methanol pump is actuated thereby it is ensured to remain the cell voltage eat a certain value. Resulting, it can be said that ANFIS could control the methanol concentration in varying working conditions such as load, methanol flow rates, temperatures and concentration in a good manner without requiring complex operations and time consuming monitoring periods.

*NOMENCLATURE*

| | |
|---|---|
| A, B | linguistic variables |
| $A^s$ | cross-sectional electrode area, $m^2$ |
| $c_i$ | concentration of component $i$ in anode compartment, $mol\,m^{-3}$ |
| $c_i^{CL}$ | concentration of component $i$ in anode catalyst layer, $mol\,m^{-3}$ |
| $c_i^F$ | feed concentration of component $i$, $mol\,m^{-3}$ |
| $C$ | double layer capacity, $F\,m^{-2}$ |
| $D_{CH_3OH}^M$ | diffusion coefficient of methanol in membrane $= 2.9*10^{-10} m^2\,s^{-1}$ |
| $d^M$ | thickness of membrane $= 200\mu m$ |
| $F$ | Faraday's constant $= 96485 C\,mol^{-1}$ |
| $f_1$, $f_2$ | consequents of the rules |
| $f$ | rule output |
| $i_{cell}$ | cell current density, $Am^{-2}$ |
| $k_j$ | rate constant of reaction $j$, $mol\ m^2 s$ |
| $k^{LS}$ | mass transfer coefficient, $m\,s^{-1}$ |

| | |
|---|---|
| $K_j$ | equilibrium constant of reaction *j* |
| $n_i^M$ | mass flux density of comp. $i$ in membrane, $mol\ m^2 s$ |
| $O$ | output of the ANFIS node |
| $p$ | pressure, Pa |
| q | methanol feed flow rate, $\text{cc min}^{-1}$ |
| Q | air feed flow rate, $\text{cc min}^{-1}$ |
| $r_j$ | rate of reaction $j$ related to electric area, $mol\ m^2 s$ |
| $R$ | universal gas constant $= 8.314\ J\ mol^{-1} K^{-1}$ |
| $t$ | time, s |
| $T$ | cell temperature, *K* |
| $U$ | electric potential, V |
| $U_{cell}$ | total cell voltage, V |
| $U_{cell}^{\theta}$ | standard cell voltage $= 1.2V$ |
| $V$ | compartment volume, $m^3$ |
| $v$ | flow velocity in membrane, $m\ \text{s}^{-1}$ |
| $w$ | weight of the membership function |
| $\overline{w}$ | average weight of the rule |
| x, y | input(linguistic) variables |

*Greek letters*

| | |
|---|---|
| β, γ, δ | parameters of consequent of the rules |
| ε, σ, λ | membership function parameters |
| $\alpha$ | charge transfer coefficient = 0.5 |
| $\eta$ | electric overpotential, V |
| $\kappa^M$ | conductivity of membrane $= 17\Omega^{-1} m^{-1}$ |
| $\tau$ | mean residence time in anode compartment, s |
| $\Theta_i$ | surface fraction covered by component i |
| μ | membership function |

*Subscripts*

| | |
|---|---|
| $a$ | anode |
| $c$ | cathode |
| $CH_3OH$ | methanol |
| $CO_2$ | carbon dioxide |
| $H^+$ | proton |
| $O_2$ | oxygen |
| Pt | free active sites of platinum catalyst |
| $Pt_3$ - COH | sites occupied by $Pt_3$-COH |
| Pt-COOH | sites occupied by Pt-COOH |
| Ru | free active sites of ruthenium catalyst |
| Ru-OH | sites occupied by Ru-OH |

*Superscripts*

| | |
|---|---|
| *CL* | catalyst layer |
| *LS* | from liquid bulk to membrane surface |

$\theta$ at standard conditions

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