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OPERATION CHARACTERISTIC CONTROL OF DIRECT METHANOL FUEL CELL LIQUID FEED FUEL SYSTEM USING ADAPTIVE NEURO-FUZZY INFERENCE SYSTEM MODEL

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Abstract. *Currently, Direct Methanol Fuel Cell (DMFC) technology suffers from the low power density caused by slow reaction rate and undesired "methanol crossover", which limits its commercialization application. At this study, Adaptive Neuro-Fuzzy Inference System (ANFIS) can predict the needed amount of methanol fuel from the relationship of current and voltage curve of DMFC under different operational conditions and keep high power density. The ANFIS is a collaborative data bank from repeated experiments results under different amount of methanol fuel liquid. The model is a control scheme for predicting of supply to a fuel cell system under dynamic loading conditions, with a high accuracy in an easy, rapid and cost effective way to regulate the concentration of a liquid feed fuel cell without any fuel concentration sensor. The control scheme uses operating characteristics of fuel cell, such as potential, current and temperature. Our previous study has presented a fuel sensorless control algorithm (IR-DTFI) to calculate the quantity of fuel liquid required at each monitoring cycle. Furthermore, we develop ANFIS to strengthen the concentration regulating process. The ANFIS had been verified by systematic experiments, and the experimental results proved that the scheme can effectively control the fuel supply of a liquid feed fuel cell with reduced response time, even while the Membrane Electrolyte Assembly (MEA) deteriorates gradually.*
Keywords: Fuel sensorless control, ANFIS, Methanol crossover

1. Introduction.

Fuel cells are green power source of energy due to their high efficiencies and quiet operation. They convert chemical energy into electricity via an electrochemical reaction. The direct methanol fuel cell offers special benefits as a power source for transportation, such as potential high energy density, no need for a fuel reformer and a quick response. The operating characteristics of a DMFC, such as methanol concentration, reactant flow rates, and temperatures of the stack and environment, all considerably influence the behaviors of the DMFC system. Among those operating characteristics, methanol concentration is one key factor that significantly affects the performance and fuel utilization of the DMFC.

Methanol crossover from anode to cathode through a Nafion membrane, which causes a mixed potential on the cathode and reduces the overall cell voltage, is a well-known problem that hinders the development of DMFC [1,2]. Due to methanol crossover, practical operation of a DMFC requires accurate control of the methanol concentration within a predetermined range. The conventional approach is to use a methanol concentration sensor in a closed loop of fuel circulation. However, many requirements exist for methanol concentration sensors for DMFC [3]. Existing affordable products that meet all desirable criteria are not yet readily available. Furthermore, methanol sensors that have been developed using electrochemical methods [4] have issues, such as performance degradation of the MEA, which result in poor stability and durability. Methanol sensors based on physical properties, such as sound speed, density, or refractometry, are also sensitive to carbon dioxide bubbles and the pulse wave of the circulation pump in the fuel loop. In addition, methanol concentration sensors, regardless of whether they are designed using physical or electrochemical principles, exhibit a marked dependency on temperature. Measurement efforts and data sets will be large, complicated, and costly for development. Furthermore, when a methanol sensor is used, experimental tasks, such as calibration for each sensor, are necessary. Although the fuel sensor approach can be used to control fuel concentration, it nevertheless has the shortcomings of increased weight, size, complexity, and cost of the liquid feed fuel cell. Accordingly, the development of fuel sensorless control of liquid feed fuel cells has received much more attention in recent years.

Our previous papers [4] have presented a fuel sensorless control algorithm (IR-DTFI) with a fixed injection quantity at each cycle, while the specific monitoring period was modulated to regulate the concentration such that the fuel concentration and power output were controlled within an acceptable range. The specific monitoring periods were equal to or longer than 40 s; namely, the IR-DTFI control algorithm controlled the DMFC system behavior through the system responses of the last cycle of 40 s. The specific monitoring periods depend on the total number of methanol solution in the mixing tank and the anode compartment of the fuel cell system. We have successfully demonstrated a 40 W DMFC power pack for power sources in notebooks and DVD players that are embedded with the IR-DTFI control scheme for control of fuel supply.

Control computing techniques, including Artificial Neural Networks (ANN), Fuzzy Logic, neuro-fuzzy systems, on the other hand, can be used as an alternative to a physical model especially for complex nonlinear systems. ANNs are able to learn and generalize from examples and experience to produce meaningful solutions to problems [5]. Fuzzy Logic provides inference mechanisms that enable approximate reasoning and model human reasoning capabilities to be applied to knowledge-based systems [6]. A number of applications of both ANN and Fuzzy Logic can be found in the fuel cell literature, e.g., [7-10]. The neuro-fuzzy techniques, such as the Adaptive Neuro-Fuzzy Inference System (ANFIS) is the combination of the Fuzzy Logic and ANN and captures the advantages of both in the sense that the membership functions and rules of the fuzzy systems is defined and optimized by ANN so that unknowledge of the system is required. Sun et al. [11] applied ANFIS in order to build temperature model for PEMFC fuel cells. Entchev and Yang [12] have applied both ANN and ANFIS model in order to predict the performance of a solid oxide fuel cell in the microgeneration instillation.

In this paper, a modified control method based on the ANFIS model is presented. The ANFIS model is used to predict the performance of the DMFC for multi-input variables, shortened monitoring period is explored for faster system response and greater stability, and the model is used to predict control for the methanol fuel supply under various conditions to verify the model at dynamical load operating conditions. The paper has been organized as follows. At first, we analyze methanol crossover, then describe the

ANFIS model by method and application in the methanol fuel supply situation. The results and discussion section present and discuss the results of the ANFIS model and the effect of the operational conditions on the cell performance, for calculating the fuel injection quantity is proposed and validated, which is then successfully combined with the ANFIS control algorithm to control the fuel supply in DMFCs for low power automotive applications such as E-Wheel chairs and handicap cars.

2. **DMFC Methanol Crossover Analysis.** DMFC consists of an anode at which methanol is electro-oxidized to CO_2 through the reaction and a cathode, at which oxygen (usually as air) is reduced to water or steam. Each electrochemical reaction on the anode (1) and the cathode (2) sides of a cell is:

$$\mathrm{CH_3 + H_2O \rightarrow CO_2 + 6H^+ + 6e^-} \tag{1}$$

$$\mathrm{3/2O_2 + 6H^+ + 6e^- \rightarrow 3H_2O} \tag{2}$$

Total cell reaction:

$$\mathrm{CH_3OH + 3/2O_2 \rightarrow CO_2 + 2H_2O} \tag{3}$$

The thermodynamic efficiency of the process is given by the ratio between the Gibbs free energy, that is, the maximum value of electrical work (ΔG°) that can be obtained, and the total available energy for the process, that is, the enthalpy (ΔH°) and temperature (T). Under standard conditions:

$$\Delta G^\circ = \Delta H^\circ - (T \times \Delta S^\circ) \tag{4}$$

The total electrochemical reaction is related to the ideal cell potential by

$$\Delta G^\circ = -nF \times \Delta E_{rev} \tag{5}$$

where E_{rev} is the reversible standard potential under thermodynamic equilibrium, n is the number of electrons involved in the reaction, and F is the Faraday's constant (96,487 coulombs per mole of electron). At 25°C, 1 atm and with pure oxygen feed, the reversible potential for methanol oxidation is 1.18 V [13,14]. It does not vary significantly in the operating range 40 $\sim$ 130°C and 1 $\sim$ 3 bar abs pressure. Usually, the open circuit voltage of a polymer electrolyte DMFC is significantly lower than the thermodynamic or reversible potential for the overall process, the terminal voltage (E_{cell}) of the cell is deconvoluted into the anode and cathode polarizations:

$$E_{cell} = E_{\text{cathode}} - E_{\text{anode}} \tag{6}$$

This is mainly due to methanol crossover that causes a mixed potential at the cathode and to the irreversible adsorption of intermediate species at electrode potentials close to the reversible potential [6-19]. The over cell voltage can be written as

$$V_{cell} = E_{cell} - \eta_{\text{anode}} - \eta_{\text{cathode}} - \eta_{\text{ohmic}} - \eta_{\text{crossover}} \tag{7}$$

where η_{anode} and η_{cathode} are the anode and cathode overpotentials, η_{ohmic} is the overpotential due to the ohmic drop in the system and $\eta_{\text{crossover}}$ is the overpotential due to de methanol crossover through the membrane.

The methanol concentration on the anode side has a decisive influence on the current/voltage characteristic of a DMFC. Too high a methanol concentration leads to high methanol permeation so that methanol is oxidized at the cathode. The methanol permeation does not only reduce the mass efficiency but is furthermore also responsible for the formation of a mixed potential at the cathode, which reduces the cell voltage and thus decreases the voltage efficiency [15,16].

Methanol crossover occurs when methanol solution permeates from the anode to the cathode through the electrolyte membrane. Most of the permeated methanol is reacted at the cathode creating a "mixed potential", which reduces the cathode potential and consumes some of the oxidant. Therefore, this methanol crossover phenomenon significantly reduces the cell's performance. The crossover of methanol is also a major cause of inefficiency as it is essentially wasted and the cathode catalysts are poisoned by the carbon atoms in the methanol [17-20]. Figure 1 shows an illustration of the methanol crossover phenomenon in a DMFC.

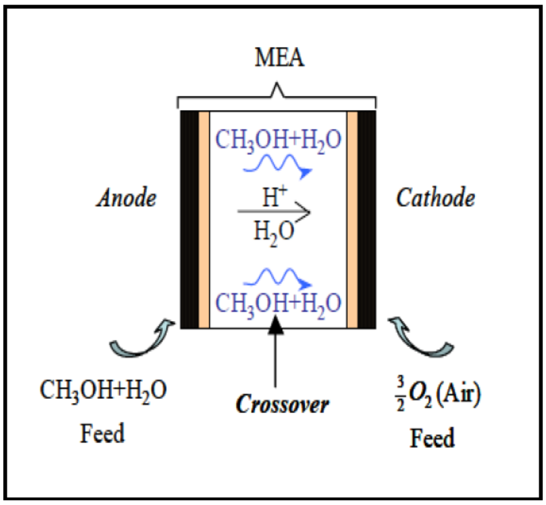

FIGURE 1. An methanol crossover in a DMFC

Methanol permeation causes losses due to additional methanol consumption and due to the formation of a mixed potential at the cathode decreasing the cell power [21].

The overall methanol consumption is:

$$\Delta MeOH_{(Total)} = \Delta MeOH_{(electric)} + \Delta MeOH_{(\mathrm{permeation})} \tag{8}$$

where $\Delta MeOH_{(Total)}$ is total molar flow of methanol (mol/s), $\Delta MeOH_{(electric)}$ is molar flow of methanol due to electric current (mol/s), and $\Delta MeOH_{(\mathrm{permeation})}$ is molar flow of methanol due to permeation (mol/s). According to Faraday's law, the $\Delta MeOH_{(electric)}$ can be expressed as:

$$\Delta MeOH_{(electric)} = \frac{I_{\mathrm{electric}}}{6F} \tag{9}$$

where I_{electric} is electric current, the methanol permeation $\Delta MeOH_{(\mathrm{permeation})}$ can be write as a corresponding parasitic current $I_{\mathrm{permeation}}$ flow Faraday's law:

$$\Delta MeOH_{(\mathrm{permeation})} = \frac{I_{\mathrm{permeation}}}{6F} \tag{10}$$

The mass efficiency $\eta MeOH$ defines the ratio of the methanol

$$\eta MeOH = \frac{\Delta MeOH_{(electric)}}{\Delta MeOH_{(Total)}} = \frac{\Delta MeOH_{(electric)}}{\Delta MeOH_{(Total)} + \Delta MeOH_{(\mathrm{permeation})}} \tag{11}$$

The methanol permeation in DMFCs decreases the voltage and fuel efficiency of the cell. When the methanol reaches the cathode catalytic layer, it is potentially reacted with the oxidant creating a mixed potential that decreases the cell potential. In addition, this reaction masks the cathodic catalytic sites that are needed for the oxygen reduction half-reaction. As stated previously, the diffusion contribution to methanol crossover limits the practical (i.e., allowable) fuel concentration, thus limiting the energy density metrics actually achievable by DMFC systems.

The diffusion increases at lower current densities due to smaller electro-oxidation rates that allow a greater reactant presence at the fuel side of the MEA; hence, a larger diffusional driving force. Thus, it is expected that the maximum crossover occurs at OCV when there is no current density drawn. Another factor that affects the amount of methanol crossover due to diffusion is the fuel mixture concentration, too low a methanol concentration prevents methanol losses and the formation of a mixed potential, however, on the other hand, it is associated with a rise in anodic diffusion overvoltage [22]. Figure 2 is shows the power density characteristics of a DMFC where the methanol concentration is varied. The lowest methanol concentration of 1 wt.% shown leads to overvoltage in the high current load (> 2 A) which reduce the cell voltage. At the same time, due to reduced mixed potential formation, the cell voltage is greatest in the low current density range (< 500 mA). The concentration must, therefore, be adjusted to an optimum value representing the best possible compromise with respect to the anodic and cathodic overvoltage.

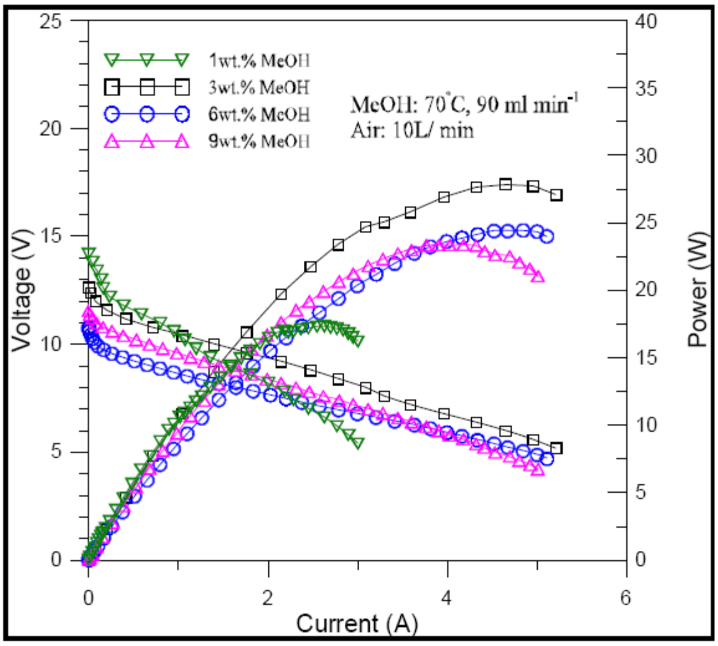

FIGURE 2. Polarization and power density curves at different methanol solution

3. Control Strategies of Method and Application DMFC System.

3.1. ANFIS control concept. ANFIS uses a hybrid learning algorithm to identify the membership function parameters of single-output. A combination of least-squares and backpropagation gradient descent methods are used for training FIS membership function parameters to model a given set of input/output data. For simplicity, it is assumed that the fuzzy inference system under consideration has two inputs and and one output. The system is an adaptive network functionally equivalent to a first-order Sugeno fuzzy inference system [23-27]. The ANFIS uses a hybrid-learning rule combining backpropagation, gradient-descent and a least-squares algorithm to identify and optimize the Sugeno system's signals. The equivalent ANFIS architecture of a first-order Sugeno fuzzy model with two rules is shown in Figure 3. The model has five layers and every node in a given layer has a similar function. The fuzzy IF-THEN rule set, in which the outputs are linear combinations of their inputs, is:

$$\begin{array}{l} \text{Rule 1: if } x \text{ is } A1 \text{ and } y \text{ is } B1 \text{ then } f_1 = p_1 x + q_1 x + r_1 \\ \text{Rule 2: if } x \text{ is } A2 \text{ and } y \text{ is } B2 \text{ then } f_2 = p_2 x + q_2 x + r_2 \end{array} \tag{12}$$

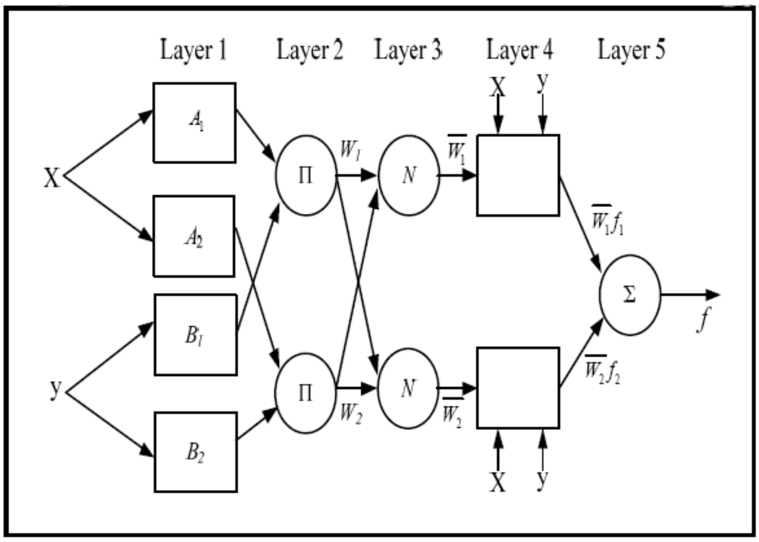

FIGURE 3. A typical structure of ANFIS

The layer 1 consists of adaptive nodes that generate membership grades of linguistic labels based upon premise parameters, using any appropriate parameterized membership function such as the generalized bell function

$$O_{1i} = \mu_{A_{iq}}(x) = \frac{1}{1 + \left| \dfrac{x - c_i}{a_i} \right|^{2b_i}} \tag{13}$$

where O_{1i} output is the output of the ith node in the first layer, x is the input to node i, A_{iq} is a linguistic label ("small", "large", etc.) from fuzzy set $A = \{A_{11}, A_{12}, \cdots, A_{1Q}, \cdots, A_{M1}, \cdots, A_{MQ}\}$ associated with the node, and $\{a_i, b_i, c_i\}$ is the premise parameter set used to adjust the shape of the membership function.

The nodes in layer 2 are fixed nodes designated Π, which represent the firing strength of each rule. The output of each node is the fuzzy AND (product, or MIN) of all the input signals

$$O_{2j} = w_j = \mu_{A_{1j}}(x) \times \mu_{A_{2j}}(x) \times \ldots \times \mu_{A_{Mj}}(x), \quad j = 1, 2, \ldots, N \tag{14}$$

The outputs of layer 3 are the normalized firing strengths. Each node is a fixed rule labeled N. The output of the jth node is the ratio of the jth rule's firing strength to the sum of all the rules' firing strengths

$$O_{3j} = \overline{w_j} = \frac{w_j}{w_1 + w_2 + \ldots + w_N}, \quad j = 1, 2, \ldots, N \tag{15}$$

The adaptive nodes in layer 4 calculate the rule outputs based upon consequent parameters using the function:

$$O_{4j} = \overline{w_j} f_i = \overline{w_i}(p_i x + q_i y + r_i) \tag{16}$$

where $\overline{w_j}$ is a normalized firing strength from layer 3, and $(p_i x, q_i y, r_i)$ is the consequent parameter set of the node. The single node in layer 5, labeled Σ calculates the overall

ANFIS output from the sum of the node inputs

$$O_{5l} = y_l = \sum_{j=1}^{N} \overline{w_j} f_1 = \frac{\sum_i w_i f_1}{\sum_i w_i}, \quad l = 1, 2, \ldots, r \tag{17}$$

Training the ANFIS is a two-pass process over a number of epochs. During each epoch, the node outputs are calculated up to layer 4. At layer 5, the outputs are calculated using a least-squares regression method. The output of the ANFIS is calculated and the errors propagated back through the layers in order to determine the premise parameter (layer 1) updates.

3.2. **ANFIS methanol fuel supply control model.** The input data to the ANFIS model for the methanol fuel supply sub-system are the operating voltage (V) and the operating current (I), used ANFIS to approximate the functional relations between input variables and responses to a desired degree of accuracy, the output data are the dosing pump active time. The performance of each model was evaluated by the root mean square error (RMSE) is defined by:

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (ei - pi)^2} \tag{18}$$

where p is the actual value from experiments, e is the predicted value from the models and N is the number of data points. According to the measured range resulted, the membership function of the variable are expressed by linguistic variables 'big (L)', 'small (S)', 'zero (Z)', 'very small (VS)', and 'medium (M)', as plotted in Figure 4, shows the input and the output data of the ANFIS methanol fuel supply control model.

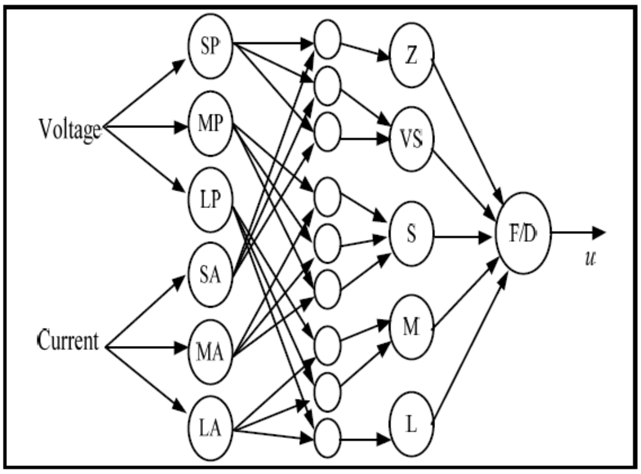

Figure 4. ANFIS methanol fuel supply control model

3.3. **Experimental setup.** An experimental setup was assembled as shown in Figure 5. Experimental system consisting of a DMFC stack, a water reservoir, a methanol reservoir, a tank for mixing water and methanol, an electronic loading, the methanol feed system consisted of a dosing pump unit for pure methanol and water. The methanol was injected into the water where it underwent mixing tank before entering the fuel cell, while a circulation pump for flow rate was fixed for each cycle. The methanol concentration was controlled by ANFIS model (PC) control the dosing pump flow rate values ranging. The overall flow rate of the methanol feed solution could be increased as desired by increasing

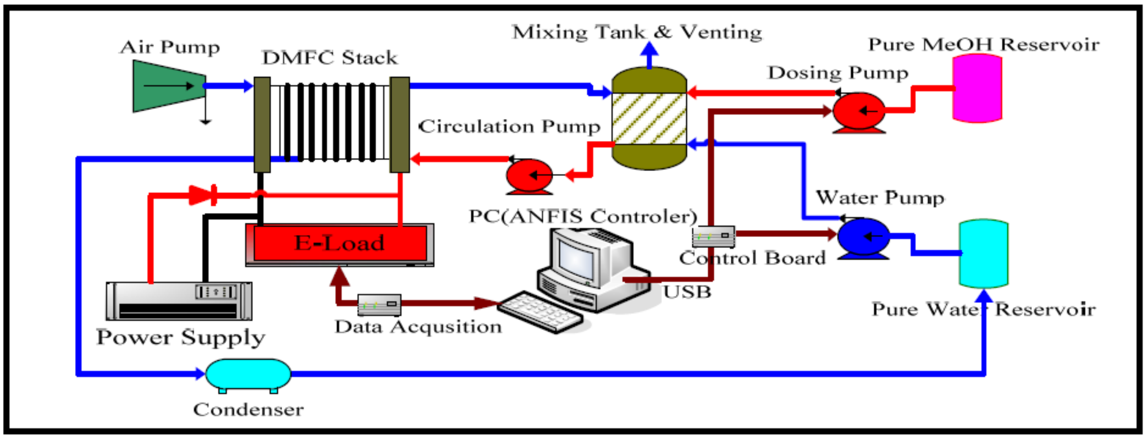

FIGURE 5. The test apparatus for evaluating the ANFIS control

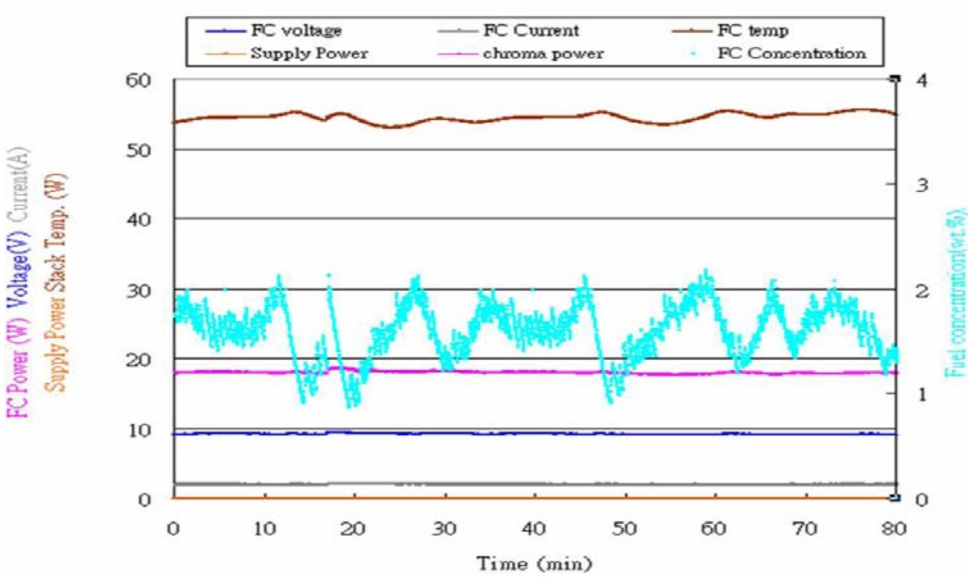

FIGURE 6. Dynamic characteristics of the fuel cell under constant current load (3 A)

the water flow rate. Load conditions on the fuel cell were maintained by DAQ system, and the operating power density was recorded by a National Instruments DAQmx^{TM} board.

4. **Results and Discussion.** Figure 6 shows the performance of the hybrid power system under constant current load (3 A) for 80 min operation, which verifying ANFIS model. The stack produced about 18 W steadily with very little fluctuation and the concentration of methanol was controlled between 1 and 3 wt.%. Figure 7 and Figure 8 plot the transient characteristics at ladder steps loads, which show good load following characteristics. The power oscillations, measured at the stack terminal, can be regulated through the DC/DC converter for steady output.

The model is successfully utilized on the power pack of 20-cell stack with active area of 50 cm^2 per cell developed using traditional bipolar plate configuration. Our DMFC team at INER make a success at DMFC/battery hybrid power system to supply an E-Wheel chair under the ANFIS control model embedded in the power management unit. The power pack on the E-Wheel chair is 4.4 L and 3 kg, which can produce about 50 W for

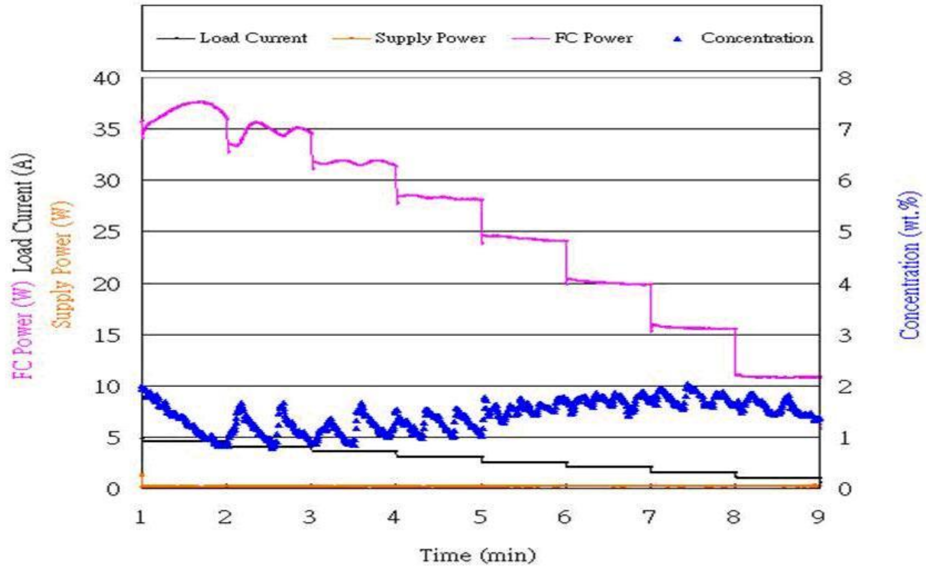

FIGURE 7. Dynamic characteristics of the fuel cell under ladder steps down load

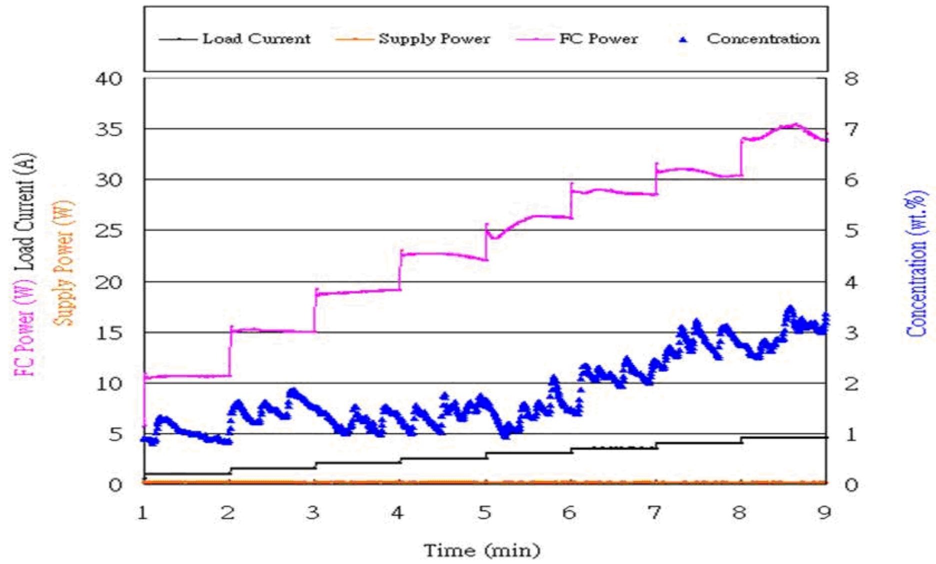

FIGURE 8. Dynamic characteristics of the fuel cell under ladder steps up load

8 to 10 hours continuous operation. Two sets of Li-ion batteries can be recharged by the power pack and served as the source of hybrid power.

5. **Conclusions.** We demonstrated the applicability of ANFIS methods for controlling DMFC methanol fuel supply. This offers the advantage that fuel management is achieved in an efficient way. Methanol crossover problems of the DMFC stack are prevented by monitoring the minimum single cell voltage. Drying of the DMFC membrane can be avoided by monitoring the ac impedance of the fuel cell stack. The complexity of a rigorous, mathematical solution of this problem cannot be handled on-line using state-of-the-art microcontrollers. On the other hand, the fuel cell characteristics depend strongly on temperature. This temperature dependence can be introduced by simple ANFIS models with appropriate transfer characteristics. Therefore, water and thermal management can be achieved by applying ANFIS logic in principle. The cost efficiency of our control

system in DMFC is well demonstrated. We find that not only methanol fuel supply, other factors such as temperature, stack decaying ratio which also can influence power density. We would investigate the effect of temperature and stack deterioration ration on DMFC in future study.

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