

Short communication

A hybrid multi-variable experimental model for a PEMFC

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Abstract

A hybrid model composed of a least square support vector machine (LS-SVM) model and a pressure-incremental model is developed to dispose operation conditions of current, temperature, cathode and anode gas pressures, which have major impacts on a proton exchange membrane fuel cell's (PEMFC) performance. The LS-SVM model is built to incorporate current and temperature and a particle swarm optimization (PSO) algorithm is used to improve its performance. The optimized LS-SVM model fits the experimental data well, with a mean squared error of 0.0002 and a squared correlation coefficient of 99.98%. While a pressure-incremental model with only one empirical coefficient is constructed to for anode and cathode pressures with satisfactory results. Combining these two models together makes a powerful hybrid multi-variable model that can predict a PEMFC's voltage under any current, temperature, cathode and anode gas pressure. This black-box hybrid PEMFC model could be a competitive solution for system level designs such as simulation, real-time control, online optimization and so on.

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1. Introduction

As a clean energy conversion technology, proton exchange membrane fuel cells (PEMFC) receive more attention because of their low operating temperature, high power density, quick start-up capability and long lifetime. PEMFC is an interesting technology for the next generation of vehicles, portable units and so on [1]. A convenient PEMFC model can help greatly to control, simulate, and diagnose its behavior.

The PEMFC system is a nonlinear, multi-variable electrochemical system that is hard to model. A large number of publications on fuel cell modeling [2] target the complicated internal phenomena at the molecular level. Among them, two-dimensional and more complex three-dimensional, two-phase and non-isothermal models have been presented [3–5], these had very complicated expressions with some key physical parameters that were even immeasurable. These mechanistic models usually focused on the electrochemistry, thermodynamics and fluid mechanics. Typically, they were centered on the membrane-electrode assembly (MEA), which could help ana-

lyze, design and optimize cell components in the laboratory context. However, generally they were not suitable for system level research [6–9].

An empirical modeling approach is more practical in some applications. Researchers can deduce a PEMFC stack's voltage responses without knowing the fuel cell's complicated internal characteristics. An active empirical modeling methodology in recent years is based on machine learning theories, such as artificial neural networks [10–14] and support vector machines (SVM) [15,16]. By mapping the fuel cell voltage as a function of various operational conditions, these black-box models agree well with experimental data. A common requirement in using these modeling approaches is that sufficiently representative data should be supplied in the training set to build a multi-variable empirical model. However, the numbers of the training data needed will increase dramatically when the numbers of input variables are increased, and these data may not be available. For example, in our previous work [15], 100 experimental data points were used to build a two-variable PEMFC voltage model, but the number of experimental data points used by Li et al. [16] had achieved 1000 to build a five-variable PEMFC temperature model. Therefore, although current density, temperature, cathode and anode gas pressures are the most important controllable variables for a PEMFC's performance, few of those

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models proposed can deal with all of them simultaneously. One important reason is simply the absence of sufficient empirical data describing a broad range of operating conditions.

Herein, a hybrid modeling approach is proposed. In this way, only limited numbers of empirical data are needed to build a model which can deal with current density, temperature, cathode and anode gas pressures simultaneously. This model consists of two parts: one is an empirical LS-SVM model that concerns current density and temperature. Li et al. [16] built a dynamic temperature model of a PEMFC by LS-SVM, incorporating temperature, cooling water speed, hydrogen speed, air speed and output power. As a least square version of SVM, LS-SVM inherits the superior generalization performance of SVM. Although its precision is slightly lower than SVM, LS-SVM significantly reduces computation time.

The key to obtaining a highly accurate SVM or LS-SVM model is to choose a proper set of hyper-parameters, but no effective guide lines have been put forward; some recommendations on how to determine them are quite contradictory and confusing. A trial method is used in [15,16], which greatly relies on the user's experience. Therefore, particle swarm optimization (PSO) [17] is adopted in this paper to automatically determine the best set of hyper-parameters.

The other part of the hybrid model is a pressure-incremental model taking account of both the cathode and anode gas pressures. This model is developed based on the work of Amphlett et al. [18] and Mann et al. [19]. After a slightly simplification, the number of empirical coefficients was greatly reduced from more than ten to only one.

2. Theoretical framework

For a given PEMFC system, the fuel cell terminal voltage V is influenced by many operating parameters: cell temperature T , cathode oxygen pressure P_{O_2} , anode hydrogen pressure P_{H_2} , relative humidity α , membrane humidity λ , etc. Accordingly, fuel cell voltage is given by

$$V = f(I, T, P_{O_2}, P_{H_2}, \lambda, \alpha, \dots) \quad (1)$$

Up to now, no model has ever been able to accommodate all these operating parameters. With the assumption that channel gas is fully saturated and membrane is fully humidified, Eq. (1) is simplified as

$$V = f(I, T, P_{O_2}, P_{H_2}) \quad (2)$$

Although Eq. (2) is a simplified equation, it is still hard to model with traditional means due to its high dimensionality. A way to deal with large scale complex systems is to break them into independent simple submodels. Thus, we further separate Eq. (2) into two parts as

$$V = V_{P_0}(I, T) + V_{\Delta P}(\Delta P_{O_2}, \Delta P_{H_2}) \quad (3)$$

The first part V_{P_0} denotes a LS-SVM model, which predicts cell voltages at different currents and temperatures under a constant cathode gas pressure $P_{O_2}^0$ and a constant anode gas pressure $P_{H_2}^0$. We refer to these constant pressures as reference pressures. The

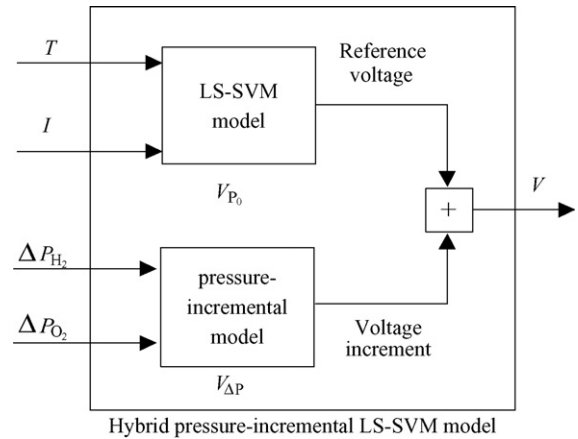


Fig. 1. Framework of the hybrid pressure-incremental LS-SVM model.

second part $V_{\Delta P}$ denotes a pressure-incremental model, which predicts voltage increment caused by oxygen pressure increment ΔP_{O_2} and hydrogen pressure increment ΔP_{H_2} . The structure of our proposed hybrid pressure-incremental LS-SVM model is illustrated in Fig. 1.

3. Optimized LS-SVM model

A support vector machine is a novel and powerful tool based on statistical learning theories. It was originally developed at AT&T Bell Laboratories by Vapnik [20] for classification in various domains of pattern recognition, then expanded successfully to deal with regression problems more recently. The SVM model possesses a high degree of precision and does not require a pre-knowledge of the fuel cell. Comparing to artificial neural networks, the SVM model has a superior capability of generalization and it is also more robust. LS-SVM proposed by Suykens and Vandewalle [21] is a least square version of the standard SVM. Compared to SVM, LS-SVM significantly reduces the computation time with a tiny precision loss. Taking advantage of the high computing efficiency of LS-SVM, PSO strategies are used to automatically optimize performance.

3.1. Modeling a PEMFC by LS-SVM

Building a LS-SVM model need three steps:

- Preparing training data.
- Selecting optimal LS-SVM parameters to train the LS-SVM model.
- Predicting with the LS-SVM model.

In this study, experimental data provided by Laurencelle et al. [22] was used to generate the training data for the LS-SVM model. This fuel cell was composed of 36 cells; each cell with a 232 cm² active area, graphite electrodes, and a Dow membrane. Air pressure and hydrogen pressure were both regulated to 3 atm. Training data were obtained at 24 °C, 31 °C, 39 °C, 56 °C and 72 °C and current densities from 0 mA cm⁻² to 1000 mA cm⁻².

A difficult and vital step in obtaining a highly accurate LS-SVM model is to choose a proper set of hyper-parameters γ and σ . γ determines the trade-off between model errors and model complexity, and σ is a internal kernel parameter. However, a few recommendations in the literature on how to determine them are quite contradictory and confusing. Past literature [15,16] used a try and trail approach to determine the optimal hyper-parameters, which needed much experience and patience by developers. Therefore, a PSO algorithm was adopted to automatically search for the best set of hyper-parameters.

3.2. Optimizing the LS-SVM model by PSO

PSO is an evolutionary computation technique proposed by Kennedy and Eberhart [17]. PSO is an extremely simple algorithm that requires only primitive and simple mathematical operators, and is computationally inexpensive in terms of both memory requirements and time. PSO is an algorithm based on population (“swarm”). Each individual, called a particle, flies in the D-dimensional problem space with a velocity that is dynamically adjusted according to the flying experience of the individual and its colleagues. Standard PSO algorithm is described as follows:

$$\mathbf{v}_i = \mathbf{w}\mathbf{v}_i + \mathbf{c}_1\text{Rand}()(\mathbf{p}_i - \mathbf{x}_i) + \mathbf{c}_2\text{rand}()(\mathbf{p}_g - \mathbf{x}_i) \quad (4)$$

$$\mathbf{x}_i = \mathbf{x}_i + \mathbf{v}_i \quad (5)$$

where \mathbf{x}_i represents the current position of particle i , \mathbf{v}_i is its velocity, \mathbf{p}_i is the best position it experienced. \mathbf{p}_g represents the best position among all particles in the swarm. \mathbf{w} is inertia weight, which regulates the trade-off between the global exploration and local exploitation abilities of the swarm. The acceleration constants \mathbf{c}_1 and \mathbf{c}_2 represent the weight of the stochastic acceleration terms that pull each particle toward self best position \mathbf{p}_i and swarm best position \mathbf{p}_g . $\text{Rand}()$ and $\text{rand}()$ are two random functions with range [0,1]. The basic idea of PSO is that every particle’s action \mathbf{v}_i is excited by self cognition \mathbf{p}_i and social experience \mathbf{p}_g to fly to the globe best position.

Using model’s mean square error (MSE) as fitness, the optimization problem is defined as follows

$$\min_{\gamma, \sigma} J(\gamma, \sigma) = \text{MSE} = \frac{1}{N} \sum_{i=1}^N (V_i - \widehat{V}_i)^2 \quad (6)$$

where N is the numbers of experimental data, V_i is experimental data, and \widehat{V}_i is predicted data. The process of implementing PSO algorithm to solve this optimization problem is as follows:

1. Initialize: initialize a swarm of particles with random positions and velocities in the three-dimensional (γ, σ) . Number of particles is 10; \mathbf{w} is set to 0.4; \mathbf{c}_1 and \mathbf{c}_2 are all set to 2. Maximal generations number is 50.
2. Evaluate each particle’s fitness: train the LS-SVM model by current position $(\gamma, \sigma)_i$, and then evaluate its fitness by Eq. (6).
3. Update each particles’ best fitness value \mathbf{p}_i according to the result of step 2. If \mathbf{p}_i is best than the swarm’s best fitness value \mathbf{p}_g , then reset \mathbf{p}_g to \mathbf{p}_i .

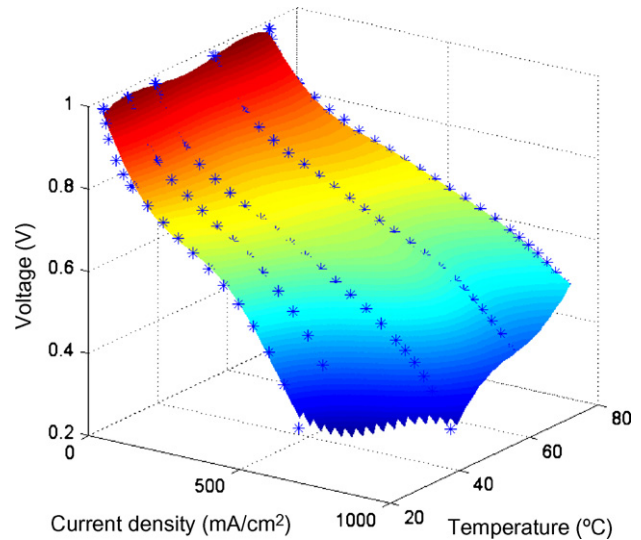


Fig. 2. Voltage predicted by LS-SVM model. Asterisk data is the experimental data.

4. Change the velocity and position of the particle according to Eqs. (4) and (5), respectively.
5. Loop to step 2 until maximal generations number is reached.

Above program is written in matlab language and implemented on Matlab 7.0. The minimum MSE of 0.0002 is found at

$$(\gamma, \sigma) = (650, 1.38)$$

As shown in Fig. 2, the LS-SVM model with optimal hyper-parameters fits the experimental data quit well. The mean squared error is 0.0002 and the squared correlation coefficient is 99.98%.

To make the model incorporate cathode and anode pressures besides current and temperature, a big problem should be considered firstly: if increasing the input variables form (I, T) to (I, T, P_{O_2}, P_{H_2}) to build a high dimensionality LS-SVM model, a huge amount of training data would be needed. Assuming n training data is needed for one variable, then n^2 training data is needed to build a 2-variable model, and n^4 training data is needed to build a 4-variable model. The numbers of training data per variable increase exponentially with the number of variables to maintain a given level of accuracy, and these data may not be available. To solve this problem, a pressure-incremental model was developed to expand our proposed LS-SVM model to other pressures.

4. Hybrid pressure-incremental LS-SVM model

4.1. Pressure-incremental model background

An influential one-dimensional PEMFC model was proposed by Amphlett et al. [18], and Mann et al. [19] further developed it to a generalized form. Herein, a simplified pressure-incremental model is developed based on their work. When current is drawn from a fuel cell, the cell voltage V_{cell} is decreased from its equilibrium thermodynamic potential E_{nemst} (open circuit volt-

age). This voltage drop includes activation loss η_{act} , ohmic loss η_{ohmic} and concentration loss η_{con} . The concentration loss is only notable when the fuel cell works with an extremely high current [23]; hence it can be neglected under normal conditions. The basic expression for the cell voltage is:

$$V_{cell} = E_{nernst} - \eta_{act} - \eta_{ohmic} \quad (7)$$

In Section 4.2, we firstly demonstrate the relationship between pressure and each of the terms in Eq. (7), and then describe how the pressure change affects them.

The first term is reversible thermodynamic potential E_{nerst} , which is described by the Nernst equation. Using literature values for the standard-state entropy change, the expression is:

$$E_{nernst} = 1.229 - (8.5 \times 10^{-4}) \cdot (T - 298.15) + (4.308 \times 10^{-5}) \cdot T \cdot (\ln P_{H_2} + 0.5 \cdot \ln P_{O_2}) \quad (8)$$

where T is cell temperature (K), P_{H_2} the partial pressure of hydrogen at the anode catalyst/gas interface (atm), and P_{O_2} the partial pressure of oxygen at the cathode catalyst/gas interface (atm).

The second term is activation overvoltage η_{act} , which is composed of anode and cathode activation overvoltages. Grouping them together, we can get a single expression as:

$$\eta_{act} = \xi_1 + \xi_2 T + \xi_3 T \ln C_{O_2} + \xi_4 T \ln I \quad (9)$$

where C_{O_2} is the concentration of dissolved oxygen at the gas/liquid interface, I is the cell current (A), and those four ξ_i ($i=1-4$) are coefficients.

The third term in Eq. (7) is ohmic polarization, which is caused by resistances in electron and proton transfers and other contact resistances. It can be expressed as:

$$\eta_{ohmic} = IR^{internal} = I(\xi_5 + \xi_6 T + \xi_7 I) \quad (10)$$

where ξ_i ($i=5-7$) are coefficients. It can be concluded from Eq. (10) that the ohmic overvoltage is independent of the pressure, which has no influence on our pressure-incremental model and could be neglected. Afterwards, a pressure-incremental model with only one coefficient is developed.

4.2. Pressure-incremental model

As shown in above discussion, hydrogen and oxygen pressures influence the cell voltage through the terms P_{H_2} , P_{O_2} and C_{O_2} . The partial pressure of hydrogen at the gas/liquid catalyst layer interface is [18]

$$P_{H_2} = P_{anode} \cdot [1 - 0.5x_{H_2O}^{sat}] \quad (11)$$

The partial pressure of oxygen at the gas/liquid catalyst layer interface is [18]

$$P_{O_2} = P_{cathode} \cdot \left[1 - x_{H_2O}^{sat} - x_{N_2}^{channel} \cdot \exp\left(\frac{0.291 \cdot I}{T \cdot 0.832}\right) \right] \quad (12)$$

Fig. 3 demonstrates the values of the exponent term $\exp(0.291 \cdot I / T^{0.832})$ at 40 °C and 80 °C. From the figure it can

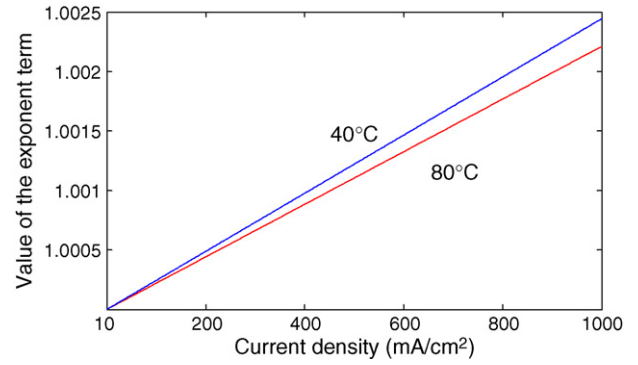


Fig. 3. Values of the exponent term at 40 °C and 80 °C.

be indicated that the difference with and without this term is no more than 0.25%, which is tiny enough to be neglected.

Hence, the partial pressure at the gas/liquid catalyst layer interface is simplified as

$$P_{O_2} = P_{cathode} \cdot [1 - x_{H_2O}^{sat} - x_{N_2}^{channel}] \quad (13)$$

The third term is the oxygen concentration C_{O_2} [18]:

$$C_{O_2} = \frac{P_{O_2}}{(5.08 \times 10^6) \cdot \exp(-498/T)} \quad (14)$$

Let $P_{O_2}^0$ and $P_{H_2}^0$ standing for the given reference partial pressures of oxygen and hydrogen, respectively, under which the LS-SVM model predicts the fuel cell voltages at different currents and temperatures. The oxygen increment and the hydrogen pressure increment are defined as the proportional form:

$$P_{O_2} = k_{O_2} P_{O_2}^0, \quad P_{H_2} = k_{H_2} P_{H_2}^0 \quad (15)$$

where k_{O_2} and k_{H_2} are ratios that represent the pressure increments to $P_{O_2}^0$ and $P_{H_2}^0$. With the substitution of Eq. (15) into Eq. (8), the thermodynamic potential can be rewritten as:

$$\begin{aligned} E_{nernst} &= 1.229 - (8.5 \times 10^{-4}) \cdot (T - 298.15) \\ &\quad + (4.308 \times 10^{-5}) \cdot T \cdot (\ln k_{H_2} P_{H_2}^0 + 0.5 \cdot \ln k_{O_2} P_{O_2}^0) \\ &= E_{nernst}^0 + (4.308 \times 10^{-5}) \cdot T \cdot (\ln k_{H_2} + 0.5 \cdot \ln k_{O_2}) \\ &\quad \text{with } E_{nernst}^0 \\ &= 1.229 - (8.5 \times 10^{-4}) \cdot (T - 298.15) \\ &\quad + (4.308 \times 10^{-5}) \cdot T \cdot (\ln P_{H_2}^0 + 0.5 \cdot \ln P_{O_2}^0) \end{aligned} \quad (16)$$

where E_{nernst}^0 is the thermodynamic potential under the reference pressures $P_{O_2}^0$ and $P_{H_2}^0$. With the substitution of Eqs. (15) and (14) into Eq. (9), the total overvoltage can be rewritten as

$$\begin{aligned} \eta_{act} &= \xi_1 + \xi_2 T + \xi_3 T \ln k_{O_2} C_{O_2}^0 + \xi_4 T \ln I \\ &= \xi_1 + \xi_2 T + \xi_3 T \ln C_{O_2}^0 + \xi_4 T \ln I + \xi_3 T \ln k_{O_2} \\ &= \eta_{act}^0 + \xi_3 T \ln k_{O_2} \quad \text{with } \eta_{act}^0 \\ &= \xi_1 + \xi_2 T + \xi_3 T \ln C_{O_2}^0 + \xi_4 T \ln I \end{aligned} \quad (17)$$

where η_{act}^0 is the overvoltage under the known pressure P^0 . With the substitution of Eqs. (16) and (17) into Eq. (7), the fuel cell

Table 1
Errors between actual voltage and predicted voltage

No.	P_{O_2}/k_{O_2} (atm)	P_{H_2}/k_{H_2} (atm)	Actual voltage (V)	Predicted voltage (V)	Error (%)
1	1.40/–	2.50/–	0.824	–	–
2	3.10/2.2143	3.10/1.2400	0.851	0.8586	0.92
3	0.60/0.4286	2.00/0.8000	0.781	0.7924	1.38
4	0.60/0.4286	3.10/1.2400	0.792	0.7988	0.83
5	3.10/2.2143	2.00/0.8000	0.847	0.8473	0.04

voltage can be rewritten in an incremental format as:

$$V = V_{P_0} + V_{\Delta P} \quad \text{with } V_{\Delta P} = \xi_3 T \ln k_{O_2} + (4.308 \times 10^{-5}) \cdot T \cdot (\ln k_{H_2} + 0.5 \cdot \ln k_{O_2}) \quad (18)$$

where V_{P_0} is the voltage predicted by LS-SVM model under the reference pressures $P_{O_2}^0$ and $P_{H_2}^0$. Eq. (18) is our proposed pressure-incremental model. By neglecting exponent term in Eq. (12), above algebraic manipulations greatly simplifies the electrochemical model by reducing the empirical coefficients $\xi_1, \xi_2, \xi_3, \xi_4, \xi_5, \xi_6, \xi_7$ and several other numeric empirical constants to only one coefficient ξ_3 . All those non-pressure-dependent terms are effectively aggregated into a single measured reference pressure. ξ_3 is a coefficient which relates only to chemical parameters of the cathode reaction and does not varies with operation conditions. Simple experiments can easily determine it. Mann et al. [19] provided a range of $(7.6 \pm 0.2) \times 10^{-5}$, and the middle value of 7.6×10^{-5} is adopted in this paper.

To verify our proposed model, experimental data proposed by Amphlett et al. [24] is used, as listed in Table 1. These data are obtained with temperature of 70°C and current density of 1.03 A cm^{-2} . The first group is used as reference data for our predictions, and the other four groups as data to verify our predictions. Based on the reference data, Eq. (14) can help predict voltages under other pressures. Although our model is a simplified form of Amphlett's model, the modeling results are still highly consistent with the same measurements.

Fig. 4 shows the fuel cell voltages at 70°C and 6.66 A. It is shown that the oxygen partial pressure bears a stronger relationship to the fuel cell voltage than the hydrogen partial pressure does. Thus, it will improve a fuel cell's output power more efficiently if the cathode pressure is increased rather than the anode pressure.

4.3. Hybrid model

In Section 3.2, the optimized LS-SVM model has been built to predict the voltages at air pressure and hydrogen pressure are both 3 atm with temperature from 24°C to 72°C . Using this prediction as reference data, our pressure-incremental model can be made to take the prediction at other cathode and anode pressures. As an example, the predictions at cathode/anode pressure at 1/1 atm and 5/5 atm are demonstrated in Fig. 5.

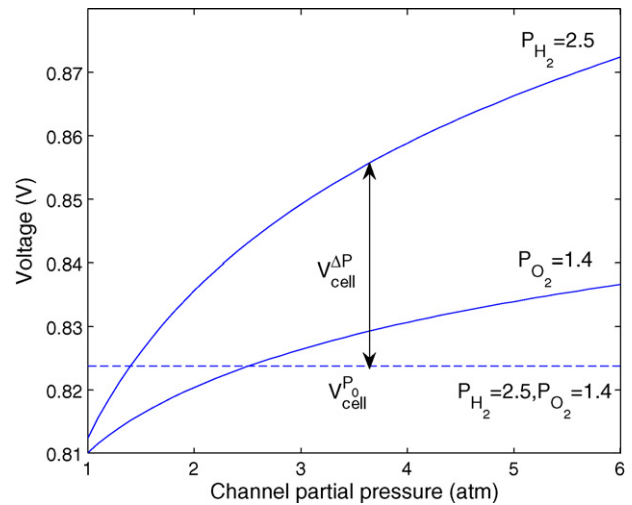


Fig. 4. Fuel cell voltages at 70°C and 6.66 A. The dot line represents the reference cell voltage under the working conditions in Table 1. In the upper curve, P_{O_2} varies from 1 atm to 6 atm with P_{H_2} kept at 2.5 atm. In the lower curve, P_{H_2} varies from 1 atm to 6 atm with P_{O_2} kept at 1.4 atm.

In order to get exact prediction results for different PEM-FCs, ξ_3 should be calibrated based on corresponding benchmark experimental data. Since ξ_3 is a constant for a given fuel cell, only small numbers of experimental data should do this procedure well.

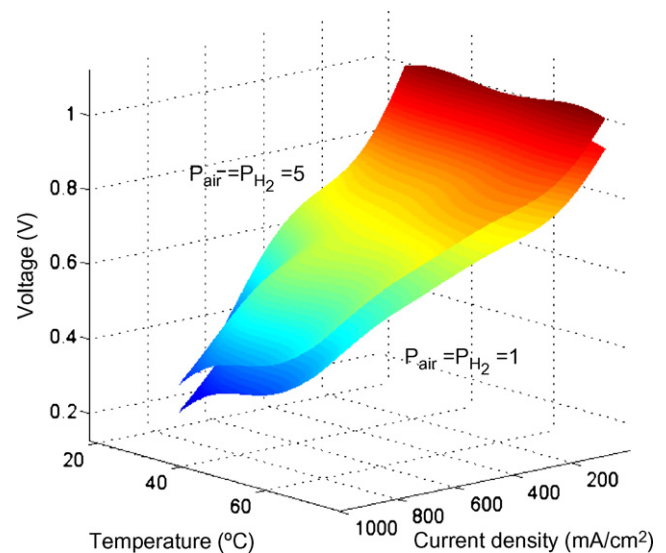


Fig. 5. Voltage predicted by the hybrid model under different pressures with temperature from 25°C to 70°C .

5. Conclusions

In this study, we have developed a system-level hybrid model of a PEMFC incorporating current, temperature, cathode and anode pressure. This model consists of two parts. One is an empirical LS-SVM model that concerns current and temperature. After optimization by a PSO algorithm, this LS-SVM model possesses excellent agreement between the predicted and experimental results. Off-line optimizing of the LS-SVM model took 68 min on a Pentium IV 1.6 GHz computer with 512 MB RAM, but the predicting time was no more than 1 ms. The other one is a pressure-incremental model that concerns cathode and anode pressures with only one empirical parameter. With little increase of benchmark experimental data in the two-variable LS-SVM model, the hybrid model could deal with all four operational variables. This hybrid black-box modeling methodology is most applicable for system level research such as simulation, real-time control, online optimization and so on.

One flaw of our hybrid model is that it should be used under modest changes in pressure, as significant changes in pressure will affect both the voltage and current, thus moving the fuel cell away from the reference conditions under which the simplified pressure-model was calibrated. It also will be invalid in low humidity or under extremely high current density operations. Hence, further studies are still needed.

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