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A self-adaptive supply method of micro direct methanol fuel cell

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ABSTRACT

In this paper, a novel self-adaptive anode feeding pattern is introduced to optimize the output and the dynamic response of the μDMFC (micro direct methanol fuel cell). The RSM (response surface methodology) is applied to optimize the anode flow rate, with two input parameters considered in this study, i.e., the methanol concentration and the operating current. In addition, the hardware circuit of anode self-adaptive supply system is realized based on the calculation results from RSM. Furthermore, the tests of system functionalities, including the pulse control test of micro peristaltic pump, the current collection test, and the flow rate monitoring of the self-adaptive feeding module are conducted. Finally, a metal-based μDMFC is designed and fabricated to evaluate the output performance when the cell patterned with or without the self-adaptive system. The experimental results reveal that the novel feeding pattern can increase the output voltage under different operating conditions, and reduce the voltage response time during the current transition.

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1. Introduction

Conventional batteries have disadvantages of self-discharge and serious environmental impact. Meanwhile, with the advantages of environmental-friendly, charging-free and high efficiency, the fuel cell becomes an attractive power source for various applications [1–7].

The micro direct methanol fuel cell has been considered as a prime candidate due to the advantages of low-emission, silent-operation and simplicity [8,9]. However, the μDMFC is a complex system in which the inner operating parameters have a significant influence on performance and dynamic characters of active μDMFC. Therefore, significant attentions have been devoted to the effects of operating parameters on μDMFC performance recently [10,11]. Seo et al. [12] analyzed the DMFC (direct methanol fuel cell) performance using air or oxygen as the oxidant gas under various operating conditions, including cell temperature, methanol concentration, flow rate, cathode humidification temperature, and cathode pressure. The results revealed that the DMFC exhibited the maximum performance with the constant anode flow rate of 3.0 ml min^{-1}. Alizadeh et al. [13] investigated the effects of various operating conditions on in-house fabricated DMFC with 10 cm by 10 cm active reaction area. In their study, the cell temperature, methanol concentration, and oxygen flow rate were considered with a fixed anode flow rate. Ge et al. [14] studied the effects of temperature, methanol concentration, anode flow rate, air flow rate, and cathode humidification on DMFC performance. The results indicated that, except the cathode humidification, all other operating parameters had significant influence on cell performance.

In most previous studies, an assumption was made that the operating parameters would affect the performance of μDMFC independently. Therefore, traditional experiments were generally carried out by varying only one parameter at a time while maintaining others constant. This procedure leads to misleading results. In fact, in the discharge process from the open-circuit state to the limiting current state, the optimal value of a particular operating parameter will be affected by other operating parameters due to the interaction among them. Therefore, for different portable applications with different fuel cell output mode, real-time optimized operating condition is needed to maximize the cell energy output.

DOE (design of experiment) and statistical techniques are widely used to optimize process parameters [15,16]. In this paper, based on the analysis results of response surface method, an anode flow rate self-adaptive supply system for active μDMFC is fabricated. The system can monitor the operating parameters such as the discharge current and the methanol concentration, and then the corresponding optimal supply rate is calculated from RSM

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(response surface methodology). Self-adaptive supply is realized through a micro peristaltic pump, which is driven by a square-wave voltage controlled by MCU (micro controller unit). Equipped with the proposed self-adaptive supply system, the cell performance shows a substantial improvement in output voltage and the voltage response time.

2. Optimization of the self-adaptive supply

2.1. *The response surface model*

In this paper, the relationship between the optimal anode flow rate and the methanol concentration as well as the operating current is established given the air-breathing μDMFC operates at room temperature. The response surface method, also known as the surface analysis method, works by approximating the implicit state function based on a series of random uncertainty experiments [17]. The CCD (central composite design) is applied using Design-Expert to simulate the relationship between the selected input parameters. A quadratic model is established as follows:

$$Y = \gamma_0 + \sum_{i=1}^{2} \gamma_i X_i + \sum_{i=1}^{2} \gamma_{ii} X_{ii}^2 + \sum_{i=1}^{2} \sum_{j=i+1}^{2} \gamma_{ij} X_i X_j \quad (1)$$

where Y is the optimal flow rate; X_i represents two main input parameters: methanol concentration ($i = 1$), operating current ($i = 2$). γ_0, γ_{ii}, γ_{ii} and γ_{ij} are the constant, the linear, the quadratic and the second order interaction coefficients, respectively.

Based on our previous study [18], operating parameters in Eq. (1) are selected randomly by Design-Expert as listed in Table 1. The possible choices of methanol solution concentration are 0.5 mol L^{-1}, 1.0 mol L^{-1}, 1.5 mol L^{-1} and 2.0 mol L^{-1}. The median of the loading current value is 120 mA cm^{-2}, while the low and high values are 60 mA cm^{-2} and 180 mA cm^{-2}, respectively.

2.2. *The establishment and significance test of the RSM*

The data in Table 1 were fed into the RSM to construct an empirical model for the optimal flow rate in terms of methanol concentration (denoted as parameter A) and discharge current (denoted as parameter B). The quadratic model was used to fit the observed data by the least squares analysis. Table 2 shows the ANOVA (analysis of variance) result of the quadratic model. As can be seen from the table, the predicted values match with the measured values accurately, indicating a significant relationship between the factors and the responses. R-Squared value of the model is 0.9532, which implies the model is significant.

Table 1
The CCD procedure and results.

	Concentration (mol L^{-1})	Operating current (mA cm^{-2})	Optimal flow rate (ml min^{-1})
1	2.0	120	0.5
2	1.0	120	1.0
3	2.0	180	0.5
4	1.5	120	0.5
5	2.0	60	0.5
6	1.0	180	1.0
7	1.0	120	1.0
8	1.5	120	0.5
9	0.5	180	1.5
10	0.5	60	0.5
11	1.0	60	0.5
12	0.5	120	1.5
13	1.0	120	1.0

Table 2
Error results for the velocity model.

Source	Squares	df	Square	Value	Prob > F
Model	1.58	5	0.32	12.03	0.0025
A-concentration	0.90	1	0.90	34.24	0.0006
B-current	0.29	1	0.29	11.15	0.0024
AB	0.24	1	0.24	9.15	0.0193
A^2	0.08	1	0.08	3.19	0.0173
B^2	0.11	1	0.11	4.06	0.0437
Residual	0.18	7	0.02		
Lack of fit	0.18	4	0.04		
Pure error	0	3	0		
Cor total	1.77	12			
R-squared	0.9532				

In the CCD method, the correlation level between the two operating parameters can be quantified by P-values. The P-value less than 0.05 indicates that the corresponding variable has a significant effect on the response with the degree of confidence of more than 95%. On the other hand, the P-value greater than 0.10 indicates the model terms are not significant. If the P-value sits between the above two mentioned values, the variables have marginal effects on the output and should not be neglected. In this model, A, B, AB, A^2 and B^2 are all significant model terms, which reveals that the two parameters are complexly interacted. The final equation in terms of actual factors was obtained as:

$$Y = 1.08 - 0.13 \times A + 0.33 \times B + 0.05 \times A \times B - 0.028A^2 - 0.028B^2 \quad (2)$$

2.3. *The RSM results and discussion*

The experimental data versus the simulated data from the RSM model are shown in Fig. 1. It can be seen that the predicted responses are close to the observed ones in the range of the operating variables, which is in agreement with the analysis of variance analysis. The residual analysis was conducted and it can be seen that normal probability point was distributed in a straight line, with only a very small range of float, shown as Fig. 1(a), which indicates that the residuals follow a normal distribution. Fig. 1(b) compares the predicted and experimental values for the model. From this figure, all the points are located in the vicinity of a straight line. The fitting result is also consistent with the results of the above residual analysis.

In order to intuitively analyze the interactions among the optimal flow rate and the methanol solution as well as the operating current, a three-dimensional response surface as shown in Fig. 2(a) and a flat contour as shown in Fig. 2(b) were generated.

From the analysis of 3D stereogram and the contour, it is obvious to notice that the optimal flow rate of μDMFC is affected significantly by the methanol concentration and the discharge current. When the μDMFC is supplied with the lower methanol concentration of 0.5 mol L^{-1}, the cell performance is hardly affected at the low current region. Considering the utilization efficiency, the optimal supply flow rate of 0.5 ml min^{-1} was adopted. The methanol consumed in the anode electrochemical reaction and the CO_2 gas produced by anode oxidation increased with the increasing of discharge current. Therefore, the anode flow rate must be elevated to 1.0 ml min^{-1} and 1.5 ml min^{-1} to maintain the high performance. As the cell supply concentration is increased to 1.0 mol L^{-1}, the limiting current is also increased. When the cell operated in the low current region, the performance difference corresponding to the flow rate of 0.5 ml min^{-1} and 1.0 ml min^{-1} is minimal. Again, the

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