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PEMEC performance evaluation through experimental analysis of operating conditions by response surface methodology (RSM)

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Abstract: The optimum current value of the proton exchange membrane electrolysis cell (PEM-EC) mainly depends on various operational factors, such as temperature, operating pressure, water flow rate, and membrane water content. Therefore, this study aims to maximize performance related to the current of PEM-EC by determining the optimal operating conditions of the PEM electrolysis cell having a $9\ cm^2$ active layer. In this regard, response surface methodology (RSM) and central composite design (CCD) were applied using Design-Expert (trial version) software to identify the optimal combination of operating variables such as temperature, pump speed, and cell voltage. Temperature, pump speed, and cell voltage were the independent variables to have ranged from 40-80 °C, 1-8, and 1.8-2.3 V, respectively. Also, the individual and combined effects of operational parameters on cell performance will be included in this study by ANOVA (analysis of variance). The optimal parameters are 80 °C, 1, and 2.3 V, respectively, temperature, pump speed, and cell voltage corresponding to the maximum current output of PEM-EC. This RSM tool found that the maximum current was 16.778 A. In addition, it was concluded that the most influential parameter on cell performance was the cell voltage, followed by the temperature.

Keywords: PEM electrolysis cell, Operational conditions, Optimization, Response surface methodology (RSM)

1. INTRODUCTION

Most research on renewable energy technologies became a widespread issue last decades due to the rapid depletion of fossil resources, the negative impact of conventional fossil fuel-based energy sources on human health and the environment, and fluctuations in natural gas or oil prices. The most crucial property of renewable energy sources is their environment friendly. Hydrogen will soon become one of the cleanest, non-toxic, and most sustainable energy carriers with this feature. Low-polluting and high-purity hydrogen and oxygen can be produced from water electrolysis compared with traditional hydrogen production methods such as alkaline water electrolysis, ammonia cracking, and fossil fuel reforming [1-3]. Hydrogen and oxygen from water electrolysis can be used directly in fuel cells and industrial applications. Additionally, water electrolysis utilizes DC power from renewable energy sources such as wind, solar, and geothermal. The cost of hydrogen production may differ depending on the renewable and nuclear energy sources used for water electrolysis. Compared to other renewable energy sources (geothermal, nuclear, and wind), solar energy has the highest cost, ranging from 10 to 30 $/kg [4].

The performance and durability of the PEM electrolysis system have been dependent on the design and operating parameters, membrane, and electrode characteristics. With the increasing demand for green hydrogen, the PEM electrolysis cell has become a trending topic in recent years. However, due to the low hydrogen production rate and high energy consumption, the efficiency of the PEM electrolysis cell remains low in terms of economic competitiveness. Therefore, many studies have been conducted to enhance efficiency and cell performance. Mass transport losses at high current densities are essential in reducing efficiency [5-7]. According to Faraday's law, oxygen gas production increases at the anode electrode of the PEM electrolysis cell at high current densities. The formation of oxygen bubbles over time interferes with the electrolysis reaction of water in the anode catalyst layer, which degrades PEM electrolysis cell performance [8]. The physical properties of the porous transport layer, such as the thickness and pore size, must be controlled to minimize mass transport limitations. Many studies have also been conducted to examine the performance of PEM electrolysis at low current densities (about $2\ A/cm^2$) to reduce mass transport loss, such as [9-10]. To increase

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the performance of the PEM electrolysis cell, voltage losses must be minimized. Afshari et al. [11] proposed a zero-dimensional mathematical model to investigate voltage losses. The results indicated that the activation over-potential contributes the highest to the cell voltage, while the concentration over-potential contributes the lowest. Therefore, activation and ohmic over-potentials need to be decreased to boost the performance of PEM electrolysis cells. Temperature, pressure, and water flow rate are the operating conditions that significantly affect the performance of the PEM electrolysis cell. Santarelli et al. [12] conducted performance tests of a 160 cm² active area PEM electrolyzer stack. They found that decreasing the water temperature or increasing the cathode pressure can reduce efficiency. At the maximum electrical power supplied to the PEM electrolysis cell, the power difference between the best (700 kPa, 58 °C) and worst (7000 kPa, 42 °C) conditions was approximately 0.5 kW. Upadhyay et al. [13] developed a steady, three-dimensional computational fluid dynamics (CFD) model. They discovered that the best water temperature range of the PEM electrolysis stack is 40 °C-80° C. Increasing the cathode pressure augmented the demand for electrical input power, which decreased cell performance. Numerical results showed that low gas diffusion layer porosity and water flow rate require high power. Lin et al. [14] numerically compared the parallel, triple-serpentine, and pin flow field configurations. They found that compared to other flow field designs, the best cell performance is obtained in the parallel flow field plate due to its good mass transfer, uniform pressure distribution, and low contact impedance characteristics. As a result of orthogonal experiments, the flow field design most affected the PEM electrolysis performance. Lee et al. [15] optimized the working conditions of the PEM electrolysis cell to improve its efficiency and performance PEMEC. Their study investigated sulfuric acid flow, direction, and flow rate in the anode and cathode compartments of the PEM-EC, catalyst coating, and various catalytic materials. Lickert et al. [16] examined the impacts of temperature, pressure, and water flow rate on cell performance on two stacks of PEM electrolyzers, one with the flow field in the anode chamber and the other without the flow field arrangement. It was concluded that the operating conditions have a crucial effect on the polarization performance of the PEM electrolyzer stack, which has no flow field design under the porous transport layer. In addition, it was observed that the mass transfer loss decreased with increasing temperature, pressure, and water flow rate. Although many numerical and experimental studies have been conducted in the literature to improve the efficiency of PEM electrolysis cells, the current research in which the design or operating parameters affecting the cell performance were optimized with an optimization tool has been carried out in the last few years. This study investigated the effects of temperature, water feed rate or pump speed, and cell voltage on cell performance by experimental design. In addition, the optimization of input factors for high current values based on response surface methodology (RSM) has been brought to the literature.

2. MATERIAL AND METHOD

The performance tests of single-cell PEM-EC with an active area of 9 cm² were carried out in the experimental test setup in the TUBITAK MAM Energy Institute laboratory. The main components of the experimental test bench consisted of a PEM-EC, Heidolph heater, TDK-Lambda DC power supply (760 W), ENDA temperature controller, Masterflex peristaltic pump, and a data acquisition system (see Figure 1). Design-of-experiment (DOE) and RSM are excellent tools to reveal the objective function's optimum value and minimize the number of experiments. Applying the RSM method to the experimental system reduces the number of experimental trials and saves time and cost. The RSM method reveals the response-input parameter relationship with three-dimensional contour plots. It combines strategy and experimental designs to create a novel data set with first or second-order polynomial equations. RSM is a mathematical and statistical tool that describes the effects, contributions, and interactions between independent variables on the dependent variable. In this study, temperature, pump speed, and cell voltage are independent variables. Current is the dependent variable. To investigate the effect of input factors on the output factors in the region of investigation, a central composite design (CCD) was applied using Design-Expert (trial version). CCD fits the second-order response surface, including axial point runs, center point runs, and cube point runs. The total number of experiments with three variables is 17. This paper's response surface consists of a second-order model with a minor numerical error (see Equation 1).

$$y = \beta_0 + \sum_{i=1}^{3} \beta_i X_i + \sum_{i=1}^{3} \beta_{ii} X_i^2 + \sum_{i=1}^{3} \sum_{j=i+1}^{3} \beta_{ij} X_i X_j + e$$

Where y is the predicted response (current and hydrogen flow rate), β_0 is the constant coefficient, X_i $(i = 1-3)$ is the main factor, β_i is the linear term, β_{ii} is the quadratic term, and β_{ij} $(i \, and \, j = 1-3)$ is the second-order interaction coefficient.

3. RESULTS AND DISCUSSION

This study optimized operating conditions using the CCD and the RSM to obtain the maximum current value. The effects of temperature (A), pump speed (B), and cell voltage (C) on the response (current) in the PEM electrolysis cell were analyzed in Design-Expert (trial-version) software. The RSM tool used low and high values of each input factor to identify significant parameters, as seen in Table 1. Analysis of variance (ANOVA) was employed for the central composite experiment design. ANOVA results of responses affected by operating conditions are given in Table 2. If p-values are greater than 0.1000, the model term is insignificant; if less than 0.0500, the model term is significant. In this case, cell temperature (A), cell voltage (C), and cell temperature-cell voltage (AC) were considerable model terms for current. The Pred R² of 0.9944 good agreed with the Adj R² of 0.9989 since the

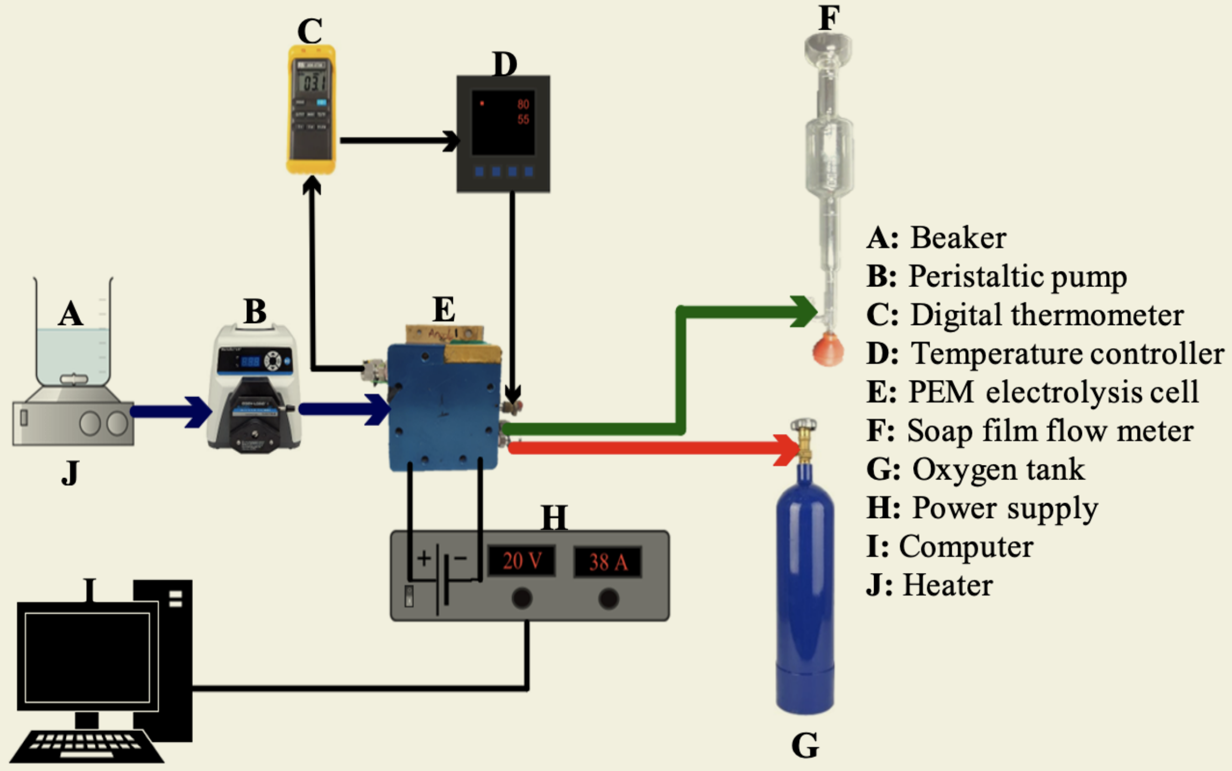

Figure 1. Experimental setup

difference between them is much smaller than 0.2. The regression analysis was carried out in Design-Expert (trial version), and the regression analysis of the input factors is given in a quadratic equation as follows:

Figure 2 (a-c) illustrates the interrelationships between temperature and pump speed (a), temperature and cell voltage (b), and pump speed and cell voltage (c) on current. The most influential parameter on cell performance is voltage, followed by temperature and pump speed, respectively. PEM-EC performance improves with an increase in cell voltage and temperature. On the other hand, operating temperatures above 80 °C cause damage to the membrane, which negatively affects cell perfor-

Table 1. Design table of experiments

Std order	Run	Factor 1 Temperature (°C)	Factor 2 Pump Speed	Factor 3 Cell Voltage (V)	Response 1 Current (A)
14	1	60	4	2.3	14.72
4	2	80	8	1.8	3.48
9	3	40	4	2	6.52
17	4	60	4	2	7.46
7	5	40	8	2.3	12.58
6	6	80	1	2.3	16.77
3	7	40	8	1.8	2.72
2	8	80	1	1.8	3.83
10	9	80	4	2	8.2
12	10	60	8	2	7.37
11	11	60	1	2	7.55
1	12	40	1	1.8	2.54
8	13	80	8	2.3	16.6
15	14	60	4	2	7.48
5	15	40	1	2.3	12.8
13	16	60	4	1.8	3.3
16	17	60	4	2	7.5

Table 2. ANOVA results for current

Source	Sum of Squares	df	Mean Square	F-value	p-value
Model	359.39	9	39.93	1632.67	<0.0001
A: Cell temperature (°C)	14.37	1	14.37	587.64	<0.0001
B: Pump speed	0.0561	1	0.0561	2.29	0.1736
C: Cell voltage (V)	331.36	1	331.36	13548.07	<0.0001
AB	0.0219	1	0.0219	0.8961	0.3754
AC	4.58	1	4.58	187.12	<0.0001
BC	0.0049	1	0.0049	0.1997	0.6684
Residual	0.1712	7	0.0245		
Lack of fit	0.1704	5	0.0341	85.20	0.0116
Pure error	0.0008	2	0.0004		
Cor Total	359.56	16			
Standard deviation	0.1564			R^2	0.9995
Mean	8.32			Adj R^2	0.9989
C.V.%	1.88			Pred R^2	0.9944

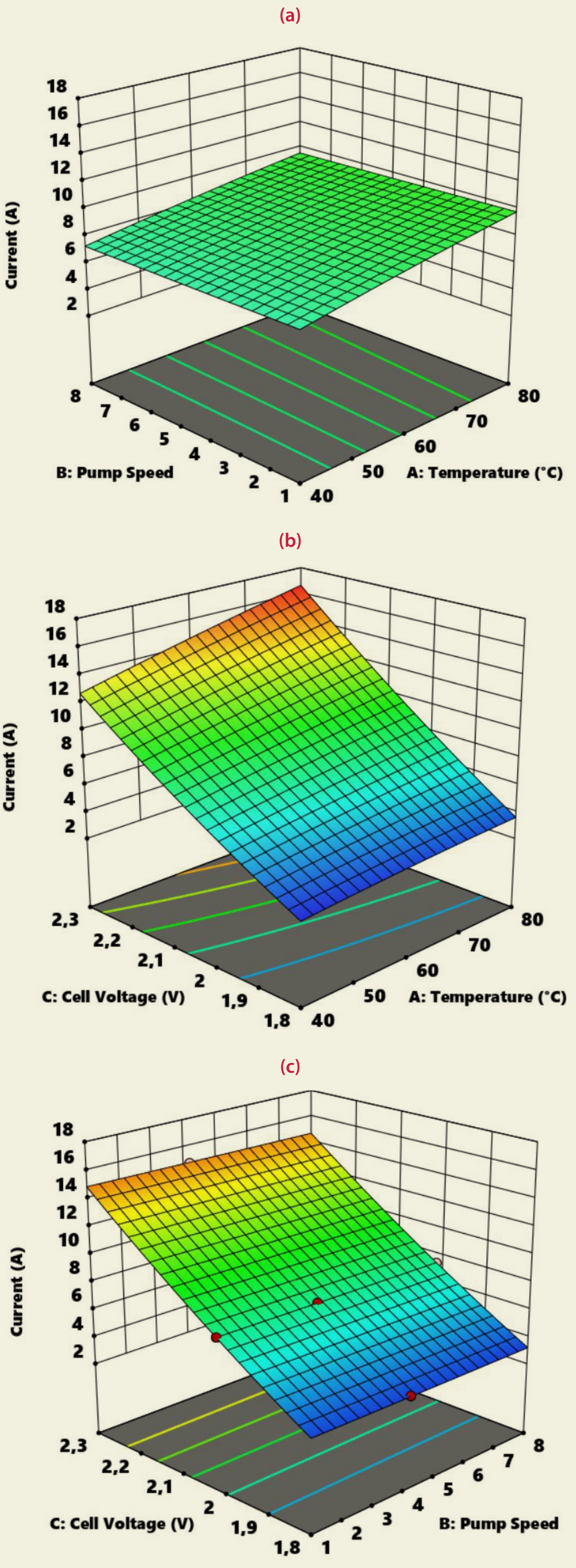

Figure 2. Current as a function of temperature and pump speed (a), temperature and cell voltage (b), pump speed and cell voltage (c)

mance. Variation in pump speed had no crucial effect on the performance of the PEM-EC.

At the end of this experimental study, optimum operating conditions for maximum cell performance were found. In the optimization process, we maximize the current and ensure that the control or input factors remain within the specified range (see Table 3). The importance and weight values of the input and output factors are given in Table 3. Optimum points with a desirability value greater than 0.995 are shown in Table 4. Maximum current (16.778 A) was obtained with a cell temperature

Table 3. Optimization for maximum current

Variable	Goal	Lower limit	Upper limit	Lower weight	Upper weight	Importance
Temperature (°C)	is in range	40	80	1	1	3
Pump speed	is in range	1	8	1	1	3
Cell voltage (V)	is in range	1.8	2.3	1	1	3
Current (A)	maximize	2.54	16.77	1	1	5

Table 4. Optimized results

Run	A: Temperature (°C)	B: Pump speed	C: Cell voltage (V)	R1: Current (A)	Desirability
1	80.000	1.000	2.300	16.778	1.000
2	79.997	1.042	2.300	16.773	1.000
3	79.966	1.068	2.300	16.772	1.000
4	80.000	1.637	2.300	16.748	0.999
5	79.603	1.000	2.300	16.742	0.999
6	80.000	1.687	2.300	16.745	0.999
7	80.000	1.755	2.300	16.742	0.998
8	79.136	1.034	2.300	16.699	0.997
9	80.000	2.242	2.300	16.719	0.996
10	80.000	2.332	2.300	16.715	0.995

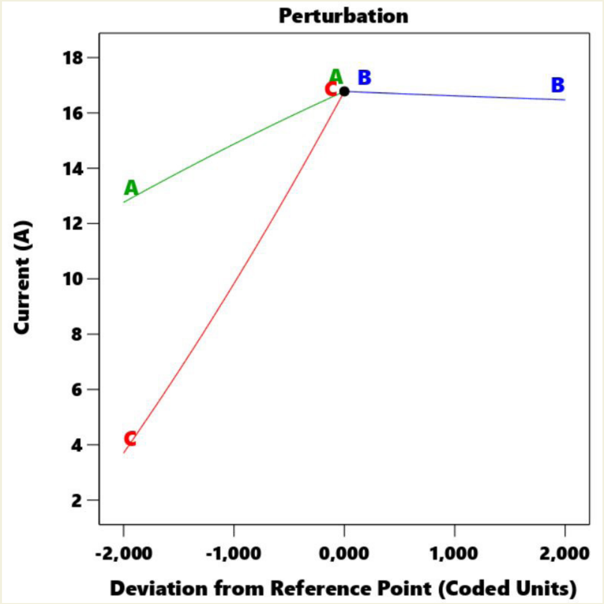

Figure 4. The perturbation plot for current

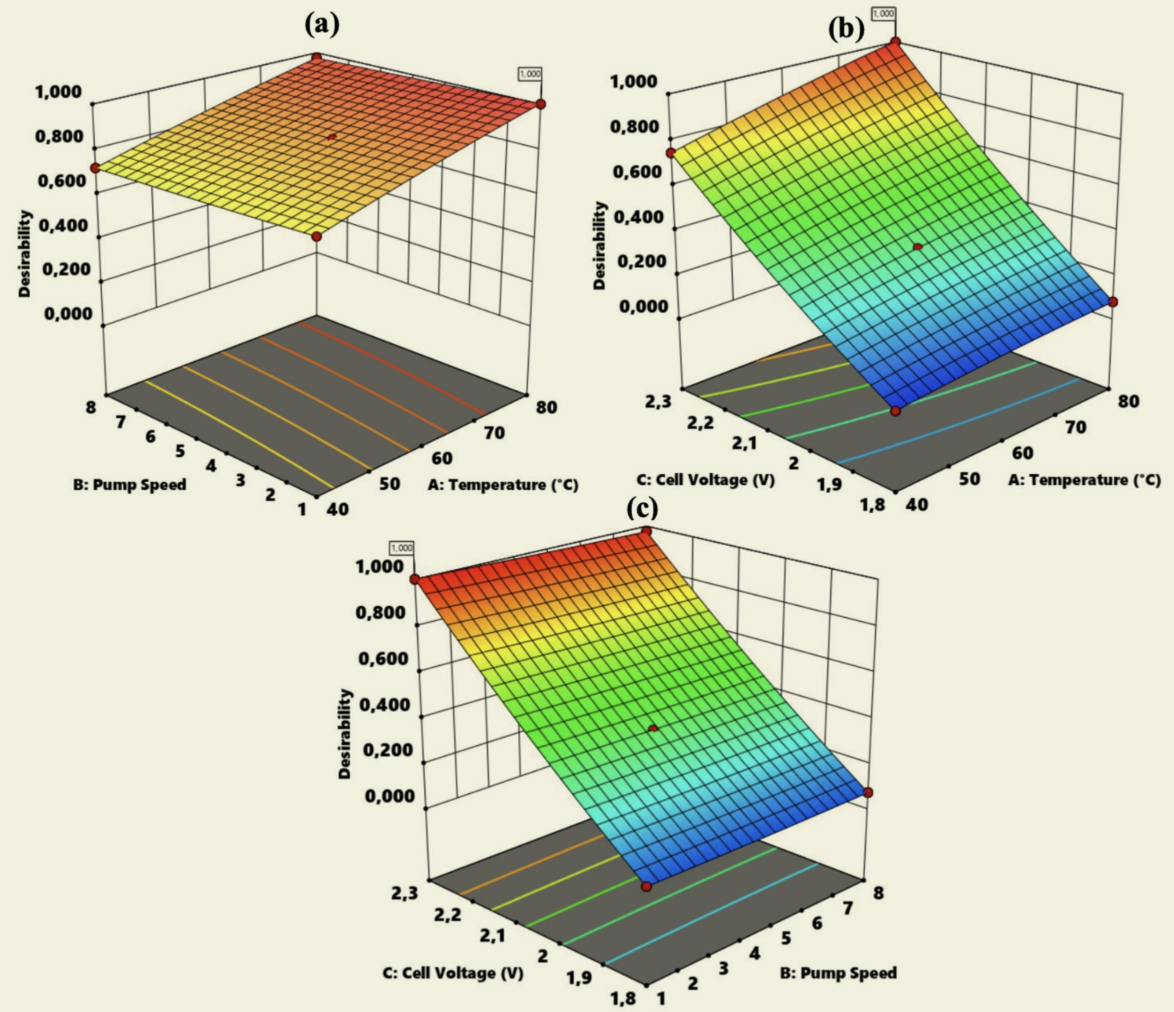

Figure 3. The desirability plots as a function of temperature and pump speed (a), temperature and cell voltage (b), pump speed and cell voltage (c)

of 80 °C, pump speed of 1, and cell voltage of 2.3 V. Figure 3 shows the desirability plots as a function of control factors. The excellent agreement of the experimental and optimization results and the high desirability value show that a reliable and accurate model has been developed. The intersection point of the operating conditions shows the optimum point and is shown on the perturbation plot (see Figure 4). The cell voltage has the steepest slope compared to the others, which means it is the most influential factor on the current.

4. CONCLUSIONS

The performance tests of a PEM-EC having a 9 cm^2 active area layer under different operational conditions have been conducted to investigate the influences of temperature, pump speed or water flow rate, and cell voltage on the cell performance. The central-composite design (CCD) and response surface methodology (RSM) were used to identify the optimal operating conditions for boosting the output current. It was concluded that the main factors affecting the performance of the PEM-EC are the cell voltage and temperature. On the other hand, the increase in pump speed did not have a significant effect on cell performance. The rise in temperature and cell voltage accelerates the electrolysis of water, and the formation of oxygen bubbles in the PEM electrolysis cell increases, so the evacuation of oxygen bubbles from the outside of the cell is provided with a low pump speed. The maximum current value of 16.778 A was obtained at 80 °C, pump speed of 1, and cell voltage of 2.3 V.

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