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Sensitivity analysis of fuel cell operating conditions based on LOESS-Sobol method

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Abstract

Decoupling the influence of operating factors such as temperature, humidity, and back pressure is essential for improving fuel cell efficiency. This study employs a regression model combining Locally Weighted Scatterplot Smoothing (LOESS) with the Sobol index method to analyze the effects of two major parameter categories: temperature and humidity, and stoichiometry and back pressure, on fuel cell output voltage. Using limited experimental data, it uncovers complex interactions and sensitivities among these factors. The results indicate that the stack temperature has the greatest impact, accounting for 60%, with minimal influence from the changes in the current density. Cathode humidity impacts output voltage by about 10%, while anode humidity accounts for approximately 5%. Significant interactions between temperature and both anode and cathode humidity contribute around 9% each. Among stoichiometries, cathode stoichiometry has the largest impact, exceeding 50% at low current density and growing to over 70% as current increases. Back pressure and anode stoichiometry each have an impact of around 10%, with minimal mutual influence. This study highlights the extent to how different operating parameters influence fuel cell performance, offering valuable insights for optimizing fuel cell operating conditions.

Keywords

Fuel Cell, Sensitivity Analysis, LOESS Regression, Sobol Index, Operating Conditions

Introduction

Fuel cells have garnered significant attention in the energy sector due to their high efficiency and environmental friendliness[1,2]. They convert chemical energy directly into electrical energy through an electrochemical reaction, with water and heat as the only byproducts. This clean energy conversion process positions fuel cells as a promising alternative to conventional fossil fuel engines. However, the performance of fuel cells is highly sensitive to various operating conditions in practical applications. Factors such as temperature, pressure, humidity, and stoichiometry can significantly influence the output voltage and consequently the efficiency of the fuel cell[3,4]. For example, maintaining an optimal temperature is crucial because it affects the reaction kinetics, membrane hydration, and overall cell resistance. Similarly, the relative humidity of the reactant gases, particularly in polymer electrolyte membrane (PEM) fuel cells, plays a vital role in maintaining membrane conductivity and preventing dehydration. Recent studies, using both simulation and experimental analyses, have investigated the effects of different operating conditions on the performance of the fuel cell stack[5–7].

Salva et al. used neutron imaging to validate cell voltage and water content in a PEM fuel cell model under various operating conditions, providing insights into the internal water distribution and its impact on cell performance[5]. Ozen et al. explored the impact of stack temperature and humidity on fuel cell performance through experimental methods, finding that increased humidity positively affects the fuel cell, and performance improves significantly with rising stack temperature until a certain threshold, beyond which performance fades[6,8]. Using numerical analysis, Jeon

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et al. discovered that cathode humidity strongly influences fuel cell performance, primarily due to electrode kinetics, including reaction rates and mass diffusion[7,9]. Kim et al. simulated a three-dimensional non-isothermal fuel cell stack, revealing that ohmic losses are affected by the humidity of the anode and cathode inlet gases, while concentration losses are mainly only influenced by cathode inlet gas humidity[10]. Zhang et al. experimentally demonstrated that fuel cell performance is highly dependent on operating temperature, back pressure, and stoichiometry[11]. Xing et al. through a two-dimensional model, discovered that while an initial increase in stoichiometry significantly boosts performance, additional increments lead to only marginal gains[12]. Wang et al. observed that fuel cell performance improves with increasing pressure through experimental studies[13]. In contrast, Askaripour et al., using a two-phase flow model, noted that for medium to high current density, the performance of fuel cells decreases with increasing cell pressure[14]. In recent years, some researchers employed sensitivity analysis methods to quantify the importance of various parameters in fuel cell models[15,16]. Brahim et al. applied global sensitivity techniques to a proton exchange membrane fuel cell (PEMFC) power output simulation model, identifying current density, temperature, membrane thickness, gas diffusion layer thickness, and porosity as sensitive parameters, with current density and membrane thickness having the most significant impact[17]. Zhang et al. used orthogonal experiments combined with the entropy weight method to comprehensively evaluate the performance of PEMFC under multiple performance objectives[18].

The Sobol index, a variance-based global sensitivity analysis technique, assesses the influence of each input parameter and their interactions on model output by calculating their contributions to output variance. Qian et al. conducted a global sensitivity analysis on an electrochemical model, computing the first-order, second-order and total Sobol index to quantify the individual impact, pairwise interactions and overall coupling effects of each parameter across the entire input parameter space[19]. Zhou et al. using a two-dimensional real-time modeling approach for PEMFCs combine with Sobol indices, analyzed parameter sensitivities and interactions[20]. Fan et al. performed a global sensitivity analysis using the Sobol index on a one-dimensional, two-phase, non-isothermal PEMFC model, finding that cathode humidity has the most significant impact on output performance among the operating parameters[21]. These studies quantified the effects of different parameters on fuel cell performance through numerical modeling and global sensitivity analysis. However, actual fuel cell operation often involves more complex water-thermal coupling. For instance, water management is critical in PEMFCs because excess water can lead to flooding, which obstructs gas transport, while insufficient water can cause membrane dehydration and increased resistance. Moreover, humidity and thermal cycling have substantial effects on the structural changes in the catalyst layer of fuel cells[22,23]. From the perspective of reaction kinetics, an increased back pressure enhances performance by improving the reactant gas distribution and increasing the partial pressures of the reactants, which accelerates reaction kinetics, leading to higher cell voltage and efficiency. However, excessive back pressure introduces mechanical stress and results in parasitic power losses. Optimizing stoichiometry is crucial to ensure an adequate supply of hydrogen and oxygen, which helps to maintain optimal reaction kinetics by reducing fuel starvation and ensuring a uniform concentration distribution across the catalyst layer[24–26]. These intricate interactions can significantly affect the overall performance of fuel cells. Current research lacks sufficient quantification of the impact of various operating conditions on fuel cell output performance under real-world conditions.

To address this challenge, this study employs a novel combination of the local weighted scatterplot smoothing (LOESS) regression model and Sobol sensitivity analysis. The LOESS regression model effectively simulates the non-linear relationship between different operating conditions and the fuel cell output voltage under experimental conditions, predicting the output performance under new operating conditions. Experimental data, combined with prediction results, are used to calculate Sobol indices for a comprehensive global sensitivity analysis. The experiment utilizes the controlled variable method, categorizing the operating conditions into two primary modules: the temperature and humidity module, which includes stack temperature, anode and cathode humidity, and the gas supply module, including back pressure and anode and cathode stoichiometry, with each module encompassing three independent variables. This research offers valuable insight into the sensitivity of fuel cell output performance to a range of operating parameters. The innovative fusion of experimental data with data science techniques has facilitated a comprehensive global sensitivity analysis of the operating parameters, providing insightful guidance for the design of more efficient and dependable fuel cell operating conditions.

The remainder of this paper is organized as follows. In Section 2, the experimental setup and procedure are introduced. Section 3 presents the LOESS model and

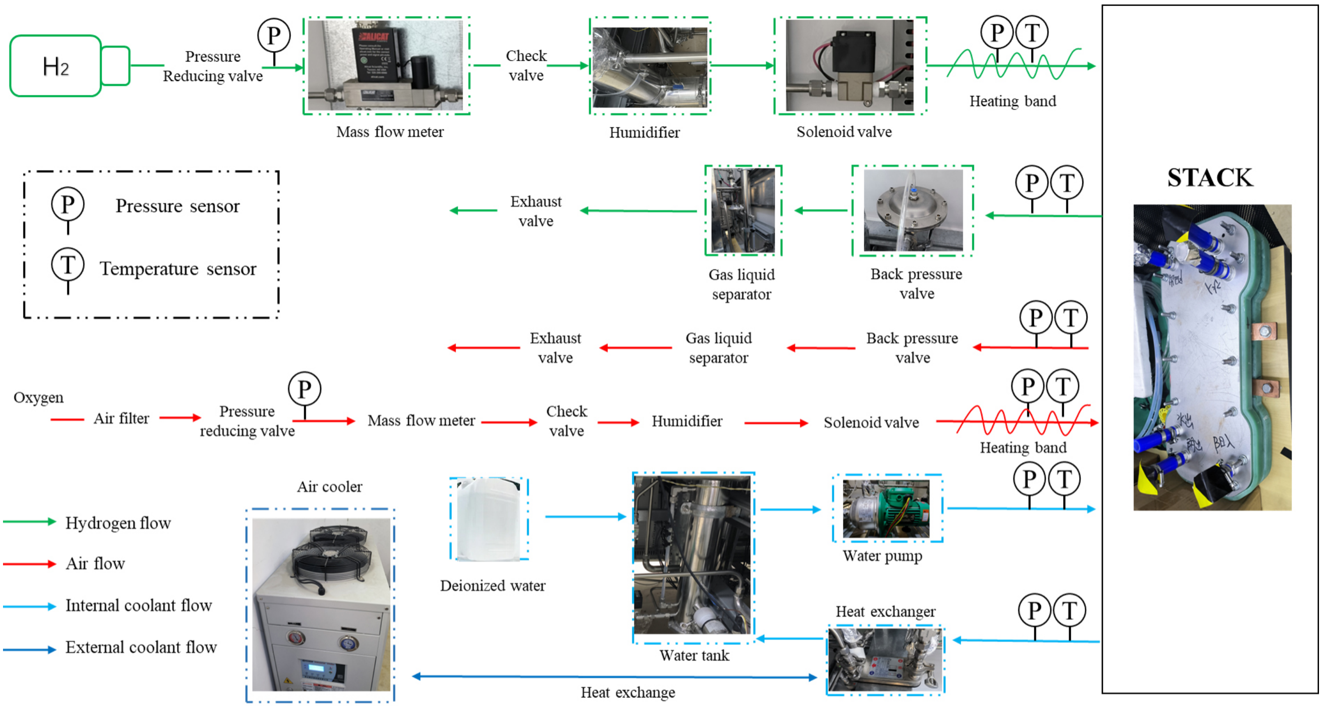

Figure 1. Experimental setups.

the Sobol sensitivity analysis method. In Section 4, the influence of various factors is quantified, the significance and interaction of each factor are analyzed, and variations in the impact of these factors are investigated at different current levels. Finally, conclusions are drawn in Section 5.

Table 1. Specific parameters of the PEMFC.

Feature Parameter	Value
Number of cells	3
Rated power	1 kW
Active area	300 cm^2
Proton exchange membrane thickness	12 μm
Catalyst loading	0.35 mg/cm^2
Cooling method	Water-cooled

Experimental setup

PEMFC Equipment

The study utilizes a fuel cell stack with a capacity of approximately 1 kW. The fuel cell stack has been used intermittently for more than half a year and has experienced degradation, making it more sensitive to changes in operating parameters. The PEMFC components consist of commercial membrane electrode assembly (MEA) and metallic bipolar plates. The cathode features a straight channel flow field, while the anode employs a serpentine flow field, with an inlet size of 3/8 inches. Figure 1 shows the test bench for the 2 kW fuel cell stack, with an electronic load current range of 0 - 600 A and voltage range of 0.1 - 40 V. The specific parameters are listed in Table 1.

The stack is water-cooled, with the test bench using internal deionized water and external cooling water to control the stack outlet temperature through heat exchange. The gas is humidified using a combination of bubbling and spraying, and heated using heating tapes to control the intake air temperature. The humidity is controlled by adjusting the dew point and intake air temperature. The flow rates of hydrogen and air are controlled by high-precision mass flow meters. A diaphragm back pressure valve controls the intake pressure, and the anode is equipped with a gas-liquid separator to reduce the impact of anode flooding on performance.

Experimental Procedure

To avoid the curse of dimensionality, which can reduce the performance of the model, the operating conditions are categorized into two groups based on their correlation: one group consists of the temperature and humidity of the anode and cathode, while the other group includes the back pressure and stoichiometry of both. To obtain the output voltage under various operating conditions, the control variable method is used in the design of experiments (DoE) process. Prior to sensitivity testing, a standard operating condition is established as a baseline, based on the optimal operating point recommended by the MEA supplier, as shown in Table 2.

During each sensitivity test, only one operating condition is altered to its preset value, while the other conditions

Table 2. Baseline operating conditions.

Parameter	Value
Stack temperature (C)	70
Cathode humidity (%)	90
Anode humidity (%)	90
Back pressure (kPa)	0
Cathode stoichiometry	1.5
Anode stoichiometry	3

are kept at their standard values. The parameter settings for each test condition are listed in Table 3 and Table 4 (Appendix A). Each test lasts approximately 15 minutes and involves altering six dependent variables and recording the corresponding voltage values. To ensure stability, the fuel cell is operated under stable conditions for 20 minutes prior to each test. The impact of varying influences on the output voltage of the fuel cell is measured at different current levels. Specifically, currents of 60A and 120A correspond to low to medium current densities of 200 mA/cm^2 and 400 mA/cm^2, respectively, while currents of 180A and 240A correspond to medium to high current densities of 600 mA/cm^2 and 800 mA/cm^2, respectively.

LOESS-Sobol Method

In this section, the theoretical foundations and calculation methods of the LOESS regression model and the Sobol index are introduced separately. Then, it is explained how these two are fused to obtain a more detailed and practical analyzing tool.

LOESS regression model

The LOESS model is employed to capture the non-linear relationships between the input parameters and the output voltage[27]. LOESS fits local polynomial curves to data subsets, allowing it to flexibly model complex, nonlinear data patterns. It applies a locally weighted regression to each data point[28], tailoring the model to the local characteristics of the data. The LOESS fit at a given data point is expressed as:

$$\hat{f}\left(x_0\right) = \Sigma_{i=1}^{N} \omega_i\left(x_0\right) y_i \tag{1}$$

where $\hat{f}\left(x_0\right)$ is the estimated value at the prediction point x_0, $\omega_i\left(x_0\right)$ is the weight at the given point x_i, and y_i is the observed value of the i^{th} data point. The weights $\omega_i\left(x_0\right)$ are typically based on the distance between the data point and the point of interest, with closer points receiving higher weights. The tricube weight function is commonly used:

$$\omega_i\left(x_0\right) = \left(1 - \left(\frac{d_i}{D}\right)^3\right)^3 \tag{2}$$

where d_i is the distance from the point x_i to x_0, and D is the maximum distance within the neighborhood. Within each local region, LOESS uses weighted least squares to fit a low-order polynomial (usually a first or second order polynomial). For a first order polynomial, the fitting process is expressed as:

$$\min_{\beta_0,\beta_1} \sum_{i=1}^{N} \omega_i(x_0)\left(y_i - (\beta_0 + \beta_1(x_i - x_0))\right)^2 \tag{3}$$

where β_0 and β_1 are the regression coefficients. By minimizing the weighted sum of squared residuals, the local regression coefficients are obtained, leading to the estimated value $\hat{f}\left(x_0\right)$ at the prediction point x_0.

Sobol index estimation

To quantify the impact of different operating conditions on the output voltage of the fuel cell, this research uses the Sobol sensitivity analysis method[29]. This method is advantageous for its ability to decompose the variance of the output into contributions from each input variable and their interactions, thereby providing a comprehensive global sensitivity analysis. It is applicable for quantifying the impact of various operating conditions on the voltage variations of the fuel cell during actual operation. The Sobol index is a variance-based method quantifying the sensitivity of the output with respect to each input parameter.

Firstly, the total variance of the output $V(\mathbf{Y})$ can be decomposed as follows:

$$\begin{aligned} V\left(\mathbf{Y}\right) = & \sum_{i=1}^{k} V_i + \sum_{1\leq i<j\leq k} V_{ij} \\ & + \sum_{1\leq i<j<l\leq k} V_{ijl} + \cdots + V_{12\cdots k} \end{aligned} \tag{4}$$

where $\mathbf{Y}$ is the combination matrix of operating conditions of the fuel cell, V_i is the variance contribution of the main effect of the i input variables, V_{ij} is the variance contribution from the interaction between the input variables i and j . The calculation method of Sobol index is to be introduced.

Then, the total Sobol index S_{T_i} represents the total effect of the input variable i, including its main effect and all interactions with other variables.

$$S_{T_i} = 1 - \frac{V_{\sim i}}{V(\mathbf{Y})} \tag{5}$$

where $V_{\sim i}$ is the variance of the output when the i input variable is fixed.

The first-order Sobol index S_i represents the individual effect of the given input variable i:

$$S_i = \frac{V_i}{V(\mathbf{Y})} \tag{6}$$

Similarly, also known as the interaction Sobol index, the second-order Sobol index S_{ij} represents the variance contribution of the interaction between the input variables i and j:

$$S_{ij} = \frac{V_{ij}}{V(\mathbf{Y})} \tag{7}$$

In addition, in practical applications, Monte Carlo method are frequently employed to estimate V:

$$S_i = \frac{\frac{1}{N}\sum_{j=1}^{N}\left(f(A_{B_j}^{(i)}) - f(A_j)\right)f(B_j)}{V(\mathbf{Y})} \tag{8}$$

$$S_{T_i} = \frac{\frac{1}{2N}\sum_{j=1}^{N}\left(f(A_{B_j}^{(i)}) - f(A_j)\right)^2}{V(\mathbf{Y})} \tag{9}$$

where A and B are matrices, each containing N samples of $O(d)$ input variables. $A_{B_j}^{(i)}$ is a matrix where the i^{th} column is from B, while the remaining columns are from A.

LOESS-Sobol fusion analysis

The construction process of the LOESS-Sobol index analysis method is as follows: The temperature block (stack temperature, cathode humidity, anode humidity) and the gas supply block (back pressure, cathode stoichiometry, anode stoichiometry) each contain three variables, collectively forming the feature matrix. To ensure consistency, the experimental data are normalized. The LOESS model is then trained using the standardized feature matrix and voltage data, with a specified bandwidth parameter that determines the smoothness of the fitted curve. This results in a locally weighted regression model to predict voltage values under different operating conditions. After normalizing the sample results, they are input into the LOESS model to predict the output voltage for each sample. Subsequently, the Sobol sensitivity analysis is performed to calculate the first-order Sobol index, total Sobol index, and second-order Sobol index, quantifying the impact of each operating condition on fuel cell output voltage variations.

To validate the error margins of the model under new operating conditions, additional experimental data are collected. This involves eight groups of experimental data where only one condition is changed at a time: back pressure set to 50 kPa and 100 kPa, cathode stoichiometry of 2.5 and 3, and anode stoichiometry of 2 and 3, while keeping other operating conditions constant. The predicted error results are shown in Figure 2. According to the experimental results, errors are found to be within 5%, demonstrating that the regression model reliably fits the data trends.

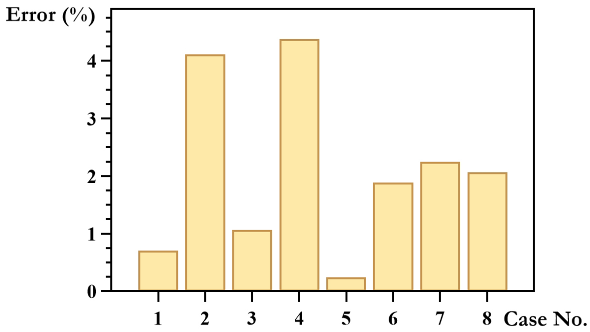

Figure 2. The predicted error of LOESS method.

Results and Discussion

In this section, the experimental results of output voltage under different operating conditions are presented. The analysis is primarily divided into two major parts: one examines the impact of temperature and humidity on the output voltage, and the other investigates the effects of back pressure and the stoichiometry of anode and cathode on the output voltage. The output voltage under various combinations of operating conditions is predicted using the LOESS method, and the contribution of each operating condition to the output voltage is quantified using the Sobol index. Furthermore, the Sobol index under different current densities is also analyzed.

Performance impact of temperature and humidity

Temperature and humidity play pivotal roles in fuel cell performance, affecting electrochemical reaction rates, catalyst activity, and membrane hydration. Optimal conditions enhance reaction kinetics and maintain proper water management within the cell, crucial for preventing operating issues like flooding or dehydration.

Through controlled variable experiments, it is observed that at low current density, the performance of fuel cells initially increases with rising temperature. This is because the increase in temperature enhances the activity of the fuel cell catalyst and accelerates the reaction rate. Additionally, the conductivity of the PEM is also temperature-dependent. At lower temperatures, poor hydration of the PEM results in decreased conductivity. As the temperature increases, improved hydration of the

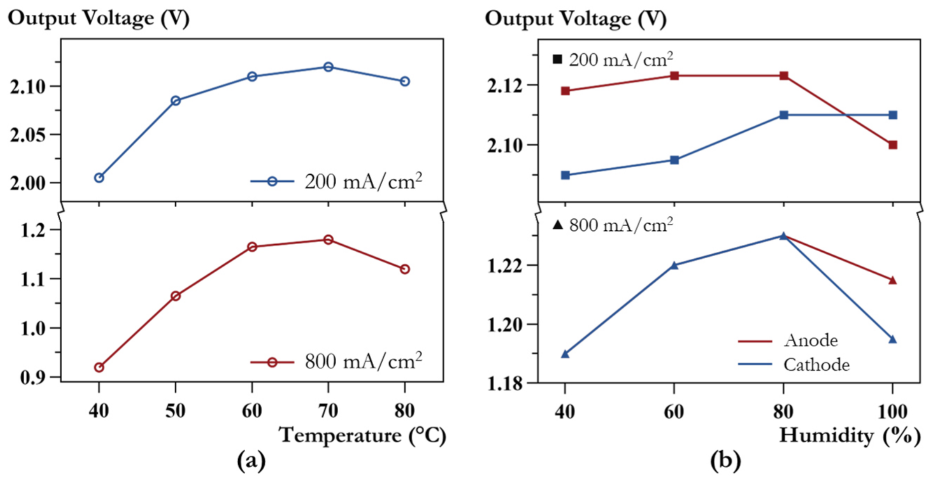

Figure 3. The impact of operating conditions on the FC output voltage: (a) Temperature, and (b) humidity of anode and cathode.

PEM leads to increased conductivity, thus increasing the voltage. Shown in Figure 3a, when the temperature reaches approximately 70C, the performance of the fuel cell reaches its peak. Further increases in temperature result in a decrease in fuel cell voltage, consistent with observations reported in the literature[8]. This decline may be due to the negative effects of excessive temperature on the thermal stability and proton conductivity of the PEM, which may cause thermal degradation or reduced chemical stability. At high current densities, a similar trend is observed, but the output voltage fluctuates more significantly with temperature. This could be due to the increased challenges in thermal and water management at higher current densities. The greater heat generation at high current densities necessitates more effective cooling mechanisms, otherwise temperature fluctuations can have a more pronounced impact on performance.

Humidity also makes sense on the output voltage (Figure 3b). At low current density, increases in the anode and cathode humidity improve the hydration of the PEM and catalyst, thereby increasing the fuel cell output voltage. The impact of cathode humidity on the fuel cell output voltage is greater than that of anode humidity because the oxygen reduction reaction at the cathode requires a well-hydrated environment to maintain reaction rates and conductivity. When the humidity reaches 90%, the performance of the fuel cell peaks. Further increases in humidity lead to a decrease in output voltage as a result of excessive water blocking the flow channels and diffusion layer pores, causing localized flooding within the fuel cell and hindering the transport of reactants. However, at high current densities, the fuel cell output voltage is less sensitive to changes in anode and cathode humidity compared to low current densities. This may be because the increased water production during high current density operation provides sufficient internal hydration, reducing the need for external humidification. Therefore, the higher water production at high current densities may alleviate the dependency on external humidity adjustments[30].

The predicted voltage map at different temperatures and multiple anode and cathode humidity levels at low current densities is visualized by Figure 4. Optimal performance regions appear when the temperature is between 60 - 70C, the cathode humidity is between 70 - 90%, and the anode humidity is between 65 - 90%. This indicates that under these conditions, the fuel cell achieves a balanced thermal and water management, resulting in stable operation. Even with minor disturbances, the fuel cell can still output a high voltage. In contrast, the worst performance is concentrated in regions of low temperature and high humidity. At low temperatures, the reaction kinetics of the fuel cell is slower, and high humidity leads to flooding, further reducing the performance. Further observations show that along the temperature axis, there are more significant changes in the color scale, followed by the cathode humidity axis, and finally the anode humidity axis. This indicates that temperature has the most pronounced effect on fuel cell performance, followed by cathode humidity and anode humidity. As shown in Figure 4b, at high current densities, the impact of the anode and cathode humidity on the output voltage of the fuel cell is not significant. Performance is poor under conditions of excessive humidity. Therefore, the optimal performance region at high current densities is found at temperatures of 60 - 70C and humidity levels of 70 - 90%. Similarly, the worst performance is concentrated in regions of low temperature and high humidity.Another area of poor performance is concentrated in the low-humidity region, possibly due to the insufficient moisture in the gas leading

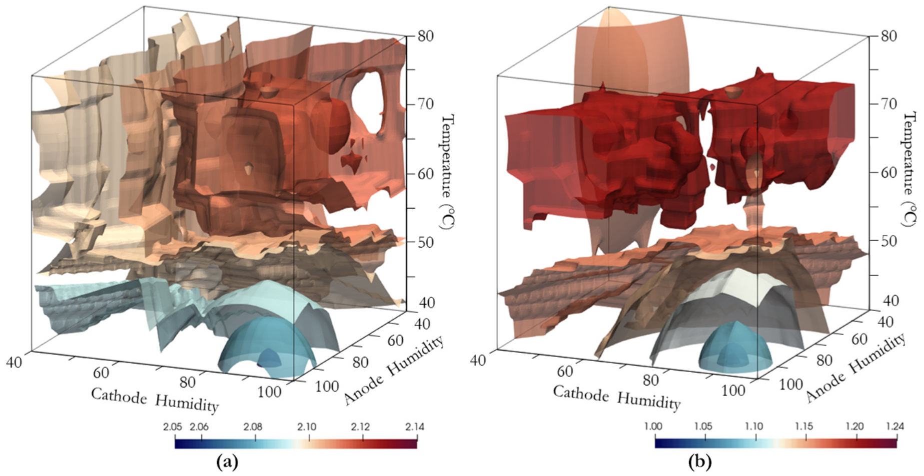

Figure 4. Predicted output voltage contours under different temperature and humidity of anode and cathode: (a) at low current density and (b) high current density. Another visualization by slices showing the same dataset is shown in Figure 11 (Appendix B).

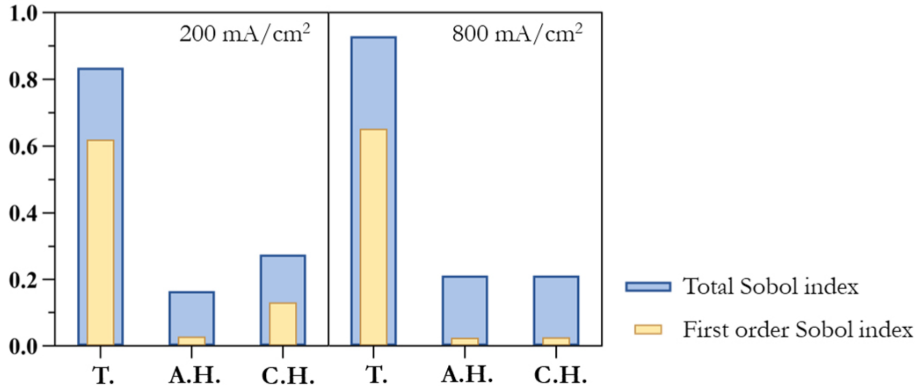

Figure 5. Sobol sensitivity analysis results. T, A.H., and C.H. are representing temperature, anode humidity, and cathode humidity, respectively.

to reduced proton conductivity and increased resistance within the cell. The output voltage of the fuel cell shows a more pronounced temperature stratification, indicating that the impact of temperature on the fuel cell continues to be enhanced.

Sensitivity analysis of temperature and humidity

Sobol index analysis reveals the quantitative effects of temperature and humidity on the performance of electrochemical systems. By examining the variations in Sobol indices at different current levels, the sensitivity of fuel cell output voltage to temperature and humidity of anode and cathode under various currents can be identified.

Figure 5 plots the total Sobol index of temperature and electrode humidity as well as their first-order component. Under low current density conditions, the total Sobol index of stack temperature in output voltage is 83.5%. It indicates that temperature is a decisive factor in influencing fuel cell performance. It can be explained by the significant impact of temperature on the electrochemical reaction rate and proton conductivity. Higher temperatures can accelerate electrode reactions and improve proton conductivity, thereby enhancing fuel cell performance. The total impact coefficients of the cathode humidity and the anode humidity on the performance of fuel cells are 27. 5% and 16. 5%. Humidity affects the hydration state of the membrane and ion conductivity; appropriate humidity can optimize membrane functionality and improve fuel cell efficiency and stability[9]. The first-order Sobol index indicates that the individual impact of temperature on fuel cell voltage is 61.9%, which is significantly higher than that of cathode humidity (13.1%) and anode humidity (2.9%), further confirming the dominant role of temperature in fuel cell performance. The second-order Sobol index shows that the interactions between temperature and cathode humidity, and temperature and anode humidity are 7.6% and 8.5%, respectively, while the interaction between cathode and anode humidity contributes only 0.5%. This suggests that temperature and humidity have a significant synergistic effect on optimizing the hydration state of the membrane and the reaction environment, even surpassing the individual

Figure 6. Different order of Sobol indices along with temperature and humidity at different current densities.

impact of the anode humidity. However, the interaction between cathode and anode humidity is relatively minor.

Under high current density conditions, the total Sobol index for the stack temperature's effect on fuel cell performance increases to 92.9%, underscoring its critical role, which aligns with previous experimental findings. The first-order Sobol index indicates that the direct impact of temperature on fuel cell voltage remains stable at 65.3%, while the influence of cathode humidity decreases to 2.6%, and that of anode humidity stays relatively constant at 2.5%. These results suggest that, under high current density, the dominant effect of temperature on fuel cell performance is not solely due to its independent contribution but rather through its synergistic interactions with humidity. The second-order Sobol index highlights that the interaction between temperature and cathode humidity accounts for 10.8%, while the interaction between temperature and anode humidity contributes 11.2%. Additionally, the interaction between anode and cathode humidity accounts for 2.3%. These findings illustrate that temperature and humidity exhibit a synergistic effect in enhancing membrane hydration and optimizing the reaction environment under high current density conditions, with the combined influence of temperature and humidity, particularly with both cathode and anode humidity, becoming more pronounced.

In practical applications, maintaining an optimal operating temperature and appropriate external humidification is crucial for enhancing fuel cell performance and stability. Among these, maintaining an optimal temperature plays a decisive role, as increasing the temperature can significantly boost the electrochemical reaction rate. However, excessively high temperatures can disrupt membrane hydration and other critical factors. Therefore, complementing temperature control with appropriate external humidification to maintain water balance within the fuel cell can effectively ensure operation within a high-performance range.

The trends of the total Sobol index and the first-order Sobol index exhibit a consistent pattern across different currents (Figure 6). As the current increases, the output voltage of the fuel cell becomes more sensitive to temperature. Concurrently, the differential impact of anode and cathode humidity on fuel cell performance decreases with increasing current, which may be related to water transport between the anode and cathode. In the second-order Sobol index, it is observed that all three indices increase with the current. This indicates that at higher currents, the interaction between thermal and humidity effects becomes more pronounced, significantly influencing the output voltage of the fuel cell. Higher currents exacerbate water management challenges, necessitating effective control of temperature and humidity to avoid performance degradation.

It is concluded that temperature is indicated to be the main factor affecting the global system performance, either through a direct impact on fuel reaction kinetics or through its combined effect with humidity. This influence becomes more significant at higher currents. Although the impact of humidity may not be as significant, its combined effect with temperature is crucial. This underscores the necessity of integrated thermal and water management strategies to achieve optimal system operation.

Performance impact of back pressure and stoichiometry

Back pressure and stoichiometry significantly influence fuel cell performance. Higher back pressure can enhance reactant utilization and power density but may also increase polarization losses. Optimal stoichiometry ensures efficient fuel and oxidant supply, balancing power output and fuel consumption. Therefore, selecting the appropriate back pressure and stoichiometry is crucial for maximizing fuel cell output and efficiency.

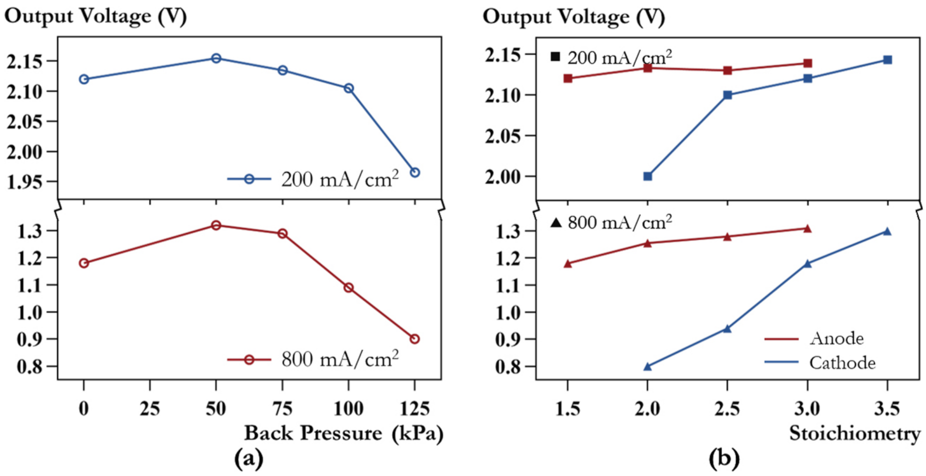

Figure 7. The impact of operating conditions on the FC output voltage. (a) Back pressure, and (b) stoichiometry of anode and cathode.

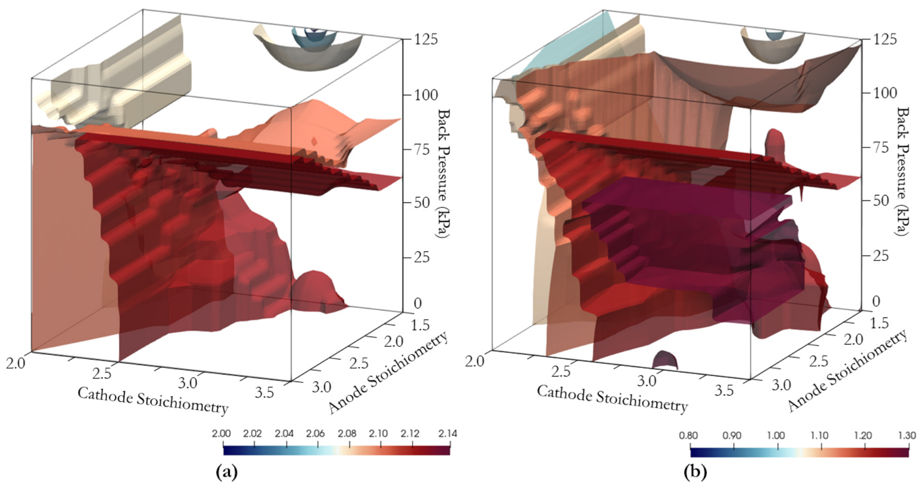

Figure 8. Predicted output voltage contours under different pressure and stoichiometry of anode and cathode: (a) at low current density and (b) high current density. Another visualization by slices showing the same dataset is shown in Figure 12 (Appendix B).

First, back pressure is experimentally studied and discussed. Figure 7a shows the impact of varying pressures on the output voltage. The output voltage of the fuel cell gradually increases with increasing back pressure, reaching its peak at 50 kPa. Beyond this point, further increases in back pressure cause a rapid decline in the output voltage. An appropriate back pressure improves the electrochemical reaction rate [13]. However, when the back pressure is too high, the resistance to gas transport increases, making it difficult for oxygen and hydrogen to effectively reach the reaction sites, which leads to a reduction in the reaction rate [14]. Additionally, high back pressure can cause water accumulation inside the cell. Since the back pressure equipment of the test bench is located at the fuel cell outlet, high pressure prevents moisture from being promptly expelled, resulting in water buildup within the cell. This accumulation obstructs gas transport, leading to local oxygen deficiency and thus a decrease in the output voltage.

As the cathode stoichiometry increases, the fuel cell output voltage rises rapidly at first, then the rate of increase gradually slows down. This is because, at lower cathode stoichiometry, oxygen supply is insufficient, leading to a low oxygen reduction reaction (ORR) rate and consequently a low output voltage. With an increase in cathode stoichiometry, the oxygen supply improves, enhancing the ORR rate and significantly increasing the output voltage. When the oxygen supply approaches its saturation state, further increases in the cathode stoichiometry have little effect on the output voltage, causing the voltage rise to level off. This is because the reaction sites are adequately supplied with oxygen, and the additional supply of oxygen does not significantly improve the reaction rate [12].

The impact of anode stoichiometry on fuel cell performance is relatively insensitive but generally shows a positive correlation. Figure 8a shows the predicted heatmap of voltage under different back pressures, anode and cathode stoichiometry at low current densities. The performance of

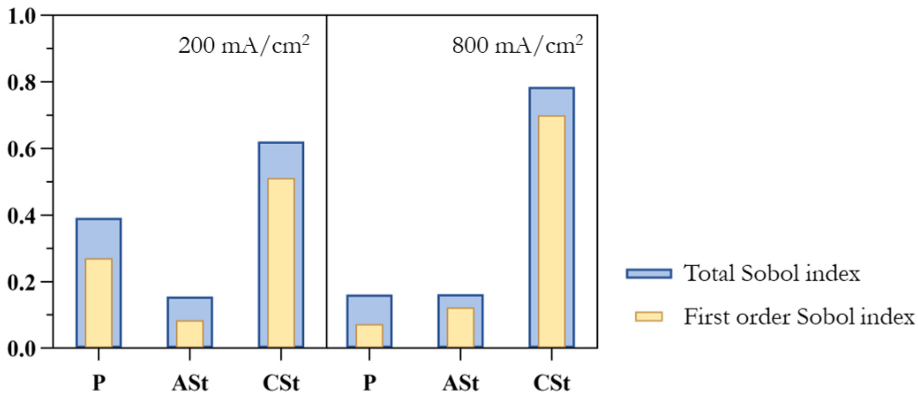

Figure 9. The Sobol sensitivity index of pressure and stoichiometry of anode and cathode.

fuel cells is notably suboptimal in regions characterized by low anode stoichiometry and high back pressure. Optimal performance areas are identified at a back pressure of 50 kPa and with higher anode and cathode stoichiometric ratios. Further increases in the stoichiometric ratios of the cathode and anode lead to continued, albeit less significant, improvements in the output voltage, as the reactant gases for the electrochemical reactions approach saturation. It is evident that the output voltage of fuel cells fluctuates significantly with changes in the cathode stoichiometry, particularly when the cathode stoichiometry is below 2.5.

Figure 7b shows the impact of varying the stoichiometry of the anode and cathode on the output voltage. At high current densities, the demand for air by the fuel cell significantly increases, making it more sensitive to changes in back pressure and stoichiometry of anode and cathode. Consequently, the fuel cell becomes more sensitive to changes in cathode and anode stoichiometry. As the cathode stoichiometry increases, the output voltage rises rapidly due to sufficient reactant gas supply, markedly improving the ORR rate. The trend in changes in anode stoichiometry is also more pronounced, with an increase in the hydrogen supply that enhances the performance of the fuel cell. High current densities generate more heat and water, making the effect of back pressure changes more drastic. Appropriate back pressure can facilitate water management and prevent water accumulation, but excessive back pressure can lead to water accumulation and local oxygen deficiency, affecting performance. Figure 8b shows the predicted heatmap of the voltage under different back pressures, the stoichiometry of the anode and cathode at high current densities. Areas of unsatisfactory performance can be observed under conditions of low anode and cathode stoichiometry, while optimal performance regions are identified at pressures below 50 kPa and with high cathode stoichiometric ratios. As the current density increases, the impact of cathode stoichiometry on the output voltage is further exacerbated, with more pronounced color changes in the direction of cathode stoichiometry.

Sensitivity analysis of back pressure and stoichiometry

Quantifying the impact of back pressure and stoichiometric on output voltage can aid in gaining a deeper understanding of the working principles and performance optimization of fuel cells. Through Sobol index, it is possible to determine how these factors affect the voltage and power output of the fuel cell. Figure 9 displays the Sobol indices as the back pressure and stoichiometry change.

At low current density, cathode stoichiometry is the primary factor that affects fuel cell performance, accounting for 62.1% of the total impact on performance. This is because that the cathode oxidation reaction is the rate-determining step, making variations in the cathode stoichiometry have the most significant effect on fuel cell output voltage. The back pressure contributes 39.2% to performance, as increasing the back pressure can enhance reaction rates according to the principles of reaction kinetics. The anode flow ratio contributes 15.6%. The first-order Sobol index indicates that the individual impacts of the cathode flow ratio, back pressure, and anode flow ratio on voltage are 51.1%, 27.0%, and 8.5%, respectively. The second-order Sobol index shows interactions between back pressure and cathode flow (5.0%), back pressure and anode flow (2.9%), and cathode and anode flow (0.5%), indicating that the mutual effects of back pressure and stoichiometry are relatively low.

At high current density, the impact of cathode stoichiometry on fuel cell output voltage further increases, with its total Sobol index reaching 78.5%. The impact of back pressure on performance decreases to 16.1%, while the anode stoichiometry slightly increases to 16.3%. The first-order Sobol index shows that the impacts of the cathode stoichiometry,

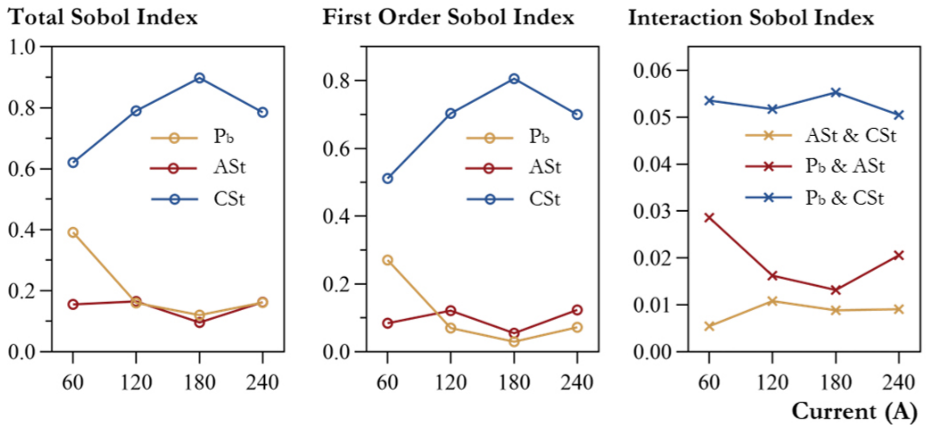

Figure 10. Different order of Sobol indices along with back pressure and Stoichiometry at different current densities.

back pressure, and anode stoichiometry on voltage are 70%, 7.3%, and 12.3%, respectively. As the current increases, the demand for hydrogen and oxygen in the fuel cell increases, and changes in the stoichiometry significantly affect the output voltage, especially when the stoichiometry is low, leading to oxygen starvation and a sharp decline in output voltage. Therefore, changes in back pressure have a relatively smaller effect on the output voltage of the fuel cell. The second-order Sobol index shows that the interactions between pressure and electrode stoichiometry are small, with the back pressure-cathode stoichiometry interaction at 5%, the back pressure-anode stoichiometry interaction at 2%, and the cathode-anode stoichiometry interaction at only 0.9%.

Overall, it is evident that the cathode stoichiometry directly influences the oxidation reaction rate of the fuel cell, which makes it the primary factor affecting output voltage, particularly when cathode gas supply is insufficient, causing significant voltage fluctuations. Appropriate back pressure can increase the partial pressure of gases in the fuel cell, significantly reduce the mass transfer resistance and cathode activation resistance, and lower the ohmic resistance and anode activation resistance, thus improving the reaction kinetics.

The impact of back pressure and stoichiometry on fuel cell output often exhibits differences under different current densities. Shown in Figure 10, the exploration of the Sobol indices for back pressure and cathode and anode stoichiometry under different current densities reveals significant trends. From the total Sobol index and the first-order Sobol index, it is evident that as the current increases, the influence of the cathode stoichiometry increases significantly by approximately 25%, while the influence of the anode stoichiometry slightly increases, and the impact of back pressure decreases. This is primarily due to the substantial increase in gas demand by the fuel cell at high current densities, with the cathode oxidation reaction being the rate-determining step. Consequently, the fuel cell becomes more sensitive to changes in the cathode stoichiometry. The cathode stoichiometry ensures an adequate supply of oxygen for the oxygen reduction reaction, which is crucial for maintaining high performance and preventing voltage drop due to oxygen starvation.

The second-order Sobol index indicates that the interactions between back pressure, cathode stoichiometry, and anode stoichiometry are relatively small, with values consistently below 5%, even as the current changes. This suggests that the effects of back pressure, cathode stoichiometry, and anode stoichiometry on fuel cell output voltage are relatively independent, with minimal mutual influence. The low interaction values confirm that optimizing each parameter can be approached independently, focusing primarily on the cathode stoichiometry at higher currents due to its significant impact.

Conclusion

The study employs experimental methods coupled with LOESS regression models and the Sobol index to analyze the trends and quantitative relationships of the impact of various operating conditions on the fuel cell output performance. Specifically, it examines the combinations of anode and cathode temperature and humidity, as well as the combinations of back pressure and stoichiometry of the anode and cathode. Furthermore, it explores how these operating conditions impact the output voltage of the fuel cell at different current levels. This work yields the following conclusions:

- Temperature and humidity of anode and cathode: The stack temperature is the decisive factor affecting fuel cell output voltage, independently accounting for 60% of the influence. When coupled with humidity, their combined effect accounts for 20% of the influence on fuel cell output voltage.

- Back pressure and stoichiometry of anode and cathode: The analysis indicates that cathode stoichiometry is the most critical factor, independently contributing 60% to 80% of the performance variation.
- Impact of current: As the current increases, the influence of temperature and cathode stoichiometry on fuel cell output voltage fluctuations becomes more significant.
- Interactivity: Temperature affects the output voltage indirectly through its coupling effect with humidity, while the stoichiometry of anode and cathode primarily influences the output voltage independently.
- Application: Based on our research findings, engineers are enabled to adjust the operating conditions of fuel cells to achieve optimal performance or energy consumption ratios with limited trials, and taking into account the practical constraints at the same time. Out research method is also applicable to other fuel cells, thereby providing a universal strategy for optimizing fuel cell performance.

Declaration of conflicting interests

The authors have no conflicts of interest to declare that are relevant to the content of this article.

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Appendix A: Operating conditions

Table 3. Stack temperature, cathode humidity and anode humidity during experimental studies.

I (A)	Stack temperature (C)	Cathode humidity (%)	Anode humidity (%)
60, 120	40, 50, 60, 70, 80	90	90
180, 240	40, 50, 60, 70, 80	90	90
60, 120	70	40, 60, 80, 100	90
180, 240	70	40, 60, 80, 100	90
60, 120	70	90	40, 60, 80, 100
180, 240	70	90	40, 60, 80, 100

Table 4. Back pressure, cathode stoichiometry and anode stoichiometry during experimental studies.

I (A)	Back pressure (kPa)	Cathode stoichiometry	Anode stoichiometry
60, 120	0	1.5, 2, 2.5, 3	3
180, 240	0	1.5, 2, 2.5, 3	3
60, 120	0	1.5	2, 2.5, 3, 3.5
180, 240	0	1.5	2, 2.5, 3, 3.5
60, 120	0, 50, 75, 100, 125	1.5	3
180, 240	0, 50, 75, 100, 125	1.5	3

Appendix B: Slice representations of results

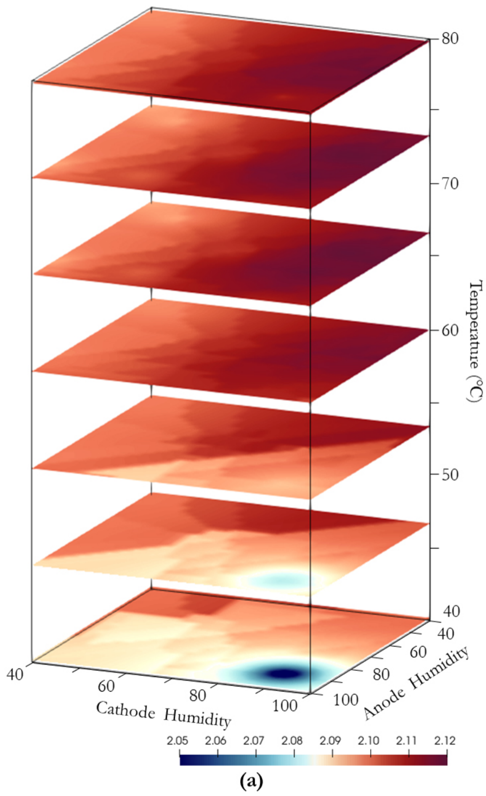

Figure 11. Slices of predicted output voltage under different temperature and humidity of anode and cathode: (a) at low current density, and (b) at high current density.

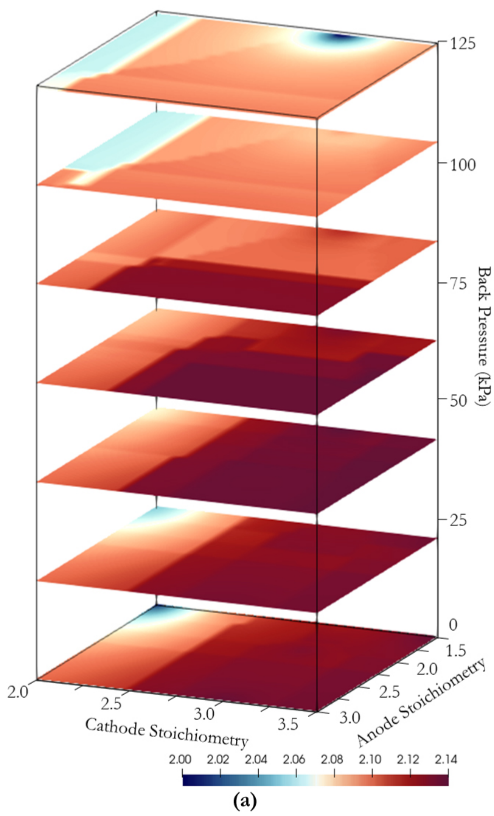

Figure 12. Slices of predicted output voltage under different pressure and stoichiometry of anode and cathode: (a) at low current density, and (b) at high current density.