

Simulation and Analysis of the Effects of Pressure and Temperature on the Output Voltage of Proton Exchange Membrane Fuel Cells

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Abstract

Proton exchange membrane fuel cell has advantages of high energy conversion efficiency, high reliability, no pollution, low operating temperature and rapid start-up. It has become an ideal method of hydrogen energy utilization and is also ideally suited to be used as the main source of energy for automobiles. Currently, it constitutes a research hot spot in the field of new energy vehicles. Based on the working mechanism of proton exchange membrane fuel cells and empirical models, a terminal voltage dynamic model, an open circuit voltage model and three voltage loss models are established. Matlab/Simulink software is utilized to simulate the model and perform analyses in response to the impact of operating temperature and pressure on its performance. To enhance the efficiency of the proton exchange membrane fuel cell, the operating temperature should be increased in the medium and low current density zones and the operating pressure should be increased in the high current density zone.

Keywords

Hydrogen Energy, New Energy Vehicles, Empirical Models, Voltage Dynamic Model

1. Introduction

Proton exchange membrane fuel cells have the advantages of high conversion efficiency, low operating temperature, high reliability and fast response. They are the most sought after, widely used and commercialized of many types of fuel cells. Proton exchange membrane fuel cell was first used on a small scale in aer-

ospace, military equipment and other fields [1] [2] [3] [4]. With the reduction of production costs and the advancement of related technologies, they are likely to become the main power source of new energy vehicles, and their application prospects are generally promising. However, to achieve large-scale commercialization, it requires extensive research into improving performance, reducing production costs and extending service life. Establishing a proton exchange membrane fuel cell model is an essential step to analyze its output characteristics and improve work efficiency. Fuel cell models are divided into mechanism models and empirical models [5]. The empirical model is simpler than the mechanism model, can explain the performance of proton exchange membrane fuel cells to a certain extent, and is widely used in commercial fuel cell pack performance simulation [6].

The main empirical models include Srinivasan model [7], Amphlett model [8] and Pukrushpan model [9]. This article Study, introduces the structure and working principle of proton exchange membrane fuel cell output voltage based on Pukrushpan empirical model, it establishes the open circuit voltage model, activation voltage loss model, ohmic voltage loss model, concentration voltage loss model, and fuel cell terminal voltage dynamic model of the proton exchange membrane fuel cell. Using the proton exchange membrane fuel cell operating temperature and pressure as the input and the voltage value as the output, simulation and analysis are conducted to study the effects of operating temperature and pressure on the voltage of the proton exchange membrane fuel cell to improve his performance.

1.1. Structure of the Proton Exchange Membrane Fuel Cell

The proton exchange membrane fuel cell is composed of several fuel cells. Each fuel cell consists of a bipolar plate (BP), a gas diffusion layer (GDL), a catalytic layer (CL) and a proton exchange membrane (PEM) [10]. The structure and working principle of the fuel cell are shown in Figure 1.

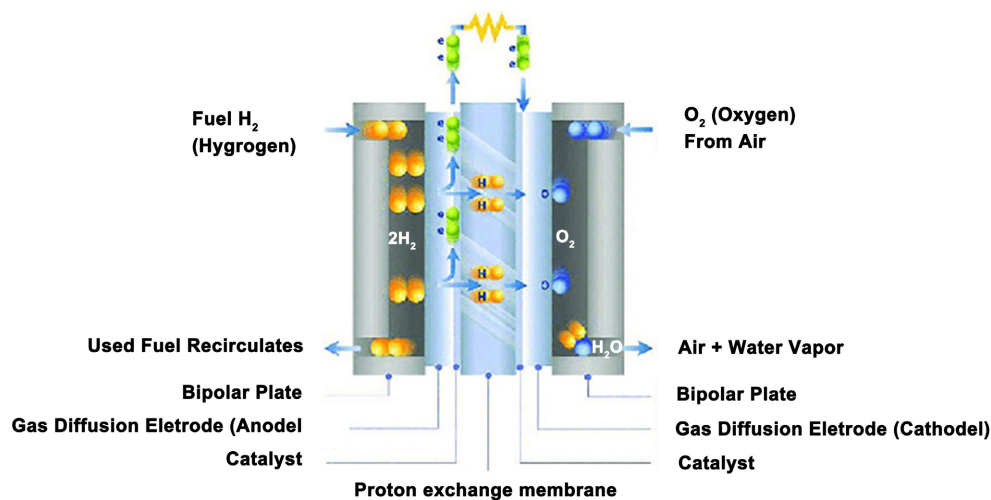


Figure 1. Fuel cell structure and working principle diagram.

The proton exchange membrane is the heart of the proton exchange membrane fuel cell, and the membrane must have relatively high proton conductivity. The catalyst layer may be partially located in the gas diffusion layer, partially located in the proton exchange membrane, or entirely located in the proton exchange membrane, depending on the manufacturing process. The gas diffusion layer is located between the catalytic layer and the bipolar plate. The fuel cell bipolar plate electrically connects fuel cells in series, has the function of heat conduction and heat dissipation, and provides a flow field channel for gas transmission. The hydrogen is stored in a hydrogen cylinder, is decompressed by a pressure reducer, and then enters the anode flow channel via a pipeline. Air is drawn from the environment by the air compressor, passes through the filters, intercoolers, humidifiers and other components, and then enters the cathode flow channel of the fuel cell.

1.2. Working Principle of Proton Exchange Membrane Fuel Cell

After hydrogen and oxygen reach the anode flow channel and cathode flow channel, respectively, they pass through the gas diffusion layer and reach the catalytic layer. Under the action of the anode catalytic layer catalyst, hydrogen breaks the chemical bond between hydrogen protons and electrons and decomposes into hydrogen ions and electrons. The sulfonic acid group in the proton exchange membrane is hydrophilic. Hydrogen ions combine with water molecules to form hydronium ions H^+ (xH_2O) and transfer from one sulfonic acid group to another in the proton exchange membrane, finally reaching Cathode, realizing the transfer from the anode side to the cathode side. This transfer causes the accumulation of negatively charged electrons in the anode, forming the negative electrode of the fuel cell. At the same time, the electrons reach the cathode through the external circuit [11].

Hydrogen undergoes an oxidation reaction at the anode, and the reaction formula is (1):



Oxygen undergoes a reduction reaction under the action of the cathode catalyst. Oxygen reacts with hydrogen ions and electrons to generate water (2). The reaction formula is:



The electrode reaction generates water, and most of the generated water is discharged from the tail gas. Some of the generated water diffuses through the membrane to the anode under the action of the pressure difference, and some of it can be used to humidify the gas.

The overall reaction equation of the fuel cell is (3):



2. Proton Exchange Membrane Fuel Cell Voltage Modeling

The output voltage of the proton exchange membrane fuel cell is related to the current density, reaction gas pressure, fuel cell temperature, proton exchange membrane humidity, etc. We first establish the fuel cell open circuit voltage model, activation voltage loss model, ohmic voltage loss model, concentration voltage loss model and terminal voltage dynamic output model as the basis for simulation and analysis.

2.1. Open Circuit Voltage Model

The theoretical open circuit voltage of proton exchange membrane fuel cell is a function of pressure and temperature. The calculation method is shown in equation (4), which is the Nernst electromotive force:

$$E = 1.229 - 0.85 \times 10^{-3} (T_{fc} - 298.15) + 4.3085 \times 10^{-5} \times T_{fc} \times \left[\ln(p_{H_2}) + \frac{1}{2 \ln(p_{O_2})} \right] \quad (4)$$

In the formula: T_{fc} is the fuel cell temperature, the unit is K; p_{H_2} and p_{O_2} are the pressures of hydrogen and oxygen, the unit is 1.013×10^5 Pa.

All single cells of a proton exchange membrane fuel cell are connected in series, so the current of a single fuel cell is equal to the total current of the fuel cell. The current density i is defined as the ratio of the fuel cell single cell current I_{st} (A) to the effective area A_{fc} (cm^2) (5):

$$i = \frac{I_{st}}{A_{fc}} \quad (5)$$

Figure 2 is fuel cell output voltage variation curve with current density obtained through modeling and simulation, that is, the typical polarization curve of fuel cell. The ordinate is the fuel cell output voltage V_{fc} , in volts (V), and the abscissa is the current density i , in $\text{A}\cdot\text{cm}^2$. It can be seen from the simulation results that as the current density increases, the output voltage of the fuel cell gradually

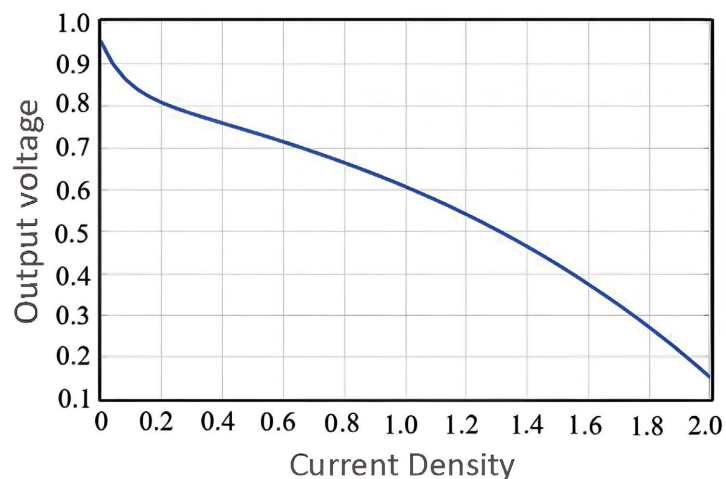


Figure 2. Typical polarization curve of fuel cell.

decreases. This is because the fuel cell has activation voltage loss, ohmic voltage loss and concentration voltage loss [12].

2.2. Activation Voltage Loss Model

A functional formula applicable to i is adopted to express the activation voltage loss (6):

$$V_{act} = V_0 + V_a \times (1 - e^{-c_1 i}) \quad (6)$$

In the formula, V_0 is the voltage when the current density is 0, and c_1 is a constant.

The values of V_0 and V_a are functions of oxygen partial pressure and temperature, as shown in Equations (7)-(9):

$$V_0 = 0.279 - 8.5 \times 10^{-4} (T_{fc} - 298.15) + 4.308 \times 10^{-5} T_{fc} \left[\ln \left(\frac{p_{ca} - p_{sat}}{1.01325} \right) + 0.5 \ln \left(\frac{0.1173 \times (p_{ca} - p_{sat})}{1.01325} \right) \right] \quad (7)$$

$$V_a = (1.8 \times 10^{-4} T_{fc} - 0.166) \left(\frac{p_{O_2}}{0.1173} + p_{sat} \right) + (-5.8 \times 10^{-4} T_{fc} - 0.5736) + (-1.618 \times 10^{-5} T_{fc} + 1.618 \times 10^{-2}) \left(\frac{p_{O_2}}{0.1173} + p_{sat} \right)^2 \quad (8)$$

$$c_1 = 10 \quad (9)$$

Figure 3 shows the activation voltage loss simulation curve. From the simulation results, it can be seen that the activation voltage loss increases rapidly when the current density is less than 0.4 A·cm⁻², and almost no longer increases at higher current densities.

2.3. Ohmic Voltage Loss Model

Ohmic voltage loss follows Ohm's law (10), where voltage equals ohmic resistance times current density:

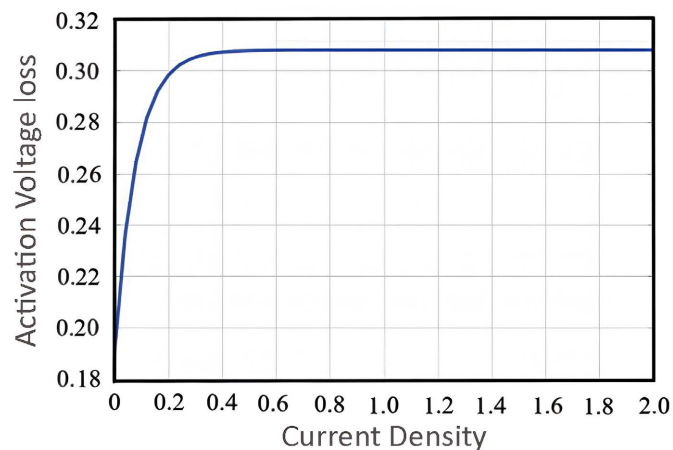


Figure 3. Activation voltage loss curve.

$$V_{ohm} = i \times R_{ohm} \quad (10)$$

In the formula, R_{ohm} is the ohmic resistance, the unit is $\Omega \cdot \text{cm}^2$. The ohmic resistance is mainly affected by the humidity of the proton exchange membrane and the fuel cell temperature (11). A large number of studies have shown that the ohmic resistance is a function of the exchange membrane conductivity σ_m :

$$R_{ohm} = t_m / \sigma_m \quad (11)$$

In the formula: t_m is the thickness of the proton exchange membrane; σ_m is the conductivity of the proton exchange membrane. The relationship between the humidity of the proton exchange membrane and the temperature (12)-(13) is as follows:

$$\sigma_m = b_1 \exp\left(b_2 \left(\frac{1}{303} - \frac{1}{T_{fc}}\right)\right) \quad (12)$$

In the formula: b_1 is a function of the proton exchange membrane water content constant λ_m :

$$b_1 = 0.005139\lambda_m - 0.00326 \quad (13)$$

The value of the proton exchange membrane water content constant λ_m varies between 0 and 14 with relative humidity. When the relative humidity is 100%, $\lambda_m = 14$ and the value of b_1 is 0.068686; b_2 is a constant here 350.

Figure 4 is the simulation curve of ohmic voltage loss. From the simulation curve, it can be seen that the value of ohmic voltage loss is small when the current density is low, and it increases linearly with the increase of current density.

2.4. Concentration Voltage Loss Model

The calculation of concentration voltage loss uses a formula (14)-(16):

$$V_{conc} = i \left(C_2 \left(\frac{i}{i_{max}} \right) \right)^{c_3} \quad (14)$$

In the formula, c_3 and i_{max} are both empirical parameters, and C_2 is the function of the fuel cell temperature and reaction gas pressure:

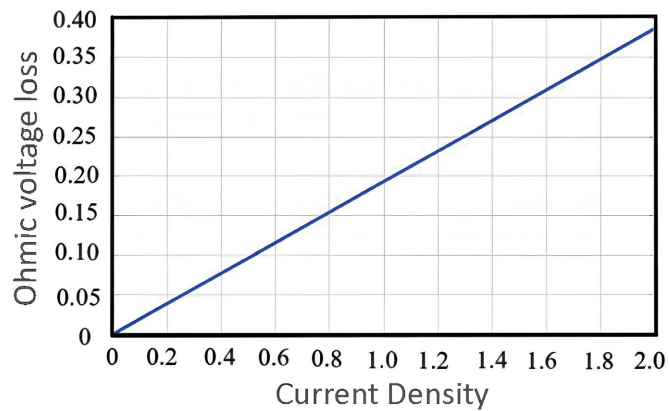


Figure 4. The curve of ohmic voltage loss.

$$\text{When } \left(\frac{P_{O_2}}{0.1173} + p_{sat} \right) < 2.026 \times 10^5 P_a$$

$$c_2 = (7.16 \times 10^{-4} T_{fc} - 0.622) \left(\frac{P_{O_2}}{0.1173} + p_{sat} \right) + (-1.45 \times 10^{-3} T_{fc} + 1.68) \quad (15)$$

$$\text{When } \left(\frac{P_{O_2}}{0.1173} + p_{sat} \right) \geq 2.026 \times 10^5 P_a$$

$$c_2 = (8.86 \times 10^{-5} T_{fc} - 0.068) \left(\frac{P_{O_2}}{0.1173} + p_{sat} \right) + (-1.6 \times 10^{-4} T_{fc} + 0.54) \quad (16)$$

The parameter i_{max} is the current density that causes a sudden voltage drop. In practical engineering applications, in order to maintain the safe operation of the fuel cell, the maximum output current is usually defined as 85% of the current when the sudden drop occurs.

Figure 5 shows the concentration voltage loss simulation curve. From the simulation curve, it can be seen that the concentration voltage loss value is small when the current density is low, and increases exponentially as the current density increases, especially when the current density is high.

2.5. Fuel Cell Terminal Voltage Model

From the Nernst electromotive force, activation voltage loss, ohmic voltage loss, and concentration voltage loss (17), the fuel cell output voltage can be obtained as [13]:

$$\begin{aligned} V_{fc} &= E - V_{act} - V_{ohm} - V_{conc} \\ &= E - [V_0 - V_0(1 - e^{-c_1 i})] - [i * R_{ohm}] - i \left(C_2 \frac{i}{i_{max}} \right)^{c_3} \end{aligned} \quad (17)$$

In the formula, E is the Nernst electromotive force, and V_{fc} is the output terminal voltage of a single fuel cell. A fuel cell is composed of multiple single cells, so the fuel cell stack voltage is the product of the single cell voltage to the number of cells (18):

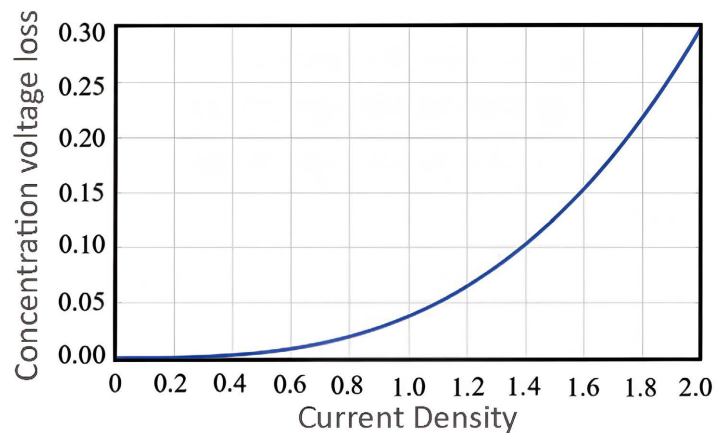


Figure 5. The curve of concentration voltage loss.

$$V_{st} = n * V_{fc} \tag{18}$$

The parameters of the fuel cell terminal voltage can be obtained using nonlinear regression equations based on the polarization data of the fuel cell. The nonlinear curve fitting function in the optimization toolbox in Matlab can be used to solve nonlinear curve fitting problems [14]. **Figure 6** shows the fuel cell terminal voltage model.

2.6. Dynamic Characteristics of Fuel Cells

The fuel cell has a fast dynamic property, called a “charge double layer” phenomenon, where the layer behaves like a capacitor. This layer generates a voltage by collecting the charge between the two poles. When the current changes suddenly, the activity voltage drop and concentration voltage drop will not change immediately with the change of current, but will lag behind the change of current; while the ohmic voltage drop will change with the change of current [15] [16]. **Figure 7** is the equivalent circuit diagram of the fuel cell dynamic model.

In **Figure 7**, R_{act} is the active resistance (19) and R_{conc} is the tolerance resistance (20).

$$R_{act} = \frac{1}{i} \left[V_0 + V_a (1 - e^{-c_1 i}) \right] \tag{19}$$

$$R_{conc} = \left(C_2 \frac{i}{i_{max}} \right)^{c_3} \tag{20}$$

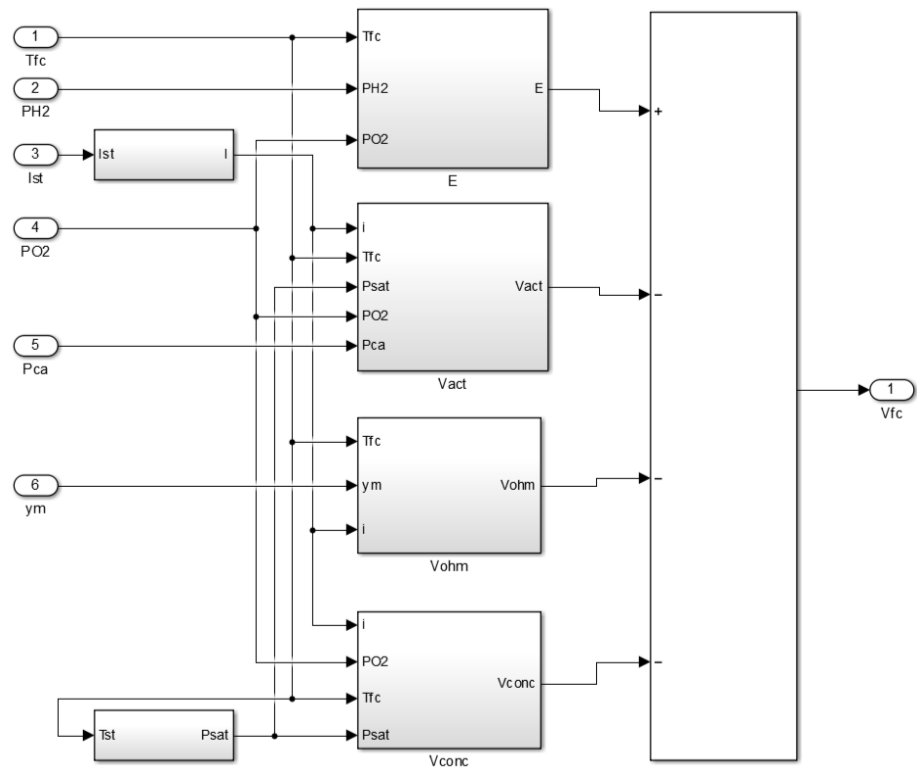


Figure 6. Fuel cell voltage model.

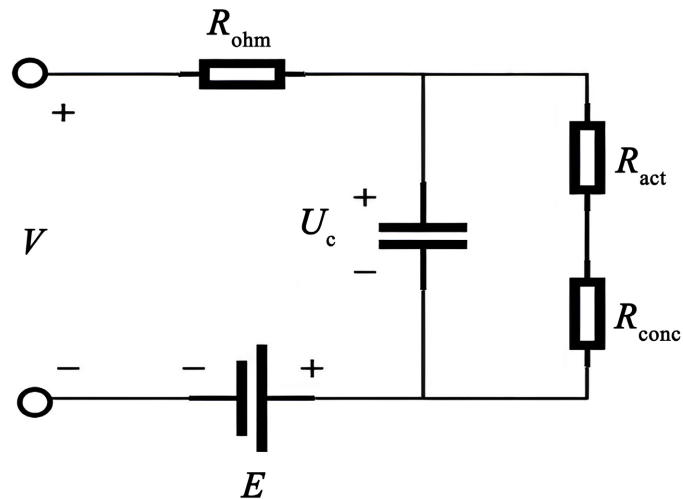


Figure 7. Equivalent circuit model of proton exchange membrane fuel cell.

Fuel cell dynamic voltage characteristics (21)-(22):

$$C \frac{dV_c}{dt} + \frac{V_c - V_0}{R_{act} + R_{conc}} = i \quad (21)$$

$$V_{fc} = E - V_c - iR_{ohm} \quad (22)$$

Based on the above basic modeling theory, a proton exchange membrane fuel cell voltage model is established.

3. Methodology and Results of Research

3.1. Methodology

Using the model established in Section 2, with the fuel cell operating pressure and cell temperature as input variables, the fuel cell polarization voltage and three voltage losses are simulated, and the effects of pressure and temperature changes on the cell voltage are analyzed.

For analyzing the influence of pressure on the fuel cell voltage, the operating temperature is kept constant at 80°C and the pressure is varied from 1×10^5 Pa to 3×10^5 Pa (1×10^5 Pa, 1.5×10^5 Pa, 2×10^5 Pa and 3×10^5 Pa respectively). Afterwards, the influence of pressure on activation voltage losses, ohmic voltage loss and concentration voltage loss are analyzed. For analyzing the influence of temperature on the fuel cell voltage, the operating pressure is kept constant at 2.5×10^5 Pa and the temperature is varied from 0°C to 100°C respectively with an interval of 20°C. Afterwards, the influence of temperature on activation voltage losses, ohmic voltage loss and concentration voltage loss are analyzed.

3.2. Results

3.2.1. Analysis of the Influence of Pressure on Voltage

Figure 8 is the polarization curve of the fuel cell when the cathode working pressure changes from 1×10^5 Pa to 3×10^5 Pa under the condition that the fuel cell operating temperature is 80°C. The working pressures of the cathode from

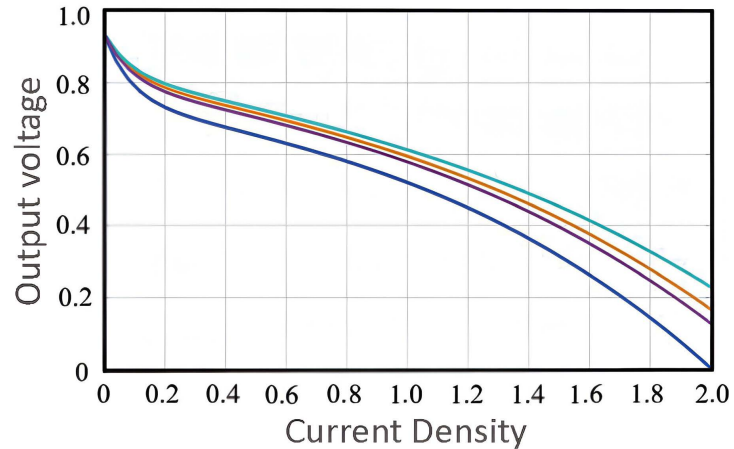


Figure 8. Fuel cell polarization curve from 1×10^5 Pa to 3×10^5 Pa at 80°C .

bottom to top in this curve are 1×10^5 Pa, 1.5×10^5 Pa, 2×10^5 Pa, and 3×10^5 Pa respectively. From the simulation curve results, it can be seen that as the working pressure increases, the fuel cell voltage increases. When the working pressure increases from 1×10^5 Pa to 1.5×10^5 Pa, the voltage increases about 3 times compared to when the working pressure increases from 1.5×10^5 Pa to 2×10^5 Pa. It shows that increasing the working pressure when the pressure is low has a significant effect on increasing the output voltage.

Figure 9 is the activation voltage loss simulation curve when the cathode working pressure changes from 1×10^5 Pa to 3×10^5 Pa under the condition that the fuel cell operating temperature is 80°C . The cathode working pressures from top to bottom of the curve are 1×10^5 Pa, 1.5×10^5 Pa, 2×10^5 Pa, and 3×10^5 Pa respectively. The activation voltage loss is maximum when the current density is greater than $0.4 \text{ A}\cdot\text{cm}^{-2}$. The values are 0.390 V, 0.365 V, 0.353 V and 0.338 V respectively. It can be seen from the simulation results that when the operating pressure of the fuel cell increases, the activation voltage loss decreases. This is because the increase in pressure is conducive to the progress of the reaction and reduces the activation energy consumed by the reaction. When the working pressure increases from 1×10^5 Pa to 1.5×10^5 Pa, the reduction of activation voltage loss is approximately twice that of when the working pressure increases from 1.5×10^5 Pa to 2×10^5 Pa. When the working pressure increases from 2×10^5 Pa to 3×10^5 Pa, the activation voltage loss reduction value is 1.25 times that of when the working pressure increases from 1.5×10^5 Pa to 2×10^5 Pa. It shows that when the pressure is small, increase the pressure has a significant effect on reducing the activation voltage loss and the effect is weakened when the pressure is high.

Figure 10 is the ohmic voltage loss curve when the cathode working pressure changes from 1×10^5 Pa to 3×10^5 Pa under the condition that the fuel cell operating temperature is 80°C . Ohmic voltage loss is the product of the fuel cell current density and the total internal resistance of the fuel cell. The total internal resistance includes ionic resistance, electronic resistance and contact resistance,

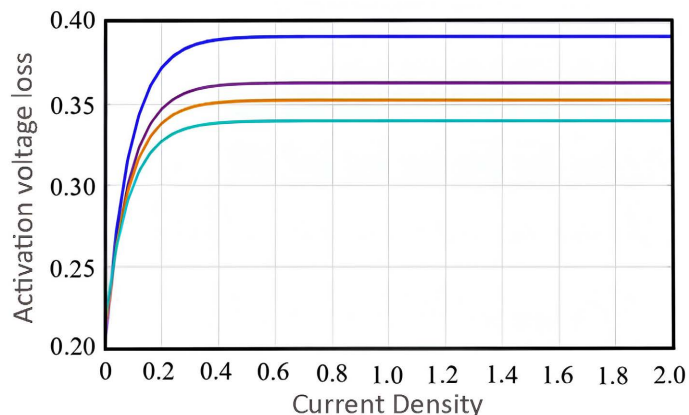


Figure 9. The curve of Activation voltage loss from 1×10^5 Pa to 3×10^5 Pa at 80°C .

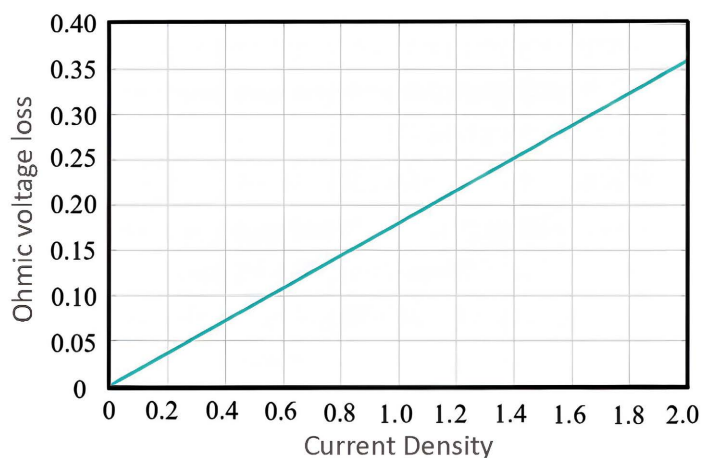


Figure 10. The curve of ohmic voltage loss from 1×10^5 Pa to 3×10^5 Pa at 80°C .

which electronic resistance is negligible, Ion resistance and contact resistance have nothing to do with the operating pressure of the fuel cell. Therefore, changes the working pressure has no impact on the ohmic voltage loss of the fuel cell.

Figure 11 shows the concentration voltage loss curve when the cathode working pressure changes from 1×10^5 Pa to 3×10^5 Pa when the fuel cell operating temperature is 80°C . The cathode working pressures from top to bottom of the curve are 1×10^5 Pa, 1.5×10^5 Pa, 2×10^5 Pa, and 3×10^5 Pa respectively. It can be seen from the simulation results that as the working pressure increases, the concentration voltage loss decreases rapidly. This is because under higher pressure conditions, on the one hand, sufficient reactant concentration is provided, and at the same time, the reactant partial pressure is increased, and the reactant diffusion rate is increased.

3.2.2. Analysis of the Influence of Temperature on Voltage

Figure 12 is the polarization curve of the fuel cell when the operating temperature changes from 0°C to 100°C under the condition that the cathode working pressure of the fuel cell is 2.5×10^5 Pa. The polarization curve in the figure shows

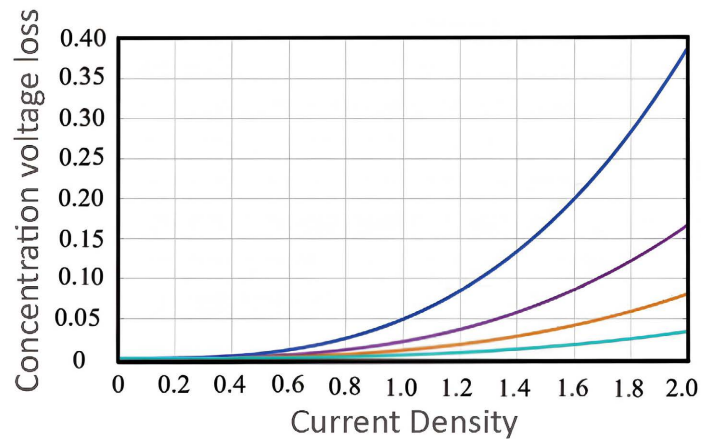


Figure 11. The curve of concentration voltage loss from 1×10^5 Pa to 3×10^5 Pa at 80°C .

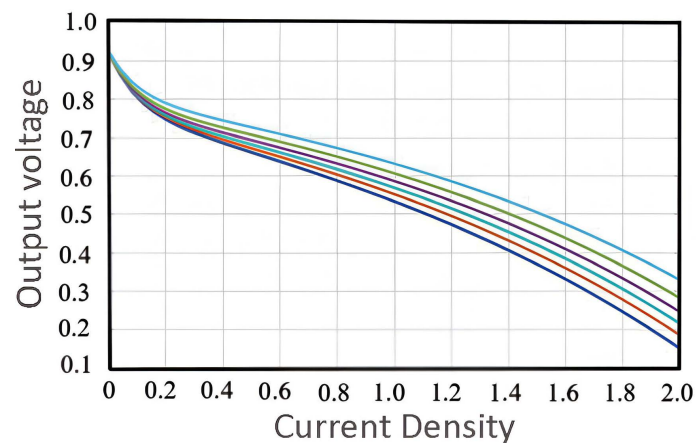


Figure 12. Polarization curve from 0°C to 100°C under 2.5×10^5 Pa.

that the operating temperature from bottom to top is 0°C to 100°C respectively, the interval is 20°C .

It can be seen from the simulation results that as the operating temperature increases under the condition of constant pressure, the output voltage of the fuel cell increases. This is because increasing the operating temperature of the fuel cell within a certain range under the condition of constant pressure can increase the Nertz electromotive force, reduce the activation voltage loss, ohmic voltage loss and concentration voltage loss, and increase the fuel cell output voltage. **Figure 13** is the activation voltage loss curve of the fuel cell when the operating temperature changes from 0°C to 100°C , when the cathode working pressure of the fuel cell is 2.5×10^5 Pa.

At the curve in **Figure 13**, the operating temperatures from bottom to top are 0°C to 100°C , with an interval of 20°C . It can be seen from the simulation results that as the operating temperature increases under the condition of constant pressure, the activation voltage loss of the fuel cell decreases. Because the increase in temperature increases the electrochemical reaction activity, it is conducive to the transfer of anode protons through the proton exchange membrane

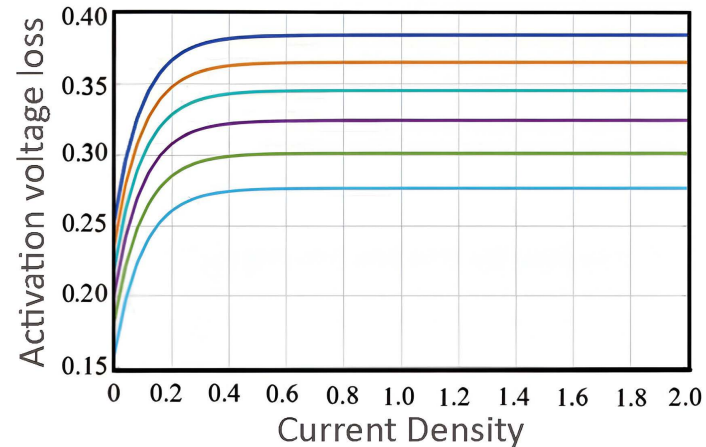


Figure 13. The curve of activation voltage loss from 0°C to 100°C under 2.5×10^5 Pa.

and electrons through the plate and other components to the cathode, reducing the activation over potential, promoting the electrochemical reaction, and reducing the reaction activation energy. So the activation voltage loss is reduced.

When the current density is 0, the initial value of activation voltage loss decreases with increasing temperature. When the temperature is 0°C, the initial value of the activation voltage loss is 0.26 V, and when the temperature is 100°C, the initial value of the activation voltage loss is reduced to 0.16 V. When the current density is greater than $0.4 \text{ A}\cdot\text{cm}^2$, for every 20°C increase in temperature, the maximum activation voltage loss decreases by approximately 0.022 V.

Figure 14 is the ohmic voltage loss curve of the fuel cell when the operating temperature changes from 0°C to 100°C under the condition that the fuel cell operating pressure is 2.5×10^5 Pa. The working temperatures of the curves in the figure are from 0°C to 100°C from bottom to top, with an interval of 20°C. It can be seen from the simulation curve that under the condition of constant pressure, the ohmic voltage loss of the fuel cell decreases as the temperature increases, and the slope of the ohmic voltage simulation curve is 0.212, 0.193, 0.180, 0.169, 0.158, 0.150. Because proton conductivity has a direct impact on the resistance, and proton conductivity is a function of temperature. When the temperature increases, the proton conductivity increases. Under the condition that the thickness of the proton exchange membrane is certain, the ohmic resistance of the fuel cell decreases, which reduces the ohmic voltage loss.

Figure 15 is the activation voltage loss curve of the fuel cell when the operating temperature changes from 0°C to 100°C under the condition that the fuel cell operating pressure is 2.5×10^5 Pa. The curves range from 0°C to 100°C from bottom to top, with an interval of 20°C. It can be seen from the simulation curve that the voltage loss of the fuel cell concentration decreases as the temperature increases when the pressure remains unchanged, because the uniformity of reactant distribution increases after the temperature increases, but the effect is not obvious. This is because the concentration difference is extremely small. Chemicalization is mainly related to concentration and pressure.

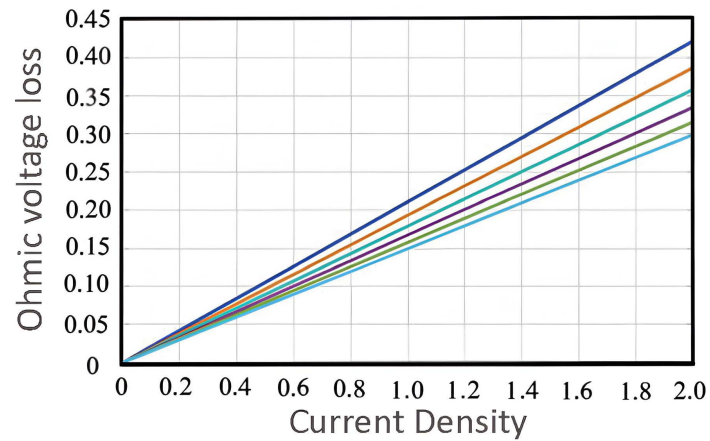


Figure 14. The curve of ohmic voltage loss from 0 °C to 100 °C under 2.5×10^5 Pa.

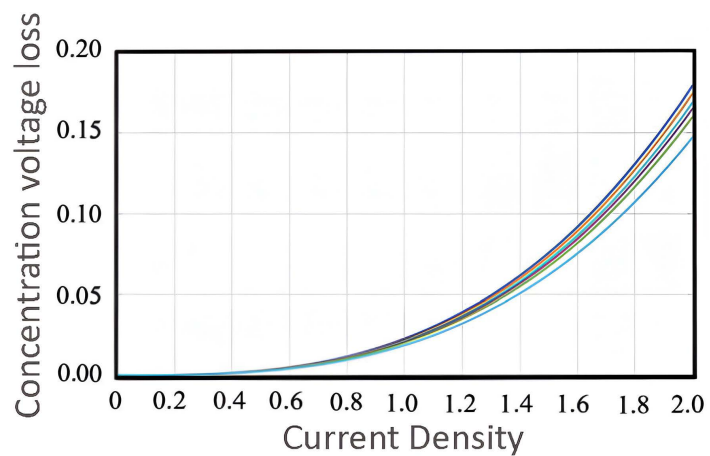


Figure 15. The curve of concentration voltage loss from 0 °C to 100 °C under 2.5×10^5 Pa.

4. Conclusions

It can be seen from the simulation results that increasing the operating pressure and temperature of the fuel cell within a certain range can reduce the voltage loss, increase the polarization voltage of the proton exchange membrane fuel cell, and improve the output performance.

Increased operating pressure can reduce the activation voltage loss of proton exchange membrane fuel cells. When the working pressure is 1×10^5 Pa, 1.5×10^5 Pa, 2×10^5 Pa, 2×10^5 Pa, and 3×10^5 Pa, the maximum activation voltage loss is 0.390 V, 0.365 V, 0.353 V, 0.338 V respectively, and the maximum drop rate (V/Pa) is 12.8%, 6.6%, 4.2% respectively. When the pressure is low, increasing the working pressure activates the voltage loss reduction effect more obviously; Due to the characteristics of ohmic voltage loss, pressure changes have no effect on its values; an increase in pressure has a significant effect on reducing the concentration voltage loss of the proton exchange membrane fuel cell.

Temperature changes have an impact on the three voltage losses of proton exchange membrane fuel cells. However, increasing temperature has a relatively poor effect on reducing concentration voltage loss. Increasing the temperature

within the allowable operating range of the fuel cell can reduce the activation voltage loss and ohmic voltage loss of the fuel cell to a certain extent, and improve the output performance of the fuel cell. In the range of 0°C to 100°C, for every 20°C increase in temperature, the maximum activation voltage loss decreases by approximately 0.022 V; For every 20°C increase in temperature, the slopes of the ohmic voltage simulation curves are 0.212, 0.193, 0.180, 0.169, 0.158, and 0.150 respectively.

According to the fuel cell polarization curve and three voltage loss simulation curves, the activation voltage loss is the main voltage loss in the low current density zone of 0 to 0.4 A·cm⁻². Ohmic voltage loss is the main voltage loss in the current density zone of 0.4 - 1.2 A·cm⁻². The concentration voltage loss is the main voltage loss in the high current density zone of 1.2 to 2.0 A·cm⁻². Therefore, in the medium and low current density zone, the solution of increasing the temperature can be adopted to reduce the voltage loss of the fuel cell. In the high current density zone, the solution of increasing the working pressure can be adopted to reduce the concentration voltage loss and improve the fuel cell output performance.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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