Research Article

Reaction Gas Pressure, Temperature, and Membrane Water Content Modulate Electrochemical Process of a PEMFC: A Simulation Study

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Proton exchange membrane fuel cells (PEMFC) are widely used in transportation systems owing to their desirable characteristics such as high efficacy and low operating temperature. However, the fuel cell systems exhibit load changes as well as voltage and power losses so as to reduce dependence on the battery. The aim of the present study was to explore the composition and basic working principle of PEMFC. A PEMFC electrochemical reaction model was then established according to the electrochemical reaction principle of fuel cell to evaluate the effects of Nernst electromotive force, activation overvoltage, Ohmic overvoltage, concentration overvoltage, and electric double layer. The effects of activation loss, concentration loss, and Ohmic loss on the fuel cell were evaluated through simulation analysis. The effect of various factors on the dynamic output of a 60 kW PEMFC was explored through dynamic simulations. The findings showed that a change in current modulated a change in voltage through the Ohmic loss equivalent resistance. The activation loss equivalent resistance and the concentration loss equivalent resistance decreased the voltage loss owing to the presence of the capacitor. The output voltage of the fuel cell decreased with an increase in load current, whereas the output power increased with an increase in load current. Increase in partial pressure of oxygen caused an increase in output power and output voltage of the cell. The internal chemical reaction rate and the voltage output of the fuel cell increases with an increase in the working temperature. The findings of this study provide a basis for conducting further studies to produce efficient fuel cells for application in various systems.

1. Introduction

A fuel cell is an electrochemical reactor that directly converts chemical energy into electrical energy without burning it [1–3]. Fuel cells exist in different types, and each type has distinct temperature requirements. Low-temperature fuel cells typically work at temperatures below 200°C, and excessive temperatures damage the electrolyte in these types of fuel cells. Low-temperature fuel cells include alkaline fuel cell (AFC), phosphoric acid fuel cell (PAFC), and proton exchange membrane fuel cell (PEMFC). Low-temperature fuel cells use platinum (Pt) as catalyst. Platinum is very sensitive to carbon monoxide; therefore, this type of fuel cell uses pure hydrogen as fuel. High temperature fuel cells can

directly oxidize other hydrocarbon fuels (such as methane). Fuel cells that directly oxidize hydrocarbon fuels include molten carbonate fuel cells (MCFC) and solid oxide fuel cells (600–1000°C). The operation temperature range of these fuel cells is 600–700°C.

The PEM fuel cell is a fuel cell type that works under a temperature range of 50–80°C. This fuel cell consists a polymer electrolyte membrane placed between two gas diffusion layers and two electrodes. The layers facilitate hydrogen and oxygen reactions at the anode and cathode, respectively. The PEM fuel cell type is characterized by high working efficiency (40%–50%), fast startup, relatively good power density, and high reliability. PEM fuel cell is widely used in various fields. Therefore, the current study sought to
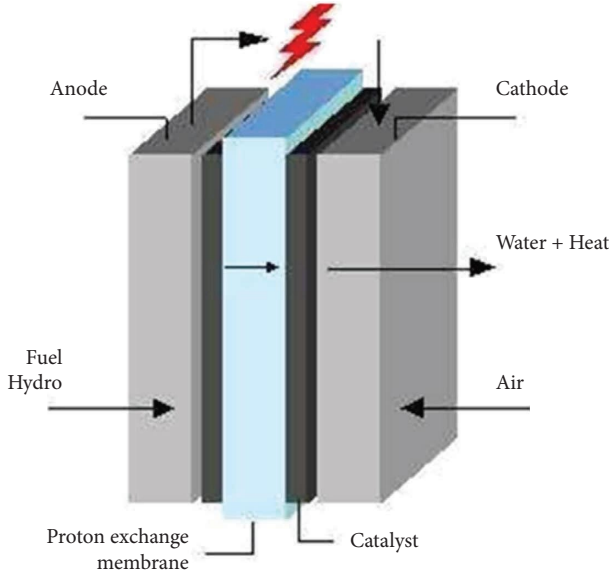
Figure 1: A representation of the working principle of PEMFC.

explore the potential application of PEMFC as a vehicle energy source.

The working principle of the hydrogen-oxygen fuel cell is the reverse process of electrolysis of water. The electrochemical reaction between hydrogen at the anode and air at the cathode leads to conversion of chemical energy into electrical energy that can be used in various equipment. A continuous supply of hydrogen and air must be supplied to the anode and cathode of the proton exchange membrane fuel cell, respectively, for generation of electricity. Hydrogen and air penetrate the flow field structure of the bipolar plate and diffuse to the anode and cathode, respectively, through the diffusion layer (Figure 1). Hydrogen at the anode releases an electron to become a proton under the action of the catalyst. An electric current is generated through flow of electrons along the external circuit, which drives the load and is transferred to the cathode. The electron combines with oxygen to become oxygen ions in the presence of a catalyst. The protons are transferred to the cathode through the selective passage across the proton exchange membrane. The protons combine with cations at the cathode to generate water molecules [4, 5]. The water generated at the cathode is released through the flow channel under the purging action of the cathode air. The electrochemical reaction in the fuel cell is characterized by generation of heat. Therefore, it is imperative to include cooling channels on the bipolar plate to regulate the temperature of the fuel cell.

2. PEMFC Electrochemical Model

The aim of this study was to explore the overall output performance and factors that affect operation of PEMFC. In the current study, the effect of the spatial location difference of variables in the internal structure of PEMFC was not evaluated. The lumped parameter modeling method was adopted in this study to study performance of the PEMFC system. The fuel cell stack is referred as a DC power supply if

the fuel cell air, hydrogen, and cooling water output are stable. The fuel cell stack voltage is an essential performance indicator of the fuel cell. Each single cell is assumed to be consistent when establishing the lumped parameter PEMFC model. In addition, the external output voltage of the fuel cell is defined as the sum of the levels of voltage generated by all the cells.

Therefore, the fuel cell voltage V_{st} is expressed as shown in the following formula:

$$V_{st} = n_{cell} V_{fc}, \tag{1}$$

where n_{cell} represents the number of single cells and V_{fc} represents the voltage of single cells.

The current generated by the fuel cell is directly proportional to the level of fuel consumed. This implies that n moles of electrons are provided for each mole of fuel. As a result, a decrease in the fuel cell voltage is associated with a decrease in the electrical power produced per unit of fuel. The fuel cell output voltage can thus be used to determine the efficiency of the fuel cell. Notably, maintaining a high voltage under current load is challenging, and the output voltage of actual fuel cells is lower than the voltage output predicted under thermodynamic theory. Previous studies report three primary types of loss of output voltage of fuel cells, namely activation loss, Ohmic loss, and concentration loss. The definite output voltage of the fuel cell is obtained by the difference between the thermodynamically predicted voltage output and the voltage drop due to the three types losses, as shown below.

$$V_{fc} = E - v_{act} - v_{ohm} - v_{conc}, \tag{2}$$

where E denotes the thermodynamically predicted open circuit voltage, also referred as the Nernst voltage; v_{act} represents the activation overvoltage; v_{ohm} indicates the Ohmic overvoltage; v_{conc} represents the concentration overvoltage.

The reversible voltage E^0 of the hydrogen-oxygen fuel cell under the standard state and the Gibbs free energy ΔG^0 released by the chemical reaction of hydrogen oxidation are governed by the following relationship:

$$\Delta G^0 = -nFE^0, \tag{3}$$

where the number of electrons transferred by the hydrogen reaction is 2 mol, thus n is 2. ΔG^0 is −273.2 kJ in thermodynamics. Gibbs free energy represents the maximum amount of work performed by a thermodynamic system at a constant pressure and temperature.

The reversible voltage is affected by temperature changes, and the effect by the change in temperature is expressed as shown in the following formula:

$$E^T = E^0 + \frac{\Delta S}{nF} (T_{st} - 298.15), \tag{4}$$

where ΔS represents the thermodynamic entropy change of the chemical reaction of hydrogen oxidation, which is equal to 164.025 J/mol•K, and T_{st} indicates the operating temperature (K) of the fuel cell stack.

Change in reversible voltage with the concentration of reactants is expressed as shown in the following formula:

$$E^C = E^0 - \frac{RT_{st}}{2F} \ln \frac{1}{p_{an}^{H_2} p_{ca}^{O_2 1/2}}, \tag{5}$$

where $p_{an}^{H_2}$ represents the partial pressure of hydrogen and $p_{ca}^{O_2}$ represents the partial pressure of oxygen.

The reversible voltage at different temperatures and reactant concentrations can be determined by combining formulas (3)–(5) to obtain the Nernst voltage of the electrochemical reaction of hydrogen and oxygen, as presented in the following formula:

$$E = -\frac{\Delta G^0}{2F} + \frac{\Delta S}{2F}(T_{st} - 298.15) + \frac{RT_{st}}{2F}\left[\ln\left(p_{an}^{H_2}\right) + \frac{1}{2}\ln\left(p_{ca}^{O_2}\right)\right]. \tag{6}$$

Activation polarization is irreversible and is the main cause of decrease in voltage. Electrons should break and form covalent bonds during transfer from the anode to the cathode. Therefore, the hydrogen oxidation reaction at the anode occurs very fast, whereas the oxygen reduction reaction at the cathode is relatively slow, implying that the cathode plays a significant role in the activation overvoltage. Some energy is lost during transfer of electrons from or to the electrode, thus the activation overvoltage is represented as the potential deviation from the original equilibrium position when the electrode surface is about to initiate the electrochemical reaction. The Tafel equation was used to establish an empirical model for the activation overvoltage as shown in the following formula:

$$v_{act} = v_0 + v_a\left(1 - e^{-\xi_1 i}\right), \tag{7}$$

where i represents the current density, which is the current generated per unit of the effective active area of the proton exchange membrane and was obtained using formula (8) below; the voltage drop at zero current density of v_0 was determined using formula (9),ξ_1 represent the empirical parameter, and v_a was determined using equation (10).

$$i = \frac{I_{st}}{A}, \tag{8}$$

$$v_0 = 0.279 - \frac{\Delta S}{2F}(T_{st} - 298.15) + \frac{RT_{st}}{2F}\left[\ln\left(\frac{P_{ca} - P_{sat}}{P_{atm}}\right)\right.$$
$$\left. + \frac{1}{2}\ln\left(\frac{0.1173(P_{ca} - P_{sat})}{P_{atm}}\right)\right], \tag{9}$$

$$v_a = \left(-1.618 \times 10^{-5}T_{st} + 1.618 \times 10^{-2}\right)\left(\frac{P_{O_2}}{0.1173} + P_{sat}\right)^2$$
$$+ \left(1.8 \times 10^{-4}T_{st} - 0.166\right)\left(\frac{P_{O_2}}{0.1173} + P_{sat}\right)$$
$$+ \left(\frac{\Delta S}{2F}T_{st} + 0.5736\right), \tag{10}$$

P_{sat} represents the saturated vapor pressure of water, which is a function of temperature, and can be obtained as shown in the following equation:

$$\lg(P_{sat}) = 0.0295 \times (T - 273.15) - 9.18 \times 10^{-5}(T - 273.15)^3$$
$$+ 1.44 \times 10^{-7}(T - 273.15)^3 - 2.18 \tag{11}$$

Ohmic overvoltage is the voltage drop caused by the ionic resistance and electronic resistance inside the fuel cell. It comprises two parts: the voltage loss caused by resistance of the proton exchange membrane to hinder passage of protons and resistance of the electrode or collector plate to electron transfer. Ohm's Law is used to express Ohmic overvoltage as shown below:

$$v_{ohm} = R_{ohm}I_{st} = \left(R_e + R_p\right)I_{st}, \tag{12}$$

where R_{ohm} represents the fuel cell impedance (Ω), R_e denotes the electron flow impedance (Ω), and R_p represents the membrane impedance, which can be determined using formula (13).

$$R_p = \frac{\rho_M * t_m}{A}, \tag{13}$$

where ρ_M denotes the membrane resistivity, which is correlated with the temperature and humidity of the proton exchange membrane and is determined using formula (14) and t_m represents the thickness of the electrolyte membrane (cm).

$$\rho_M = \frac{181.6\left[1 + 0.03 * (I_{st}/A) + 0.062 * (T_{st}/303)^2 * (I_{st}/A)^{2.5}\right]}{\left[\lambda_m - 0.634 - 3 * (I_{st}/A)\right] * \exp\left[4.18 * (T_{st} - 303/T_{st})\right]}, \tag{14}$$

where λ_m denotes the water content of the membrane.

Concentration overvoltage is associated with changes in the concentration of the reactants consumed during the reaction. The rapid consumption rate of reactants causes a decrease in the partial pressure of reactants and decrease in the reaction rate at high current density, which results in voltage loss in the fuel cell. The concentration overvoltage is expressed as shown in the following equation:

$$v_{conc} = i\left(\xi_3\frac{i}{i_{max}}\right)^{\xi_2}, \tag{15}$$

where ξ_2 denotes the equation coefficient, which is correlated with the properties of the fuel cell and its working environment, i_{max} represents the maximum current density that the fuel cell can achieve under the working state, usually 2.2, ξ_3 denotes the operating temperature of the fuel cell stack. Coefficients related to the partial pressure of oxygen are expressed as shown below:
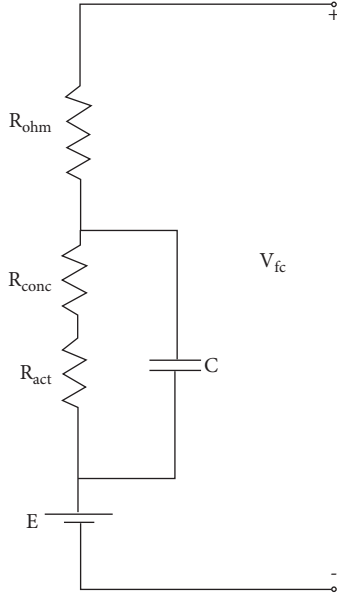
Figure 2: A PEMFC equivalent circuit.

$$\xi_3 = \begin{cases} \left(7.16 \times 10^{-4} T_{st} - 0.622\right)\left(\dfrac{P_{ca}^{O_2}}{0.1173 \times 10^5} + \dfrac{P_{sat}}{10^5}\right) \\[2em] + \left(-1.45 \times 10^{-3} T_{st} + 1.68\right)\dfrac{P_{ca}^{O_2}}{0.1173} + P_{sat} < 2P_{atm} \\[2em] \left(8.66 \times 10^{-5} T_{st} - 0.068\right)\left(\dfrac{P_{ca}^{O_2}}{0.1173 \times 10^5} + \dfrac{P_{sat}}{10^5}\right) \\[2em] + \left(-1.63 \times 10^{-3} T_{st} + 0.54\right)\dfrac{P_{ca}^{O_2}}{0.1173} + P_{sat} \geq 2P_{atm} \end{cases}$$

$$(16)$$

3. PEMFC Dynamic Characteristics

The fuel cell exhibits dynamic behavior when it releases power to the outside [6–9]. Contact between two dissimilar materials at the electrode and electrolyte interface in a fuel cell produces a charge layer that resembles a capacitor. This charge layer is referred as a "charge double layer" and can be used to store charge and energy [10]. Abrupt change in the current caused by the presence of the electric double-layer structure results in the activation overvoltage by the blocking effect of the capacitor. Subsequently, change in the concentration overvoltage is gradual and exhibits a hysteresis effect.

The activation overvoltage and concentration overvoltage in the equivalent circuit of the fuel cell are markedly affected by the electric double layer. An equivalent circuit diagram of the fuel cell is presented in Figure 2. The Ohmic loss equivalent resistance R_{ohm} rapidly causes a change in the voltage when the current changes. The activation loss equivalent resistance (R_{act}) and the concentration loss

equivalent resistance (R_{conc}) reduced the voltage drop of the resistance due to the presence of a capacitor. The charge double-layer effect can be expressed as shown below:

$$R_a i = R_{act} i + R_{conc} i = v_{act} + v_{conc},$$
$$C\frac{dV_c}{dt} = i - \frac{v_c}{v_{act} + v_{conc}},$$

$$(17)$$

where R_a represents the equivalent resistance, C represents the equivalent capacitance, which is a constant, and v_c denotes the dynamic overvoltage under the action of the electric double layer. The single cell output voltage of the fuel cell can be expressed as presented below:

$$v_{fc} = E - v_{ohm} - v_c.$$

$$(18)$$

4. Establishment and Simulation of PEMFC Model

MATLAB is a software widely used in the engineering field and is effective for application in nonlinear and linear dynamic simulations. Simulink is a toolbox in MATLAB software. System models built using the Simulink toolbox is characterized by simplicity, strong operability, and convenient maintenance. Simulink has been widely used in academic and industrial fields for simulation studies. In the present study, the PEMFC model was established using Simulink in MATLAB software as shown in Figure 3. The model parameters are presented in Table 1.

The input section of the model comprises fuel cell load current, anode hydrogen partial pressure, cathode oxygen partial pressure, cathode pressure, PEMFC stack temperature, and water content of the proton exchange membrane. The system output constitutes the output voltage and power of the fuel cell. The steady-state conditions used for the simulation are shown in Table 2.

5. Results and Discussion

The fuel cell polarization curve obtained after simulation is shown in Figure 4. The polarization curve exhibited typical characteristics of the fuel cell performance. The simulation results indicated that the polarization curve of the fuel cell can be divided into three regions, namely activation overvoltage region, Ohmic overvoltage region, and concentration overvoltage region. The three regions result from the different effects exerted by the three losses on the output voltage of the fuel cell under increase in the current density. The effects of the three losses on the Nernst electromotive force when the three losses act together and individually are presented in Figure 5. The activation overvoltage represents the region of low current density and indicates the energy limit that the chemical reaction should overcome. The results indicate that when the current density is very small, the activation overvoltage is the main source of voltage loss when the energy from the fuel cell is released to the outside (Figure 6). The fuel cell efficiency in this region is not high due to the lower current density. The
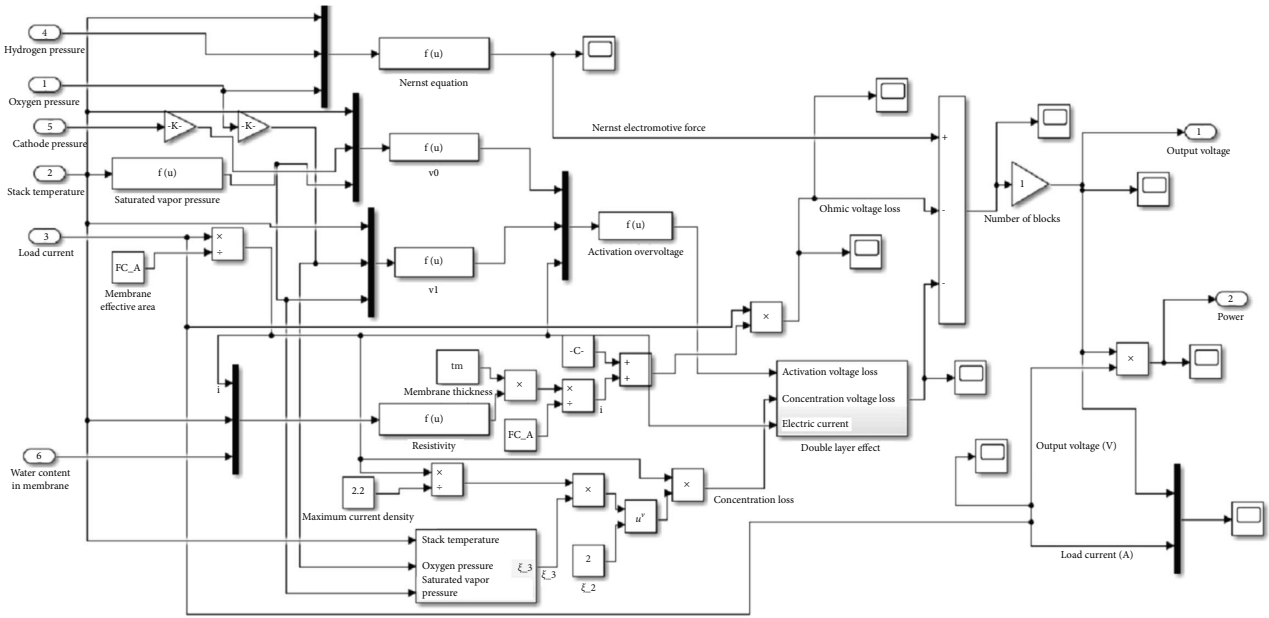
FIGURE 3: PEMFC electrochemical model simulated using Simulink toolbox.

TABLE 1: PEMFC system model simulation parameters.

Parameter	Description	Numerical value	Unit
T_{st}	Stack temperature	348.15	K
A	Single-cell activation area	220	cm^2
F	Faraday constant	96485	C/mol
P_{atm}	Standard atmospheric pressure	101325	Pa
T_{atm}	Atmospheric temperature	298.15	K
t_m	Proton exchange membrane thickness	125	μm
ξ_1	Empirical parameters	10	
ξ_2	Empirical parameters	2	
C	Double-layer charge equivalent capacitance	2.5	F
R	Universal gas constant	8.314472	
$\rho_{m,dry}$	Dry film density	0.002	kg/cm^3
$M_{m,dry}$	Dry film molar mass	1.1	kg/mol
A_{fc}	Effective area of proton exchange membrane	220	cm^2

TABLE 2: Steady state simulation conditions.

Parameter	Numerical value	Description
T_{st} (K)	348.15	Stack temperature
$P_{ca}^{O_2}$ (Pa)	3×10^4	Cathode oxygen partial pressure
P_{ca} (Pa)	3×10^5	Cathode pressure
$p_{an}^{H_2}$ (Pa)	2.6×10^5	Anode hydrogen partial pressure
λ_m	14	Proton exchange membrane water content

Ohmic overvoltage is the main voltage loss when the current density increases, which is attributed to the resistance loss of the electrolyte and electrodes (Figure 5(b)). The change in current density in this exhibits a linear trend according to the Ohm's law. The
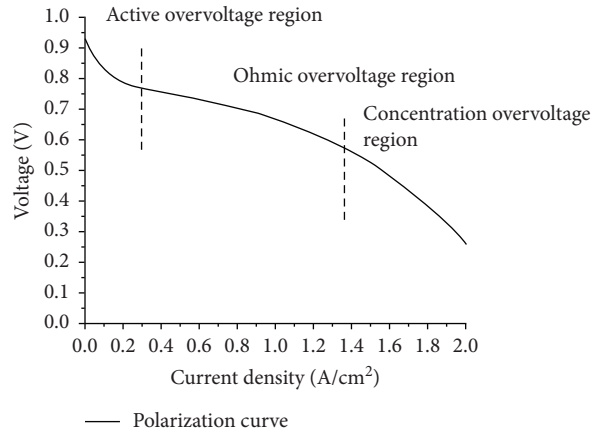

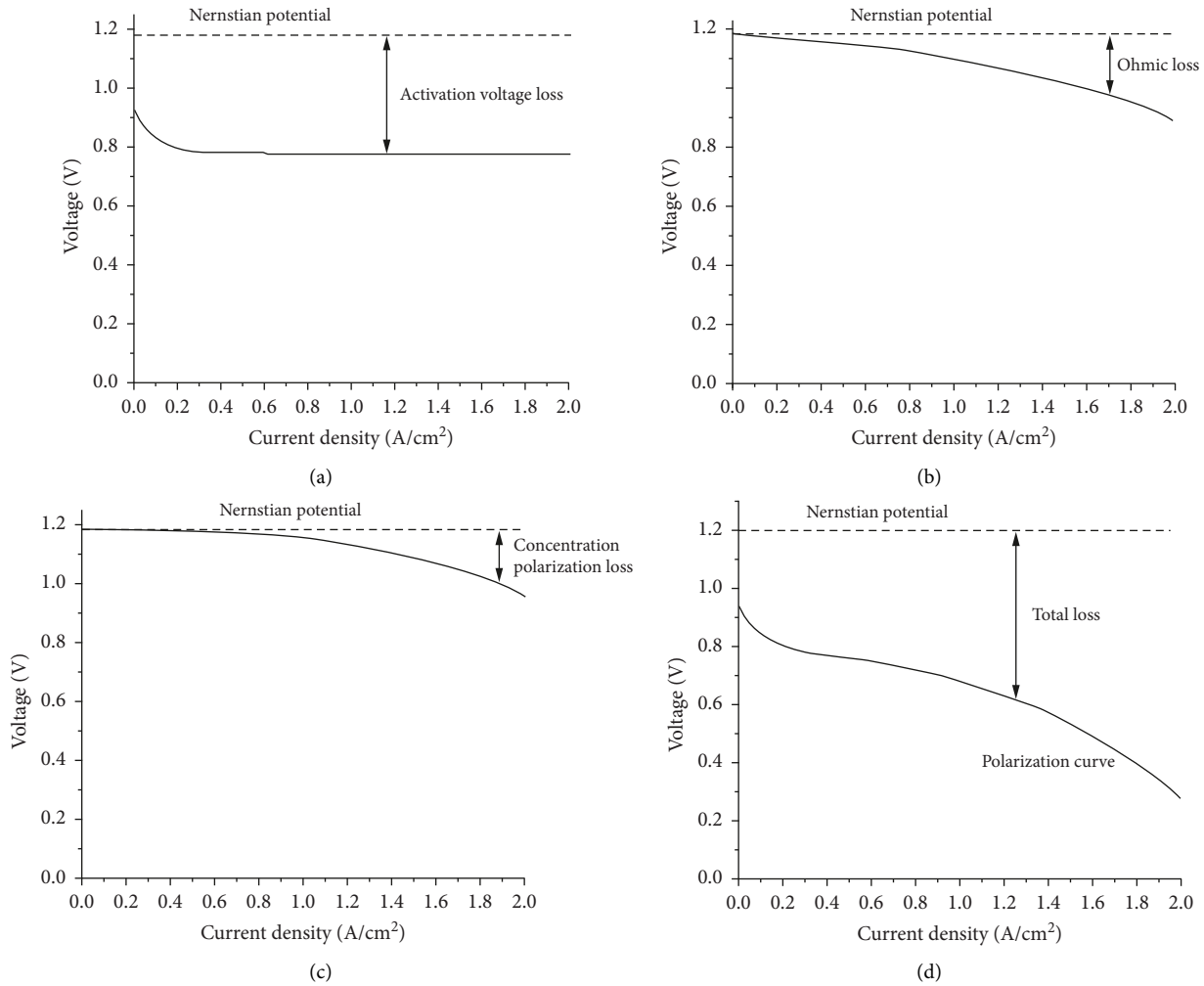
FIGURE 4: Fuel cell polarization curve.

(a)

(b)

(c)

(d)

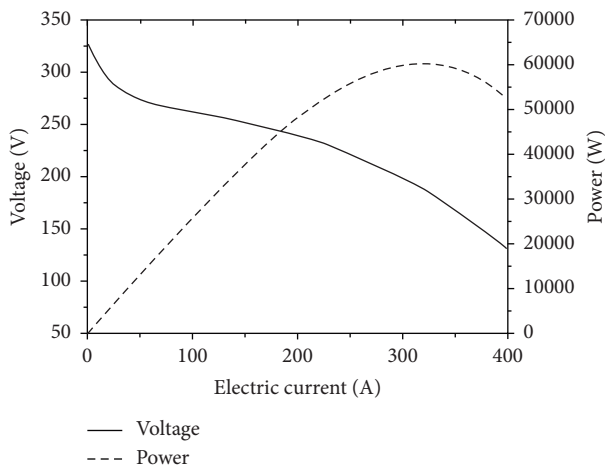FIGURE 5: effect of the three voltage losses on output voltage.

FIGURE 6: Current-voltage-power curve of a 60 kW PEMFC.

reactants on the catalyst surface rapidly decrease when the current density increases, and the replenishment speed of the reactants reduces the output voltage of the fuel cell. The concentration overvoltage is the main voltage loss in this region (Figure 5(c)). The overall loss of the output voltage of the fuel cell is presented in Figure 5(d). The polarization curve of the fuel cell was obtained under the combined action of the three losses.

The effective activation area of the PEMFC electrochemical model determined in this study was 220 cm^2, and the number of PEMFC cells was set to 350. Changes in the output voltage and output power with the load current were obtained after simulation. The findings showed that the maximum output power of the fuel cell was 60.42 kW when the load current was 317.19 A.

The dynamic performance of the 60 kW fuel cell was simulated, and the dynamic changes of the load current, oxygen partial pressure, hydrogen partial pressure, operating temperature, and membrane water content are presented in Table 3. The simulation results are presented in Figures 7 and 8.

The initial load current was 80 A, and the output voltage significantly decreased from the initial 322.2 V to a steady state of 269.8 V, under the action of activation polarization. Notably, the load current first jumped increased to 180 A between 5 s and 10 s, then decreased to 150 A. The output voltage changes were −20.39 V and 7.02 V. The output power

TABLE 3: Dynamic simulation conditions.

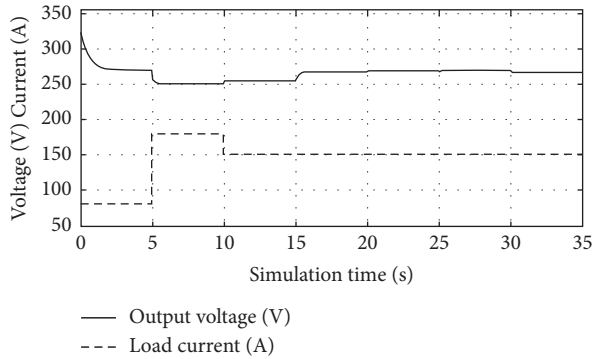| Simulation time (s)	0–5	5–10	10–15	15–20	20–25	25–30	30–35
Load current (A)	80	180	150	150	150	150	150
Partial pressure of oxygen (kPa)	30	30	30	40	40	40	40
Hydrogen partial pressure (kPa)	260	260	260	260	300	300	300
Operating temperature (°C)	75	75	75	75	75	80	80
Membrane water content	14	14	14	14	14	14	12

FIGURE 7: Dynamic response characteristics of PEMFC output voltage.
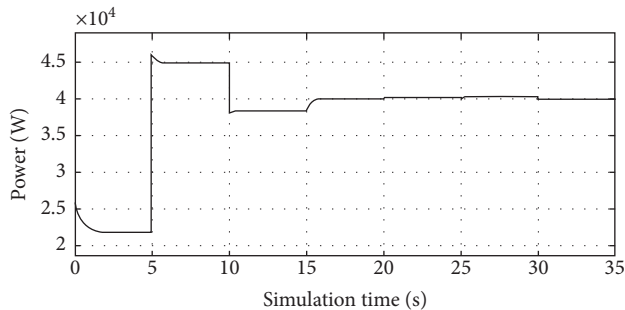

FIGURE 8: Dynamic response characteristics of PEMFC output power.

change values were 23.30 kW and −6.425 kW. The results show that the output voltage of the fuel cell decreases with an increase in the load current, and the output power increased with an increase in the load current under these working conditions. The findings indicate transitional changes in output voltage and output power.

The oxygen partial pressure and hydrogen partial pressure changed between 15 s and 20 s, whereas other conditions did not change. The output voltage increased by 10.95 V and the output power increased by 1.642 kW when the partial pressure of oxygen increases by 10 kPa. Moreover, the output voltage increased by 0.751 V whereas the output power increased by 0.113 kW when the partial pressure of hydrogen increases by 40 kPa. The simulation results indicate that the oxygen partial pressure had a greater impact on the fuel cell output compared with the effect by the change in hydrogen partial pressure. This is because activation overvoltage is the main factor that affects the output voltage, and

oxygen reduction, which is the main factor that modulates the activation overvoltage was relatively low on the cathode side. Therefore, the partial pressure of oxygen had a significant effect on the voltage drop of the fuel cell compared with the effect of the partial pressure of hydrogen. Hydrogen is typically in excess in the fuel cell anode. Nitrogen from air is the main gas on the cathode side, thus the partial pressure of oxygen in the cathode is relatively low. Therefore, it is necessary to compress the air using an air compressor to improve the output performance of the fuel cell. However, excessive gas partial pressure adversely affects the airtightness of the stack, and increases the energy consumption of the air compressor, so the oxygen partial pressure should be maintained in an appropriate range.

The operating temperature of the fuel cell increased by 5°C at 25 s, the output voltage increased by 2.07 V, and the output power increased by 0.3109 kW whereas other conditions did not change. The internal chemical reaction rate of the fuel cell increased with an increase in the working temperature. In addition, the output voltage and the output power increases with an increase in the working temperature. Notably, a large amount of liquid water in the proton exchange membrane will be vaporized under too high temperature, thus reducing the performance of the fuel cell, and even, in serious cases, eventually damaging the fuel cell. The water content of the proton exchange membrane decreased from 14 to 12 at 30 s, the output voltage decreased by 3.528 V, and the output power decreased by 0.52 kW whereas other parameters did not change. The water content of the proton exchange membrane serves as a medium for protons to pass through the exchange membrane and take part in the reaction. A lower water content is associated with a greater effect on the reaction rate and a significant polarization effect. However, too high water content, above 14, causes "flooding" of the proton exchange membrane ultimately blocking the channel and stopping the reaction.

6. Conclusion

The simulation results in the present study show that the electrochemical reaction process of the fuel cell is modulated by various factors such as reaction gas pressure, temperature, and membrane water content. Therefore, the working parameters of the fuel cell should be relatively stable and within an appropriate range to ensure efficient and reliable functioning of the fuel cell.

Data Availability

The datasets used and analyzed during the current study are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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