Progress of proton exchange membrane fuel cell

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Abstract. The rapid development of the economy of China leads to the significant growth of energy production and consumption demand. At present, the total amount of greenhouse gas emissions has ranked second in the world. To meet the challenges caused by global climate change and environmental pollution as well as realize the sustainable development of our country’s economy, carbon neutrality should be achieved. As a new type of low-temperature fuel cell, proton exchange membrane fuel cell (PEMFC) attracts a lot of attention due to its safe and environmentally friendly characteristics. However, the operating efficiency of the current commercial PEMFC is rather low (about 50%), which means that about half of the energy will be released as thermal energy. The stable and reliable heat dissipation of the fuel cell has a decisive impact on its performance and life, i.e., thermal management issues are particularly important for the efficiency and stability of the PEMFC stack. This article reviews the thermal management of PEMFC and analyzes the specific applicable conditions of cooling methods as well as the latest developments in thermal management methods. Specifically, it focuses on the thermal characteristics of PEMFC and its effects on performance and life. Besides, the PEMFC thermal management schemes and control strategies under different cooling methods are also summarized.

1. Introduction
There is a long tradition in the study of energy and environmental issues. Especially in recent years, with the aggravation of the greenhouse effect, clean energy technology has received attention from all countries. Contemporarily, the major energy sources are non-renewable energy sources that generate plenty of carbon emissions. At present, the main energy consumption in China is dominated by coal. The increasing consumption of fossil energy has led to an increasingly serious greenhouse effect and energy crisis. Lack of energy and low energy efficiency are the characteristics of energy use in developing countries, so the energy transition is becoming more and more important [1]. Hydrogen energy is recognized as clean energy and is known as the most promising secondary energy in the 21st-century, which has the characteristics of clean, pollution-free, convenient storage and transportation, and high utilization rate. Meanwhile, it has a wide range of sources and various production methods [2]. Fuel cells are devices that directly convert the chemical energy of the fuel into electrical energy without the combustion process, which posses the advantages of high energy conversion efficiency and low pollutant emissions [3]. Among them, proton exchange membrane fuel cell (proton exchange membrane fuel cell, PEMFC), as a new type of low-temperature fuel cell, has the characteristics of low operating temperature, low noise, high power density, safety, and reliability, etc [4].

During the working process of PEMFC, hydrogen is supplied to the anode and split into protons and electrons. Protons are conducted to the cathode through the polymer membrane, and electrons are
conducted to the cathode through the external circuit and react with oxygen (from the air) to generate water and heat [5]. So far, the energy conversion efficiency of commercial PEMFC is generally about 50%, which means that 50% of the chemical energy is released in the form of thermal energy [6]. Heat generation by the fuel cell comprises entropy heat of reaction, irreversible heat of electrochemical reaction, ohmic heat, and condensation heat of water vapor, which will cause the internal temperature of the stack to rise. As shown in Figure 1, the first three heats accounted for 35%, 55%, and 10% of the total heat generation [7]. To ensure that the temperature in the fuel cell stack is maintained at a stable level, PEMFC needs to dissipate the heat in time during operation [8].

In order to ensure the efficiency and stability of the PEMFC stack, thermal management is particularly important. Its main purpose is to ensure the uniformity of internal temperature and that the stack operates at a reliable and efficient temperature level as well, avoiding local high temperatures [9]. Peng and Lee [10] developed a three-dimensional single-phase non-isothermal numerical model for the single cell of PEMFC to investigate the thermal effects on fuel cell performance. They strongly recommend active thermal management to maximize battery performance. A simple way to improve fuel cell performance is to operate at the highest temperature allowed by the system. At high temperatures, increased electrochemical activity and reaction rate is conducive to the improvement of performance. In addition, the operating temperature will affect the theoretical electromotive force of the fuel cell. The higher the temperature, the lower the activation energy required for the electrochemical reaction [5]. The temperature control of the fuel cell is mainly reflected in the cooling of the battery. To improve energy efficiency, it is very important to design a reasonable cooling device to release the heat of the fuel cell. The cooling of traditional fuel cells mainly compromises air cooling and liquid cooling. Generally, for high-power (>5kW) fuel cells, liquid cooling is the most common cooling method, which has the advantages of high heat transfer capacity, low flow rate, etc [11]. However, it will increase the volume of the fuel cell system, complicating the structure and increasing the cost. Air cooling is mainly suitable for the heat dissipation of small fuel cell stacks, with simple structure, convenient operation, relatively easy control, and low cost [12]. For low-power stacks (~100 W), the cooling method is often based on increasing the air supply volume of the cathode. The advantage is that the structure is simple, but it does not have sufficient precise temperature control, which depends largely on the temperature and humidity of the ambient air. Due to the poor heat dissipation capacity of the air and the relatively low heat capacity, the cooling power is limited. Generally, the power of the stack is required to not exceed 5 kW. This cooling method must use a fan to force air convection. However, this will easily cause the unevenness of the temperature in the stack, increasing the voltage difference between batteries, which eventually reduce the total power of the entire stack [13]. The air-cooled PEMFC integrates the cooling and the oxidizer subsystem, which reduces the quality, volume, and cost of the fuel cell, and reduces the control complexity [14]. Wang C S et al. [15] established a single-channel fuel cell temperature field model, obtaining the temperature change of the reactant gas in the process of passing through the channel and the diffusion layer to the electrochemical reaction zone of the catalytic layer. Li Xi et al. [16] proposed a nonlinear model
between battery temperature and operating parameters through fuzzy clustering identification methods. Based on the analysis of heat transfer, Wang B R et al. [17] constructed an air-cooled fuel cell temperature field thermal model equation and obtained the best temperature value of the fuel cell output power at different current and voltage loads through experiments. For liquid cooling, the core of thermal management lies in the design of the flow field [18]. The flow method of the liquid in the cooling flow field will affect the temperature distribution in the PEMFC stack. The more uniform the coolant flow, the more uniform the temperature distribution will be [19]. Nowadays, the commercial fuel cell stack uses the mixture of deionized water and ethylene glycol as the cooling medium, which has high specific heat capacity and low price. However, during the working process, the cooling medium will ionize, which further affects the conductivity of the stack and corrodes the liquid channel. Therefore, deionization equipment must be added to the cooling medium [20]. In recent years, it has been discovered that nanofluid is also a good choice [21] in addition to using deionized water as a cooling medium. Liquid cooling is currently the most stable, effective, and widely used thermal management method. This method can make the internal temperature distribution of the fuel cell more uniform, which is conducive to the stability and durability of the battery [22]. Three fuel cell vehicles including Toyota Mirai, Honda Clarity, and Hyundai Nexo all use liquid cooling to remove waste heat from the reactor [23]. This article reviews the thermal management issues in PEMFC recently, and aims to find out the effects of temperature on PEMFC stacks. Subsequently, the best performance of stacks under different thermal management levels are found out.

2. Electrothermal output characteristics of PEMFC

PEMFC is composed of cathode, anode, electrolyte diaphragm, and bipolar plate, as illustrated in Figure 2. The electrolyte membrane adopts a proton exchange membrane, which plays a dual role in separating fuel and oxidant and conducting protons [24]. The hydrogen reaches the anode through the duct or gas guide plate. Under the action of the anode catalyst, it decomposes into hydrogen ions (protons) and releases electrons. The basic reaction is as follows:

Anode: \[ H_2 \rightarrow 2H^+ + 2e^- \] (1)

Cathode: \[ \frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O \] (2)

Total response: \[ H_2 + \frac{1}{2}O_2 \rightarrow H_2O + \text{heat} \] (3)
The heat (or enthalpy) of a chemical reaction is the difference between the heat of formation of the product and the reactant, namely:

\[ \Delta H = (h_f)_{H_2O} - (h_f)_{H_2} - \frac{1}{2} (h_f)_{O_2} \]  

(4)

The heat of forming liquid water is -286 kJ/mol (at 25°C), and the heat of forming these elements is zero by definition. The enthalpy in a chemical reaction is negative, which means that heat is released in the reaction, that is, it is an exothermic reaction. Therefore, the formula (4) can be written as:

\[ H_2 + \frac{1}{2} O_2 \rightarrow H_2O(l) + 286 \text{kJmol}^{-1} \]  

(5)

Equation (5) is only valid at 25°C, indicating that both the reactant gas and product water are at 25°C, and the enthalpy (286 kJ/mol) is also called the high heating value of hydrogen. When the product water exists in gaseous form, there is about 241 kJ/mol corresponding heat is released, which is the low heating value of hydrogen. However, since every chemical reaction produces some entropy, part of the high heating value of hydrogen cannot be converted into useful work, that is, electric energy. The partial reaction enthalpy (or the high heating value of hydrogen) that can be converted into electrical energy in a fuel cell corresponds to the Gibbs free energy, which is determined by the following formula:

\[ \Delta G = \Delta H - T \Delta S \]  

(6)

where \( \Delta H \) characterizes the energy of the material system; \( \Delta S \) is a measure of the degree of chaos in the system.

The theoretical potential of the fuel cell is:

\[ E = \frac{G}{nF} \]  

(7)

where \( F \) is the product of Avogadro's constant and an electron's charge; \( \Delta S \) is a measure of the degree of chaos in the system.
Since $\Delta G$, $n$, and $F$ are all known, the theoretical potential of hydrogen/oxygen fuel cell can be calculated:

$$E = -\frac{DG}{nF} = \frac{237340 \text{ J mol}^{-1}}{2.96485 \text{ A mol}^{-1}} = 1.23 \text{ V}$$

(8)

At 25°C, the hydrogen/oxygen fuel cell has a theoretical potential of 1.23V. If formula (6) is substituted into formula (8), the following formula can be obtained:

$$E = -\left(\frac{DH}{nF} - \frac{TDS}{nF}\right)$$

(9)

Obviously, the increase in battery temperature will reduce the theoretical potential of the battery. However, during the operation of the fuel cell, the battery temperature is usually higher. This is because the voltage loss will decrease as the temperature rises during the operation of the fuel cell, which is greater than the compensation value of theoretical potential loss.

The performance and durability of the battery are affected by temperature. Within a certain range, the performance of the battery will be improved when the temperature of the battery rises. Besides, temperature also affects catalyst activity (improving the activity and increasing the reaction rate), fuel gas diffusion (improving gas mass transfer), and moisture diffusion in the proton exchange membrane (reducing membrane resistance and increasing conductivity) [25]. Temperature can also affect the gas permeability of the proton exchange membrane. If the cell temperature is too low, the polarization phenomenon in the electrode will be intensified and the output voltage will be reduced, which will lead to the deterioration of the cell performance. Whereas, if the cell temperature is too high, the membrane will dehydrate, shrink or even rupture, and the strength will be reduced to a certain extent. Moreover, the partial pressure of water vapor in the fuel gas will increase, diluting the concentration of the reaction gas. In the most serious case, it will cause the loss of water in the proton exchange membrane. Therefore, maintaining a good operating temperature is essential to keep the normal operation of the electrochemical reaction inside the battery, and the long-term stable operation of the proton exchange membrane [26].

3. Active air-cooled PEMFC

3.1. Air-cooled PEMFC stack structure

During the operation of the air-cooled PEMFC, excess air is extracted into the cathode to provide the oxygen required for the reaction and take away the reaction heat. Air cooling is mainly used for heat dissipation of small fuel cell stack, with simple structure, convenient operation, relatively easy control and low cost. The stack structure simplifies the cooling, humidification, air compressor and air pump system [27]. It should be noted that for air-cooled PEMFC, the rotational speed of the radiator should be appropriate, which is conducive to the stability and reliability of the system. If the speed is too fast and the heat dissipation is large, the temperature may be too low. Thereby, the internal temperature of the battery stack to be not high enough, and the battery performance will be seriously degraded. If the situation is opposite, the system temperature may be too high and affect the stability of the system [12].

The experimental stack of Wei D et al. [28] is composed of 22 single cells. The temperature and humidity are controlled by adjusting the voltage of the air supply fan and the exhaust gas emission cycle. Cao S Y et al. [29] took a 500 W air-cooled proton exchange membrane fuel cell stack as the research object. Thermocouples were evenly inserted on the cathode side to record and observe the changes in the internal temperature of the stack. Based on the electronic load test, the output performance of the stack and the cell voltage distribution under different loads are recorded to comprehensively analyze the performance of the stack. The experimental equipment is shown in Figure 3.
3.2. Thermal characteristics of air-cooled fuel cells

Based on the air-cooled technology, Wei D et al. [28] investigated the impacts of ambient temperature and load change on the working temperature and humidity of the reactor. Specifically, it obtained the influence law of temperature characteristics on the output performance, and acquired the optimal working temperature under the optimal performance output. The experimental results of the stack show that the environmental temperature change has a direct impact on the optimal operating temperature of the same current output, especially in the low current section, and the impact gradually weakens as the current increases. The optimal operating temperature is greatly affected by the high-voltage output section, and the influence gradually weakens as the voltage output decreases. The root cause of this phenomenon is that the electrochemical reaction speed determines the heat release, and the stack temperature affects the hydration state of the proton membrane. In order to obtain the best output performance of the stack, temperature control is required. The effect of temperature adjustment is not only related to the control algorithm, but also limited by the cooling method and the cooling capacity of the fan. The traditional PID algorithm cannot meet the requirements of the rapidity of load changes, i.e., the adaptive fuzzy method is used to design the control system. Cao S Y et al. [29] conducted an experimental study on the internal temperature distribution of the 500W air-cooled PEMFC stack. According to the results, the internal temperature distribution of the stack is uneven, which has a lot to do with the position. The temperature at the bottom of the stack is higher than the temperature at the top. This is because the top of the stack is located at the hydrogen inlet, and the water generated by the electrochemical reaction inside the stack gathers at the bottom under the action of gravity. Hence, it leads to the concentration loss or flooding easily, which in turn leads to increased local heat generation. Since the probability of flooding at the top is small and the entry of hydrogen at room temperature will reduce the inlet temperature, the temperature near the hydrogen inlet is lower. Pei H C et al. [30] detected the temperature by placing a miniature thermocouple in the cathode flow field, and found that the highest temperature in the battery stack is at the exit of the stack. Besides, it is noted that the greater the temperature difference of each single cell with the increase of current density. In the meantime, the temperature difference between the single cells is getting bigger and bigger. Setareh Shahsavari et al. [31] designed the three-dimensional thermal model of the air-cooled PEMFC stack, and measured the changes of selected parameters (e.g., intake speed, bipolar plate thermal conductivity, and GDL thermal conductivity, etc.) on the maximum temperature and temperature distribution of the stack. The experimental results indicate that the air velocity and the in-plane thermal conductivity of the bipolar plate are the key factors affecting the air-cooled PEMFC stack.
4. Active liquid-cooled PEMFC

4.1. Liquid-cooled PEMFC structure

For high-power (>5kW) fuel cells and automotive fuel cells, liquid cooling is the most common cooling method. Compared with air cooling, it has the advantages of high heat transfer capacity and low flow rate [11]. The specific heat capacity of liquid is larger than that of air, i.e., the use of liquid cooling makes the distribution of fuel cells more even. Liquid cooling makes it possible to design independent cooling channels between the anode and cathode stages of the fuel cell, relying on the forced convection heat exchange of the coolant to take away the heat generated during the working process [32]. Fuel cells have very strict requirements on ion concentration. When the ion concentration exceeds a certain value, the performance of the fuel cell will be affected. The liquid coolant can be deionized water, a mixture of water and ethylene glycol, or a nanofluid containing nanoparticles, because they are non-conductive and anti-freezing [20]. I. Zakaria et al. [33] analysed the application of nanofluid coolant in the single cooling plate of polymer electrolyte membrane fuel cell. Based on the results, it is found that in the volume ratio of 6:4 and 5:5 water and ethylene glycol mixture, 0.1% concentration of Al2O3 is the best nanofluid coolant material. However, in the 5:5 mixture of that, which is the second best one. Chen W R et al. [34] proposed a temperature control strategy with cooling water inlet pressure as the adjustment target. By adjusting the speed of the cooling water pump and the speed of the radiator fan, the cooling water flow rate and the stack cooling water inlet temperature are respectively controlled. The control object is the operating temperature of the fuel cell, and the actuator is the circulating water pump and the radiator fan. Both the cooling water pump and the radiator fan are driven by DC motors. The circulating water pump drives the cooling water circulation to take out the heat generated during the PEMFC power generation process. Afterward, the high-temperature cooling water flowing out of the stack passes through the radiator, and the radiator fan rotates to convect the air, thereby reducing the temperature of the cooling water.

Niu Z et al. [35] proposed an improved temperature control strategy with flow following power to realize the decoupling of cooling water pump and cooling fan. The actuators for stack temperature control are cooling water pump and radiator fan. By controlling the voltage of the cooling fan and the frequency of the cooling water pump, the stack inlet temperature and the temperature difference between the stack cooling water inlet and outlet are respectively adjusted. Besides, the stack working temperature is stabilized through the coordinated control of the cooling water pump and the cooling fan. Chen X W et al. [36] designed a liquid-cooled HT-PEMFC with a net output power of 6kW (running on pure H2) illustrated in Figure 4. For the supply of cathode oxidant, a corresponding amount of ambient air is blown into the stack according to the output current. The air damper is located at the cathode exhaust to regulate the air flow and keep the stack warm under low load conditions. For stack heating, the coolant is heated by the heater and circulated through the coolant circuit (stack, radiator, and water tank) by a pump.

![Figure 4. Simplified schematic diagram of HT-PEMFC module [36]](image-url)
Tae-Won Song et al. [37] also designed a cooling device for liquid-cooled HT-PEMFC, which is composed of a heat exchanger, a liquid storage tank and a solenoid valve. When the stack coolant leaves the stack, the water in the liquid phase enters the liquid storage tank, while the steam in the steam phase enters the heat exchanger. The secondary coolant passes through the heat exchanger to cool the steam of the stack coolant. Due to the difference between the water level inside the reservoir and the cooling plate, condensate enters the cooling plate from the reservoir at the simultaneously. In this case, the internal circulation of the stack coolant in the cooling channel is realized by natural convection.

4.2. Thermal characteristics of air-cooled fuel cells
Tae-Won Song et al. [37] also designed the basic parameters required for a liquid-cooled HT-PEMFC cooling device. According to the experimental results, the pumpless cooling method provides a more uniform temperature distribution in the reactor regardless of the direction of the coolant. In order to solve the strong coupling in the actual operation of the PEMFC stack and avoid the short-time high temperature caused by the large current load, an improved temperature control strategy is proposed by Chen W R et al. [34]. The traditional strategy can only track the temperature change, but the temperature itself has a great delay, which leads to a long adjustment time. In order to simultaneously consider the impact of temperature on the fuel cell and the pressure withstanding capacity of the fuel cell plate, the temperature control strategy as shown in Figure 5 is proposed. The temperature control strategy, aiming at the difference between the cooling water inlet pressure and the air inlet pressure, shall be satisfied by adjusting the cooling water inlet temperature by controlling the speed of the radiator fan:

$$\left| \begin{array}{cc} P_{air, in} & P_{coolant, in} \\ P_{coolant, in} & P_{air, in} \\ P_{coolant, in} & P_{air, in} \end{array} \right| \leq P_{ws}$$

where $P_{coolant, in}$ is the inlet pressure of the stack cooling water; $P_{air, in}$ is the air inlet pressure of the stack; $P_{ws}$ is the maximum unidirectional pressure that the plate can withstand.

The experimental results demonstrate that the improved control strategy is superior to the traditional control strategy in temperature fluctuation, system coupling strength, fuel cell performance and lifetime.

5. Other thermal management methods of PEMFC
In addition to the above conventional air-cooled, water-cooled active heat management, other heat management methods, including phase change cooling, heat pipe cooling, evaporative cooling, belong to non-active cooling. Phase-change cooling eliminates waste heat in fuel cells by means of enthalpy
of vaporization. Applications in automotive cooling systems can be classified as heat pipe cooling and evaporative cooling [38]. Heat pipe cooling is to embed the heat pipe into the bipolar plate, in the absence of external power input, heat pipe will be a large amount of heat through the cross-section for long-distance transferring heat. Evaporative cooling is the introduction of liquid water into the flow channel to remove waste heat from the fuel cell through the phase change of water evaporation. Compared with other cooling methods, evaporative cooling can dissipate heat and humidify the battery simultaneously, and evaporative water can be collected in the water tank for use. It does not need an external humidifier or a separate cooling plate to be added to the battery stack. Generally, deionized water is chosen as the coolant [39].

6. Research conclusions
In conclusion, this paper summarizes the structure and temperature control strategy of air-cooled and water-cooled for state-of-art PEMFC. Although the temperature affects the performance and life of PEMFC, it is necessary to maintain the optimal output performance of PEMFC at a good temperature. Non-active cooling mode is just beginning in fuel cell field. It is worthwhile to explore the path to combine non-active cooling mode with PEMFC more effectively to achieve better heat dissipation effect.

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