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Research Progress on Thermal Management of Proton Exchange Membrane Fuel Cells based on Application Scenarios

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Abstract

Proton exchange membrane fuel cell (PEMFC) is a clean and efficient chemical energy power generator. Based on the working principle and thermal balance management mode of PEMFC, this paper analyzes the way that PEMFC overcomes the obstacles of high and low temperature operation. In order to provide support and help for the thermal design of PEMFC based on application scenarios, it analyzes the performance features and advantages of PEMFCs in different application scenarios, such as domestic energy supply, aircraft, vehicle and ship. Furthermore, it analyzes the application technology, thermal design difficulties in different working environments, the research progress and development prospects of different platform projects.

Keywords

Proton Exchange Membrane Fuel Cell; Thermal Management; Application Scenario.

1. Introduction

Fuel cells are power generation devices that directly convert the chemical energy stored in fuel and oxidizer into electrical energy with high efficiency and pollution-free [1]. As PEMFC's energy conversion is not limited by the Carnot cycle, its theoretical efficiency can be as high as 80% or more [2]. Meanwhile, because hydrogen and oxygen are used as fuel and oxidant, the product is only water, which is clean and low-carbon. Among various types of fuel cells, PEMFC has the advantages of high energy conversion rate, environmental friendliness, and low operating temperature. It is an ideal clean energy technology. It is known as the future energy and has received great attention.

In the working process, hydrogen loses electrons at the anode and is oxidized. At the cathode, oxygen combines with hydrogen ions that passing through the proton exchange membrane (PEM) and electrons transmit from the external circuit to form water and release a lot of heat [3]. The sources of heat include irreversible electrochemical reaction heat, ohmic heat, and phase change heat [4]. Maintaining a stable operating temperature has an important impact on the performance and life of the fuel cell. Poor heat dissipation, which will further affect the gas supply and electrochemical reaction, will cause the temperature to exceed the membrane's design tolerance temperature when it's working and affect the temperature uniformity. The transport characteristics of PEM and the management of the water produced will ultimately affect the performance and stability of the PEMFC [5]. For this reason, attention must be paid to the design of thermal management methods. At present, the main thermal management solutions of PEMFC include air cooling, liquid cooling, phase change cooling, passive cooling, etc., which are suitable for different working environments [3].

Due to the limitations of objective conditions in different application scenarios, it is necessary to design a suitable fuel cell for a specific application scenario, including its power level, cooling method, size and other technical indicators. Under the same energy density, the higher the power is, the larger the volume of the fuel cells are. The cooling method of fuel cells is also related to the system

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complexity. For example, the system complexity of air-cooled fuel cells is lower than that of water-cooled fuel cells. In certain application scenarios, the system needs to be lightweight to ensure the system's running time is as long as possible, which requires the system to be as simple as possible. For example, fuel cells used in Unmanned Aerial Vehicles (UAVs) need to be lightweight and the thermal management system as concise as possible to ensure long time fly for UAVs.

For example, in 2019, a four-rotor fuel cell UAV developed by South Korea's MetaVista uses a 6L ultra-light liquid hydrogen storage tank that stores 390g of hydrogen and Intelligent Energy's 650W light fuel cell power module to provide power to the UAV. The flight time of 10h 50min 5s broke the world record of multi-rotor UAV flight time [6], and its fuel cell power system only weighs 1kg. The system adopts a direct air-cooling cooling method with characteristics of simple system structure and low control complexity.

For application scenarios such as automobiles and ships, high-power fuel cells are required to drive the vehicle. The power level is ~10kW to ~100kW generally. For example, the hydrogen fuel cell supply vessel "Viking Lady", which was put into use in 2009, uses power up to 320kW fuel cell system [7]. At the 20th China International Maritime Exhibition in 2019, China Shipbuilding 712 Research Institute released the country's first 500 kW marine fuel cell system solution with independent intellectual property rights [8].

In recent years, home-based local energy Internet solutions have gradually attracted attention, it owns broad potential application prospects in unstable grid or remote areas. The home scene has high requirements on the volume and stability of the fuel cell. A typical household fuel cell combined heat and power system (commercial name ENE-FARM) hot water tank has a volume of 200L and a mass of 250kg. Its hot-electric conversion efficiency is 39%. The waste heat generated in the process of power generation can be used to supply hot water and heating, with a heat recovery efficiency of 56%. The overall efficiency of the whole process is as high as 95% [9]. This kind of devices can be applied to many family houses.

Based on the working principle and thermal management mode of the PEMFC, this paper analyzes the development direction of adapting to various operating environments through different types of cooling modes and assembly designs and summarizes the current design focus and development under various working conditions and its developing prospect.

2. Structure and working mode of PEMFC

2.1 The structure and working principle of PEMFC

The current mainstream fuel cells include solid oxide fuel cells (SOFCs) and PEMFCs. Compared with earlier fuel cell technologies such as phosphoric acid fuel cells (PAFCs), besides the environmental advantages of materials, the PEMFC technology has a low working temperature and higher safety and is currently more widely used.

PEMFC is an energy conversion device based on electrochemical principles that directly converts the chemical energy stored in fuel and oxidant into electrical energy. The components of PEMFC include a PEM, catalyst layers, microporous layers, gas diffusion layers, and bipolar plates. The principle of power generation is closed to the principle of the primary battery. Take a hydrogen fuel cell as an example. After losing electrons at the cathode, hydrogen becomes hydrogen ions (protons) that pass through the PEM to reach the anode. Electrons flow from the cathode to the anode from the external circuit and react with oxygen and hydrogen ions at the anode to produce water. This forms a loop and generates current. The electrochemical reaction equations are as follows:

Cathodic Reaction:

$$H_2 \rightarrow 2H^+ + 2e^-; \tag{1}$$

Anodic Reaction:

$$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O; \tag{2}$$

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Overall Reaction:

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O\,; \tag{3}$$

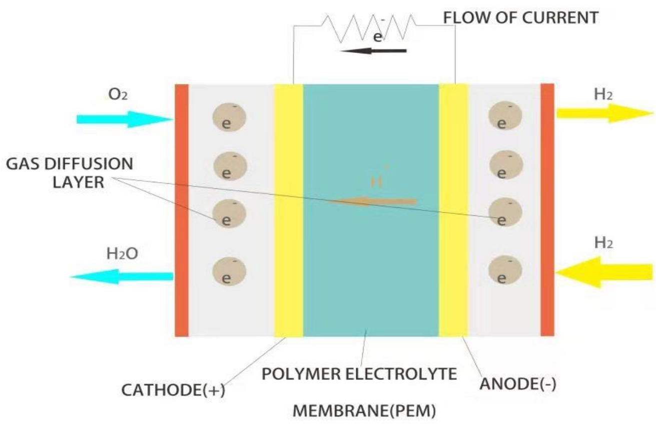

Figure 1. PEMFC's working principle and structure diagram

2.2 The source of battery heat

The energy conversion efficiency of PEMFC is about 50% [3], so nearly half of the chemical energy is released in the form of heat.

The entire reaction equation of the fuel cell is the same as the hydrogen combustion reaction. At 25°C, 100kPa one mole of hydrogen burns to release 286kJ of heat, and that is an exothermic reaction.

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O + 286.02kJ/mol \tag{4}$$

There is no combustion in the fuel cell, and the maximum energy (heat energy) released by the hydrogen combustion reaction cannot be completely converted into electrical energy. Due to the generation of entropy ΔS, there will be irreversible losses in the energy conversion process, which can be expressed by the following formula:

$$\Delta G = \Delta H - T\Delta S \tag{5}$$

ΔH and ΔS are the difference between the heat of the product and the reactant and the difference between the entropy,

$$\Delta H = (h_f)_{H_2O} - (h_f)_{H_2} - 1/2\,(h_f)_{O_2} \tag{6}$$

$$\Delta S = (s_f)_{H_2O} - (s_f)_{H_2} - 1/2\,(s_f)_{O_2} \tag{7}$$

The h_f and s_f values of the reactants and products in the hydrogen combustion reaction at 25°C and 100kPa are shown in the following table:

Table 1. Enthalpy and entropy of fuel cell products and reactants (at 25°C and 100kPa)

	h_f/(kJ mol^{-1})	S_f/(kJ mol^{-1})
H_2	0	0.13066
O_2	0	0.20571
H_2O (l)	-286.02	0.06996
H_2O (g)	-241.98	0.18884

It can be seen that in this condition, amid the 286.02 kJ mol^{-1} energy generated, the fuel cell can convert up to 237.34 kJ mol^{-1} energy into electric energy, and the remaining 48.68 kJ mol^{-1} energy into heat energy.

When expressing efficiency, the low heating value of hydrogen is usually used to express the efficiency of the fuel cell. At this time:

$$\Delta G = 228.74 kJ\ mol^{-1} \tag{8}$$

The theoretical maximum efficiency of a fuel cell is:

$$\eta = \Delta G / \Delta H_{LHV} = 228.74/241.98 = 94.53\% \tag{9}$$

3. Thermal design of PEMFC based on different scenarios

3.1 Solutions for the normal operation of PEMFC in high and low temperature environments

Many fuel cell application platforms face large temperature changes during operation, and the performance of fuel cells will be affected by different temperatures. When the ambient temperature is 0°C, the water in the fuel cell will freeze, which is not conducive to the normal operation of the battery. As water freezes, its volume expands, squeezing the internal structure of the battery, and even damages the internal components. The ice inside the fuel cell may prevent the gas from reaching the surface of the catalyst, causing low-temperature start up failure. The ice in the cell may damage the membrane electrode and cause the performance of the fuel cell to decrease [10]. Therefore, after the fuel cell is shut down, it needs to be purged to remove water. Otherwise, after multiple freeze-thaw (F-T) cycles, the PEMFC will be severely damaged [11].

The components in the low-temperature PEMFC can operate normally under high temperature conditions except for the PEM [12]. Under temperature conditions of 100°C~200°C, most fuel cells operating in this temperature range use phosphoric acid (PA) as the proton carrier, and the catalytic efficiency of the electrode reaction is significantly improved compared to that at low temperatures. The heat generated by the battery reaction does not need to be dissipated in time, so its thermal management system can be simplified. The water in the battery mainly exists in the form of gas phase at high temperatures, which facilitates the discharge of water and simplifies the water and heat management system. Under high temperature conditions, impurity molecules such as CO and SO_2 are less likely to be adsorbed on the surface of precious metal catalysts such as platinum (Pt), which greatly improves the electrode's tolerance to impurity molecules.

The current commonly used HTPEM material polybenzimidazole (PBI) itself has low proton conductivity, and it usually needs to be doped with proton acid to be used as a PEM. When doped with the same amount of proton acid, the order of the proton conductivity of the proton acid doped PBI proton exchange membrane is $H_2SO_4 > H_3PO_4 > HClO_4 > HNO_3 > HCl$. Although the PBI proton exchange membrane doped with sulfuric acid has the highest proton conductivity, it requires high environmental humidity, so it is not suitable for use under high temperature conditions [13]. PA doped PBI (PBI/PA) composite membrane can maintain high proton conductivity under high temperature and low humidity, so it has attracted wide attention [12]. However, phosphoric acid, as a water-soluble electrolyte, is easily soluble in the by-product water when the fuel cell is working, resulting in a decrease in the proton conductivity of HT-PEM and degradation of battery performance.

3.2 Application of PEMFC in Household Scenes

Household PEMFC systems have been widely used in countries with frequent natural disasters such as Japan and are supported by government subsidies. Among them, the household fuel cell combined heat and power system (ENE-FARM) produced by Panasonic has been successfully commercialized, and its improved version has entered the market in the second half of 2020. ENE-FARM generally adopts liquid cooling. The cooling liquid is water, and the cooling water can be directly used as domestic water after being heated, which improves the overall energy utilization rate of the system. It is very suitable for households for Its working noise is less than 40 decibels. Considering that it is difficult for individual households to obtain a safe and stable source of pure hydrogen supply, this equipment uses natural gas to produce hydrogen. The use of waste heat recovery devices to assist household heating increases the overall energy utilization efficiency.

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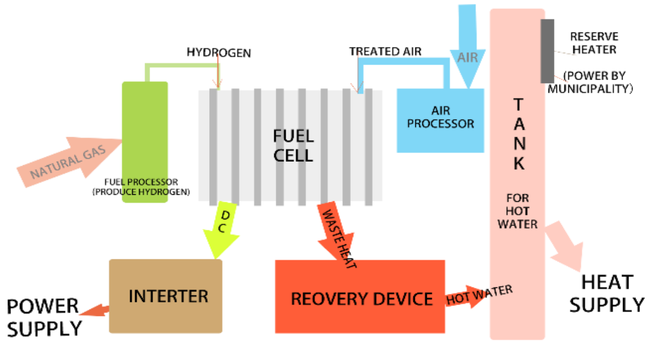

Figure 2. Working principle and structure of ENE-FARM system

ENE-FARM can generate electricity for eight consecutive days in the event of a power failure. The maximum AC generation power is 500W [14]. It can be used to power heating and hot water, as well as home appliances such as smart phones, to maintain the minimum electricity required for people's lives, and to generate power of shelter for people in the event of a disaster. In the course of its development, the equipment is becoming more compact and the floor space is shrinking. The combined footprint of the fuel cell and hot water tank in the domestic fuel cell system is reduced to less than $1m^2$, and it can even be used in smaller houses, so it can also be applied in cities with a small per capita living area such as Tokyo. The power generation efficiency is about 39%, the heat recovery efficiency is about 56%, and the overall efficiency is about 95% [9]. ENE-FARM is a very efficient clean energy power supply system. It uses the principle of hydrogen fuel cell to significantly reduce household carbon dioxide emissions.

It must be noticed that the application market for the ENE-FARM system designed by Panasonic at this stage is still relatively narrow. According to the research and investigation of Shen Ruibao et al. [9], the current price of this system is quite high, the design life is limited, and the short-term breakeven can only be achieved in households in developed countries such as Japan.

However, considering the soaring demand for environmental protection in the world, the continuous economic development of various countries in the world, the living standards and electricity consumption habits of residents are gradually rising closer to developed countries, the equipment cost and market pricing of such equipment still have a considerable decline in prospects, and the market prospects are considerable.

3.3 Application of PEMFC in aircraft

The fuel cell-driven aircraft system has the advantages of environmental friendliness, low operating cost, and high safety. With the breakthrough of its key technologies, it will bring greater economic benefits [15].

The power source of the aircraft is required to generate a large amount of power when starting, and needs stable power during normal cruising. Lithium batteries have high power density but limited power and are suitable for high-power discharge application scenarios; fuel cells has high energy density but small output power change, which is suitable for long-term discharge. Currently, most of the hydrogen energy aircrafts use the above two types of batteries to meet their work needs. For example, the air-cooled fuel cell modules with rated power ranging from 650W to 2.4kW manufactured by Intelligent Energy have been widely used in power supply systems for UAVs and other portable devices.

Fuel cell technology is a good solution to the high endurance energy supply system for aircraft, but there are obvious obstacles at present. For example, the hydrogen energy UAV mentioned above can only guarantee the reliability of operation under the working condition of 20-50℃, and the operation ability at high altitude is not ideal. Taking the UAV application environment as the representative. Due to the high working conditions and strong internal environment requirements of the fuel cell stack itself, and in order to extend the flight time of the UAV as longer as possible, the UAV's heat dissipation system generally adopts an air-cooled structure. The air-cooled structure of fuel cell has two types: open cathode and closed cathode. The open cathode structure is simple and has high energy density, so it is more suitable for UAV application scenarios [16]. Due to the extremely compact mechanical system of the UAV and the relatively small space, it is difficult for the gas pressurizer used to carry more auxiliary devices to reduce the impact of the cooling air on the working conditions. When the flight altitude increases, the water content of the compressed air provided by the air supply system is difficult to control either, and the use of bottled liquid oxygen supply is undoubtedly a dangerous and bloated solution. At present, most of the public designs still manage to improve the air compression system. In terms of energy supply, most of them use hydrogen for energy, whose difficulties are the limitation of hydrogen production technology and the of manufacturing and storing hydrogen cylinders. In general, the cooling system, gas supply system, hydrogen cylinder size and replacement restrictions on the PEMFC solution make it difficult to reduce the cost of aircraft and the application environment is limited. However, as a clean energy aircraft, it has the advantages of lightweight that are currently difficult to be replaced. It is more suitable for the design of small and medium-sized aircrafts under commercial and military conditions.

At present, the improvement of intake control system is an important research direction. In order to obtain the maximum battery output power and take advantage of the high efficiency function of the flight system, the gas oxygen concentration in the stack and the working environment pressure should be kept at the optimal value when the PEMFC is working. From curve fitting analysis, the best compressor power can be obtained as a cubic function of fuel cell current [17]. As shown in equation (10), the optimal compressor power under different fuel cell currents under curve fitting analysis is:

$$P_{com}^{op} = f(I_{fc}) = -0.0035I_{fc}^3 - 0.007I_{fc}^2 + 1.6I_{fc} + 0.02 \quad (10)$$

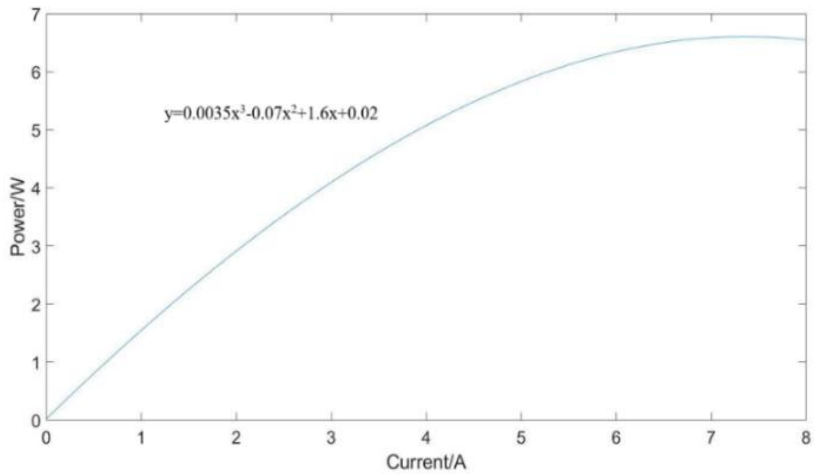

Figure 3. Optimal compressor power

Yang Wengang’s research [17] optimizes the control method of the fuel cell system to improve performance based on the adaptive neuro-fuzzy inference algorithm, and realizes the control of the compressor power to improve the energy-saving effect under relatively ideal aircraft conditions.

The current design schemes of hydrogen storage cylinders are generally divided into the following categories [18]. The aircraft system prefers the design schemes of type III and IV cylinders with low weight-to-volume ratio. The use of carbon fiber technology to make composite material high-performance oxygen cylinders can be effective to resists load and working environment temperature and pressure, and has good application prospects in aircraft.

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Table 2. Comparison of performance parameters of hydrogen storage cylinders

Type	Craftsmanship	Work pressure /MPa	Weight-to-volume ratio /(kg/L)	Service Life /year	Cost
I	Pure steel metal	17.5~20	0.9~1.3	15	Low
II	Steel liner, carbon fiber winding	26.3~30	0.6~0.95	15	Medium
III	Aluminum inner liner, carbon fiber winding	30~70	0.35~1	15~20	The highest
IV	Plastic liner, carbon fiber winding	70 above	0.3~0.8	15~20	High

At the same time, referring to the current supply of hydrogen production companies, hydrogen production and hydrogen perfusion technologies should be popularized as soon as possible in the current GB/T 37244-2018[19] standard. Although the hydrogen concentration requirement of 99.999% is reduced to 99.97% compared to the GB/T 37244-2012 standard, the requirements for CO, sulfur, particulates and other impurities have been greatly improved or proposed for the first time, aiming to reduce the poisoning of the platinum catalyst used in the PEMFC and increase the operating life of the system effectively.

3.4 Application of PEMFC in Automobile

As the main commercial platform for the development of fuel cells, new energy vehicles have considerable representative significance in design and experiments, and show the development of small and micro PEMFC technology.

Fuel cells for vehicles face a similar dilemma as fuel cells for UAVs, that is, fuel cells are suitable for stable power discharge occasions but not for high-power discharge occasions. Therefore, the current fuel cell vehicles are generally a hybrid of fuel cells and batteries or internal combustion engines.

Compared with UAVs, cars have more space to install fuel cells and heat dissipation modules to achieve better performance. Therefore, vehicle fuel cells generally use liquid cooling for heat dissipation. Since both the fuel cell and the battery are high-voltage electrical devices, the problem of coolant conductivity cannot be ignored. The main cause of the increase in the conductivity of the coolant is that the flux sprayed during the production of the radiator separate out conductive ions during use. After analyzing the flux composition, Zhang Shaopeng [20] found that the main reason for the increase in the conductivity of the coolant was the dissolution of aluminum fluoride, and the process of releasing conductive ions to increase the conductivity was mainly in the initial soaking stage of the radiator. These phenomena can be solved by cleaning and surface repair.

PEMFC usually works in the temperature range of 60℃~80℃ [21]. Below this temperature range, the catalyst activity will decrease, and temperatures higher than this range will cause dehydration of the PEM. The heat dissipation methods of vehicle fuel cells include exhaust heat dissipation, radiation heat dissipation and liquid cooling heat dissipation. Since the exhaust temperature is around 70℃, its heat dissipation efficiency is far lower than that of the internal combustion engine whose exhaust gas is about several hundred Celsius. The heat emitted by exhaust only accounts for 3% to 5% of the total heat dissipation [22]. Radiation heat is a very small part of fuel cells and internal combustion engines, only about 1% [22]. The remaining 95% of the heat needs to be discharged by the radiator with the coolant. The heat dissipation of the radiator is shown in the following formula under the experimental study [23]:

$$Q_w = c \cdot \rho \cdot V_{H_2O} \cdot (T_{out} - T_{in}) \tag{11}$$

Where c is the specific heat capacity of water; ρ, the density of water; V_{H_2O}, volume flow of water; T_{out}, outlet temperature of fuel cell stack's circulating water; T_{in}, inlet temperature of fuel cell stack's circulating water.

The operating temperature of the internal combustion engine is about hundreds of degrees Celsius. When the coolant temperature is the same, the temperature difference between the PEMFC and the

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coolant is smaller, so it is harder to discharge heat. Therefore, fuel cell vehicles need to improve the layout of the radiator to meet its work needs.

Xia Mingzhi [2] and others found that by increasing the fan power, enlarging the radiator area, and changing the radiator position, the heat dissipation effect of the fuel cell can be improved. However, increasing the fan power will increase the heat production of auxiliary facilities such as battery packs. Although extending the radiator area is effective, the area that can be increased in a compact application scenario such as a car is very limited, and sometimes even the body structure needs to be modified.

The cold start of vehicle fuel cells is also one of the problems that need to be solved. According to experience, when the temperature is above -5°C, the fuel cell can be warmed up by the heat generated by the stack, and the water produced during operation will not freeze. When the temperature is below -10℃, other methods are needed to warm up the fuel cell stack [24]. In addition, water purge treatment before shutdown is also essential. The current cold start methods include heat preservation method, heating method and self-start method [11]. Heat preservation method is to reduce the heat exchange with the outside as much as possible or use an auxiliary heat source after shutdown so that the temperature of the fuel cell after shutdown will not lower than 0℃. For example, the fuel cell stack is immersed in the coolant after the shutdown, and the heat of the coolant is used to maintain the temperature of the fuel cell stack. But this method only works in situations with short downtime. If the downtime is long, the auxiliary heat source may exhaust the battery of the car and eventually cause the car to fail to start [25]. The heating method can be divided into cooling liquid heating (adding hot cooling liquid to the heat dissipation system), intake heating (heating the gas passing into the anode and the anode), and catalytic combustion (passing the mixed gas with low hydrogen content to the cathode and catalytic combustion) [11]. The self-starting rule applies to environments above -5°C.

3.5 The application of PEMFC on ships.

At present, ships in the world mainly use diesel engines as power equipment, and most of the fuel used is oil, causing serious pollution [26]. For example, fuel leakage will damage the ecological environment of the water area and pollute aquatic products. Nowadays, clean energy ships are the key research objects of countries all over the world. Among them, fuel cells have attracted much attention because of their low noise and low emission characteristics.

Fuel cell systems are usually mixed with other power systems on ships, such as solar cells, storage batteries, and internal combustion engines. For example, a cruise ship powered by solar energy will use storage batteries and fuel cells as auxiliary power to provide power when the photovoltaic system is insufficient to generate power [27]. Countries around the world have successively launched their own ships powered by fuel cells in this century. For example, the hydrogen-wind-light hybrid ship "Energy Observer" built by France in 2017 uses a 22kW hydrogen fuel cell system. China has also developed a 500kW fuel cell system with independent intellectual property rights.

Due to their compact structure, high energy conversion efficiency and good silent performance, fuel cells have been used in the design and manufacture of air independent propulsion systems (AIP) since the middle of the last century. Now it's one of the research directions of the power system of submarine.

Since this system has low operating noise and its main product is water, it has good concealment and anti-tracking capabilities. When applied to submarines, it is suitable for high-power fuel cell systems for power supply, because it works underwater, the fuel cell system has good heat dissipation performance. However, the fuel cell system requires a large-capacity energy storage system to store high-purity compressed hydrogen gas and oxygen. This design is quite difficult and insecure for submarines. The current development direction is mainly to improve the hydrogen production technology and hydrogen storage tank technology, while continuing to optimize the battery efficiency and control system, and to reduce the cost of the program [28].

4. Conclusion

Fuel cells have attracted attention and have been used in various scenarios due to their clean and efficient characteristics. When people solve the needs of different application environments, various important technologies have been developed in response to meeting requirements of different application environments and external working conditions, and they had achieved great progress in the last decades.

In general, new energy vehicles are more inclined to use liquid-cooled design of small stacks, emphasizing compact structure. Air-cooled fuel cell stacks are widely used in aircraft, and more attention is paid to optimizing system thermal management and improving various parts of the battery cell technology. However, due to the high cost, only preliminary commercialization has been achieved at present, and there is still a certain distance from popularization. Household fuel cell systems have been widely used in Japan and some countries, but in other countries, such as China, they are still not concerned. On ships, especially submarines, fuel cells have been maturely used. At present, fuel cells are developing in the direction of pursuing modularity, combination, high reliability, and long life in various application scenarios to improve their competitiveness.

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