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Current status of cold start research on proton exchange membrane fuel cells

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Abstract Proton exchange membrane fuel cell vehicles have attracted widespread attention for their outstanding advantages such as high energy conversion efficiency, long range, and zero emissions. However, the cold start pro blem in low-temperature environment has become a major obstacle limiting its commercialization and wider application. Therefore, researchers have conducted a large number of experiments and simulations to investigate the fuel cell water transport phase change and heat transfer mechanisms as well as the optimization of start-up strategies. By studying the distribution, transport and phase transition processes of water and ice during the initiation process, it was found that the reaction product water first saturates the water content of the proton exchange membrane and catalytic layer with membrane state water, and finally discharges the cell or ices suddenly. By studying the heat transfer process of cold start, it is found that the middle part of the reactor cell and the middle region of the single cell heats up the fastest and is the main region of heat generation, and the irreversible reaction heat generated by the cathode oxygen reduction reaction is the main source of heat generation. Based on the cold start mechanism of fuel cells, researchers have developed various self-start and assisted-start strategies, and conducted numerous simulations and experiments to verify the superiority of the strategies. The purpose of this paper is to summarize the research on fuel cell cold start.

Keywords Proton exchange membrane fuel cell ; Cold start ; Water transport and phase change ; Control Strategy

1. Introduction

In recent years, the development of new and renewable alternative energy sources has become urgent due to the increase in energy demand [1], the fluctuation of fossil fuel prices [2], and the greenhouse gas emissions generated by fossil fuel-powered vehicles and industries [3]. Hydrogen, as an odourless, non-toxic, zero-emission fuel, has become a good energy carrier by virtue of its high energy content, which has attracted extensive attention from countries around the world [5]. The development of "hydrogen economy" [6] and the construction of "hydrogen society" [7] is a huge industrial revolution involving transport, construction, industry, manufacturing, power and general energy use in the whole society. Fuel cell vehicles, with their outstanding advantages such as high energy conversion efficiency, short fuel refuelling time, long range and zero emission, have become an important part of building a "hydrogen society", and are considered to be a promising alternative to traditional fossil-fueled vehicles [8]. At present, fuel cell vehicles have entered the commercialisation stage from the demonstration stage. However, there are still many key factors constraining the further commercialisation and promotion of fuel cell vehicles [9]. Among them, the cold-start problem of fuel cells due to cold regions and climatic conditions is one of the major barriers to the commercialisation of fuel cells [10,11]. To address the cold-start problem of fuel cell vehicles, the U.S. Department of Energy has proposed the ultimate cold-start target for an 80 kW fuel cell system: 50% of the rated power of the system should be reached within 30 s at -20° C, and the energy consumption of the startup and shutdown process should not be more than 5 MJ; meanwhile, it is necessary to achieve the cold-start at -30° C without assistance and the cold-start at -40° C without assistance. At the same time, unassisted cold start at -30° C and assisted cold start at -40° C should be achieved. Based on these objectives, fuel cell cold start technology has developed rapidly in recent years. At present, the parameters of some typical fuel cell vehicles are shown in Table 1, and the typical fuel cell models of Toyota and Honda in Japan and Hyundai in South Korea, which are at the forefront of the world, have achieved the requirement of fast cold start at -30° C within 30s without mentioning the -40° C cold start. On the other hand, the cold start performance of fuel cell vehicles in Europe, America and China is much worse, and some models have not even disclosed the cold start performance parameters.

Although some fuel cell vehicles can meet the cold-start requirements to a certain extent, and can achieve fast cold-start in most environments, there is still a big gap between them and the required target, and it is difficult to solve the problems of durability and performance degradation of fuel cells caused by cold-start [12]. Therefore, researchers have conducted a lot of studies on the output performance change and durability degradation mechanism of fuel cells during cold start [13]. The results showed that the formation of ice inside the cell at low temperatures is the main cause of the cold-start performance and cell durability degradation. Through a large number of experimental characterizations and modelling simulations, researchers have conducted in-depth analysis of the distribution, transport and phase transition of water and ice as well as the heat transfer process, and optimized the fuel cell cold-start strategy for the fuel cell cold-start mechanism. This paper summarises and reviews the current status of fuel cell cold start research at home and abroad.

Table 1: Cold start performance parameters of a typical fuel cell vehicle

Vehicles	Country	Status	Output power(kw)	Driving range(km)	Cold start temperature (℃)	Cold start time(s)
Toyota Mirai	Japan	Mass production in 2014	114	550	-30	30
Toyota II Mirai	Japan	Announced 2020	128	650	-	-
Honda Clarity	Japan	Mass Production 2015	130	700	-30	30
Hyundai Nexo	Korea	Mass Production 2018	120	800	-30	30
Mercedes-Benz GLC F-cell	Germany	Mass Production 2018	155	478	-	-
GM Chevrolet Colorado ZH2	United States	Announced in 2016	132	-	-	-
SAIC Rongwei 950 Fuel Cell	China	Displayed in 2014	36	430	-20	-

2. Study of water transport and phase change mechanism in cold start-up

Since the degradation of cold-start output performance and durability of fuel cells is mainly related to the formation of ice inside the cell, it is of great importance to study the distribution of water and ice in the cell, and the process of water transport and phase transition. The researchers combined the direct observation of water and ice in the cold-start experiment and the simulation of water and ice distribution, transport and phase transition based on the fuel cell cold-start model to analyse the mechanism of water transport and phase transition.

2.1 Observation and characterisation of water in different forms

With the continuous development of experimental and characterisation techniques, in addition to indirectly investigating the transport and phase transition properties of water during cold start by measuring the output performance of cold start under different start parameters, researchers also use various advanced observation and characterisation means to directly observe the changes of internal water during the start process

Ge and Wang [14] employed a silver mesh as the GDL of the cathode to directly observe the water transport and phase transition process on the CL surface of the cathode, and it was found that only ice existed in the CL during start-up at 20 mA/cm2 and -5°C; liquid water droplets were found at the CL interface at start-up temperatures above -3°C. Several small holes of about 400 um diameter distributed at different locations in the actual cathode GDL were also attempted to provide a good optical pathway for direct observation of the behaviour of water or ice on the CL surface during cell start-up [15].

In addition to transparent cells, neutron imaging has been widely used in the observation of cold-start water and ice in fuel cells by virtue of its non-destructive nature and sensitivity to hydrogen-containing compounds [16].Wang et al. [17] directly observed the change of water content in the flow channel and underneath the ridges during cold-starting by neutron images.Mishler et al. used high-resolution neutron radiography to detect the behaviour of ice formation during cold start in PEMFC and found that the peak ice thickness exists on the cathode side of the MEA.Santamaria et al. observed the condensation and redistribution of water in the GDL and flow channels during cell cooling with neutron imaging, followed by 3D tomography and reconstruction of the images into a 3D model. The results of tomography imaging can well show the regions of ice formation and blockage in the flow field. The results show that ice is mainly formed in the flow channel due to the repulsion of water by the GDL; under the effect of gravity, larger ice mainly accumulates in the lower cathode outlet region.

2.2 Simulation of fuel cell cold start process

In addition to the direct observation and characterisation of water and ice, the simulation study of fuel cell cold start is also helpful to analyse the internal change process of the cell and the distribution of each parameter, reduce the cost of experiments and shorten the experimental cycle.

Mao et al [18] established a multiphase transient model of PEMFC during cold start by comprehensively considering the formation of ice in the cathode CL and GDL, water transport at very low temperatures, phase change heat transfer, oxygen mass transfer, electrochemical kinetics, and the interactions between these factors. Validated by a large amount of experimental data, the model was used to predict the cold-start performance of the PEMFC, which could reveal the distribution of current density, temperature, membrane water content and ice volume fraction in the CL, and the simulation results emphasised the effect of water icing in the cathode CL.Meng [19] developed a transient multiphase multidimensional PEMFC model within a hybrid domain framework to elucidate the fundamental physical fuel cell cold start characteristics. Simulations of cold start at -20°C under constant current and pressure conditions showed that the water vapour concentration within the cathode gas path affects ice formation in the cathode CL, and the membrane plays an important role in successful cold start by absorbing the reaction product water to hydrate during cold start.

At present, a systematic and complete cold-start mechanism system verified by multiple sets of experiments and multiple characterisation methods has not yet been formed, so it is still necessary to improve the simulation hardware equipment and technology level. In the future, we can try to build a three-dimensional simulation model for cold start of microporous structures involving the water phase transition process, and carry out an in-depth study of the phase transition mechanism of water transport in the microporous structures during the cold-start process through more direct multi-physics field coupling simulation, and at the same time, we will continue to develop more advanced characterisation and testing methods to validate the results of the simulation.

3. Heat transfer mechanism study of cold start

3.1 Heat transfer process of cold start

In fuel cell cold-start simulation, a one-dimensional model is suitable for calculating the heat production of the fuel cell and the mutual heat transfer of the internal structure, especially when the specific distribution of the ice volume fraction is not taken into account, and the use of a one-dimensional model can better balance the modelling details and the computational efficiency [20].Khan-delwal et al. [21] established a transient one-dimensional thermal model of a general-purpose PEMFC power stack, considering the phase change of water and the effect of recirculation of coolant flow on the thermal mass and temperature distribution of the power pile, and investigated the cold-start capability and the corresponding energy demand under different operating and environmental conditions. The results show that for the selected power stacks, there exists an optimal range of operating current densities that allows the stacks to start quickly at low temperatures and suggests thermal insulation at the end plates to shorten the cold start time.

Luo et al [22] developed a three-dimensional multiphase PEMFC stack model to study the cold start process of an automotive PEMFC stack. The simulation results show that the higher the number of cells in the stack, the faster the temperature rises, and the temperature of the cells in the middle is higher and more uniformly distributed than that of the cells on both sides of the stack and a single cell.Chippar and Ju[23] developed a fuel cell stack model to simulate the heat generation during cold start. The results showed that the irreversible reaction heat dominated by the cathode oxygen reduction reaction contributed most of the heat production.

Ishikawa et al [24] investigated the behaviour of supercooled water during cold start of a fuel cell by cross-sectional imaging with visible and infrared images, and found that liquid water and supercooled water freezing exothermic as the same phenomenon, and that the heat of condensation diffuses at the GDL/MEA interface when the cell performance is degraded.Lin et al. have successively investigated the cold-starting behaviour of a five-chip fuel cell short stack by using a partitioned test technique. The experimental results showed that, for a monolithic cell, the initially most reactive region for startup moved from the inlet end to the middle of the cell, and then until the cell operated normally, the current density in the middle was always the highest, which played a dominant role in cell warming. In the stack, the two cells at the outermost side, which are close to the end plates, have the worst reaction performance and warm up the slowest. This indicates that the heat dissipation from the end plates to the outside world is the primary factor affecting the thermal insulation performance of the fuel cell, which is consistent with the simulation results of Chippar and Ju [23]. Meanwhile, it is also found that, for the two outermost cells, the performance of the cell near the inlet end is worse than that of the cell near the outlet end, which indicates that the convective heat transfer between the low-temperature gas and the cell is also a major factor leading to the loss of heat from the cell.

3.2 Heat transfer mechanism of cold starting

Without considering the additional heat sources that may be added in the auxiliary starting method, the sources of heat generated during cold starting of a fuel cell include Irreversible reaction heat dominated by cathode redox, Joule heat due to the electric current, and latent heat released due to the phase change processes such as sublimation, freezing, and liquefaction of different forms of water [23]. The main ways of consuming heat are the constant heat dissipation from the outside of the fuel cell, especially the end plates, into the cryogenic environment, the convective heat exchange between the cryogenic reactive gases with the cell as they pass through the flow channels, and the heat absorption by melting of the ice previously present in the interior of the cell when the temperature rises above freezing point.

On the parallel plane of the MEA, the inlet is the main reaction area at the very beginning of startup and produces the most heat and water. Since the short time of heat production cannot raise the cell temperature above freezing, the product water in this part of the cell rapidly freezes, providing part of the latent heat of phase transition while inhibiting the reaction at the inlet. Next, the middle of the cell begins to dominate the cell warming as the main reaction region. If the temperature successfully rises above freezing before this part of the reaction fails, the reaction is able to continue and the cold start is successful; conversely, as the reaction in the main reaction region fails, the cell is unable to provide enough reaction heat to sustain the continuation of the warming, and thus the cell begins to cool down under the influence of the ambient low temperature and the cold start fails.

In the vertical plane of the MEA, due to the higher thermal conductivity and lower specific heat capacity of the GDL relative to the PEM, the conduction of heat generated by the reaction at the cathode CL to the PEM and the performance of temperature rise in this part of the PEM are worse than that of the cathode GDL, and the performance of temperature rise in the anode GDL region is even worse. During the low-temperature startup of the PEMFC, the reaction-generated heat at the cathode CL will be conducted to the lower-temperature cathode GDL side faster, and the temperature in this part of the region can be driven up when the reaction-generated heat is sufficient. When the start-up temperature is low and the reaction heat is insufficient, this limited heat energy is also more easily conducted and consumed outward from the cathode GDL side, and cannot effectively drive up the temperature in this part of the area. For the stack, the battery in the middle plays a leading role in warming up the battery due to less contact with the outside world and slower heat dissipation, and once the temperature of the battery in the middle successfully rises above freezing, it can drive the whole stack to successfully complete cold start.

4. Optimisation of fuel cell cold start strategy

With the deepening of research and the knowledge of fuel cell cold start mechanism, more and more cold start optimisation strategies have been proposed. According to the different heat sources, these optimisation strategies can be divided into self-starting strategy and assisted cold-starting strategy. The self-starting strategy is to use the heat released by the electrochemical reaction inside the cell for heating, in which the cold-starting strategies involved mainly include the optimisation of the starting method and control strategy as well as the optimisation of the internal structure of the cell itself. Among them, the starting methods can generally be divided into constant current starting, constant voltage starting and maximum power mode starting [25], while the control strategy is adjusted or combined on the basis of these three starting methods to pursue the optimal way. The self-start scheme has received extensive attention from researchers in the automotive industry because it does not use an external heat source, can avoid the complexity and power consumption brought by the auxiliary heating system, and radically reduces the cold start time.

4.1 Optimisation of self-start mode and control strategy

According to the contents of cold start experimental research, it is known that the most common start-up mode for cold start is constant current start-up.Zang and Hao [26] investigated the performance of cold start-up under different current density operation modes, and obtained corresponding current density thresholds that can achieve successful start-up.Lei et al [27] used a one-dimensional transient numerical model to study the constant-current loading mode, the slope loading mode, the variable-slope loading mode , sawtooth current loading mode and stepwise variable current loading mode on the cold start of a power reactor at -20°C, respectively. The results show that the stepwise variable current loading mode has better start-up time, temperature rise rate, ohmic impedance, heat production rate, ice volume fraction and water production efficiency.

For the cold start of the electrostack, since the output current of the electrostack in the constant-current mode is constant, when the single cell in the stack is unable to provide enough electrons through the hydrogen-oxygen electrochemical reaction, it tends to provide electrons by other means, such as electrolysis of water, carbon corrosion, and oxidative corrosion of the catalyst, which results in the damage of the fuel cell structure, and seriously affects its durability [28]. Therefore, involving the electrostack level, the constant voltage start-up approach can avoid the above problems to a certain extent.Jiang and Wang compared constant voltage start-up with constant current start-up and concluded that constant voltage start-up is more advantageous than constant current start-up in terms of heating time versus energy demand, and concluded that the cold start-up solution with constant voltage is one of the best approaches proposed so far in terms of energy demand and system cost.

However, the constant voltage start method has not yet been applied on a large scale in the automotive field due to a number of limitations such as the stringent procedure for determining the value of the start voltage, the dependence on the state of the PEMFC, and the inability to adapt to changes in attenuation and operating conditions. Therefore, in recent years, researchers have tried to develop an integrated starting method by combining the respective advantages of constant current and constant voltage.Hu et al [29] reported a cold-start control strategy for a metal plate fuel cell electric stack with 30 cells and a rated power of 7 kW. It was found that the use of current-voltage synergistic control method (controlling the current to speed up the startup and limiting the voltage to ensure the safety of the fuel cell stack) could maximise the heat production efficiency of the fuel cell stack and significantly reduce the time of the startup process.

The various start-up schemes and control strategies tried by researchers in constant-current mode, constant-voltage mode and maximum-power mode are all aimed at pursuing the optimal hydrothermal management scheme, i.e., generating as much heat as possible through electrochemical reactions in a short time, while avoiding a large amount of water accumulating inside the cell after the saturation of the PEM to inhibit the reaction. With the updating and optimisation of real-time monitoring means and control algorithms, the cold start strategy has evolved from simple open-loop control to closed-loop regulation with real-time feedback. The core idea can be summarised as in the early stage of start-up, the water content of the membrane is small, by increasing the start-up current to make the reaction quickly produce a large amount of heat and thus rapidly increase the temperature of the battery, and when the water content of the membrane is close to saturation, the current density is adjusted to reduce the amount of icing of water in the reaction products to ensure the normal reaction, and to achieve the balance between the production of water and the production of heat.

4.2 Optimisation of auxiliary cold-start strategy

The optimisation of the self-start scheme can certainly improve the cold-start performance of the fuel cell fundamentally, but when it comes to the extremely low temperature environment and the fast cold-start demand proposed in the engineering practical application, the self-start scheme alone cannot enable the cell to achieve a fast cold-start from a very low temperature. montaner et al. [30] studied the cold-start performance of the 4 kW power stack at -30°C. They found that for fast cold-start, the self-start strategy is only applicable to cold-start temperatures above -15°C. cold start and found that for fast cold start, the self-start strategy is only applicable for cold start temperatures above -15°C. For cases below -15°C, the assisted start strategy can avoid icing for a more favourable start, despite the extra energy required to use a heater. Therefore, in practice, more efficient assisted cold-start strategies are often used based on batteries with better self-starting capabilities [31].

There are many ways to achieve an external heat source for the assisted cold-start strategy, the simplest being the addition of a heater.Li et al [32] proposed a localised heating method to improve the cold-start performance of a fuel cell by placing a heater wire under some of the ridges of the cathode plates. Coolant heating is more effective than air heating due to its higher specific heat. By circulating the coolant, the battery pack can be heated more uniformly, resulting in better voltage uniformity of the battery pack.Wei et al [33] considered the flow and heat transfer in the coolant circulation during cold start of an on-board fuel cell and established a three-dimensional transient cold start model. The simulation results show that increasing the coolant flow rate or coolant tank capacity has little effect on the battery voltage, but it increases the inhomogeneity of temperature distribution along the flow direction. And although increasing the coolant flow rate will lead to a more uniform current density distribution, it will also lead to an increase in the amount of icing and will affect its positional distribution.

In addition to the strategy of adding an external heat source to assist cold start, the choice of purge strategy is also important for cold start as the water content in the cell before start-up has a significant effect on the cold start performance, and as it is necessary to remove as much water as possible by gas purge during the shutdown period of the electric stack, the choice of purge strategy is also very important for cold start.

Sinha and Wang [34] investigated the effect of blowdown conditions on blowdown effectiveness and found that the use of low relative humidity and high flow rate gas blowdown with high cell temperature resulted in better water removal and helped to improve the cold-start performance.Kim et al [35] improved the effectiveness of blowdown by adding a small amount of hydrogen to the cathode gas stream. The results showed that the hydrogen addition blowdown method was effective in removing the residual water near CL. Cold start experiments were performed after this blowdown method, and it was found that there was little attenuation of CL and the cold start performance was improved.

5. Summary

Fast and safe startup of PEMFC at low temperatures is a key technology to ensure the normal operation of the fuel cell and extend its lifetime. The lack of a systematic and complete water transport and phase change mechanism system, the lack of a large-area fuel cell stack simulation model at the automotive system level, the poor self-starting performance, and the high cost and energy consumption of assisted fast cold-starting have limited the large-scale commercialisation of fuel cells. Researchers have conducted in-depth studies on the water distribution, heat transfer, mass transfer, phase transition and performance degradation of fuel cells during the whole cold-start process by various means such as experiments and simulations, and tried and optimised a variety of cold-start schemes, which have achieved fruitful results. The establishment of a fuel cell stack cold start model can simulate the heat transfer process on the vertical plane of the MEA during the cold start process, and it is concluded that the main source of heat is the irreversible reaction heat of the cathode redox reaction, and the cell in the middle of the stack warms up the fastest. The temperature distribution and heat transfer process on the parallel plane of the MEA can be measured practically in the cold-start experiment by using the partitioning test technique, and it is found that the region of the main reaction is shifted from the air inlet at the beginning of the startup to the middle region of the cell.

Fuel cell cold start strategy can be divided into self-start strategy and assisted start strategy. Among them, the self-starting strategy includes the optimisation of the starting method and control strategy with the optimisation of the internal structure of the cell itself; the auxiliary start-up mainly includes the strategies of gas blowing, heater, reaction heating, gas heating and coolant heating. For large-area on-board PEMFCs, matching the hydrothermal distribution area on the parallel plane of the MEA will also have an impact on the low-temperature starting performance, and at the same time, there are problems such as the lack of maturity of the fast cold-start technology, and the difficulty of balancing output performance and durability.

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