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Review

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Research Progress on Aging Prediction Methods for Fuel Cells: Mechanism, Methods, and Evaluation Criteria

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Abstract: Due to the non-renewable nature and pollution associated with fossil fuels, there is widespread research into alternative energy sources. As a novel energy device, a proton exchange membrane fuel cell (PEMFC) is considered a promising candidate for transportation due to its advantages, including zero carbon emissions, low noise, and high energy density. However, the commercialization of fuel cells faces a significant challenge related to aging and performance degradation during operation. In order to comprehensively address the issue of fuel cell aging and performance decline, this paper provides a detailed review of aging mechanisms and influencing factors from the perspectives of both the PEMFC system and the stack. On this basis, this paper offers targeted solutions to degradation issues stemming from various aging factors and presents research on aging prediction methods to proactively mitigate aging-related problems. Furthermore, to enhance prediction accuracy, this paper categorizes and analyzes the degradation index and accuracy evaluation criteria commonly employed in the existing fuel cell aging research. The results indicate that specific factors leading to aging-related failures are often addressed via targeted solving methods, corresponding to specific degradation indexes. The significance of this study lies in the following aspects: (1) investigating the aging factors in fuel cells and elucidating the multiple aging mechanisms occurring within fuel cells; (2) proposing preventive measures, solutions, and aging prediction methods tailored to address fuel cell aging issues comprehensively, thereby mitigating potential harm; and (3) summarizing the degradation index and accuracy evaluation standards for aging prediction, offering new perspectives for resolving fuel cell aging problems.

Keywords: fuel cell; aging prediction; degradation index; failure factors

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1. Introduction

In order to mitigate environmental pollution, the development of new energy sources has become a global priority [1]. Hydrogen, as a pollution-free green energy source, is regarded as a crucial component of new energy [2]. As a novel energy device and the carrier of hydrogen energy, fuel cells have undergone extensive research due to their advantages, including zero carbon emissions and high energy density [3].

There are various types of fuel cells, where proton exchange membrane fuel cells (PEMFCs) have advantages such as low operating temperature, low noise, and ease of integration [4]. In addition, proton exchange membrane fuel cells (PEMFCs) are characterized by their exceptional efficiency, low emissions, rapid start-up capabilities, and compact, lightweight nature [5]. These qualities render them highly suitable for a wide range of potential applications, including automotive, portable devices, and backup power sources, among others [6].

The proton exchange membrane fuel cell (PEMFC) is an electrochemical device employed for the conversion of chemical energy into electrical energy. The fuel used in PEMFC is hydrogen gas (H_2), while the oxidant is oxygen gas (O_2). These two gases undergo reactions within the fuel cell. At the anode, hydrogen gas (H_2) undergoes a process of decomposition into protons (H^+) and electrons (e^-). At the cathode, oxygen gas (O_2) combines with protons and electrons to produce water (H_2O). The chemical reactions of the anode and cathode are as follows:

$$H_2 \rightarrow 2H^+ + 2e^- \qquad O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$$

Electrons flow through an external circuit, traveling from the anode to the cathode, thereby generating an electric current. This electric current can be harnessed to perform work or provide power. PEMFC efficiently and cleanly converts hydrogen and oxygen gas into electrical energy, thus achieving zero carbon emissions.

Figure 1 shows the components of PEMFC. As shown in Figure 1, the PEMFC stack consists of an end plate, a current collector plate, and multiple individual single cells together. Each individual cell is composed of seven major components: the proton exchange membrane, catalyst layers on the anode and cathode, gas diffusion layers, and bipolar plates [7]. During operation, PEMFCs require the collaborative functioning of auxiliary system components such as an air compressor, hydrogen recirculation pump, DC/DC converter, etc., to ensure the stable chemical reactions of hydrogen and air within the fuel cell stack.

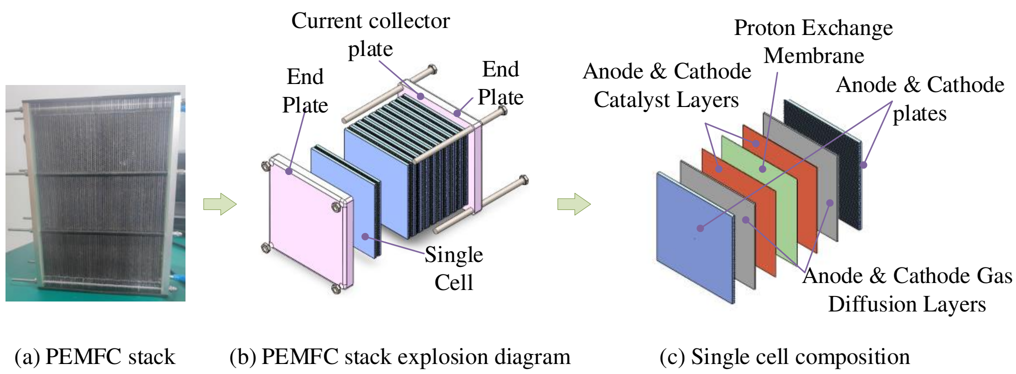

Figure 1. Components of PEMFC.

However, the lifetime of PEMFC is a significant limiting factor for their large-scale commercialization. There are multiple reasons that contribute to the limited lifetime of PEMFC [8]. Due to the complexity and multi-component properties of both the PEMFC stack and system, the challenges related to poor durability and aging failures in fuel cells have multifaceted origins. The factors that cause PEMFC aging can be categorized into two groups: common failures of auxiliary components within the PEMFC system [9] and degradation factors affecting the seven components of the PEMFC stack [10].

Once the fuel cell occurs aging phenomena, a series of internal faults in the stack are caused. For example, the catalyst activity is decreased, as well as the ion transport rates. As a result, the energy conversion efficiency and power output of the fuel cell are degraded. In such circumstances, the lifespan of PEMFC is markedly decreased. Moreover, the aging of the fuel cell system precipitates unpredictable failures, imparting a deleterious impact on system reliability and engendering hazards to critical applications like transportation and standby power provision. Therefore, it is significant to proactively circumvent and mitigate fuel cell aging with the goal of sustaining optimal fuel cell output performance and averting potential safety perils stemming from abrupt system failures.

In order to solve the fuel cell aging issue, the optimization of control strategies of each subsystem can effectively deal with the corresponding aging problem. This optimization allows the PEMFC stack to operate under optimal conditions. As a result, key components such as the proton exchange membrane, catalyst layers, and gas diffusion layers can avoid the aging mechanism occurring. Thus, the lifespan of the PEMFC is prolonged. On the other hand, it is also necessary to regularly check hardware faults such as air compressor surges, air filter blocks, and water pump failures. In addition, the regular maintenance and parameter updating of the software is also important work. Only in this case can the calibration parameters of the system adapt to the degradation curve of the PEMFC to meet the load demand and set the most reasonable output power. Section 2.3 gives a brief literature review of these solutions.

Prognostic and health management (PHM) can predict aging failure and facilitate informed decision making [11]. Figure 2 shows the whole process of PHM. As a crucial component of PHM, prognostic enables the early prediction of fuel cell aging trends, mitigating the risk of sudden failures. The prediction methods are primarily categorized into model-based and data-driven approaches [12].

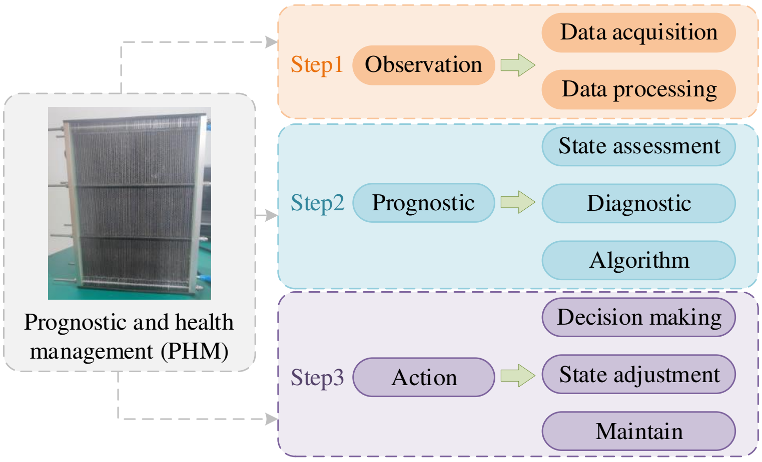

Figure 2. The whole process of prognostic and health management (PHM).

To quantitatively assess the extent of fuel cell degradation resulting from the aforementioned factors, researchers have extensively explored degradation indexes (DIs) [13]. Serving as a metric for fuel cell health, an appropriate DI provides an accurate reflection of the aging condition, enabling reliable aging prediction and mitigating sudden failure risks. The DI of PEMFC can be categorized into three groups: measurement data-based DI, major component-based DI, and hybrid characteristic-based DI.

Furthermore, in order to quantitatively analyze the accuracy of prediction methods, it is important to establish appropriate accuracy evaluation criteria in addition to selecting suitable DI [14].

This paper provides a comprehensive review and investigation of fuel cell aging, addressing the following distinct sub-problems: aging factors and mechanisms, mitigation strategies for various aging causes, and aging prediction research, along with aging index and accuracy evaluation criteria for individual components. The main contributions of this work are as follows:

(1) The aging factors and mechanisms of fuel cells are thoroughly presented, encompassing both system and stack levels. This comprehensive review exposes the underlying causes of performance degradation in fuel cells. It enables the implementation of appropriate measures by researchers to mitigate or prevent aging, ultimately enhancing the RUL, thus reducing maintenance costs and maximizing performance potential.

(2) Targeted mitigation strategies and aging prediction methods based on different aging factors and mechanisms are proposed. These strategies and methods represent the

most advanced solutions and advanced prognostic techniques to effectively solve the aging problem of fuel cells.

(3) An investigation into the correlation between the degradation index and various aging factors is conducted. This investigation forms the basis for the quantitative assessment of fuel cell aging. The research defines the appropriate degradation index for the various aging factors, providing guidance for the quantitative evaluation of aging predictions.

The rest of this paper is organized as follows. Section 2 provides a detailed description of the aging factors in fuel cells. Section 3 comprehensively classifies the aging prediction methods. In Section 4, the selection basis for the degradation index and accuracy evaluation criteria are outlined for different prediction methods. Section 5 discusses the challenges and future prospects of fuel cell aging prediction methods.

2. Aging Factors and Lifespan Prolongation Strategy for PEMFC

The variability of the aging failure factor arises due to the multi-component property of both the PEMFC stack and system. The causes of fuel cell aging failure can be categorized into two groups: the PEMFC system layer and the stack layer. In this section, the aging factors of the fuel cell system and stack level are first introduced, followed by the solving measures and lifespan prolongation strategy (LPS) for the PEMFC.

2.1. PEMFC System Level

Figure 3 shows the aging factors in the PEMFC system. It can be seen from Figure 3 that the failure of auxiliary components encompasses malfunctions in various subsystems within the fuel cell system: the air subsystem, hydrogen subsystem, hydrothermal management subsystem, and electronic control subsystem.

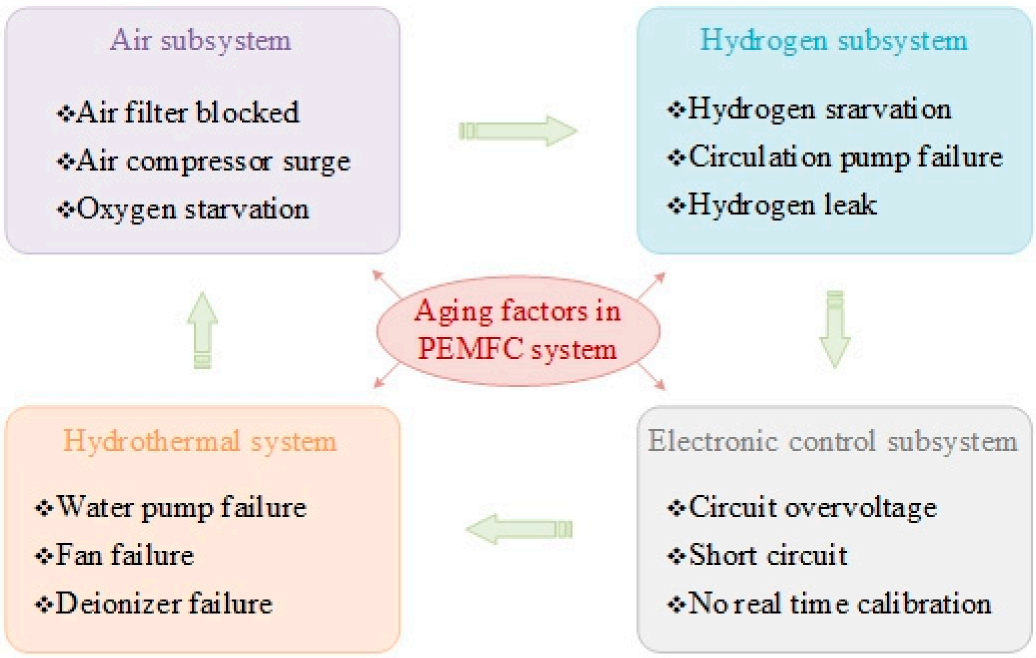

Figure 3. Aging factors in PEMFC system.

2.1.1. Air Subsystem Failure

The primary role of the air subsystem is to ensure the provision of adequate and purified air. Failure in critical components of the air subsystem can result in an excess of impurities and inadequate air supply, ultimately leading to aging-related failures. Typical failures and factors observed in the air subsystem are provided below.

Air filter clogging results in the contamination of the supplied gas, thereby causing the pollution of the proton exchange membrane and accelerating the decay of the remaining useful life (RUL) of the fuel cell [15]. On the other hand, air compressor surge introduces instability in the gas supply to the fuel cell, negatively impacting its output performance [16]. In addition, oxygen starvation is a frequently encountered malfunction, particularly in poor air, which renders it vulnerable to oxygen starvation instances [17]. Oxygen starvation gives rise to irregular current distribution [18]. Furthermore, this condition accelerates catalyst loss and contributes to the aging failure of the fuel cell [19].

Numerous fault diagnosis strategies have been extensively investigated to address the aforementioned common failures encountered in the air subsystem. To mitigate the harm caused by air compressor surges, Han et al. [20] developed a high-speed calculation control algorithm that ensures a sufficient air supply. Reference [21] devised a flow controller that prevents oxygen starvation by accurately managing the oxygen excess ratio. Liu et al. [22] developed a modified super-twisting sliding mode algorithm to estimate fault signals within the air subsystem under variable load conditions, effectively mitigating the problem of oxygen starvation within the air subsystem.

2.1.2. Hydrogen Subsystem Failure

The hydrogen subsystem determines the fuel supply of PEMFC. Within the hydrogen subsystem, failures are frequently encountered, which cause insufficient hydrogen supply and hydrogen pollution. These conditions have significant ramifications, leading to accelerated fuel cell aging.

Inadequate control over crucial components results in decreased power, primarily attributed to the occurrence of hydrogen starvation phenomena. This insufficiency in hydrogen supply accelerates carbon corrosion [23]. Consequently, the degradation of the carbon carrier in the catalytic layer ensues, accompanied by the dislodging and agglomeration of platinum particles. These combined effects lead to a reduction in the ECSA and irreversible degradation of the PEMFC stack lifetime [24]. Furthermore, apart from the detrimental effects of hydrogen starvation, hydrogen leakage poses a significant safety hazard, making it prone to causing severe accidents.

In addition, reference [25] introduced a two-step method and a mitigation approach to alleviate hydrogen starvation. In the context of fuel cell vehicles, Maeda et al. [26] proposed a methodology for diagnosing hydrogen leakage by utilizing sound analysis to identify the distinctive acoustic signatures associated with hydrogen leakage. Additionally, reference [27] developed a real-time gas monitoring method in the surroundings as a preventive measure against hydrogen leakage.

2.1.3. Hydrothermal Management Subsystem Failure

To ensure optimal performance of PEMFC, it is important to maintain the temperature of the PEMFC within a specified range [28]. Excessive operating temperatures can result in membrane drying, while excessively low temperatures can lead to increased activation loss and ohmic loss, resulting in decreased output performance. Furthermore, both excessively high and low temperatures can adversely affect the chemical reactions occurring within the proton exchange membranes [29].

To accurately regulate the operating temperature of fuel cells, reference [30] developed a dynamic model for the PEMFC hydrothermal management system. This approach establishes the relationship between temperature and polarization characteristics to simulate dynamic loads. In terms of fault diagnosis, reference [31] presented an online computational component based on a neural network. Zhao et al. [32] developed a thermoelectric control strategy for fuel cells. They also developed an experimental system for model validation to mitigate temperature fluctuations, thereby enhancing the operational stability of the fuel cells.

2.1.4. Electronic Control Subsystem Failure

The electronic control subsystem plays a vital role in maintaining a stable voltage output for fuel cells. A periodic calibration of the fuel cell is essential to avoid algorithm failures in the controller and ensure its optimal functioning.

Excessive voltage in the PEMFC stack is a prevalent failure occurrence. The overvoltage in the stack circuit accelerates carbon corrosion and catalyst degradation, which can result in the detachment and loss of platinum particles. This leads to a significant catalyst loss and significantly reduces the PEMFC stack lifetime [33].

Guilbert et al. [34] introduced a fault-tolerant control approach to mitigate membrane drying resulting from DC/DC converter failures. This method effectively prevents DC/DC converter failures. Mohammadi et al. [35] devised a fault diagnosis technique for DC/DC converters. Their approach integrates neural network (NN) modeling and numerical simulations, encompassing comprehensive considerations of water flooding and membrane drying conditions.

2.2. PEMFC Stack Level

The lifetime of a fuel cell is influenced not only by the malfunction of auxiliary components within the system but also by the degradation of critical components in the PEMFC stack. The failure occurs in the PEMFC stack, and the classification is shown in Figure 4.

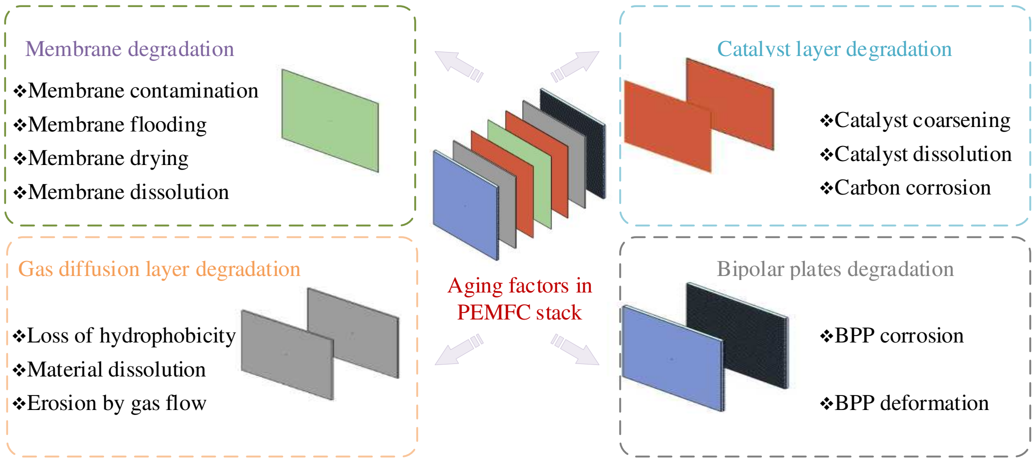

Figure 4. PEMFC stack failure classification.

2.2.1. Membrane Failure

The proton exchange membrane is subject to failure in three primary forms: membrane contamination, membrane flooding, membrane drying, and membrane dissolution.

Membrane contamination is one of the most severe faults in fuel cells. The primary causes of this fault include impurities present in reactants (hydrogen source and air source) as well as the degradation of fuel cell stack components such as the gas diffusion layer (GDL) and catalyst layer. Even small amounts of contaminants can lead to irreversible damage to the fuel cell stack, resulting in phenomena like catalyst poisoning and proton exchange membrane degradation, significantly impacting the lifespan [36]. As shown in Table 1, a detailed classification of membrane contamination is provided.

For the hydrogen source, common contaminants arise from residual carbon oxides, CH_4, and sulfides remaining from the hydrogen reformation process [37]. In the case of the air source, nitrogen gas, nitrogen oxides, and sulfides are the primary sources of contamination. Furthermore, when PEMFC systems operate in harsh environments, some toxic gases such as carbon monoxide and methane can also severely pollute the proton exchange membrane [38,39]. Beyond reactant sources, critical components degradation, like metal bipolar plates, sealing rings, and gas diffusion layers, can release metal ions such as Fe^{3+} and Cu^{2+}, inhibiting H^+ reactions. Simultaneously, these metal ions can promote the formation of metal oxides, accelerating membrane degradation and catalyst degradation [40].

Table 1. Detailed classification of membrane contamination.

Contamination Classification	Source	Contamination Component	Factor	Impact
Reaction gas	Hydrogen	Carbon oxides, CH_4, Sulfides	Residual contaminants from catalytic steam reforming of hydrogen	PEM and electrode degradation
	Air	Nitrogen gas, Nitrogen oxides, Sulfides, Toxic gas	Operation environment and air quality determine	
Key components	Membrane	Na^+	Membrane degradation	Diluting reactant concentrations accelerates aging
	BPP	Fe^{3+}, Cu^{2+}	Wear and corrosion	
	Sealing gasket	Si	Wear and corrosion	

The occurrence of membrane flooding is observed when the PEMFC operates at high current densities. A large load current forces a large amount of hydrogen to react chemically, thus producing more water. In this case, if the purging and drainage characteristics of the stack are poor, membrane flooding is easy to occur. Liquid accumulation blocks the gas flow channel, and the difficulty of a chemical reaction is increased. [41].

Membrane drying is another degradation phenomenon observed in the PEMFC stack. Membrane drying is easy to occur under high temperatures and small loads. Prolonged operation in a dry state can result in the expansion of the drying area within the membrane. Consequently, irreversible damage like membrane rupture occurs due to membrane drying [42]. Membrane dissolution is mainly caused by chemical factors. The presence of hydrogen peroxide during fuel cell operation and the subsequent breakdown of free radicals are the principal contributors to membrane dissolution [43].

2.2.2. Catalyst Layer Failure

Catalyst coarsening refers to the enlargement of Pt particles, resulting in a decrease in ECSA. This reduction in ECSA adversely affects the fuel cell output performance and accelerates its aging failure [44]. Borup et al. [45] observed from cyclic measurement experiments that the rate of catalyst coarsening exhibits a linear increase with temperature.

Catalyst dissolution, specifically the dissolution of platinum, has been identified as a significant degradation mechanism [46,47]. Reference [48] discovered factors such as potential cycling aggravate platinum dissolution. Reference [49] developed a model that effectively describes Pt dissolution and mobility within the membrane electrode assembly, aligning well with existing experimental data in the literature. Similarly, the literature proposed a model describing the movement of Pt catalysts during dissolution, offering valuable insights into the Pt dissolution process [50].

Carbon corrosion, as a crucial degradation mechanism, results in the depletion of the carbon support material in the presence of a Pt catalyst, leading to the detachment and loss of Pt particles [51]. Once carbon corrosion occurs, the gas transmission capacity will be weakened [52]. Furthermore, it promotes the material hydrophilicity, thereby triggering membrane flooding [53].

2.2.3. Gas Diffusion Layer Failure

Carbon corrosion can also manifest in the gas diffusion layer (GDL) alongside the catalyst layer, as the GDL primarily comprises carbon fibers. These carbon fibers are prone to structural deterioration due to corrosion [54]. In addition, insufficient shutdown purging leads to the formation of a hydrogen–air interface, thus accelerating the carbon corrosion process [55]. Consequently, the initiation and cessation of fuel cell operation can expedite the occurrence of carbon corrosion in the GDL.

GDL dissolution occurs when the fuel cell operates and the GDL comes into contact with a complex mixture comprising water, hydrogen, and air [56]. As a result, the diverse components present in the GDL interact, leading to the formation of hydroxides, oxides, and other substances that contribute to the dissolution of carbon material within the GDL [57].

Gas flow erosion is a phenomenon that occurs when high-pressure hydrogen and air are supplied to the fuel cell stack, resulting in the mechanical degradation of the GDL due to the impact of elevated gas pressure. Wu et al. [58] find that the loss of hydrophobicity significantly increases the gas transmission resistance at high current density, which is not conducive to the full progress of chemical reactions. Moreover, their research demonstrates that thermal or mechanical stress weakens the strength of the GDL and exacerbates material loss caused by gas cycling.

2.2.4. Bipolar Plates Failure

Bipolar plate corrosion results in corrosive substances accumulating in the MEA, triggering membrane aging failure. Moreover, the corrosion of BPP increases the ohmic resistance of the PEMFC, resulting in an elevated level of ohmic polarization loss. Consequently, the performance of the PEMFC stack is adversely affected by the corrosion of BPPs, leading to a decrease in overall system performance.

BPP deformation can occur when high mechanical pressure seals are utilized, resulting in the fracture and deformation of the BPP [59]. Furthermore, electrical characterizations reveal that the process conditions, design of carbon fabric, and incorporation of nanofillers exert a significant influence on the bulk conductivity of the BPP [60].

2.3. Lifespan Prolongation Strategy for PEMFC

2.3.1. Fault Handling Measures for System Level

In order to prevent PEMFC system failures and minimize the degradation of fuel cell lifespan due to the reasons mentioned above, more and more lifespan prolongation strategies have been proposed [61]. It is evident that regular hardware inspections and maintenance are essential at the system level. On this basis of robust hardware foundation, appropriate lifespan prolongation strategies and fault tolerance control (FTC) are implemented to prevent severe damage to the fuel cell lifespan, such as membrane flooding, membrane drying, electrode degradation, overvoltage, short circuits, and other detrimental phenomena.

To address the challenge of synchronously tracking and controlling the hydrogen subsystem under complex operating conditions, Zhu et al. [62] designed a nonlinear model predictive control method. This approach enhances the system's dynamic response speed while improving control system robustness as well as hydrogen utilization efficiency. For the air subsystem, Yang et al. [63] introduced an approach employing a linear parameter-varying observer and an air stoichiometry ratio estimator for fault diagnosis and management in the air subsystem. However, for certain special fault signals in the supply manifold, such as sinusoidal signals, it resulted in significant performance degradation, with a maximum power deviation of up to 4 kW. Different from the observer–estimator method, Wang et al. [64] devised an FTC method based on a dynamic triple-step approach. This method effectively maintains air pressure within an appropriate range even in the presence of air subsystem faults.

Li et al. [65] proposed an active FTC approach. This method integrates meta-learning with base learners to independently control hydrogen, air, and thermal management using base learners. In addition, for the hydrothermal management system, a meta-learner is adopted to identify different fault states, thereby achieving fault tolerance control under various operating conditions. Thus, the fuel cell lifespan is prolonged effectively. In order to solve the sensor failure in PEMFC systems, Oh et al. [66] compared feedforward and feedback-based FTC methods, thus avoiding real-time correction of sensor data. The experimental results indicate that passive FTC can effectively enhance response speed, while active FTC can significantly improve control precision. Both methods contribute to enhancing system efficiency and preventing sensor failure issues. Similarly, to address sensor faults in hydrothermal management, Yan et al. [67] demonstrated an active FTC strategy based on sliding mode control; the temperature control precision is ensured within ± 0.5 °C. In this way, the membrane drying phenomenon is avoided to some extent. Thus, the lifespan is prolonged. As a result, an FTC strategy based on performance recovery strategy and rapid fault diagnosis has been devised. This approach further investigates three issues: membrane flooding, membrane drying, and air starvation. The method has demonstrated effective performance recovery under all three fault conditions [68]. Furthermore, FTC strategies have also been employed to optimize the design of the hydrothermal management, ensuring the fuel cell operates within a reasonable temperature range and consequently prolonging the operational lifespan of fuel cell [69,70].

Furthermore, in addressing the potential overvoltage or short-circuit problems stemming from the instability in the output of the DC/DC converter, Ahmed et al. [71] developed a linear robust control method. The experimental results demonstrate that this approach ensures real-time performance and robustness of the DC/DC converter under stochastic conditions. Wang et al. [72] presented a coordination control strategy, and the fluctuation of bus voltage is alleviated. In this way, the stable operation state of PEMFC is ensured, as well as the lifespan.

2.3.2. Fault-Handling Measures for Stack Level

In addition to system-level fault-handling measures, improvements in the chemical composition and materials of key components in PEMFC stacks also contribute to mitigating the degradation of these components, thereby extending the overall lifespan of PEMFCs.

For proton exchange membranes, membrane contamination stands out as one of the most severe and common issues. To address membrane contamination problems, attention should be given to the purity of the reactant gases, preventing the presence of impurities such as sulfides and carbon monoxide. Furthermore, critical components within the fuel cell stack should be selected based on the ability to inhibit membrane contamination.

To investigate the physical mechanisms of membrane contamination in PEMFC, Zamel et al. [73] developed a transient mechanistic model based on membrane contamination. The model results indicate that the addition of oxygen to CO can mitigate the harmful effects of CO poisoning. In addition to mechanistic research at the physical level, studies in the fields of chemistry and materials science provide solutions to mitigate the damage caused by membrane contamination. Postole et al. [74] compared Pt adsorption properties for three different gases: CO, NH_3, and H_2. The experimental results demonstrate that, under identical conditions, Pt exhibits significantly stronger adsorption affinity for CO and the weakest affinity for hydrogen gas. This further presents the inhibitory effects of poisoning on the internal chemical reactions within PEMFC. Jackson et al. [75] proposed online and offline cleaning methods based on ozone (O_3). By introducing O_3 during the oxygen purging process, it effectively alleviates the damage caused by membrane contamination from CO and sulfide poisoning.

3. Aging Prediction Method

In order to address the issue of sudden aging failure in fuel cells from the aforementioned problems, this section provides a detailed description of the aging prediction methods for fuel cells. As shown in Figure 5, fuel cell aging prediction methods are classified into model-based and data-driven methods, and various approaches are further categorized and reviewed.

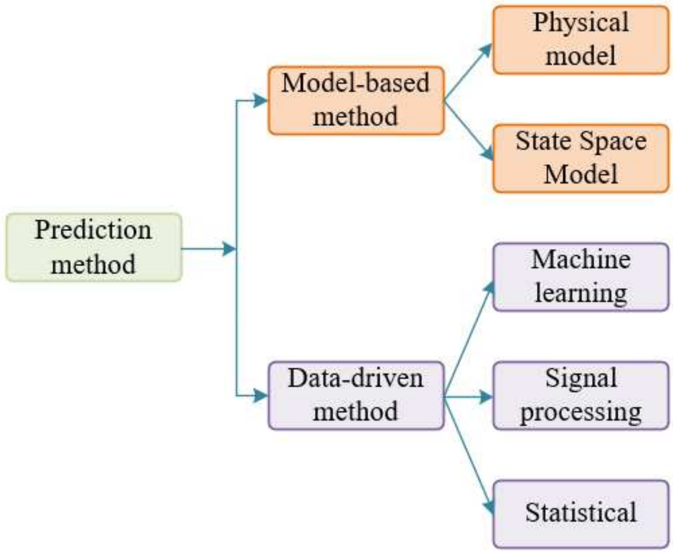

Figure 5. Fuel cell aging prediction methods classification.

3.1. Model-Based Method

This method is categorized into physical models and system state observer methods and predicts fuel cell aging by constructing mathematical representations of the physical and chemical processes [12].

3.1.1. Physical Model Method

This method is a modeling approach based on the internal mechanism of PEMFC. It involves establishing complex equations that capture the intricate relationships between various factors and predicting aging phenomena from a mechanistic standpoint. Robin et al. [76] developed a theoretical method to display the degradation of platinum surfaces, investigating operational dynamics using numerical simulations. Ao et al. [77] focused on microscopic particle-level phenomena to predict fuel cell performance. Reference [78] presented the ECSA model to provide the lifespan decrease trend. Hu et al. [79] considered ECSA and resistance degradation mechanisms to simulate actual operation mode and evaluate algorithm performance.

3.1.2. State Space Model Method

State space approach involves constructing a system state equation based on an empirical formula of the key parameter for PEMFC. Parameter identification techniques are then employed to update parameters of real-time running process. Zhang et al. [80] described the mathematical relationship between degradation rate and current. The model's aging coefficient is identified through direct fitting. Similarly, reference [81] employed fitting approach to acquire the degradation information and achieve fuel cell aging prediction using an empirical model. However, the direct fitting method exhibits low accuracy because of the dynamic nature of model parameters, which cannot be fully captured through direct fitting alone.

To enhance the fitting accuracy, adaptive updates are applied and optimized for the above methods. Reference [82] adopted extended Kalman filter (EKF) to prognostic time-varying parameters, with high prognostic accuracy. In addition, adaptive Kalman filtering was proposed to acquire the degradation phenomenon in reference [83]. However, the real-time performance cannot be ensured. To address this issue, unscented Kalman filter (UKF) was adopted to speed up the calculation [84]. Nevertheless, the accuracy decreased compared with former. Ao et al. [85] proposed a frequency domain Kalman filter (FDKF) to balance accuracy and real-time with different operation modes. Zhou et al. [86] presented a particle filter (PF) method unlike EKF. This approach incorporates typical aging coefficient and updating coefficient during the prediction stage to forecast the fuel cell stack voltage.

The results demonstrate the method's ability to effectively track the fuel cell aging process. Additionally, various advanced algorithms, such as UPF [87], APF [88], RPF [89], etc., have been implemented for prognostic approaches, exhibiting high performance.

3.2. Data-Driven Method

Data-driven approach constructs the input–output relationship using a large amount of data. This method can make predictions without knowing the internal mechanism. This method can be categorized into (1) machine learning methods, (2) signal processing methods, and (3) statistical methods [12].

3.2.1. Machine Learning Method

Machine learning excels at capturing key characteristics and is commonly employed in prediction tasks involving missing or complex models. Wang et al. [90] combined the Monte Carlo dropout method and deep neural network to predict the fuel cell health state. Based on wavelet transform, Chen et al. [91] developed an extreme learning machine with genetic algorithm optimization. This method performed much better than ELM. In addition, recurrent neural network (RNN) is often used in time series prediction in recent years because it considers real-time and past inputs [92].

Long short-term memory (LSTM), an improved method of recurrent neural networks (RNNs), is widely used in fuel cell aging prediction due to its ability to address the issues of gradient disappearance and explosion. Liu et al. [93] applied LSTM to predict remaining useful life (RUL), achieving higher accuracy compared to the backpropagation neural network (BPNN). To optimize the LSTM network structure, Ma et al. [94] proposed a grid long short-term memory (G-LSTM) method with superior prediction accuracy over single LSTM in experimental datasets. Similarly, references [95–100] also conducted in-depth research on the LSTM algorithm.

3.2.2. Signal Processing Method

Signal processing approach involves data splitting and filtering to remove noise and irrelevant information [101]. It serves as a valuable tool in achieving accurate predictions of fuel cell aging.

Reference [91] utilized GA to optimize the global parameters, taking into account the impact of various key conditions for PEMFC degradation. On this basis, wavelet transform with nonlinear autoregressive exogenous neural network is proposed. This method incorporates previous value into the present state, enhancing its predictive capabilities [102]. Discrete wavelet transform (DWT) with various prognostic algorithms is commonly adopted in fuel cell health management [103]. Hua et al. [104] introduced a method that combines DWT with echo state networks. The prediction accuracy of fusion after decomposition is greatly improved. Furthermore, the authors further enhanced the prediction accuracy by utilizing GA to optimize the key parameters of ESN [105].

3.2.3. Statistical Method

Statistical methods are established to research the uncertainty of entire series using random statistical models. Typically, there are the auto-regressive moving average model (ARMA), Gaussian process (GP), grey model (GM), and other prediction methods.

Detti et al. [106] applied the ARMA to research the degradation phenomenon. Although the general trend can be captured, it is difficult to capture the volatility characteristics. Zhou et al. [107] combined the time delay neural network to address the volatility component, thus enhancing the robustness of prognostic approach. Different ARMA, Zhu et al. [108] adopted GP to describe the degradation trend. Deng et al. [109] presented a double mathematical model to address this degradation prognostic issue. In addition, as a statistical method, the grey theory model is often applied to predict fuel cell aging [110,111].

The data-driven approach discards the construction of complex physical models. Once sufficient training set data are given. However, to ensure computational speed, a trade-off between prediction accuracy and computation time is inevitably required. Additionally, the selection of parameters for the "black box" is a primary condition for prognostic performance. Thus, optimization and parameter sensitivity analysis are crucial for determining appropriate model parameters. The summary of characteristics of aging prediction methods is shown in Table 1.

4. Degradation Index and Accuracy Evaluation Criteria

An accurate assessment of the overall health status of PEMFC and its individual components in real time can be achieved using an appropriate degradation index (DI). In addition, the accuracy of the prediction results can be quantitatively analyzed by adopting appropriate accuracy evaluation criteria. Figure 6 shows the degradation index and accuracy evaluation criteria. In this section, DI and accuracy evaluation criteria are summarized, and the prediction methods adapted to various indexes are briefly analyzed.

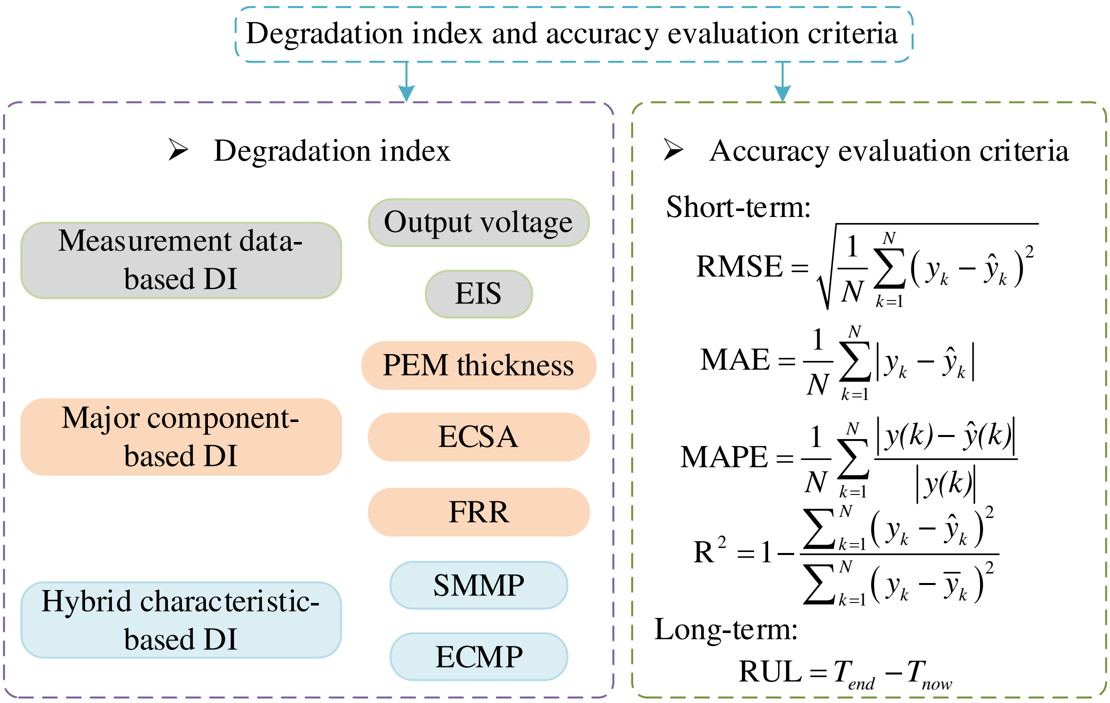

Figure 6. The degradation index and accuracy evaluation criteria.

The utilization of an appropriate DI is crucial for the development of an effective PHM for PEMFC. Presently, the DI for PEMFC can be categorized into three types: measurement data-based DI, component-based DI, and hybrid characteristic-based DI.

4.1. Degradation Index

The measurement data-based DI encompasses two main components: sensor data DI and auxiliary device measurement DI. The sensor data-based DI primarily relies on output voltage and power measurements, while the auxiliary device measurement-based DI predominantly utilizes electrochemical impedance spectroscopy (EIS).

4.1.1. Measurement Data-Based DI

The output voltage of PEMFC can be readily obtained and is known to undergo considerable degradation during fuel cell operation. Output voltage and output power are commonly employed as degradation indexes based on measured data for PEMFC. In fact, the IEEE PHM Data Challenge [112] employs the percentage decrease in output power as a failure index. Various aging prediction studies have also utilized output voltage or output power as degradation indices. Notably, the most frequently referenced datasets for aging analysis are the voltage measurements collected under static and dynamic current conditions.

In addition to output voltage and output power, the characteristics of polarization curves and EIS can also serve as DI to assess the health status of fuel cells, provided that adequate experimental devices are available. Bezmalinovic et al. [113] established a correlation between polarization curves and the occurrence of aging failure phenomena in fuel cells. This highlights the potential of polarization curves and EIS as alternative DIs for evaluating the condition of fuel cells.

Wang et al. [114] utilized polarization curves and EIS to determine the aging condition of fuel cells. They extracted polarization curves and EIS data under various operating conditions and employed the marginal distance method to evaluate the aging state based on the geometric properties of EIS. Similarly, Li et al. [115] proposed an approach to estimate the aging state of PEMFCs by utilizing characteristic metrics related to aging extracted from polarization curves and EIS. Additionally, Vianna et al. [116] assessed and predicted the aging state of PEMFCs using real and imaginary impedance components at different frequencies as DI. These studies demonstrate the potential of polarization curves and EIS in quantifying and predicting the aging status of fuel cells.

The deterioration of pivotal components within the PEMFC plays a pivotal role in hastening the onset of aging failure. Therefore, it becomes imperative to establish distinct degradation indices for each component, as it serves to enable a direct and comprehensive depiction and analysis of the aging state of individual components. By delineating these degradation indices, researchers can effectively characterize and evaluate the deteriorative condition of each component, thus enhancing the understanding and management of PEMFC aging phenomena.

4.1.2. Stack Component-Based DI

The degradation assessment of proton exchange membranes often relies on a set of commonly employed degradation indices, namely PEM thickness, fluoride release rate (FRR), and ECSA. To address the estimation of PEM aging, Karpenko et al. [117] proposed an empirical model that captures the progressive decay of PEM thickness and electrical conductivity throughout the aging process. This model takes into account various operational parameters, including temperature, relative humidity, hydrogen pressure, and initial PEM thickness, which collectively influence the extent of PEM degradation. Within the scope of this study, PEM thickness and oxygen permeability are adopted as DI, effectively characterizing the degree of PEM aging. By incorporating these parameters, researchers can gain valuable insights into the aging progression of PEMs and further enhance their understanding of PEMFC performance and durability.

Inaba et al. [118] extensively examined the mechanisms underlying the aging of PEM and performed durability tests to analyze the evolution of hydrogen permeability and fluoride release rate (FRR) throughout the aging process. Likewise, Liu et al. [119] conducted long-term aging tests on PEM fuel cells under cyclic current load conditions, utilizing hydrogen permeation rate and FRR as DI for the PEM. On this basis, Xu et al. [120] further investigated the influence of relative humidity on the performance and aging characteristics of PEMFC stacks. These studies collectively contribute to a comprehensive understanding of the aging mechanisms and performance degradation of PEM, facilitating the development of effective strategies for prolonging the durability and reliability of PEMFC systems.

Burlatsky et al. [121] considered the mechanical properties of membranes and the magnitude of relative humidity cycling as additional indicators to predict the lifetime of PEM. Their study emphasized the importance of incorporating mechanical factors and environmental cycling in assessing PEM durability. Furthermore, Macauley et al. [122] developed an empirical model to estimate the lifetime of PEMs, specifically in the context of fuel cell vehicles, taking into account various factors, such as operating conditions, temperature, and humidity. These research efforts contribute to a more comprehensive understanding of PEM lifetime prediction and aid in the development of strategies to enhance the longevity of PEM-based fuel cell systems.

4.1.3. Hybrid Characteristic-Based DI

To investigate the aging characteristics of fuel cells, researchers have integrated aging data with aging parameters using empirical modeling to assess the aging state of PEMFC through a hybrid degradation index. Mao et al. [123] developed parameters derived from the voltage decay semi-mechanical model parameter (SMMP) as a degradation index to capture the aging trend of PEMFC. These parameters include exchange current density, internal current density, mass transfer loss, and membrane resistance parameters. Zhou et al. [86] utilized the ohmic resistance coefficient, gas diffusion coefficient, and exchange current density coefficient from the voltage decay semi-mechanical model to characterize the aging state of the PEM, electrode, and GDL in PEMFC, respectively. Chen et al. [87] developed a hybrid degradation index that incorporates internal resistance, output voltage, and power to characterize the aging of PEMFC under static load conditions. Kim et al. [124] proposed an equivalent circuit model parameter (ECMP) to fit EIS measurements of PEMFC at various aging stages. The ECM utilizes four resistance parameters to characterize the aging phenomenon in PEMFC. By integrating aging data with specific degradation indices, these studies enable a comprehensive understanding of the aging behavior of PEMFC and contribute to the development of accurate aging prediction models. The summary of characteristics of aging prediction methods is shown in Table 2.

Table 2. Summary of characteristics of aging prediction methods.

Approach Category	Subclass	References	Degradation Index	Characteristic
Model-based methods	Physical model	[76–79]	PEM thickness, ECSA, FRR, and other component properties.	Theoretical description of the actual physical degradation phenomena; modeling process is complex.
	State space model method	[80–89]	Output voltage, output power, SMMP, and ECMP.	Simple and easy implementation.
Data-driven methods	Machine learning method	[90–100]	Mainly output voltage, output power, and EIS.	Sensitive to the data quality and quantity.
	Signal processing method	[91,101–105]	Mainly output voltage, output power, and EIS.	Suitable for non-stationary time series.
	Statistical method	[106–111]	Mainly output voltage, output power, and EIS.	Good generalization capability; Stationary series.

It can be seen from Table 2 that the prediction method based on a physical model, PEM thickness, ECSA, and FRR is usually used as degradation index. The output voltage and empirical model parameters are usually used as the degradation index. For data-driven methods, output voltage, output power, and electrochemical impedance spectrum are mainly adopted as degradation index.

4.2. Accuracy Evaluation Criteria

To assess the prediction performance, four commonly used standards are employed, namely root mean square error (RMSE), mean absolute error (MAE), mean absolute percentage error (MAPE), and coefficient of determination (R^2) [125].

RMSE measures the average difference between predicted and actual values, providing an overall assessment of prediction accuracy. MAE calculates the average absolute difference between predicted and actual values, indicating the magnitude of prediction errors. MAPE represents the average percentage difference between predicted and actual values, allowing for the evaluation of relative prediction errors. R^2 quantifies the proportion of the variance in the observed data that can be explained by the prediction model, indicating the goodness of fit. The calculation process of these accuracy evaluation criteria is as follows:

$$\mathrm{RMSE} = \sqrt{\frac{1}{N}\sum_{k=1}^{N}(y_k - \hat{y}_k)^2} \tag{1}$$

$$\mathrm{MAE} = \frac{1}{N}\sum_{k=1}^{N}|y_k - \hat{y}_k| \tag{2}$$

$$\mathrm{MAPE} = \frac{1}{N}\sum_{k=1}^{N}\frac{|y(k) - \hat{y}(k)|}{|y(k)|} \tag{3}$$

$$\mathrm{R}^2 = 1 - \frac{\sum_{k=1}^{N}(y_k - \hat{y}_k)^2}{\sum_{k=1}^{N}(y_k - \overline{y}_k)^2} \tag{4}$$

$$\mathrm{RUL} = T_{end} - T_{now} \tag{5}$$

where y_k is the voltage value, and $\hat{y}_k$ is the prognostic value. N is the amount of data. T_{end} is the operating time at the end of the life, and T_{now} is the time that the fuel cell has been in operation.

By utilizing these evaluation criteria, researchers can comprehensively assess the prediction performance of different models and determine their effectiveness in capturing the desired outcomes. It should be noted that for short-term aging prediction, such as predictions within 10 h, the prediction errors are typically set at the minute level, requiring higher accuracy. This necessitates the use of accuracy evaluation criteria with higher resolution. Therefore, evaluation criteria, such as RMSE, MAPE, MAE, and R^2, which offer high-resolution precision assessment, are commonly applied in short-term prognostic scenarios. Conversely, for long-term aging conditions, the prediction errors are typically set at the hour level. Therefore, RUL is commonly adopted as the accuracy evaluation criterion for long-term aging prediction.

5. Challenges and Future Prospects

Although great efforts have been made in developing aging prognostic techniques, there are several major challenges and corresponding future work in this field:

The standardization of PHM mechanisms: Due to the complexity and variability of fuel cell systems, establishing accurate predictive models and health management strategies remains challenging. Furthermore, a deeper understanding of aging mechanisms and failure modes is needed to enhance the accuracy of prediction and diagnosis. Additionally, the lack of standardized evaluation methods and metrics also limits the development of fuel cell predictive and health management technologies. Therefore, further efforts are required to address these issues and improve the reliability and performance of fuel cell systems.

Whole-lifecycle establishment: The whole-lifecycle establishment entails the comprehensive monitoring and assessment of fuel cell systems during the stages of design, manufacturing, operation, and maintenance. The real-time monitoring and evaluation of system performance, combined with advanced fault diagnosis and prediction techniques, facilitate the early detection of issues and the implementation of maintenance measures. Regular inspection and maintenance are also crucial for maintaining the health of fuel cell systems. A whole-lifecycle approach will provide a more accurate system condition assessment, enhance system reliability and performance, and support the development of effective maintenance strategies.

Research on the health management strategy: There is little research on health management strategy. On the basis of prediction, an advanced health management strategy is also very important. By incorporating advanced sensing technologies and data analysis methods, the real-time monitoring of fuel cell systems can be achieved, enabling timely fault detection and maintenance decision making. In addition, the integration of cloud computing and internet of things technologies allows for the centralized monitoring and management of large-scale fuel cell systems, thereby enhancing the remaining useful life of the fuel cells.

6. Conclusions

In order to address the aging failure issues caused by multiple fault factors in fuel cells, this paper comprehensively reviews and investigates the aging problem of fuel cells, including the aging factors and mechanisms, various measures to counteract aging causes, aging prediction research, degradation index of various components, and accuracy evaluation criteria for aging prediction. On this basis, the relationship between various aging prediction methods, degradation index, and accuracy evaluation criteria is established. Based on an extensive literature review, it is evident that aging failure issues caused by specific factors are often addressed with targeted approaches corresponding to specific degradation indexes. This review results indicate that physical model prediction methods typically employ degradation indexes such as PEM thickness, ECSA, and FRR. State-space model prediction methods typically utilize degradation indexes such as output voltage, SMMP, and ECMP. Data-driven methods generally adopt degradation indexes such as output voltage, power, and electrochemical impedance spectra. To further analyze the accuracy of aging prediction methods, short-term aging prediction usually adopts accuracy evaluation criteria such as RMSE and MAPE, while long-term aging prediction typically uses Remaining Useful Life (RUL) as an accuracy evaluation criterion. Finally, the challenges and future directions in aging prediction are proposed to guide the prolongation of fuel cell lifespans.

In future work, it will be interested in continuing to study the multi-objective lifespan prolongation strategy and aging prediction method based on multi-aging index fusion, it is of great significance for improving the service life and commercialization of fuel cells.

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