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Review

Comprehensive Analysis of Solid Oxide Fuel Cell Performance Degradation Mechanism, Prediction, and Optimization Studies

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Abstract: Solid oxide fuel cell (SOFC) performance degradation analysis and optimization studies are important prerequisites for its commercialization. Reviewing and summarizing SOFC performance degradation studies can help researchers identify research gaps and increase investment in weak areas. In this study, to help researchers purposely improve system performance, degradation mechanism analysis, degradation performance prediction, and degradation performance optimization studies are sorted out. In the review, it is found that the degradation mechanism analysis studies can help to improve the system structure. Degradation mechanism analysis studies can be performed at the stack level and system level, respectively. Degradation performance prediction can help to take measures to mitigate degradation in advance. The main tools of prediction study can be divided into model-based, data-based, electrochemical impedance spectroscopy-based, and image-based approaches. Degradation performance optimization can improve the system performance based on degradation mechanism analysis and performance prediction results. The optimization study focuses on two aspects of constitutive improvement and health controller design. However, the existing research is not yet complete. In-depth studies on performance degradation are still needed to achieve further SOFC commercialization. This paper summarizes mainstream research methods, as well as deficiencies that can provide partial theoretical guidance for SOFC performance enhancement.

Keywords: solid oxide fuel cells; degradation mechanism analysis; degradation performance prediction; degradation performance optimization

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1. Introduction

A solid oxide fuel cell (SOFC) is an all-solid-state power generation device that can directly convert chemical energy into electrical energy. It has the advantages of low pollution, high efficiency, wide fuel applicability, high cogeneration capacity, and relatively low cost [1]. However, the reliability of SOFC systems is difficult to guarantee due to their complex structure. Performance degradation may occur on various subsystems and components. Moreover, many failures, such as air leakage and impedance increase, evolve gradually from smaller defects [2]. In this case, how to ensure efficient, stable, and long-life operation of the system has become an urgent problem in the current SOFC commercialization process [3].

Currently, the SOFC system degradation mechanism has been summarized in several literatures. Løye et al. [4] reviewed SOFC degradation and lifetime studies based on more than 50 parameters and 1 million hours of cumulative test time from 150 publications. The authors concluded that the degradation rate has been steadily decreasing in recent years and is rapidly approaching official targets. Tu et al. [5] summarized in detail the current status of SOFC development progress and the aging mechanism of the stack. They pointed out that the SOFC aging mechanism depends heavily on the material, microstructure, and operating conditions. Therefore, developing new materials, optimizing the microstructure, and reducing the operating temperature are beneficial for SOFC durability enhancement. Sreedhar et al. [6] reviewed the degradation mechanisms in the anode, cathode, electrolyte, interconnect, and sealant of the SOFC stack. Moreover, they summarized the recent progress of research on measures to mitigate degradation. However, they only summarized the degradation mechanisms and mitigation measures in the SOFC stack and did not review the degradation in the overall system. Anwar et al. [7] reviewed the studies using X-ray photoelectron spectroscopy (XPS) to understand the SOFC cathode degradation mechanism. Furthermore, they discussed the strategies to improve the structural stability and electrochemical performance of the cathode. However, the study has a relatively small scope of application and can only provide theoretical guidance for SOFC cathode degradation. Subotic et al. [8] reviewed the degradation studies of SOFC in terms of health condition diagnosis, failure modes, degradation mitigation, and performance regeneration. They provide guidelines for dealing with SOFC degradation issues running in real-world environments. However, their review is not comprehensive enough. In terms of degradation mechanisms, only degradation in the stack anode, cathode, and electrolyte are reviewed. In terms of degradation mitigation and performance regeneration, there is no review of controller design studies.

To the authors' knowledge, the relevant literature available so far has only analyzed the materialistic degradation mechanism of SOFC stack. Although understanding the SOFC degradation mechanism can help improve the system's structure, it cannot quickly improve the performance of SOFC systems [9]. Predictive studies of degradation are equally important if one wishes to avoid the occurrence of degradation or to optimize system performance quickly. Predicting the SOFC system degradation trend can help to take measures to mitigate the degradation in advance [10]. This can avoid unplanned downtime and help improve system life. In addition, once the degradation mechanism is obtained and the degradation performance is predicted, an effective performance optimization strategy is a key step to extend the system's lifetime [11]. Improving the material structure of the SOFC system, or designing the corresponding health controller, can provide performance improvements to the degraded system.

The degradation mechanism, degradation performance prediction, and performance optimization strategy design of SOFC systems have been investigated in numerous studies. However, the current research work is still scattered and has not been systematically organized. In this paper, previous studies on degradation mechanism analysis, degradation performance prediction, and performance optimization schemes for SOFC systems are carefully reviewed and summarized. First, the work on the degradation mechanism analysis of SOFC is summarized at the stack level and system level, respectively. Then, model-based, data-based, electrochemical impedance spectroscopy (EIS)-based, and image-based degradation prediction methods are analyzed, respectively. Finally, the performance optimization strategies of SOFC systems are reviewed in terms of material structure improvement and health controller design. This paper also presents the current SOFC system performance degradation research shortage and analyzes future research directions. The structure of this paper is shown in Figure 1.

The novelty of this paper is as follows.

(1) This paper reviews the degradation mechanism, degradation performance prediction, and degradation performance optimization studies at the same time. This paper provides a comprehensive summary of the entire process of degradation from being detected to being optimized. This can provide a theoretical basis for the SOFC system life extension.

(2) The degradation mechanism analysis studies are reviewed at the stack level and the system level, respectively. In addition to the stack, other SOFC system components degradation can also affect SOFC performance. Therefore, the system-level degradation

mechanism analysis is equally important. However, there are relatively few reviews of system-level degradation.

(3) This paper summarizes the advantages, disadvantages, and applicability of model-based, data-based, EIS-based, and image-based degradation prediction methods. This can help researchers to quickly find the most suitable prediction means.

(4) The degradation performance optimization research is reviewed in terms of both material structure improvement and health controller design. Material structure improvement can improve degradation from the root cause, but the optimization period is long. Controller design can optimize performance without system downtime. Therefore, the design of the controller is also worth studying.

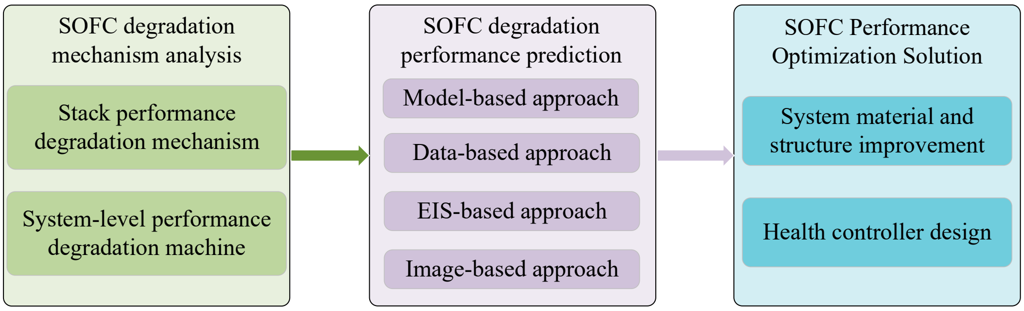


Figure 1. Solid oxide fuel cell (SOFC) performance degradation study process.

2. SOFC Degradation Mechanism Analysis

The lack of long-term stability is one of the main problems in commercializing SOFC technology. The SOFC system consists of a stack and the corresponding balance of plant (BOP) [12]. The stack is the core component of the SOFC system. Its main function is to convert the chemical energy in oxygen and fuel directly into electrical energy through electrochemical reactions. The BOP subsystem consists of reformer, heat exchanger, exhaust gas combustion chamber, desulfurizer, dehumidifier, air compressor, air storage tank, electric lighter, cooling water tank, and monitoring system, as shown in Figure 2. They have functions such as gas transport, heat exchange, and exhaust gas treatment. The current research on SOFC degradation mechanisms can be mainly divided into stack performance degradation studies and system-level performance degradation studies.

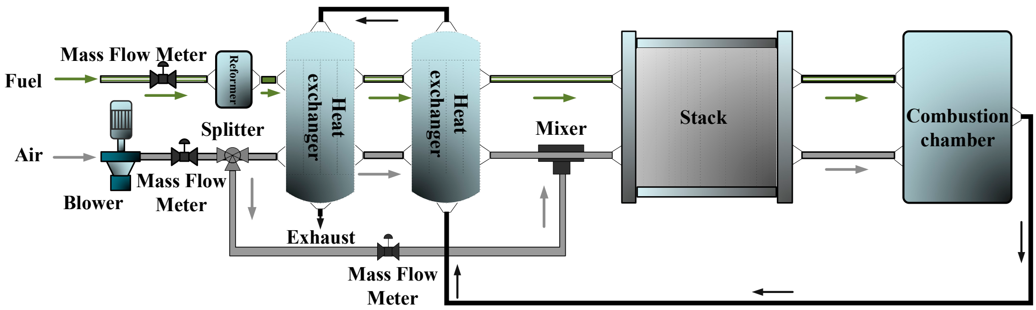


Figure 2. SOFC system structure.

2.1. Stack Performance Degradation

SOFC stack is assembled from multiple single cells. The voltage generated by a single cell is very low, typically less than 1 V (operating voltage is about 0.6 to 0.9 V), and its power density is less than 2 W/cm^2. As a result, the output voltage and output power of a single cell are very limited. To obtain the power output to meet the demand, the cells are connected in series or parallel to form SOFC stack. The single cell consists of electrolyte, cathode, and anode. The single cell is connected by the interconnect to form the stack. Moreover, since gas tends to leak from the edge of the electrode, the edge of the electrode is sealed by sealing material. Therefore, the components of the stack mainly include the electrolyte, cathode, anode, interconnect, and sealing material, as shown in Figure 3. In high temperature operating environments, the components of the SOFC stack are subject to performance degradation both due to material changes and chemical reactions with each other.

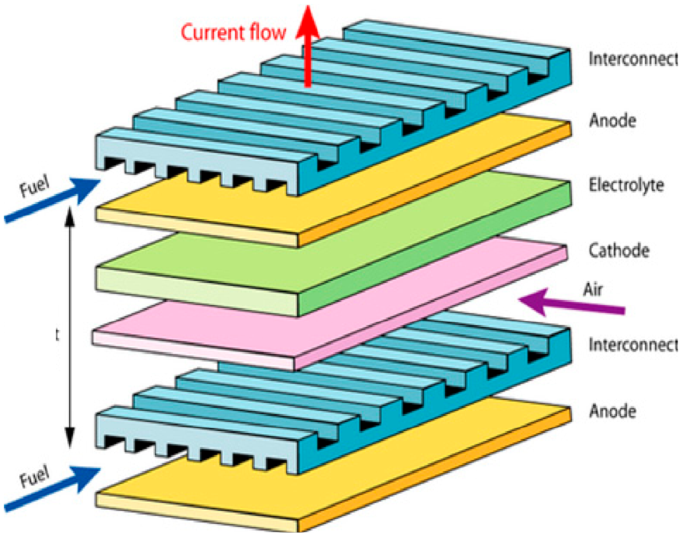


Figure 3. SOFC stack composition structure.

2.1.1. Cathode Performance Degradation

In SOFC, the cathode mainly plays the role of transporting air, catalyzing electrode reactions, and conducting current. In the cathode-supported SOFC, the cathode also takes on the role of supporting the entire SOFC cell structure. Therefore, the cathode material needs to have a strong electrochemical catalytic ability for oxygen reduction to reduce the activation loss of the cell and improve cell performance. The cathode material also needs to have a high electrical conductivity to reduce ohmic losses. Meanwhile, due to the high operating temperature of SOFC, the cathode material needs to have good chemical stability in the oxidizing atmosphere. At the same time, it cannot react chemically with adjacent materials.

The cathode of SOFC is usually composed of chalcogenide oxides. The most common cathode material in medium and high-temperature SOFCs is $La_{1-x}Sr_xMnO_3$ (LSM) [13]. LSM combined with electrolyte material yttrium oxide stabilized zirconium oxide (YSZ) to form a composite electrode that has good electrical conductivity and ion conductivity. Meanwhile, the expansion coefficient of LSM is similar to that of YSZ electrolyte, so they have excellent thermal matching.

However, a chemical reaction may occur between the LSM cathode and the YSZ electrolyte to form $La_2Zr_2O_7$ during high-temperature sintering. It is an insulating oxide that can cover the active sites of electrochemical reactions. Moreover, it can increase the system ohmic resistance, which reduces the electrochemical performance of SOFC. Yokokawa et al. [14] analyzed the reaction between the lanthanide manganite cathode and yttrium-stabilized zirconia electrolyte by the chemical potential diagram. They concluded that the change in manganese valence causes the formation of $La_2Zr_2O_7$, which can lead to a drastic change in interfacial chemistry.

Cr poisoning is another cause of SOFC cathode degradation. Cr poisoning occurs because Cr_2O_3 on the surface of the interconnect produces gaseous oxides (e.g., CrO_3, $Cr(OH)_2O_2$, etc.) through surface reactions. These oxides enter the interior of the cathode by volatilization and are chemically reacted on the surface of the cathode particles to re-form Cr_2O_3. Cr_2O_3 covers the surface of cathode particles, which can cover the active reaction sites of the cathode. Moreover, it will hinder the adsorption and diffusion of oxygen on the cathode surface, which decreases the performance of the cathode. Fang et al. [15] test the performance degradation of a planar SOFC stack for more than 100,000 h. The results showed that the degradation in the first 40,000 h was mainly due to Cr poisoning in the cathode and oxidation of the metal interconnects. Chen et al. [16] studied Cr deposition and poisoning in $(La_{0.8}Sr_{0.2})_{0.95}(Mn_{1-x}Co_x)O_{3\pm\delta}$ (LSMC, $0.0 \leq x \leq 1.0$) cathodes. The results show that as the Co content in LSMC increases from 0.0 to 1.0, the deposition of chromium on the electrolyte surface decreases, while the deposition on the electrode surface increases. On the other hand, the chromium poisoning effect is most pronounced at the LSMC cathode with x = 0.4, as shown in Figure 4. The occurrence of maximum chromium poisoning is most likely due to the combined poisoning effect of chromium deposits on the electrode surface and the electrolyte surface.

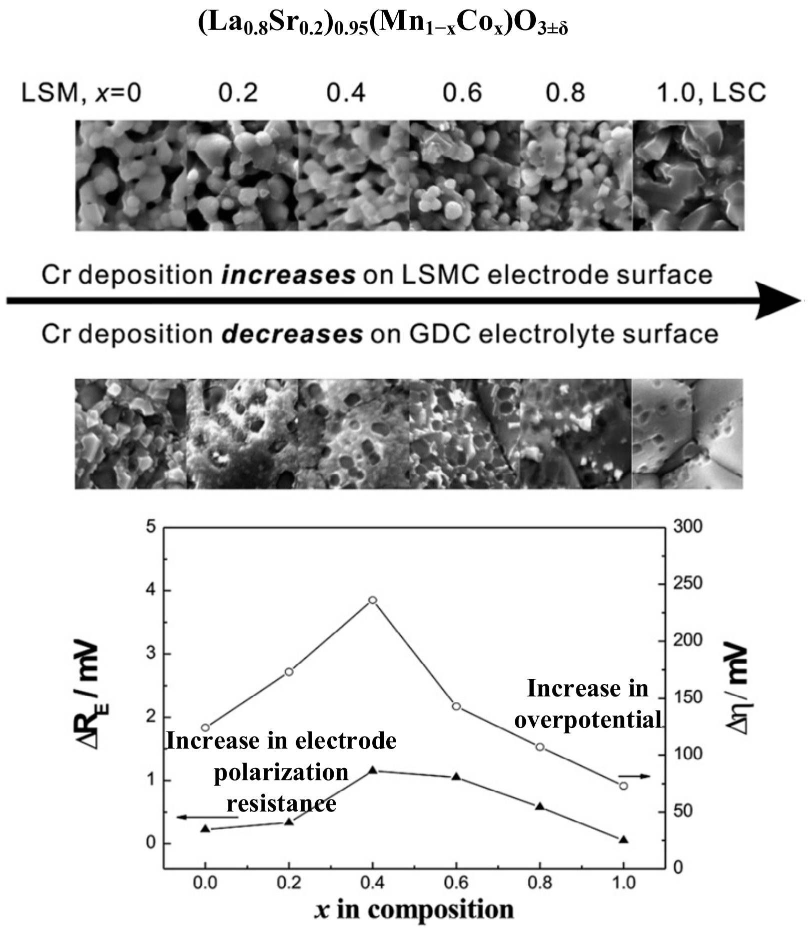


Figure 4. Effect of Co content on Cr poisoning. Adapted with permission from Ref. [16].

In addition, gaseous impurities in the air (such as SO_2), can also cause cathode performance degradation. These impurities can react with the cathode material to produce new reactants. These reactants can block the activation sites of electrochemical reactions. They may also corrode the cathode structure, which can change the cathode's mechanical properties and lead to cathode performance degradation. Riesgraf et al. [17] developed a kinetic model to predict the effect of sulfur poisoning on SOFC performance degradation. The model proved its validity in $H_2/H_2O/H_2S$ and $CH_4/H_2/H_2O/H_2S$ fuel mixtures, respectively. It verifies the effect of adsorbed sulfur on charge transfer processes and on complex methane reforming chemistry. The results show that the sulfur surface coverage increases with increasing current density. Moreover, the addition of small amounts of H_2S to methane can significantly reduce the fuel conversion rate and correspondingly reduce carbon formation.

2.1.2. Anode Performance Degradation

The anode mainly plays the role of transporting fuel gas, catalyzing electrode reactions, and conducting current. In anode-supported SOFC, the anode also plays the role of supporting the entire SOFC cell structure. The most commonly used SOFC anode material is nickel-doped YSZ composite electrode (Ni-YSZ) [18]. As a metallic material, Ni has a very high electronic conductivity and thermal conductivity. Meanwhile, Ni acts as a catalyst for the oxidation of fuels such as hydrogen and carbon monoxide. It can perform direct internal reforming of hydrocarbons such as methane. In addition, compared to precious metals such as Pt, Ni is relatively inexpensive and can reduce the cost of anodes. The thermal expansion coefficient of the Ni-YSZ composite electrode is consistent with the YSZ electrolyte and can meet the requirements in terms of thermal stress. Ni particle coarsening, Ni particle redox, carbon deposition, and gas impurity poisoning are the main causes of SOFC anode performance degradation.

Ni particle coarsening is the main reason for the SOFC anode performance degradation. Ni particle coarsening is caused by surface diffusion between interconnected Ni particles. This phenomenon is due to the higher interfacial energy causing Ni to diffuse on the YSZ surface. Then, under the action of surface diffusion, multiple interconnected Ni particles fuse to form one particle, increasing the average radius of the Ni particles. With the coarsening of Ni grains, the triple-phase line length and conductivity of the anode gradually decrease, leading to the degradation of SOFC performance. Khan et al. [19] prepared a double-cell anode-supported flat-tube SOFC stack and designed an accelerated lifetime test experiment to investigate the causes of SOFC system performance degradation. The results show that coarsening of Ni particles is the main degradation mechanism of the anode of the stack. Moreover, in the first 200 h, the stack exhibits a rapid performance degradation due to the rapid increase in ohmic and polarization resistance. Subsequently, the SOFC stack voltage decreases slowly, along with a small increase in stack resistance. Sehested et al. [20] showed that temperature has a great influence on the coarsening of Ni. When the temperature is low, the surface diffusion of Ni is slower and the particle coarsening is inhibited, as shown in Figure 5.

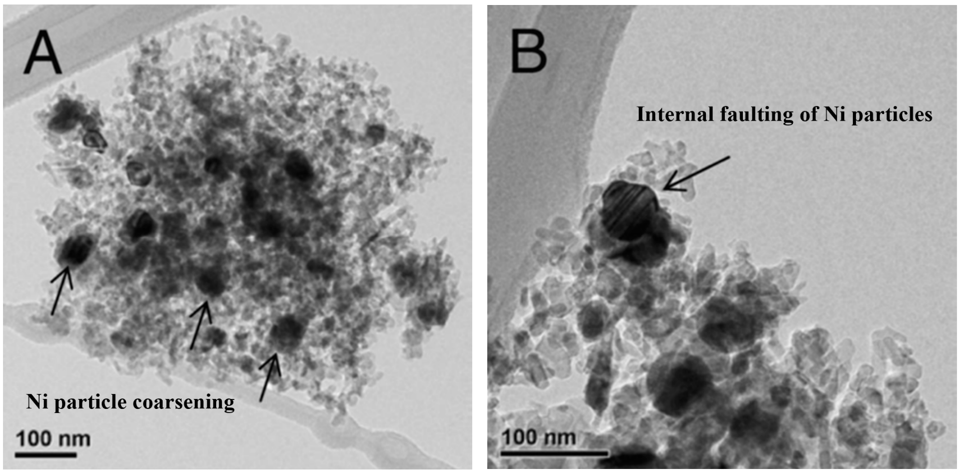


Figure 5. Ni particle coarsening failure: (**A**) Ni particle coarsening. (**B**) Internal faulting of Ni particles. Adapted with permission from Ref. [20].

Ni redox cycling is another reason for SOFC anode performance degradation. This degradation occurs because the Ni in the anode may be oxidized to NiO during the start-up and shutdown of the SOFC system. In turn, during system operation, NiO is reduced to Ni. However, due to the difference in volume between Ni and NiO particles, switching between the two may cause anode rupture or delamination at the intersection of electrolyte and anode. Klemensø et al. [21] investigated the performance degradation of SOFC systems during redox processes. The results show that the change in Ni particle volume can generate stress in the anode and electrolyte, which increases the probability of mechanical damage.

Carbon deposition can also cause SOFC system performance degradation. When SOFC systems use hydrocarbons as fuel, the fuel undergoes a cracking reaction catalyzed by Ni to produce carbon. The carbon deposition on the anode surface may cover the active site of the electrochemical reaction, which prevents the occurrence of the electrochemical reaction. Lanzini et al. [22] investigated the effect of carbon deposition on the performance degradation of nickel-based anode-supported SOFCs, as shown in Figure 6. The results show that carbon deposition can lead to an increase in stack ohmic and activation polarization. Carbon formation can be mitigated by adding the right percentage of carbon dioxide to the fuel.

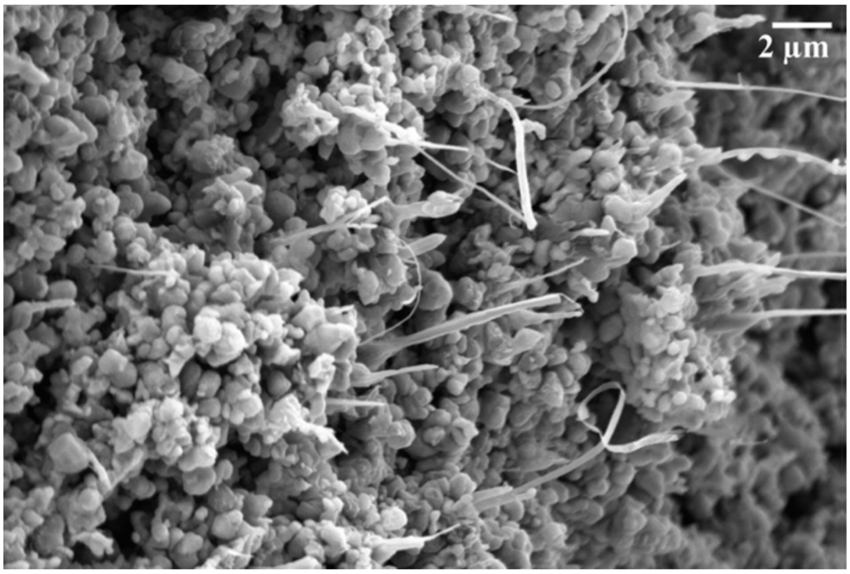


Figure 6. Carbon deposition in the anode. Reprinted with permission from Ref. [22].

Gas impurities in the anode can also cause degradation of the SOFC system performance. When impurities such as phosphide and sulfide react chemically with Ni in the anode, it may produce other substances. These substances can block the active reaction sites of the electrode, resulting in increased ohmic impedance, electrode corrosion, and changes in the electrode surface. Sezer et al. [23] developed a transient 3D planar cell model to simulate and analyze the cell performance degradation due to the fuel contaminant phosphine. The results show that the coverage of contaminants significantly alters the current and temperature distribution. Meanwhile, a dramatic decrease in the electrochemical performance of the cell anode is observed. Yoshizumi et al. [24] investigated the degradation mechanism of H_2S poisoning at the Ni anode. The results showed that the degradation of SOFC performance caused by H_2S poisoning is related to the increase of the O/Ni ratio and the growth of nickel particles in the anode.

2.1.3. Electrolyte Performance Degradation

In SOFC, the electrolyte mainly serves to transfer to oxygen ions and to isolate air and fuel gas. In electrolyte-supported SOFCs, the electrolyte is also responsible for supporting the entire cell structure. To prevent direct contact between air and fuel, the structure of the electrolyte should be dense and have considerable mechanical strength. In addition, the electrolyte should be able to isolate electrons so that electrons can be transferred via an external circuit, which creates a closed loop of current. Therefore, the electrolyte material needs to have high ionic conductivity and low electronic conductivity. The most commonly used electrolyte material is 8% Y_2O_3-stabilized ZrO_2 (YSZ). YSZ has a relatively high ionic conductivity at high temperatures, while it has relatively stable chemical properties. In addition, YSZ has good bending resistance and sufficient mechanical strength to reduce the occurrence of mechanical damage.

The decrease in ionic conductivity of YSZ is the main reason for SOFC electrolyte degradation. After high-temperature sintering, YSZ materials may change from cubic phase to tetragonal phase or other phases. However, the ionic conductivity of the tetragonal phase is not as good as that of the cubic phase. Therefore, the material phase change may cause a decrease in the ionic conductivity of the electrolyte, which in turn leads to a decrease in the SOFC system performance. Haering et al. [25] studied the degradation phenomenon of ionic conductivity of YSZ by high-resolution X-ray diffraction, neutron diffraction, and STEM. The results indicate that at high temperatures, a transformation occurs within the associates in the zirconia specimens leading to electrolyte performance degradation.

2.1.4. Interconnect Performance Degradation

In SOFC, interconnects connect multiple single cells to form a stack. The interconnects are generally bipolar plate constructions to isolate the gas and oxygen gas paths. The bipolar plate structure can simultaneously supply fuel to the upper single cell and oxygen to the lower single cell. Meanwhile, the interconnects also have the function of collecting current. Therefore, the interconnects need to meet the following characteristics: good densities, high electronic conductivity, good physical and chemical stability, and sufficient structural strength. Currently, high-temperature SOFC (800–1000 °C) mainly uses $LaCrO_3$ as the material for interconnects. Medium-temperature SOFC (600–800 °C) can use relatively inexpensive metal alloy materials.

The disadvantage of $LaCrO_3$ materials is a high price for interconnects, which increases the cost of SOFC. Moreover, its high hardness makes it difficult to process and shape. The disadvantage of metallic materials (such as SS430, corfer22, etc.) is that oxidation occurs at high temperatures, resulting in higher ohmic resistance of the stack. In addition, the Cr in stainless steel may volatilize into the cathode, leading to cathodic Cr poisoning. Geng et al. [26] showed in their study that when stainless steel materials are used to develop interconnects, oxidation of stainless steel at high temperatures can form Cr_2O_3 oxide layers. The Cr_2O_3 oxide layers can cause an increase in the impedance of the stack. Furthermore, Cr_2O_3 may evaporate and enter the cathode to form volatile Cr(VI) species, leading to cathodic Cr poisoning.

2.1.5. Sealing Material Performance Degradation

The function of the sealing material is to prevent gas leakage between the cell and the interconnect and between the interconnect and the stack frame. Therefore, the sealing material needs to have good gas tightness to ensure the sealing effect. In addition, the sealing material needs to have good thermal stability so that there is good thermal matching among the components of the stack. For SOFC, sealing performance is an important indicator. Once the cell has gas leakage, it will not only lead to cell performance degradation but may even cause danger. Currently, the types of SOFC seals are divided into two main categories, adhesion-type seals and compaction-type seals.

Adhesive seals ensure gas tightness by bonding the sealing material to the adjacent components. A glass-ceramic type seal is the most common type of SOFC adhesion seal. The fluidity of the glass body at high temperatures ensures a good seal, while the cost of the glass material is very low. However, the glass material may react chemically with the electrode material, which causes cell performance degradation. Chen et al. [27] found that the reaction between the sealing glass and the chromium on the interconnects results in the formation of chromate. The formation of chromate causes stress between the sealing glass and the metal interconnect, which in turn leads to the delamination of the stack, as shown in Figure 7.

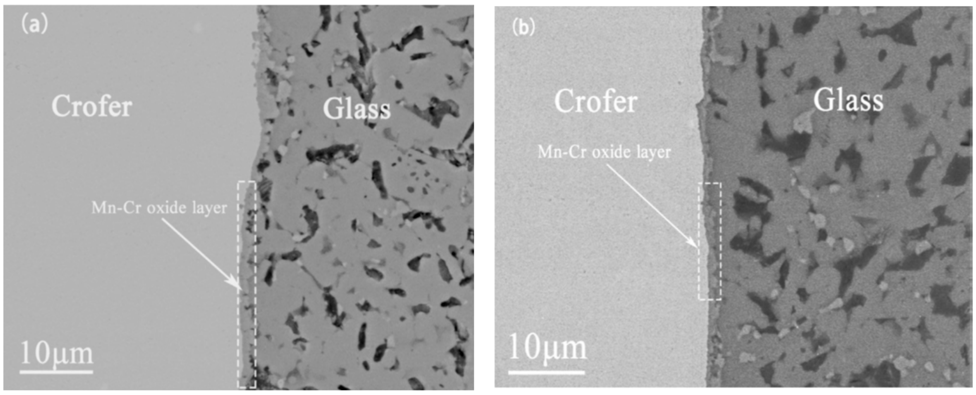


Figure 7. Reaction images of sealed glass and interconnects at different time lengths: (**a**) Reaction images at 100 h. (**b**) Reaction images at 500 h. Reprinted with permission from Ref. [27].

The compaction-type seal is used to seal the structure by applying pressure. Therefore, the compaction seal material does not need to match the thermal expansion coefficient of the cell element. The current mainstream compaction seal material is the mica seal. The strength of the mica material and the surface defects determine the sealability. The greater the strength and the fewer the surface defects of the mica material, the better the gas tightness. Chou et al. [28] tested Phlogopite mica flakes for leakage at compressive stresses of 3 to 100 psi and helium pressures of 0.2 or 2 psi. The results showed that seal rupture is associated with high-temperature leakage.

The current research on the mechanism of SOFC degradation is mainly focused on the degradation of stack materials. There are several possible causes of degradation in the stack, as shown in Table 1.

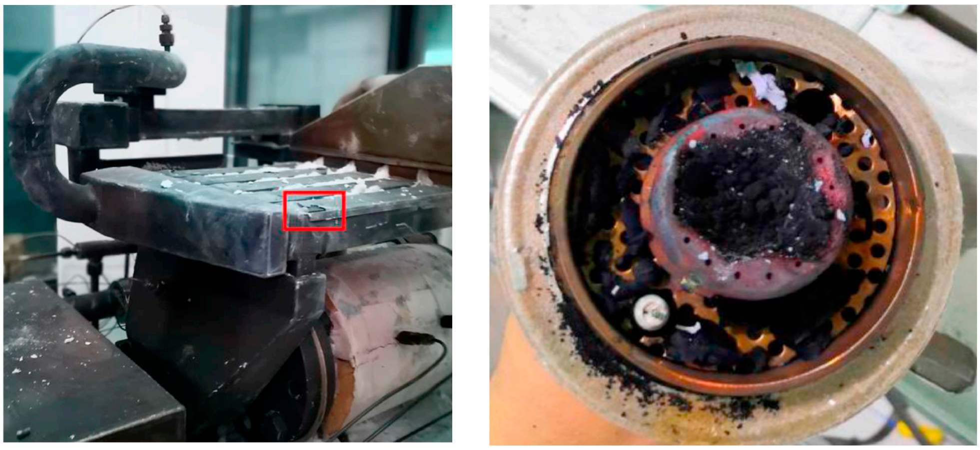
Table 1. Reasons for stack material degradation.

Stack Components	Reasons for Degradation
Cathode	Oxidation of LSM cathode, Cr poisoning, poisoning due to gas impurities
Anode	Ni particle coarsening, Ni redox, carbon deposition, poisoning due to gas impurities
Electrolytes	Decrease in ionic conductivity of YSZ
Interconnect	High-temperature oxidation of metal interconnects
Sealing material	The chemical reaction between glass sealing material and electrode material, mica seal leakage

2.2. System-Level Performance Degradation

From an application-oriented point of view, stacks in thermostatic test furnaces are difficult to commercialize. Therefore, the operation of the stack needs to be moved from the test furnace to the power generation system. The detailed discharge process of natural gas in the SOFC system is as follows: The natural gas is first injected into the reformer through a pressure-reducing valve. In the reformer, methane gas and deionized water are catalyzed to produce hydrogen and carbon monoxide. Then, the fuel gas is preheated by a heat exchanger and enters the SOFC stack anode. The air is preheated by a heat exchanger and then enters the cathode of the stack. Next, the fuel and air undergo an electrochemical reaction in the stack to generate electricity. However, the exhaust gas from the anode of the stack still contains a certain percentage of combustible material. This part of the combustible material will be fully burned in the exhaust combustion chamber, releasing more heat. The heat generated in the combustion chamber is used in the heat exchanger to preheat the fuel and air. Therefore, each component of the SOFC system is very important. The degradation of any component can affect the performance of the whole system.

Peng et al. [2] studied the effect of heat exchanger rupture failure and reformer carbon deposition failure on SOFC performance degradation, as shown in Figure 8. The results show that heat exchanger rupture can lead to an increase in combustion chamber temperature and a decrease or even disappearance of the stack cathode inlet pressure. Moreover, it increases the system's air demand. Carbon deposition failures in the reformer can lead to incomplete reforming reactions that do not produce the desired gas required by the stack, which can further lead to the degradation of SOFC system performance. Moreover, it can cause frequent dithering of the methane flow rate and combustion chamber temperature. Nakajo et al. [29] investigated the effect of constant power output mode and dynamic scheduling mode on the performance degradation of SOFC systems. The results show that the dynamically scheduled SOFC system proves to be more durable and less degraded than a system operating at a constant full power output. Moreover, the power generated in the SOFC device and its delivery efficiency are highest in the constant half-power mode. Wu et al. [30] developed a system-level degradation prediction method for SOFC based on the Elman neural network. The results showed that the method was able to infer potential degradation information from the voltage data. Wu et al. [31] investigated the effect on SOFC system lifetime during standby and shutdown. The results of the study showed that the cooling rate is a major factor affecting the remaining lifetime of the system. Tucker et al. [32] investigated the effect of integration with a gas turbine system on the lifetime of the SOFC. The results showed that when SOFC performance degrades, transferring power to the gas turbine can significantly extend the SOFC life.



(**a**) (**b**)

Figure 8. Failures occurring in SOFC systems: (**a**) Heat exchanger rupture. (**b**) Reformer carbon deposition. Reprinted with permission from Ref. [2].

Most of the current studies on the degradation mechanism of SOFC systems focus on the stack itself but ignore the influence of BOP components such as the reformer, exhaust combustion chamber, and heat exchanger. To commercialize SOFC, it is equally important to investigate the SOFC system-level degradation mechanism. However, there are relatively few studies on SOFC system-level degradation. Therefore, research efforts on SOFC system-level performance degradation need to be enhanced.

3. SOFC Degradation Performance Prediction

The SOFC system degradation performance prediction technique is an advanced monitoring and diagnostic function. The technology is designed to assess the health of critical components and predict their degraded performance. An effective degradation performance prediction strategy can provide the basis for system maintenance. Furthermore, predictive techniques can help reduce costs by reducing unplanned maintenance. Therefore, degradation performance prediction is a prerequisite for SOFC system performance optimization. It can get more time for SOFC system health control to inhibit the deterioration of system performance. Currently, degradation performance prediction methods for SOFC systems can be classified into model-based methods, data-based methods, electrochemical impedance spectroscopy (EIS)-based methods, and image-based methods.

3.1. Model-Based Degradation Performance Prediction

The model-based approach describes the evolution of SOFC performance by building a physical model. The model predicts the degradation of the system by simulating the microscopic parameters and material properties of the SOFC. The model-based prediction method can capture the trend of voltage degradation over a longer prediction time.

Dolenc et al. [9] used a nonlinear SOFC stack model with aggregate parameters and a Kalman filter to predict the degradation of the stack. The results show that the proposed degradation model can adapt to changes in degradation rates. Even if the operating conditions change, the model still has a good prediction. Gemmen et al. [33] built a mechanistic model to describe the polarization loss and predict the degradation performance. Specific area resistance (ASR) and degradation rate (DR) are also defined to characterize SOFC stack degradation. Experimental results show that ASR proves to be insensitive to certain changes in test conditions and can be used to compare performance differences due to incremental changes in design/materials. DR is the optimal parameter to determine the efficiency change during the life of the cell. Mason et al. [34] proposed a multiphysics model of SOFC to predict the performance degradation caused by electrode coarsening of LSM-YSZ/YSZ/Ni-YSZ SOFC. The results show that increasing the operating temperature significantly increases degradation when coarsening is considered as the only degradation mode. Moreover, there exists an optimal temperature that maximizes lifetime performance. Zaccaria et al. [35] developed a real-time one-dimensional SOFC model to predict the effect of operating parameters on the output voltage in the presence of degradation. Fu et al. [36] developed a multi-physics field model to predict the performance degradation of direct methane reforming (DIR) SOFC due to Ni particle coarsening. The results show that the model can quantitatively predict the effects of Ni particle coarsening on Ni particle percolation probability, effective TPB area, effective electron conductivity, maximum power density, and DIR-SOFC degradation rate. Zhu et al. [37] predicted the effect of Ni coarsening on SOFC performance by building a transient multi-physical field model. The results showed that the high operating temperature can accelerate the growth of Ni particles and intensify SOFC performance degradation. Meanwhile, the increase of vapor to carbon ratio also can intensify the coarsening process of Ni particles and deteriorate the SOFC transient performance. However, multi-physics field simulations require a long computational time and are difficult to achieve fast predictions. More model-based studies on SOFC degradation performance prediction are shown in Table 2.

Table 2. Model-based study of SOFC degradation performance prediction.

Prediction Method	Predicted Objects	Models	Ref.
Model-based approach	Predicting future degradation trends and remaining service life of SOFC stack.	Nonlinear SOFC power stack integration model	[9]
Model-based approach	Predicting the long-term performance degradation process of SOFC stack under accelerated operating conditions.	SOFC multi-physics field degradation model	[19]
Model-based approach	Predicting cell performance degradation due to fuel contaminant phosphine.	Transient 3D planar SOFC model	[23]
Model-based approach	Predicting the impact of scheduling methods on SOFC system performance.	Degradation model of each SOFC component	[29]
Model-based approach	Predicting the performance degradation of the stack when coal syngas is the fuel.	A mechanistic model of the stack	[33]

Table 2. *Cont.*

Prediction Method	Predicted Objects	Models	Ref.
Model-based approach	Predicting performance degradation caused by electrode coarsening.	SOFC multi-physics field model	[34]
Model-based approach	Predicting the effect of operating parameters on SOFC output voltage in the presence of degradation.	1D real-time SOFC model	[35]
Model-based approach	Predicting degradation of direct methane reforming (DIR) SOFC performance due to Ni particle coarsening.	SOFC multi-physics field model	[36]
Model-based approach	Predicting the effect of Ni coarsening on SOFC performance.	Transient multi-physics field model for SOFC	[37]
Model-based approach	Predicting the degradation of the electrochemical performance of SOFC anodes due to Ni grain growth.	SOFC anode degradation prediction model	[38]
Model-based approach	Predicting SOFC performance degradation caused by nickel content, oxidation state, and temperature.	Multi-physics field model of SOFC stack	[39]

The model-based approach is the most commonly used method to predict SOFC performance degradation. However, the application of model-based prediction methods is affected by two factors. First, the mechanism model lacks accuracy due to an incomplete understanding of the degradation mechanisms in electrodes, electrolytes, interconnects, and seals [40]. In addition, models have difficulty simulating the inhomogeneity of materials, fabrication, operation, and degradation. Model building requires a large number of assumptions to be made in advance, which can make the degradation too idealized [41].

3.2. Data-Based Degradation Performance Prediction

Data-based prediction is achieved by building black-box models. It learns the system behavior from the system's sensor monitoring data to predict the future system state. It does not require any physical understanding of the system's behavior. The data-based prediction method has better adaptability and accuracy, which is easy to implement.

Rao et al. [42] proposed a state prediction model for SOFC systems based on a long short-term memory network (LSTM). The model uses a two-layer LSTM structure, which supports multiple sequential feature inputs and flexible multi-step prediction outputs. The model is compared with the traditional ARIMA model and the LSTM recursive prediction model. The results show that the multi-step LSTM prediction model has excellent prediction performance in the multi-step prediction of SOFC system states. Chi et al. [43] proposed a link function degradation model to predict the degradation of SOFC. Moreover, a cyclic batch identification process based on the great likelihood method is used to identify its parameters online. The results show that the method can identify long-term degradation trends. The method statistically improves the accuracy and stability of the prediction compared to the constant degradation velocity model. Wu et al. [44] used a least squares support vector machine (LS-SVM) classifier for SOFC fault type identification. Then, the remaining lifetime of the SOFC is estimated using the hidden semi-Markov model (HSSM). Zu et al. [45] proposed a method to predict SOFC performance degradation by combining an empirical model and a data-based algorithm. The method can capture the degradation trend of aging data while maintaining details of short-term nonlinear degradation characteristics. This improves the accuracy of the prediction results. Pahon et al. [46] used the wavelet transform method to predict eigenvolumes that are sensitive to anomalous states. Moreover, wavelet energy and wavelet entropy are combined as performance indicators to predict the lifetime of SOFC. Yang et al. [47] proposed a method combining machine learning and relaxation time (DRT) distribution to predict the degradation of SOFC performance caused by Cr poisoning. The results show that the method has a strong predictive capability and the predicted data are in good agreement with the experimental results. Li et al. [48] proposed an encoder-decoder-based recurrent neural network (RNN) state prediction model to predict the degradation performance of SOFC systems. The results show that the method has high accuracy. However, the implementation of the method requires a large amount of training data. More data-based studies on SOFC degradation performance prediction are shown in Table 3.

Table 3. Data-based study on SOFC degradation performance prediction.

Prediction Method	Predicted Objects	Algorithm	Ref.
Data-based Approach	Predicting the degradation performance of SOFC systems.	Double-layer LSTM model	[42]
Data-based Approach	Predicting long-term degradation trends in SOFC.	Link function, great likelihood algorithm	[43]
Data-based Approach	Identifying the SOFC fault types and predicting the remaining SOFC lifetime under the faults.	Least squares support vector machine, semi-hidden Markov algorithm	[44]
Data-based Approach	Predicting heat exchanger rupture failures in SOFC systems.	SVM algorithm	[46]
Data-based Approach	Predicting the SOFC performance degradation caused by Cr poisoning.	Machine learning combined with relaxation time (DRT) distribution	[47]
Data-based Approach	Predicting the SOFC system's degradation performance.	A state prediction model based on encoder-decoder RNN	[48]
Data-based Approach	Finding the effective characteristic quantity for judging the abnormal operation status and predicting the remaining lifetime of SOFC.	Wavelet transform algorithm	[49]
Data-based Approach	Forecasting performance degradation of proton exchange membrane fuel cells (PEMFC).	Grid long and short-term memory, recursive neural network	[50]
Data-based Approach	Predicting the output voltage trajectory of SOFC.	Neural network (NN) algorithm	[51]
Data-based Approach	Predicting the impact of uncertainty in SOFC during degradation.	An approximate randomized algorithm based on Taylor series expansion	[52]
Data-based Approach	Predicting SOFC performance degradation at seven different temperatures and four different fuel operating conditions.	The semi-empirical degenerate prediction algorithm	[53]

Data-based methods are currently developing rapidly in the field of SOFC degradation performance prediction. However, there are still the following drawbacks: first, the accuracy of data-driven models depends on the amount of training data. However, the acquisition of training data is expensive and time-consuming. Second, the quality of the training data also affects is the accuracy of the algorithm. Good training data should contain the degradation trend of the system. Third, data-driven algorithms require the selection of appropriate algorithm structures and parameters to achieve high accuracy. This affects the practicality of data-based methods.

3.3. EIS-Based Degradation Performance Prediction

Electrochemical impedance spectroscopy works by injecting a sinusoidal current stimulus into the SOFC system and recording the voltage response. The ratio between the voltage and the input current represents the electrochemical impedance of the system. The impedance information can be used to predict the performance changes of the system.

Khan et al. [38] used EIS, scanning electron microscopy (SEM), and two-dimensional image analysis techniques to quantify the particle size, phase ratio, and TPB point distribution of the anodes. Moreover, SOFC performance degradation is predicted. The results show that the variation of particle size and TPB length in the anode with time fits well with the power-law coarsening model. The model can be used to gain insight into the effects of the electrochemical degradation of SOFC anodes. Muñoz et al. [54] predicted the degradation of SOFC by EIS, polarization curves, distributed relaxation time (DRT) methods, and equivalent circuit modeling (ECM). The results show that the method can predict the performance degradation of a single cell when operating under predetermined conditions. Gallo et al. [55] developed a SOFC online aging estimation algorithm combining an envelope dynamic model with EIS. The algorithm can diagnose faults during the operation of the stack, as well as to predict the remaining useful life (RUL). Gazzarri et al. [56] predicted the effect of electrode delamination failure of the cathode on the SOFC degradation performance by analyzing the change in the EIS of the series and polarization resistances.

There are two main disadvantages of using EIS to predict the SOFC degradation: first, the performance of SOFC can be affected by EIS during operation. Second, additional equipment is required, which increases the cost of the system.

3.4. Image-Based Degradation Performance Prediction

Image analysis methods are a powerful and reliable tool for predicting performance degradation. The image method can effectively characterize the microscopic features, mechanical properties, and thermal properties of SOFC. The performance parameters observed through the images can be used to quantify the degradation phenomenon in SOFC. The image-based degradation prediction method first needs to acquire the image information of the SOFC interior by scanning electron microscopy (SEM), transmission electron microscopy (TEM), and focused ion beam (FIB) techniques. Then, the obtained images are processed and quantitatively analyzed to obtain information about the microstructure, including phase fraction, grain size, particle size pattern, composition shape factor, the spatial organization of the phases, and descriptive features. Finally, the degradation of the system is analyzed by the obtained information on the internal microstructure of SOFC. Compared to model-based, data-based performance prediction methods, image analysis is a simple and effective technique.

Ananyev et al. [57] developed a method for the detection of degraded areas based on image line autocorrelation function analysis. The method successfully detected the degradation of SOFC system performance due to chromium poisoning and nickel agglomeration. Simwonis et al. [58] proposed an image analysis technique to predict the degradation of Ni/YSZ anodes in SOFCs. The results show that after 4000 h of exposure to Ar/4% H_2/3% H_2O at 1000 °C, a 33% decrease in initial conductivity is measured. This is related to the aggregation of metal particles. Yan et al. [59] developed a low-cost and fast image analysis method to predict the degradation of the SOFC anode microstructure. The results show that the method not only covers more areas than the FIB-SEM method, but also has less dependence on the device.

The main drawback of the image-based approach is that the equipment is expensive and can significantly increase the cost of system performance analysis. In addition, the image method can generally only analyze and observe the images inside the system after the equipment has been shut down. Therefore, the method can only perform offline analysis of SOFC performance degradation.

4. SOFC Performance Optimization Scheme

After predicting the performance degradation of the system, SOFC systems need to avoid the impact of degradation effectively. To extend the operational lifetime of SOFC systems, it is necessary to develop performance optimization strategies to suppress SOFC performance degradation. The optimization strategy can be designed in terms of both hardware architecture and software strategy. The improvement of the hardware structure can be specifically divided into the improvement of the SOFC system material and the improvement of the structure. The optimization of the software strategy mainly refers to the design of the SOFC system health controller.

4.1. System Material and Structure Improvement

Under practical operating conditions, material and microstructural instabilities can lead to SOFC degradation. Therefore, the development of new materials and optimization of microstructure are desirable for the long-term stability of SOFC.

Qi et al. [60] find that TiC/Hastelloy alloy composites used as interconnects can generate dense NiO (or TiO_2-Cr_2O_3) layers, which can prevent the SOFC performance degradation caused by interconnect oxidation. Hannes et al. [61] developed a Crofer 22APU high-temperature ferritic stainless steel for use as SOFC interconnects. This material can be used to solve the performance degradation problems such as creep and rupture in conventional interconnects. Cheng et al. [62] used molecular dynamics methods for YSZ electrolyte material improvement to mitigate the degradation problems caused by the reduced ionic conductivity of YSZ. They eventually obtained the optimal Y_2O_3 doping concentration to mitigate the degradation. Chen et al. [63] mitigated the performance degradation caused by Ni particle coarsening by incorporating an interstitial layer consisting of nanoparticles in the SOFC electrode. Jiang et al. [64] investigated the Ni particle coarsening in Ni-YSZ anodes at different YSZ contents. The results showed that increasing the content of YSZ can effectively suppress the coarsening of Ni particles. Aphalet et al. [65] provided a scheme for SOFC using aluminum oxide and coatings to build air handling systems (heat exchangers, ducts, manifolds). This scheme can prevent chromium evaporation and associated electrode poisoning. More studies on SOFC performance optimization based on material improvement are shown in Table 4.

Table 4. SOFC performance optimization scheme based on material improvement.

Optimized Solutions	Degradation Type	Improvement Methods	Ref.
Material Improvement	Interconnect oxidation	TiC/Hastelloy alloy composites are used to develop the interconnects.	[60]
Material Improvement	Creep and rupture of interconnects	Crofer 22APU high-temperature ferritic stainless steel is used to develop the interconnects.	[61]
Material Improvement	Decrease in electrolyte YSZ ionic conductivity	Changing the doping concentration of Y_2O_3.	[62]
Material Improvement	Anode Ni particle coarsening	An interstitial layer consisting of nanoparticles is added to the composite electrode.	[66]
Material Improvement	Cathode Cr poisoning	A dense and uniform alumina protective layer is generated on the surface of the interconnect to reduce the evaporation of Cr.	[64]

Table 4. *Cont.*

Optimized Solutions	Degradation Type	Improvement Methods	Ref.
Material Improvement	Anode sulfur poisoning	Transition metals such as Cu, Pd, Au, Ag, and Rh are doped in Ni anodes to reduce anode sulfur poisoning.	[65]
Material Improvement	Redox of anode Ni	Nickel is infiltrated into the prefabricated porous yttrium oxide stabilized zirconia structure.	[67]

Schluckner et al. [68] discussed the effects of co-current, cross-flow, and cross-current structures on the electrical characteristics of SOFC stack. The results show that the cross-flow stack structure can lead to a more uniform current density and temperature distribution throughout the SOFC stack, which is beneficial to extend the SOFC stack lifetime. Canavar et al. [69] experimentally analyzed the temperature distribution and performance of SOFC stacks with two different anode flow channel designs. One is a conventional rectangular machined design and the other is an arbitrary flow path layout based on a woven structure of nickel mesh. The results show that the fuel flow is more uniformly distributed when using a woven structure based on nickel mesh. This can reduce the peak stack temperature and improve the stack performance. Seo et al. [70] investigated the effect of different electrical contact methods on SOFC performance. They found that the type of contact significantly affects the stability of the cell during long-term operation and thermal cycling. Moreover, stable operating conditions can be achieved by using platinum-based contact paste. Chen et al. [71] investigated the effect of gas channel shape on the performance of micro-SOFC. Numerical simulations revealed that the finger-shaped anode support structure is more conducive to improving the stack performance. More studies on SOFC performance optimization based on structural improvements are shown in Table 5.

Table 5. SOFC performance optimization scheme based on structural improvement.

Optimized Solutions	Improvement Program	Improvement Effect	Ref.
Structural improvement	The gas flow path of the stack uses a cross-flow structure.	It can make the current density and temperature distribution of the whole SOFC stack more uniform, which helps to extend the life of the SOFC stack.	[68]
Structural improvement	Adopting the anode flow channel layout based on the woven structure of nickel mesh.	The fuel flow in the stack is more uniformly distributed. The stack peak temperature is lower.	[69]
Structural improvement	Adding platinum-based contact paste to the electrical contact points.	The output performance of the stack becomes more stable.	[70]
Structural improvement	Finger-shaped anode support structure is adopted as the air channel shape for micro SOFC.	The finger-shaped anode support structure is more conducive to improving the stack performance.	[71]

Table 5. *Cont.*

Optimized Solutions	Improvement Program	Improvement Effect	Ref.
Structural improvement	Adding coin cell batteries to SOFC systems.	The coin cell can assist in the thermal conversion of the stack and can effectively improve the dynamic performance of the SOFC system.	[72]
Structural improvement	X-shaped column interconnects.	It can significantly increase the oxygen concentration of the porous cathode under the rib, reduce the cathode concentration difference polarization loss, and improve the performance of the stack.	[73]
Structural improvement	Applying an air bypass valve to the air input side of the stack.	It is sufficient to increase the adjustment range of air input, which in turn increases the adjustment range of system temperature and improves the overall system performance.	[74]

Performance optimization schemes based on system material and structural improvements are offline optimization methods. This method optimizes the downtime system after degradation has occurred. The advantage of this method is that it can solve the degradation problem more thoroughly which improves the reliability of the SOFC system from the root. However, the method requires significant time and cost to validate new materials and structures. The method is not suitable for systems that are actually working.

4.2. Health Controller Design

In recent years, SOFC performance degradation has become an increasingly important issue. Therefore, system health controllers for mitigating performance degradation occupy an increasingly important position. SOFC system health control aims to optimize the performance degradation problem by adjusting the system parameters.

Wu et al. [10] developed a new strategy combining prediction and dynamic optimization to quantitatively extend the lifetime of SOFCs under nickel coarsening and oxidative degradation mechanisms. The results show that this strategy can effectively extend the fuel cell life without significantly reducing the system power generation efficiency. Nakajo et al. [75] developed a control strategy to extend the operational lifetime of SOFC systems. The results show that the Cr contamination of the cathode dominates the SOFC stack degradation. The control strategy can improve the durability of SOFC by optimizing the operating parameters. Moreover, controlling the SOFC stack to operate at a lower system-specific power and higher temperature can extend the lifetime by 10 times. Spivey et al. [76] proposed a constrained control method to extend the lifetime of tubular SOFCs. The method uses minimum cell temperature and maximum radial thermal gradient as key control variables to extend fuel cell life. The optimization results show that the method extends the SOFC operating life by reducing the thermal stress inside the stack. Parhizkar et al. [77] proposed a degradation-based optimization (DBO) framework to find the optimal operating conditions for the target lifetime. The results showed that the target lifetime has a significant effect on the system productivity, optimal operating temperature, and current density. Wu et al. [78] developed four model predictive controllers for load tracking and temperature safety of SOFC systems under fuel leak failure, air compressor failure, simultaneous failure, and health, respectively. Sangtongkitcharoen et al. [79] reduced the formation of carbon deposition by controlling the proportion of water vapor within the fuel in the SOFC system. The results show that carbon deposition on the surface of the anode can be avoided when the ratio of water vapor to methane in the fuel is higher than 1.6. However, too high a water vapor ratio can lead to a decrease in system efficiency. Mueller [80] developed a control method based on constant fuel utilization. This method prevents the fuel deficit phenomenon by limiting the fluctuation of fuel utilization under instantaneous power demand. Table 6 summarizes the research on the design of SOFC health controllers.

Table 6. SOFC system health controller design study.

Control Purpose	Control Solutions	Control Effect	Ref.
Quantitatively extending the lifetime of SOFC under nickel roughening and oxidative degradation mechanisms.	Control strategy combining prediction and dynamic optimization	It can effectively extend the life of SOFC without significantly reducing the efficiency of power generation.	[10]
Optimizing the performance degradation of SOFC in terms of lifetime and electrical efficiency.	Operation parameter optimization strategy	Operating at a lower system-specific power and higher stack temperature can extend the lifetime by 10 times.	[75]
Extending fuel cell life by controlling minimum cell temperature and maximum radial thermal gradient.	Constrained control method for the lifetime of tubular SOFC	This method extends SOFC operating life by reducing the thermal stress inside the stack and reduces operating costs by 5%.	[76]
Finding the optimal operating conditions for the target operating time of the anode-supported SOFC.	Degradation-based optimization (DBO) framework	The target life has a significant impact on system productivity, optimal operating temperature, and current density.	[77]
Implementing load tracking and temperature safety for SOFC systems.	Neural network-based predictive controller	Load tracking and temperature safety are achieved under different fault states.	[78]
Reducing the formation of carbon deposits by controlling the proportion of water vapor in the fuel in the SOFC system.	SOFC system water-to-carbon ratio controller	When the ratio of water vapor to methane in the fuel is higher than 1.6, carbon deposition on the surface of the anode Ni particles can be avoided.	[79]
Preventing fuel deficits.	Control method based on constant fuel utilization	Successfully preventing fuel deficits by limiting fluctuations in fuel utilization under instantaneous power demand.	[80]
Reducing the thermal stress in the stack.	Model predictive controller based on the generalized predictive control algorithm	The controller can regulate and control the temperature difference of the cells in the SOFC stack to reduce the thermal stress in the stack.	[81]
Minimizing SOFC stack space temperature.	H-infinity-based feedback control strategy	The control strategy achieves load following and space temperature minimization under fast and large load disturbances by controlling the air flow rate and the cathode inlet temperature.	[82]
Controlling the temperature gradient inside the SOFC stack.	Composite nonlinear controller based on higher-order sliding mode observer	The temperature inside the stack can be observed by the observer and the temperature gradient can be adjusted.	[83]

The performance optimization scheme based on health controller design is an online optimization method. This method can optimize the degradation performance of SOFC by adjusting the parameters during the system operation. This method is cheap to implement.

Moreover, it can avoid system downtime for maintenance when degradation is not serious. However, health controllers may sacrifice output performance such as the electrical efficiency of the system to mitigate degradation.

5. Conclusions and Future Remarks

To make SOFC systems operate with long life and high performance, degradation mechanism analysis, degradation performance prediction, and performance optimization strategy design are necessary. In this study, the degradation mechanism analysis study of SOFC is first reviewed from the stack level and system level, respectively. Then, the model-based, data-based, EIS-based, and image-based methods for SOFC degradation performance prediction are summarized. Finally, the performance optimization scheme is reviewed in two aspects: material structure improvement and health controller design.

A large amount of research work already exists in these three directions, but there are still shortcomings in these studies. The research work on SOFC degradation mechanism analysis is mainly focused on the degradation study of stack materials and structures. There is still relatively little work on degradation studies of the entire SOFC system. However, it is equally important to investigate the mechanisms of degradation at the system level. This is because the system-level degradation mechanism study is a macroscopic degradation study, which is more conducive to the commercialization of SOFC.

The four degradation performance prediction techniques, model-based, data-based, EIS-based, and image-based, all have their characteristics and different scope of application. Model-based prediction methods have the widest range of applicability and are capable of predicting different types of degradation. However, the model-based approach lacks accuracy because the mechanism model is too complex. Data-based methods do not require advanced knowledge of degradation mechanisms and have better adaptability and accuracy. However, the accuracy of data-based methods depends on the quantity and quality of training data. Both EIS-based and image-based prediction methods are capable of predicting the degradation trend of system performance directly at the microscopic level. However, both methods require expensive equipment, which can significantly increase the cost of experiments. If several methods can be used in combination, better results may be achieved.

Material structure improvement and health controller design are offline optimization methods and online optimization methods, respectively. Material structure improvement can solve the degradation problem more thoroughly at the micro level and improve the reliability of the SOFC system from the root. However, this method requires a lot of time and cost. The health controller is a direct mitigation of SOFC degradation during system operation. However, the mitigation of degradation may require sacrificing other performances of the system.

There is still a gap between the current state of the SOFC industry and the existing research. If we want to achieve further commercialization of SOFC, we still need to conduct in-depth research in degradation mechanism analysis, degradation performance prediction, and performance optimization strategy design.

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