Catalyst Degradation in Proton Exchange Membrane Fuel Cells: Mechanisms, Investigation Techniques, and Predictive Modelling

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ABSTRACT
Proton Exchange Membrane Fuel Cells (PEMFCs) represent a promising solution for sustainable energy. However, the degradation of their catalyst layer (CL), particularly platinum-based catalysts, remains a major barrier to their efficiency and long-term durability. This review summarizes the key degradation mechanisms within the CL, including platinum dissolution, Ostwald ripening, carbon support corrosion, and nanoparticle agglomeration. It further examines experimental and characterization techniques employed to investigate these mechanisms. Specifically, it discusses accelerated stress tests (ASTs) and life tests used to mimic long-term performance loss under realistic conditions, alongside electrochemical and microscopic methods for monitoring structural and chemical transformations. In addition, it highlights predictive modelling approaches, both model-based and data-driven, that offer effective strategies to forecast degradation trends and guide catalyst design. Finally, this work proposes the use of hybrid modelling, integrating both physics-based and data-driven approaches, as a holistic framework to improve the understanding and mitigation of CL degradation, ultimately contributing to the development of more durable and efficient PEMFC systems.

***Keywords*: PEM Fuel Cells, Catalyst Layer Degradation, Model-Based, Data-driven Model, Investigation Techniques**

I. INTRODUCTION

Hydrogen-based energy technologies have gained increasing attention as the world transitions toward sustainable energy solutions. Among these, Proton Exchange Membrane Fuel Cells have emerged as a promising alternative due to their high efficiency, high power density, low emissions, and reliable energy output[1], [2], [3], [4]. However, their widespread adoption is still limited by two critical challenges: high production costs and limited durability, primarily caused by degradation of the CL[5].

The CL, which consists of platinum (Pt) nanoparticles dispersed on a carbon support, combined with an ionomer matrix[6], [7], plays a crucial role in facilitating the oxygen reduction reaction (ORR) at the cathode and the hydrogen oxidation reaction (HOR) at the anode[8]. However, prolonged PEM fuel cell operation leads to CL degradation through multiple pathways, including platinum dissolution and redeposition, carbon support corrosion, and nanoparticle agglomeration[9]. These mechanisms collectively reduce the electrochemically active surface area (ECSA), which quantifies the active sites on the catalyst surface, directly impairing catalyst efficiency, and ultimately degrading fuel cell performance[10], [11], [12].

To tackle these challenges, researchers have used various approaches to investigate CL degradation. ASTs and life tests are commonly employed to assess fuel cell durability under both controlled conditions and real-world operational scenarios[10]. Additionally, characterization techniques such as cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), and transmission electron microscopy (TEM) provide valuable insights into catalyst layer transformations[11]. Furthermore, predictive modeling frameworks, which include both physics-based models and data-driven techniques, have been developed to simulate and anticipate CL degradation, supporting the development of more resilient fuel cell designs.

Although significant progress has been made in understanding CL degradation, several challenges remain in effectively addressing and mitigating it. Experimental techniques such as ASTs and life tests provide valuable insights into degradation mechanisms but are often time-consuming and may not accurately replicate real-world operating complexities. Conversely, modeling approaches offer complementary benefits: physics-based models enhance mechanistic understanding but demand complex parameterization and significant computational resources, while data-driven models deliver accurate predictions but lack physical interpretability, limiting their application into system-level-design.

To address these limitations, this review systematically examines both experimental and modeling strategies used to study CL degradation. It explores key degradation mechanisms, including platinum dissolution, Ostwald ripening, carbon support corrosion, and nanoparticle agglomeration, along with the characterization techniques used to detect them. Additionally, it compares physics-based modeling approaches with data-driven methods to assess their reliability and practical relevance. Ultimately, this paper highlights the emerging potential of hybrid modeling, which integrates physics-based and data-driven methods, as a holistic framework for a better understanding of CL degradation. This integration aims to support the development of more durable and efficient PEMFC systems and proposes future directions for advancing hybrid modeling specifically tailored to CL degradation analysis.

II. BASIC OPERATION OF PROTON EXCHANGE MEMBRANE FUEL CELLS

A PEM Fuel Cell generates electricity through an electrochemical reaction between hydrogen and oxygen, as illustrated in Fig. 1. At the anode, hydrogen molecules dissociate into protons and electrons via an oxidation reaction (Equation 1). The protons then migrate through the proton exchange membrane to the cathode, while the electrons travel through an external circuit, generating electrical power. At the cathode, oxygen reacts with the incoming protons and electrons in a reduction reaction (Equation 2) to form water as the only byproduct. This electrochemical process enables PEMFCs to operate with high efficiency and minimal environmental impact, making them ideal for transportation and stationary power applications.

Anode Reaction: $H_2 \rightarrow 2H^+ + 2e^-$ (1)

Cathode Reaction: $(1/2)\ O_2 + 2H^+ + 2e^- \rightarrow H_2O$ (2)

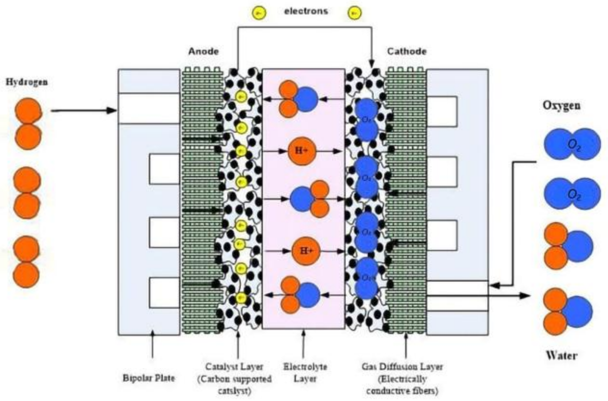

Fig. 1: The working principle of PEMFCs[12]

III. ROLE OF THE CATALYST LAYER IN PEMFCS

The catalyst layer is a critical component of PEMFCs because it facilitates the electrochemical reactions necessary for energy conversion[13], [14]. Positioned between the gas diffusion layer (GDL) and the proton exchange membrane (PEM), the CL enables the HOR at the anode and the ORR at the cathode, as illustrated in Figure 2. These reactions are fundamental to the operation of PEMFCs, making the CL a key factor in determining the overall efficiency and durability of the system[15], [16].

Typically, the CL consists of Pt nanoparticles dispersed on a porous carbon support, which provides a high surface area for electrochemical reactions (see Fig. 2)[17]. Additionally, this structure includes an ionomer matrix, usually made of Nafion®, which enhances proton conductivity and facilitates the efficient transport of reactants[18]. Due to its exceptional catalytic activity and stability under acidic conditions, Pt remains the most widely used catalyst in PEMFCs[19].

Despite its essential role, the CL is highly susceptible to degradation, especially under harsh operating conditions, such as potential cycling, elevated temperatures, and reactant starvation[20]. These stressors lead to structural damage within the CL, including Pt dissolution, agglomeration, carbon support corrosion, and ionomer deterioration. As a result, these factors significantly reduce catalyst stability and overall fuel cell efficiency[21]. The following section will examine these degradation mechanisms in detail.

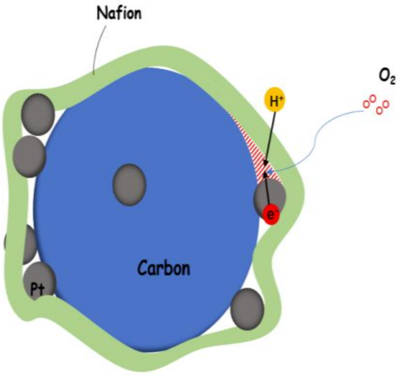

Fig 2. Schematic diagram of three-phase boundary of PEMFC cathode[22].

IV. CATALYST LAYER DEGRADATION MECHANISMS

The CL in Proton Exchange Membrane Fuel Cells is subjected to various electrochemical, thermal, and mechanical stresses during operation. These conditions initiate several degradation mechanisms, as shown in Fig. 3, which impair catalyst activity, compromise structural integrity, and ultimately reduce fuel cell performance and durability. The most critical mechanisms include:

(i) platinum dissolution and redeposition
(ii) carbon support corrosion
(iii) nanoparticle agglomeration and sintering

The following subsections explore each mechanism in detail.

1. Platinum dissolution and redeposition

Platinum dissolution plays a crucial role in the degradation of CL in PEMFCs [23]. During operation, variations in electrode potential, particularly at high voltages and under dynamic load cycling, lead to the electrochemical dissolution of Pt into soluble Pt^{2+} ions[24], [25], [26]. These ions can then migrate through the ionomer matrix, where they may either redeposit onto existing particles, forming large, less active clusters, or accumulate within the membrane to form a platinum band[24]. Both processes contribute to a gradual loss of ECSA and a reduction in ORR efficiency [27].

1.1. Electrochemical Mechanism of Platinum Dissolution

Platinum dissolution in PEMFCs occurs through two main electrochemical pathways: direct electrochemical dissolution and oxidation-induced dissolution. When the electrode potential exceeds 0.9 V vs RHE, platinum is oxidized, forming platinum oxide (PtO) as an intermediate species (Equation 3). Under dynamic load conditions, where the voltage fluctuates frequently, the PtO layer dissolves, releasing soluble Pt^{2+} ions into the ionomer phase (Equation 4). At even higher potentials, particularly above 1.188 V vs RHE, platinum undergoes direct oxidation to Pt^{2+}, which further accelerates the dissolution process (Equation 5)[28].

$$Pt + H_2O \rightarrow PtO + 2H^+ + 2e^- \quad E^0=0.8\text{-}0.95V \text{ vs RHE} \quad (3)$$

$$PtO + 2H^+ \rightarrow Pt^{2+} + H_2O \quad E^0 = 0.98V \quad (4)$$

$$Pt \rightarrow Pt^{2+} + 2e^- \quad E^0 = 1.188V \quad (5)$$

1.2. Ostwald Ripening and Nanoparticle Growth

Ostwald ripening is a secondary degradation mechanism observed in PEMFCs, typically occurring after the dissolution of platinum. This process is driven by differences in chemical potential, as smaller platinum nanoparticles have higher surface energy, and are therefore more susceptible to dissolution and reprecipitation under dynamic operating conditions[29]. The resulting Pt^{2+} ions migrate through the ionomer phase and preferentially redeposit onto larger particles, which leads to catalyst coarsening[29], [30].

The rate of this process is influenced by several factors, including high cathode potential (greater than 0.9 V vs. RHE), frequent voltage cycling within 0.6–1.0 V range, elevated temperatures (above 80°C), and high water content in the ionomer, all of which increase Pt ion mobility and redistribution[31], [32]. Over time, this results in a gradual change in the particle size distribution, leading to increased aggregation, a loss of ECSA, and reduced catalytic efficiency, ultimately accelerating the overall performance degradation of PEMFCs[9].

1.3. Impact of Platinum dissolution and redeposition on the PEMFCs

Platinum dissolution and Ostwald ripening significantly contribute to the loss of ECSA in Proton Exchange Membrane Fuel Cells (PEMFCs), thereby reducing the number of active sites for the ORR and increasing activation overpotential[24]. Platinum dissolution leads to the formation of platinum bands within the membrane, which hinder proton conductivity and elevate oxygen transport resistance, resulting in irreversible catalyst loss as material migrates to non-catalytic regions. In contrast, Ostwald ripening leads to catalyst coarsening through the redeposition of dissolved Pt onto larger particles, reorganizing the catalyst structure into less reactive clusters and reducing both dispersion and activity[1], [33], [34]. Consequently, these degradation pathways shorten the operational lifespan of PEMFCs, elevate maintenance costs, and present significant challenges to their long-term commercial viability[35].

2. Carbon Support Corrosion

Carbon support corrosion is a critical degradation mechanism in the cathode catalyst layer (CCL) of PEMFCs. The carbon support, typically made of high-surface-area carbon black (e.g., Vulcan XC-72 or Ketjenblack), serves as the substrate for Pt nanoparticles, providing electrical conductivity and structural integrity to the catalyst layer[36], [37]. However, under harsh operating conditions, such as high electrode potentials (greater than 1.0 V vs. RHE) and low humidity, the carbon can undergo electrochemical oxidation, gradually weakening its support structure[33]. Ultimately, this degradation compromises catalyst stability and contributes to the gradual decline in PEMFC performance over time[38].

2.1. Electrochemical Mechanism of Carbon Corrosion

Carbon support corrosion is primarily driven by electrochemical oxidation at elevated electrode potentials (greater than 1.0 V vs. RHE), particularly during start-up, shut-down, and transient load conditions[28]. Under these scenarios, the carbon support reacts with water, to form carbon dioxide (CO_2), as shown in Equation 6, which weakens the CL's structural integrity[39], [40], [41]. Unlike platinum dissolution, which primarily reduces the number of active catalytic sites, carbon corrosion weakens the entire catalyst support structure, leading to Pt nanoparticle detachment and loss of electrical connectivity[13], [14], [20], [42].

$$C + 2H_2O \rightarrow CO_2 + 4H^+ + 4e^- \quad E^0 = 0.207V \quad (6)$$

2.2. Impact of Carbon Corrosion on PEMFC Performance

The degradation of the carbon support has a profound impact on PEMFC durability and efficiency, primarily through the progressive loss of rated power output due to decreased electrical connectivity[21]. As carbon oxidation advances, the mechanical integrity of the CCL deteriorates, leading to structural collapse and reduced adhesion to the PEM[43]. This weakening accelerates Pt nanoparticle detachment, reducing the ECSA and hindering ORR kinetics[21]. Furthermore, the breakdown of the porous carbon network restricts oxygen diffusion to active sites and disrupts the electrical pathways within the catalyst layer[44]. These effects not only increase ohmic resistance but also exacerbates voltage losses, particularly under dynamic operating conditions[45].

As a result, the fuel cell's ability to maintain its maximum rated power output is compromised, necessitating additional Pt and carbon loading to maintain long-term performance. For instance, a 1 kW-rated fuel cell may only be able to generate 950 W after prolonged operation due to reduced conductivity. This degradation is particularly problematic during transient operations, where batteries are often deployed to buffer voltage fluctuations and power losses. The combined effects of catalyst loss, structural degradation, and mass transport limitations make carbon corrosion a critical failure mode affecting the long-term performance and stability of PEMFCs[43].

3. Nanoparticle Agglomeration and Sintering

Agglomeration and sintering are significant degradation mechanisms in PEM fuel cells. These processes occur when platinum nanoparticles cluster together due to high operating temperatures, transient load cycling, and fluctuations in electrochemical potential[46]. As a result, this clustering reduces the number of active sites available for the ORR[25], [47], [48], [49], ultimately lowering fuel cell efficiency and durability[5], [50], [51].

3.1. Electrochemical Mechanism of Nanoparticle Agglomeration and Sintering

Nanoparticle agglomeration and sintering in PEMFCs occur through distinct but related mechanisms that involve the migration, clustering, and coalescence of Pt nanoparticles under electrochemical and thermal stress conditions[34]. Agglomeration is primarily initiated by electrostatic interactions, van der Waals forces, and poor catalyst dispersion, which lead to the formation of loosely bound Pt clusters on the carbon support[10], [52], [53], [54], [55]. At high operating temperatures and during potential cycling, these clusters migrate and fuse, accelerating the sintering process[39].

Sintering occurs when Pt nanoparticles physically merge due to surface diffusion and atomic rearrangement, forming larger, less active structures. Unlike Ostwald ripening, which involves dissolution and redeposition, sintering is irreversible and leads to a substantial reduction in catalytic efficiency and a gradual decline in the rated power output of the fuel cell over time[40].

3.2. Impact of Nanoparticle Agglomeration and Sintering on PEMFC Performance

Nanoparticle agglomeration and sintering degrade the performance and durability of PEMFCs by reducing the ECSA and limiting catalyst utilization[41]. As platinum nanoparticles merge into larger clusters, the number of active sites available for the ORR declines, increasing activation overpotential and reducing catalytic efficiency[56]. Additionally, agglomeration disrupts catalyst dispersion, hindering oxygen transport and creating localized oxygen-depleted regions that further impair ORR kinetics. The densification of the catalyst layer also restricts reactant accessibility and raises ohmic resistance, intensifying mass transport limitations. These combined effects accelerate performance loss, leading to reduced power output, lower long-term efficiency, and compromised operational stability of PEMFCs.

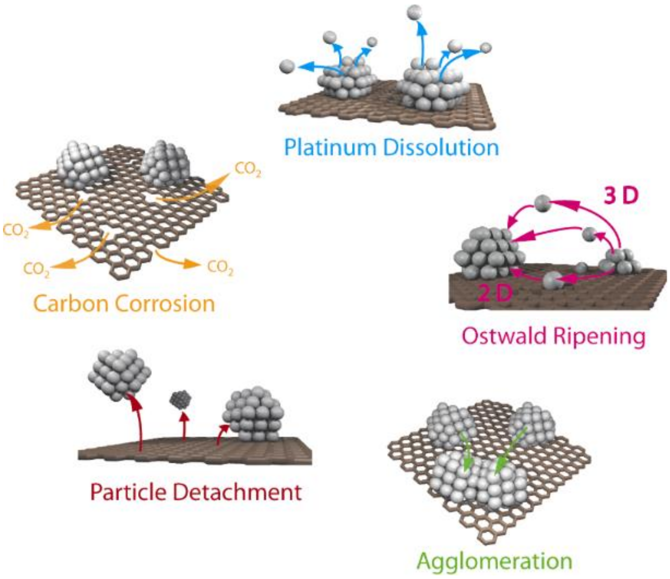

Fig. 3: Simplified representation of suggested degradation mechanisms for platinum particles on a carbon support in fuel cells[57].

V. TECHNIQUES FOR INVESTIGATING CL DEGRADATION

The degradation of the catalyst layer significantly affects the durability of PEMFCs[58]. Enhancing fuel cell longevity requires a thorough understanding of the degradation mechanisms under varying operating conditions[10]. This understanding is essential not only for identifying the sources of degradation but also for developing effective mitigation strategies. Achieving this goal involves a systematic investigation using experimental approaches that replicate real-world conditions and evaluate catalyst performance over time, as illustrated in Table 1. These investigations are complemented by advanced characterization techniques that analyze degradation at both microscopic and atomic scales, offering insights into key mechanisms such as active site loss, nanoparticle aggregation, and surface chemistry changes[59]. A comprehensive exploration of these mechanisms helps identify critical degradation pathways, which in turn support the development of more durable catalysts and effective mitigation strategies. The following section will explore these investigation techniques in detail, highlighting their role in uncovering underlying degradation processes and improving PEMFC performance and longevity.

1. Experimental Approaches

1.1. Accelerated Stress Test

Accelerated stress tests play a critical role in PEM fuel cell research by enabling the evaluation of catalyst durability under controlled operating conditions[10]. Rather than relying on prolonged operational testing, ASTs apply intensified stressors, such as voltage cycling, temperature fluctuations, and chemical contamination, to replicate long-term degradation within a significantly shorter timeframe [60]. This approach allows researchers to rapidly identify performance loss and key failure mechanisms in platinum-based catalysts, which would otherwise take thousands of operational hours to observe under normal conditions[61].

Several experimental studies have demonstrated the effectiveness of ASTs in evaluating ECSA loss and identifying catalyst degradation patterns under accelerated conditions. For example, Sharma et al.[62] applied a square wave voltage AST protocol and reported a 72% decline in ECSA after 14,000 cycles, attributing this primarily to nanoparticle coalescence and Ostwald ripening. Their findings provided early quantitative evidence of how aggressive cycling protocols can replicate long-term degradation mechanisms. Building on this, Birkner et al. [50] introduced a dynamic AST applied to a complete MEA system, observing a 40% ECSA loss within just 1,000 hours. This study extended the understanding of AST impacts by including full assembly effects and dynamic load stresses. Schneider et al.[51], further contributed by systematically investigating the role of Pt loading and catalyst layer thickness under voltage cycling conditions. Their results demonstrated that lower loadings and thinner CLs increase Pt dissolution and ECSA loss, emphasizing the trade-offs in material design. Together, these studies highlight the evolution of AST application, from single-variable protocols to more system-representative testing and underscore their role not only in material screening, but also in shaping real-world design strategies.

Despite their usefulness, ASTs have several limitations when it comes to accurately predicting the degradation behaviour of PEMFCs in real-world scenarios[63]. In practical applications, fuel cells operate under complex and dynamic conditions, including fluctuating loads, varying temperatures, humidity levels, and exposure to contaminants, which ASTs often struggle to fully replicate. Most AST protocols isolate individual stress factors, which can lead to either overestimating or underestimating degradation rates compared to those observed during real-world operation[64]. For example, Khedekar et al.[47] demonstrated that while ASTs provide valuable insights into the effects of humidity and gas feed conditions on catalyst degradation, they do not adequately capture the multi-factor interactions that occur in real-world fuel cell systems. Another critical issue is the lack of repeatability of ASTs; even tests conducted under nearly identical conditions can yield significantly different degradation rates. For instance, a study comparing two fuel cell stacks tested under the same conditions, except for slight variations in relative humidity ($\varphi_{air, in,1} = 70\%$ vs. $\varphi_{air, in,2} = 50\%$), showed markedly different degradation rates (471µV h^{-1} vs. 244µV h^{-1}). Such sensitivity to minor parameter changes complicates internal comparisons within the same study[48]. Beyond repeatability, reproducibility across laboratories also remains a major challenge. The absence of standardized AST protocols limits cross-study comparisons and hinders the development of generalizable degradation models[18], [19]. This challenge is further compounded by variations in test setups, material sources, and operating conditions, all of which introduce additional inconsistencies and make it difficult to draw universal conclusions. As highlighted in recent reviews, current AST protocols have been successfully adopted for durability testing, but their application for system lifetime prediction is still limited due to persistent difficulties in aligning accelerated and real-world aging conditions[65]. Although some studies have attempted to address this by adapting load cycling protocols to better mimic real operational profiles, these efforts still lack robust validation. Finally, the nonlinear relationship between AST conditions and actual degradation rates introduces additional uncertainty in lifetime prediction models[10], [52], [53], limiting their effectiveness in defining clear benchmarks for catalyst reliability and durability[54]. Addressing these issues calls for both improved AST design and a deeper understanding of how multiple stressors interact to better replicate real-world fuel cell degradation.

1.2. Life Test

Life tests are widely considered the most reliable approach for assessing the long-term durability of PEM fuel cells. In contrast to accelerated stress tests, which focus on specific degradation mechanisms, life tests expose the fuel cell system to complex, real-world conditions, such as load cycling, temperature variations, and humidity fluctuations. This exposure enables the observation of gradual catalyst degradation over extended periods, providing valuable insights into long-term performance loss under practical usage scenarios[55].

Several key studies have demonstrated the effectiveness of life testing in identifying catalyst degradation patterns in PEMFCs. For example, Hengge et al.[66], conducted a systematic life test on MEAs, periodically interrupting operation to evaluate the progression of degradation. Their analysis revealed a gradual loss of ECSA and thinning of the CL, primarily due to platinum dissolution and nanoparticle coalescence. Notably, the study found that structural changes increased over time, indicating a nonlinear degradation trajectory that short-term testing could not adequately capture. These findings highlight the importance of high-resolution diagnostics in life testing to reveal the sequence and intensity of degradation mechanisms. In another study, He et al.[67] provided further insights by conducting life tests at 100, 225, and 650 hours under constant conditions (80 °C, 120 kPa, 80% RH). Their results confirmed a two-stage degradation pattern. In the initial phase (0–100 hours), Pt agglomeration was the dominant mechanism, accounting for 78%-80% of total ECSA loss. In later stages (225–650 hours), proton connectivity decay and ionomer degradation became more dominant, contributing to nearly half of the observed performance drop. This shift reveals the evolving nature of degradation mechanisms as MEAs age. In a broader context, Schneider et al. [68] emphasized that long-term degradation is influenced not only by operating time but also by the structural and compositional features of the CL, such as Pt loading and layer thickness. Their comparative analysis demonstrated that thinner CLs and lower Pt loadings experienced faster ECSA loss under extended cycling, underscoring the need to optimize both material design and operating protocols for durability. Together, these studies demonstrate the critical role of life tests in accurately capturing degradation pathways over time and informing the development of PEMFCs with enhanced long-term durability[69].

Despite their reliability in simulating real-world conditions[19], [70], life tests face several limitations. One major issue is poor repeatability, as even slight variations in parameters such as cathode pressure, temperature, or humidity can significantly affect degradation outcomes. Inconsistencies in test bench environment, cell construction, and operational history further contribute to variable results[71]. Additionally, reproducibility across laboratories is limited due to the absence of standardized protocols, making it difficult to compare degradation rates or draw general conclusions. Furthermore, most testing procedures struggle to distinguish between reversible and irreversible degradation effects, and there is currently no universal method to quantify reversible losses or optimizing recovery strategies[71].

Beyond the technical challenges, life tests are also constrained by their long duration and high costs. These factors restrict their scalability for large-scale implementation and delay the process of material screening and fuel cell optimization[72]. To overcome these limitations, it is recommended to adopt an integrated degradation assessment approach, that combines ASTs for rapid evaluation with life tests for real-world validation. This hybrid strategy ensures a balance between accuracy in degradation assessment and practical feasibility in testing[73]. Additionally, the adoption of standardized testing protocols is essential to improve result comparability and enhance the predictive accuracy of long-term degradation models.

Table 1: Experimental Approaches for Investigating CL Degradation

Technique	Advantages	Disadvantages	References
Accelerated Stress Tests	Rapid evaluation; identifies degradation mechanisms quickly; useful for material screening.	Does not fully replicate real-world conditions; lacks standardized testing protocols.	[60], [62]
Life Tests	Provides the most realistic degradation trends; essential for long-term fuel cell optimization.	Time-consuming and costly; impractical for large-scale material screening.	[10], [29], [52], [53], [66], [68], [70]

2. Characterization Techniques

The CL plays a vital role in facilitating electrochemical reactions in PEM fuel cells; however, it remains highly susceptible to degradation, which adversely affects both performance and fuel cell longevity. To study these degradation processes, a variety of characterization techniques have been used to monitor structural and chemical changes within the CL. Recent studies have classified these diagnostic approaches into two main categories: electrochemical techniques and microscopic/spectroscopic techniques

2.1. Electrochemical Characterization

2.1.1. Cyclic Voltammetry

Cyclic voltammetry is a widely used technique for quantifying the ECSA, making it essential for characterizing platinum catalyst degradation in PEMFCs. This method calculates ECSA by integrating the peaks of hydrogen adsorption and desorption, which reflects the number of available active catalytic sites. For example, He et al.[67], applied CV during 650 hours of dynamic operation and identified a two-stage degradation pattern. They observed a rapid 40% loss in ECSA within the first 200 hours due to Pt agglomeration, followed by a slower decline associated with a loss of proton connectivity. Similarly, Wang et al.[73] reported a 52% decrease in ECSA under simulated bus operation, which they attributed to Pt dissolution and electrochemical Ostwald ripening. In both cases, CV effectively captured the degradation trend. However, since CV cannot differentiate between specific degradation mechanisms, such as carbon corrosion versus Pt loss, it is often necessary to complement it with structural diagnostics like transmission electron microscopy (TEM) or X-ray diffraction (XRD) for more accurate interpretation of CL deterioration[45].

2.1.2. Electrochemical Impedance Spectroscopy

Electrochemical impedance spectroscopy is a powerful diagnostic tool for analysing performance degradation in PEMFCs CLs. It enables the separation of electrochemical phenomena by quantifying changes in charge transfer resistance (R_{ct}), mass transport resistance (R_{mt}), and ohmic resistance (R_{Ω}) under real or accelerated operating conditions. These parameters help identify whether performance loss originates from catalyst degradation, carbon corrosion, or mass transport limitations. For instance, Meyer et al.[74] used EIS under accelerated stress tests to distinguish between catalyst and carbon support degradation. In catalyst-degraded cells, a gradual increase in R_{ct} at high current densities was observed, rising by 33% after 30,000 cycles, indicating reduced catalytic activity due to Pt nanoparticle dissolution and agglomeration. In contrast, the carbon-corroded cells displayed a rapid increase in R_{ct} (up to 225%) along with a six-fold rise in R_{mt} after 500 cycles, indicating pore structure collapse and compromised gas transport pathways. Similarly, He et al. [67] applied EIS to investigate catalyst degradation over 650 hours of dynamic operation. Their data showed a significant rise in cathode impedance, primarily due to carbon support corrosion, which disrupted electronic connectivity within the CL. Equivalent circuit modelling revealed that R_{ct} steadily increased over time, aligning with reductions in ECSA and indicating that catalyst particle sintering and detachment were primary degradation drivers. Furthermore, Schneider et al.[68] performed high-resolution impedance measurements under varying humidity and load conditions. Their results demonstrated that R_{ct} and mass transport behaviour strongly depend on CL structure. They also emphasized that accurate EIS modelling must account for through-plane inhomogeneities, especially in high-performance and low-Pt-loaded configurations.

Despite EIS being an indispensable tool for quantifying degradation kinetics and identifying degradation origins of performance loss in PEMFC catalyst layers, interpreting EIS data remains challenging. Accurate analysis requires sophisticated equivalent circuit modelling and careful separation of overlapping resistance contributions[73].

2.1.3. Linear Sweep Voltammetry

Linear sweep voltammetry is a commonly used technique in the diagnostic of PEMFC, particularly for assessing hydrogen crossover and membrane degradation. Although LSV does not directly evaluate catalyst degradation, it provides critical insights into the structural integrity and gas barrier properties of the membrane-electrode assembly, which are closely related to long-term catalyst stability.

Kocha et al. [75] conducted one of the earliest and most comprehensive studies on gas crossover using LSV. Their results showed that hydrogen crossover currents increased with temperature and pressure, indicating that thinner membranes are more susceptible to gas permeation. This increased crossover not only leads reduces fuel cell efficiency but also contributes to chemical degradation mechanisms that accelerate the deterioration of the cathode catalyst layer. The study highlighted LSV's sensitivity to membrane thinning and its importance in detecting early signs of structural failure, such as gas leakage and reduced performance.

Additionally, Pivac et al. [76] employed LSV as part of a broader electrochemical diagnostic suite during accelerated stress testing. Their findings revealed that hydrogen crossover remained relatively stable throughout the test period, suggesting that the observed degradation was primarily within the catalyst layer rather than the membrane. However, the inclusion of LSV allowed the researchers to monitor membrane integrity in real-time and confirm that performance losses were not due to increased permeability. This application demonstrates LSV’s utility as a safeguard when studying catalyst degradation under operational stress.

Similarly, Schneider et al. incorporated LSV into a multi-modal in-situ diagnostic protocol to investigate catalyst degradation across 36 different material configurations. Their use of LSV provided essential information about the influence of ionomer thickness and composition on membrane durability and crossover behaviour[68]. Cells with optimized ionomer distribution exhibited more stable crossover currents, which helped isolate the role of catalyst degradation in overall performance decline.

In summary, while LSV does not directly detect platinum dissolution or carbon support corrosion, it remains a critical complementary technique in CL characterizations. When combined with electrochemical impedance spectroscopy and cyclic voltammetry, LSV enhances the understanding of degradation mechanisms by identifying membrane-related factors that may indirectly affect catalyst performance. Its key strength lies in providing a non-invasive, real-time assessment of gas crossover and structural resilience, both of which are essential for long-term PEMFC durability.

2.2. Microscopic and Spectroscopic Techniques

2.2.1. Transmission Electron Microscopy

Transmission electron microscopy is a pivotal technique for visualizing platinum nanoparticle degradation at the nanoscale, providing detailed insights into mechanisms such as agglomeration, particle growth, and detachment. In a systematic post-mortem study, Prokop et al. used TEM to analyse catalyst layers operated at various voltages in high-temperature PEMFCs. Their study revealed that lower cell voltages accelerated Pt nanoparticle growth via electrochemical Ostwald ripening. TEM images showed a broader particle size distribution at lower voltages, indicating that operating voltage strongly affects degradation rate and morphology of Pt particles during fuel cell operation[77]. Similarly, Meng et al. employed TEM to study catalyst degradation under dynamic cyclic loading. They observed significant Pt agglomeration near the oxygen outlet, with average particle sizes grew from 2.67 nm to 4.78 nm after 2500 cycles. This growth corresponded with significant losses in ECSA and overall fuel cell performance. Their study also confirmed that Pt migration and detachment from the catalyst layer were intensified under fluctuating current conditions, reinforcing TEM's role in identifying spatial degradation patterns[78]. Despite its high resolution, TEM has limitations in degradation studies. Very small particles (~2 nm) may remain undetected due to poor contrast between Pt and carbon support, while overlapping particles can complicate interpretation. Additionally, TEM images represent only small sample areas (1–10^2 μm^2), which may not reflect the overall CL condition. Therefore, combining TEM with complementary techniques like SAXS or XRD is necessary for robust analysis of catalyst layer degradation[77].

2.2.2. Scanning Electron Microscopy

Scanning Electron Microscopy (SEM) is critical characterization technique used to assess morphological degradation in the catalyst layer due to its ability to provide cross-sectional imaging of structural features at the micron scale as shown in Fig. 4. It is particularly valuable for evaluating layer thinning, Pt particle migration, and support collapse that occur during prolonged operation or accelerated stress testing. For instance, Meng et al. used SEM under cyclic loading conditions and observed significant thinning of the CL at the oxygen outlet, highlighting the role of oxygen starvation in accelerating Pt dissolution and carbon corrosion[78]. Another study employed cross-sectional SEM to examine the impact of Pt loading and layer thickness on degradation. After 45,000 voltage cycles (0.6–0.95 V), low-Pt-loaded catalyst layers exhibited severe Pt depletion near the membrane interfaces. However, no major porosity changes were detected, suggesting that ECSA loss was not solely due to carbon corrosion[51]. These findings underscore SEM's utility in distinguishing between structural and electrochemical degradation pathways. Nevertheless, SEM cannot resolve nanoscale features or differentiate Pt particles from carbon support clearly, and its limited field of view may lead to sampling bias. Therefore, it is often used in conjunction with TEM or EDX for comprehensive characterization.[51].

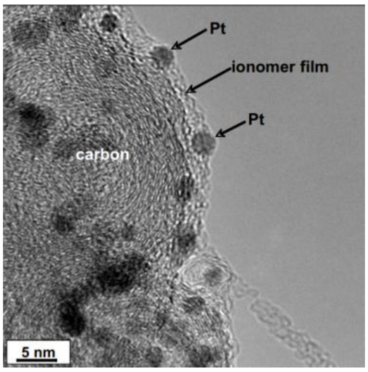

Fig. 4. Microstructure of Pt particle/ionomer/carbon support as shown by scanning electron microscope (SEM)[79]

VI. CATALYST LAYER DEGRADATION MODELLING

As discussed above, ensuring the long-term durability of PEM fuel cells remains a significant challenge in the advancement of hydrogen energy technologies[80]. It is now well established that degradation of the catalyst layer directly affects both the performance and lifespan of fuel cells[21]. However, conventional short- and long-term laboratory-based diagnostic degradation techniques are often costly, requiring significant investments in specialized equipment and skilled personnel. To address these limitations, Prognostics and Health Management (PHM) has emerged as a promising approach for assessing degradation mechanisms and estimating the remaining useful life (RUL) of CL in PEMFCs[52], [54], [55].

These prognostic models are typically classified into two main categories: model-based and data-driven approaches, as shown in Fig. 5 and Table 2 [81]. Model-based techniques rely on physics-based equations to simulate electrochemical reactions and structural changes within the CL [82]. In contrast, data-driven approaches use experimental datasets and machine learning algorithms to identify degradation patterns and predict performance loss[83].

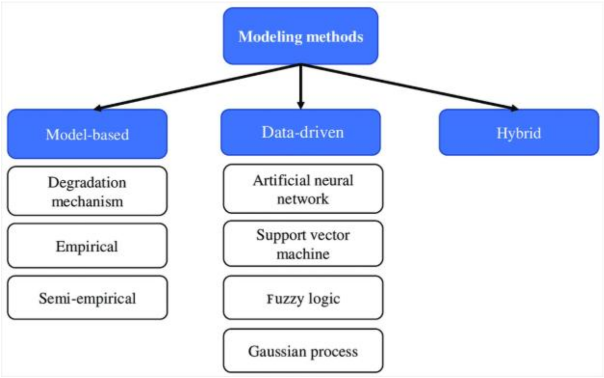

Fig. 5. Degradation Modelling Methods[84]

1. Model-Based

Physics-based models, also known as mechanistic models, utilize fundamental physical principles, including electrochemical kinetics, thermodynamics, and mass transport equations, to describe the degradation processes occurring in the CL of PEMFCs[85]. These models help identify key degradation mechanisms, such as platinum dissolution, Ostwald ripening, and carbon corrosion, while also predicting which regions within the catalyst layer are most susceptible to failure under specific operating conditions.[86].

Early physics-based models primarily focused on capturing platinum dissolution dynamics. Darling and Meyers[87] developed the first numerical degradation model for Pt/C catalysts, using a one-dimensional transient framework to simulate the ESCA loss due to platinum dissolution, redeposition, and ion transport within the MEA. However, this model did not account for other key degradation mechanisms, such as the platinum "band" formation mechanism, where dissolved platinum migrates into the membrane and precipitates, leading to catalyst layer thinning and performance degradation. To address this limitation, Bi and Fuller et al. [88] extended the model by incorporating this mechanism. Their model improved the predictions of cathode platinum mass, catalyst particle size, and platinum surface area, thereby enhancing the understanding of catalyst degradation.

Despite these advancements, early models remained limited in capturing the full complexity of catalyst degradation, as they could not simultaneously account for atomic-scale dissolution, particle evolution, and macroscopic transport phenomena. To overcome these challenges, researchers have developed multiscale models that integrate degradation mechanisms across various length and time scales. For example, Moore et al.[89] introduced a multiscale framework coupling a one-dimensional micro-scale ionomer-filled agglomerate model with a two-dimensional macro-scale MEA model. This approach enabled a more comprehensive analysis of catalyst layer behaviour, including its effects on charge, mass, and kinetic transport phenomena. Their model provided insights into how microscale agglomerate properties influence local catalyst effectiveness and current density distribution while considering macroscopic effects within the MEA. Another significant multiscale modelling approach was introduced by Jahnke et al.[90], who developed a coupled 2D performance and multiscale catalyst degradation model for PEMFCs. Their framework addressed key degradation mechanisms, including platinum oxidation, dissolution, particle growth, and platinum band formation. By incorporating spatially resolved catalyst degradation and ECSA changes under varying operating conditions, including accelerated stress tests, their model improved predictive accuracy and provided deeper insights into the catalyst aging process.

Recent advancements have further refined these models by incorporating environmental factors such as temperature, humidity, and voltage cycling, which were previously overlooked. For instance, Kregar et al.[91], developed a temperature-dependent model that demonstrates how platinum dissolution is more prevalent at lower temperatures, while carbon corrosion becomes increasingly significant at higher temperatures. Their study highlights the importance of thermal effects on catalyst layer degradation, which is essential for optimizing operating conditions. In addition to temperature effects, researchers have also focused on the impact of voltage cycling on catalyst layer degradation. Li et al.[92] developed a model that predicts catalyst layer degradation during startup and shutdown cycles by linking carbon corrosion to structural changes in the catalyst layer. Their findings indicate that rapid voltage fluctuations can accelerate carbon loss, resulting in a reduction of the ECSA and overall performance deterioration. By integrating these environmental factors, modern models offer a more holistic view of catalyst layer degradation, enhancing the ability to predict and mitigate catalyst deterioration under real-world operating conditions.

Despite these advancements, physics-based models encounter several challenges that restrict their predictive accuracy and practical applicability. One of those limitations is their strong dependence on material-specific parameters, making them sensitive to small variations in material properties such as platinum particle size and carbon morphology. These variations can significantly alter degradation behaviour, as demonstrated by Ahluwalia et al.[93] and Goshtasbi et al.[94], complicating both model calibration and generalization across different PEMFC designs. In addition to this, repeatability and reproducibility in model-based analyses present other critical challenges. Physics-based degradation models often rely on assumptions or fitted parameters that vary across experimental setups, making it difficult to achieve consistent results under different conditions. For instance, Bi and Fuller[88] showed that their catalyst degradation model consistency failed to predict platinum loss due to uncertainties in Pt ion diffusion rates and unmodeled mechanisms like particle coarsening. Similarly, Dong et al.[95] reported significant deviations between simulation results and experimental observations when using oversimplified carbon corrosion model. Another significant challenge is that many physics-based models tend to analyse degradation mechanisms in isolation, overlooking their complex interdependencies. For example, some models primarily focus on platinum dissolution and redeposition[89], [90], while others emphasize carbon corrosion[95]. However, experimental evidence indicates that these degradation processes are highly interconnected: carbon corrosion accelerates the detachment of Pt nanoparticles, leading to irreversible catalytic activity loss[69], [96], while Pt dissolution alters the local electrode environment, exacerbating both carbon degradation and ionomer instability. Ignoring these feedback loops limits the predictive accuracy of such models, especially for long-term fuel cell performance. Beyond mechanistic limitations, the high computational cost of physics-based simulations presents a major barrier to real-world implementation. These models must solve non-linear partial differential equations governing mass transport, electrochemical reactions, and degradation kinetics across multiple spatial and temporal scales. As demonstrated by Kregar et al.[97], although such simulations provide high predictive accuracy, they require substantial computational resources, making them impractical for real-time fuel cell monitoring and control.

In response to these challenges, researchers are increasingly adopting data-driven modelling approaches that utilize experimental datasets to improve prediction accuracy and adaptability. Unlike physics-based models, which rely on simplifications and often struggle to capture the full range of degradation factors, data-driven methods extract patterns directly from the data. This reduces dependence on predefined assumptions and enhances the reliability of degradation predictions across diverse operating conditions[98].

2. Data-driven model

Data-driven models, often referred to as non-physics-based models, utilize experimental datasets to predict degradation in the catalyst layer of PEM fuel cells. In contrast to physics-based models, which rely on fundamental electrochemical equations and parameterized degradation mechanisms, data-driven approaches identify patterns and correlations directly from observed data, eliminating the need for explicit knowledge of the underlying physical processes[99]. This enables them to capture nonlinear relationships, accommodate real-world variability, and improve predictive accuracy in modelling PEMFC degradation.[100].

One of the earliest data-driven models for predicting CL degradation was developed by Maleki et al.[101]. Their study employed an artificial neural network (ANN) trained on experimental degradation data to forecast the loss of ECSA over time. The proposed model followed a two-step approach: first, inputs such as temperature, relative humidity, and potential cycling were used to estimate the Pt dissolution rate. Then, this predicted rate, along with ion diffusivity, were used to determine Pt mass loss, surface area loss, and Pt particle radius change, as illustrated in Fig. 6. The experimental data were obtained through accelerated degradation tests conducted using square-wave voltammetry between 0.87 V to 1.2 V. Degradation was characterized using cyclic voltammetry for ECSA measurement, inductively coupled plasma mass spectrometry (ICP-MS) for Pt dissolution quantification, and scanning electron microscopy with energy-dispersive spectroscopy (SEM-EDS) to evaluate Pt distribution across the membrane electrode assembly.

Maleki et al.'s results demonstrated that well-trained ANN models can accurately predict catalyst degradation, showing a strong correlation with experimental data. Their model achieved high reliability with minimal error (e.g., RMSE, MRE, MAE), and R^2 values exceeding 99.9% across all outputs.

While these results highlight the predictive potential of ANNs, they also raise concerns regarding the reproducibility and repeatability of such data-driven models. As highlighted by Nguyen et al.[84] , the performance of machine learning models in PEMFCs degradation prediction is highly sensitive to the quality and pre-processing of training data, model architecture, and validation strategies. Variations in data collection methods, experimental conditions, and feature selection can lead to inconsistencies in model performance when applied across different studies or real-world scenarios. Therefore, ensuring repeatability and generalizability in data-driven CL degradation modelling requires rigorous cross-validation, transparent reporting of training protocols, and the use of diverse, representative datasets.

To date, data-driven models have been predominantly applied at the system level in PEM fuel cell research, with a primary focus on predicting performance-related metrics such as power loss, voltage decay, and overall efficiency[74], [75], [76]. These models have also contributed to optimizing CL design and operating conditions, thereby improving catalyst utilization and electrochemical stability[102], [103]. However, despite their growing use, their application specifically for modelling catalyst layer degradation remains scarce. To the best of author’s knowledge, beyond the work of Maleki et al., no further studies have developed data-driven frameworks dedicated to understand and predict CL degradation. This gap highlights an important research opportunity, as leveraging data-driven models to predict CL degradation could provide deeper insights into failure mechanisms, enabling more accurate lifetime predictions and the development of more resilient fuel cell designs.

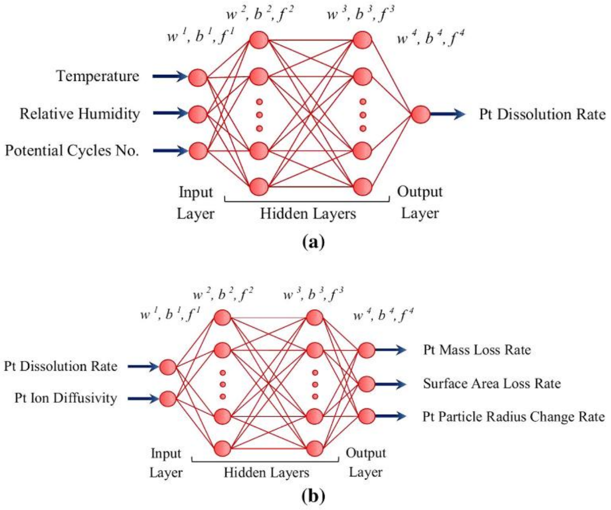

Fig. 6. Schematic ANN architecture for modeling of (a) Pt dissolution rate and (b) Pt mass loss rate, surface area loss rate, and Pt particle radius change rate[101].

Table 2: Comparison of Model-Based and Data-Driven Approaches for Catalyst Layer Degradation Modelling

Aspect	Model-Based Approach (Physics-Based)	Data-Driven Approach (Machine Learning)
Definition	Uses electrochemical and physical laws to simulate degradation mechanisms[85].	Relies on experimental datasets and statistical models to predict degradation trends[99].
Advantages	Provides detailed mechanistic insights into platinum dissolution and carbon corrosion[86].	Captures nonlinear dependencies, adaptable to different datasets, and requires less prior physical knowledge[100].
Disadvantages	Requires extensive parameterization, high computational cost, and is less adaptable to real-world variations[93].	Lacks physical interpretability and requires large, high-quality datasets for accuracy[101].

VII. PROSPECTS AND FUTURE DIRECTIONS

The durability of PEMFCs remains a critical challenge, particularly due to the degradation of the CL. Current research has focused on experimental and modelling approaches to understand degradation mechanisms and predict performance losses. However, significant gaps persist in both methodologies, which must be addressed to enhance the reliability of fuel cell lifetime predictions.

ASTs rapidly assess catalyst degradation under controlled extreme conditions but fail to replicate real-world operating environments, leading to discrepancies in performance predictions. In contrast, life tests provide accurate long-term degradation data but are costly and impractical for large-scale optimization.

To overcome these limitations, an integrated degradation assessment is proposed, combining ASTs for accelerated degradation assessment with strategically optimized life tests for validation, thereby balancing efficiency and accuracy. Instead of relying on full-duration life tests, this method periodically monitors key degradation indicators such as ECSA loss, catalyst thinning, and platinum dissolution through targeted checkpoints. These intermittent validations ensure that AST-derived trends remain representative of actual fuel cell behaviour. By balancing the speed of ASTs with the accuracy of real-world life tests, this approach enhances the reliability of degradation assessments while remaining practical for fuel cell research and development.

Current modelling techniques for predicting CL degradation in PEMFCs rely on two primary approaches: physics-based models and data-based models. Physics-based models describe degradation mechanisms using electrochemical and thermodynamic principles. However, they often involve simplifications and assumptions that limit their accuracy in real-world conditions. In contrast, data-driven models can analyse large experimental datasets to detect degradation trends but lack physical interpretability and struggle with limited CL-specific experimental data.

To address the limitations of current degradation models, a hybrid approach is proposed, integrating physics-based models for mechanistic accuracy with data-based models for predictive adaptability. Model-based outlines key degradation processes, while data-driven models, trained on AST and life test datasets, refine these predictions by capturing nonlinear interactions and operational dependencies, which are often missed in those simplified models. This hybrid modelling enables real-time self-correction, where data-driven dynamically updates physics-based estimates with experimental data, ensuring that degradation predictions are both accurate and physically interpretable.

Although no published studies to date have implemented hybrid modelling specifically for catalyst layer degradation in PEMFCs, promising results have been reported in the broader context of PEMFC degradation. For example, Zhou et al.[104] developed a multi-stage hybrid framework that combined a physical aging model with an autoregressive moving average (ARMA) method and a time-delay neural network (TDNN) to capture both linear and nonlinear aspects of fuel cell voltage degradation over time. Their model outperformed traditional physics-based and standalone ML approaches in long-term forecasting accuracy and robustness, particularly under complex degradation patterns. Similarly, Wang et al.[105] proposed a fusion prognostics framework that extracted degradation indicators from a physics-based model and used them as inputs to a symbolic long short-term memory (LSTM) network for predicting the remaining useful life (RUL) of PEMFCs. Their method achieved a low RUL prediction error (11.4%) and demonstrated strong performance across variable load conditions, confirming the practical promise of hybrid modelling approaches.

While these studies do not target CL degradation directly, they provide compelling evidence that combining mechanistic modelling with data-driven models improves prediction fidelity and generalizability. Many of the challenges encountered at the system level, such as nonlinear dependencies, sparse experimental data, and dynamic operating conditions, are equally present in CL degradation modelling. Therefore, adapting hybrid strategies to the CL scale is not only methodologically justified but potentially transformative. The encouraging outcomes reported in related PEMFC applications establish a strong foundation for future research aimed at developing hybrid models tailored to catalyst layer degradation, to enhance interpretability, reliability, and predictive power in fuel cell diagnostics.

Despite these advancements, further improvements are necessary to enhance the reliability, accuracy, and real-world applicability of CL degradation models. Future research should focus on further refining integrated degradation assessment methodologies and hybrid modelling approaches by expanding high-quality experimental datasets and improving model accuracy across different PEMFC operating conditions. Additionally, the integration of multi-scale modelling and advanced experimental validation will be crucial to ensuring that predictive models remain both computationally efficient and physically interpretable. Ultimately, the effectiveness of a hybrid model depends on the accuracy and resolution of the experimental data used to calibrate and validate it. This underscores the importance of advancing diagnostic techniques for catalyst layer degradation. Although electrochemical techniques (such as CV, EIS, and LSV) and microscopic or spectroscopic methods (such as TEM and SEM) are essential for characterizing CL degradation in PEMFCs, each technique presents distinct limitations that constrain their effectiveness. Electrochemical methods often provide global metrics like ECSA loss or resistance shifts but lack spatial resolution, making it difficult to pinpoint localized degradation or distinguish between overlapping degradation mechanisms. On the other hand, imaging techniques such as TEM and SEM offer nanoscale structural insight but are limited to ex-situ, static observations and are restricted to a narrow field of view and particle size range, which may prevent the full visualization of fine-scale degradation processes across the entire catalyst layer. In addition, both types of methods can suffer from limited repeatability due to sample variability, operator dependence, or uncontrolled environmental factors. To overcome these challenges, future research should prioritize the development of integrated diagnostic platforms that combine multiple complementary techniques in a synchronized and standardized framework. This includes improving the temporal resolution of electrochemical methods, enhancing the reproducibility of imaging protocols, and adopting data fusion strategies that correlate electrochemical signals with spatially resolved morphological changes. By advancing these techniques, the development of next-generation PEMFCs with enhanced durability and reliability will be accelerated, supporting broader commercialization efforts in automotive, stationary, and portable energy applications.

VIII. CONCLUSION

Catalyst layer degradation remains one of the most significant barriers to the long-term durability and commercial application of Proton Exchange Membrane Fuel Cells. This review has explored the major degradation mechanisms such as platinum dissolution, Ostwald ripening, carbon support corrosion, and nanoparticle agglomeration, along with the experimental and modelling approaches used to study them.

Electrochemically active surface area is the most commonly used metric to measure CL degradation, as it directly indicates the loss of active sites available for electrochemical reactions. However, based on the studies reviewed, there is no consistent ECSA value or range that can be generalized. For instance, some studies reported ECSA losses exceeding 70% after 14,000 cycles, while others observed around 40% loss after 1,000 hours of operation. Likewise, the dominant degradation mechanism reported also differs from one study to another. These variations are not only due to differences in test setups or materials, but also reflect the absence of standardized experimental protocols and the lack of general models to asses degradation reliability across different systems.

This lack of standardization not only limits experimental comparability but also creates a significant gap in current modelling approaches. Physics-based models can provide mechanistic insight into specific degradation phenomena, but they often require detailed system-specific input and are computationally intensive. In contrast, data-driven models offer faster prediction and broader applicability, but they typically lack physical interpretability and rely heavily on the quality of available data.

To address this gap, future research should focus on the adoption of hybrid modelling approaches that combine the strengths of both methods, integrating physical understanding with the flexibility of machine learning for CL degradation prediction. This should be supported by standardized testing frameworks and generalizable model architectures to improve consistency, accuracy, and reliability in degradation assessment. Advancing in this direction will be essential for improving PEMFC durability and enabling their broader adoption in hydrogen-based energy systems.

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