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Toward Hydrogen Mobility: Challenges and Strategies in Electrocatalyst Durability for Long-Term PEMFC Operation

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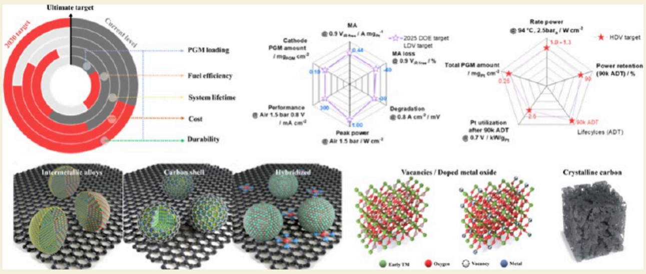

ABSTRACT: Proton exchange membrane fuel cells (PEMFCs) are emerging as a key technology in the transition to hydrogen-based energy systems, particularly for heavy-duty vehicles (HDVs) that face operational challenges, such as frequent startup-shutdown cycles and fuel starvation. However, the widespread adoption of PEMFCs has been limited by their durability and long-term performance issues, which are crucial for heavy-duty applications. This Perspective focuses on recent advancements in PEMFC catalysts and supports, with an emphasis on strategies to enhance their durability. We introduce Pt-based intermetallic catalysts, including Pt transition metal (TM) alloys, which offer improved stability and activity through regular atomic arrangements and strengthened metal−support interactions. Hybrid catalysts combining Pt with M-N-C (M = Fe, Co) have shown promise in boosting performance by enhancing the catalytic activity while reducing the platinum content. Moreover, stringent conditions must be met to meet the HDV requirements. Consequently, alternative support materials, such as metal oxides and graphitized carbons, have been introduced to enhance both the corrosion resistance and the electrical conductivity, thereby addressing the limitations of conventional carbon supports. Structural innovations and material advancements are essential for optimizing catalysts and supports to achieve long-term PEMFC performance. This Perspective provides a comprehensive overview of key developments in catalyst and support design, offering insights into current challenges and future directions for achieving durable and cost-effective PEMFCs.

KEYWORDS: *proton exchange membrane fuel cell, oxygen reduction reaction, electrocatalyst, catalyst stability, intermetallic, carbon shell, hybrid catalyst, catalyst support*

1. INTRODUCTION

With the growing emphasis on hydrogen energy as a solution to climate change, proton exchange membrane fuel cell (PEMFC) technology has attracted significant attention as a key technology.[1] PEMFC technology is versatile, with applications ranging from power plants to various types of vehicles, and it has already been commercialized in light-duty vehicles (LDVs), including the Hyundai Nexo and Toyota Mirai.[2,3] Despite the launch of these commercial models, sales remain low and have yet to drive significant growth in the hydrogen market. Currently, the main barriers include the limited availability of hydrogen refueling infrastructure and high hydrogen costs.[4] However, in the long term, catalyst durability is expected to become a critical challenge. In the context of complementary growth in battery technologies, PEMFCs are projected to become increasingly specialized for heavy-duty applications, including heavy-duty vehicles (HDVs), stationary power generation, aerospace, and vessels.[5] Consequently, research aimed at enhancing the durability of PEMFCs is expected to become increasingly important.

Ensuring the long-term performance of PEMFCs requires stable catalysts that can operate under harsh conditions such as high voltages and elevated temperatures, which is particularly crucial for HDVs compared to LDVs (Table 1).[5−8] In fuel cells, cathodic oxygen reduction reactions (ORRs) play a key role in determining the overall performance because of the slower reaction rate compared with anodic hydrogen oxidation reactions (HORs).[9] Although anodic catalysts face challenges such as resistance to poisoning (from CO, H_2S, SO_2, etc.) and cell reversal issues, this Perspective focuses on cathodic catalysts. Currently, Pt/C catalysts have been used as ORR catalysts in commercial PEMFCs due to their high activity but are expensive and have limited durability.[10] Researchers have focused on alloying Pt with transition metals (TMs) to adjust its electronic structure and increase the number of active sites, leading to enhanced activity.[11] However, the degradation of TMs, along with the corrosion of carbon supports, remains an

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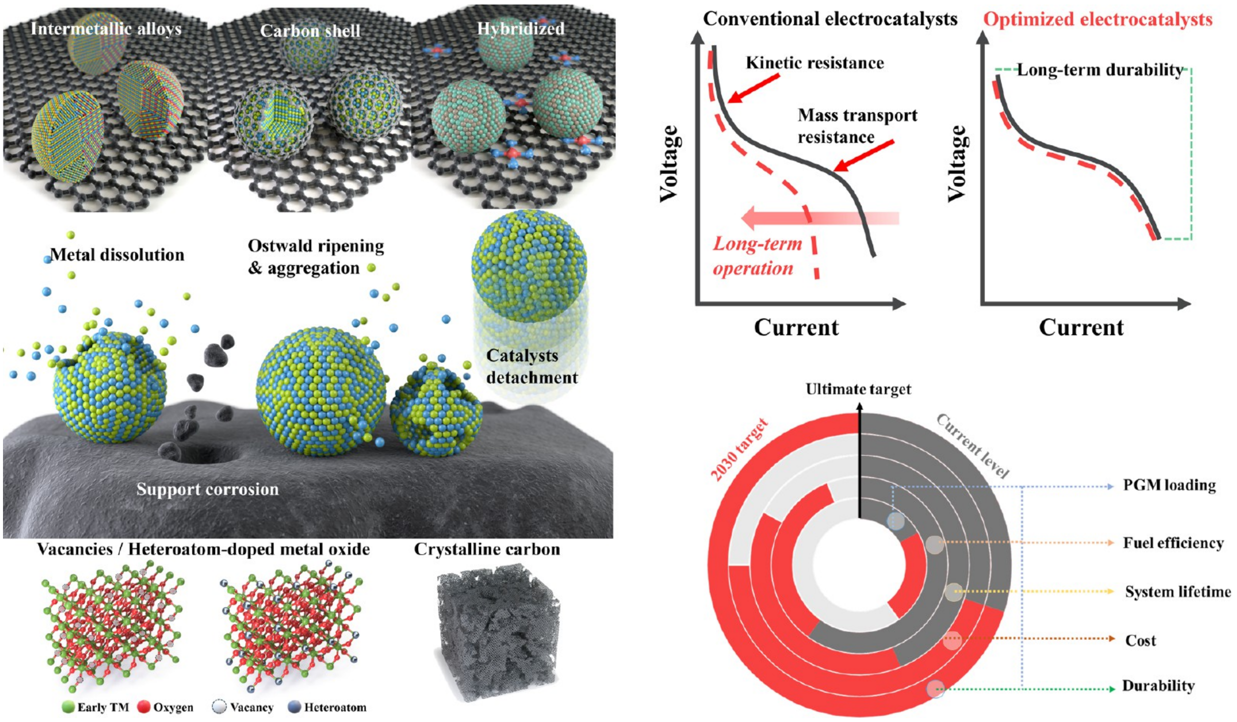
Table 1. Comparison of HDV and LDV Targets

	operation temperature (°C)	outlet pressure (kPa)	ADT cycles	rated power density (W cm^{-2})	Pt utilization after ADT @0.7 V (kW g_{Pt}^{-1})	power loss (%)	total PGM (mg_{Pt} cm^{-2})		
HDV targets	94	250	90,000	1.30	2.5	10	0.25		
	operation temperature (°C)	outlet pressure (kPa)	ADT cycles	current density @0.8 V air condition (A cm^{-2})	MA (A mg_{Pt}^{-1})	peak power (W cm^{-2})	0.8 A/cm^2 degradation (mV)	MA loss (%)	total PGM (mg_{Pt} cm^{-2})
LDV targets	80	150	30,000	0.3	0.44	1.00	−30	40	0.125

Scheme 1. Degradation Mechanisms in Electrocatalysts and Strategies for Enhancing Catalyst Durability, Representative *I*−*V* Polarization Curves of Conventional and Optimized Electrocatalysts under Long-Term Operation, and Radar Graph of Current Fuel Cell Technology Levels and DOE 2030 Targets Compared to DOE Ultimate Sector Targets[5]



issue.[12] As a result, next-generation PEMFC catalysts and support structures have been a subject of intensive research.[13] This Perspective aims to present key emerging technologies in this area.[13]

We first focused on Pt-TM intermetallic catalysts, which prevent TM dissolution through regular atomic arrangements and strong metallic bonding.[14] Recent studies on the synthesis of intermetallic catalysts, such as PtCo and PtNi alloys, have highlighted the use of small molecules to precisely control the particle size and structure, making them ideal for PEMFCs.[15] To further enhance the performance, research has expanded beyond binary intermetallic systems to explore diverse (intermetallic) alloying catalysts including ternary, high-entropy, and doped structures. Second, the carbon shell coating strategy plays a critical role in enhancing the fuel cell performance by protecting the catalyst particles and increasing their stability.[16] Thin carbon layers prevent the dissolution and agglomeration of catalyst particles, thus preventing long-term performance degradation. Third, hybrid-structured catalysts that combine Pt nanoparticles (NPs) with M-N-C (M = Fe, Co, etc.) support the catalytic activity and stability of pristine Pt.[17] These hybrid catalysts increase the ORR activity through strong interactions between the components and mitigate catalyst degradation through the spillover effect. This structure reduces the Pt content while maintaining a high performance, contributing to the economic efficiency of PEMFCs.

The development of stable and durable materials that can withstand PEMFC operating conditions is critical for enhancing the durability of fuel cells. Traditional carbon-based supports, although commonly used, face limitations in harsh operational environments, particularly in HDVs. Carbon supports undergo electrochemical corrosion during PEMFC operation, leading to the agglomeration and detachment of catalyst particles.[18] To address this issue, metal oxides and crystalline carbon supports are being explored as promising alternatives. Metal oxide supports, such as TiO_2, SnO_2, WO_3, and CeO_2, exhibit high corrosion resistance and strong metal−support interactions (SMSIs), inhibiting Pt particle migration and agglomeration and enhancing long-term stability.[19,20] The SMSI effect optimizes the *d*-band center of the catalyst by adjusting the electron-donating ability of the support, thereby lowering the energy barrier at the reaction sites and significantly enhancing the catalytic performance.[21] Additionally, crystalline carbon supports improve the structural durability and electrical conductivity through high-temperature treatments, thereby meeting the stringent durability require-

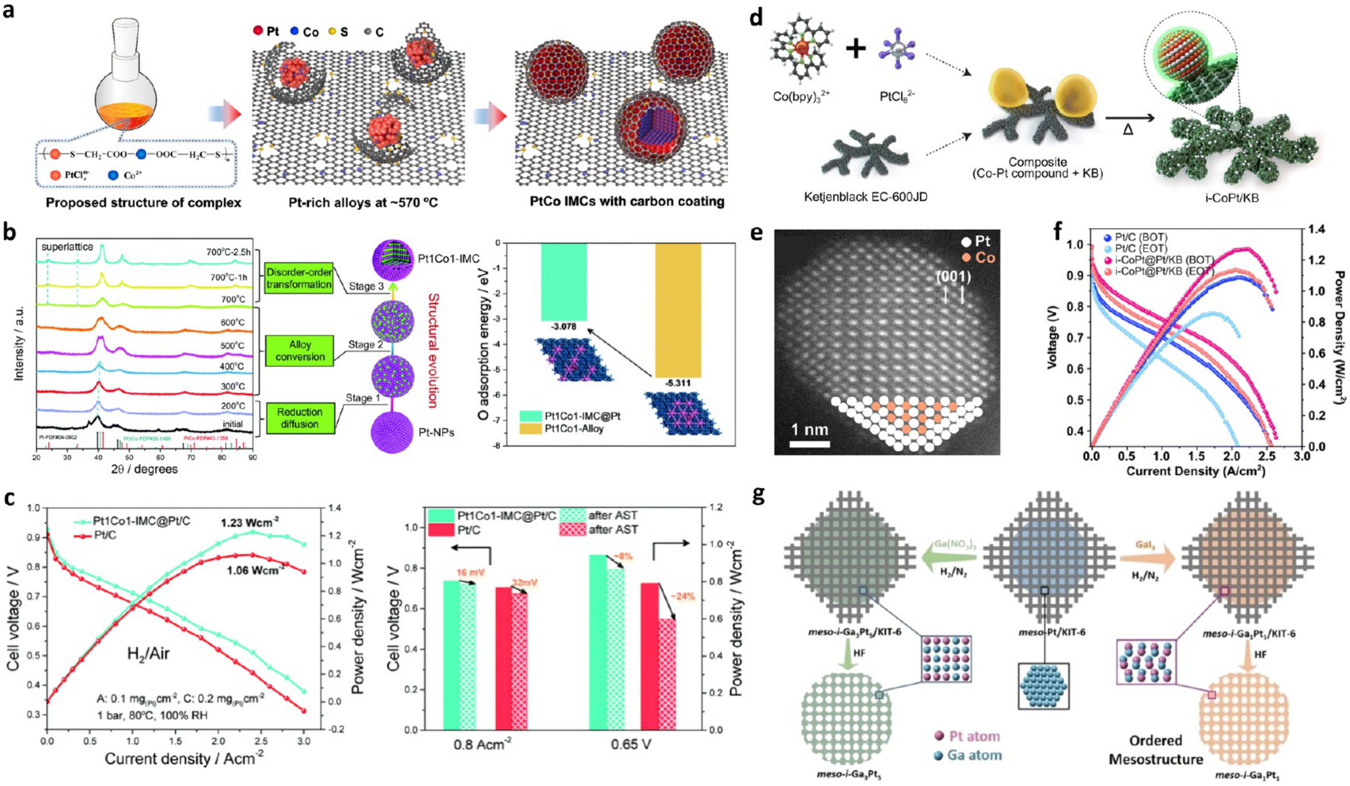


Figure 1. (a) Proposed antisintering mechanism for Pt_1Co_1-IMC@Pt/C. Reproduced from ref 27. Available under a CC-BY 4.0 license Copyright 2022. (b) Temperature/time-dependent ex situ X-ray diffraction (XRD) patterns of Pt_1Co_1-IMC@Pt/C catalyst during the annealing process with possible structural evolution mechanism and O adsorption energy of Co sites in the ordered (greenish cyan) and disordered (yellow) Pt_1Co_1 alloy structure. (c) Pt_1Co_1-IMC@Pt/C beginning of life *I*−*V* curves and degradation results (voltage drops and power density losses) after the ADT test. Reproduced with permission from ref 28. Copyright 2022, Royal Society of Chemistry. (d) Bipyridine-assisted scalable method of i-CoPt@Pt/KB. (e) Pt-skin layers and the internal atomic arrangement of Co and Pt. (f) PEMFC ADT tests of i-CoPt@Pt/KB. Reproduced with permission from ref 30. Copyright 2023, Royal Society of Chemistry. (g) Schematic illustration of mesoporous intermetallic GaPt nanoparticles control by different Ga salts. Reproduced with permission from ref 31. Copyright 2023, Wiley-VCH GmbH.

ments of PEMFCs.[22] These support materials are expected to significantly expand the commercial applicability of fuel cells by offering resilience to high temperatures and repeated load changes in HDVs.[23,24]

Although several reviews have been published on PEMFCs, particularly in the context of HDVs, most have focused on the associated market and system considerations. However, this Perspective addresses more technical aspects, focusing on the challenges and recent advancements in PEMFC catalysts and supports. Scheme 1 provides key degradation mechanisms that compromise catalyst performance during long-term operation, such as particle agglomeration, dissolution, and support corrosion. Additionally, it highlights strategies to mitigate these issues, including the use of advanced support materials, alloying, and surface modifications. The included representative *I*−*V* polarization curves compare the performances of conventional and optimized electrocatalysts. A radar graph contextualizes current fuel cell technology levels against the Department of Energy's (DOE) 2030 targets (red circles) and ultimate targets (gray circles) across key metrics such as price, efficiency, and lifetime, emphasizing the progress made and the remaining gaps to achieve these ambitious benchmarks.[5] By exploring the latest developments in catalyst durability, support materials, and strategies to overcome key performance barriers, this Perspective provides a comprehensive overview of the critical factors shaping the future of PEMFC technology.

2. CATALYSTS

2.1. Binary and Ternary Intermetallic Catalysts

Intermetallic catalysts composed of Pt and TMs such as Co and Ni are widely recognized as effective approaches for enhancing the durability and performance of PEMFC catalysts. Their long-range atomic ordering strengthens metal−metal bonding and reduces TM dissolution compared to that in disordered alloys.[14] Furthermore, the intermetallic structure usually downshifts the *d*-band center of Pt, thereby improving the ORR kinetics.[25] Owing to the demand for next-generation fuel cells with high power densities and long-term durability increases, interest in intermetallic catalysts is expected to increase. Despite their potential, several issues remain unaddressed, including the identification of optimal morphologies, development of mass-production methods, and prevention of aggregation during heat treatments.[26]

Among the various TMs, Co is suitable for Pt intermetallic catalysts because of its low dissolution rate. In addition, it can form an ordered face-centered tetragonal (FCT) structure instead of a disordered face-centered cubic (FCC) structure, which enhances the catalytic activity and durability. Intermetallic FCT PtCo can be synthesized using small-molecule-assisted techniques, which successfully control the particle size without sintering issues during high-temperature annealing (Figure 1a).[27] Furthermore, the size-controlled PtCo FCT showed resistance to oxidation and Co dissolution, as

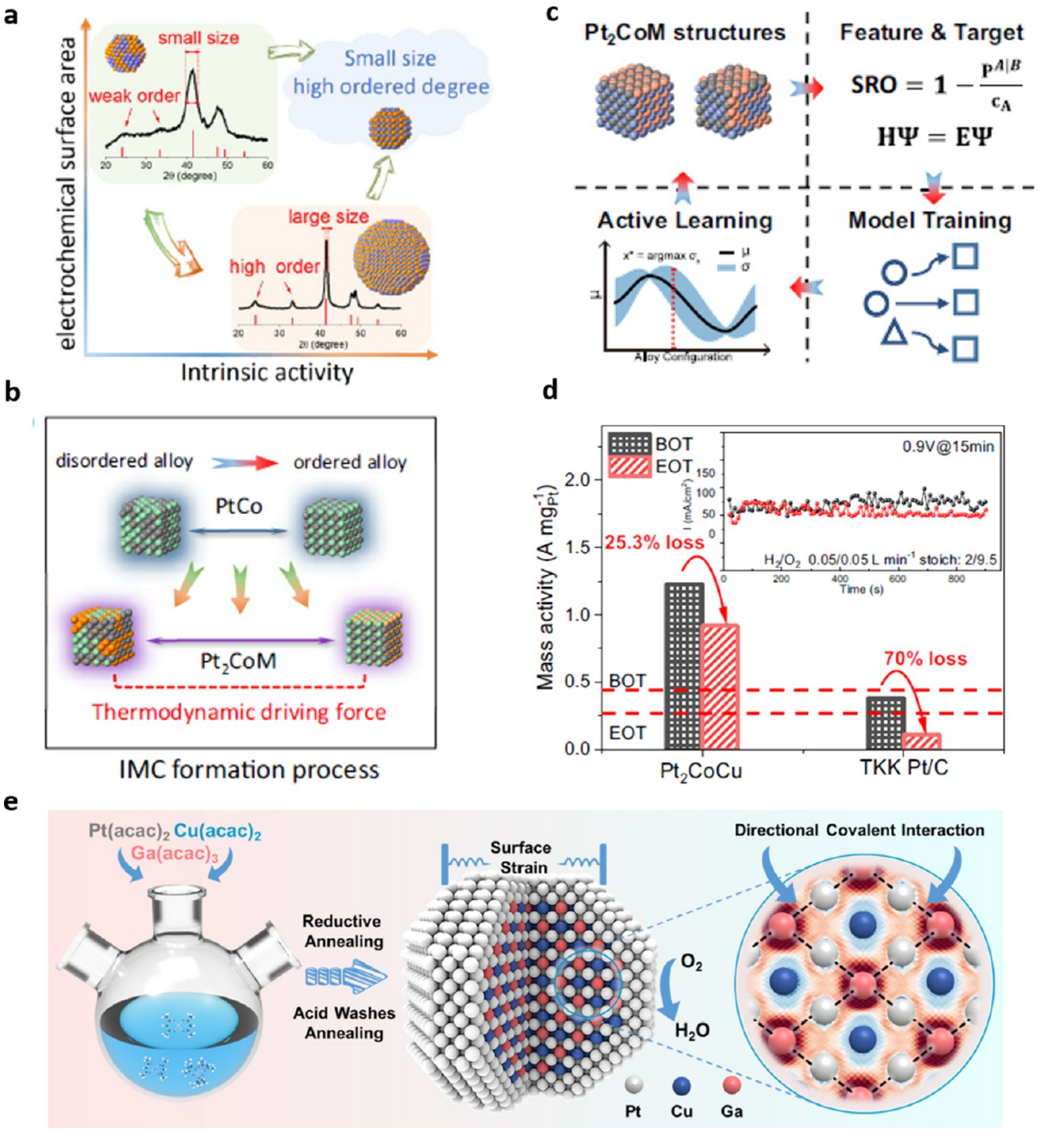


Figure 2. (a) The dilemma in a typical Pt-TM intermetallic catalyst synthesis. (b) Schematic illustration depicting the presence of a Pt_2CoM combination with an increased thermodynamic driving force for the disordered-to-ordered transition. (c) Active learning procedures to construct machine learning prediction model for the relative energy of Pt_2CoM. (d) The loss of MA during ADT. Reproduced from ref 32. Available under a CC-BY 4.0 license Copyright 2024. (e) Schematic illustration of synthesis of ternary intermetallic alloy through directional covalent interaction induced by Ga. Reproduced with permission from ref 34. Copyright 2023, Wiley-VCH GmbH.

supported by density functional theory (DFT) calculations (Figure 1b,c).[28] Moreover, the application of synthetic methods that enable large-scale production, along with excellent catalyst performance for PEMFCs, is a significant advantage.[29] Similarly, the incorporation of bipyridine ligands enables particle size control, thereby enhancing PEMFC performance (Figure 1d−f).[30] Regarding the structure, while most bimetallic intermetallic catalysts exhibit $L1_0$ and $L1_2$ FCT structures, researchers have sought to identify structures with improved activity and stability. Consequently, several interesting structures have been reported, including an orthorhombic Ga_5Pt phase. The orthorhombic intermetallic catalyst exhibited asymmetric atomic arrangements that increased the surface electron density and enhanced both the activity and the durability (Figure 1g).[31]

Beyond binary systems, introducing a third element is regarded as an effective strategy for precisely tuning the properties of binary intermetallic compounds to enhance the activity and durability of intermetallic catalysts.[7] With the growing interest in ternary systems, recent efforts have focused on identifying the optimal third element. To improve efficiency, machine learning (ML) techniques have been introduced to enable the rapid screening of thousands of combinations. Using ML, optimal compositions (e.g., Pt_2CoM (M= Cu, Ni)) were predicted based on thermodynamic stability and phase transition temperatures (Figure 2a−c).[32] The Warren−Cowley short-range order (SRO) is used to characterize the degree of chemical ordering, with a negative SRO indicating a preference for ordered configurations. Figure 2b shows that the introduction of a third element (M) increases the thermodynamic driving force for the disorder-to-order transition compared with binary PtCo, enhancing the nucleation of intermetallic phases. This additional stabilization energy from Co−M interactions promotes a highly ordered structure, improving both durability and performance. Figure 2c details the active learning procedures used to construct the

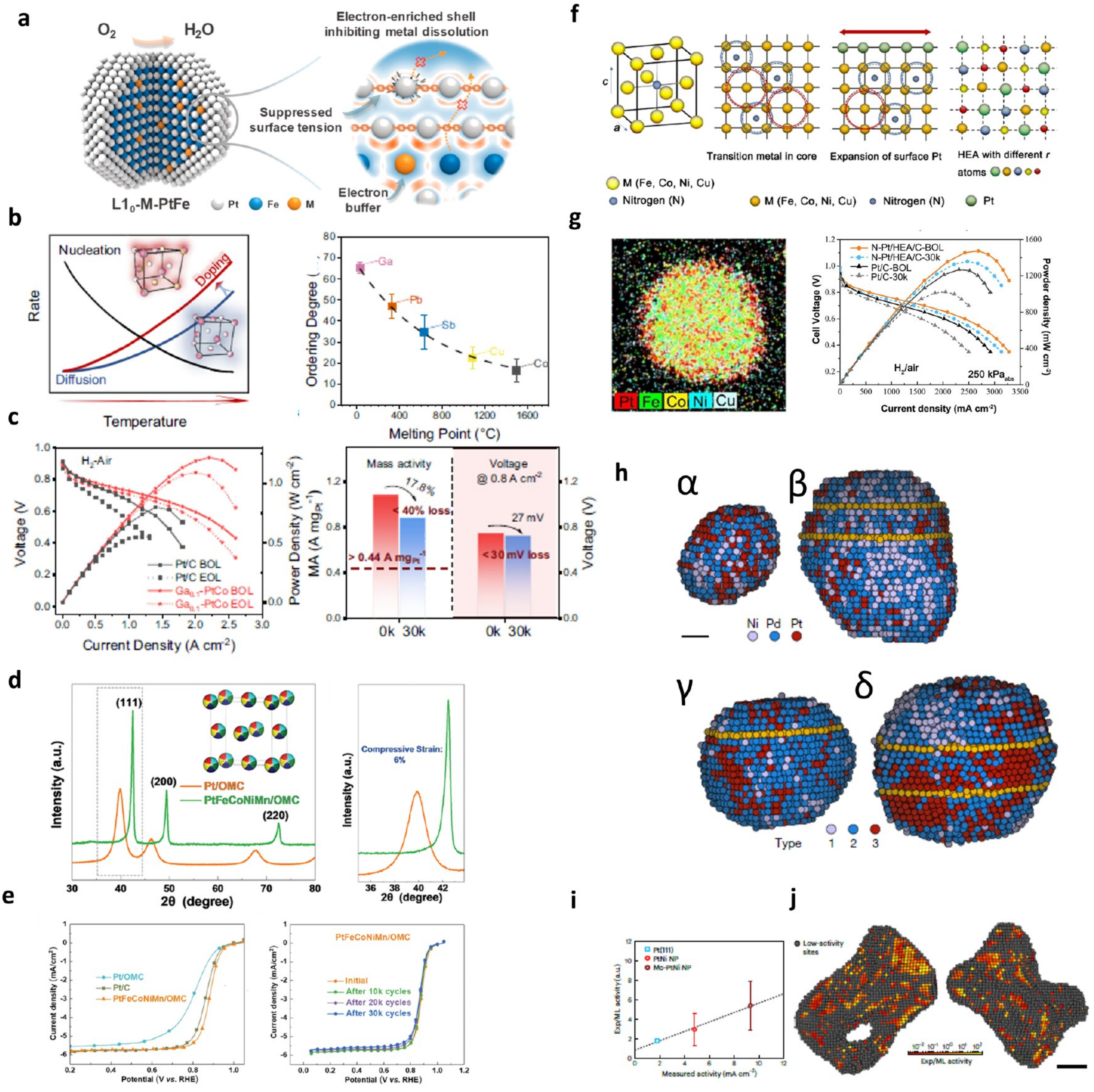


Figure 3. (a) Mechanism of electron buffers for relieving surface polarization in L10-M-PtFe catalysts through early transition metal doping. Reproduced from ref 35. Copyright 2024, American Chemical Society. (b) The dependence of nucleation and diffusion kinetic rate on temperature in the disorder-to-order transition and ordering degree of the M-PtCo catalysts versus melting point of M. (c) PEMFC performance before and after ADT of Ga-doped PtCo intermetallic alloys. Reproduced from ref 36. Available under a CC-BY 4.0 license Copyright 2023. (d) XRD patterns of Pt/OMC and PtFeCoNiMn/OMC (inset is the cell structure of structurally disordered FCC HEA nanoparticles) and the enlargement of the selected area. (e) ORR activity characterizations of PtFeCoNiMn/OMC and ORR stability characterizations. Reproduced with permission from ref 37. Copyright 2023, Wiley-VCH GmbH. (f) N-Pt/HEA atomic model with FCC metals and N atoms. (g) Energy dispersive spectroscopy (EDS) elemental mapping of the N-Pt/HEA/C nanoparticle and PEMFC performance during durability cycles. Reproduced from ref 8. Copyright 2024, American Chemical Society. (h) The 3D atomic structure using AET techniques. Experimental atomic models of two medium-entropy alloys (MEAs), (α) and (β), and two high-entropy alloys (HEAs), (γ) and (δ). Reproduced with permission from ref 38. Copyright 2023, Springer Nature. (i) Comparison between the specific activity at 0.9 V_{RHE} and the machine learning-identified activity. (j) Identification of the active sites of the PtNi and Mo-PtNi nanocatalysts using AET (scale bar, 2 nm). Reproduced with permission from ref 39. Copyright 2024, Springer Nature.

ML prediction model, which guides the selection of compositions with optimized energy and stability.

The addition of a third element has recently shown promising results in enhancing catalyst durability.[7] For example, the Pt_2CoCu catalysts demonstrated an excellent durability as well as a high power density in PEMFC tests, with minimal performance loss in the accelerated degradation test (ADT) (Figure 2d). This durability enhancement can be

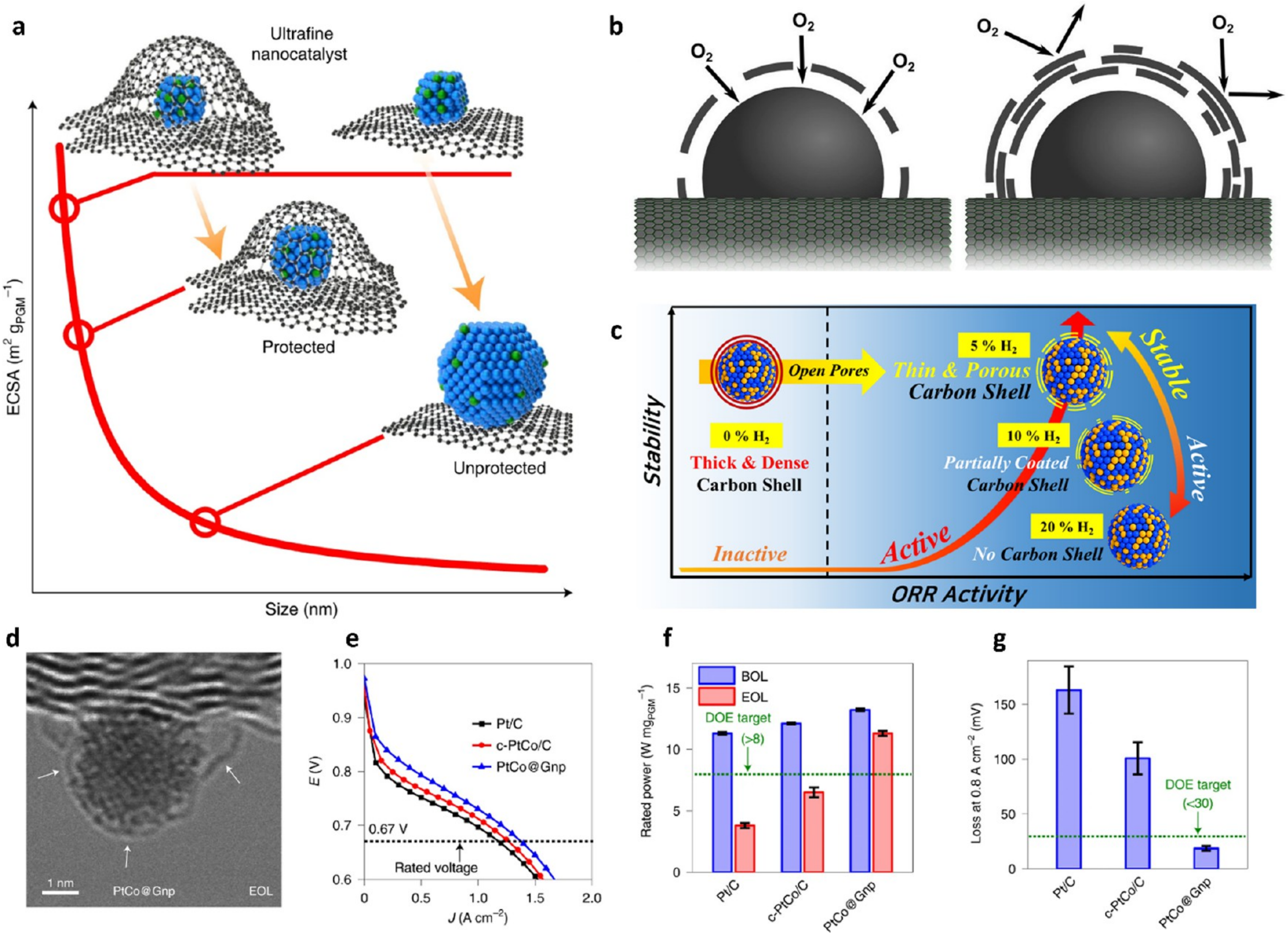


Figure 4. (a) Schematic illustration of ultrafine Pt nanocatalysts encapsulated in carbon shells and their effect on ECSA and particle size retention after ADT. Reproduced with permission from ref 42. Copyright 2022, Springer Nature. (b) Schematic diagram of the transport of oxygen to the catalyst surface as a function of carbon shell thickness. Reproduced from ref 41. Copyright 2015, American Chemical Society. (c) Schematic illustration of the target-customized carbon shell structure of carbon-encapsulated PtFe nanoparticles tuned using the H_2 content in the annealing gas. Reproduced with permission from ref 43. Copyright 2021, Royal Society of Chemistry. (d) Bright-field STEM image of carbon shell catalyst after PEMFC operation (white arrows indicate carbon shells). (e) Initial performance comparison of carbon-shell-coated catalyst (PtCo@Gnp) and conventional catalysts. (f) Comparison of initial and post-operational performance at a specified voltage, normalized to Pt loading. (g) Comparison of voltage losses at 0.8 A cm^{-2} (before and after operation). Green dotted line represents DOE target. Reproduced with permission from ref 42. Copyright 2022, Springer Nature.

attributed to the strengthened Pt–Co bonds and/or atomic ordering within the catalyst, which results from the introduction of a third element (e.g., Cu or Ni).[33] For another example, the introduction of Ga into PtCu intermetallics leads to directional covalent bonding during low-temperature phase transitions. As the covalent bonding between Pt and Ga is stronger than that between Pt atoms, Pt dissolution is significantly suppressed. Furthermore, the induced biaxial compressive strain optimizes the oxygen binding energy, thereby increasing the ORR activity (Figure 2e).[34] Additionally, the low melting point of Ga facilitates atomic mobility between Pt and Cu, enabling A1–L1$_0$ phase transitions at a low temperature of 350 °C, preventing particle agglomeration at higher temperatures, and maintaining structural stability.

2.2. Diverse Alloy Catalysts: Doped Structures and High-Entropy Alloys

Introducing early TMs (such as Cr, V, Ti, and Nb) as dopants can enhance the durability of Pt intermetallic catalysts. These early TMs usually reduce the energy barrier for forming ordered Pt–TM bonds, facilitating ordered L1$_0$-PtM (M = Co, Ni) intermetallic formation at lower annealing temperatures. Lowering the annealing temperature is advantageous for preventing particle sintering, which is a key challenge in the production of small intermetallic catalysts. Moreover, the dopants serve as electronic buffers that increase the electron density on the Pt shell, and this is beneficial for stabilizing the catalyst structure (Figure 3a).[35] In addition to early TMs, doping with main-group elements (p-block metals such as Ga, In, and Sn) facilitates fast ordering at high temperatures by lowering the energy barrier for atomic diffusion (Figure 3b).[36] Ga-doped L1$_0$-PtCo catalysts demonstrated high power densities at low Pt loadings and superior durability in ADT (Figure 3c).

High-entropy alloy (HEA) catalysts, which consist of more than five elements, have recently gained significant attention as a strategy for improving catalyst durability. By increasing the configurational entropy, these catalysts thermodynamically stabilize the structure, thereby reducing the likelihood of TM

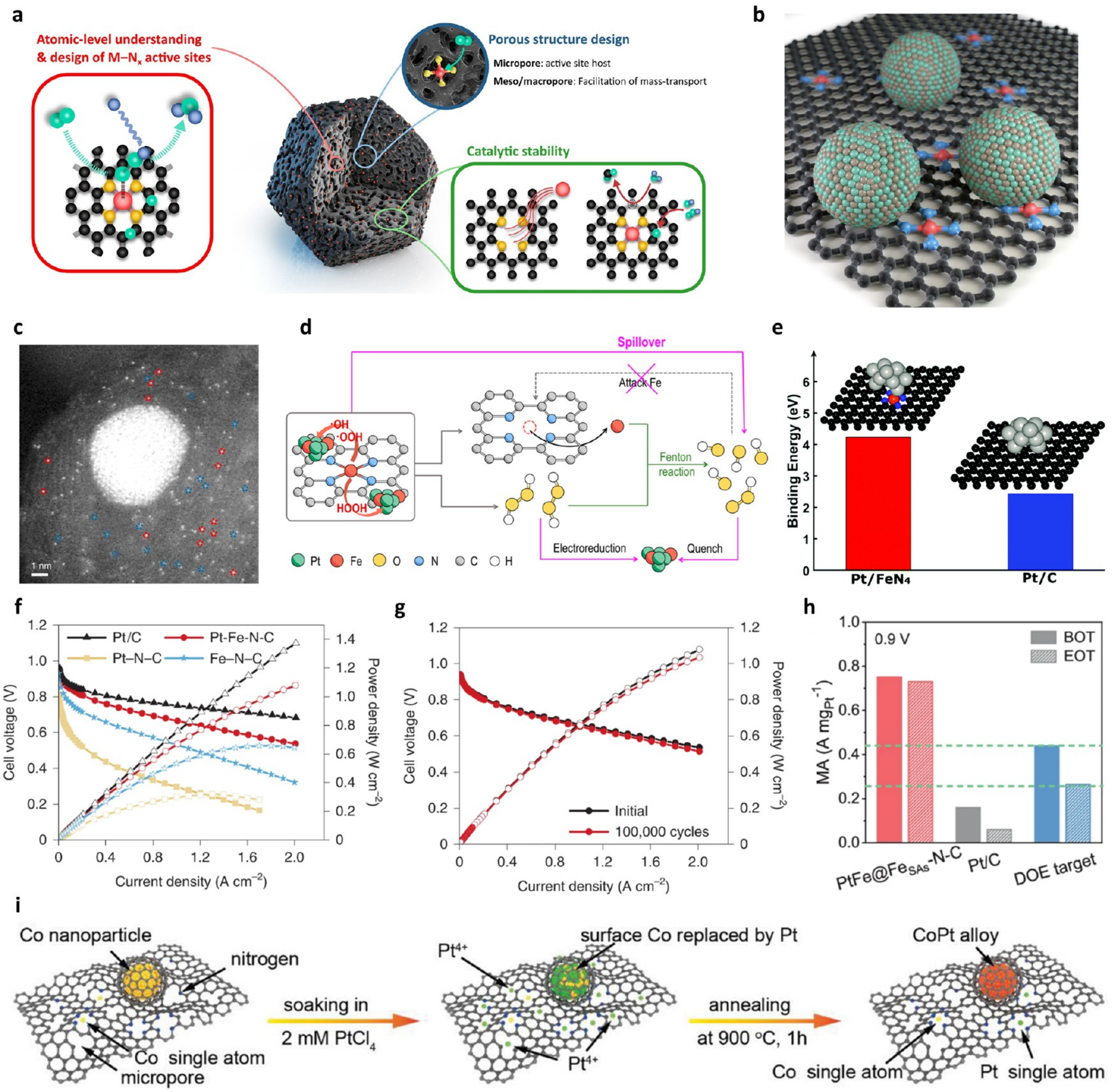


Figure 5. (a) Schematic illustration of the structure and design factors to consider for single-atom M-N-C catalysts for PEMFCs. Reproduced with permission from ref 47. Copyright 2021, Elsevier. (b) Schematic illustration of a hybrid catalyst using Pt nanoparticles and M-N-C catalyst as a support material. (c) HAADF-STEM image of a hybrid of Pt nanoparticles and M-N-C single-atom catalysts. Reproduced from ref 50. Available under a CC-BY 4.0 license Copyright 2022. (d) Schematic illustration of the performance and durability enhancement analysis of PtFe/Fe-N-C hybrid catalysts. Reproduced from ref 52. Copyright 2024, American Chemical Society. (e) Comparison of the calculated binding energies of Pt nanoparticles adsorbed on FeN_4 moieties embedded in graphene layers and Pt particles embedded in conventional graphene layers. Reproduced with permission from ref 51. Copyright 2021, Royal Society of Chemistry. (f) Comparison of PEMFC performance metrics for hybrid catalysts, conventional Pt/C catalysts, and single-atom Fe-N-C, Pt-N-C catalysts. (g) Long-term durability performance of PEMFCs over 100,000 cycles for hybrid catalysts. Reproduced from ref 50. Available under a CC-BY 4.0 license Copyright 2022. (h) Comparison of durability between the DOE long-term operational targets and hybrid catalysts in PEMFCs. Reproduced from ref 54. Available under a CC-BY 4.0 Copyright 2024. (i) The schematic diagram of the CoPt@C with Co and Pt single-atom catalyst synthesis. Reproduced from ref 55. Available under a CC-BY 4.0 Copyright 2023.

dissolution.[8] Recently, HEAs have been adopted as catalysts in various electrocatalytic applications due to their enhanced stability. Wang et al. demonstrated that the HEA catalyst exhibits enhanced ORR performance and long-term stability under harsh conditions (Figure 3d,e).[37] This could be due to lattice distortion effects and sluggish diffusion. The electronic structure of HEAs allows inactive atoms, such as Fe, Co, and Ni, to transform into ORR-active sites through electronic interactions, further enhancing the catalytic efficiency and durability. Additionally, Zhao et al. suggested that N-doping in

HEAs can lead to the formation of chemically stable metal−nitrogen (M−N) bonds, which enhance stability and prevent TM leaching, while also optimizing the surface electronic structure for selective oxygen reduction (Figure 3f,g).[8] The Pt-rich shell and synergistic effects of the M−N bonds contribute to the enhanced corrosion resistance and long-term operational stability of HEA-based catalysts in PEMFCs. However, the complex structures of high-entropy catalysts make their characterization and identification of optimal designs challenging. Hence, recent studies have introduced advanced tools, such as atomic electron tomography (AET), to verify the local structure and chemical ordering (Figure 3h).[38] Sophisticated atomic-level analyses provided insights into how lattice distortions or strain stabilize the structure (Figure 3i,j).[39] With advancements in analytical tools and the optimization of component compositions, high-entropy catalysts are expected to meet the stringent requirements of PEMFCs.

2.3. Carbon-Shell-Coated Catalysts

Carbon-shell-coated catalysts are recognized as an effective strategy for improving the durability and performance of PEMFC catalysts.[16] Carbon shells exist in various forms on the top surface of catalysts, including graphene, amorphous carbon, and porous structures. They protect the catalyst by physically stabilizing it and reducing potential damage that may occur during the electrochemical reaction.[40] However, if the carbon coating is too dense, it may restrict oxygen diffusion and potentially reduce catalyst activity. In contrast, porous carbon shells, which allow better oxygen access, are more vulnerable to degradation, particularly at the high temperatures and potentials encountered in PEMFCs.[41] To balance these factors, various carbon shell structures have been developed to achieve effective protection while minimizing the oxygen diffusion limitations.

Although Pt-based catalysts have been widely used in PEMFCs because of their high catalytic activity, they undergo degradation (e.g., dissolution, reorganization, and aggregation) during reactions. The particle growth reduces the electrochemical surface area (ECSA), resulting in a decrease in the active surface area (Figure 4a).[42] Migration and redeposition processes can be suppressed by encapsulating Pt NPs within a carbon shell, thereby minimizing changes in the particle size. Because performance deterioration begins with Pt migration, the introduction of a carbon shell can significantly extend the lifespan of the catalyst.

The thickness of the carbon shell is an essential factor that influences the catalyst stability.[43] However, if the shell is too thick, it may restrict the diffusion of oxygen molecules, resulting in decreased catalyst activity (Figure 4b).[41] In fuel cells, oxygen must diffuse to the catalyst surface. If the carbon shell impedes oxygen access to the particles, the electrochemical reaction efficiency decreases, which negatively affects the overall system performance. However, if the carbon shell is too thin, then the protective effect is insufficient. This can weaken the defense against the external environment and reduce the ability to prevent changes in the particle size and degradation of the Pt. Therefore, the formation of a carbon shell of appropriate thickness is crucial for effective oxygen diffusion while providing protection to the Pt particles; typically, the thickness is set to a few nanometers. Carbon shells are usually formed via the pyrolysis of organic molecules such as polydopamine, oleylamine, aniline, or 2,2′-bipyridine. According to previous studies, the thickness of the carbon shell can be efficiently controlled by adjusting the hydrogen content of the treatment gas (Figure 4c).[43,44]

For example, PtCo NPs have been encapsulated within graphene nanopockets to maximize the activity while reducing Pt usage.[42] A transmission electron microscopy (TEM) analysis of the NPs confirmed that the carbon shell remained intact, effectively encapsulating the Pt NPs (Figure 4d). PtCo@Gnp catalysts demonstrated superior voltage−current performance at various current densities compared with conventional commercial Pt/C and cobalt-based Pt catalysts (c-PtCo/C), particularly in maintaining high initial voltages at low current densities (Figure 4e). PtCo encapsulated within graphene nanopockets showed superior PEMFC performances that exceeded the DOE target of 8 W mg_{PGM}^{-1}, achieving a high rated power of 13.2 W mg_{Pt}^{-1} even under a low loading condition of 0.07 mg_{Pt} cm^{-2} (Figure 4f). Additionally, after rigorous durability testing, the voltage loss at 0.8 A cm^{-2} was only 18.8 mV, achieving the DOE's voltage loss target of 30 mV (Figure 4g). Recent reports on various Pt-based catalysts with different types of carbon shells highlight the potential of carbon-shell-coated catalysts as a promising solution for achieving a long lifespan in PEMFCs.[45,46]

2.4. Hybrid-Structured Catalysts

In PEMFC development, the use of Pt catalysts has consistently limited their widespread adoption because of their high cost and scarcity. To address this, research into nonprecious metal catalysts (NPMCs) has steadily progressed, with M-N-C emerging as the most viable alternatives under laboratory conditions.[47] However, several challenges remain for their widespread application in practical fields for both LDVs and HDVs. One of the primary challenges is the durability of NPMCs in acidic environments. Fe-based NPMCs, for instance, are prone to degradation due to Fenton reactions with H_2O_2, leading to structural breakdown and performance loss over time. Furthermore, despite significant advancements in material design, NPMCs still fall short of achieving the desired mass activity (MA) and long-term stability. Therefore, it is essential to optimize the structural and electronic properties of NPMCs to enhance stability, explore hybrid structures that integrate NPMCs with Pt or Pt alloys to improve durability, and leverage computational methods to identify new dopants or configurations that can mitigate degradation pathways (Figure 5a).[47,48]

Moreover, hybrid catalysts that combine M-N-C with Pt-based NPs have recently gained attention due to their synergistic effects.[49] The incorporation of Pt or Pt alloy NPs (e.g., Pt_3Co or Pt_3Fe) not only enhances the density of active sites but also facilitates oxygen adsorption and the overall reduction process, thereby improving both activity and durability (Figure 5b,c).[50] This is attributed to the modified electronic structure of Pt resulting from the SMSIs between Pt and M-N-C, which weaken the oxygen adsorption energy and lower the ORR activation energy, thereby enhancing the intrinsic activity of the catalyst.[23] Computational chemistry studies have demonstrated that in hybrid catalysts such as Pt/Fe-N-C, the O_2 adsorption energy at Pt active sites can be reduced by approximately 0.15 eV, facilitating easier O−O bond dissociation.[51] In addition, hybrid catalysts enable ORRs through multiple pathways by leveraging the interactions between multiple active sites. The cooperative mechanism between Pt and Fe-N-C promotes a gradual reduction of oxygen molecules through a stepwise reaction pathway, where

Pt reduces intermediate H_2O_2 produced via the two-electron pathway.[52]

Hybrid catalysts demonstrated enhanced electrocatalytic and structural stability compared to bare Pt and M-N-C catalysts. In Fe-N-C catalysts, H_2O_2 generated during ORRs induces structural damage to the catalyst. However, in hybrid catalysts, such as PtFe/Fe-N-C, H_2O_2 produced at the Fe−N sites migrates to the PtFe alloy surface, where it undergoes further reduction. This H_2O_2 spillover process effectively removes harmful oxygen radicals, suppressing catalyst degradation and thereby ensuring long-term stability (Figure 5d).[52] DFT calculations indicate that PtFe alloys have ·OH and ·OOH adsorption energies lower than those of Fe-N-C, effectively scavenging oxygen radicals. This ROS mitigation effect reduces the oxidative damage to the hybrid catalyst, allowing it to maintain a stable ORR performance. Additionally, the $Fe-N_4$ sites in M-N-C serve as anchoring sites for Pt clusters, which enhance the stability of the Pt catalyst (Figure 5e).[51]

In PEMFC tests, the hybrid catalyst exhibited a performance comparable to that of Pt/C, significantly outperforming the Fe-N-C and Pt-N-C catalysts (Figure 5f).[50] Notably, the durability remained exceptionally high, with little to no performance drop, even after 100,000 cycles (Figure 5g). Moreover, various hybrid catalysts have demonstrated impressive durability results that satisfy most durability targets set by the U.S. DOE (Figure 5h).[53,54] Therefore, Pt/M-N-C hybrid catalysts are promising alternatives for achieving both cost-efficiency and long-term performance. Recent advancements in catalyst design have demonstrated the effectiveness of hybrid strategies that integrate multiple approaches to enhance both activity and durability. In particular, catalysts that combine intermetallic alloying with carbon shell encapsulation have attracted significant attention due to their ability to improve stability, mitigate corrosion, and sustain high performance under demanding conditions. For example, PtCo alloy NPs protected by a graphene shell have been reported as an effective design strategy, leveraging the synergistic benefits of alloying and carbon-based protection to enhance both catalytic activity and durability (Figure 5i).[55] Similar approaches have been explored in various studies, highlighting the role of carbon shells in stabilizing metal NPs while promoting SMSI.[30] These findings highlight the significance of integrated catalyst architectures that combine intermetallic catalysts, carbon-based protective layers, and optimized electronic structures to enhance the electrocatalytic performance. Although these hybrid catalysts show potential for next-generation fuel cell applications, further optimization of catalyst structures and exploration of diverse metal−support interactions are essential to minimize the Pt dependence and maximize long-term stability. Additionally, rigorous evaluation under real-world operating conditions is crucial to assess their practical viability and identify pathways for further enhancement. The key parameters for the catalysts discussed in this Perspective are presented in Table 2.

3. SUPPORTS

3.1. Metal Oxide Supports

Previous studies have analyzed the design and preparation of metal oxide supports, such as TiO_2, SnO_2, ZrO_2, WO_3, CeO_2, and MoO_2, which generally exhibit much higher corrosion resistance than carbon in harsh acidic environments (pH < 1) and at high oxidation potential (1.0−1.5 V), as indicated by

Table 2. Summary of Catalyst Types, Properties, Advantages, Limitations, and Challenges for PEMFCs

catalyst type	definition	pros	cons	major challenges
conventional Pt/C	commercially used Pt NPs on carbon supports	(1) high initial catalytic activity and ESCA (2) well-established synthesis techniques	(1) susceptible to carbon corrosion (2) Pt degradation	mitigating carbon corrosion and enhancing durability
intermetallic catalysts	highly ordered Pt-based alloys (e.g., PtCo, PtNi, PtNiCu)	(1) excellent ORR activity (2) suppressed TM dissolution	(1) limited scalability (2) difficulty in morphology control	establishing scalable methods and preventing NP sintering during high-temperature process
carbon shell	carbon protective layer-coated NPs	(1) migration of Pt degradation (2) enhanced electron transfer	mass transfer restriction due to suboptimal shell thickness	optimizing carbon shell thickness to enhance oxygen diffusion while maintaining protection
hybridized structure	combination of different materials (e.g., Pt-M-N-C) with strong metal−support interactions	improved ORR performances via enhanced anchoring of Pt catalyst	(1) complex synthesis (2) potential electronic mismatch (3) challenging surface optimization	stabilizing electronic interactions while reducing Pt dependence

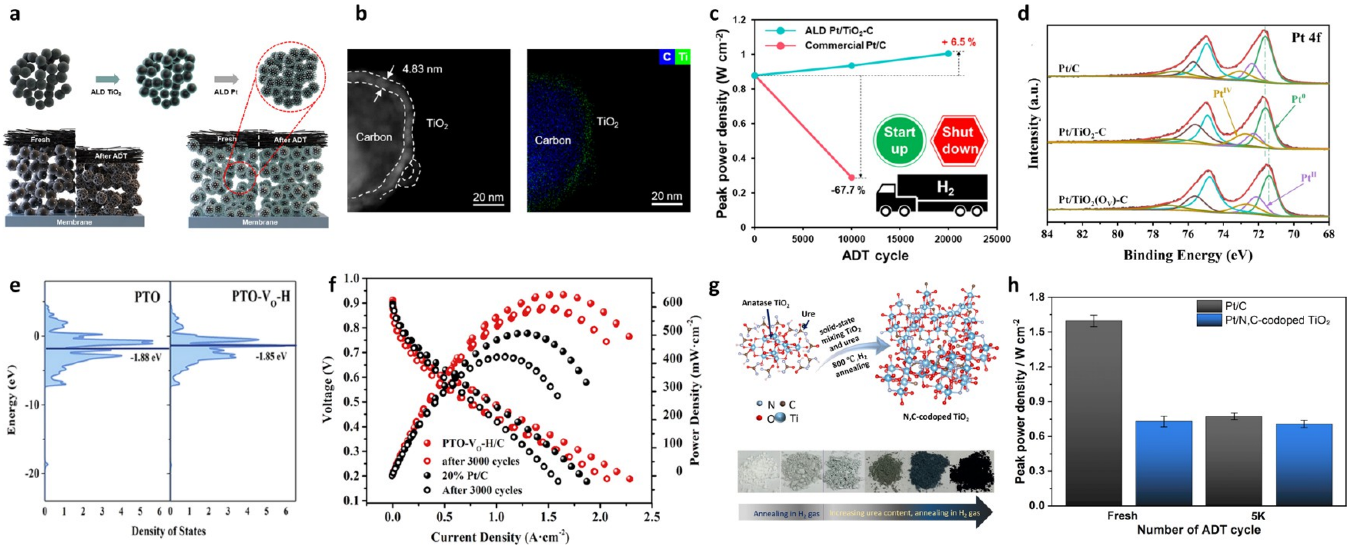


Figure 6. (a) Schematic illustration of the ALD process used to form ALD Pt/TiO_2-C, along with a comparison of the catalyst layer after ADT with commercial Pt/C and ALD TiO_2-C. (b) STEM and EDS elemental mapping images of ALD TiO_2-C. (c) Comparison of ALD Pt/TiO_2-C and commercial Pt/C materials with respect to long-term catalyst support, as represented by peak power density over a number of ADT cycles. Reproduced from ref 59. Copyright 2024, American Chemical Society. (d) XPS spectra of Pt 4f in the Pt/C, Pt/TiO_2-C, and Pt/TiO_2(O_V)-C. Reproduced with permission from ref 63. Copyright 2023, Elsevier. (e) PDOS for d-orbitals of Pt atoms of PTO and PTO-V_o-H. (f) *I*−*V* polarization curves of PTO-V_o-H/C and commercial Pt/C catalysts in single cells before and after 3000 cycles. Reproduced from ref 64. Copyright 2022, American Chemical Society. (g) Synthetic process of N,C-codoped TiO_2 by annealing method. (h) Reduction degree results of max power density of Pt/N,C-codoped TiO_2. Reproduced from ref 65. Copyright 2020, American Chemical Society.

Pourbaix diagrams, primarily due to the high valence states of metal oxides.[20,56] One of the key advantages of metal oxide supports is their ability to have high electrochemical stability, which helps prevent Pt catalyst aggregation, a common challenge in Pt/C catalysts. Under high potentials caused by repetitive startup/shutdown cycles and fuel starvation, carbon support corrosion occurs, leading to irreversible damage to the catalyst layer.[57] Metal oxide supports are expected to achieve an operational lifetime of up to 30,000 h, meeting one of the critical requirements for HDV applications. Moreover, Pt catalysts can be immobilized on oxide supports to prevent Pt migration and agglomeration. The anchored Pt can withstand the high potentials encountered during the startup and shutdown phases of PEMFCs.[12] Among the various oxides, titanium oxide (TiO_x) is one of the most widely studied noncarbon support materials for PEMFCs owing to its stability, low cost, and nontoxicity.[58] The strong interaction in Pt/TiO_2 is expected to improve both the catalytic activity and the stability compared to those of conventional Pt/C. However, despite these advantages, the insulating nature and small surface area of TiO_2 hinder its overall performance in PEMFCs.

To address this, a recent study demonstrated that a Pt/TiO_2-C electrode fabricated via atomic layer deposition (ALD) exhibited PEMFC performance comparable to that of commercial Pt/C (Figure 6a).[59,60] By incorporation of carbon, good electrical conductivity was achieved, and the thin TiO_2 coating on carbon effectively prevented carbon corrosion (Figure 6b). The Pt NPs were strongly anchored to the TiO_2-C surfaces, which inhibited their growth and aggregation. Compared to commercial Pt/C, Pt/TiO_2-C shows excellent durability, meeting the DOE target for support durability after ADT, which is attributed to the stabilization of Pt NPs through the SMSI effect (Figure 6c).[59] However, under high-current-density conditions (>1.0 A cm^{-2}), additional ohmic losses from electron transport across the TiO_2 layers can significantly hinder the performance, highlighting the need to further enhance the electrical conductivity of TiO_2.[61]

The introduction of oxygen vacancies (O_V) and doping with other elements is a key strategy to improve electrical conductivity and tune the electronic structure, thereby enhancing ORR activity.[62] O_V promotes a strong Pt−TiO_2 interaction, which in turn promotes the electron transfer between Pt and TiO_2, as evidenced by a shift in the Pt 4f X-ray photoelectron spectroscopy (XPS) peak of Pt/TiO_2(O_V)-C toward lower binding energies (Figure 6d).[63] The calculated partial density of states (PDOS) revealed that highly dispersed Pt anchored on TiO_x/C composites with abundant oxygen vacancies (PTO-V_o-H/C) exhibited a higher *d*-band center than the non-V_o supports, indicating a reduction in the binding strength of Pt and oxygen-containing intermediates, mainly due to the SMSIs triggered by O_V (Figure 6e).[64] PEMFC single cells equipped with PTO-V_o-H/C delivers a maximum power density of 643.5 mW cm^{-2}, which is superior to that of Pt/C-based PEMFCs. After ADT, the maximum power density of the PTO-V_o-H/C-based PEMFC was reduced by only 6.9%, which was approximately one-fifth of the degeneration of the Pt/C-based PEMFC (Figure 6f). Previously, Lee et al. reported the introduction of dopants and O_V into a TiO_2 support to maximize the SMSI effects (Figure 6g).[65] A N,C-doped TiO_2 support with abundant defect sites showed a stronger interaction between Pt and the support via SMSIs. As a result, although the initial peak power density of Pt/N,C-doped TiO_2-based PEMFC was lower than that of Pt/C-based PEMFC, it demonstrated remarkable durability in PEMFC tests, showing only a 4% decrease in the maximum power density compared to the 52.4% reduction observed in commercial Pt/C (Figure 6h). The introduction of O_V and doping strategies for TiO_2 support demonstrate effective approaches to enhancing PEMFC performance by improving the electrical conductivity

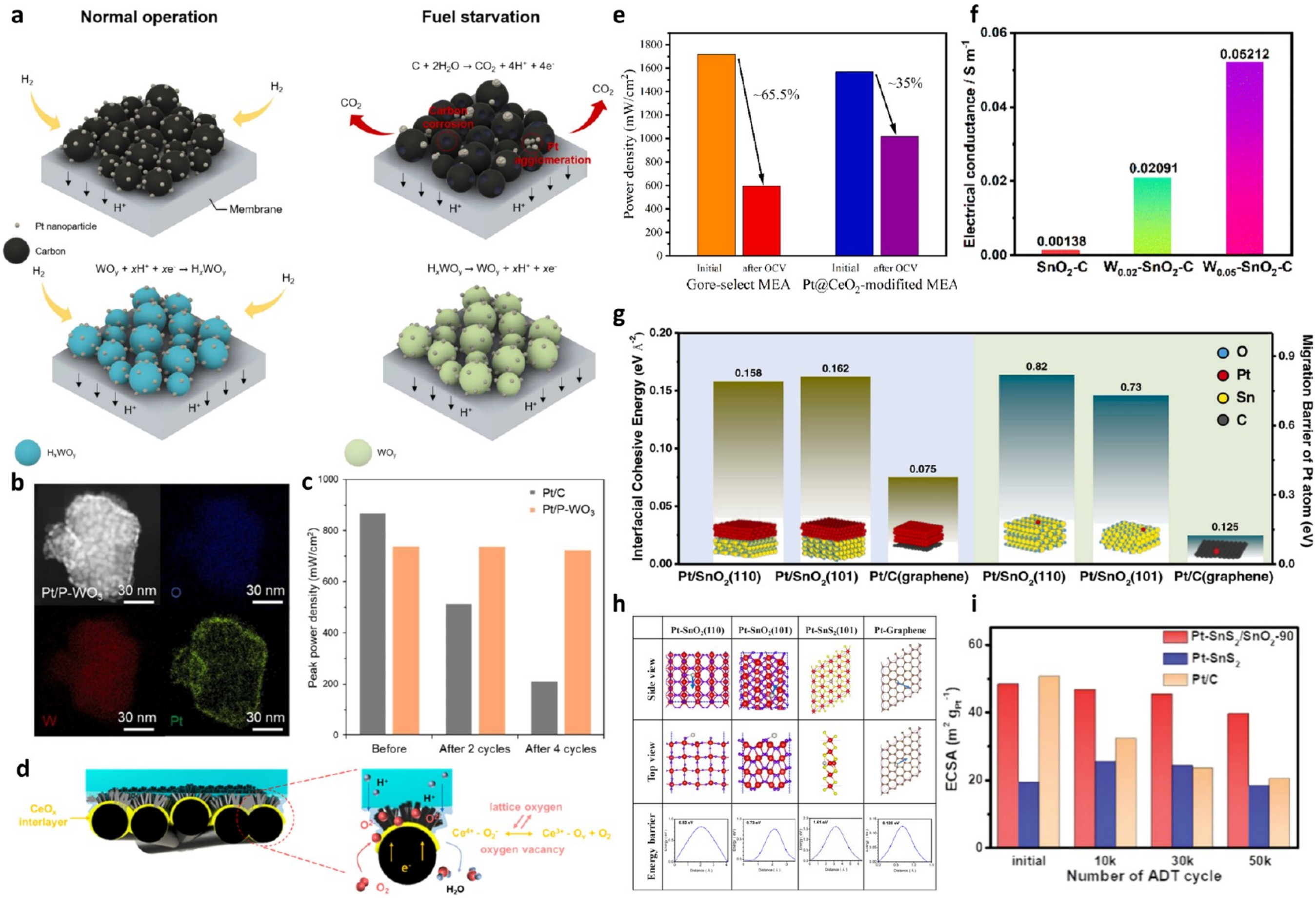


Figure 7. (a) Schematic design of processes occurring in PEMFC anode under normal operation and fuel starvation conditions for Pt/C and Pt/WO_3. (b) EDS mapping data from STEM image of Pt/P-WO_3 catalyst. (c) Peak power density changes following fuel starvation durability tests. Reproduced with permission from ref 67. Copyright 2023, Elsevier. (d) Schematics illustrating the redox reaction of CeO_X interlayer during ORR. Reproduced from ref 68. Copyright 2023, American Chemical Society. (e) Power density decay comparison for commercial MEA (Gore-select MEA) and Pt@CeO_2-modified MEA. Reproduced with permission from ref 69. Copyright 2023, Elsevier. (f) Comparison of electrical conductivity of W-doped SnO_2-C and SnO_2-C. Reproduced with permission from ref 70. Copyright 2024, Royal Society of Chemistry. (g) Comparison of interfacial cohesive energies and energy barriers of a Pt atom of Pt/SnO_2(110), Pt/SnO_2(101), and Pt/graphene. Reproduced with permission from ref 71. Copyright 2022, Elsevier. (h) DFT calculated energy barriers of Pt-SnO_2(110), Pt-SnO_2(101), Pt-SnS_2(101), and Pt/graphene for migration barrier of Pt atom. (i) ECSA values of Pt-SnS_2/SnO_2-90, Pt/C, and Pt-SnS_2 before and after ADT. Reproduced with permission from ref 72. Copyright 2022, Wiley-VCH GmbH.

of the support material, utilizing the SMSI effect, and achieving optimized electronic modulation.

Tungsten oxide (WO_3) is considered another promising support material for PEMFCs, due to its excellent acidic stability, which is achieved through the rapid formation of a special hydrogen tungsten bronze (H_xWO_3, $0.3 < x < 0.5$), also known as the hydrogen spillover effect (eqs 1 and 2).[66]

$$WO_3 + x\text{Pt–H} \rightarrow H_xWO_3 + \text{Pt} \quad (1)$$

$$H_xWO_3 \rightarrow WO_3 + xe^- + xH^+ \quad (2)$$

The reversible hydrogen spillover effect of H_xWO_3 helps maintain the Pt active sites and prevents corrosion, especially under fuel starvation conditions in PEMFCs (Figure 7a).[67] When the hydrogen fuel is insufficient, typical Pt/C catalysts degrade via carbon corrosion reactions. In contrast, when hydrogen is scarce, H_xWO_3 generates the necessary hydrogen ions through a reverse reaction to meet the power demand without performance degradation, demonstrating the excellent resilience of PEMFC under fuel starvation and actual driving conditions. Recently, Park et al. demonstrated that uniformly distributed Pt can be deposited on conductivity-enhanced WO_3 supports via ALD, resulting in Pt/P-WO_3 (Figure 7b).[67] Compared to the Pt/C-based PEMFCs, which showed a significant decrease in peak power density after fuel starvation in PEMFC cycles, the Pt/P-WO_3-based PEMFC demonstrated superior durability, retaining over 98% of its initial peak power density owing to the corrosion resistance of P-WO_3. This was attributed to the hydrogen spillover effect (Figure 7c).

In addition to catalyst degradation, membrane degradation also poses major challenges under harsh PEMFC operating conditions. This degradation is typically initiated by free radicals formed from the reaction of H_2O_2 with metal ions. To address this issue, cerium oxide (CeO_2) has gained significant attention because of its ability to effectively scavenge these harmful radicals. The transition between Ce^{3+} and Ce^{4+} states facilitates the formation of oxygen vacancies and the intrinsic SMSI effect for the metal oxide support, which is crucial for enhancing the catalytic performance and long-term durability of PEMFCs (Figure 7d).[68] The lower degradation of the power density of the Pt-CeO_2-based PEMFC after an OCV test compared to that of the commercial membrane-based

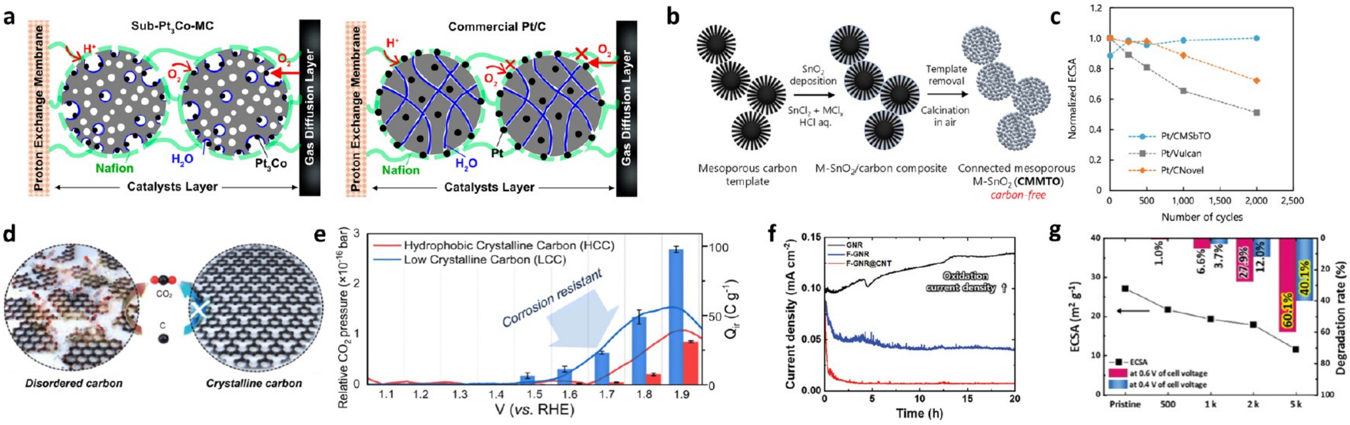


Figure 8. (a) Schematic design of processes occurring in PEMFC for mesoporous support-based MEA and Pt/C-based MEA. Reproduced from ref 74. Available under a CC-BY 4.0 license Copyright 2021. (b) Synthesis scheme of connected mesoporous M-doped SnO_2 (CMMTO). (c) Normalized ECSA (normalized by its maximum value) as a function of the number of potential cycles for the start−stop ADT. Reproduced from ref 75. Copyright 2024, American Chemical Society. (d) Differences in the carbon corrosion resistance depending on the degree of crystallinity. (e) Differential electrochemical mass spectrometry and irreversible charge results showing the corrosion points of low-crystalline carbon and crystalline carbon. Reproduced with permission from ref 76. Copyright 2023, Royal Society of Chemistry. (f) Current density per time at a constant 1.8 V for 20 h of GNR, F-GNR, and F-GNR@CNT. (g) Quantitative analysis of ECSA values with decreasing performance rates measured at cell voltages of 0.6 and 0.4 V for Pt/C+F-GNR@CNT during 5k ADTs. Reproduced from ref 77. Available under a CC-BY 4.0 license Copyright 2024.

PEMFC indicates that Pt-CeO_2 can effectively scavenge free radicals in the PEMFC and prevent the chemical degradation of PEM to enhance the durability of the PEMFCs (Figure 7e).[69] Challenges remain in uniformly forming a CeO_2 support with highly dispersed Pt NPs on the MEA. Further design of the CeO_2 support to create a more heterogeneous interface could improve the long-term durability of PEMFC membranes by preventing free radical attacks.

From the perspective of the excellent acid resistance properties of metal oxides, tin oxide (SnO_2) is promising as an acidic ORR support due to its strong Sn−O bonds and inherent durability. However, despite these advantages, the low electron conductivity of SnO_2 ($<10^{-5}$ S cm^{-1}), is one of the main factors reducing electrochemical efficiency. To address this issue, a strategy of doping SnO_2 with other elements can be used. For example, doping SnO_2 with W (W-SnO_2-C) can effectively enhance the electronic conductivity (Figure 7f), achieving a higher peak power density than commercial Pt/Co-based PEMFCs.[70] Recently, Li et al. reported cable-like core@shell CNTs@SnO_2, where the CNT core provides the necessary electron conductivity, while the SnO_2 shell acts as a protection layer, shielding the CNTs against corrosion and stabilizing the Pt NPs under PEMFC operation.[71] The interfacial cohesive energy and energy barriers of a Pt atom migrating on the SnO_2 surface are higher than those on graphene, suggesting that Pt growth is effectively suppressed by the strong interaction between Pt and SnO_2 (Figure 7g). Similarly, heterostructures incorporating extrinsic conductive materials, such as SnS_2, can be applied.[72] The SnS_2/SnO_2 heterostructured support provides an effective Pt diffusion barrier, as the diffusion rate on either side of the SnS_2/SnO_2 heterointerface is significantly lower than that on graphene supports (Figure 7h). Consequently, after ADT, Pt migration and aggregation at the SnS_2/SnO_2 interface are suppressed, which hinders Pt particle growth and the dissolution rate of the active sites, resulting in the preservation of the ECSA (Figure 7i). In conclusion, to leverage the excellent durability characteristics of SnO_2 in PEMFCs, combining it with conductive materials to form a heterostructure is effective. This approach, through its high electrical conductivity and well-dispersed Pt, ensures enhanced efficiency and durability in PEMFCs.

Although metal oxide supports have been reported to exhibit superior durability compared to Pt/C, they generally show inferior activity compared to Pt/C. This is primarily due to the lower electrical conductivity of metal oxides. Despite this challenge, the highly durable metal oxide can be adopted in HDVs, where stability under long-term performance and in dynamic operating environments is required. To achieve this, the challenges associated with metal oxide supports, particularly their low electrical conductivity, should be addressed. For example, applying a modified metal oxide coating with enhanced properties to graphitized carbon supports featuring mesopores could result in an ideal advanced material that integrates the full range of the desired support characteristics.

3.2. Mesoporous-Structured Support

Mesoporous supports (including carbon and metal oxides) offer significant advantages for enhancing the initial performance of PEMFCs by addressing catalyst poisoning issues from sulfonic acid groups in the ionomer and improving oxygen transport across Pt/ionomer interfaces.[73] Moreover, the pore structure provides the catalysts with stable sites that prevent catalyst dissolution and agglomeration under harsh PEMFC operating conditions. As shown in Figure 8a, a Pt_3Co NP catalyst embedded within the mesopores experienced less direct coverage by the ionomer, which minimized poisoning by sulfonic acid groups.[74] This reduced direct contact with the catalyst surface enhanced the durability of the PEMFCs compared to that of commercial Pt/C. Recently, Inaba et al. reported an effective strategy using a mesoporous Sb-doped SnO_2 support (CMSbTO) with well-controlled mesopore sizes ranging from 4 to 11 nm through a carbon template, which not only enhanced the electrical conductivity via Sb-doping but also ensured excellent durability for PEMFCs by constructing a mesoporous-structured metal oxide support (Figure 8b).[75] After ADT, Pt/CMSbTO maintained a stable ECSA, even showing a slight increase, possibly owing to the prevention of Pt growth and improved Pt utilization from the altered

Table 3. Summary of Support Types, Properties, Advantages, Limitations, and Challenges for PEMFCs

support type	definition	pros	cons	major challenges
metal oxides	oxide-based supports (e.g., TiO_x, WO_x, SnO_x)	(1) high stability (2) modification of catalyst via metal−support interactions	(1) low electrical conductivity (2) challenges in achieving uniform Pt dispersion	enhancing intrinsic electrical conductivity and Pt dispersion
mesoporous materials	porous materials with tunable pore structure (e.g., mesoporous carbon, metal oxides)	(1) large ECSA (2) enhanced Pt dispersion (3) improved mass transport (4) resistance to ionomer poisoning	(1) structural degradation under harsh conditions (2) challenges in achieving optimal position of Pt	maintaining structural integrity and optimizing the impregnation method
graphitized carbon support	highly ordered carbon materials (e.g., graphene, crystalline carbon)	(1) high electrical conductivity (2) enhanced corrosion resistance	(1) requires high-temperature process (2) challenges in achieving uniform Pt dispersion (3) intrinsic carbon defects	ensuring consistent structural properties under PEMFC operating conditions

ionomer distribution or enhanced wettability of the mesoporous support (Figure 8c). Mesoporous structures are expected to surpass the properties of commercially available mesoporous carbons, such as Ketjen Black, by improving oxygen transport and reducing catalyst poisoning from ionomers, ultimately enhancing the durability of PEMFCs.

3.3. Graphitized Carbon Support

Considering the conductivity of carbon-based supports, anticorrosive carbons, such as crystalline carbon and graphene, are viable alternatives to commercial carbon supports. These materials not only offer high conductivity but also ensure superior corrosion resistance, making them effective for application in PEMFCs. Generally, commercial carbon supports such as Vulcan XC-72R are susceptible to corrosion at high potentials of up to 1.5 V due to frequent voltage spikes under dynamic PEMFC conditions. However, crystalline carbon, converted from disordered to crystalline through high-temperature treatments, shows significantly improved corrosion tolerance (Figure 8d,e).[76] Recently, an anticorrosive carbon material for ORR-fluorine-doped graphene nanoribbons incorporated with graphitic carbon nanotubes (F-GNR@CNT) and anticorrosion properties attributable to the F-doping effect was reported (Figure 8f).[77] Following the DOE-recommended ADT protocol, the PEMFC with Pt/C-containing F-GNR@CNT (Pt/C+F-GNR@CNT) exhibited a low degradation rate and good ECSA retention, effectively suppressing carbon corrosion (Figure 8g).

Although recent studies have demonstrated promising results with graphitized carbon-supported catalysts, most of them do not meet the performance targets set for commercial applications. Despite their higher conductivity, carbon-based supports have inferior stability compared with metal oxide supports, particularly under the frequent voltage spikes caused by PEMFC startup and shutdown cycles. Therefore, utilizing carbon supports under harsh conditions requires more rigorous verification and strategies. Further research is needed on hybrid approaches, such as heterostructures that combine the high conductivity of graphitized carbon with the excellent durability of metal oxides. The key parameters for the supports in this Perspective are presented in Table 3.

4. CONCLUSIONS AND PERSPECTIVES

Despite the remarkable progress in PEMFCs over the past few decades, the transition from LDVs to HDVs presents new challenges, especially for catalyst durability (Table 1). LDVs focus on power density to keep the fuel cell stack compact while meeting performance demands for acceleration and frequent stop−start cycles in urban environments. Typically, LDVs operate at cell voltages around 0.675 V and temperatures between 80 and 90 °C, as they require low speeds and low average power output. In contrast, HDVs demand constant high power due to their driving characteristics, such as long-distance, high-speed driving and heavy load transport. To enable constant high-power operation, the operating conditions are carefully optimized. Compared to LDVs, HDVs

(i) Operate at higher cell voltages ($\geq$0.7 V) to achieve efficiency levels of 50% or more, which significantly reduces hydrogen consumption and heat generation.
(ii) Require long-term stability of 30,000 h (equivalent to 90,000 cycles in ADT) under HDV operating conditions. Additionally, they must withstand dynamic current loading modes, including startup and shutdown cycles.
(iii) Operate at operating temperatures above 90 °C, which improves waste heat rejection by increasing the temperature gradient between the fuel cell and ambient air and enables more compact thermal management systems by reducing humidification needs.

Therefore, unlike the catalyst development approach used for LDVs, catalysts for HDVs must be developed to meet the specific conditions required for HDV applications. For example, the HDV necessitates higher Pt loadings, whereas LDV requires lower Pt loadings ($<$0.125 mg_{Pt} cm^{-2}). LDVs require high power density and low Pt usage to achieve enhanced ORR mass activity (MA $\geq$ 0.44 A mg_{PGM}^{-1} @0.9 V, O_2). In contrast, HDVs prioritize long-term stability, necessitating highly durable Pt alloy catalysts, corrosion-resistant supports, and stable ionomer/catalyst interfaces to minimize degradation. Specifically, durable catalysts designed to withstand harsher conditions, such as elevated temperatures and voltages, should be developed. Additionally, there is a lack of validation under HDV operating conditions, particularly under adverse conditions such as fuel starvation that can accelerate degradation. Therefore, the establishment of comprehensive durability assessments is essential. These assessments should include *I*−*V* testing at high temperatures and pressures, cyclic load variations, and platinum utilization assessments at voltages exceeding 0.7 V.

In this Perspective, alongside conventional Pt-based catalysts, we highlight promising strategies, including ternary intermetallic, carbon-shell-coated, and hybrid catalysts, all of which have shown superior performance and durability. However, several technical barriers hinder their practical application in HDVs. One of the key limitations is the scalability of the synthesis process. Only when the synthesis method is simple and cost-effective can the developed catalysts be successfully adopted in commercial applications, especially for HDVs, which require large amounts of catalysts. To achieve reliability and scalability, collaboration with industry stakeholders is essential to bridge the gap between laboratory innovations and commercialization.

In conclusion, continued advancements in PEMFC technology require improvements not only in catalysts and supports but also in their optimal combinations and precise property tuning. The integration of machine learning and computational modeling is expected to accelerate the identification of optimal materials and design strategies while enabling the control of low-temperature phase transitions to enhance the catalyst stability. As the hydrogen economy expands, HDVs are expected to become a primary application for PEMFCs, making the development of high-durability, high-efficiency catalysts increasingly vital.

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Author Contributions

$^{\nabla}$S.P., E.L., and Y.P. contributed equally. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. CRediT: **Subin Park** resources, visualization, writing - original draft; **Eungjun Lee** conceptualization, resources, validation, writing - original draft; **Yoonsu Park** resources, validation, writing - original draft; **Myeong-Geun Kim** conceptualization, supervision, validation, writing - review & editing; **Sung Jong Yoo** conceptualization, investigation, resources, writing - review & editing.

Notes

The authors declare no competing financial interest.

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