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Research Progress and Prospects of Multi-Dimensional Optimization of Hydrogen Fuel Cell Catalysts

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Abstract. Against the backdrop of the global transition to clean and low-carbon energy, hydrogen fuel cells have attracted widespread attention because of their high efficiency and zero pollution characteristics. At present, its large-scale commercialization still faces challenges such as the high cost of noble metal catalysts and insufficient activity and stability. Especially in proton exchange membrane fuel cells, the cathode oxygen reduction reaction (ORR) relies on a large number of platinum-based catalysts, which restricts the development of the technology. This essay systematically reviews the latest research progress of hydrogen fuel cell catalysts, with a focus on the optimization strategies of platinum-based catalysts, as well as the key breakthroughs and challenges faced by low-platinum and platinum-free catalysts. Finally, the future technological development direction and commercial prospects are prospected, with the aim of providing references for related research and promoting the further development and application of hydrogen fuel cell technology.

Keywords: Platinum-Based Catalysts, Hydrogen Fuel Cells, Review, Optimization Strategies

1. Introduction

Against the backdrop of the global energy structure's transition towards green energy and clean low-carbon, hydrogen fuel cells, as an efficient and zero-emission energy conversion device, have received unprecedented attention [1,2]. The hydrogen fuel cells energy conversion principle is as follows: the overall reaction is $2H_2 + O_2 \rightarrow 2H_2O$; the anode reaction is $2H_2 \rightarrow 4H^+ + 4e^-$; the cathode reaction is $O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$; and the only by-product is water. Therefore, they have demonstrated tremendous application potential in various fields such as transportation, distributed power generation, and portable electronic devices. Especially in the automotive industry, PEMFCs are regarded as the ideal technology to replace traditional internal combustion engines in the future and achieve the goal of "zero emissions [3,4]. This is because PEMFC has significant advantages, low operating temperature, fast start-up speed, and high power density

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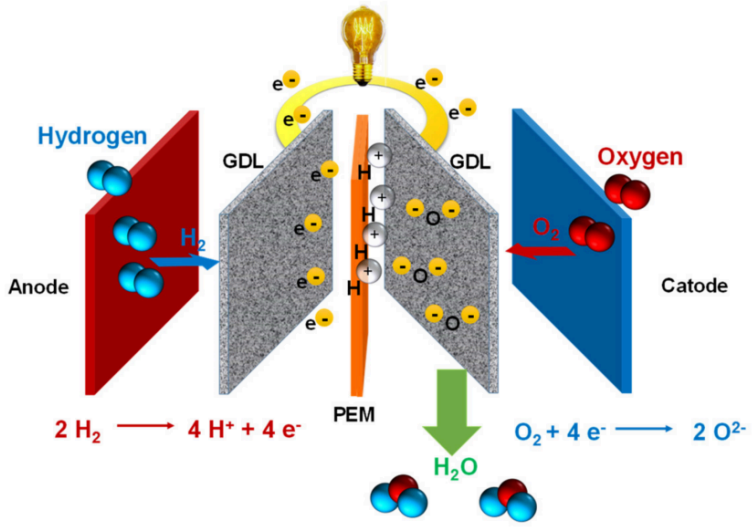


Figure 1. The structure diagram of a proton exchange membrane fuel cell (PEMFC) includes components such as the cathode, anode, gas diffusion layer (GDL), and proton exchange membrane. This figure shows the working process of generating current from the reaction gas under the action of a catalyst [5]

However, the large - scale commercialization of hydrogen fuel cells still confronts numerous challenges. Among these, the cost, stability, and activity of catalysts are the core constraints impeding their development. In PEMFCs, The oxygen reduction reaction (ORR) at the cathode relies on an efficient electrocatalyst. Owing to the slow kinetics and intricate reaction mechanism of the oxygen reduction reaction (ORR), the cathode of proton exchange membrane fuel cells (PEMFCs) necessitates a substantial amount of platinum (Pt) as a catalyst. This not only escalates the cost of fuel cells but also impedes their sustainable advancement.

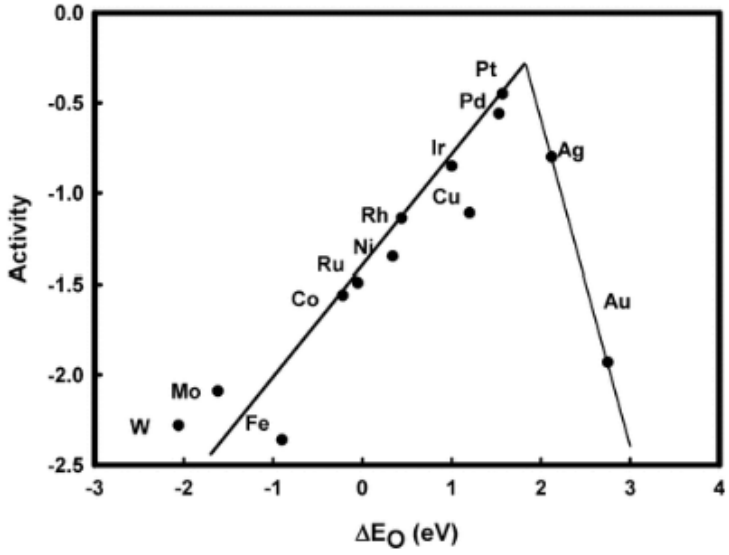


Figure 2. Volcanic plot showing the activity of oxygen reduction reactions of different elements [6]

It can be seen from this that to overcome the above challenges, in-depth research on the catalytic mechanism of hydrogen fuel cells and efforts to develop new catalyst systems with low costhigh

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performance, high performance and long life are the core challenges and important directions of current research in this field. This article systematically reviews the latest research progress of hydrogen fuel cell catalysts. The performance optimization methods of Pt-based catalysts, as well as the technological breakthroughs and existing difficulties of low platinum content and platinum-free catalysts were mainly discussed, and a comparative analysis of the performance of various catalysts was carried out. On this basis, the article further looks forward to the future development direction and commercial potential of such catalysts. Through the discussion of key issues such as the structural design, performance regulation, stability improvement and industrialization bottlenecks of catalysts, the aim is to provide useful reference value for researchers in the field and promote the progress and commercialization of hydrogen fuel cell technology.

2. Application basics and optimization strategies of Pt-based catalyst

At present, Pt-based catalysts are indispensable core materials in fuel cells of hydrogen fuel vehicles, mainly proton exchange membrane fuel cells (PEMFCs), due to their excellent catalytic effects. Its basic structure is generally composed of platinum (Pt) nanoparticles with a size ranging from 2 to 5nm, which are loaded on a carbon carrier. The Pt nanoparticles in traditional Pt/C catalysts mostly have a face-centered cubic structure and are attached to the surface of carbon carriers through physical or chemical methods [1,2]. To enhance the utilization rate and catalytic activity of Pt, researchers designed various structured Pt-based catalysts. Pt nanocrystals with specific crystal plane exposure (e.g., cubes, octahedrons, icosahedrons) can alter the atomic arrangement on the surface to optimize the adsorption of ORR intermediates in oxygen reduction reactions, thereby achieving the optimization of intrinsic activity [3,4].

2.1. Morphology and core-shell structure regulation

Morphology and core-shell structure regulation is one of the key strategies to enhance the stability and activity of platinum-based catalyst and reduce platinum usage. By precisely controlling the size, shape, exposed crystal facets of platinum-based nanoparticles, and constructing core-shell structures, the electronic properties of surface can be effectively tuned, the total number of active sites increased, platinum atom utilization improved, and the core metal protected from corrosion and dissolution [7-9].

The morphology of nanoparticles directly determines their exposed crystal facets and surface energy, thereby affecting catalytic activity. For example, platinum-based nanocrystals can be synthesized into various morphologies such as cubes, octahedra, tetrahedra, and nanosheets. As shown in Figure 3, nanostructures with high-index crystal facets (for instance, PtNi nanowires and PtCuNi tetrahedra) generally exhibit higher catalytic activity because higher-index facets contain more low-coordinated atoms (steps, kink sites), which are often the active centers for electrocatalytic reactions like the ORR [10-12]. Core-shell structure design usually refers to coating one or several layers of platinum atoms or platinum alloy shells on the surface of non-precious metal or inexpensive transition metal cores (such as Ni, Co, Cu, Fe, Pd, etc.) [8,9,13]. The advantages of this structure are: (1) the core material can reduce the amount of platinum used, thereby lowering costs; (2)strain effect (compressive or tensile strain caused by lattice mismatch) and the electronic effect (ligand effect) between the core and shell can modulate the center of d-band of the platinum surface, optimizing the adsorption energy of intermediates in the ORR and thus enhancing intrinsic activity; (3) The platinum casing can effectively prevent the internal core metal from dissolution and

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corrosion, while the core can also inhibit platinum migration and aggregation, improving catalyst stability [9,14,15].

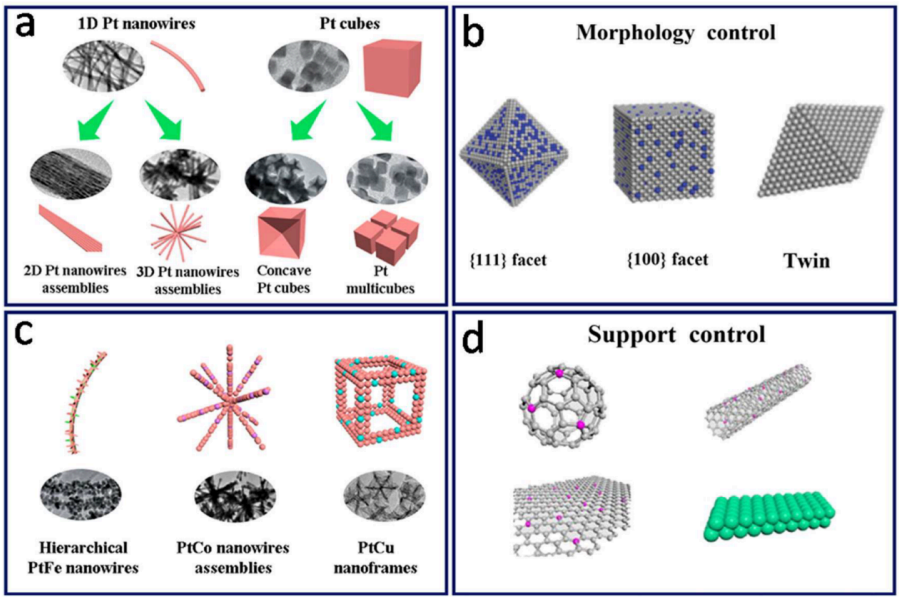


Figure 3. (a) Structural adjustment (b) Morphological control (c) Nanostructure (d) Carrier improvement [2]

There are various methods for synthesizing core-shell structures, including underpotential deposition followed by displacement, chemical reduction, thermal annealing, and dealloying. For example, PtCo or PtNi alloy nanoparticles with a Pt-enriched surface layer can be prepared via dealloying, which exploits the different dissolution rates of metal elements under corrosion conditions to form a high-Pt layer on the surface. Additionally, using metal-organic frameworks (MOFs) or other template-assisted synthesis allows precise control of the core-shell structure. As an example, a catalyst composed of sub-5 nm PtCo nanoparticles coated with a platinum skin and supported on a single-atom Co-N-C carrier achieves structural stability through the barrier effect of Co anchor points, with a mass activity as high as 2.71 A mgPt^-1 and negligible stability loss after 40,000 cycles.

The "skin effect" of core-shell structures (Pt-skin) is one of the important mechanisms for enhancing ORR activity. When the platinum shell is thinned to 1-3 atomic layers, the electronic and strain effects of the core metal can be maximally transmitted to the platinum surface, effectively modulating the electronic structure of platinum [8,9,13]. The oxygen reduction (ORR) activity of the catalyst with a Pt/Fe alloy core and a Pt rich shell structure strongly depends on the degree of order. Among them, the highly ordered PtFe/Pt catalyst demonstrated outstanding mass activity (0.92 A $mgPt^{-1}$) in H_2-O_2 fuel cells [8]. Density functional theory (DFT) calculations indicate that the high activity of this catalyst is because the nuclear-induced compressive strain on the platinum shell and its optimized D-band center [8,13].

In addition to binary core-shell structures, multi-core-shell structures have been extensively studied to further optimize performance. For example, In alkaline media, the PT-Trimer /CoPd catalyst exhibits outstanding mass activity for cathodic reactions, significantly outperforming commercial Pt/C catalysts (30.6 times that of commercial Pt catalysts) and excellent durability (no activity degradation after 322,000 potential cycles) [16]. This unique structural design achieves a performance leap through the charge localization effect of the platinum trimers and the stability of

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the core-shell structure. Furthermore, combining core-shell structures with other optimization strategies, such as support engineering, can produce synergistic effects. For example, loading PtCo core-shell nanoparticles on a Co-N-C single-atom support leverages strong interactions of electrons between the support and the core-shell, this strategy significantly enhanced the material's bifunctional catalytic activity for oxygen reduction reaction and ethanol oxidation reaction , demonstrating a synergistic effect [13].

2.2. Carrier material optimization

Carrier materials play a significant role in platinum-based catalysts. They can not only disperse and fix metal nanoparticles, but also enhance the number of active sites due to their large specific surface area. At the same time, they also have an impact on electron conduction, mass transfer, catalyst stability and metal-carrier interaction (SMSI) [17-19]. Although traditional carbon carriers (such as carbon black) have the advantages of cost and conductivity, they are difficult to withstand the high potential and acidic corrosion in the working environment of fuel cells. Carrier corrosion can directly lead to the agglomeration and shedding of platinum nanoparticles, which in turn causes catalyst deactivation [17,20,21]. Therefore, the optimized design of support materials, including the development of novel carbon materials, non-carbon materials, and the modification of traditional carbon materials, can significantly improve the overall performance of platinum-based catalysts [7,19,22].

Carbon materials, because of their excellent conductivity, chemical stability, and high specific surface area, remain the most important type of support for platinum-based catalysts. Besides traditional carbon black, new carbon materials such as carbon nanotubes (CNTs), graphene, reduced graphene oxide (RGO), ordered mesoporous carbon (OMCs), carbon nanofibers (CNFs), and heteroatom-doped carbons (e.g., nitrogen-doped carbon) have been widely studied and applied as supports for platinum-based catalysts [17-19]. For example, carbon nanotubes, with their unique one-dimensional tubular structure and high mechanical strength, can effectively inhibit the agglomeration of Pt nanoparticles and promote rapid electron transfer. By etching carbon nanotubes, more surface defects and functional groups can be introduced, enhancing their interaction with Pt nanoparticles. A Pt3Fe alloy catalyst synthesized at low temperature loaded on etched carbon nanotubes demonstrated excellent ORR activity and stability [23]. Graphene and reduced graphene oxide (RGO) possess an ultra-large specific surface area and excellent electronic conductivity, enabling the uniform dispersion of Pt nanoparticles and improving Pt utilization. Studies showed that Pt/RGO catalysts prepared by an optimized reduction method illustrate higher Pt utilization, electrochemical active surface area, and stability of Pt/RGO catalysts compared to conventional catalysts, enhancing 17% of the maximum power of density when applied to PEMFC cathodes [19].

Single-atom metal-nitrogen-carbon (such as Co-N-C, Fe-N-C) material has gathered widespread attention in recent years as a special type of carbon-based support or synergistic catalytic component. Loading Pt-based nanoparticles onto M-N-C materials can utilize the synergistic effects between atomically dispersed metal sites in M-N-C materials and platinum-based system, as well as strong metal-support interactions, to further increase ORR stability and activity [13,24,25]. As shown in Figure 4, PtCo intermetallic compound nanoparticles loaded on a single-atom Co-N-C carrier, leveraging direct electron transfer between PtCo and Co-N-C and the barrier effect of Co anchoring sites, achieve extremely high ORR mass activity (2.71 A $mgPt^{-1}$) and excellent stability [13].

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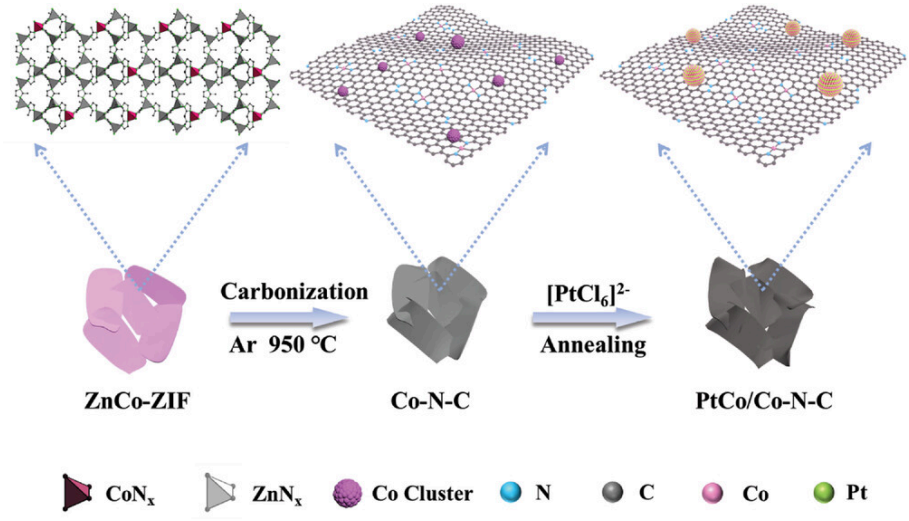


Figure 4. Schematic diagram of Pt-Co/Co-N-C synthesis [12]

2.3. Single-atom optimization strategy

Single-atom catalysts refer to catalysts composed of metal active centers dispersed at the atomic level on a carrier. They have the highest atom utilization efficiency (theoretically up to 100%), well-defined active site structures, and unique electronic properties. Platinum-based single-atom catalysts and their synergistic systems with other single atoms provide new ideas for further reducing platinum usage and improving catalytic performance [26].

The core of platinum single-atom catalysts (Pt SACs) lies in dispersing Pt atoms individually on suitable carriers, the fixation of each single platinum atom through strong metal-carrier interaction (SMSI), and the adjustment of its electronic structure to optimize the adsorption energy of reaction intermediates. For example, Pt atoms fixed on chromium nitride nanoparticles (Pt SACs/CrN) via atomic layer deposition (ALD) exhibit extremely high mass activity in the hydrogen oxidation reaction and excellent CO tolerance [27]. DFT indicate that the interaction between oxygen atoms and Pt single atoms in the CrN support weakens Pt's adsorption of CO, thereby enhancing CO tolerance [28].

Constructing a composite system of single atoms and nanoparticles is an important strategy for optimizing single atoms. Platinum single atoms, platinum-based nanoparticles, and other metal nanoparticles are combined to leverage their respective advantages and achieve a synergistic effect, thereby enhancing the overall catalytic performance [13,29,30]. Figure 4 shows a symmetry-breaking Ru nanoparticle catalyst, where Co single atoms are embedded into Ru nanoparticles to form parasitic RU-CO double single atoms (Co1Ru1, n/rGO) on reduced graphene oxide (rGO). In this structure, oxygen-affinity Co atoms disrupt the local symmetry of ruthenium nanoparticles, promoting the formation of *H2O and accelerating the HOR kinetics [30].

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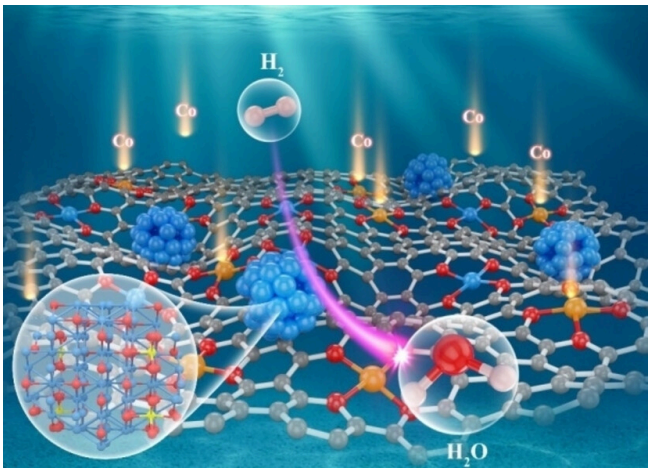


Figure 5. The symmetry of Ru nanoparticles is affected by the embedding of Co atoms, inducing the release of Ru atoms and facilitating the Co1Ru1/n-rGO structure formation. This structural modulation effectively adjusts the interaction between active sites, enhance the kinetics of reduction reactions, improves the activity of the catalyst, and ensures durability [30]

The selection and modification of carriers can further improve the catalytic performance of single-atom platinum. Commonly used carriers include N-doped carbon materials (such as CNT, graphene, MOF-derived carbon), metal oxides (such as CeO_2, TiO_2), and metal nitrides (such as CrN, TiN), etc. [27,28,30]. The N atoms in N-doped carbon carriers can act as anchor sites to stabilize monoatomic metals by forming M-Nx (such as Pt-N4) structures [27,31,32].

2.4. Strain engineering

Strain engineering is an important strategy that optimizes the electronic structure and surface chemical properties of platinum-based catalysts by regulating their lattice strain state (compressive strain or tensile strain), thereby enhancing catalytic activity and stability. Lattice strain mainly arises from differences in atomic radii during alloying, lattice mismatch between the shell and core in core–shell structures, and size effects or exposure of specific crystal facets of nanoparticles. This strain alters the bond length and electron cloud distribution of Pt atoms, thereby affecting the position of the Pt d-band center. According to the Sabatier principle, it optimizes the adsorption energy of reaction intermediates (such as O* and OH* in the oxygen reduction reaction (ORR)) on the Pt surface, enabling the catalytic reaction to reach the optimal kinetic state [33].

Alloying is one of the main methods to introduce lattice strain. When Pt forms an alloy with metals having less atomic radii (such as Fe, Co, Ni),because of the difference in atomic radii (e.g. Pt: 139 pm Co: 125 pm, Ni: 124 pm,), the smaller metal atoms cause the Pt lattice to contract, generating compressive strain [8,13,14]. As shown in Figure 5, in PtCo or PtNi alloys, Co or Ni atoms integrate into the Pt lattice, leading the length of Pt-Pt bond decreases and the d-band center of Pt has a downward shift that weakens the adsorption strength of intermediates like OH*, thereby accelerating the kinetics of the reduction reaction [8,10]. Studies have shown that PtCo intermetallic compound nanoparticles with an L_1 ordered structure exhibit more uniform and stronger compressive strain compared to disordered alloys, thus demonstrating higher ORR activity and stability [8,10].

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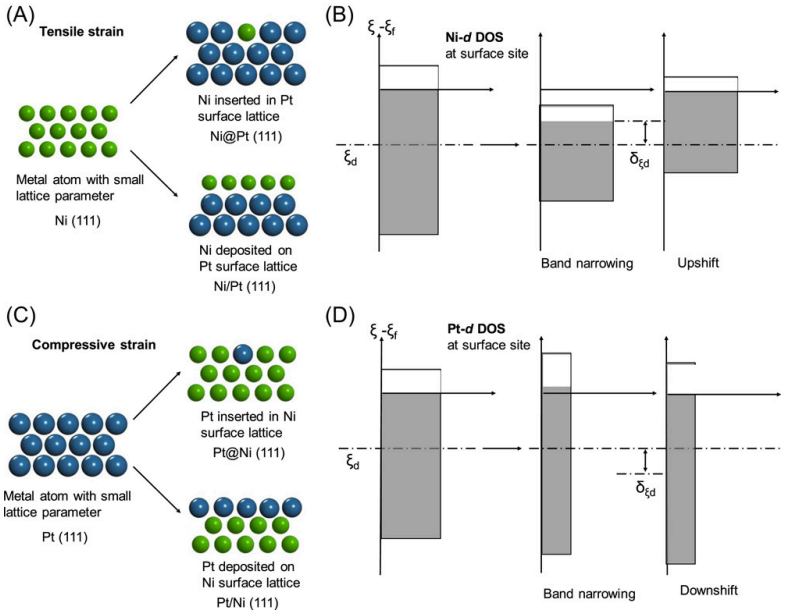


Figure 6. (A) Lattice strain - stretching. (B) The center of the d band of Ni - stretching. (C) Lattice strain - compression. (D) The center of the d band of Pt - compression [3]

Core-shell structure design is an effective means to achieve controllable strain engineering. By selecting core materials with different lattice parameters and coating their surfaces with Pt or Pt alloy shells, controllable compressive or tensile strain can be introduced through lattice mismatch between the core and shell. In the core-shell structure with Pd as the core and Pt as the shell (Pt/Pd), since the lattice constant of Pt (3.92 Å) is slightly higher than that of Pd (3.89 Å), the Pt shell experiences slight compressive strain, thereby optimizing its ORR activity [9]. A more typical example is the nanostructure with an ordered intermetallic compound core (such as L1-PtCo) and a Pt-rich shell, where the ordered arrangement and smaller lattice constant of the core provide Pt shell stable compressive strain [10,13].

The synergistic effect of strain with other effects (such as electronic effects, ligand effects, and metal-support interactions) is an important research direction for optimizing platinum-based catalysts through strain engineering in the future. For example, the electronic effect and strain effect introduced by alloying often coexist, jointly regulating the d-band center of Pt. It should be noted that in core-shell structures, the core not only provides strain effects but also influences the Pt shell electronic properties through electronic transfer. Metal-support interactions (such as electron transfer between Pt and N-doped carbon supports) can also synergize with strain effects to further optimize catalytic performance [34].

3. Challenges faced by platinum-based catalysts

3.1. Resource scarcity and cost issues

Platinum, as a rare precious metal, its content in the Earth's crust is only 5×10^{-7}%. This scarcity directly leads to its high market price, becoming the core factor restricting the cost reduction of proton exchange membrane fuel cells (PEMFCs) [35]. The cathode reaction in PEMFCs is kinetically very slow, often requiring a higher loading of platinum-based catalysts to ensure sufficient output power, which further intensifies the dependence on platinum resources and the overall cost of the cell [4,19]. Statistics show that catalyst costs dominate 30%-50% of the total cost

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of PEMFCs, with platinum costs being the largest component [35]. This high cost makes it difficult for PEMFCs to have price advantages in competition with traditional internal combustion engines and other battery technologies, severely hindering their large-scale promotion in transportation, distributed power generation, and other fields [36].

Although we have made lots of significant progress in the research of low-platinum catalysts, developing high-performance non-platinum catalysts remains a long-term goal to completely eliminate reliance on platinum. In recent years, non-platinum ORR catalysts, such as single-atom catalysts (SACs), metal-nitrogen-carbon catalysts, high-entropy alloys, and non-metal-doped carbon materials, have attracted widespread attention due to their performance in specific reactions that is comparable to or even superior to platinum-based catalysts. Atomically dispersed cobalt cluster catalysts (CoAC/NC) exhibit excellent ORR activity in alkaline media, with a half-wave potential (0.887 V vs. RHE) higher than that of commercial catalyst, demonstrating much higher power density and long-thermostability [37,38]. However, the long-term stability and activity of these non-platinum catalysts in operating conditions, as well as their integration and performance in membrane electrode assemblies (MEAs), still require further improvement to meet the PEMFCs stringent requirements [39].

In summary, the scarcity of platinum-based catalysts and their high cost are multifaceted issues, involving multiple aspects from raw material supply to catalyst design, preparation processes, and the development of alternative materials. Although approaches such as nanostructure regulation, alloying, and support optimization can improve platinum utilization and reduce usage to some extent, seeking truly low-cost, high-performance non-platinum alternative catalysts, as well as developing new fuel cell systems that demand less catalyst (such as AEMFCs), is the long-term solution to fundamentally address this problem [40].

3.2. Limitations of catalytic stability and activity

The catalytic activity and stability of platinum-based catalysts in PEMFCs are key factors determining the lifespan and performance of the cells. Although many strategies such as morphology control, alloying, core-shell structure design, and support modification can significantly enhance their initial catalytic activity, The reduced activity and inadequate stability of platinum-based catalysts under the harsh operating conditions encountered in fuel cell applications—such as exposure to acidic or alkaline electrolytes, high potentials, repeated charge-discharge cycles, and temperature fluctuations—continue to represent a major bottleneck hindering their widespread commercialization [41].

3.3. Inherent limitations and influencing factors of catalytic activity

The activity of platinum-based catalysts still faces many challenges under practical operating conditions. Firstly, the kinetic limitations of ORR and HOR vary in different electrolyte environments. In acidic PEMFCs, ORR kinetics are slow, requiring highly active catalysts; whereas in alkaline AEMFCs, although ORR kinetics improve, HOR kinetics significantly decrease, posing different demands for catalyst design [42,43]. Secondly, the electrochemically active surface area (ECSA) of the catalyst is a key factor affecting mass activity. Reducing the size of platinum nanoparticles can increase the ECSA, but when the particle size becomes too small (such as less than 2 nm), due to quantum size effects and increased surface energy, the stability of platinum atoms sharply decreases, making dissolution and agglomeration likely, which in turn leads to activity loss [44].

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4. Low-platinum / non-platinum

4.1. Tuning adsorption strength

The ORR has the highest performance through the adsorption of oxygen-family intermediates onto the catalyst surface not too weakly and not too strongly. This “just-right” window appears as the peak on a textbook volcano plot and can be modified by changing the metal’s surface electrons (the d-band), by alloying or design of the structure [44]. Given that goal, recent computational efforts demonstrate how to move binding around in reality: either they pair single-atom sites that stabilize OOH in an alternate way from OH or they pair nearby sites to work together, and both strategies may bring the system to the top of the volcano [45].

Two concrete paths are primary on the Pt side. Firstly, alloys containing Pt (Pt–Ni/Co/Fe) reduce the effective oxygen-binding energy through shifting the d-band which permits OO(H)* to bind more at “just right” and this explains why the mass-activity is increased toward lower Pt loading [44]. Second, core–shell catalysts consist in a very thin Pt shell on a cheap core, causing strain that tunes adsorption onto the Pt surface, and recent PtNi at Pt/C has reached 0.58 mA μg Pt^{-1} at 0.9 V and maintained 90% of its activity after 10000 cycles, indicating both binding-tuning effect and durability requirement – that the shell is not cracked [46]. To decrease the usage of Pt more, single-atom/cluster Pt enhances atom efficacy, but only if the atoms are heavily anchored down so that they don’t move, a point highlighted in a 2024 single-atom Pt review [47] and nicely illustrated by (in another reaction) by substituting Pt atoms into α-MoC, where lattice anchoring allows isolated Pt atoms to remain in place; the chemistry explains why “anchoring” governs adsorption on an ultra-dilute metal site [47,48].

The non-Pt side employs the same adsorption approach, though it provides specific coordination of earth-dominated metals. In Fe–N–C, the FeN_4 site is also a fixed “seat” for binding ORR intermediates; a 2024 investigation demonstrated that adding a small Br^- effect can enhance FeN_4 (ligand field and Fe–N bond length) and therefore allow OOH/OH to bind optimally and elevate both half-wave potential and MEA power, demonstrating a “straightforward model of coordination control to fine-tune adsorption” [49]. In their companion in-situ paper, a companion analysis follows Fe–N–C during pyrolysis to demonstrate how FeN_4 motifs will be formed to become genuinely active configurations, once again linking performance with the type of manner in which the site binds oxygen pieces in a single molecule [50]. Going a Step Further, dual-atom sites (e.g., Fe–Co, Fe–Cu) allow two adjacent metals to co-regulate and release intermediates, interrupting some of the normal OOH-OH scaling and therefore shifting the volcano peak towards less costly metals, theoretical and screen studies suggest a number of dual-atom combinations that are also located within or approaching the peak of activities [45,51]. The bottom line, regardless of whether we are alloying Pt, constructing core–shell strain, pin Pt into single-atom sites or engineering FeN_4/dual-atom motifs, the consistent theme is to tune adsorption to the “just-right” range and control this within the “just-right” window [44,45] for the fuel-cell at its full extent (the common point of fixation/reduction).

From the low-Pt alternatives, alloy/core–shell seems more ready for deployment in the form of MEA durability reports, when we consider the performance, cost and safety as compared to Pt only on the basis of papers. PtNi at Pt/C work combines micro-kinetics and an experiment to connect strain-weakened HO* binding with the high mass activity and then shows only small after long-term cycling loss despite Ni DE alloys, because the structure and performance of Pt skin and carbon anchor is intact [46]. So, effectiveness is high, the usage of Pt is reduced, with safety risks, such as Ni leaching, are controlled by design. In contrast, the single-atom Pt review emphasizes the fact that,

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although Pt utilization is excellent, site migration and support corrosion still represent the durability barriers in realistic MEAs [47]. Overall, alloy/core–shell route is safer (not lifetime proven) for PEMFC stacks while single-atom Pt looks promising [44,46,47].

Under the PGM-free framework, Fe–N–C along with tuned FeN_4 has also been the driver performance on device-based activity, while dual-atom ideas have not yet been established. The Br-assisted Fe–N–C paper, following the coordination-level optimization approach proposed this protocol, achieves the current-density target of the DOE by integrating coordinated level adsorption tuning using a more efficiently developed ionomer interface [49]. Yet the acid-side durability threat of Fe–N–C is not clear, and hence the same literature now focuses on lifetime (as well as activity) for future studies [50]. The dual-atom sites might share intermediates and enhance the scaling limits, although the MEA-maturity at this stage is lower [45,51]. We do not state this out on the open stage where available research does not offer enough support to make the performance ranking strict. For instance, single-atom Pt cannot yet be taken as superior to Pt alloy/core–shell in terms of full-stack durability, due to the reviews primarily relying on migration and changes in the state to be obtained in response to PEMFC cycling [47]. Just as the evidence for MEA is still early, the dual-atom design cannot be considered as better than Fe–N–C in acid MEAs [51]. These aren't negatives for the concept, but rather signs that this technology isn't mature and that real MEA testing is still catching up to RDE/DFT promise.

4.2. Change to catalyst environment

The present section shifts the attention away from the active site to its environment: the structure which supports particles; the ionomer film to host protons; the pores carrying oxygen and water; the chemistry that creates and/or degrades peroxide. In real MEA, these details determine whether a site that seems flawlessly ideal on paper will remain easy to reach and stable in high current. Reviews on PEMFC durability and on catalyst-layer interfaces, for example, show that at high potential, acidity, and humid heat the Pt can dissolve and damage ionomer. Thus, engineering the local environment is a first-order design lever for lifetime [52,53].

Support–ionomer pair is the natural starting point due to these materials touching virtually every catalytic site. When the carbon support is highly graphitized and covered as much or as little as a thin layer, it will withstand oxidation, the nanoparticles are stable, and the electrochemically active area of the surface decays more slowly; meanwhile, a proper placement of an ionomer layer dispenses protons on the surface without smothering it, reducing transport blockages at elevated currents [52]. Durability studies link these materials selections closely to operating conditions: expanding the potential window or forcing a very high humidity and temperature facilitates Pt dissolution and promotes corrosion, while decreasing the window, and avoiding sustained flooding, delay loss of area and mass activity [53]. In practice all of which does is mean that the "best" micro-environment for a "low-Pt" alloy or core–shell catalyst, both carbon-wise and for an intelligent ionomer film, is a better carbon one and also a disciplined operation that always keeps voltages and humidity at safety-considered levels. Once the support-ionomer scaffold is installed, the water/oxygen flow is then opened a gate. When the pores and surface energies are not fine-tuned, cathodes either flood (oxygen is unable to get to the sites) or dry out (protons cannot pass across the film). The interface discussion favors a regulated hydrophobic/hydrophilic equilibrium, as well as the design of pores capable of removing water without starving the ionomer of hydration which would preserve the simultaneous arrival of O_2 and protons at a high current density [52]. Since flooding raises local potentials and promotes corrosion, the smart transport design and operating

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window control above reinforce each other — one holds access open, the other avoids the electrochemical conditions that would reverse it [53].

By contrast, for PGM-free cathodes in acid, the environmental problem moves from transport to peroxide and radical chemistry. Fe–N–C and other M–N–C-related materials can also be produced for the manufacture of H_2O_2 and, from Fenton reactions, radicals capable of destroying the ionomer, the carbon and occasionally the metal site. For that reason, the interface review recommended the graphitization of the carbon with skin or coatings protection, ionomer–catalyst boundary adjustment that suppresses/scavenges reactive oxygen species prior to their harming the layer; i.e., we engineer the environment to protect the site and only then stress on transport. At the full electrode scale, architecture and loading merge the above concepts. The graded ionomer through thickness, proper inter-particle distance, and well-chosen pore sizes allow all active sites to behave in much the same manner, which reduces mass transport losses at the same time as ultra-low PGM loadings. Durability data again link these structural choices to operating practices: limiting upper potential limits and preventing long, wet high-temperature holds decrease Pt dissolution and particle growth, so the architectural advantages are not wiped away by cycling [53]. While we are targeting acidic forms of PEMFC, the geometry of the micro-environment also explains why certain alkaline routes appear attractive. In base, carbon corrosion decreases, and many oxides become more stable, thus Mn-oxide families are a cheaper, less toxic option if their conductivity and site density can be designed according to their nature. The findings have contributed to research-based work on the stability of ORR of MnOx in alkaline and even for a broad pH range using its composition as conductive ink to demonstrate that once the membrane/ionomer stability of AEMFCs has been addressed, micro-environment design will allow these earth-abundant catalysts to retain activity for extended time periods of time [54,55].

The benefits of NPF in these systems are not transferable to acid PEMFCs unchanged; in acid the peroxide problem for M–N–C is still the dominant lifetime barrier and so the success in alkaline of non-Pt technology is supportive rather than substitutive [52,54]. Based on these links, a reasonable horizontal comparison is made. In acid and lower Pt cathodes, the most deployable micro-environment device is corrosion compatible support and the well positioned ionomer film and the controlled operating window, the three acting on the recognised degradation channels and already introduced into the MEA protocols to improve the lifetime and without introducing new materials risk [52,53]. While water and oxygen management are important in increasing current, it performs very well as supplementary elements to the support–ionomer fix rather than as a single solution as transport cannot prevent electrochemical dissolution [52,53]. For PGM-free cathodes in acid, the fundamental tool is peroxide-aware interface design–graphitized or coated carbons and tuned ionomer boundaries–as a means of protection for Fe–N–C sites, while for alkaline systems the material environment makes Mn-oxides functional by inhibiting corrosion and permitting pH-robust inks, even though this is not yet able to solve acid-side durability [52,54,55].

A careful conclusion must be drawn. The papers do not support a strict ranking in which water-management architectures alone beat support/ionomer engineering for lifetime performance in acidic PEMFCs, since potential-driven Pt dissolution and carbon corrosion are strong unless the interfaces are stabilized; alkaline Mn-oxide environments cannot be claimed to outperform Fe–N–C in acidic PEMFCs because the strongest Mn evidence is found in alkaline or pH-flexible devices [52-55]. We can confidently state, now, that two micro-environment strategies are the most feasible: first, for low-Pt PEMFCs, support/ionomer engineering is required, and the operating window should be tightly controlled as this combination prongs into the main degradation pathways while leaving the transfer path intact; second, for PGM-free cathodes, the design of interface using peroxide-aware

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interface design in acid and the prospect for alkaline Mn-oxide routes where stable oxide and well-pH-robust inks already demonstrate long-life, low-cost operation [52-55].

4.3. Alternatives to PEMFC based on AEMFC

Switching the membrane environment from acidic (PEMFC) to alkaline (AEMFC) changes rules about the active site and about the site of action. When the membrane surface is more oxide-covered at base, and ORR potential is lower versus SHE, non-PGM surfaces can easily take part, at least thermodynamically, at the same time as real cells face new constraints: water is forced to back-diffuse from anode to cathode, CO_2 from air carbonates the OH^- and causes very large voltage losses, the polymer: ionomer chemistry has to survive at high pH [56]. In short, AEMFCs broaden the materials palette, but also lead to water/CO_2/membrane problems that need to be engineered together with the catalyst. From the point of view of cathode (ORR), two PGM-free families are especially noticeable in the alkaline medium.

The first one is transition-metal oxides based on spinel (AB_2O_4) or perovskite (ABO_3) structures. They may also be tuned by the B-site metal, eg-orbital filling, oxygen vacancies, and by coupling to conductive carbon to solve low conductivity; several Mn/Co/Ni spinel's and La-based perovskites demonstrate heavy ORR/OER bifunctions in alkaline and relatively resistant to acid [55]. Mechanistically, these lattices allow for oxygen intermediates to be placed at near-optimal binding through B-site chemistry and the carbon composite to provide electronic pathways. To put it briefly, oxide–carbon composites provide you an easily modifiable adsorption and alkaline-stable scaffold.

The second method is M–N–C and earth abundant single-atom catalysts, SACs. Alkaline isolated Fe/Co/Ni–N_4 motifs exhibits $4e^-$ ORR selectivity and great intrinsic turnover, due to the site geometry ensuring OOH* and OH* adsorption within a narrow range of "just-right", and the SAC literature summarizes how the well-defined coordination and strong host–guest bonding determine binding energy and sintering resistance [49,50]. SACs show high site precision with atom economy compared to oxides but require further development of carbon/ionomer interfaces for mass transport and for extended durability. On the anode (HOR), AEMFC is expected to use PGM-free catalysts (e.g., Ni-based), although the literature suggests the current situation is clear: the fully PGM-free AEMFCs have not achieved performance or stability, and cell containing Pt/C or PtRu/C at one or both electrodes generally obtain multi-W cm^{-2} power. On the flip side, the perspective suggests that, in order to outperform PEMFCs on price, AEMFCs must eventually go PGM-free at both electrodes, but it also emphasizes their inherent limitation in being PGM-free HOR, which is mechanistically linked up with ionophilicity and interfacial water in base [56].

Having these threads, we can assess which one of the PGM-free cathode approaches has more potential to be developed compared today. Spinel/perovskite–carbon composites appeal for their robustness and simplicity-the tuning knobs (B-site substitution, oxygen vacancies) offer chemically scalable and alkaline stability is by nature superior to that of acid; these come at the price of lower conductivity unless you balance carbon coupling, and where particle dispersion is manipulated to prevent the thick films from forming [55]. Earth-rich SAC/M–N–C cathodes on the other hand provide excellent site efficiency with prospects to transition toward $4e^-$ ORR selectivity, but lifetime is determined by the carbon, and the ionomer boundary so micro-environment engineering (hydrophobic balancing, peroxide management) will continue to be at least as important as site design [49,50]. Oxides make for materials that are tough, SACs make for sites which are precise; both are PGM free, both of which require an excellent electrode structure to be designed to convert activity into cell electricity. A fair conclusion (with room to maneuver inside the papers) is that we cannot yet determine whether one cathode family is more or less superior by cost–safety–

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effectiveness. Oxides appear manufacturable and chemically rough-hewn in alkaline, but are subject to transport and conductivity penalties; SAC/M–N–C appears highly efficient and tunable but has to demonstrate MEA-level durability over thousands of hours. As anode, PGM-free HOR is nascent, the present AEMFC stacks developed with real power target are still low-PGM HOR whilst the community is investigating nickel-centered designs and interfacial water control [56]. AEMFCs are attractive only because alkaline environments will give PGM-free cathodes real competition, but full systems must be the resolution of HOR catalysis, water routing, and CO_2 scrubbing as coexisting with the catalyst. The papers here generally agree on that priority arrangement and arrange the research pathway accordingly.

5. Conclusion

Significant progress has been made in the research of platinum-based catalysts for hydrogen fuel cells, but it still faces many challenges and opportunities. Over the past few decades, the activity and stability of platinum-based catalysts have been significantly enhanced, and the usage of platinum has also been greatly reduced. Through strategies such as alloying, nanostructure design, carrier optimization and single-atom catalysis, the performance of platinum-based catalysts has continuously broken through its limits. Future catalyst research should pay more attention to scenario orientation, carry out material design and performance optimization in combination with specific application requirements, and strengthen the collaborative innovation of catalysts and MEA integrated processes to promote the commercialization process of hydrogen fuel cell technology. Meanwhile, the research on low-platinum and non-platinum catalysts will also provide more directions and ideas for future fuel cell technologies.

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