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Research Progress on Pt-based Catalysts for Proton Exchange Membrane Fuel Cells

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Abstract. With high power density, fast low-temperature response and no carbon emissions, proton exchange membrane fuel cells (PEMFCs) are widely employed for transport and distributed power generation. However, their large-scale commercialization is currently impeded by high manufacturing costs and limited durability. One major obstacle is sluggish cathodic oxygen reduction, which currently requires a substantial loading of costly platinum (Pt) catalyst. Therefore, researchers come to focus on the development of advanced Pt-based catalysts that can minimize Pt loading while maintaining high performance and intrinsic stability. This article provides an overall review of recent progress in platinum-based PEMFC catalysts, offering a comprehensive overview of strategies designed to improve the electronic structure and adsorption behavior of platinum surfaces. Specifically, the review concentrates on four key areas: the scale and morphology effects of platinum nanoparticles, the development of platinum alloys (PtM), core-shell structure design strategies, and the catalytic mechanisms of single-atom catalysts (SACs). Furthermore, the paper discusses future development directions and the industrialization prospects of these advanced materials.

Keywords: PEMFC, Pt-based catalyst, ORR

1. Introduction

Proton exchange membrane fuel cells (PEMFCs) are promising for clean and distributed power because they combine high power density with rapid start-up at relatively low temperatures. However, high manufacturing costs and limited-service life remain the core bottlenecks which restrict the large-scale commercial application of PEMFCs. Because cathodic oxygen reduction is intrinsically slow, PEMFCs depend on relatively high loadings of platinum (Pt) to obtain an adequate reaction rate [1].

Commercial PEMFC cathodes generally employ Pt nanoparticles dispersed on carbon, although other Pt-rich materials have also been investigated. However, the prohibitive cost of Pt hinders the large-scale application of them, so it's a critical direction for the research and development of Pt-based catalysts to reduce the size of Pt nanostructures, prepare single-atom catalysts (SACs), as well as synthesize core-shell structures to enhance the inherent catalytic activity of Pt [2]. Recent research has mainly concentrated on optimizing the surface electronic structure and adsorption

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behavior of Pt by improving synthesis pathways, alloying, or microstructure design to meet that demand.

This review summarizes the latest research progress in platinum-based catalysts for PEMFCs systematically, paying more attention to the size/morphology effects of Pt nanoparticles, PtM-based transition metal alloy catalysts, and the construction strategies and catalytic mechanisms of SACs. Additionally, this review provides an outlook on the future development directions and industrialization prospects of this field.

2. Research background

2.1. The reaction mechanism of PEMFC

Figure 1 shows the earliest prototype of a fuel cell [3]. Hydrogen and oxygen are separately introduced into dilute sulfuric acid, where they can react to generate a small electric current. The anode undergoes an oxidation reaction where hydrogen diffuses to the catalyst layer, reacting catalytically to produce electrons and protons (equation 1). The cathode of the cell undergoes an oxygen reduction reaction, and air diffuses through the cathode catalyst layer reacting with protons and electrons to produce water (equation 2). Cathodic oxygen reduction is reported to be approximately six orders of magnitude slower than anodic hydrogen oxidation; combining the two half-reactions gives water as the overall product (equation 3).

$$H_2 \rightarrow 2H^+ + 2e^- \qquad E_1 = 0\ V_{SHE} \tag{1}$$

$$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O \qquad E_2 = 1.229\ V_{SHE} \tag{2}$$

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \quad \Delta E = E_2 - E_1 = 1.229\ V_{SHE} \tag{3}$$

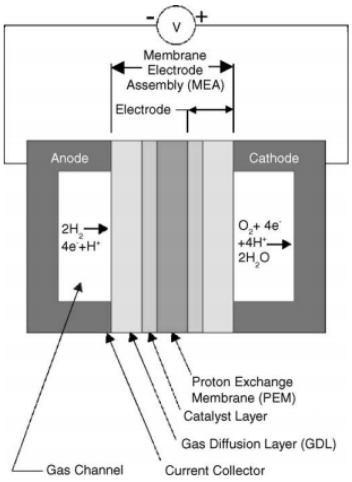


Figure 1. Schematic of a single typical proton exchange membrane fuel cell [3]

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2.2. The ORR mechanism of Pt catalyst in PEMFC

Oxygen reduction is a multielectron process that can follow either a four-electron route or an incomplete two-electron route. The four-electron pathway shown in equation (4) is preferred in fuel cells and is also important in metal-air batteries.

$$In\ acidic\ medium:\ O_2 + 4H^+ + 4e^- \rightarrow 2H_2O,\ \ \varphi^\theta = 1.229V \tag{4}$$

Occurring at the cathode, this process involves oxygen molecules acquiring electrons and being reduced under the influence of the cathode electrocatalyst. Based on the four-electron electrocatalytic oxygen reduction mechanism, the rational development of efficient ORR electrocatalysts to overcome the challenge of slow kinetics rate inherent to ORR has become a key priority in promoting the sustainable energy technologies.

3. Improvement strategies for platinum-based catalysts

There are various types of improvement strategies, and different improvement strategies have different dimensions. Therefore, this article will clarify the advantages of different improvement strategies from perspectives of Pt atom utilization, ORR mass activity, and durability.

3.1. Pt nanoparticles

Platinum nanoparticles in the approximate size range of 2–20 nm may be dispersed in water or other liquid media. Their large surface-to-volume ratio exposes many catalytic sites. ORR behaviour also varies with particle size and surface orientation because high-index planes contain a greater proportion of under-coordinated atoms than low-index facets [4].

To accelerate the application of devices with its zero-emission power of PEMFCs and take full advantage of their power characteristics, Paperzh et al. [5] tested a novel N-doped carbon support, synthesizing Pt/C nanoparticles with a platinum loading of approximately 37.3%. Researchers conducted an experiment in the oxygen atmosphere by the accelerated stress testing (AST) with pulse potential changes within the range of 0.4; 1.0 V. The platinum loading current commercial JM40 analog was 40%, while this material exhibited a larger electrochemical active surface area (50 m^2/g Pt-1), and the electrochemical parameters retained after AST were nearly double those of JM40. Thus, this Pt/C catalyst holds broad prospects for commercial application in PEMFCs.

Darab et al. [6] investigated the impact of various methods for synthesizing on the durability and electrochemical activity of the carbon-supported platinum nanoparticles as catalysts for PEMFCs. Utilizing a same carbon support, they synthesized samples with 10% to 30% platinum loading using four distinct synthesis methods and characterized them for use as electrocatalysts in PEMFCs. Their experiment demonstrated that the 20 wt% sample synthesized via the adsorbed polyol method had superior electrochemical activity, particularly at lower potentials. The membrane electrode assembly (MEA) equipped with this catalyst produced a significantly higher current density than other samples. Its current density was almost twice that of the 10 wt.% sample at a cell voltage of 0.5 volts. The adsorbed polyol (AP) method produced the most homogeneous Pt/C nanoparticles and the best activity among the tested methods with high compositional homogeneity and including only individual particles.

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In general, the nanoscale transformation of platinum helps reshape its electronic configuration, surface geometry, and thermodynamic state. This endows nano-platinum with more active sites, superior molecular adsorption capability, higher intrinsic activity, and better selectivity in catalytic reactions.

3.2. Ptm alloy

Platinum alloy (PtM) is an alloy material composed of platinum as the matrix and one or more metal elements. Further research on PtM alloys can effectively enhance the ORR activity and durability of PEMFCs. Adding a second metal redistributes charge around Pt and can impose lattice strain because the alloying elements differ in electronegativity and atomic size. The resulting shift of the Pt d-band centre changes the binding strengths of reactants and oxygenated intermediates such as *OOH.

Woo-Jae et al. [7] proposed a synthetic scheme for preparing uniform metal alloy nanoparticles of active PtNi alloy structure by the fluidized bed reactor atomic layer deposition (FBR-ALD). His scheme shown in Figure 2 employs the ALD supercycle method to control the component of PtNi alloy, whose experimental data demonstrates that the $Pt_{78}Ni_{22}$ alloy structure synthesized by ALD exhibits higher electrochemical specific surface area (ECSA), durability, and ORR activity compared to the currently commercialized Pt_3Ni alloy.

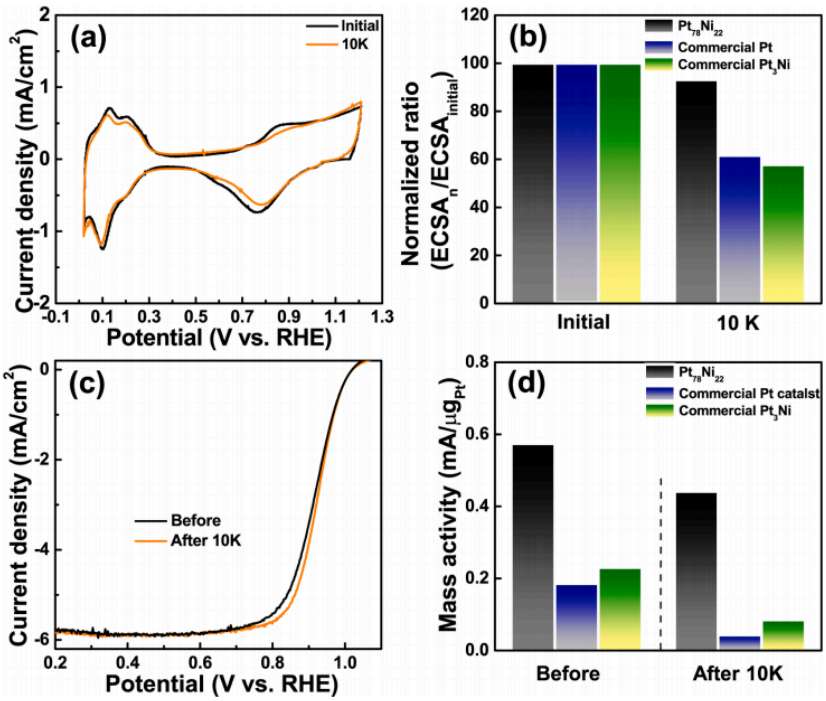


Figure 2. (a) Cyclic Voltammetry durability test of the Pt78Ni22, (b) standard ECSA ratio with ADT testing, (c) ORR polarization curve of Pt78Ni22, (d) the change of quality activity [7]

Geng et al. [8] have designed a platinum-nickel high-entropy alloy nanowire (Pt3Ni @ HEA NWs) as an efficient bifunctional catalyst in PEMFCs. Compared with the currently commercialized Pt/C catalyst, this catalyst exhibits a higher mass ratio in redox reactions. When it's used as both a cathode and anode catalyst, Pt3Ni @ HEA NWs / C can fulfill a high membrane electrode assembly power density which reach 1653.5 mW cm-2 and its long-term stability for 280 hours. This research

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provides a new paradigm for developing comprehensive and efficient Pt-based functional catalysts in fuel cells and other fields.

In addition to PtNi alloy, Belenov et al. [9] had new ideas for the manufacturing method of PtCo/C catalyst. They created a new way for multi-step synthesis of PtCo/C materials with composite Co_xO_y/C, which combined the benefits of different liquid-phase synthesis techniques. The bimetallic nanoparticles of PtCo/C catalyst synthesized by this method are distributed on the surface of the support in a uniform manner, and the size is about 3 nm, so it has high ESA and ORR activities. This synthesis method involving multiple steps by composite carbon oxide support can synthesize the bimetallic nanoparticles (NPS) PtCo/C catalyst with an average particle size of 2.7 nm, and the synthesized PtCo/C catalyst has a specific ORR activity and high active surface area, which is 1.75 times higher than that of commercial Pt/C.

3.3. Core-shell structure

The core-shell structure is an advanced nanomaterial design aimed at reducing the use of precious metals, improving catalytic activity, and strengthening the utilization of platinum atoms. This structure typically manifests as a "platinum-rich shell encapsulating a non-platinum core". However, in a few specific applications, platinum can also serve as the core, surrounded by an inert shell layer such as silica or metal oxide. This structure primarily utilizes the outer shell to shield the internal platinum from harsh environments, thereby enhancing the stability of the catalyst, such as the research carried out by Zhou et al.

Zhou et al. [10] designed a core-shell catalyst named Pt/C@NCL300 to recover the lost ORR activity through a self-assembly process. They made a thin layer derived from Nafion on the surface of Pt/C catalyst, which would coat Pt particles and separate Nafion membrane from Pt particles, as shown in figure 3. Through the reduction of Co replacement coverage of Pt/C@NCL300, the researchers proved that due to the existence of NCL300, the sulfonate adsorbed on the Pt surface decreased, which made the catalyst show a specific activity reaching 434.3μA $cmPt^{-2}$ with excellent ORR activity in RDE test.

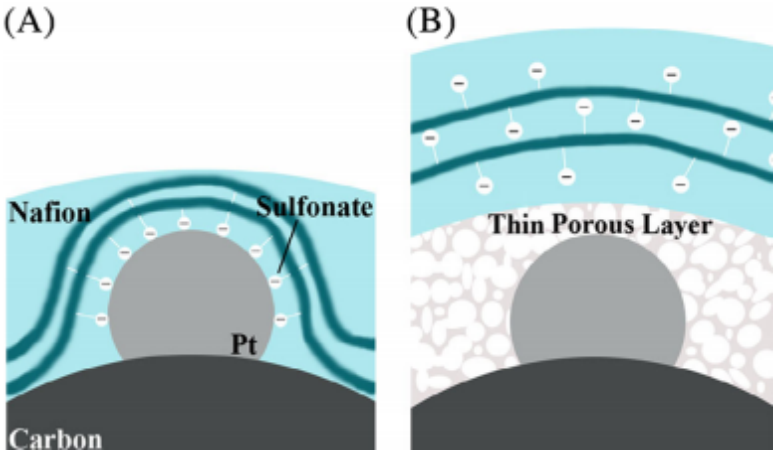


Figure 3. (a) Traditional schematic diagram and (b) new interface structures of the catalyst layer [10]

Shahgaldi et al. [11] and others synthesized a carbon titanium dioxide (C—TiO_2) core-shell structure by hydrothermal method to coat Pt as its carrier. At the same time, researchers used different chemical reagents to increase the surface area of the carrier with controlling the porosity of the carrier. By analyzing the data, researchers found that the maximum power density of 10 wt% C

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—TiO_2 was the highest, about 410 mW/cm^2, with the catalytic action of titanium dioxide. Regarding stability, the surface area of the activated carbon nanospheres decreased by 80% after 3000 cycles, while the surface area reduction of the encapsulated structure and core-shell structure increased to 46% and 27% respectively.

3.4. Single-atom catalysts

Single-atom catalysts (SACs) are attractive for ORR because isolated metal centres can maximize atom utilization. Their catalytic behaviour is governed by the interaction of adsorbed intermediates with the central atom and its coordination environment, including neighbouring heteroatoms in the support.

Choi et al. [12] used S-doped three-dimensional network graphene nanoribbons as carriers to prepare Pt SACs with a mass fraction of 5% Pt. This catalyst has excellent ORR performance, with an initial potential of 0.95 V, comparable to commercial Pt/C catalysts, a current density of 1.7 mA/cm^2, and an electron transfer number of 3.5 in ORR catalysis, approaching the ideal four electron process. The results of density functional theory (DFT) calculations show that the introduction of S element increases the loading of Pt atoms, forming Pt-S_4 active sites with efficient catalytic activity, reducing Pt atom aggregation, and improving the stability of the catalyst.

Liu et al. [13] subjected carbon black 2000 to hydrothermal treatment in a sealed ethanol solution at 200 ℃, and synthesized defective carbon (referred to as BP defect) using the hydrothermal method. With the porous structure of BP defect, it was used as a carrier to anchor Pt atoms, where the mass fraction of Pt loaded was 1.1%. The obtained catalyst was referred to as $Pt_{1.1}$/BP defect. The power density curves and polarization curves of commercial Pt/C, BP, $Pt_{1.1}$/BP, and $Pt_{1.1}/BP_{defect}$ has shown that the maximum power density of this catalyst in acidic H_2/O_2 fuel cells is as high as 520 mW/cm^2, which is prior to the commercial Pt/C and $Pt_{1.1}$/BP catalysts. The Tafel slope is 96 mV/dec, similar to the commercial ones, and it can achieve ideal four electron ORR processes.

4. Conclusions

Nowadays, researchers have made significant progress in four modification strategies, including nano platinum particles, platinum transition metal (PTM) alloys, core-shell structure design, and single atom catalysis, aiming to explore effective strategies to reduce platinum dosage and improve catalytic activity and stability. The future development direction of platinum-based catalysts will focus on the integration of multi-scale structural regulation and advanced synthesis methods. Precise control of atomic arrangement and local electronic structure may improve Pt utilization while mitigating catalyst degradation, thereby supporting broader PEMFC deployment. However, how to achieve a leap from laboratory environments to large-scale industrial production and ensure high product quality remains a key issue that urgently needs to be addressed, while cost, large-scale synthesis, and durability assessment remain major challenges.

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