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MINI REVIEW

Mesoporous carbon materials: Synthesis and applications in proton exchange membrane fuel cells

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Abstract Proton exchange membrane fuel cells (PEMFCs) have attracted significant attention as sustainable energy technologies due to their efficient energy conversion and fuel flexibility. However, several challenges remain, such as low catalytic activity of fuel cell membrane electrode assembly (MEA), insufficient mass transfer performance, and performance degradation caused by catalyst deactivation over long period of operation. These issues are especially significant at high current densities, limiting both efficiency and operational lifespan. Mesoporous carbon materials, characterized by a high specific surface area, tunable pore structure, and excellent electrical conductivity, are emerging as crucial components for enhancing power density, mass transfer efficiency, and durability of PEMFCs. This review first discusses the properties and advantages of mesoporous carbon and outlines various synthetic strategies, including hard template, soft template, and template-free approaches. It then comprehensively examines the applications of mesoporous carbon in PEMFCs, focusing on their effects on the catalyst and gas diffusion layer. Finally, it concludes with future perspectives, emphasizing the need for further research to fully exploit the potential of mesoporous carbon in PEMFCs.

Keywords mesoporous carbon materials, proton exchange membrane fuel cells (PEMFCs), nanocomposites, electrocatalysis, ionomer

1 Introduction

The rapid advancement of society is driving a growing demand for energy. However, around 80% of the global energy mix is still reliant on non-renewable fossil fuels, the excessive use of which leads to a range of environmental issues [1–3]. To mitigate global environmental pollution, it is essential to develop efficient, stable, and environmentally friendly energy sources and technologies. Significant resources and efforts have already been devoted to the development of sustainable energy conversion and storage devices. Among these, proton exchange membrane fuel cells (PEMFCs) stand out as effective energy conversion devices, transforming the chemical energy of fuels into electrical energy. PEMFCs offer several notable advantages, including high power density, rapid startup and response times, and adaptability to various fuel sources [4–6]. However, challenges such as high costs and relatively short lifetimes remain obstacles to their widespread commercialization [7]. Research has demonstrated that specific key functional materials such as proton exchange membranes, gas diffusion layers, and catalysts significantly influence the performance and longevity of fuel cells [8]. Therefore, advancing and optimizing these critical materials is crucial for enhancing fuel cell performance and durability, reducing costs, and making them more commercially viable.

In recent years, mesoporous carbon materials, characterized by pore dimensions between 2 to 50 nm, have gained considerable attention due to their high specific surface area, tunable pore structure, and excellent electrical and thermal conductivity [9]. These properties enhance energy density, improve electrochemical reaction efficiency, facilitate charge transport, and increase cycling stability, positioning mesoporous carbon as a promising candidate for energy conversion and storage applications [10–12]. Additionally, various mesopore morphologies provide specific advantages in different applications. For example, a uniform

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mesoporous structure provides regularly arranged pore channels, facilitating the homogeneous distribution of platinum (Pt) during the oxygen reduction reaction (ORR) process. This arrangement increases active sites and enhances the electrochemical surface area [13]. A multilevel pore structure, containing channels of different sizes facilitates faster transport pathways and larger reactive surfaces, aiding rapid ions migration during high-power charging and discharging, ideal for supercapacitors with extremely excellent rate performance [14]. A divergent gradient pore structure, with pore sizes increasing from the inside to the outside, enhances electrolyte penetration and active site utilization, exhibiting excellent enzyme-like activity [15]. The core-shell structure adds an extra buffer layer for mesoporous carbon materials, reducing volume expansion during cycling and improving structural stability, particularly as anode materials for sodium-ion batteries [16]. Honeycomb mesoporous carbon offers excellent mechanical stability at the nanoscale, while its honeycomb structure minimizes diffusion resistance and facilitates mass transfer in fuel cell membrane electrode assembly (MEA) [17]. Furthermore, mesoporous carbon materials with a unique Janus structure allow for multi-step catalytic reactions within the same material, with each step confined to a specific component, significantly improving the catalytic efficiency of aqueous hydrogenation reactions [18]. In summary, mesoporous carbon materials with a wide variety of mesoporous morphologies show considerable promise for applications in catalysis, energy storage, specific adsorption, and functional encapsulation [19–22].

In recent years, numerous reviews have been published focusing on the synthesis, modification, and applications of mesoporous carbon materials, with detailed discussions by Mehdipour-Ataei and Aram [23], Lei et al. [24], and Li et al. [25]. For electrochemical energy storage and conversion devices, Zhu et al. [26] and Yuan et al. [11] have thoroughly discussed the advantages of mesoporous carbon materials. However, a comprehensive and detailed discussion on the specific role of mesoporous morphology in PEMFCs, along with the underlying mechanisms by which mesoporous carbon materials enhance energy conversion reactions, is still lacking.

This review summarizes recent advancements of the applications of mesoporous carbon materials in PEMFCs. It introduces the properties of mesoporous carbon and three primary synthesis methods—hard template method, soft template method, and template-free method—for mesoporous carbon materials. In addition, it discusses the correlation between the structure and properties of mesoporous carbon materials, including their catalytic properties, ionomer adsorption and distribution, and gas/water transport efficiency in the gas diffusion layer. Finally, it outlines the future challenges and perspectives of mesoporous carbon materials for PEMFCs.

2 Synthesis of mesoporous carbon

2.1 Challenges in synthesizing mesoporous carbon

Despite the successful synthesis of various mesoporous carbon materials with specialized pore size structures, precise control over pore size remains a significant challenge [27,28]. One common method, the hard-template approach, often suffers from complications during the template removal process. For example, hard templates such as SiO_2 typically require strong acids or bases for template removal, which can damage the carbon skeleton, leading to material collapse and pore structure loss. Moreover, this procedure is hazardous, environmentally polluting, and results in low yields and high costs. A typical case is the synthesis of ordered mesoporous carbon (CMK-3) using SBA-15 as a template, which requires multiple rounds of template etching [29].

Similarly, the soft-template method, which relies on the assembly of amphiphilic block copolymers, faces issues related to high template costs and poor thermal stability, often leading to disordered or contracted pore structures. For instance, when F127 is used as a template to prepare mesoporous carbon, the high carbonization temperatures (above 400 °C) cause template decomposition, resulting in a broader pore size distribution and less uniform pore structures [30].

In anticipation, research on hard-template methods may focus on developing easily soluble templates or designing reusable templates to reduce costs and improve efficiency. In contrast, soft-template approach may benefit from exploring biomass-derived surfactants as alternatives to block copolymers, or from developing new technologies that could lower the carbonization temperature, preventing template decomposition. By combining the advantages of existing methods, and continually improving synthesis techniques, more efficient processes could be developed, facilitating the widespread application of mesoporous carbon materials. These areas merit further investigation.

2.2 Synthesis methods

The development of mesoporous carbon materials dates back to the 1990s, when researchers began exploring carbon materials with ordered pore structures. In 1999, Ryoo et al. [31] made a significant breakthrough by preparing mesoporous carbon materials with high specific surface areas and tunable pore sizes, using silica as a hard template and sucrose as a carbon source. Thereafter, Liang et al. [32,33] further advanced the synthesis of mesoporous carbon by using self-assembled copolymer molecular arrays and carbon precursors. Additionally, researchers developed the soft template method, where surfactants or block copolymers were used as templates to form mesoporous structures through self-assembly, followed by the removal of templates via high-temperature calcination to obtain mesoporous carbon materials [34,35].

Other synthesis techniques for mesoporous carbon include physical and chemical activation, polymer precursor carbonization, the sol-gel method, microwave synthesis, and phase transition methods [36–40]. These approaches can be combined to produce mesoporous carbon materials with varying pore sizes and distributions, tailored for specific practical applications. The following sections will explore these synthesis strategies for mesoporous carbon materials in the context of their applications in PEMFCs, focusing on hard template methods, soft template methods and template-free methods.

2.2.1 Hard template methods

In 1992, Kresge et al. [41] successfully synthesized the M41S series of ordered mesoporous molecular sieves for Mobil Corporation by using alkyl quaternary ammonium cationic surfactants as templates. These surfactants were co-assembled with inorganic silica sources under alkaline conditions, and the templates were then removed, introducing the concept of templating in the preparation of mesoporous materials for the first time. Since then, the hard template method has become widely adopted for the synthesis of mesoporous carbon materials [23].

The typical process for the hard-template method involves synthesizing a hard template with a specific structure, filling it with a carbon precursor, carbonizing the precursor at high temperatures, and then removing the template through physical or chemical methods. This results in mesoporous carbon materials with well-defined pore structures. This method is akin to casting in the metal processing industry, though nanoscale casting presents additional challenges compared to macroscale processes. The main advantage of the hard template method is that it allows for precise control over pore structure and size. However, a significant disadvantage is that the template removal process can be complex and may damage the pristine structure of the material.

Primary hard template materials include silica [42,43], metal oxides [44,45], zeolite molecular sieves [46,47], and thermoplastic elastomers [48,49]. Among these, mesoporous silica materials such as SBA-15 and MCM-48 are the most widely used hard templates. As shown in Fig. 1(a), Kwak et al. [50] prepared iron-sulfur co-doped mesoporous carbon nanostructures using ordered mesoporous silica SBA-15 as a hard template, with porphyrinic iron and sulfur as precursors. The hard template method allows the mesoporous carbon materials to exhibit highly ordered mesoporous structures, and the pore size can be precisely controlled by adjusting the synthesis temperature and precursors molar ratios. The result is mesoporous carbon materials with large specific surface area, high pore volumes, and high doping levels (Fig. 1(b)).

Zhang et al. [51] introduced a top-down template strategy, in which commercially available metal oxides

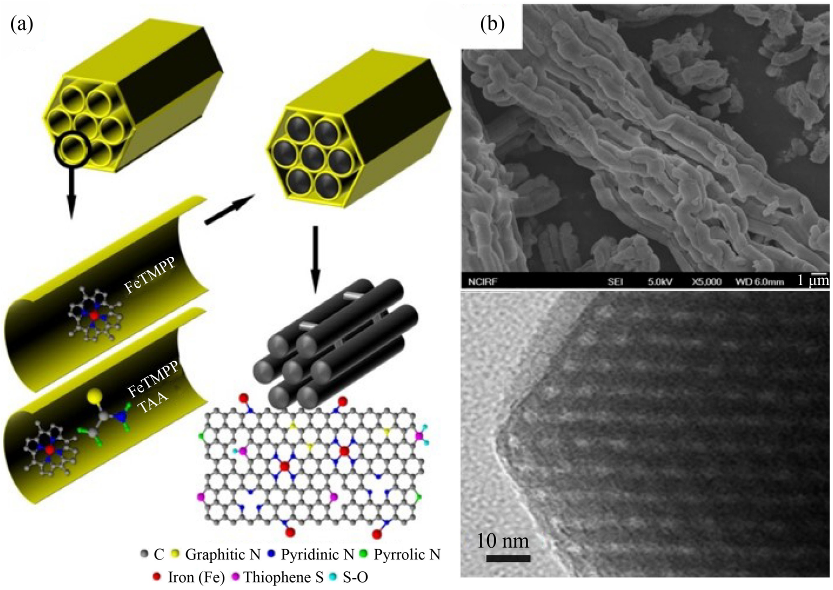


Fig. 1 Preparation of ordered mesoporous carbon materials using SBA-15 as a hard template (adapted with permission from Kwak et al. [50], copyright 2017, Elsevier).

(a) Synthetic schematic; (b) SEM and HR-TEM images of meso-SiO_2.

are mixed with organic precursors and subjected to high-temperature carbonization. As shown in Fig. 2(a), during high-temperature carbonization, the metal oxides decompose into nanoparticles, acting as *in situ* templates to create nanoscale pores in the carbon matrix. By adjusting the composition of metal oxides and organic precursors, the pore size and the incorporation can be finely controlled. This approach provides a universal method for producing atomically doped mesoporous carbon materials. Figure 2(b) shows the specific surface areas (SSAs) and pore volumes of mesoporous carbon materials prepared using other commercially available metal oxide powders and organic precursors.

In addition to conventional three-dimensional mesoporous carbon materials, the hard template method can also be used to create two-dimensional mesoporous carbon materials. Leyva-García et al. [52] skillfully utilized ordered mesoporous silica films as templates to synthesize continuous mesoporous carbon thin films on graphite current collectors. As shown in Fig. 2(c), these films exhibit a lamellar thin-film structure with pore sizes around 2–3 nm, along with certain structural defects. The unique two-dimensional mesoporous structure provides the material with exceptionally high specific capacitance and rapid charge-discharge capabilities, making it highly promising for various applications.

Overall, the preparation of mesoporous carbon materials using the hard template method is evolving toward greater template diversification. Initially, templates like SBA-15 and MCM-48 were commonly used, but now novel templates such as nanosheets, nanowires, and silicon carbide are being explored [53–55]. This shift also includes moving from hard templates with specific structures to those with diverse

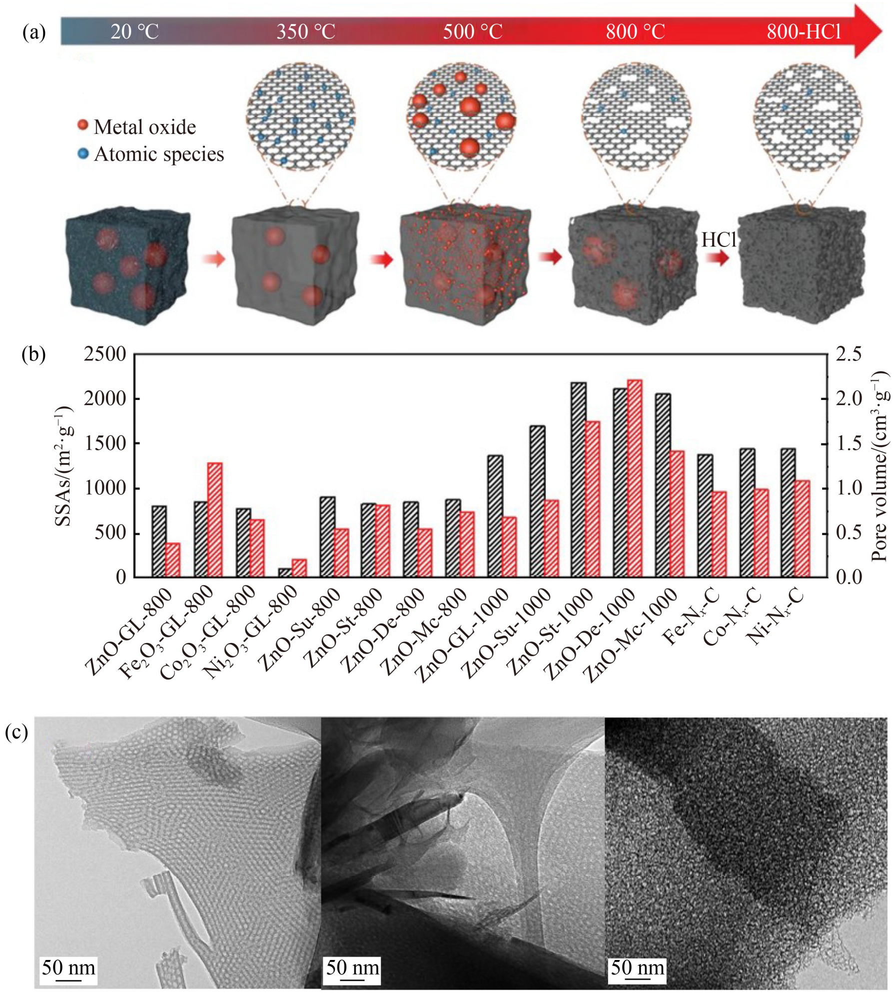


Fig. 2 Mesoporous carbon materials prepared by using the hard-templating method.

(a) Schematic illustration of pore generation mechanism based on carbonation of ZnO/glucose mixtures (adapted with permission from Zhang et al. [51], copyright 2022, Wiley-VCH Verlag); (b) SSAs and pore volumes of mesoporous carbon materials obtained by top-down templating strategy using other metal oxides and organic precursor preparation (adapted with permission from Zhang et al. [51], copyright 2022, Wiley-VCH Verlag); (c) TEM of mesoporous silica thin film, composite silica/carbon thin film and mesoporous carbon thin film (adapted from Leyva-García et al. [52] under the terms of CC BY-NC license).

morphologies, and from simple templates to multifunctional, multi-component ones. By integrating these templates with other functional materials, the electrochemical performance, mechanical strength, and electrical conductivity of mesoporous carbon materials have been further enhanced [56,57]. Additionally, efforts are underway to investigate renewable and recyclable hard templates, aiming to reduce the costs associated with mesoporous carbon production through the hard template method [58–60]. In the future, the focus will be on improving the controllability of template synthesis and ensuring the recyclability of these templates, addressing emerging needs, and driving further advancements in this field.

2.2.2 Soft template methods

Soft templating is another widely used method for preparing mesoporous carbon materials. This method usually utilizes the co-assembly behavior of amphiphilic molecules, such as surfactants or specific block copolymers, to form mesoporous structures [61]. Compared to the hard template method, soft templating has advantages of lower cost and the ability to precisely control the physicochemical properties of the resulting mesoporous carbon by adjusting the physicochemical composition of the amphiphilic molecules [62,63].

Liang et al. [64] summarized the key factors that contribute to the successful preparation of mesoporous carbon using the soft templating method. First, the system must contain at least one carbon-forming component and one pore-forming component, which must interact and self-assemble into nanostructures. The carbon-forming component should have a high carbon yield at carbonization temperatures, and the nanostructures formed during self-assembly must remain stable during the carbonization process. In contrast, the pore-forming component should decompose easily at carbonization temperatures and contribute minimally to the carbon content.

The main advantages of the soft template method are its simplicity and ease of template removal. However, a significant disadvantage is the relatively greater difficulty in accurately controlling the pore structure. Recent research on soft templating has increasingly focused on fine-tuning the interactions between the carbon precursor and the pore-forming template. Several strategies are being developed to achieve this goal [65–67].

Liu et al. [15] proposed a solvent-mediated polymerization-induced self-assembly (PISA) strategy to precisely control the hydrogen bonding and interfacial interactions between pore-forming micelles and the carbon precursor, resulting in nitrogen-doped mesoporous carbon spheres with adjustable pore sizes. SEM images of the intermediate morphologies during the growth of the carbon spheres are shown in Fig. 3(a). Initially, only mesopores existed in the carbon spheres, but after 24 h, the spheres developed a layered mesoporous structure, with macropores in the center having a pore size of 1.2 μm, and smaller pores on the walls of the macropores measuring about 95 nm.

Beyond the more conventional spherical mesoporous carbon, the soft templating method can also be applied to fabricate one-dimensional linear mesoporous structures (Fig. 3(b)). Peng et al. [68] proposed a kinetic-controlled micelle-directed self-assembly strategy at high temperatures, where hexamethylenetetramine decomposes *in situ* to release ammonia and formaldehyde. Ammonia acts as a pH buffer, while formaldehyde regulates the hydrogen bonding interactions and self-assembly kinetics between the carbon precursors and pore-forming agents. By adjusting the stoichiometric ratios of the reactants, this approach yields one-dimensional carbon nanofibers with uniform diameters, high aspect ratios, and an ordered mesoporous structure (Fig. 3(c)), providing a novel pathway for synthesizing advanced porous materials.

Additionally, the soft templating method can be combined with the hard templating method to create more complex structures. As shown in Fig. 3(d), Zhao et al. [69] proposed a self-assembly strategy utilizing single micelle interfacial confinement to prepare a composite mesoporous N-doped carbon superstructure. In this approach, a silica macrosphere serves as a hard template, creating central macropores with a diameter of approximately 300 nm. Meanwhile, block copolymer micelles in the surface layer function as a soft template, generating surface mesopores with a pore size of about 22 nm. These surface mesopores are interconnected with the central macropores through smaller internal pores, resulting in a unique hierarchical pore structure.

Currently, the interactions between the pore-forming component and the carbon-forming component in the soft templating method are mainly weak interactions, such as hydrogen bonding, electrostatic interactions, and hydrophilic-hydrophobic interactions. These interactions ensure that the templating agent binds tightly to the carbon source and facilitates the formation of an ordered mesoporous structure during the subsequent carbonization process [70–73]. In the future, the development of additional dimensional interactions, such as spatial site resistance effects and solvent effects, could enable more precise regulation of the co-assembly behavior, allowing for the creation of desired mesoporous structure at will.

2.2.3 Template-free method

The template-free method is a technique for preparing mesoporous materials without the use of templating

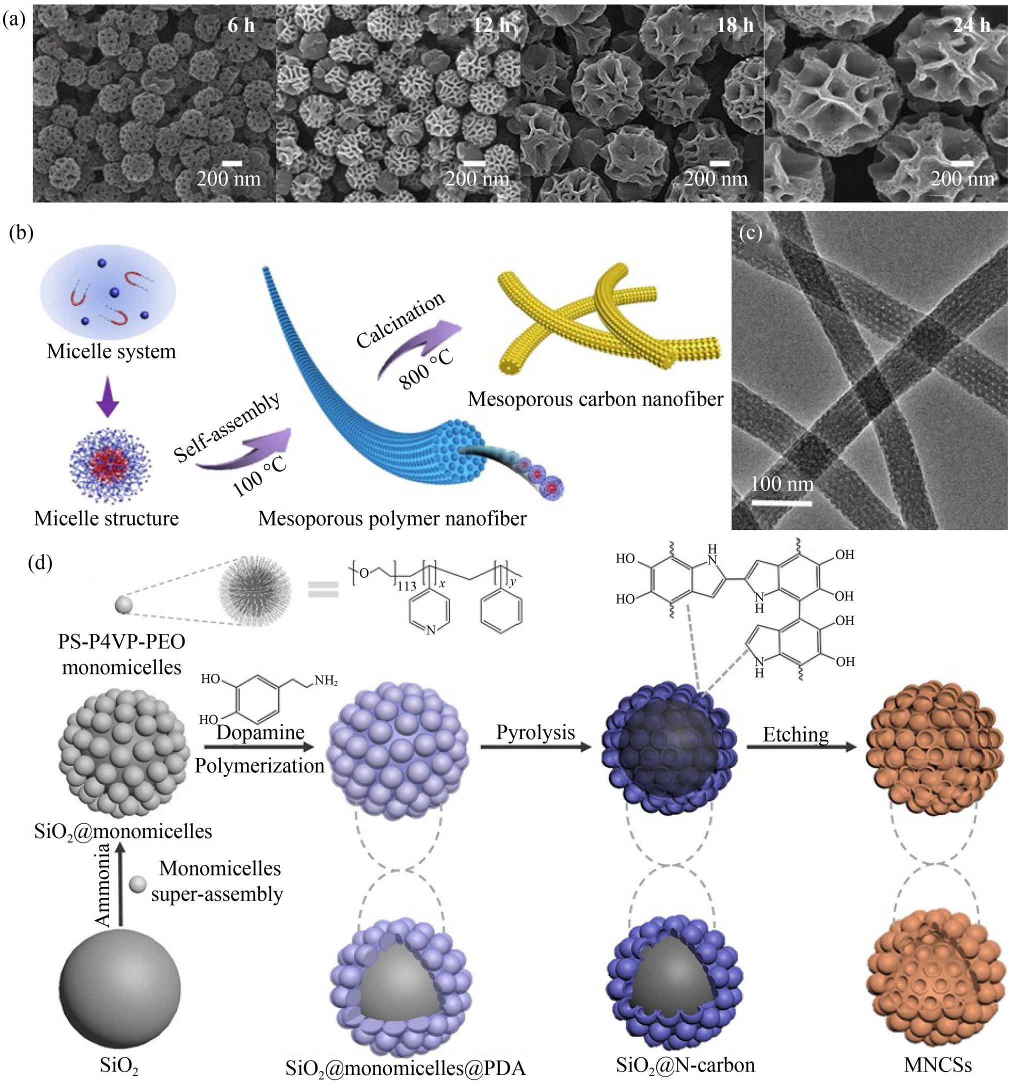


Fig. 3 Preparation of mesoporous carbon materials by using the soft-templating method.

(a) SEM images of intermediate morphologies during growth of carbon spheres (adapted with permission from Liu et al. [15], copyright 2023, American Chemical Society); (b) schematic illustration; (c) TEM image of one-dimensional ordered mesoporous carbon nanofibers (adapted from Peng et al. [68] under the terms of CC BY license); (d) schematic illustration of the process for synthesis of three-dimensional hollow layered nitrogen-doped carbon superstructures with spherical mesopores via a monomicelle interface confined assembly (adapted with permission from Zhao et al. [69], copyright 2022, American Chemical Society).

agents. This approach typically involves the direct formation of mesoporous structures through chemical processes. One of the main advantages of the template-free method is that it eliminates the need for introducing and later removing templating agents, which simplifies the preparation process and can reduce costs [64,74]. However, the template-free method may be less effective than hard and soft templating methods in controlling pore size distribution and ensuring homogeneity. It requires more precise regulation of reaction conditions, including temperature, time, and reagent concentration, to achieve the desired mesoporous structure [75].

Zheng et al. [76] proposed a straightforward template-free method for synthesizing N, S co-doped hierarchical mesoporous carbon materials as ORR electrocatalysts. The distinct hollow spherical mesoporous structure was generated *in situ* from a meticulously designed covalent triazole framework through a thermally induced cavitation mechanism. In this process, the oligomers at the core of the sulfur-bonded covalent triazole framework decompose and volatilize under heat, diffusing to the outer shell. These oligomers are then etched by gases released during pyrolysis, creating numerous mesopores. Notably, the thickness of outer shell of the mesoporous carbon spheres can be precisely controlled by adjusting the pyrolysis temperature. The N, S co-doped hierarchical mesoporous carbon spheres feature high nitrogen and sulfur content, large specific surface areas, and a uniform distribution of abundant mesopores. These unique properties contribute to exceptional electrocatalytic performance and superior longevity for oxygen reduction reactions in PEMFCs.

Gong et al. [77] utilized carbohydrates as carbon sources and kayexalate as a structural directing agent to synthesize carbon spheres with three-dimensional mesoporosity and short-range microporosity. During the hydrothermal process, kayexalate suppresses excessive cross-linking of the carbon precursor, facilitating the formation of uniformly dispersed carbon spheres with sizes ranging from 60 to 80 nm. This method produces mesoporous carbon materials with an exceptionally high specific surface area of up to 1306 m^2/g and significant pore volume. Robertson et al. [48] proposed an innovative method for synthesizing ordered mesoporous materials through by pyrolyzing polystyrene-block-poly(ethylene-ran-butylene)-block-polystyrene (SEBS) (Fig. 4(a)). As shown in Fig. 4(b), the PEB segments in the thermoplastic elastomer undergo crosslinking at high temperatures in concentrated sulfuric acid, forming a carbon framework during pyrolysis. The lower molecular weight PS segments decompose and exit the system due to their lower carbon yield, resulting in sulfur-doped mesoporous carbon materials. This method effectively integrates the two key components for mesoporous carbon synthesis, enabling precise control over carbonization and pore formation by adjusting the polymerization degree of each copolymer segment. This strategy also provides a promising direction for future developments in mesoporous carbon material synthesis.

In addition to the methods discussed above, recent years have witnessed the development of a variety of novel synthesis methods that break through the limitations of traditional template-based methods. These innovations are driven by the growing demand for diversified design and application of mesoporous carbon materials. For instance, the directional assembly of monomers with magnetic properties under the influence of an applied magnetic field enables precise control over the orientation and size of the mesoporous channels. This method addresses the inherent lack of flexibility in regulating complex structures using traditional template methods [78]. Moreover, the biomass-derived method utilizes the spontaneous assembly properties of natural biomolecules combined with a carbonization process to generate hierarchical mesoporous structures in a single step. This approach eliminates the need for template removal and significantly enhances the sustainability of the resulting materials [79]. These innovative methods not only expand the range of the preparation techniques of mesoporous carbon materials but also open up new research directions and potential applications for the future development of mesoporous carbon materials.

3 Applications of mesoporous carbon in PEMFCs

Fuel cells are crucial for the efficient energy conversion and utilization of hydrogen, with PEMFCs being the most representative type. PEMFCs have significant advantages, including independence from the Carnot cycle, high energy conversion efficiency, and the production of non-toxic and harmless byproducts, making them highly promising for a wide range of applications [80–82]. The MEA is the core component of PEMFCs, where electrochemical reactions take place. At the anode, hydrogen is oxidized, while at the cathode, oxygen is reduced. These processes are fundamental to energy conversion and multiphase material transport within the fuel cell, directly affecting its performance, lifespan, and

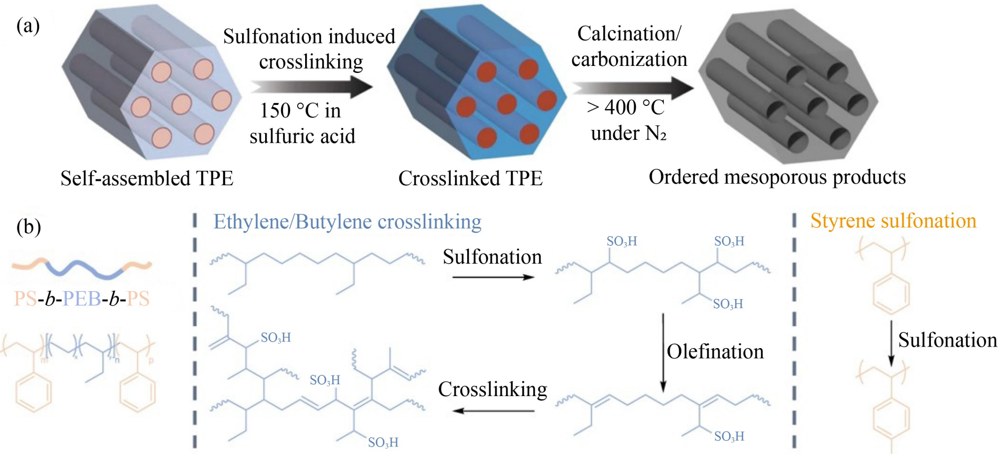


Fig. 4 Preparation of mesoporous carbon materials by using the template-free method.

(a) Schematic illustration of sulfonation-induced crosslinking reaction in thermoplastic elastomers, converting them into carbon precursors; (b) crosslinking reaction schemes for both PEB and PS blocks in the SEBS118 precursor (adapted from Robertson et al. [48] under the terms of CC BY license).

cost [83]. The MEA structure mainly consists of a proton exchange membrane, a catalyst layer, and a gas diffusion layer. A schematic illustration of the PEMFC and MEA structure is shown in Fig. 5.

Currently, PEMFC technology has reached a relatively mature stage, enabling it to serve as a power source for large commercial vehicles, thereby complementing pure electric vehicles, which are primarily used for small passenger cars [7]. However, challenges such as high costs and limited durability of fuel cell stacks have impeded the broader commercialization of PEMFCs [84]. As a key functional material, mesoporous carbon materials present significant potential in reducing costs, enhancing durability, and improving PEMFC performance [5]. With ongoing research and development, mesoporous carbon materials are expected to play a crucial role in the successful commercialization of PEMFCs.

3.1 Mesoporous carbon as catalysts support

3.1.1 Optimizing the distribution of ionomer and mitigating the poisoning of Pt

Efficient proton transport is crucial for the continuous operation of PEMFCs [85,86]. For instance, in the ORR at the fuel cell cathode, oxygen molecules react with protons and electrons to form water. The efficient migration of protons, as reactants, directly influences the reaction kinetics. Any hindrance to proton conduction can dramatically reduce the reaction rate, ultimately decreasing the output performance of PEMFCs [87]. To enhance proton mobility, ionomers with ionic conductivity are typically incorporated into the catalytic layer. These ionomers often contain polar groups, such as sulfonic acid (–SO_3H), in their molecular structure, which bind water molecules and create a continuous proton transfer channel to facilitate the ORR [88].

However, while ionomers enhance proton conduction, their irregular and uneven distribution significantly hinders the transport of O_2, which is another reactant in the ORR. As shown in Fig. 6(a), the long diffusion distance required for O_2 to travel from the gas flow channel to the catalyst surface further impedes its effective diffusion and contacts with the catalyst [89]. Moreover, in areas where the catalyst particles are directly covered by ionomers, the sulfonate groups within the ionomers can poison the active sites of the catalyst, leading to a substantial decrease in catalytic activity. This ultimately limits the reaction rate of ORR and diminishes the overall performance of PEMFCs [90]. Therefore, optimizing ionomer distribution to balance proton conductivity and oxygen diffusion, while minimizing catalyst poisoning, is a key challenge in fuel cell research.

Mesoporous carbon materials can address the issue by influencing the distribution of ionomers and their spatial arrangement with catalyst particles. As shown in Fig. 6(b), the second generation of Toyota Mirai fuel cell vehicles employs dendritic mesoporous carbon carriers, which

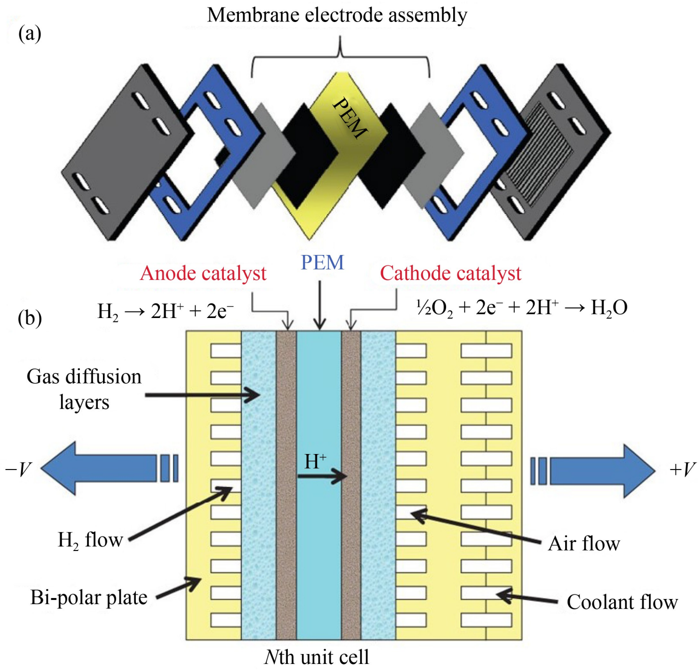


Fig. 5 Internal structure diagram of a fuel cell unit within a fuel-cell stack.

(a) Schematic diagram of the structural components of PEMFC power pile; (b) schematic diagram of reaction principle in MEA (adapted with permission from Debe [83], copyright 2012, Springer Nature).

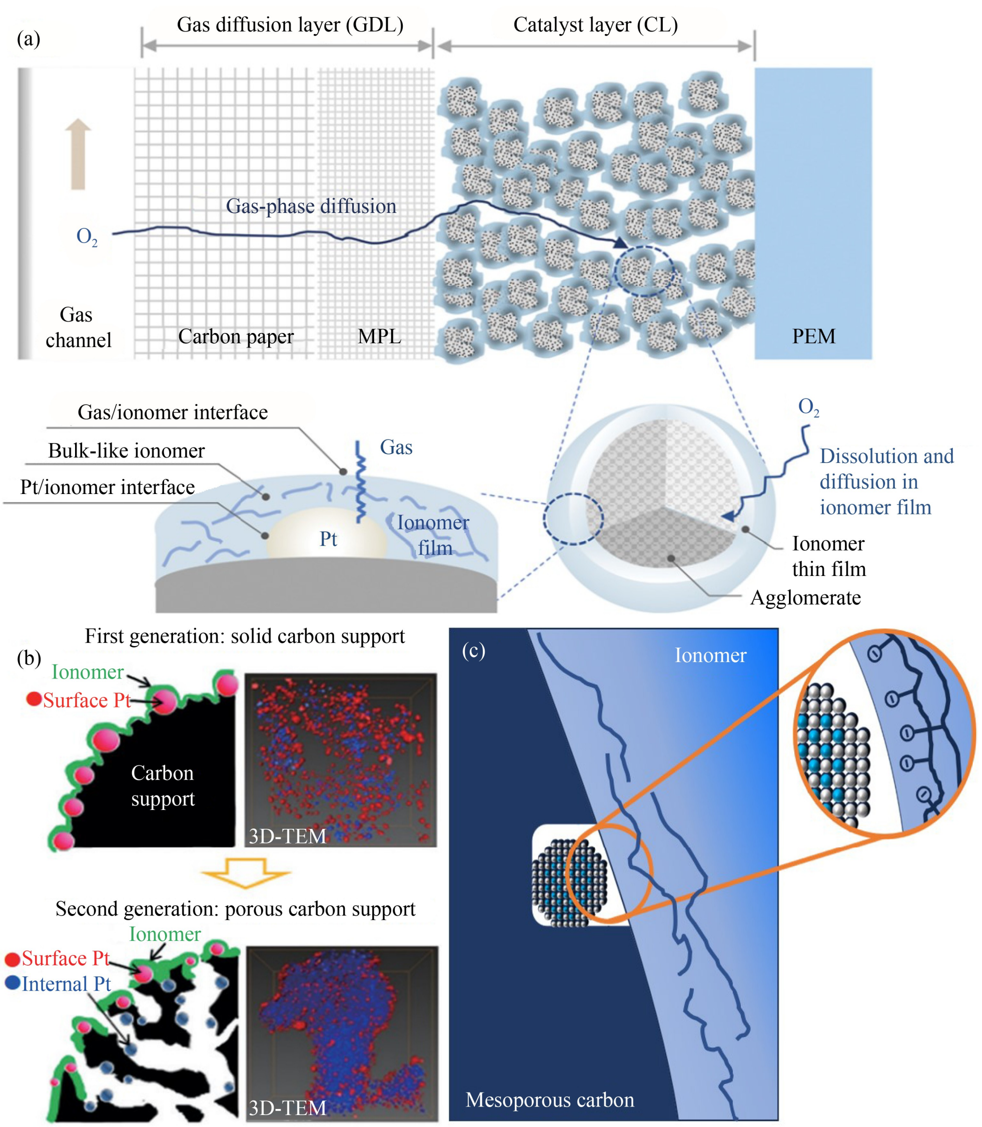


Fig. 6 Mesoporous carbon structure design for optimizing ionomer distribution and spatial arrangement with catalyst nanoparticles. (a) O_2 transport paths in PEMFCs (adapted with permission from Tang et al. [89], copyright 2022, Royal Society of Chemistry); (b) dendritic mesoporous carbon carriers used in Toyota's second-generation Mirai fuel cell vehicle (adapted with permission from Mizuno et al. [91], copyright 2021, Toyota Motor Corporation); (c) suitable mesoporous structure allowing ideal spatial arrangement of catalysts and ionomers in "proximity but not in contact."

provide optimal pore diameters for catalyst loading. This mesoporous structure avoids direct contact between the catalyst and the ionomer (Fig. 6(c)), effectively reducing the poisoning effect of the ionomer on the catalyst and enhances the mass activity (MA) of the ORR without compromising proton conduction, ultimately improving the overall performance of the fuel cell [91].

An effective indicator for estimating the proportion of particles located on the outer surface of carbon is the measurement of Pt utilization in a MEA during a CO-stripping test conducted under low humidity conditions (10%–20% relative humidity) [92,93]. Padgett et al. [94] loaded Pt nanoparticles onto two commercially available carbon carriers, XC-72 and EC-300J, to obtain catalysts with similar average Pt particle sizes and electrochemical active surface area (ECSA). The dry proton accessibility of the catalyst using XC-72 as the carrier was found to be as high as 95%, significantly higher than that of the Pt/EC-300J catalyst. This suggests that the vast majority of Pt nanoparticles in the Pt/XC-72 catalyst are located on the surface of the carbon carrier, while most of the Pt nanoparticles in the Pt/EC-300J are located in the pores of the carrier, reducing the direct contact between the Pt and the ionomer. However, the tortuous pore structure of commercial porous carbon EC-300J hinders adequate contact between oxygen and the catalysts within the pores, increasing oxygen transport resistance and potentially leading to a degradation in fuel cell performance, particularly at high current densities [95,96].

To address this issue, General Motors (USA) developed two types of accessible mesoporous carbons, HSC-e and HSC-f, by applying specific treatments to commercial Ketjen Black EC-300J carbon supports [97]. The structural schematics of accessible mesoporous carbons, commercial solid carbons, and porous carbons are illustrated in Fig. 7(a). Compared to catalysts supported on commercial solid carbon (XC-72) and porous carbon (EC-300J), those supported on HSC-e and HSC-f demonstrate superior performance in both the high-voltage and high-current regions (Fig. 7(b)). This improvement can be attributed to the unique short mesopore structure of the accessible mesoporous carbons, which reduces O_2/H^+ transport resistance while mitigating the toxic effects of ionomers on the catalysts.

The surface chemistry of carbon carriers significantly affects the distribution of ionomers. As shown in Fig. 7(c), Ott et al. [98] demonstrated that nitrogen (N) doping on commercial EC-300J carbon carriers resulted in a catalytic layer with highly interconnected pores and a uniform ionomer distribution. This improvement was attributed to the electrostatic interactions between the sulfonic acid groups in the ionomers and the nitrogen sites on the carbon carrier. The Pt/N-C catalyst not only mitigates the poisoning effect of ionomers but also effectively reduces local oxygen transport resistance (R_{local}). Compared to undoped carbon carriers, N-doped porous carbon carriers reduce the voltage loss by up to 45 mV at a high current density of 2 A/cm^2 and increase power density by 31%. Similarly, Orfanidi et al. [99] introduced –NH$_x$ groups onto carbon carriers, which led to a uniform ionomer distribution and significantly enhanced fuel cell performance (Fig. 7(d)).

To precisely control the interactions between carbon carriers and ionomers, the initial step involves developing advanced methods for preparing mesoporous carbons. This allows for better control over the mesoporous structure, which in turn influences the adsorption and distribution of the ionomers. Concurrently, interfacial engineering techniques can be utilized to optimize

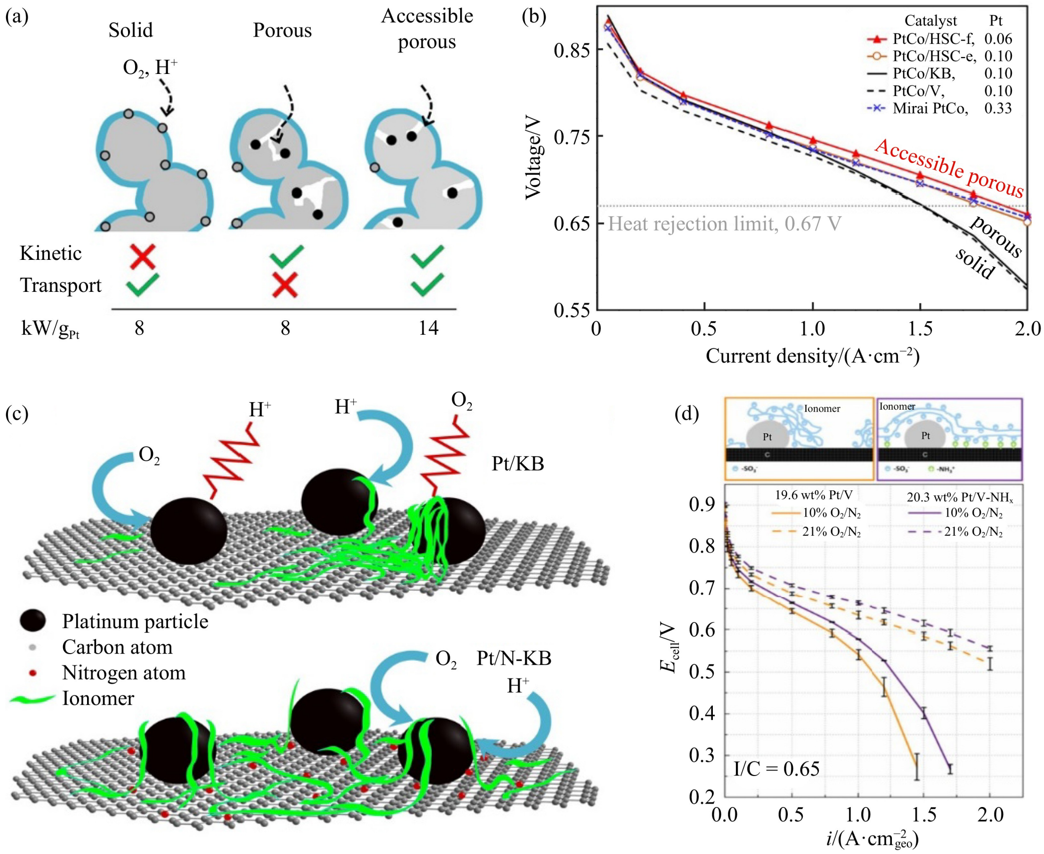


Fig. 7 Mesoporous carbon structure design for reducing O_2 transport resistance and mitigating ionomer toxicity on catalysts.

(a) ORR kinetics and substance transport properties of catalytic layer structures obtained from three different carbon carrier systems; (b) comparison of fuel cell performance with different platinum-cobalt-on-carbon catalysts ((a) and (b) adapted with permissions from Yarlagadda et al. [97], copyright 2018, American Chemical Society); (c) electrostatic interactions due to nitrogen doping on carbon carrier surface resulting in a more homogeneous ionomer distribution (adapted with permission from Ott et al. [98], copyright 2020, Springer Nature); (d) surface doping with –NH$_x$ groups leading to better fuel cell performance (adapted from Orfanidi et al. [99] under the terms of CC BY license).

interactions between the mesoporous carbon carriers and ionomers by adjusting the surface chemistry through surface modification, functionalization, and heteroatom doping. Additionally, introducing strong interactions such as π-π stacking and ionic coordination can facilitate selective interactions between the ionomers and carbon carriers without compromising the activity of the catalyst particles [100]. This approach enhances both the adsorption stability and proton conduction efficiency of the ionomers on the carbon carriers while preserving catalyst functionality.

3.1.2 Lowering the O_2 transport resistance

In PEMFC, Pt is the most widely used catalyst [101]. However, Pt reserves on Earth account for only 5% of those of gold, leading to extremely high catalyst costs due to its limited availability. According to estimates by the US Department of Energy, when the annual shipment volume of PEMFC stacks reaches 500000 units, the catalyst cost will account for over 40% of the total stack cost [102], presenting a significant barrier to the broader commercialization of PEMFCs. To address this issue, various approaches have been developed to reduce the loading of precious metals in catalysts, including alloy catalysts, single-atom catalysts, and non-precious metal catalysts [103–105]. However, simply enhancing the intrinsic activity of the catalyst cannot effectively reduce the Pt content without compromising the performance of PEMFCs.

As shown in Fig. 8(a), reducing the Pt loading by decreasing the Pt/C ratio, while keeping the thickness of the ionomer film constant, increases the oxygen contact for each Pt nanoparticle, which consequently increases the R_{local} [106]. Greszler et al. [108] demonstrated that when the Pt content in the MEA of a fuel cell is reduced to a certain threshold, significant voltage loss occurs, especially in the high current density region. Sgarbi et al. [109] further emphasized that, in the low current density region of the polarization curve, decreasing Pt content diminishes fuel cell performance. The voltage loss becomes even more pronounced in the high current density region, which is characterized by mass transport limitations due to increased resistance of oxygen and proton transport. These voltage losses are attributed to the localized increase in oxygen flux due to the reduced Pt content in the catalytic layer, which subsequently increases oxygen transport resistance and reduces overall fuel cell performance [110]. Figure 8(b) illustrates the dominant factor of overpotential in low Pt-loaded PEMFC across different current regions [89]. It is evident that in the high current density region, the R_{local} is a significant factor contributing to the polarization loss.

Mesoporous carbon materials, known for their extremely high specific surface area and unique pore structures, can significantly enhance catalyst dispersion

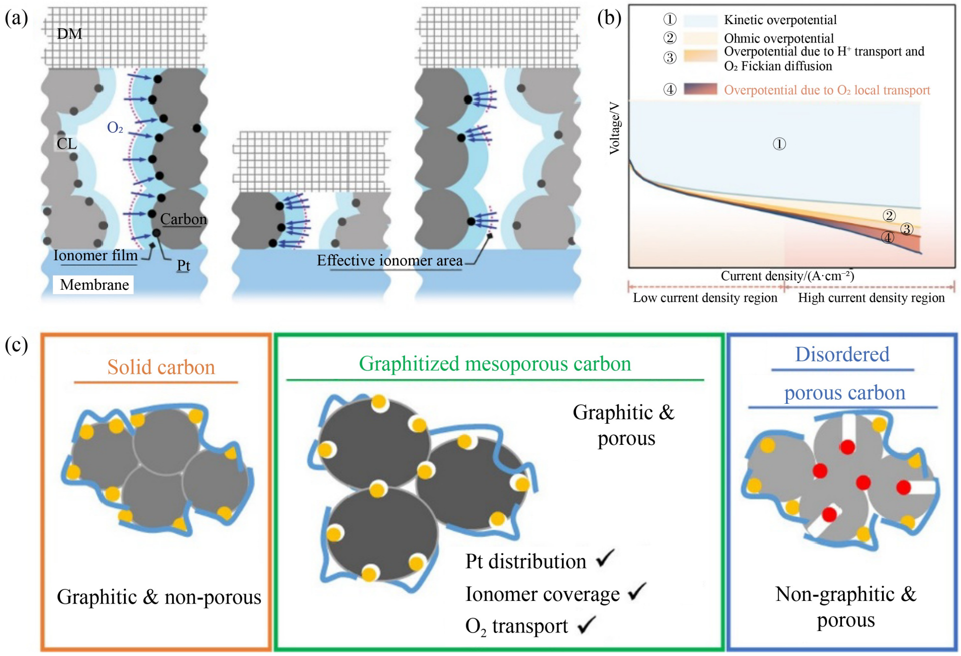


Fig. 8 Mesoporous carbon structure design for reducing oxygen transport resistance in the catalytic layer.

(a) Lower Pt loading results in greater localized oxygen flux and R_{local} (adapted with permission from Nonoyama et al. [106], copyright 2011, Electrochemical Society, Inc.); (b) overpotential dominant factors in PEMFC for different current regions (adapted with permission from Tang et al. [89], copyright 2022, Royal Society of Chemistry); (c) schematic structure of catalysts with Vulcan, ECS4005 and HSC as carriers (adapted with permission from Ramaswamy et al. [107], copyright 2023, American Chemical Society).

and promote oxygen transport within the catalyst layer when used as catalyst supports. This effectively improves the performance of low noble metal-loaded catalysts, mitigating performance degradation under high current density conditions [111,112].

Yarlagadda et al. [113] demonstrated that using ordered mesoporous carbon (CMK-3) as a support for Pt catalysts can significantly reduce R_{local}. The pressure-independent O_2 transport resistance is measured through O_2-limiting current density experiments. By normalizing this resistance with the measured roughness factor, the R_{local} can be determined, corresponding to the O_2 diffusion resistance near or on the Pt surface within the catalyst layer. Specifically, the R_{local} of Pt/CMK-3 is approximately 3 s/cm, markedly lower than the 15.9 s/cm of high surface area carbon (HSC) and the 8.3 s/cm of commercial Vulcan. The exceptionally low R_{local} is attributed to the ordered mesoporous structure, which provides more efficient oxygen transport pathways compared to disordered pores, allowing oxygen to more easily reach the surface of active sites. Additionally, CMK-3 features a smaller micropore volume compared to Vulcan and HSC. Since micropores (pores smaller than 2 nm) can impede the transport of oxygen and protons, reducing the micropore volume helps further decreases oxygen transport resistance [114]. Similarly, Ramaswamy et al. [107] investigated the effect of mesoporous structures on mass transport resistance using a novel graphitized mesoporous carbon support, ECS4005, developed by Pajarito Powder (USA). The mesopores of ECS4005 are primarily distributed in two size ranges: 10 nm and 40–80 nm. The 10 nm pores facilitate the dispersion of Pt nanoparticles, while the larger 40–80 nm pores enhance oxygen transport. This unique hierarchical pore structure greatly enhances reactant accessibility, reduces ionomer-induced catalyst poisoning, and prevents the deep embedding of Pt nanoparticles in disordered mesopores, thereby improving their utilization (Fig. 8(c)). Experimental results show that the R_{local} of the Pt/ECS4005 catalyst is 22 S/cm, significantly lower than the 31 S/cm observed for both Pt/HSC and Pt/Vulcan.

3.1.3 Enhancing stability and activity of catalysts

Maintaining catalyst stability is a significant challenge for PEMFCs. For instance, the commonly used commercial Pt/C catalyst undergoes notable degradation under actual PEMFC operating conditions, particularly at high potentials (> 0.9 V). At these elevated potentials, Pt can be partially oxidized ($\mathrm{Pt} \rightarrow \mathrm{Pt}^{2+}$), leading to its dissolution and migration from the electrode surface. According to the Ostwald ripening theory, dissolved Pt^{2+} ions diffuse through the solution and preferentially deposit onto the surfaces of larger platinum particles, as these particles are thermodynamically more stable and have lower surface energy. This process accelerates the loss of catalytic active sites while promoting particle growth, which in turn reduces the specific surface area and catalytic activity. Consequently, this process negatively impacts the performance and durability of PEMFCs [115]. Additionally, the carbon carriers used in the catalysts are susceptible to oxidation in high-voltage, oxygen-rich environments, where carbon can be oxidized to CO_2. This corrosion of the carbon carrier contributes to the detachment of catalysts, further compromising both the ECSA and stability of the overall system [116]. A schematic illustrating the mechanism of ECSA decay in Pt nanoparticles is presented in Fig. 9(a).

The mesoporous structure of carbon carriers has been demonstrated to inhibit the dissolution, migration, and recrystallization of catalyst nanoparticles through domain-limiting effects. Cheng et al. [117] prepared sub-sized Pt alloy catalysts with an average particle size of 2.3 nm by impregnation using commercial porous carbon Ketjenblack-600 as a carrier, with the surface of the catalysts covered by a layer of Pt-skin. After undergoing an accelerated durability test for 30000 cycles, the MA decay of sub-$\mathrm{Pt_3Co}$-MC was only 18.5%, significantly lower than the 67% observed for commercial Pt/C catalysts. Additionally, in fuel cell tests, sub-$\mathrm{Pt_3Co}$-MC exhibited an MA retention of 81.5%, far surpassing the 33% retention of commercial Pt/C (Fig. 9(b)). The remarkable stability of sub-$\mathrm{Pt_3Co}$-MC is attributed to the domain-limiting effect of its mesoporous structure, which effectively prevents catalyst agglomeration under actual operating conditions (Fig. 9(c)).

Zhou et al. [118] integrated titanium carbon-nitride (TiCN) nanoparticles into one-dimensional mesoporous carbon nanofibers, facilitating direct contact and strong interactions between the Pt nanoparticles and TiCN (Fig. 9(d)). This interaction enabled electron transfer from TiCN to Pt, effectively delaying the pre-oxidation of Pt. As a result, the initial step of Pt nanoparticle degradation was suppressed, enhancing the catalyst's overall stability. As illustrated in Fig. 9(e), the Pt@m-TiCN nanofibers exhibited remarkable durability, losing only 0.04 $\mathrm{W/cm^2}$ after 30000 accelerated durability test cycles in fuel cell evaluations, outperforming commercial Pt/C and other control samples. Furthermore, mesoporous carbon materials exhibit superior oxidation resistance, as their pores could effectively mitigate fluctuations in local potentials, thereby preventing oxidation reactions of the carbon carriers at elevated potentials. Tian et al. [119] successfully developed a series of hollow mesoporous carbon spheres. Finite element simulation analyses demonstrate that the unique hollow structure creates a closed internal microenvironment, which helps to stabilize local potentials and prevent oxidation reactions of the carbon carriers at high potentials.

Modifying the surface of mesoporous carbon materials

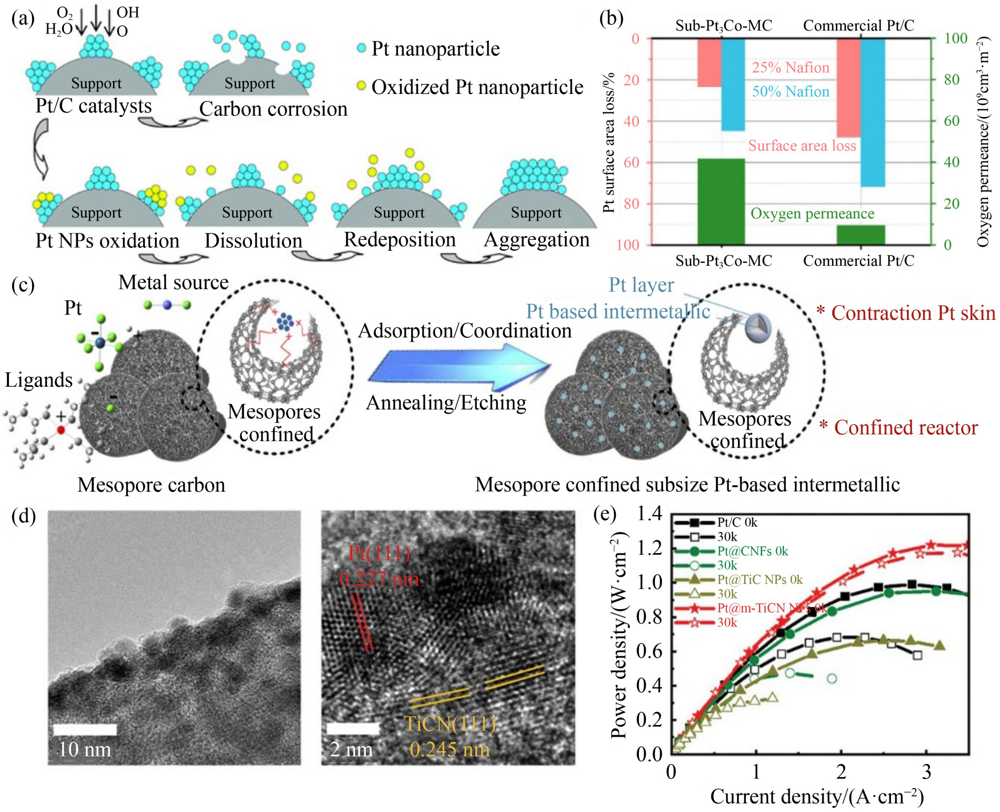


Fig. 9 Mesoporous carbon application for enhanced catalyst stability and activity.

(a) Schematic diagram of the mechanism for the loss of Pt ECSA (adapted with permission from Li et al. [115], copyright 2012, Royal Society of Chemistry); (b) sub-Pt_3Co-MC has superior performance and stability over commercial Pt/C in fuel cell testing; (c) illustration of synthesis methods for sub-Pt intermetallic-MC ((b) and (c) adapted with permissions from Cheng et al. [117], copyright 2021, National Academy of Sciences); (d) HRTEM image of Pt@m-TiCN NFs; (e) H_2–O_2 fuel cell power density plots comparisons before and after 30000 cycles (0.6–0.95 V) Pt@m-TiCN NFs and other control samples ((d) and (e) adapted with permissions from Zhou et al. [118], copyright 2024, Wiley).

enables further adjustment of the catalyst's electronic structure, thereby enhancing the ORR in fuel cells [120]. Zhu et al. [121] reported a nitrogen-doped mesoporous carbon material derived from metal-organic frameworks and nano-modified FeCo alloy. This catalyst features highly exposed bimetallic active sites and a core-shell structure. In alkaline half-cell tests, the optimal sample, $Fe_{0.25}Co_{0.75}$/NC-800, exhibited a kinetic current density of 156.25 mA/cm^2 at 0.8 V, far surpassing the 19.45 mA/cm^2 achieved by commercial Pt/C, demonstrating faster reaction kinetics and effective reactant penetration. This outstanding performance is primarily attributed to the formation of the FeCo alloy, which adjusts the electronic structure of Co active sites, optimizes the binding strength of oxygen reduction intermediates (OOH* and OH*), and significantly enhances catalytic activity. Table 1 summarizes the activity and stability of mesoporous carbon and traditional carbon materials as catalyst supports in PEMFCs, providing a clearer and more intuitive representation of the distinct advantages that mesoporous carbon possesses over traditional carbon materials.

3.2 Application of mesoporous carbon in gas diffusion layer

The gas diffusion layer (GDL) is a critical component of PEMFCs, situated between the catalytic layer and the bipolar plate. Its primary function is to facilitate the efficient transport of reactive gases, such as oxygen and hydrogen, to the catalyst layer while also aiding in the removal of generated water and heat from the fuel cell [124,125]. The GDL typically consists of a substrate layer and a microporous layer. The substrate, typically made of carbon fiber paper or cloth with specific hydrophobic properties, provides the necessary pore structure for the GDL, ensuring uniform gas distribution and transfer while facilitating effective water drainage [126]. The microporous layer, positioned between the catalytic layer and the substrate, commonly employs materials such as carbon black powder and hydrophobic polytetrafluoroethylene.

Traditional carbon black consists of irregular spherical particles and features a basic structure made of amorphous carbon with minor graphitized carbon

Table 1 Performance comparison of mesoporous carbon and conventional carbon materials as catalyst supports in PEMFCs

Catalyst	ECSA/ $(\mathrm{m^2 \cdot g^{-1}})$	MA at 0.9 V/$(\mathrm{A \cdot mg_{Pt}^{-1}})$		MA retention	Dry proton accessibility	Ref.
		BOL	EOL			
Pt/EC300J△	87	0.50	NA	NA	35%	Padgett et al. [94]
Pt/Acetylene Black°	99	0.30	NA	NA	49%	
Pt/Vulcan XC-72°	85	0.18	NA	NA	95%	
PtCo/EC300J△	67	0.60	NA	NA	55%	
PtCo/Acetylene Black°	74	0.53	NA	NA	49%	
PtCo/Vulcan XC-72°	50	0.27	NA	NA	97%	
Pt/HSC-d△	73.3	0.27	NA	NA	42%	Yarlagadda et al. [97]
Pt/HSC-e△	48.7	0.30	NA	NA	61%	
Pt/HSC-f△	79.2	0.36	NA	NA	51%	
Pt/Vulcan XC-72°	67.5	0.11	NA	NA	95%	
E-Pt/NC$_5$△	217	0.48	NA	NA	42%	Derendyaev et al. [110]
commercial Pt/C°	69	0.15	NA	NA	80%	
Pt@m-TiCN△	169.26	0.30	NA	NA	NA	Zhou et al. [118]
Pt@CNFs°	86.17	0.10	NA	NA	NA	
L1$_0$-PtCo/Co-N-PCNF△	41.3	1.82	1.80	98.9%	92%	Lai et al. [122]
L1$_0$-PtCo/Vulcan XC-72°	42.8	0.48	0.36	75%	78%	
Pt/GMC1300-1800△	106.1	0.212	0.141	66.5%	NA	Wu et al. [123]
Pt/C-JM°	68.5	0.101	0.041	40.6%	NA	

Notes: △—Mesoporous carbon support; °—Conventional carbon support; BOL—Beginning of Life; EOL—End of Life.

content. This structure microscopically appears as graphite-like layers with irregularly arranged carbon atoms [127]. While conventional carbon black materials offer some electrical conductivity and structural support, they have drawbacks such as uneven pore structure and insufficient water management properties [128]. In contrast, ordered mesoporous carbon materials, characterized by a well-defined pore structure and high porosity, can effectively address these limitations by enhancing gas and water transport channels.

Sahu et al. [129] synthesized ordered mesoporous carbon (MC) with a high specific surface area by using the soft template method, followed by treatment with 2 mol/L HNO_3. This treatment enhanced the hydrophilicity of the mesoporous carbon by introducing acidic groups on its surface. After treatment, the mesopore volume of the small pores (3.8 nm) nearly doubled, while the volume of larger pores remained largely unchanged, with their diameter decreasing from 7.6 to 6.4 nm. Due to the presence of mesopores with different pore sizes in the GDL, the water generated from the fuel cell reaction can be rapidly discharged through the small mesopores under the capillary effect, which reduces the water accumulation, while the reactive gases can be transported and permeated to the catalytic layer to participate in the reaction through the large mesopores. Consequently, PEMFCs using ordered mesoporous carbon as the GDL microporous layer show significantly improved performance compared to the commercial Vulcan XC-72R. At a load current density of 1.1 A/cm^2, the electrode with 2 mol/L HNO_3-treated MC achieved a peak power density of 0.53 W/cm^2, while the electrode using Vulcan XC-72R reached only 0.47 W/cm^2 at a lower load current density of 0.9 A/cm^2.

Selvarani et al. [130] modified commercial Vulcan XC-72R using sucrose as a pore-forming agent and investigated the influence of GDL porosity on fuel cell performance. Increased porosity enhances oxygen transport and reduces water flooding, but excessive pore-forming agents can increase resistance due to structural loosening. The study found that a pore-forming agent content of 50 wt% yielded the optimal porosity and gas diffusion coefficient for GDL, effectively balancing gas permeability and conductivity. In H_2–O_2 PEMFC systems, electrodes made with GDL containing the optimal pore-forming agent exhibited a peak current density of up to 2.08 A/cm^2, while the control sample without a pore-forming agent only reached 1.7 A/cm^2.

While mesoporous carbon offers advantages as GDL in fuel cells, its practical application faces several challenges. For example, high-porosity materials are prone to structural collapse under mechanical stress or electrochemical corrosion over prolonged operation [131], which can adversely affect the stability and durability of the GDL. Selvarani's study also highlights the necessity of striking a balance between porosity and conductivity, as excessively high porosity may diminish the mechanical support and electronic conductivity of the

GDL. Future research will focus on developing composite materials with gradient pore structures and the integrating mesoporous carbon with nanofibrous carbon to synergistically enhance gas transport efficiency and mechanical strength. Additionally, multiscale simulations and *in situ* characterization techniques will be used to explore the structure-property relationships between porosity, mass transfer, and performance. This will provide insights into how the microscopic pore structure of the GDLs influences macroscopic properties like mass transfer efficiency and overall performance under operating conditions, further advancing the development and widespread application of mesoporous carbon-based GDLs in PEMFCs.

4 Summary and outlook

This review summarizes the application of mesoporous carbon materials in PEMFCs, focusing on their synthesis methods, advantages as catalyst supports, and potential to enhance fuel cell performance. Mesoporous carbon materials are considered pivotal for improving PEMFC performance due to their high specific surface area, tunable pore structures, and excellent conductivity. Compared to traditional carbon materials, the high specific surface area of mesoporous carbons provides numerous active sites for catalysts, thereby enhancing the energy density and electrochemical reaction efficiency of PEMFCs. Their precisely tunable mesopore structures facilitate related kinetic reactions, charge transfer, and mass transport, significantly boosting the electrochemical performance of electrodes. Moreover, the chemical properties of mesoporous carbon materials can be modified through doping and other methods, further enhancing their performance.

Despite considerable advancements in mesoporous carbon materials for PEMFCs, challenges and opportunities remain. Therefore, this review outlines possible research directions, aiming to provide inspiration for further studies.

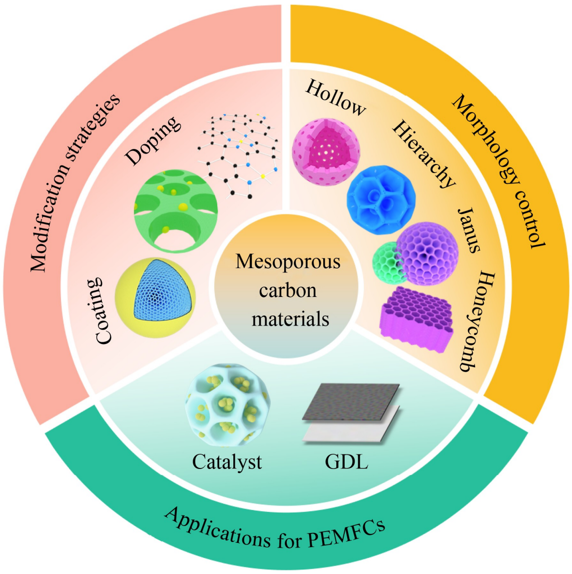
Regulation of mesoporous pore size and morphology: Different applications in PEMFCs require mesoporous carbon materials with varying pore sizes and morphologies. For example, the performance of mesoporous carbon materials for the ORR in MEA can differ greatly based on pore size and depth [97]. Achieving precise and on-demand synthesis of mesoporous carbon materials is essential. Although a variety of preparation methods, including template and template-free methods, are available, precise control of the pore size and morphology of mesopores remains a challenge. Future efforts should focus on developing new preparation techniques that meet the needs of the times. For example, novel template materials can be developed, which can make mesopores precisely replicate the template structure like hard templates and can also detach from the system during carbonization like soft templates. Furthermore, these innovative templates are designed to be recyclable, enhancing their sustainability and cost-effectiveness. It is hoped that this innovative method can simplify the synthesis steps while precisely regulating the mesopore structure and reduce the cost, which will be more advantageous in industrialized applications.

Modification of mesoporous carbon: Current modifications to mesoporous carbon materials primarily involves doping with heteroatoms and introducing functional groups to alter chemical properties. However, these singular doping methods often do not lead to a significant overall improvement in performance. Similar to the regulation of mesoporous structures, modifications should be tailored to specific reactions and applications. For instance, employing template agents containing specific elements can facilitate the precise introduction of heteroatoms into mesopores upon template removal, while simultaneously introducing functional groups to the surface of the mesoporous carbon. This approach could create distinct chemical properties both inside and outside the pores, improving the reaction kinetics.

Integration with machine-learning: Recent advancements in mesoporous carbon materials have enhanced the performance of PEMFCs, but the relationship between mesoporous structures and their electrochemical performance requires further investigation. Machine learning has the potential to accelerate the screening of large libraries of mesoporous structures and quantify the effects of different mesoporous architectures on catalyst performance and stability. By integrating methods such as finite element simulations, machine learning not only provides insights into experimental observations but also predicts the influence of pore size and morphology on PEMFC performance. Ultimately, it may facilitate the development of mesoporous carbon materials with precisely tailored structures and functionalities, improving performance.

Reducing production costs to advance commercialization: While traditional carbon sources, such as phenolic resins and bitumen, provide a suitable carbon skeletons for mesoporous carbon synthesis, their high cost and intricate preparation procedures elevate production expenses. Recent studies have explored using biomass waste, such as agricultural straw, fruit shells, and wood waste, as alternative, cost-effective carbon sources. These biomass feedstocks are not only inexpensive but also abundantly available, which can significantly reduce raw materials costs. For instance, Liu et al. [132] developed high-surface-area porous carbon materials using pomelo peels as the raw material, through a process of pre-carbonization, hydrothermal treatment, followed by segmental warming and KOH activation. The cost of pomelo peel is negligible compared to phenolic resins, making it a promising alternative for large-scale production. Combining low-cost biomass carbon source with simplified preparation process holds promise for effectively decreasing the production costs and accelerate the commercial deployment of mesoporous carbon materials in fuel cell catalysts.

In summary, mesoporous carbon materials exhibit considerable potential for application in PEMFCs, though challenges persist in terms of pore size control, chemical modification, and integration with PEMFCs. Continued advances in material design, modification techniques, and the integration of innovative research methodologies will help overcome these challenges, thereby further enhancing the performance and applicability of mesoporous carbon materials. As technology advances and understanding of mesoporous carbon materials deepens, they are expected to play a more prominent role in enhancing the efficiency and sustainability of PEMFCs, contributing significantly to the advancement of clean energy applications.



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Competing Interests The authors declare that they have no competing interests.

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