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Recent Advances in MOF-Derived Functional Materials for Electrochemical Energy Conversion and Storage

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Abstract. Driven by low-carbon energy development, electrochemical energy conversion and storage technologies, including water electrolysis and zinc-air batteries, have drawn attention. However, the oxygen evolution reaction (OER) and oxygen reduction reaction (ORR) are plagued by sluggish reaction kinetics and high overpotential, limiting the energy efficiency of electrochemical devices. Though noble metal-based catalysts deliver outstanding catalytic performance, they are constrained by high cost and resource scarcity, making them unable to meet the demands of large-scale applications. Consequently, efficient and stable non-noble metal electrocatalysts have attracted increasing research interest. Metal-organic frameworks (MOFs), featuring tunable compositions, ordered pores, and uniformly distributed metal sites, serve as promising precursors for non-noble metal-derived electrocatalysts. Thus, this review highlights three construction strategies for MOF-derived electrocatalytic materials, including composition regulation, non-metal doping, and structural engineering, and discusses their impacts on metal coordination, electronic structures, and mass transport. Besides, the research progress of MOF-derived materials in the OER of water electrolysis and bifunctional oxygen electrocatalysis for zinc-air batteries is summarized. Combined with in situ characterization and density functional theory (DFT) calculations, the active sites, catalytic evolution, and underlying reaction mechanisms are discussed. The results demonstrate that despite favorable catalytic performance, MOF-derived electrocatalysts still face challenges in the analysis of dynamic evolution of active phases, stability at high current densities, consistency in scalable preparation, and device compatibility.

Keywords: Metal-Organic Frameworks, Derived Materials, Electrocatalysis, Electrochemical Performance

1. Introduction

Driven by the global energy transition and carbon neutrality goals, electrochemical energy conversion and storage technologies, including hydrogen production via water electrolysis and rechargeable zinc-air batteries, are important for low-carbon and sustainable energy systems. However, key electrode reactions like the hydrogen evolution reaction (HER), oxygen evolution reaction (OER), and oxygen reduction reaction (ORR) suffer from sluggish kinetics and high overpotentials. Traditional noble metal-based catalysts suffer from high costs and limited reserves,

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which hinder their large-scale application. Therefore, developing highly efficient and stable non-noble metal electrocatalysts is a core issue that urgently needs to be addressed in this field [1]. Metal-organic frameworks (MOFs), featuring tunable compositions, ordered porous structures, and uniformly dispersed metal sites, are ideal precursors for high-performance derived electrocatalysts. By combining controllable conversion (pyrolysis, phosphidation, and sulfidation) with structural optimization (defect engineering and heterointerface regulation), MOF-derived materials retain precursor features, regulate electronic structures and mass transport, and improve electrocatalytic performance [2]. The combination of in-situ characterization techniques and density functional theory (DFT) calculations also provides an important research paradigm for unraveling the dynamic reconstruction behavior and authentic active sites during the reaction process. Despite their advantages, MOF-derived electrocatalysts encounter challenges in identifying active phases, achieving reproducible large-scale synthesis, and maintaining stability at high current densities. This review summarizes their design strategies, structure-activity relationships, and applications to provide insights into practical electrochemical energy technologies.

2. Design strategies and synthesis methods of MOF-derived materials

2.1. Composition-regulated MOF-derived materials

MOFs with uniformly dispersed metal nodes and tunable compositions serve as suitable precursors for controlling the composition and structure of derived materials. By varying the metal centers in MOFs, the metallic phase composition, coordination environment, and electronic structure can be tuned during derivatization, thereby regulating catalytic activity.

During pyrolysis, M-O and M-N coordination structures in monometallic MOFs undergo gradual transformation. Organic ligands are carbonized into carbon matrices, while metal species migrate and form derived phases, such as metals, oxides, and carbides. In bimetallic MOFs, differences in metal diffusion and reduction behaviors lead to local compositional variations and coordination changes, generating heterogeneous metal sites and defect structures with electronic modulation effects [3].

Beyond metal center regulation, non-metal incorporation provides an effective strategy for tuning the composition of MOF-derived materials. During phosphidation, reactive phosphorus species interact with metal centers, transforming M-O or M-N bonds into M-P coordination. Hussain et al. reported that phosphidation of Ni-MOF replaces the original Ni-O octahedral coordination with Ni-P structures, significantly altering the coordination environment [4]. In bimetallic NiCo phosphides, Ni and Co jointly participate in lattice formation, and their differences in atomic radii and valence states induce local electronic modulation. Similarly, sulfidation generates M-S coordination through sulfur–metal interactions, modifying the composition and electronic states of active metal sites.

2.2. Non-metal-doped MOF-derived materials

MOF frameworks serve as carbon precursors for the synthesis of heteroatom-doped carbon materials.

By introducing heteroatoms such as N, S, and P, the electronic structure, surface chemistry, and defect characteristics of carbon matrices can be regulated, tuning catalytic activity. Meanwhile, heteroatom incorporation methods and chemical states govern the local coordination and electronic effects.

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Nitrogen doping is commonly achieved using N-containing ligands (e.g., 2-methylimidazole) or external nitrogen precursors. During pyrolysis, nitrogen species are transformed into pyridinic N, pyrrolic N, and graphitic N. Among these configurations, pyridinic N and pyrrolic N are mainly located at carbon edges and defect sites, serving as catalytic active sites, whereas graphitic N enhances charge transfer by modifying the electronic structure of carbon layers. Furthermore, XPS analysis showed that in N-doped graphene-like carbon/MOF-74 composites, the proportion of pyridinic N increased from 12.7% in pristine MOF-74 to 29.0%, thus indicating that doping effectively regulates nitrogen species distribution and surface chemical environments [5].

Compared with N, larger atomic-radius elements such as S and P are generally introduced through post-treatment, inducing local carbon-framework distortion, defects, and asymmetric coordination environments to further modify electronic properties. Generally, heteroatom doping can be achieved through two main approaches: in situ doping and post-treatment doping. In situ doping introduces heteroatom-containing precursors during MOF synthesis, thus enabling uniform incorporation during pyrolysis, whereas post-treatment doping modifies carbon structures through vapor diffusion and impregnation [6]. The resulting doping configurations are influenced by multiple factors, including the dopant type, pyrolysis temperature, atmosphere, and precursor ratio, which collectively determine heteroatom bonding states and distributions. By optimizing these parameters, the electronic properties and catalytic performance of MOF-derived carbon materials can be further improved.

2.3. Structure-regulated mof-derived materials

By employing structural engineering strategies such as templated conversion, oriented growth, and spatial assembly, MOF-derived materials achieve precise control over morphology, architecture, and compositional distribution. The construction approaches and representative studies corresponding to different structural types are summarized in Table 1.

Table 1. Construction strategies of structure-regulated mof-derived materials

Structural Types	Formation Mechanism	Typical Synthesis Strategies	Representative Studies
Hollow Structures	Formation of internal cavities via compositional migration driven by the Kirkendall effect, selective etching, or ion exchange	MOF Sacrificial Template Method; Ion Exchange Method; Selective Etching Method	MIL-88A-Derived CoFeP-Based Hierarchical Hollow Structures
Core-Shell Structures and Heterostructures	Construction of interfacial regions via the spatial distribution of distinct components to form core-shell or multilayer heterostructures	Stepwise Growth Method; Layer-by-Layer Assembly Method; Template-Induced In-Situ Growth Method	CoFeP/CoFeP/NP-C Multilayer Heterostructure

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Table 1. (continued)

Ordered Nanoarray and Hierarchical Structures	Maintenance of oriented alignment via substrate-induced effects to realize structural ordering	Substrate-Induced Growth Method; Hydrothermal Oriented Deposition Method; Morphology Regulation Method	Zn–Co–Ni Sulfide Nanosword Arrays Supported on Nickel Foam

Among various structural designs, hollow structures are mainly fabricated through compositional migration and spatial reconstruction during the templated conversion of MOFs. For example, Zhang et al. used MIL-88A as a template to construct cavity structures via cation exchange and etching, followed by conversion into CoFeP-based hierarchical hollow structures [7]. In addition, core-shell structures and heterostructures further utilize the spatial distribution differences between components to generate interfacial regions. For instance, CoFe-LDH and CoFe-PB were successively grown on MIL-88A surfaces, followed by phosphidation to generate a CoFeP/CoFeP/NP-C multilayer structure with hierarchical component distribution [7].

Further extending structural regulation, ordered nanoarray structures facilitate the oriented growth of MOF precursors through substrate-induced effects and preserve the array morphology during subsequent conversion. Du et al. constructed Zn-Co-Ni sulfide nanosword arrays on nickel foam, while Cao et al. achieved in situ growth of ZIF-8 on $Ti_3C_2T_x$ MXene surfaces followed by sulfidation, forming a 0D/2D composite structure with uniformly dispersed ZnS nanodots [8].

3. Key applications of MOF-derived materials in electrocatalytic energy conversion

3.1. Electrocatalytic performance in water electrolysis

Water electrolysis consists of the cathodic HER and the anodic OER. In particular, the OER involves a four-electron transfer process with sluggish reaction kinetics, serving as the key step that limits the overall water-splitting efficiency. To address this limitation, MOF-derived non-noble metal catalysts improve OER performance through compositional conversion, molecular structure regulation, and interfacial construction. These strategies mainly regulate active centers, electronic structures, and charge transport processes. Among them, compositional conversion modifies the phase composition and coordination environment of MOF derivatives through post-treatment approaches, tuning the electronic structure of active centers. Wang et al. employed NiFe Prussian blue analogs as precursors to fabricate porous P-CoNiFe catalysts via low-temperature phosphidation [9]. During phosphidation, Fe-P, Ni-P, and Co-P coordination structures are formed, which facilitate the modulation of metal electronic states and increase active-site exposure through the porous structure. The catalyst requires an overpotential of 360 mV to reach 30 $mA \cdot cm^{-2}$ under alkaline conditions, showing favorable OER activity.

In contrast, molecular design strategies focus on tuning the coordination environment and intrinsic active sites based on the structure of MOF precursors. For example, Han et al. constructed NiFe-BPD-FcDA nanoflower catalysts using a donor-acceptor dual-ligand strategy. The introduction of 4,4'-bipyridine ligands alters the connection mode of metal nodes and modulates the electronic structure of Fe sites via the ligand electronic effect, thereby optimizing the adsorption process of OH* intermediates [10]. This catalyst delivers an OER overpotential of 244 mV at 10 $mA \cdot cm^{-2}$ with a Tafel slope of 99 $mV \cdot dec^{-1}$. Furthermore, interfacial engineering improves electron transport and electrode stability by coupling active materials with conductive supports. For example, Mohammadi

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Kalimani et al. supported MOF-derived $CuMn_2O_4$ on CNT-modified carbon felt. In this composite system, CNTs form a continuous conductive network that promotes electron transport and improves the dispersion of the catalytic layer, thereby reducing the electrode overpotential to 337 mV at 10 $mA \cdot cm^{-2}$ with a Tafel slope of 62 $mV \cdot dec^{-1}$ [11].

3.2. Electrocatalytic performance in metal-air batteries

With their high theoretical energy density and inherent resource advantages, rechargeable zinc-air batteries are regarded as promising energy storage systems with great application potential. However, the air cathode involves both the ORR and OER, both of which suffer from sluggish reaction kinetics, leading to increased battery polarization and degraded cycling stability. Although conventional noble metal catalysts (e.g., Pt, RuO_2, and IrO_2) exhibit high catalytic activity, they are constrained by issues such as high cost, resource scarcity, and limited bifunctional catalytic performance. Benefiting from the tailorable composition and structure, MOF-derived materials can achieve optimized ORR/OER bifunctional catalytic performance by constructing multifunctional active structures. Based on regulation mechanisms, studies mainly focus on heterointerfaces, spatial structures, and single-atom sites. Among these strategies, heterointerfacial regulation optimizes the adsorption process of oxygen intermediates mainly through electronic coupling between different components. For example, Lan et al. fabricated NiFe-MOF/$NiFe_2O_4$ hollow nanospheres via a one-step ligand-deficient solvothermal method. The heterointerface between the two phases facilitates electronic interaction while also improving oxygen species adsorption and reaction kinetics. These structural advantages enable the material to deliver an OER overpotential of 302 mV at 10 $mA \cdot cm^{-2}$, and the assembled zinc-air battery achieves a peak power density of 158.4 $mW \cdot cm^{-2}$ [12].

In addition to interfacial regulation, spatial structure optimization enhances device performance by improving mass transport and the utilization efficiency of active sites. Wang et al. employed leaf-like ZIF-L(Co) as the core and Fe-doped ZIF-L as the shell, and obtained FeCo@NC-II core-shell catalysts after pyrolysis. This structure facilitates reactant transport through hierarchical pores and modulates the electronic structure via the synergistic effect between FeCo alloy and nitrogen-doped carbon, reducing the bifunctional potential gap ΔE to 644 mV and enabling the zinc–air battery to reach a peak power density of 212.4 $mW \cdot cm^{-2}$ [13]. Furthermore, single-atom sites enhance metal utilization efficiency and oxygen electrocatalysis. Zhang et al. introduced iron porphyrin into the UiO-66 framework to construct a multicomponent MOF, which was transformed into Fe-N_4 single-atom catalysts after pyrolysis. Benefiting from stably dispersed atomic-level active sites, this material promotes both ORR and OER, yielding a bifunctional potential gap of only 0.59 V and a peak power density of 340.5 $mW \cdot cm^{-2}$ for the liquid zinc-air battery [14].

3.3. Active site regulation and reaction mechanisms

The catalytic performance of MOF-derived materials is closely correlated with the structural state of active sites. Modulation of catalytic performance mainly involves the metal coordination environment (M-N, M-O, M-P), bimetallic electronic synergy, and surface reconstruction behavior during the reaction process. Given that derived materials may undergo structural and valence state changes under operating conditions, combining in-situ characterization techniques like XAS and Raman with DFT calculations to analyze active phase evolution and reaction pathways has become a key approach for understanding catalytic mechanisms. In bimetallic systems, intermetallic electronic interactions can tune the local electronic structure, which affects the adsorption behavior of reaction intermediates. For example, in FeCo@NC-II, FeCo alloy sites and M-N coordination structures

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regulate local electron distribution and oxygen-intermediate adsorption strength [13]. DFT calculations demonstrate that the electronic coupling between Fe and Co can adjust the d-band center position of Co sites, optimizing the adsorption energy of key intermediates such as OH* [15]. Furthermore, after the introduction of sulfur-containing ligands, the coordination effect of S atoms can further modify the electronic environment of metal centers and influence the charge distribution around active sites.

Beyond initial structure regulation, surface reconstruction during the reaction is also a key factor determining the genuine active phases. In most MOF-derived OER systems, catalysts undergo surface oxidation and coordination environment adjustment under operating conditions. The actual active species are typically metal (oxy)hydroxides, while precursor structures mainly govern the formation, dispersion, and stability of reconstructed active sites [2]. In MOF-derived phosphide systems, Fe doping in the Fe–Ni_2P/C catalyst can modulate the electronic structure of Ni sites and influence the formation process of OOH intermediates; meanwhile, phosphidation-induced structural changes help improve active site exposure and electron transport [16].

4. Existing challenges and optimization strategies

4.1. Limitations in active site regulation and reaction mechanisms

Accurate determination of the active site structure and actual catalytic mechanism of MOF-derived electrocatalytic materials remains challenging. During derivation, metal species undergo migration, agglomeration, and valence changes, causing deviations between fabricated and designed structures. Under operating conditions, surface coordination environments and electronic states continuously evolve, making initial structures insufficient to represent active states. Therefore, correlating derived structures, dynamic active sites, and catalytic performance remains challenging.

At present, the regulation of single-atom sites, dual-atom sites, and defect structures represents a key strategy for identifying active centers. Among these structures, single-atom sites stabilize metal centers via coordination with MOF-derived carbon frameworks and regulate intermediate adsorption through electronic interactions between metal centers and coordinating atoms. In contrast, dual-atom sites modify local electronic structures through synergistic interactions between adjacent metal atoms [17]. In addition, defect structures can alter the metal coordination state and electron distribution, offering additional flexibility for activity regulation. In-situ techniques such as XAS and Raman enable tracking of valence and structural evolution during the reaction, and when combined with DFT calculations, they facilitate analysis of the relationship between electronic structure and reaction energy barriers [18, 19]. Nevertheless, most existing computational models are constructed based on idealized surface structures, and thus cannot adequately describe interfacial reconstruction and active phase transformation under realistic reaction conditions.

4.2. Bottlenecks in stability and electrical conductivity

MOF-derived materials often encounter challenges in maintaining structural stability and efficient charge transport during electrocatalysis. In terms of structural stability, under strongly acidic, strongly alkaline, or high-potential conditions, metal-ligand coordination changes may trigger reconstruction, active-site leaching, and performance decay. At the same time, some MOFs and their derived carbon materials suffer from insufficient electrical conductivity and low electron transport rates, which compromise the charge supply to active sites and hinder reaction kinetics.

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To improve electron transport, studies typically introduce conductive components to construct conductive networks. Materials such as graphene, carbon nanotubes, and MXene form continuous electron transport channels within catalysts, reducing charge-transfer resistance. For example, the hybridization of rGO with NH_2-MIL-125 reduces the charge transfer resistance; in the MXene/ZIF-8 system, MXene not only enhances electron transport but promotes the dispersion of MOF particles [20]. Beyond enhancing electrical conductivity, interfacial regulation modifies the local electronic structure of active sites. By constructing secondary phase interfaces between MOFs and other phases such as metal oxides and layered double hydroxides, the electronic interactions among distinct components can tune the local charge distribution and alter the adsorption behavior of reaction intermediates. Accordingly, the construction of conductive networks mainly acts on the electron transport process, whereas interfacial regulation focuses on optimizing the properties of active centers. The two strategies often function simultaneously in composite catalytic systems, yet their specific contributions to catalytic performance enhancement still need to be further distinguished via in-situ characterization techniques.

4.3. Limitations in scalable preparation and device applications

MOF-derived materials are typically fabricated via solvothermal or hydrothermal methods. However, these methods are subject to limitations in heat transfer, mass transfer, and reaction condition control during scale-up, leading to batch-to-batch variations in material structures and performances. In addition, most laboratory electrochemical tests are conducted in three-electrode systems, whereas practical devices involve different electrode structures, charge transport pathways, and mass diffusion conditions, thereby making it difficult to directly translate catalytic performance into device-level performance.

To enable scalable preparation, continuous-flow synthesis and low-solvent routes can improve production efficiency and process stability. For instance, when MOF-303 is synthesized through a continuous-flow approach, its space-time yield (STY) can reach 700 $kg \cdot m^{-3} \cdot day^{-1}$, exceeding the 287 $kg \cdot m^{-3} \cdot day^{-1}$ of conventional batch hydrothermal synthesis [21]. Furthermore, aqueous synthesis and solvent-free synthesis can reduce solvent consumption, and process costs can be evaluated through techno-economic analysis [22]. During device integration, powdered catalysts usually need to be loaded onto conductive substrates with the aid of binders, which may compromise electron transport and active site exposure. The in-situ growth strategy can reduce the interfacial resistance between the catalytic layer and the substrate. For example, directly constructing MOF-derived electrode structures on carbon materials or metal foam substrates enhances electrode stability and transport efficiency [21]. Nevertheless, the electrode structure design, consistency of scalable preparation, and long-term operational performance in practical devices still require further validation.

5. Conclusion

This review summarizes the structural design, structure-activity relationships, and applications of MOF-derived electrocatalysts. Through composition regulation, heteroatom doping, and structural engineering, MOF-derived materials regulate metal coordination, electronic structures, and pore architectures to enhance active-site exposure, charge transport, and reactant diffusion. In systems such as the oxygen evolution reaction in water electrolysis and oxygen electrocatalysis in zinc-air batteries, these materials exhibit favorable catalytic activity and stability, providing a material foundation for designing non-noble metal electrocatalysts. However, MOF-derived electrocatalysts

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still face several challenges. The dynamic active-phase evolution during reactions complicates the identification of true active sites and reaction mechanisms. Under high-current-density conditions, structural stability and charge transport require further improvement. Meanwhile, laboratory-scale preparation methods still suffer from deficiencies in batch consistency, cost control, and device compatibility during the scale-up process. Future research should integrate in-situ characterization techniques and theoretical calculations to further clarify the evolution behavior of active structures under reaction conditions. Structural optimization and interfacial regulation strategies should be adopted to enhance the stability and transport performance of the materials under practical operating conditions. In addition, synthetic methodologies compatible with scalable preparation need to be developed to promote the translation of MOF-derived electrocatalytic materials from laboratory research to practical device applications.

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