

Review: Recent Advances in Metal Organic Frameworks Functionalized with Bioinspired Ligands for Selective Catalysis and Sensing

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ABSTRACT: Metal-organic frameworks (MOFs) is one of the versatile classes of crystalline porous materials owing to their exceptionally high surface areas, tunable pore architecture and flexible metal-ligand chemistry. In recent years, the integration of bioinspired ligands and MOFs' principal design has gained attention as an effective strategy to enhance their selectivity, efficiency and functionality in catalytic and sensing applications. This review provides a comprehensive overview of recent advances in synthesis, functionalization strategies such as defect engineering, post-synthetic modification and composite formation are highlighted as powerful tools for tailoring active sites and improving stability and performance. Furthermore, recent developments in MOF-based sensing platforms are reviewed, illustrating how bioinspired MOFs function as Nanozymes, optical transducers and electrochemical sensors for the selective detection of biomolecules. This review aims to provide valuable insight for the rational design of next generation MOFs materials for sustainable catalysts and advanced sensing technologies.

Keywords: *Metal-organic framework, Catalysis, Sensing, Bioinspired ligands*

1. Introduction

Since the discovery of metal organic frameworks (MOFs) in 1990, they have become one of the important classes of modern chemistry and material science. MOFs were under the attention of the researchers because of their incredible tunability, highly porous structures and high surface areas [1-4].

The crystalline porous coordination polymers differ in shape and size. The inorganic building blocks like organic ligands and clusters or metal ions link them together. The diversity of these compounds comes from the variety of linkers and clusters, furthermore, the coordination bonds orientations, customizable functions and tunable porosities, making the MOFs highly versatile for variety of applications [1,4-6].

Compared to the classic porous materials such as activated carbons and Zeolites, MOFs have exceptional structural variety and chemical flexibility, which leads to a customized function that are needed for particular technical requirements [7-9].

MOFs were under the interest of researchers' investigations of MOFs use in the gas environment such as real time carbon dioxide (CO₂) captured from vehicle emissions or variable emissions from chemical facilities, gas separation, water purification, catalysis, sensing and even biomedical drug delivery [9-12].

It is easier to regulate the characteristics within the pore region of MOFs due to their organized crystalline structure. MOFs can be significantly altered to meet their intended uses by altering the metal ions or organic linkers [13-15].

Over the years, the synthesis methods of MOFs have developed significantly the variety of methods are used to control the size, shape and functionality. The traditional synthesis methods such as hydrothermal and solvothermal, still widely used because of their ability to produce well defined structures with highly crystalline materials. However, an alternative synthesizing method was needed to address the challenges such as, cost efficiency, environmental sustainability and scalability. The alternative methods explored included sonochemistry, electrochemical methods, the URAM process, precipitation methods and microwave assisted synthesis [16-21].

MOFs are now being hybridized with advanced materials such as graphene, carbon nanotubes and covalent organic frameworks (COFs) to create multifunctional systems that combine high selectivity, enhanced mechanical strength and improved charge transport properties [22-25].

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2. Synthesis and Functionalization

Recent progress in the synthesis and functionalization of metal-organic frameworks (MOFs) has been pivotal in overcoming early limitations in stability, processability and applicability.

Contemporary strategies can be grouped into alternative synthesis routes which enables scalability and greener processing, defect engineering and deliberate non-stoichiometry to create active sites, post-synthetic modification (PSM) techniques to introduce functionality without destroying MOF integrity and lastly formation of composites and MOF-derived materials to enhance mechanical, electrical or chemical properties [26-30].

The traditional synthesis routes of metal-organic frameworks.

2.1. solvothermal/Hydrothermal synthesis

In the low-temperature reaction organic linker and the metal salt are mixed in the solvent and heated in vials. The reaction is called hydrothermal reaction when the solvent used is water, when the solvent used have a higher boiling point it's called solvothermal synthesis [31].

2.2. Slow Diffusion method

This method to obtain MOF single crystal by mixing the ligand solution and the metal ions in an appropriate ratio to form single crystals over time due to diffusion of two different densities of solvents to form a better quality and larger single crystal [32-33].

2.3. Microwave-assisted method

The method is used in preparing an organic and nanoporous inorganic material. The advantages of this method are the low cost, the high yield and the short time required to obtain the product, another advantage to this method is that the microwave facilitate the motion of the molecules leading to the formation of the crystals with the ability to control the shape and size by adjusting the temperature and concentration of the reaction appropriately [34-37].

2.4. Mechanochemical method

This method is based on using mechanical forces instead of solvent at room temperature to form coordination bonds by using the automatic ball mills or less used by manual grinding sometimes to obtain one-dimensional, two-dimensional and three-dimensional coordination polymer by adding a small amount of solvent into the solid mixture of the reaction. This method accelerates the reaction time by facilitating heats, mass transfer, reduces particle size and locally melts the reagent, using this method in synthesizing MOFs have an important advantage being an alternative to the high temperature and pressure solvo/hydrothermal synthesis but its main disadvantage is the isolation of amorphous product, unsuitable for the X-ray structural studies of the single crystal [38-42].

3. Bio-inspired functionalization strategies

Enzymatic catalysis had the attention of the chemists because of the selectivity and high efficiency but due to the vulnerability and structural complexity hampers the potential of enzymes in the application. So, to satisfy the practical demand the researchers began trying to produce enzymes active sites molecular analogue. The concept of artificial enzymes was first promoted by Breslow and Overman, but the fruitful results and mimicking enzymes were achieved by researchers such as Groves, Gray, Lippard, Rauchfuss, Darensbourg and Halm [43-48]. Figure 1 illustrates the design of metalloenzyme-inspired active sites inside MOFs.

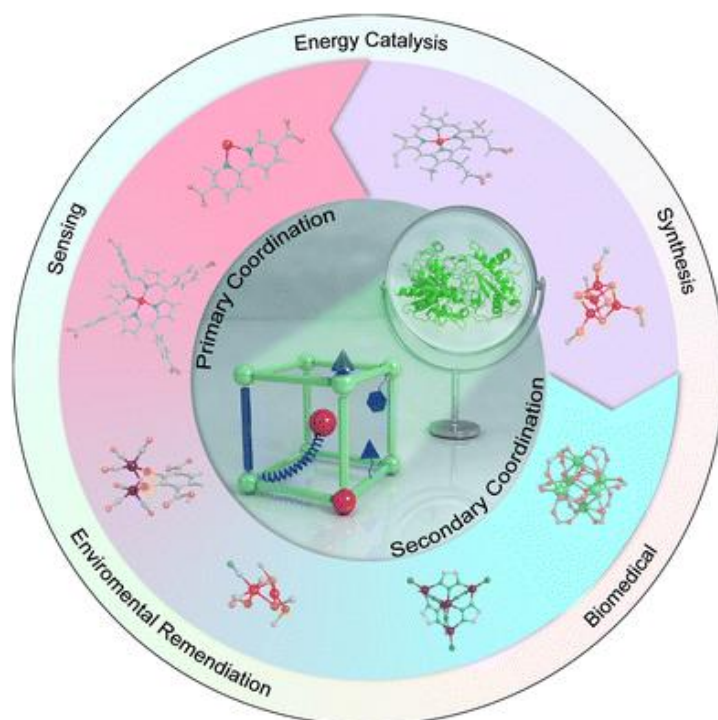


Figure 1: Design of metalloenzyme-inspired active sites inside MOFs for diverse catalytic applications at the atomic level ⁴⁴.

3.1 Bioinspired metal active site design

Metalloenzymes in nature rely heavily on metal cofactors to drive the chemical reactions required for life. Each enzymes' metal ion supports a distinct function and involvements in catalysis and the result of natural selection. Monometallic sites like heme-containing oxygenase and Zn-OH center of metallohydrolase and multi-metallic clusters like multicopper oxidases, Fe₄S₄, FeMo nitrogenase, Ni-Fe and Fe-Fe hydrogenase, make up the majority of metal cofactors among all metalloenzymes [49-50].

MOFs have the ability to mimic the catalytic sites vividly at the atomic scale due to the diversity and spatially isolated metal ions. Secondary building units (SBUs) and organic linkers are the focus two main buildings in the construction of enzyme like active pockets MOFs [49].

Various metal classes and ligands such as TCPP, Imidazole, thiolate, enable biomimetic MOFs to accurately replicate the metal coordination structures of metalloenzymes [49-51].

3.2 Alternative Synthesis Routes – Scale, Speed and Sustainability

Cambridge crystallographic data Centre (CCDC) has more than 12500 MOF structures reported. Some of these structures demonstrate highly promising outcomes in variety of

applications including Biomedicine, catalysis, water harvesting, Carbon capture and hydrogen storage [54-55].

Optimized protocols were developed in order to produce higher yields of metal-organic frameworks (MOFs) in laboratory pilot scale (kilogram scale) instead of laboratory scale (gram scale). Different parameters such as the type and speed of mixing, mass, the type of reactions, heat transfer and reactors are important for the pilot scale production process [56-61].

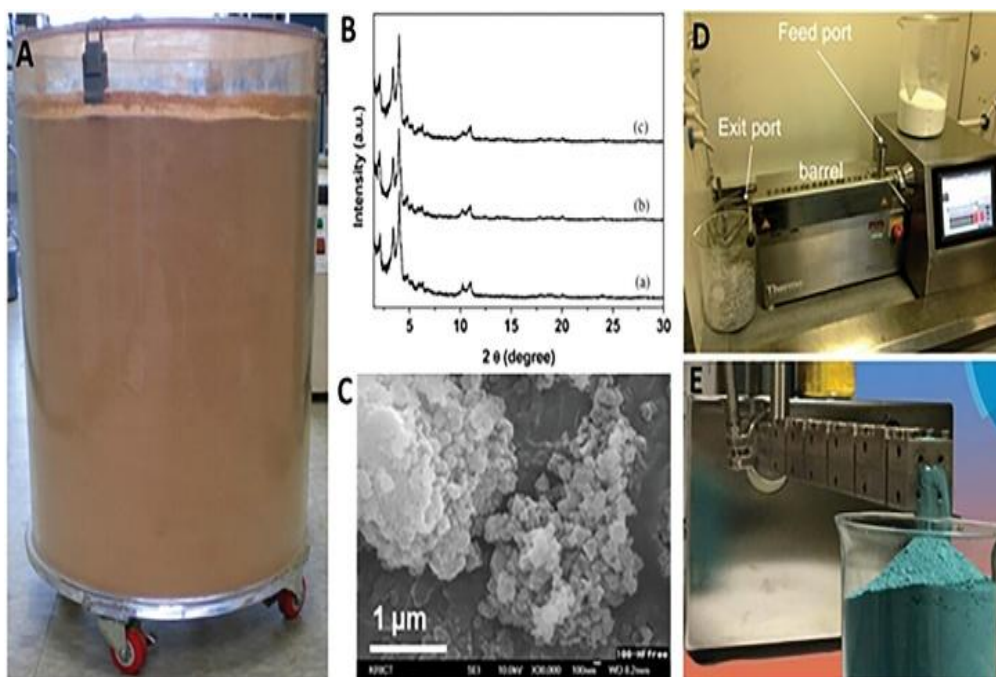


Figure 2: A. poly (methyl methacrylate) transparent box B. PXRD pattern a) before exposure to boiling water b) after 3 days of exposure c) 5 days ⁶² C. SEM image of MIL-100(Fe) D. twin-screw extruder showing the different parts. E. MOF production using the screw extruder ⁶⁴. Reproduced with permission ^{62, 64}. Copyright 2023. Elsevier.

A large-scale synthesis of MOF was explored, since the reporting of the mechanochemical synthesis of porous MOF Cu (INA)₂. Ball milling preparing the MOF was in scale less than 1g [62,64-66]. Figure 3

Green and bio-inspired synthesis- using water, benign solvents, biomass-derived linkers or room-temperature conditions has expanded producing (Bio-MOFs) and frameworks with lower environmental footprints. The movement toward greener synthesis seeks to couple material performance with lifecycle sustainability.

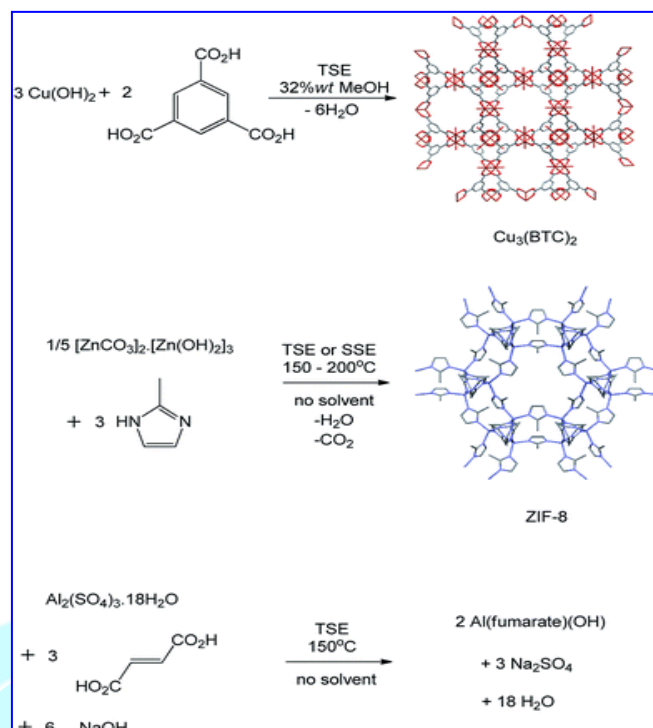


Figure 3: illustrating scheme of MOF $\text{Cu}_3(\text{BTC})_2$ (HKUST-1), $\text{Zn}(\text{methylimidazolate})_2$ (ZIF-8) synthesis reaction by TSE or SSE and $\text{Al}(\text{fumarate})(\text{OH})$ [52].

3.3 Green methodologies for MOF synthesis

The green chemistry's 12 principles can be adapted in MOF synthesis process. MOF can be synthesized by the basis of the green methodologies standard aspects such as **(a)** safe reaction media (such as water and supercritical solvents), **(b)** biocompatible building blocks, **(c)** continuous production approaches **(d)** reduced energy input and **(e)** performance design of MOFs through theoretical predictions [67-70]. Green and bio-inspired synthesis- using water, benign solvents, biomass-derived linkers or room-temperature conditions has expanded producing (Bio-MOFs) and frameworks with lower environmental footprints. The movement toward greener synthesis seeks to couple material performance with lifecycle sustainability.

a. Solvent-based synthesis: an integral step in synthesizing MOFs is selecting the solvent since its important role in regulating the coordination environment formation. Solvent can take a part in the coordination process by coordinating with metal ions as well as being structure-directing agent [69,71]. Using water as solvent resulted in the discovery of anew

variety of archetype MOFs, such as MOF-74, CaU-22(Zn) and MIL-91(Ti), yet some MOFs are water-sensitive especially those containing carboxylate group such as $\text{Cu}_3(\text{BTC})_2$ and Mg-MOF74 [69,72-74].

Due to the sensitivity of these MOFs the existence of water might result in crystallinity loss, structural decomposition and phase changes. However, UiO-66 (carboxylate containing MOF) has water stability due to several stereotypes and chemical factors. The use of water in the process would offer lower environmental impacts would be welcomed by the industry. The eco-friendly MOFs that lead the way in the direction of using MOFs in toxic adsorption from the gaseous or aqueous systems [74-78].

3.4 Defect Engineering of MOFs

This process in the synthesis of MOFs opens an avenue in terms of their physical properties such as conductance and mechanical response. Also, it provides new opportunities in the catalysis and adsorption fields [29].

Some incredible characteristics are obtained by the defects of the metal-organic frameworks. Since in MOFs these defects might have extremely subtle effects on them, the enhancement of MOFs both functional and structural can be obtained by certain degree of defects [79-81].

A change of properties might be accompanied by structural defects due to the electron localization, lattice distortion, fracture and recombination of the chemical bonds. These defects provide a potential active site because some of the atoms in MOFs crystals become unsaturated due to the defect's existence. [82-84]

Defect engineering is utilizing MOFs crystals already existence defects are considered a treasure for the enhancement of MOFs performance for potential applications [26].

3.5 Post-synthetic modification methods (PSM)

PSM is a chemical modification of linkers, grafting of functional moieties or exchange/metalation of secondary building units after framework assembly remains a powerful route to introduce catalytic sites, chelators, polymer brushes or sensing groups [85-87].

Post-synthetic modification is under the interest of many researchers for providing a solution for the unpredictable product [87,88].

PSM is utilized in synthesizing new scaffolds with enhanced properties when compared to the original frameworks. The aspect of PSM lies in the fact that the enhanced MOFs cannot be obtained by the classic synthesis methods [89-91].

By changing the chemical composition of MOFs, they gain new physical and chemical properties leading to the expansion of the applications of MOFs. [92-94].

PSM yields MOFs optimized for CO₂ chemisorption, selective separations or anchoring of catalytic nanoparticles, PSM also underpins membrane functionalization for molecular separation [95-97].

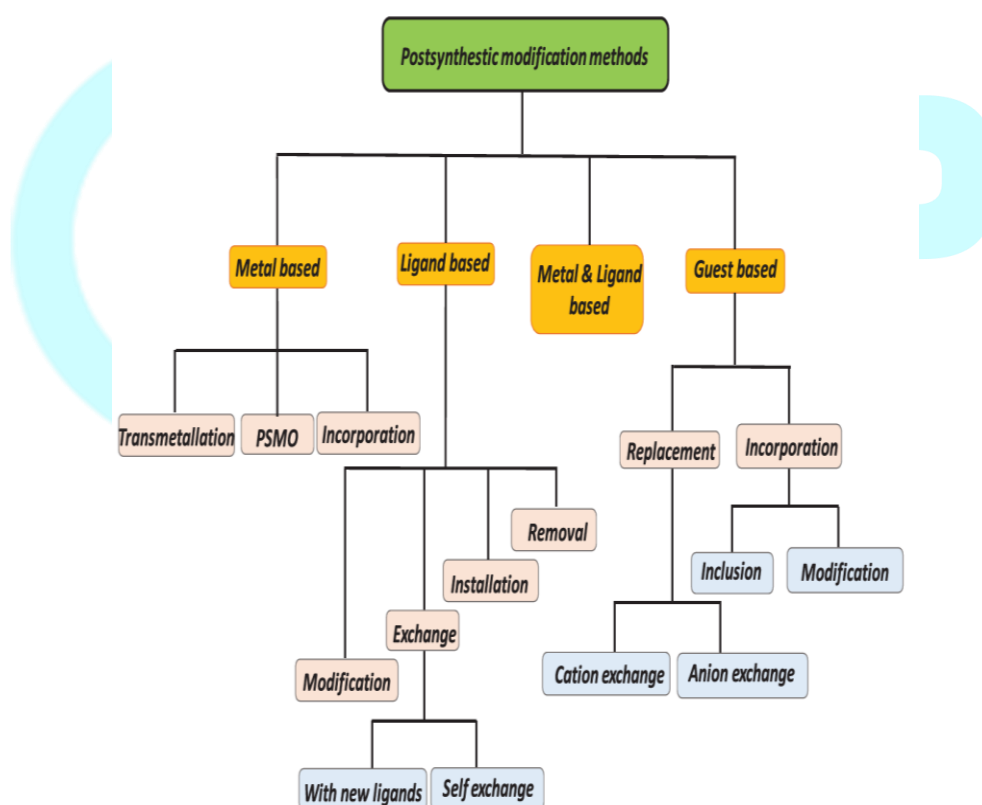


Figure 4: Photosynthetic modification methods

3.6 Composites, hybridization and MOF-derived materials

Combining MOFs with carbon materials such as graphene, carbon nanotubes (CNTs), activated carbons, polymers, nanocellulose or inorganic supports typically improves conductivity, mechanical robustness and processability [24, 98].

MOFs-derived carbons which obtained via controlled pyrolysis have the ability to preserve hierarchical porosity and embed catalytically active metal species producing highly active electrocatalysis and adsorption media [99-101].

4. Bio-inspired selective catalysis and sensing functions

MOFs have the ability to act as biosensors and enzyme mimics (nano-enzymes) due to its bioinspired functionalization. For example, Cu- and Fe- based MOFs with histidine- or catechol derived linkers catalyze oxidation and peroxidation reactions similar to natural enzymes, offering enhanced durability compared to proteins [102,103].

Redox transformation by mimicking cofactor-dependent enzymes is led by the hybrid frameworks which contain either porphyrin-like or flavin-like linkers. In biosensing, coordination sites and biomimetic ligands platforms provided by the bioinspired MOFs are utilized for the selective detection of dopamine, Glucose and nucleic acids through specific recognition [104-106].

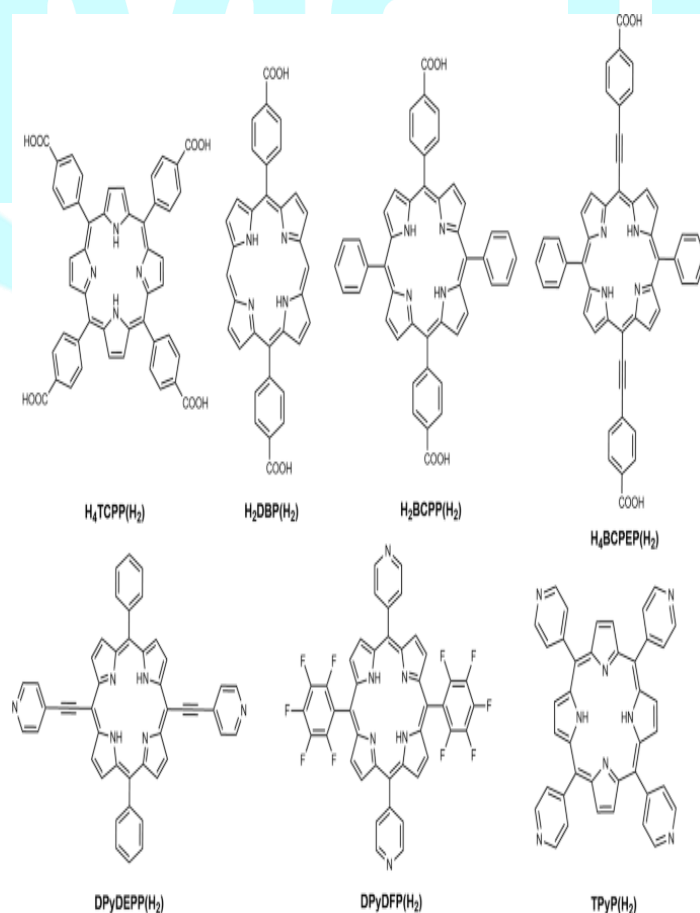


Figure 5: porphyrin derived MOF linkers [105]

4.1 Selective catalytic function

In the past two decades MOFs went through considerable effort in order to regulate its pore size/shape and porosity by optimizing the structures and components, which is conducive not only the realization of the size-selective catalysis but also, the molecules diffusion. Some researchers made modifications in order to defect the MOFs catalysis and control their size/morphologies by adding modifiers into the precursors [107-111].

- **Metal coordination mimicry**

Ligands featuring histidine, cysteine or carboxylate groups reproduce the coordination geometry of metalloproteins enabling redox reactions with high turnover rates [112,113].

- **Microenvironmental control**

Hydrophilic and hydrophobic patches around the catalytic site regulate substrate access and transition-state stabilization, leading to substrate-specific catalysis [114].

- **Cofactor-inspired reactivity**

Incorporating porphyrin-, flavin- or nicotinamide-like fragments reproduces enzymatic redox cycles, crucial for selective oxidation and reduction reactions [115].

- **Hydrogen bonding and π - π interaction**

Bioinspired linkers often utilize non-covalent interactions to control substrate orientation, reducing undesired side reactions [116].

a) **Bioinspired MOFs for selective oxidation reactions**

Selective oxidation is one of the most significant applications of bioinspired MOFs

Fe- and Cu-based MOFs functionalized with catechol or histidine ligands exhibit high activity in selective hydroxylation of phenols and alkanes, mimicking tyrosinase and methane monooxygenases enzymes [117].

Mn-porphyrins MOFs have shown remarkable selectivity for epoxidation of olefins avoiding overoxidation due to the enzyme-like confinement effects [118,119].

Incorporation of biotinylated linkers enables site-specific anchoring of substrates, enhancing product specificity [119].

b) Selective reduction and hydrogenation

Bioinspired ligands also enable controlled reduction and hydrogenation reactions. Nickel and cobalt based MOFs containing amino acid or imidazole-derived ligands mimic hydrogenases active sites, showing selective CO₂-to-CO or formate conversion with minimal H₂ evolution [112, 117, 119].

The incorporation of nicotinamide-like ligands provides hydride transfer capability akin to NADH, promoting mild and selective reduction of carbonyl compounds [16, 51, 58].

Peptide-functionalized MOFs provide chiral environments, achieving enantioselective hydrogenation of prochiral ketones and alkenes [120].

c) Enantioselective and regioselective transformations

The incorporation of chiral, bioinspired ligands has enabled MOFs to perform reactions previously limited to enzymes or homogeneous catalysts.

According to He et al., the amino acid-modified Zr-MOFs catalyze asymmetric aldol and Diels-Alder reactions with enantiomeric excesses exceeding 95%. While the DNA- and peptide-grafted MOFs exhibit substrate-shape recognition through hydrogen bonding, producing regioselective transformations in carbon-carbon coupling [121].

Biomimetic pocket engineering, where bulky side groups create substrate channels, has enhanced regioselectivity in oxidation of long-chain hydrocarbons [122].

4.2 Sensing function of MOFs

The integration of bioinspired design principles into metal-organic frameworks (MOFs) has opened a transformative pathway for developing next-generation chemical and biological sensors. MOFs- renewed for their exceptionally high surface area, tunable porosity and customizable metal-ligand chemistry offer an ideal scaffold for the selective detection of small molecules, ions and biomacromolecules. However, conventional MOFs,

while structurally versatile, often lack the molecular recognition capability required for precise sensing in complex biological or environmental systems [7,102, 112]

Recent advances demonstrate that bioinspired MOFs can act as nano-enzymes, optical transducers and electrochemical sensors capable of detecting a broad spectrum of targets from toxic gases and metal ions to glucose, dopamine and nucleic acid with high selectivity and sensitivity [114, 122].

The intrinsic catalytic sites often mimicking metalloenzymes such as peroxidase or oxidase provide signal amplification mechanism that allow for low detection limits even in complex samples. Furthermore, the incorporation of bioligands such as amino acids, peptides, cofactors and DNA fragments, endows MOFs with biocompatibility enabling their application in biosensing and medical diagnostics [49, 86]

Affinity-based recognition grafting peptides, aptamers, antibodies or small bioactive ligands to MOF surfaces introduces highly selective binding pockets for target analytes such as proteins, nucleic acids and small metabolites. Peptide-functionalized ZIF or UiO derivatives have been used as signal amplifying tags in immunoassays [120].

Nanozyme catalytic amplification MOFs bearing bioinspired ligands like catechol, histidine, porphyrin analogues or encapsulated metal centers act as enzyme mimics such as peroxidase/oxidase-like, catalyzing colorimetric or electrochemical signal generation and thus enabling low-concentration detection of H₂O₂, glucose, and other analytes [53,122].

Photoelectrochemical / luminescent transduction porphyrin and flavin like ligands or fluorophore tagged bioligands create MOFs with tailored light absorption / emission that change upon guest binding such optical shifts are used for ratiometric or turn-on sensing of metal ions, volatile organics and biomolecules [123].

Electrochemical sensing (conductive composites) bioinspired MOFs combined with conductive carbon, MXenes or noble-metal nanoparticles yield sensitive electrochemical sensors for neurotransmitters and metabolites in complex matrices (blood, sweat). Integration into flexible/wearable devices has been demonstrated for health monitoring [124].

Mass-based (QCM) and microcantilever sensing immobilizing selective bioligands on MOF-coated QCM chips yields high mass sensitivity and improved selectivity for small molecules and proteins by exploiting the porous uptake of the MOF and the ligand's affinity.

The advantages of bioinspired MOF sensors are improved selectivity through biomolecular recognition elements, Dual function recognition and amplification combining affinity ligands with nanozyme activity or optical reporters in the same MOF gives built-in signal amplification. Also, stability and protection encapsulation or immobilization within MOFs protects fragile biomolecules like enzymes and aptamers from denaturation and matrix interference.

5 Conclusion

Metal organic framework is considered a platform for bridging the gap between the synthetic materials and biological systems by integrating enzyme mimicking metal sites, and multi-functionality that were limited previously to the homogeneous and natural enzyme. In this review we highlighted the advances in MOF significant expansion in the structural diversity and practical applicability of bioinspired MOFs.

The bioinspired MOFs demonstrated an exaptational potential in chemical and biological sensing; their molecular recognition and signal amplification capability allows for the sensitive and selective detection in complex environments.

Despite these advances, challenges remain in achieving long-term stability under operational conditions, precise control over defect populations and large-scale industrial implementation.

Future research efforts should focus on rational active-site engineering, integration of computational design tools and development of sustainable, cost-effective production routes. With continued interdisciplinary collaboration, bioinspired MOFs are expected to play a pivotal role in the development in next-generation catalysts and sensors for environmental, industrial and biomedical applications.

Conflicts of Interest: “The authors declare no conflict of interest.”

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