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Recent Progress in Metal-Organic Frameworks for Dye Adsorption

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ABSTRACT

In recent years, the discharge of dye-laden wastewater has become one of the major contributors to aquatic pollution. Owing to their complex chemical structures and persistent aromatic systems, many synthetic dyes are difficult to remove by conventional treatment methods, which often fail to fully degrade or capture these contaminants. Metal-organic frameworks (MOFs), with their highly tunable porosity, abundant coordination-active sites, and advantageous chemical stability, have therefore attracted growing attention as promising next-generation adsorbents. This review begins by outlining the fundamental structural classes of MOFs, the mainstream synthetic strategies, and the key features that make them suitable for water purification. Particular emphasis is placed on recent studies that enhance adsorption performance through structural regulation, targeted functionalization, and the construction of MOF-based hybrid or composite materials. These approaches not only improve affinity toward dye molecules but also help address some of the inherent limitations of pristine MOFs. Finally, the review summarizes the remaining challenges and highlights the directions in which the field is likely to develop. Issues such as long-term stability, regeneration efficiency, and performance retention under realistic conditions still require careful attention. Nevertheless, continued advances in MOF-based composite materials point toward a broader and more reliable application of these systems in dye adsorption and wastewater treatment.

KEYWORDS

Dye wastewater; MOFs; Composites; Adsorption performance

1 Introduction

With the rapid advancement of global industrialization and urbanization, aquatic environments are facing increasingly severe contamination. Dye-containing wastewater is one of the major sources of pollution, characterized by large discharge volumes, high organic pollutant loads, intense chromaticity, significant composition fluctuations, and the presence of toxic and hazardous substances. If left untreated, such wastewater threatens aquatic life and poses risks to human health. As textile dyeing technologies continue to evolve, a growing variety of novel dyes are entering wastewater streams. For example, methylene blue (MB) is difficult to degrade or recover; when improperly treated, MB-containing industrial effluents discharged into receiving waters pose significant risks to ecosystems and public health [1]. To address this issue, various wastewater treatment technologies have been proposed, including adsorption, redox-based treatments, photocatalysis, membrane separation, and microbial biodegradation [2]. However, biological processes often require large footprints and stringent operating conditions, while chemical methods tend to be costly, generate sludge that is difficult to dispose of, and can impose additional environmental burdens. In contrast, adsorption is widely regarded as one of the most practical methods due to its low cost and operational simplicity. However, the efficiency of adsorption is influenced by the intrinsic properties of the adsorbent and the physicochemical environment; highly porous architectures and large surface areas are key to achieving high uptake and rapid kinetics.

Among the search for advanced adsorbents, porous materials have attracted significant attention, with metal-organic frameworks (MOFs) emerging as prototypical candidates. MOFs are crystalline, permeable, three-dimensional coordination networks constructed from polytopic organic linkers and metal ions/clusters. Due to their structural diversity, excellent thermodynamic and mechanical stability, tunable pore environments, exceptionally high surface areas, and reconfigurable metal sites, MOFs have been extensively researched in materials science. In recent years, they have shown considerable promise in catalysis, gas capture and storage, biomedicine, separations and adsorption, and as platforms for nanomaterials. In the context of dye adsorption, the high porosity, tunable chemistry, and topology of MOFs offer distinct advantages for pollutant recognition and removal, making them highly effective in liquid-phase adsorption of organic contaminants as well as in solid-phase extraction and decolorization applications.

This review provides an overview of the preparation methods, classification, and recent advances of MOFs in dye adsorption. We will discuss in detail the preparation techniques and structural characteristics of different types of MOFs, with a focus on their applications in dye adsorption, including the advantages and limitations of pristine MOFs. Through a review of existing studies, this paper also outlines the future directions for MOFs in the field of dye adsorption.

2 MOF Materials

2.1 Classification of MOFs

By judicious selection of metal ions and organic linkers—and by tuning synthetic conditions—diverse families of MOFs can be prepared with finely controlled pore sizes and geometries [3]. Representative classes include the isoreticular MOF

series (IRMOFs), coordination pillared-layer (CPL) architectures, the University of Oslo (UiO) series, zeolitic imidazolate frameworks (ZIFs), and the Materials of Institute Lavoisier (MIL) family.

2.1.1 Isoreticular Metal-Organic Frameworks (IRMOFs)

IRMOFs are constructed by bridging $[Zn_4O]^{6+}$ secondary building units with aromatic dicarboxylate linkers (e. g., terephthalate and its derivatives), yielding highly ordered three-dimensional octahedral lattices with tunable pore apertures and surface chemistry [4]. They can exhibit specific surface areas of several thousand square metres per gram, with pore volumes approaching nearly 90% of the crystal volume.

2.1.2 Coordination Pillared-Layer Frameworks (CPL)

CPL materials consist of two-dimensional coordination layers (typically based on hexacoordinate metal centers with carboxylate or azacyclic ligands) that are interconnected by neutral N-heterocyclic pillar ligands—such as 2,2′-bipyridine or 4,4′-bipyridine—into three-dimensional networks. The pillar ligands impart framework flexibility and enable adjustable pore metrics [5,6].

2.1.3 University of Oslo Series (UiO)

UiO frameworks are assembled from $Zr_6O_4(OH)_4$ nodes and terephthalate (BDC) or its derivatives to form highly symmetric, microporous architectures in which cages comprise octahedral and tetrahedral units. They display exceptional thermal stability (>500 ° C) and outstanding chemical robustness, tolerating strong acids/bases and hydrolytic conditions [7].

2.1.4 Zeolitic Imidazolate Frameworks (ZIFs)

ZIFs are formed by tetrahedrally coordinated Zn^{2+}, Co^{2+}, and related metal ions linked by imidazole or substituted imidazolate ligands. They combine the designability of MOFs with the thermo-chemical stability of zeolites, resisting high temperatures and a broad range of organic solvents [8].

2.1.5 Materials of Institute Lavoisier Series (MIL)

MIL frameworks employ transition or rare-earth metals (e.g., Fe, Cr, Al, V) with dicarboxylate and tricarboxylate ligands. Certain members (e.g., MIL-100, MIL-101) offer very high surface areas (>3000 $m^2 \cdot g^{-1}$) and excellent hydrothermal stability, retaining structural integrity under humid conditions [9].

2.2 Synthetic Methods for MOFs

2.2.1 Conventional Solvothermal and Nonsolvothermal Routes

Conventional syntheses most commonly use solvothermal or nonsolvothermal conditions. In solvothermal synthesis, reactions proceed in sealed vessels at temperatures above the solvent's boiling point under autogenous pressure, promoting slow crystal growth and affording MOFs with high crystallinity and large crystal size [10].

2.2.2 Microwave-Assisted and Electrochemical Synthesis

Microwave-assisted methods heat polar media volumetrically, enabling rapid temperature ramping and markedly shortened reaction times [11,12]. Using high dielectric-loss solvents enhances efficiency, often yielding uniformly sized, highly crystalline products; however, precise control of irradiation time is essential to avoid ligand decomposition. In electrochemical synthesis, anodic dissolution supplies metal ions that react with linkers under an applied field. This approach can deliver high-purity products but is constrained by the availability of suitable sacrificial anodes and requires optimization of electrolysis parameters to minimize side reactions [13].

2.2.3 Green Synthetic Strategies

Green approaches seek to reduce environmental impact through benign media, notably supercritical fluids and ionic liquids. In supercritical $scCO_2$, for example, ZIF-8 can form within nearly 5 min with high purity, and the medium is readily recyclable, though high-pressure equipment is required [14]. Ionic-liquid routes benefit from negligible vapor pressure and efficient heat transfer; for certain Zr-based MOFs, reaction times can be shortened from nearly 120 h to nearly 30 min with excellent product stability, albeit at higher cost.

2.2.4 Post-synthetic Modification (PSM) and In-situ Construction

PSM introduces functional groups into preformed frameworks via physical or chemical transformations, enabling precise incorporation of new functions without redesigning the de novo synthesis—provided that the framework's integrity is preserved [15,16]. In-situ construction incorporates functional components during MOF growth, ensuring intimate integration with the host lattice; this strategy is particularly advantageous in catalysis, flame retardancy, and related applications.

2.3 Advantages of MOFs in Water Treatment

2.3.1 Abundant Adsorptive Active Sites

Adsorption in water treatment can involve electrostatic attraction, π-π stacking, hydrogen bonding, and metal-O/N coordination, among other interactions [17]. In sulfate-radical-based advanced oxidation processes (SR-AOPs), both oxidants and pollutants can be co-enriched at catalytic active sites, markedly enhancing purification efficiency. MOFs, with high surface areas and well-defined channels, effectively concentrate oxidants and contaminants. For example, PHE can be adsorbed onto Al/Co bimetallic MOFs through electrostatic interactions and heterogeneous pore sites; pyridinic-N sites in Fe-ZIF-8/TiO_2 arrays can strengthen π - π interactions with PHE, facilitating uptake and subsequent transformation [18].

2.3.2 Coordinatively Unsaturated Metal Sites (CUS)

CUS in MOFs serve dual roles in adsorption and catalysis during pollutant degradation [19]. Their highly positive character allows strong oxidant binding, while vacant p orbitals act as Lewis-acid sites that promote oxidant activation. Common CUS include Fe^{2+}[20], Co^{2+}[21], and bimetallic pairs such as Fe^{2+}/Co^{2+}[22], Cu^{2+}/Co^{2+}[23], Fe^{2+}/Cu^{2+}[24], and Mn/Fe [25], which can undergo reversible redox cycling to activate persulfate (PS) and accelerate electron transfer.

2.3.3 Rich Surface Functionality

MOF surfaces often present hydroxyl, carboxyl, carbonyl, and pyrrolic-N groups that directly participate in oxidant activation and pollutant adsorption [26]. Electron-rich sites readily coordinate oxidants to form surface complexes that initiate pollutant degradation. For instance, hydroxyl groups in Fe-MOF@Mn_2O_3 strengthen binding with peroxymonosulfate (PMS); in MnS/Fe-MOF, multivalent metal centers adsorb water to generate metal-hydroxyl species that activate PMS to produce reactive oxygen species (ROS) for efficient degradation of organics.

3 Recent Advances in MOF Materials for Dye Adsorption

Through precise regulation of the intrinsic structure and surface chemistry of MOFs—such as increasing specific surface area, tuning the ratio of incorporated metals, and introducing functional groups (e.g.,-NH_2, -OH, -COOH)—their adsorption capacity and selectivity toward dyes can be markedly enhanced. As shown in Fig. 1(a, c), Keshta et al. [27] synthesized coordinatively unsaturated MIL-101(Cr) via an HF-free route; by simultaneously increasing specific surface area and the density of positive surface charge, they achieved highly efficient uptake of anionic dyes. MIL-101(Cr) also exhibited excellent chemical and aqueous stability, retaining more than 80% removal efficiency after five adsorption-desorption cycles. Xu et al.[28] prepared five Ni/Fe-MOFs with distinct microstructures by varying the Ni/Fe ratio, thereby demonstrating that selective adsorption of dyes can be engineered through structural optimization. With increasing Ni/Fe molar ratio, the morphology evolved from nanoflower-like (Fe-MOFs) to rhombohedral block-like (Ni-MOFs), accompanied by a gradual decrease in specific surface area. All materials showed high dye-removal performance in real water samples, with outstanding reusability and anti-interference behavior; compared with single-metal systems, the Ni/Fe-MOFs exhibited stronger overall adsorption competitiveness. As depicted in Fig. 1(b,d), Hu et al.[29] obtained a Zr-MOF (MOF-525) by solvothermal synthesis that displays strong affinity for anionic dyes. MOF-525 delivered high adsorption capacities for two benchmark anionic dyes—methyl orange (MO) and methyl red (MR)—and its capacity for both increased with rising sulfate (SO_4^{2-}) concentration; the material was also readily reusable. Using Fe-BDC synthesized at ambient temperature and pressure as the parent adsorbent, Wu et al[30]. implemented a synergistic dual modification—Cu (II) doping together with amino (-NH_2) functionalization—to enhance cation-dye selectivity. In mixed solutions of cationic methylene blue and anionic methyl orange, the optimized $Fe_{0.6}Cu_{0.4}$-BDC-NH_2 exhibited a 22.5-fold increase in the selectivity coefficient for methylene blue; moreover, in binary or multicomponent mixtures containing other cationic dyes (e.g., malachite green, rhodamine B), $Fe_{0.6}Cu_{0.4}$-BDC-NH_2 maintained excellent preferential adsorption of cationic species (Fig. 1(e)). Finally, Su et al.[31] leveraged the coordination of Zr^{4+} with formic acid (FA) by judiciously adjusting the water/FA ratio to generate Zr-O cluster solutions, enabling in-situ, room-temperature synthesis of MOF-808 within Zr_6 cluster media. The resulting MOF-808 is hydrophilic, highly stable, and possesses a high specific surface area together with a mildly positive surface, conferring ultra-high adsorption capacities for anionic organic dyes and high selectivity in cation-anion dye mixtures; it effectively separates complex dye mixtures as illustrated in Figure 1(f).

Further construction of MOF-based composite membranes or hybrid materials can integrate the exceptional specific surface area and tunable porosity of MOFs with the high throughput and mechanical robustness of membrane-separation technologies, yielding dual-functional systems that couple adsorption with separation. Such composite strategies not only enable efficient removal of complex contaminant mixtures but also markedly enhance materials' adaptability under diverse environmental conditions and their engineering applicability. As shown in Fig. 2(a,c), Peng et al. [32] fabricated a high-efficiency, environmentally benign membrane based on a MOF-COF hybrid. MOF nanofibers were grown in situ on bulk COF surfaces, substantially increasing the hybrid's specific surface area and thereby providing

abundant adsorption sites. Concurrently, functional groups on the FS-50 molecules endowed the hybrid membrane with pronounced superhydrophilicity and superoleophobicity, enabling selective, high-capacity adsorption of multiple dyes along with concurrent uptake of several pesticides; outstanding oil-water emulsion separation was achieved even for 50-milliliter emulsions. As illustrated in Fig. 2(b), Vinothkumar et al.[33] synthesized an amine-functionalized Fe-based MOF (MIL-101-NH_2(Fe)) by a solvothermal route and embedded the MOF particles into a polysulfone (PSf) matrix to construct a well-integrated Fe-MOF/PSf membrane. Introduction of amine groups enhanced optical properties and allowed effective modulation of surface charge, thereby affording efficient and selective adsorption of pollutants. The functionalized Fe-MOF was successfully incorporated into the PSf membrane, imparting significant hydrophilicity and enabling targeted adsorption, separation, and degradation of both cationic and anionic dyes. Xiang et al.[34] preinstalled hydrophobic methyl substituents on the organic linker to obtain a water-stable, porous anionic MOF (Bio-MOF-2Me) and a corresponding mixed-matrix membrane (Bio-MOF-2MeMMM). The membrane exhibited high stability, preferential adsorption, and strong separation performance; moreover, it maintained a negative electrostatic potential across different aqueous environments, thereby enabling exceptionally efficient adsorption and separation of target cationic and anionic dyes from mixed aqueous solutions. Li et al.[35] used ZIF-8 as a model filler and γ-poly(glutamic acid) (γ-PGA) as the gel matrix to prepare ZIF-8@γ-PGA hydrogels via in-situ growth. Uniform distribution of carboxylate sites improved the dispersion of ZIF-8 within the network, while ZIF-8 incorporation reduced the crosslinking density of the γ-PGA hydrogel, increasing its flexibility and adhesiveness. Adsorption tests with methylene blue as a representative cationic dye showed that the stratified, hierarchically porous architecture of the composite hydrogel accelerated adsorption kinetics and enhanced uptake efficiency—conferring clear advantages for rapid and high-efficiency adsorption of cationic dyes (Fig. 2(d,e))..

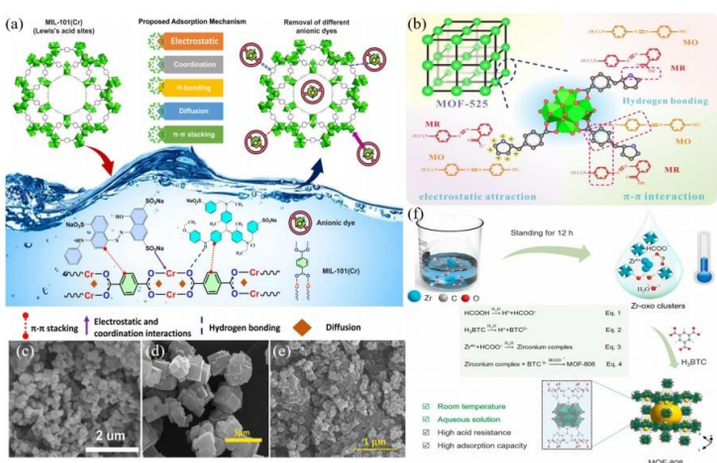

Figure 1 Modified MOF materials. (a) Schematic illustration of the adsorption mechanism of MIL-101; (b) schematic of the synthesis of MOF-525; (c) SEM image of MIL-101; (d) SEM image of MOF-525; (e) SEM image of $Fe_{0.6}Cu_{0.4}$-BDC-NH_2; (f) schematic of the synthesis of MOF-808

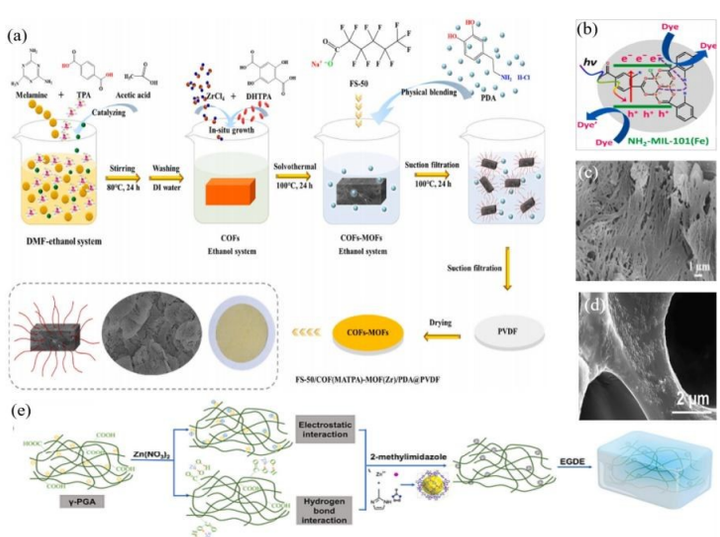

Figure 2 MOF-based composite membranes and hybrid materials. (a) Schematic of the fabrication of an FS-50/COF(MATPA)-MOF(Zr)/PDA@PVDF membrane; (b) proposed photocatalytic degradation mechanism of dyes over MIL-101-NH_2(Fe); (c) SEM image of the PVDF membrane; (d) SEM image of ZIF-8@γ-PGA; (e) schematic illustration of the preparation of the ZIF-8@γ-PGA composite

Optimization of MOF performance for adsorption-separation has evolved from single-parameter structural tuning to multidimensional, synergistic design. By introducing functional groups at the molecular scale, adjusting metal composition, and—at the macroscopic level—constructing composite membranes and hybrid architectures, researchers have achieved systematic improvements in adsorption capacity, selectivity, and stability. Nonetheless, modification can compromise framework integrity, induce pore blockage, or increase synthetic complexity, thereby limiting sustained deployment under real-world conditions. To overcome these bottlenecks, the development of MOF-based composites has become a key direction: such materials integrate the high specific surface area of MOFs with the mechanical robustness of membrane platforms, while enabling multifunctional synergistic effects.

4 Conclusions and Outlook

This review has systematically discussed recent advances in the adsorption-based treatment of dye-laden wastewater using MOFs, covering their structural characteristics, synthetic routes, modification strategies, and the construction of composite architectures. In particular, studies published in recent years have further clarified how their highly tunable porosity, abundant metal-active centers, and exceptionally large specific surface areas contribute to improved adsorption performance. These features give MOFs clear advantages in capturing organic dyes, heavy-metal ions, and other related contaminants that are typically difficult to remove through conventional processes.

Despite these benefits, pristine MOFs still encounter several practical limitations, such as restricted accessible surface area, nonuniform pore-size distribution, finite chemical stability, slower adsorption kinetics, and difficulties in achieving efficient regeneration after repeated cycles. Current efforts to address these issues mainly rely on structural regulation, targeted functionalization, and the fabrication of hybrid or composite materials that help compensate for intrinsic weaknesses. While MOFs remain a promising class of adsorbents, future development should place greater emphasis on improving recyclability and mitigating challenges such as pore blockage or gradual structural degradation during operation. Moreover, although notable progress has been made, obstacles remain for large-scale application, including insufficient stability under harsh industrial conditions, relatively high synthesis costs, and possible losses in surface area or functionality during modification or scale-up.

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