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MOF Composites as Adsorbents for Dyes: A Review

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ABSTRACT
The discharge of dye-laden effluents has become a major source of aquatic pollution. Owing to their complex chemistries and persistent aromatic ring systems, dyes are often recalcitrant to complete removal by conventional treatment technologies. Metal-organic frameworks (MOFs), featuring highly tailorable porosity, abundant coordinatively active sites, and excellent chemical stability, have emerged as promising next-generation adsorbents. This review particular emphasis is placed on the design and performance of diverse MOF composite systems—including MOF/graphene, MOF/chitosan, MOF/nanocellulose, and MOF/magnetic metal particle hybrids. These architectures realize multiscale synergistic effects that preserve the intrinsic high surface area and accessible porosity of MOFs while markedly improving mechanical robustness, cycling stability, and ease of recovery. Finally, the review discusses current challenges and delineates future research directions for MOF-based composites in dye adsorption and the large-scale deployment of MOF-based composites

KEYWORDS
Dye wastewater; MOFs; Composites; Adsorption performance

1 Introduction

With the rapid advancement of global industrialization and urbanization, the pollution of aquatic environments has become an increasingly serious issue, especially the discharge of dye-containing wastewater. Dye wastewater is characterized by large discharge volumes, high organic pollutant loads, intense chromaticity, and the presence of toxic and hazardous substances, posing a significant threat to aquatic ecosystems and human health if left untreated [1]. Although various wastewater treatment technologies exist, adsorption is widely regarded as an ideal method due to its low cost, simplicity, and operational ease. However, the choice of adsorbent significantly impacts the efficiency of adsorption. While pure MOFs materials offer high surface areas and tunable pore structures, they still face certain limitations, such as poor recyclability, insufficient mechanical strength, and uneven pore size distribution. As a result, researchers have increasingly sought to enhance the dye adsorption performance of MOFs through the development of MOF-based composites.

Research into MOF-based composites has become a critical approach for improving dye adsorption efficiency. By combining MOFs with other materials such as polymeric materials, nanoparticles, magnetic components, and carbon-based materials, the performance of MOFs has been significantly enhanced. For example, combining MOFs with graphene, chitosan, or nanocellulose has resulted in improved adsorption capacity, selectivity, and regeneration ability. These composite materials can effectively exploit the advantages of MOFs while addressing their limitations in practical applications, achieving more efficient treatment of dye-laden wastewater.

This review systematically examines recent advances in MOF-based composites for dye adsorption. We summarize the design principles and adsorption behaviors of representative composite systems, including MOF/graphene, MOF/chitosan, MOF/nanocellulose, and MOF/magnetic metal particle materials. Finally, we identify the current bottlenecks of MOF-based composites in dye adsorption and discuss future research directions.

2 MOF-Based Composites for Dye Adsorption

2.1 MOF/Graphene Composites

Graphene—a two-dimensional crystalline allotrope of carbon with sp2 hybridization—combines an exceptionally high specific surface area, outstanding electrical conductivity, and superior mechanical strength, affording broad potential in energy, environmental remediation, and catalysis. Integrating graphene with MOFs yields complementary advantages: graphene furnishes fast electron-transport pathways and structural reinforcement, whereas MOFs contribute tailorable pore systems and dense populations of adsorption/catalytic sites. The hybridization not only improves the electrical conductivity and cycling stability of MOFs but also suppresses graphene restacking and enhances interfacial reactivity. As shown in Figure 1(a,b), Xiao et al.[2] constructed a layered composite membrane GO@PTCDA-UiO-66-NH2 by intercalating PTCDA-modified UiO-66-NH2 between graphene oxide (GO) sheets. π-π conjugation between PTCDA and GO, aided by hydrogen bonding, anchored the MOF robustly within the GO galleries. The composite delivered a water permeance 1.7 times that of pristine GO membranes and achieved near-quantitative removal (nearly 100%) of methylene blue, Congo red, crystal violet, and disperse black 9. Incorporation of PTCDA-UiO-66-NH2 narrowed the interlayer nanochannels and introduced lateral adsorption sites, thereby reconciling high flux with high selectivity. The membrane retained structural integrity and separation performance under high salinity and extreme pH, and maintained excellent dye removal after

multiple cycles. As depicted in Figure 1(c), Jia et al.[3] prepared a ZIF-8/GO composite membrane whose separation derives from a synergy of size-sieving and electrostatic interactions. The membrane exhibited high water flux (56.94 L·m-2·h-1·bar-1) and strong rejection of both cationic and anionic dyes, with retention of methylene blue and methyl orange reaching 99.79% and 98.85%, respectively. After 18 h of continuous operation, dye removal remained above 90%, evidencing excellent operational stability and antifouling behavior. Figure 1(d–f), Kasula et al.[4] in situ deposited copper- and silver-based MOFs (Cu-MOF and Ag-MOF) onto GO sheets to obtain GO-Cu-MOF and GO-Ag-MOF hybrids. The room-temperature, nontoxic synthesis afforded rapid fabrication and significantly improved MOF specific surface area, surface charge characteristics, and the density of active sites. Graphene incorporation enhanced electron transport and active-site utilization, thereby strengthening adsorption toward organic dyes; for methylene blue, the GO-Ag-MOF achieved a removal efficiency of up to 98%, markedly outperforming the corresponding single-component MOFs. The hybrids retained more than 80% of their adsorption efficiency after three cycles, demonstrating good thermal stability and regenerability.

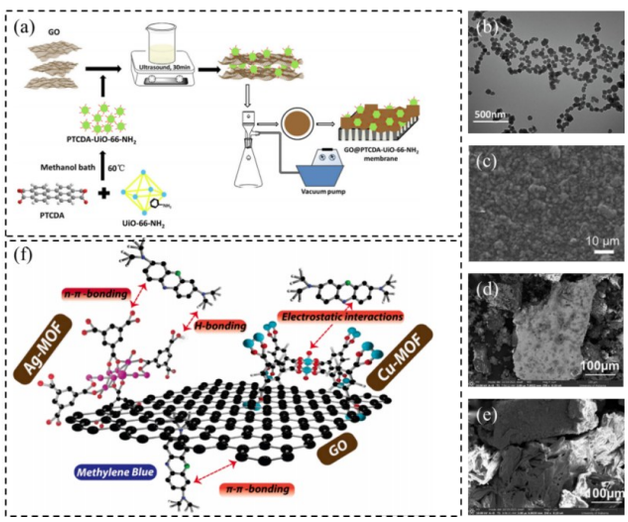

Figure 1 MOF/graphene composites. (a) Schematic of the fabrication of GO@PTCDA-UiO-66-NH2; (b) TEM image of GO@PTCDA-UiO-66-NH2; (c) SEM image of ZIF-8/GO; (d) SEM image of GO-Ag-MOF; (e) SEM image of GO-Cu-MOF; (f) schematic illustration of the adsorption mechanism of MB by GO-Ag-MOF and GO-Cu-MOF

2.2 MOF/Chitosan Composites

Chitosan (CTS) is a widely available natural polysaccharide that exhibits favorable biocompatibility, renewability, and a high density of hydroxyl and amino functionalities capable of coordinating with metal ions or organic ligands, thereby enabling efficient pollutant adsorption. Nevertheless, its intrinsically low specific surface area, limited mechanical strength, and susceptibility to dissolution under acidic conditions constrain its use in complex aqueous environments. Forming composites of chitosan with MOFs preserves the biodegradability of chitosan while markedly enhancing specific surface area, pore architecture, and chemical stability. The porous MOF lattice provides abundant adsorption and catalytic sites, whereas chitosan's functional groups strengthen complexation with metal ions and organic contaminants. The resulting porous networks combine high mechanical integrity with robust cyclic regenerability and have demonstrated high efficiency and durability for heavy-metal adsorption, organic dye degradation, and treatment of multicomponent wastewaters. As shown in Figure 2(a, c), El-Monaem et al.[5] fabricated a chitosan-modified Fe/MOF-5 bimetal-organic framework composite membrane (Fe/MOF-5@CTS). Incorporation of Fe/MOF into the chitosan matrix significantly increased the specific surface area and the density of positive surface charge, enabling high-efficiency adsorption of the anionic dye Congo red (CR). The maximum adsorption capacity reached 219.78 milligrams per gram, and after ten adsorption-desorption cycles the membrane retained 82% of its initial adsorption performance together with good structural integrity and mechanical strength, evidencing excellent reusability and durability. As depicted in Figure 2(b), Li et al.[6] employed an in-situ method to anchor Co-doped MIL-101(Fe) onto thiolated chitosan (CS-SH), yielding a bifunctional Co(II) -MIL-101(Fe)@CS-SH composite with combined adsorption and catalytic activity. Electrostatic and coordination interactions involving -SH and -NH2 groups enabled efficient adsorption of Cr(VI). In parallel, the material achieved 99.4% degradation of methylene blue within 20 minutes. In a mixed Cr(VI)-MB system, removal efficiencies for Cr (VI) and MB reached 99.1% and 88.1%, respectively; after five cycles, the composite retained 91.4% of its degradation efficiency and 74.4% of its adsorption capacity. Leaching of iron and cobalt remained below discharge limits, indicating excellent stability and regenerability. Using epichlorohydrin as a cross-linker, Yan et al.[7] covalently coupled chitosan with UiO-66-NH2 to produce a modified chitosan composite adsorbent (UNCS) that combines the high specific surface area of

the MOF with the multifunctionality of chitosan. The material exhibited outstanding Hg(II) uptake, with a maximum adsorption capacity of 896.8 milligrams per gram. Thermodynamic analysis indicated a spontaneous and exothermic process; Hg(II) removal exceeded 90%, and the composite maintained high adsorption capacity after five adsorption-desorption cycles (Figure 2(d,e)).

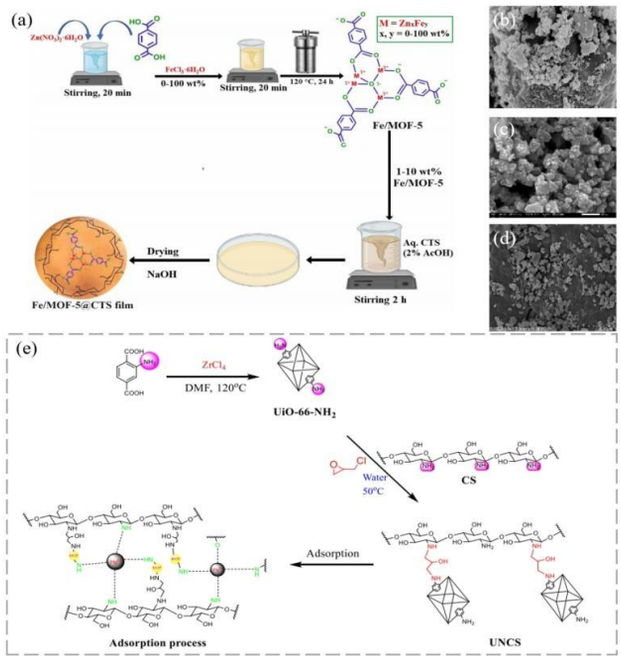

Figure 2 MOF/chitosan composites. (a) Schematic of the fabrication of Fe/MOF-5@CTS; (b) SEM image of Co(II)-MIL-101(Fe)@CS-SH; (c) SEM image of Fe/MOF-5@CTS; (d) SEM image of UNCS; (e) schematic illustration of the preparation of UNCS

2.3 MOF/Nanocellulose Composites

Cellulose is an abundant, structurally robust natural polymer with excellent biocompatibility, renewability, and a high density of hydroxyl functionalities capable of forming covalent or coordination bonds with diverse inorganic and organic components. Owing to its plentiful surface hydroxyl and carboxyl groups, cellulose exhibits useful adsorption behavior in water treatment; however, its intrinsic specific surface area is limited and the structure can collapse, yielding insufficient adsorption capacity and cycling performance when used alone. Forming composites with MOFs preserves cellulose's biodegradability while markedly increasing specific surface area, porosity, and chemical stability. The ordered porous lattices of MOFs furnish dense populations of active sites for adsorption and catalysis, whereas cellulose improves MOF dispersion and processability. Synergistically, these hybrids combine efficient adsorption, catalysis, and separation with mechanical robustness and reusability, enabling high-performance removal of heavy-metal ions and organic dyes. As shown in Figure 3(a,e), Wu et al.[8] used sodium carboxymethyl cellulose (CMC) as a template and employed a green, facile polymerization to produce highly porous MIL-100/CMC composite beads. This approach uniformly dispersed MOF crystals within the cellulose matrix, enhancing mechanical stability and mass-transport through the pore network. The resulting MIL-100/CMC-HD achieved a specific surface area of 1, 277.2 m2/g and effectively adsorbed and activated peroxymonosulfate (PMS) to degrade methylene blue (MB), attaining over 96% removal within 60 minutes. After five cycles, the material retained 93.6% of its degradation efficiency, indicating excellent structural stability and reusability. As depicted in Figure 3(d), Lin et al. [9] constructed a cross-linked network using CMC as the backbone with poly (ethyleneimine) (PEI) and sodium alginate (SA), within which ZIF-67 crystals were grown in situ to yield a CMC/SA/PEI/ZIF-67 composite. This architecture significantly improved MOF dispersion and mechanical strength, affording quantitative removal (100%) of Cr(VI) under neutral conditions. The composite membrane also activated peroxymonosulfate to generate sulfate SO4- and ·OH radicals, enabling complete degradation of the cationic dye rhodamine B within 120 minutes; after five cycles, treatment efficiency remained above 90%. Hu et al.[10] hydrothermally grew a Zr(IV)-based MOF (UiO-66) in situ on cotton fibers and subsequently modified the surface with hexadecyltrimethoxysilane (HDTMS) to obtain an ultrahydrophobic HDTMS-UiO-66@CFs composite. The material exhibited pronounced water repellence and oil affinity. The combination of the porous UiO-66 scaffold with the low-surface-energy groups introduced by HDTMS increased surface roughness and hydrophobicity, delivering adsorption capacities for both light and heavy oils exceeding 1, 100% by mass and achieving oil-water separation efficiencies up to 96%. The composite maintained stable superhydrophobicity across various oils, pH conditions, and complex aqueous matrices; after ten adsorption-compression cycles, the water contact angle remained above 150°, demonstrating excellent durability and reusability (Figure 3(b,c)).

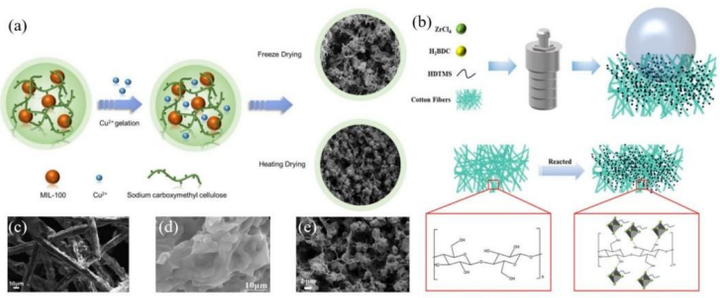

Figure 3 MOF/nanocellulose composites. (a) Schematic of the fabrication of MIL-100/CMC-HD; (b) schematic of the preparation of SH-UiO-66@CFs composite; (c) SEM image of SH-UiO-66@CFs; (d) SEM image of CMC/SA/PEI/ZIF-67; (e) SEM image of MIL-100/CMC-HD

2.4 MOF/Magnetic Metal Particle Composites

Magnetic metal oxides (e.g., Fe3O4) are widely employed for pollutant adsorption and catalytic removal owing to their strong magnetic responsiveness, chemical stability, and environmental compatibility. Under an external magnetic field, these materials can be rapidly separated and recovered, thereby simplifying process operations. However, single-component magnetic sorbents typically suffer from limited specific surface area and an insufficient density of active sites, which constrains their effectiveness in complex wastewaters containing multiple contaminants. Hybridizing magnetic phases with MOFs combines the high porosity of MOFs with the facile recoverability of magnetic components, simultaneously increasing specific surface area and adsorption capacity while imparting enhanced photocatalytic and Fenton-like catalytic activity. In such core-shell or closely integrated architectures, the magnetic core enables rapid separation, whereas the MOF shell provides abundant adsorption and reaction sites, yielding high efficiency, stability, and recyclability for heavy-metal ion capture, organic dye degradation, and photocatalytic transformations. As shown in Figure 4(a,c), Yang et al.[11] prepared a magnetic porous composite (Fe3O4-PSS@ZIF-67) by using Fe3O4 nanoparticles as the core and ZIF-67 as the shell, bridged by poly(sodium p-styrenesulfonate) (PSS). The resulting uniform core-shell structure retained the high porosity and specific surface area of ZIF-67 while exhibiting excellent magnetic responsiveness for field-assisted separation. The material showed strong affinity toward 2-nitroresorcinol, with a maximum adsorption capacity of 52.50 mg/g, and maintained structural integrity and adsorption performance over multiple adsorption-desorption cycles. As depicted in Figure 4(b, d), Zheng et al.[12] constructed a stepwise-assembled magnetic composite, Fe3O4@UiO-66@TzDa-COF, wherein Fe3O4 imparts magnetic responsiveness, the UiO-66 layer increases specific surface area and adsorption capacity, and a triazine-containing covalent organic framework (TzDa-COF) provides robust photocatalytic activity. The composite achieved very high photodegradation efficiencies for the anionic dyes malachite green and Congo red; at an initial concentration of 100 milligrams per liter, removal reached 99% within 40 minutes and 97% within 120 minutes, respectively. The corresponding apparent rate constants were 1.21 min-1 and 0.65 min-1, among the fastest values reported to date. After five cycles, the photocatalytic efficiency remained close to 99%, and the material could be magnetically recovered without loss of structural integrity. As shown in Figure 4(e), Chong et al.[13] employed a one-pot synthesis to obtain a Cu-MOF using 1,4-bis(1H-imidazol-1-ylmethyl)benzene as the linker, and introduced Fe3O4 nanoparticles in situ to produce a magnetically responsive Cu-MOF@Fe3O4 hybrid. Combining the magnetic separability of Fe3O4 with the adsorption activity of the Cu-MOF yielded highly efficient removal of perrhenate (ReO_4^-), achieving up to 97% removal under illumination—significantly higher than that of the pristine Cu-MOF. The composite maintained high adsorption efficiency over repeated cycles and could be rapidly recovered by an external magnetic field, demonstrating excellent stability and regenerability.

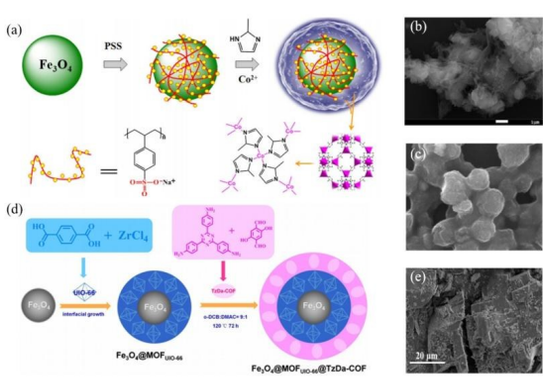

Figure 4 MOF/magnetic metal particle composites. (a) Schematic of the fabrication of Fe3O4-PSS@ZIF-67; (b) SEM image of Fe3O4@UiO-66@TzDa-COF; (c) SEM image of Fe3O4-PSS@ZIF-67; (d) schematic illustration of the preparation of Fe3O4@UiO-66@TzDa-COF; (e) SEM image of Cu-MOF@Fe3O4

3 Conclusions and Outlook

This review has provided a comprehensive overview of recent progress in MOF-derived composites for dye-laden wastewater treatment, focusing on their structural characteristics, and modification strategies. MOF-based composites, such as MOF/graphene hybrids, MOF/chitosan systems, and MOF/magnetic composites, have shown significant improvements in adsorption performance and recyclability. MOF/graphene composites enhance electron transport and surface area; MOF/chitosan and MOF/cellulose systems preserve biodegradability while improving mechanical strength and environmental tolerance; and MOF/magnetic composites provide high-capacity adsorption along with rapid magnetic separation.

Despite these advancements, several challenges remain for large-scale deployment of MOF-based composites. Firstly, many MOFs exhibit insufficient stability in harsh acidic or basic environments, which can lead to metal-node leaching or framework collapse. Therefore, developing more robust MOF architectures, such as multimetal systems, is essential for enhancing chemical durability. Secondly, many synthesis routes for MOF-based composites still rely on expensive organic solvents and high energy inputs. The development of greener and more cost-effective production methods is essential to meet industrial-scale demands. Thirdly, modifications to MOFs during composite formation may result in pore blockage or loss of surface area, which can reduce the adsorption efficiency at high dye concentrations. Furthermore, repeated regeneration can lead to structural weakening or deactivation of functional sites. To address these issues, future research should focus on the development of integrated composites that combine adsorption, catalysis, and in situ degradation, alongside standardized regeneration protocols to ensure long-term stability and performance in real-world applications.

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